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This study was conducted for the US Department of Energy by a Waste Isolation System Panel of the Board on Radioactive Waste Management under the National Research Council's Commission on Physical Sciences, Mathematics, and Resources. The panel was charged to review the alternative technologies available for the isolation of radioactive waste in mined geologic repositories, evaluate the need for and possible performance benefits from these technologies as potential elements of the isolation system, and identify appropriate technical criteria for choosing among them to achieve satisfactory overall performance of a geologic repository. Information has been acquired through examination of a large body of technical literature, briefings by representatives of government agencies and their industrial and university contractors, in-depth discussions with individual experts in the field, site visits, and calculations by panel members and staff, with deliberations extending over a period of approximately two years. The panel's principal findings are given. Chapters are devoted to: the geologic waste-disposal system; waste characteristics; waste package; conceptual design of repositories; geologic hydrologic, and geochemical properties of geologic waste-disposal systems; overall performance criterion for geologic waste disposal; performance analysis of the geologic waste-disposal system; and natural analogs relevant to geologic disposal. 336 references. (ERA citation 09:000237)

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A Study of the Isolation System for Geologic Disposal of Radioactive Wastes

Waste Isolation Systems Panel

BOARD ON RADIOACTIVE WASTE MANAGEMENT

Commission on Physical Sciences, Mathematics, and Resources

National Research Council

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This report has been reviewed by a group other than the authors according to procedures approved by a Report Review Committee consisting of members of the National Academy of Sciences, the National Academy of Engineering, and the Institute of Medicine.

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During the course of this study many individuals, representing many organizations, provided invaluable assistance to the panel, in many instances at some sacrifice and accompanied by less-than-gentle treatment by a panel impatient to get at the facts and to learn all it could on a broad range of topics relevant to the study. Space does not permit identifying all of those who so generously provided assistance.

Because of the important contributions of several individuals to the successful completion of the study, the panel wants to express its special appreciation to John Holloway, the staff officer of the National Research Council, whose diligent and thorough staff work turned a collection of committee contributions into a consolidated report; to Bruce Mann, who aided all panel members in their search for information and who made extensive contributions on radiological issues; to Paul Chambré, for his contributions to waste-form performance analysis; to Colin Heath, formerly of the U.S. Department of Energy (DOE), for establishing a high priority within that organization and with its contractor organizations in support of the panel's efforts; to Donald Vieth and Carl Cooley, also of DOE, who successively implemented this DOE policy during the course of this study; to Thomas McSweeney, of the Battelle Office of NWTS Integration, who provided invaluable assistance during the data collection phase of the study; to Donald Brown, of Rockwell Hanford Operations, for his timely and generous response to a seemingly endless series of requests for data and interpretation; to Gregg Korbin, a consultant to the panel, for his assistance in the collection and interpretation of data on rock mechanics; to consultant Harry Smedes, for similar assistance with data on tuff; and to Konrad Krauskopf, of Stanford University, for his evaluation of critical geochemical data and for his overall support of the panel's work.

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EXECUTIVE SUMMARY

1.1. INTRODUCTION

This study was conducted for the U.S. Department of Energy by a Waste Isolation Systems Panel of the Board on Radioactive Waste Management, under the National Research Council's Commission on Physical Sciences, Mathematics, and Resources. The panel was charged to review the alternative technologies available for the isolation of radioactive waste in mined geologic repositories, evaluate the need for and possible performance benefits from these technologies as potential elements of the isolation system, and identify appropriate technical criteria for choosing among them to achieve satisfactory overall performance of a geologic repository.

Information has been acquired through examination of a large body of technical literature, briefings by representatives of government agencies and their industrial and university contractors, in-depth discussions with individual experts in the field, site visits, and calculations by panel members and staff, with deliberations extending over a period of approximately two years. The panel's principal findings, in the form of overall conclusions and recommendations, are summarized in this chapter and treated in detail in subsequent chapters.

1.2. READINESS OF TECHNOLOGY FOR GEOLOGIC WASTE DISPOSAL

The current state of geologic containment technology offers two general categories of systems that are estimated, within the limits of this study, to meet the performance criterion adopted in this report. In the first category, total containment is provided by the long-term absence of flowing groundwater that would come in contact with the waste form. In the second category, low-velocity flowing groundwater is present, and adequate containment is estimated to result principally from low, solubility-limited release rates of the hazardous reactor products, geologic retardation, and a decrease in potential radiation doses to individuals that results from dispersion and dilution processes during transport and on discharge into surface water.

Conclusions

o With respect to the current options, the technology for geologic waste disposal has advanced to the state of a preliminary technical plan, suitable for testing and for further technical studies and pilot-facility confirmation. Conceptual and preliminary designs are under development. The technology is not yet ready for completing a final design, construction, and operation.

o Technical information now in hand and expected to be forthcoming, including conceptual design of repositories and analysis of repository performance, is likely to be sufficient for the selection of one or more candidate sites for in-situ testing. Following site selection, detailed exploration and underground testing at the candidate site or sites are likely to provide sufficient technical information to proceed with detailed design and construction of a repository. The extent to which further information is needed depends in part on the overall performance criterion.

o Technology that is not yet fully available could provide additional options in the future that would rely on containment within highly insoluble waste forms within longer-lived waste packages. Alternative waste forms that dissolve at a fractional rate of 10^{-7}/yr or less have been shown to be technically feasible and can offer potential advantages, particularly with respect to validation of long-term performance and possible relaxation of criteria for selecting qualified sites, e.g., water travel time to the biosphere and specified volume of surface water flow.

1.3. OVERALL PERFORMANCE CRITERION

The U.S. Environmental Protection Agency (EPA) has been working for several years to develop a standard for geologic waste disposal. The EPA staff has written a number of internal drafts and supporting technical reports that were reviewed during the panel's study. EPA issued a proposed standard for public comment after the panel's report was written. After several preliminary drafts, the U.S. Nuclear Regulatory Commission (NRC) has recently issued a proposed final rule incorporating detailed numerical criteria for individual components of the disposal system. The rule is intended to implement the yet unissued EPA standard. To the panel's knowledge, the U.S. Department of Energy (DOE) has not adopted an interim overall criterion, although DOE contractors' studies have assumed values of individual dose rate criteria for comparing with calculations of doses from radionuclide migration and release.

Lacking uniform guidance in this matter at the national level and with reservations regarding the technical basis of some parts of the standards and regulations in their current state of development, the panel has adopted its own overall performance criterion for the purpose of this study. This is described in detail in Chapter 8.

Conclusions

o Timely development of geologic waste-isolation systems and assessment of their adequacy require a general criterion that defines acceptable overall performance.

o The overall performance criterion to be used by federal agencies in designing a geologic waste-isolation system and in evaluating its performance has not yet been specified. This criterion could take the form of an upper limit on the radiation dose to an individual, the dose to a total population, or the cumulative amount of radioactivity released to the biosphere. The choice of this criterion will influence the selection and design of the repository and of the waste form and waste package.

o In the absence of an agreed-on performance criterion, one was adopted for the purpose of this study. The criterion selected is the lifetime radiation-dose commitment to the maximally exposed individual at any future time. The equivalent average lifetime dose rate selected is 10^{-4} sieverts per year (Sv/yr), about 10 percent of the whole-body dose rate from average natural gamma background. This is applied as a reasonable baseline value to compare with radiation exposures from expected future events, such as the individual doses resulting from the slow dissolution of waste solids in wet rock repositories and from the groundwater transport of dissolved radionuclides to the biosphere.

o A performance criterion based on population dose requires estimates of the numbers, location, and eating habits of future populations. It involves too many uncertainties to be useful as a primary criterion for protecting future mankind from the effects of geologic repositories. A repository sited to meet the performance criterion of a suitably low radiation dose to the maximally exposed individual is likely to meet a reasonable and realistic population-dose criterion, but a population-dose criterion does not necessarily result in suitably low radiation doses to all individuals.

o We did not adopt a time limit beyond which no future radioactivity releases and doses are to be computed, as in the 10,000-year time limit proposed by EPA. Only a small fraction of the radionuclides ultimately reaching the environment is expected to have been released during that time. Instead, we calculated future radiation doses for all times as long as potentially important doses were predicted to occur. Use of a 10,000-year time limit distorts the performance analysis of waste packages and of other components of the waste-isolation system.

1.4. GEOLOGIC WASTE-DISPOSAL SYSTEM

As described in Chapter 3, the system under consideration is a deep geologic repository for spent fuel or reprocessed waste from commercial nuclear power reactors. The system comprises the waste form and the balance of the waste package, the geologic repository for waste emplacement, and the surrounding geologic environment. Its objective is to protect humans now and in the future by isolating the waste from the environment effectively enough and for a period of time long enough that the amount of radioactive material ever reaching the biosphere will present no unacceptable hazard. To achieve this objective, a hierarchy of mechanisms exists to reduce the release of radionuclides to the biosphere and thus have the waste-isolation system meet the criterion of overall performance. One or more of the following release control mechanisms must be sufficient to meet the panel's criterion:

- delay of ingress of water
- slow dissolution of radionuclides
- slow release from the waste package
- long groundwater travel time
- delay due to sorption in the geologic medium
- dispersion
- dilution

The system must guard against an unacceptable release of the radioactive material into groundwater and the transport of this contaminated water to the biosphere--a principal pathway by which some portion of the buried radioactive material may eventually reach humans. This pathway is emphasized in this study.

The system most extensively analyzed by the panel is one for isolating waste from the reprocessing of uranium fuel discharged from commercial light-water reactors. The repositories analyzed are designed to accommodate spent fuel or 10-year-old reprocessing waste, even though most fuel to be reprocessed during the next several decades will have been stored for much longer periods of time, ranging up to 30 or 40 years. The high-level and transuranic wastes are assumed to be calcined and incorporated into a borosilicate-glass matrix or alternative waste form, stored for 10 years, and emplaced in deep underground cavities mined in basalt, granite, salt, or tuff.

The selection of the reactor fuel cycle can have an important effect on the amount of actinides in the waste. This selection and the choice of either spent fuel or waste from reprocessing as the material to be isolated will influence the performance of the geologic waste-disposal system.

The principal materials being considered for disposal are commercial spent fuel, high-level waste (HLW), and transuranic (TRU) waste. Concentrates of iodine-129, carbon-14, krypton-85, and tritium are also potential candidates, although, because of their volatility, they can be removed and treated separately from other wastes. Qualitative and quantitative characteristics of the various types of waste are described in Chapter 4.

A mechanism for reducing the release of radionuclides to the geologic medium is the time delay involved before water can penetrate to the waste form containing the radionuclides, allowing for substantial decay of those radionuclides with half-lives much shorter than the time delay in water penetration. The delay results from engineered barriers within the waste package and the time it takes for water to resaturate a filled and sealed repository, and it is affected by thermal, chemical, and hydrologic features and the design of the repository.

If water reaches the waste form, a principal mechanism available for reducing the overall rate of release of radionuclides to the geologic system is the low rate of release from the waste package. The rates at which radionuclides are released to the biosphere will be further reduced by radioactive decay during the time it takes for contaminated groundwater to travel to the biosphere and during the additional time delays due to sorption. For each radionuclide the extent of sorption delay depends on its chemistry and on the mechanical properties of the geologic environment.

Dispersion of some radionuclides during transport and their dilution on discharge to the biosphere are important processes. These processes do not affect the absolute amount of radionuclides reaching the biosphere, the details of groundwater flow, but they reduce the final concentrations and, therefore, the dose to an exposed individual. The important parameters are the total time and distance of radionuclide transport to the biosphere, the details of groundwater flow, and the volume of surface water, if any, into which the contaminated groundwater discharges as it reaches the biosphere.

Conclusions

o In repositories in basalt, granite, or tuff the processes of slow release, dispersion, dilution, and delay can become important at various times after the repository has been sealed. The relative importance of each process to waste disposal depends on the properties of the waste form and on the chemical and decay properties of each radionuclide. Our estimates of the performance of waste-disposal systems in these media predict that most of the radionuclides can be contained within the waste or the geologic media, or both, long enough to disappear by radioactive decay; but even after delays of thousands to millions of years a small proportion of the radionuclides will still reach the biosphere by groundwater transport.

o Dilution by surface water to reduce radiation doses to exposed individuals is important for a repository in basalt at Hanford, Washington, and for the generic granite repository assumed for this study, but such dilution is unavailable for a repository in the candidate tuff site.

o When groundwater is not initially present in the host rock of the repository, as in the case of a properly chosen salt deposit, containment within the waste package is complete unless humans intrude or unless groundwater intrudes into and flows through the repository. Because in such deposits water may not intrude for an indefinitely long time, dissolution and hydrogeologic transport of radionuclides to the biosphere are not expected. Processes of delayed and slow release, transport delays, dispersion, and dilution would provide additional protection if human intrusion or natural intrusion of groundwater were to occur.

o A repository in unsaturated tuff is expected to provide much longer time delays for potentially contaminated groundwater to travel to a given off-site location than is a repository in saturated tuff.

1.5. WASTE PACKAGE

Chapter 5 describes the different waste forms that have been proposed, discusses the choice of materials for canisters, overpack, and backfill, and reviews current designs of waste packages. Most important, the chapter analyzes the data and theories available for estimating rates of leakage of radionuclides into the surrounding groundwater after many years.

Conclusions

- The rates of release of radionuclides from candidate waste forms can be affected by the rate of transformation and dissolution of the waste-form matrix, by the rate of diffusion within the waste form, by the solubilities of stable compounds of low-solubility radioelements, and by the rates of diffusion and convection into the groundwater surrounding the exposed waste form. Laboratory data for the release of radionuclides from waste forms have not been shown to be applicable to predicting performance of waste packages in geologic repositories, and suitable laboratory and field experiments for determining releases from waste packages have not been carried out.
- The theory for solubility-limited dissolution of waste forms used in this study applies to the extent that effects of the formation and transport of colloids and unspecified complexes can be neglected. Based on the experimental and estimated solubilities quoted in this report, the theory predicts very low release rates for low-solubility radionuclides from the waste package. The concentrations of radionuclides later released to surface water are calculated to be low enough that the predicted doses from surface water will be several orders of magnitude below the individual-dose criterion adopted for this study.
- The effects of uncertainties in waste-package performance, including uncertainties in solubilities and unknown effects of colloids and complex formation, are important. Therefore, additional study needs to be undertaken to determine whether or in which cases the resulting rates of release to groundwater are solubility limited.
- If the fractional release rate of a candidate waste matrix is less than the solubility-limited release rate of a contained radionuclide, the rate of release of that radionuclide to groundwater will be determined by the rate of dissolution or physical/chemical transformation of the waste matrix, by the rate of diffusion of that radionuclide within the waste solid, or by both processes. It is theoretically predicted that release rates governed by diffusion within the solid could be as low as those governed by solubility.
- Uncertainties about the physical integrity of borosilicate glass exposed to leaching solutions at high temperatures and uncertainties as to the effect of physical integrity on radionuclide dissolution may require that glass high-level waste be protected from groundwater by a corrosion-resistant overpack when the repository rock is at temperatures greater than about 100°C. There are candidate overpack metals, such

as titanium alloys, that may protect the waste canister from groundwater; but these candidate metals require further testing in an assembled package under the mechanical, chemical, and thermal stresses that are expected in a geologic repository.

o Repositories are likely to be loaded initially with wastes derived from accumulated and aged discharged reactor fuel. With such loadings, maximum rock temperatures in current repository designs are predicted to be low enough, in the neighborhood of 100°C, to warrant confidence in the suitability of borosilicate-glass waste for these initial emplacements.

o There is need for additional information on the performance under repository conditions of 10-year-old high-level waste in borosilicate glass, for which repositories are currently designed. Experiments are needed to verify the predicted dissolution rate of radionuclides, to determine the extent of release by other mechanisms, and to determine the effect of long-term physical integrity on the rate of dissolution of radionuclides.

o There is need for a continuing program to develop new and better alternative waste forms.

o Borosilicate glass may be a suitable waste form for transuranic wastes. Uncertainties are similar to those for high-level waste, except that transuranic waste will be at ambient temperature.

o If the performance of borosilicate glass containing transuranic waste is to equal that of such glass containing high-level waste, it may be necessary to reduce the chemical contaminants normally associated with transuranic waste.

o Carbon-14 recovered when spent fuel is reprocessed, if converted to a concentrated insoluble form, is predicted to dissolve at a rate low enough that appreciable isolation of this radionuclide could be achieved.

o There are alternatives to geologic disposal of separated iodine-129 and carbon-14, such as sea disposal, but they have not been evaluated by the panel.

o Once the fuel cladding, canister, and overpack of a waste package containing unreprocessed spent fuel are breached by water intrusion, the rates of release of many critical radionuclides are likely to be greater than those for waste from fuel reprocessing. The resulting radiation doses from radionuclides transported to the environment are likely to be greater for unreprocessed spent fuel.

o More information is needed concerning the expected performance of a solid backfill surrounding the high-level waste canister and overpack. A dense backfill, such as compressed bentonite clay, may delay and reduce the discharge of radionuclides from the waste to the groundwater but at the expense of increasing waste temperatures. More information is needed on the chemical, physical, and radiation stability of candidate backfill materials under repository conditions and on the effective diffusion properties of backfill materials before backfill can be relied on to retard and reduce significantly the release of long-lived radionuclides to the surrounding groundwater.

o A backfill material may affect the leach rate of the waste form. Further studies are required to evaluate the synergistic and possibly antagonistic interactions among waste form, waste package, and backfill materials.

1.6. REPOSITORY DESIGN

Major factors and design features affecting the performance of geologic repositories in different rock types are discussed in Chapter 6, including proposed layouts, methods of excavation and stabilization, retrievability, plugging and sealing procedures, and the applicability of numerical codes and constitutive models to prediction of repository behavior. Particular attention has been given to the influence of temperature on short- and long-term performance.

Present conceptual designs of repositories loaded with 10-year-old commercial high-level waste show rock temperatures as high as 140°C in saturated tuff, 160°C in salt, 165°C in granite, 225°C in dry tuff, and 250°C in basalt. The basalt repository in which this high temperature will be reached is designed for a loading of heat-generating waste per area of emplacement about half that of the other repositories.

Repository temperatures can be reduced, if necessary, by additional above-ground storage of waste before emplacement, by decreasing the waste loading per package, by increasing the spacing between waste packages, by ventilation cooling, and/or by separation of the heat-generating strontium and cesium during reprocessing. About 25 additional years of above-ground storage would result in a twofold reduction in the maximum temperature rise in the repository. Since these design temperatures are much higher than the temperatures used in most of the laboratory studies of waste-form dissolution, there is need for waste-package release studies at higher temperatures.

Conclusions

- The development of repositories in candidate geologies such as basalt, granite, competent tuff, and bedded or domed salt is feasible in terms of present construction and mining technology. The costs of excavation, stabilization, and repository construction are likely to vary considerably among these different rock types. More exploration is needed to support the development of repository designs.
- Induced stresses from mining repository openings and heating will result in increased fracturing and permeability in the immediate vicinity of the repository rooms and in nearby shafts. In rocks other than salt this can result in a later increase in the local groundwater flow rates past the waste package. Under some repository conditions this greater flow may increase the dissolution rate of the waste, but at flow rates low enough that the dissolution rate will not be affected. The more important potential effect on long-term disposal is the creation of bypass pathways by which radionuclides could reach the biosphere, placing special demands on plugs and seals.

- Evaluating the performance of sealing and plugging systems and the extent of the disturbed zone around underground openings requires carefully controlled in-situ field experiments and measurements in the site-specific rock mass under consideration. Performance criteria for the underground openings and the sealing and plugging systems should be compatible with practical construction and mining procedures and should be realistic for field testing of performance.
- Retrievability of emplaced waste may be technically feasible, but the need for retrievability has not yet been substantiated. Retrieving emplaced waste in salt repositories would involve remining of backfilled storage rooms. Retrievability in other rock media appears to be feasible with or without backfill. In all media, however, retrieving the waste would be difficult, costly, and potentially dangerous. High temperatures of the waste package, the surrounding rock, and the repository rooms caused by radioactive decay heating are major concerns. In addition, a retrievability plan, if required, must include plans for handling and disposing of the retrieved waste.
- Proposals for retrievability of emplaced waste seem to have been premised mainly on the possibility of encountering geologic conditions that could render a repository site unsuitable. The need to design for retrievability could be obviated by a thorough site exploration program prior to any placement of waste. Because of reluctance to penetrate a repository horizon with a large number of drill holes from the ground surface, the exploration effort may have to be done at the repository level through drifts and drill holes.

1.7. GEOLOGY, HYDROLOGY, AND GEOCHEMISTRY

Except for human intrusion or such possible but unanticipated events as a volcanic eruption or other geotechnical cataclysms, the main pathway by which radionuclides in the waste could reach the biosphere after the repository has been sealed is by dissolution and hydrologic transport. Natural barriers to these processes include low solubilities of waste forms, sorption of key radionuclides, and lack of moving groundwater or sufficiently long water travel times. All of these, as well as repository construction, waste emplacement, and sealing, are affected by the properties of the geologic medium. The summary evaluation in Chapter 7 discusses the favorable and unfavorable aspects of some generic rock types of potential utility and some particularly well studied specific sites. Also included are an evaluation and selection of solubilities of various radioelements and their retardation coefficients due to sorption in various geologic media.

Conclusions

- Average water travel times from some potential repositories to the biosphere of several thousands to millions of years seem available, more probably in some repository rock types than in others. All these travel times are long enough for significant containment and decay of most of the radionuclides to take place.

- Although many candidate repository sites may be satisfactory, no repository rock type is certain, by itself, to provide complete containment of the radionuclides emplaced in a repository. The nearest to complete natural containment can be expected in a suitable salt deposit, whereby radionuclides could be released to the environment only by human intrusion or through some naturally disruptive event, such as water intrusion.
- Adequate repository sites can probably be most easily identified in thick bedded salt deposits.
- Adequate repository sites are probably available in volcanic tuffs. On balance, a deep repository above the water table has advantages over a repository below the water table.
- A hybrid type of geologic medium, with a repository in crystalline granitoids overlain by a regional aquifer, combines the advantages of two rock types while mitigating the disadvantages of each. This hybrid type offers the strength, continuity, and stability of granitic rocks with the favorable regional predictability of an overlying aquifer with slow flow rate and low hydrologic gradient. Some hybrid localities will have old immobile deep brines in pores, indicating hydrologic stability.
- Basaltic lava flows of the Pasco Basin, Washington, have favorable geochemical characteristics for retaining most radionuclides, i.e., low solubilities and high sorption potential. Outstanding hydrologic characteristics are travel time greater than 10,000 years and discharge into the Columbia River. Disadvantages are a relatively thin layer of host rock for constructing a repository, with likely boundary discontinuities; a relatively high ambient temperature of about 57°C; probable high horizontal stress in the repository host rock, likely to give rise to construction difficulties; proximity to permeable aquifers; and complex flow pathways in the geologic media surrounding the repository host rock.
- Adequate media for repositories are probably identifiable in granitoids that lack overlying sedimentary aquifers, in water-saturated or unsaturated rhyolitic tuffs, and in domal salt deposits; but identification of specific suitable sites will be difficult.
- The general increase of salinity of groundwater with depth may provide increased gravitational stability to reduce thermally induced flow from repository heating. Increased salinity also decreases the likelihood of future intrusion by humans to obtain water for human use. Ancient, gravitationally stable, and essentially immobile saline brines probably exist in the United States, but they have not been systematically investigated as indicators of potential repository sites.
- Uncertainties in the hydraulic properties of fracture-dominated rocks are best studied by in-situ experiments on groundwater flow.

1.8. OVERALL PERFORMANCE OF CANDIDATE REPOSITORIES

Chapter 9 presents predictions of long-term releases of radionuclides to the environment and resulting radiation doses to humans, based on data and evaluation of individual system elements elsewhere in the report. Predicted doses are compared with the performance criterion discussed in Chapter 8, and calculations are used to identify the performance features and limitations of each of the isolation mechanisms in the geologic disposal system.

Emphasis is placed on the expected long-term performance of the conceptual repositories in different rock media and the effect of uncertainties on that performance.

The following conclusions must be considered in the light of the preliminary state of any knowledge of solubilities and sorption under repository conditions. Additionally, release and transport phenomena as affected by colloids and complexes may alter these conclusions.

The following conclusions are based on the dose factors adopted and the predictions for all the geologic media considered in this study, assuming solubility-limited dissolution of waste solids.

Conclusions

o If we assume a repository completely loaded with wastes from fuel reprocessing and assume that contaminated groundwater will reach the environment within 10,000 years after the wastes are emplaced, the principal contributors to radiation dose to individuals in the future will be carbon-14, cesium-135, neptunium-237, lead-210, and selenium-79.

o For water travel times of about 100,000 years or longer, fuel-reprocessing waste can be adequately contained in granite, basalt, or tuff. Even if water intrudes into a salt repository within a few hundred thousand years after waste emplacement, water travel times of 100,000 years or longer after intrusion will ensure adequate containment of all the radionuclides in fuel-reprocessing waste.

o All radionuclides in unreprocessed spent fuel can be adequately contained. The peak doses--from carbon-14 and lead-210--are considerably greater than those from fuel-reprocessing waste.

o Essentially all of the iodine-129 in the unreprocessed spent fuel in wet-rock repositories will eventually reach the biosphere, but the resulting doses meet the performance criterion. This will also be true for a salt repository if water intrudes within 10 million to 20 million years after waste emplacement.

o There appear to be large uncertainties in the calculation of radiation doses that could result from the use of contaminated groundwater for growing food and drinking. The uncertainties are greater when estimating population doses than when estimating individual doses. A more active program to evaluate these uncertainties and to improve the accuracy of the dose estimates should be initiated, with focus on those radionuclides that are predicted to be the important contributors to radiation dose from waste in geologic repositories.

Criteria should not be expressed in terms of activity limits as long as such uncertainties exist.

o The new calculation techniques and metabolic data for estimating radiation doses from radium-226, lead-210, and neptunium-237, published in 1980 by the International Commission on Radiological Protection, provide important adjustments that have been included in this study in estimating the radiation dose from these radionuclides. This new information has not been used in many of the dose estimates for repository performance analyses supplied to the panel by the federal agencies and their contractors.

o All the repository media considered in this study provide some degree of sorption for all radionuclides other than carbon-14 and iodine-129. Sorption is most effective in retarding radionuclide transport in basalt, tuff, and granite and is least effective in the media surrounding salt. Based on the sorption data adopted for this study, to obtain the same degree of retention of cesium-135 in a salt site as is obtainable in any of the other media, and assuming water intrusion in less than 10 million to 20 million years, about a hundredfold longer water travel time is required for the salt site.

o None of the water travel times and sorption retardation properties for these different media present enough delay for significant radioactive decay of iodine-129 and uranium-238. However, the estimated solubility of uranium is low enough that it can be predicted that much of the uranium-238 and its more toxic decay daughters, radium-226 and lead-210, will be retained within the repository.

o For all the host-rock media considered herein, including a salt site with assumed intrusion of water, and based on the groundwater flow rates adopted for this study, the assumed use of the groundwater for drinking and irrigation before it mixes with surface water could result in radiation doses that are greater than the performance criterion of 10^{-4} Sv/yr. Because of the salinity of groundwater contaminated from a breached salt repository, use of this groundwater by future generations may be less likely than for other repositories.

o A large amount or high flow rate of surface water into which contaminated groundwater discharges is important in reducing the concentrations and radiation doses from radionuclides reaching the biosphere. For the basalt site the groundwater discharges into the Columbia River, resulting in the lowest calculated doses from groundwater transport of any of the repository media and potential sites considered here. Alternatively, subsurface discharge of groundwater into the ocean, as for a repository in a hybrid medium, could be equally or more effective in diluting the discharged radionuclides, and it would provide less opportunity for the discharged radionuclides to be ingested by humans in the future.

o In the region of the Nevada Test Site, where a possible repository in tuff is being considered, there is no identified moving surface water into which potentially contaminated groundwater from a repository in tuff would discharge. If the contaminated groundwater from a repository in tuff were to reach the surface without further dilution and at the groundwater flow rates and travel times assumed in this study, calculated dose rates would exceed the performance criterion of 10^{-4} Sv/yr. Because there is no flowing surface water in this region, there will be a greater incentive for future generations to use the groundwater for drinking and irrigation than in the case of a site located near a large amount of surface water.

- o Calculations indicate that a repository in tuff at the Nevada Test Site, if we assume a groundwater travel time to the accessible environment of 1,000 years or longer, would meet the EPA's proposed draft performance standard, which is based on a population-risk criterion and specifies limits to the cumulative radioactivity release of individual radionuclides over a period of 10,000 years. The ease of meeting EPA's release limits for this site and the difficulty of meeting the individual-dose criterion illustrate the differences in these two criteria as they affect the evaluation of repository performance.
- o Preliminary information indicates that the water travel time to the potentially accessible environment will be about 20,000 years longer in unsaturated than in saturated tuff. This may result in about a tenfold reduction in the estimated radiation dose to an individual who uses the groundwater, as compared with the individual groundwater dose for saturated tuff. However, the estimated individual groundwater dose for unsaturated tuff would still exceed the individual-dose criterion of 10^{-4} Sv/yr.

1.9. OVERALL RECOMMENDATIONS

In addition to specific courses of action implied by the conclusions listed above, the panel offers the following general recommendations, some of which cut across more than one area of technical endeavor:

- o The overall criterion for acceptable performance of a geologic repository should be specified by one or more appropriate federal agencies. In the absence of an overall criterion specified by other agencies, the Department of Energy should establish its own interim criterion for acceptable overall performance.
- o The primary performance criterion should be specified as an average lifetime dose rate to an individual at any future time.
- o The Department of Energy and its contractors should implement a continuing program to estimate the probable future doses from repositories that they are developing, including the estimating of uncertainties in these dose estimates.
- o Experiments should be carried out to resolve the uncertainty in the long-term physical integrity of borosilicate glass exposed to groundwater during the period of repository heating, and experiments should be carried out to determine the importance of the physical integrity of the waste form to system performance.
- o Experiments should be carried out to test the predicted solubility-limited dissolution rates of radionuclides under repository conditions and to resolve uncertainties with regard to formation and transport of colloids and complexes.

o The development program should include experiments and analyses of the effects of repository heating on the dissolution rate of radionuclides from waste forms exposed to groundwater. The temperature-dependent solubilities of the critical radionuclides should be determined.

o Experiments should be designed to determine the long-term release rate of cesium from borosilicate glass under repository conditions. Experiments are needed to determine the long-term release rate of actinides from transuranic waste in borosilicate glass and in other candidate waste forms under repository conditions.

o Even though present waste forms may be demonstrated to be satisfactory, the magnitude of the national program for geologic disposal, its extended time scale of development, design, and implementation, and the potential advantages of better waste packages justify a continuing backup program to develop better understanding of the main elements of geologic disposal and to provide even better waste-package alternatives that could be used in later stages of waste emplacement.

o The Department of Energy should support a continuing program of research and development on alternative waste forms. To provide against the contingency that release rates and uncertainties in release rates from glass prove to be considerably higher than are now estimated, alternative waste forms that are much more resistant to attack should be developed to the point where their relative merits can be more realistically evaluated. If the low release rates predicted by the theory on solubility-limited diffusion-convection are verified by appropriate experiments, then comparably low rates may be expected for some other waste-form candidates that have heretofore been considered to be relatively leachable. A more realistic assessment of leachability may modify the need for steps in some of the presently prescribed processes, and this should be investigated for more promising candidates.

o Waste packages for separated carbon-14 and iodine-129 should be designed to meet a specified performance criterion, and benefits from waste forms with low surface-to-inventory ratios should be considered. Disposal alternatives such as deep ocean disposal should also be considered.

o The manufacture of waste forms will entail the handling of very large quantities of high-level radioactive materials, coupled in some cases with high-temperature processing. The safety of this work, and the problems associated with disposal of process wastes and eventual decommissioning of the plant, can be factors in the choice among waste forms, given that performance criteria may be adequately met by several waste-form candidates. The analysis of release of radioactivity to the environment must include the entire waste-disposal cycle, beginning with waste-form manufacture, to achieve overall minimal release objectives.

o Experiments and analyses should be carried out to determine the probable failure modes of waste packages under repository conditions, with emphasis on performance of the waste package during intrusion of water into a heated repository.

o If unreprocessed spent fuel is to be emplaced in a repository, more information should be obtained on its rate of release of critical radionuclides under repository conditions.

o Continuing research, development, and testing are needed to clarify current uncertainties and to advance the design of the repository and its waste packages to the stage of technological acceptance. Even when the design has been accepted, continued research and development should be carried out to better understand the expected performance and to develop alternatives and improvements. These steps should continue through the life of the program, including repository construction, waste emplacement, and final sealing.

o A review should be made of the techniques for calculating radiation doses that could result from water contaminated by a geologic repository, and the uncertainties in these calculations should be determined. Consideration should be given to the development of standardized methods for calculating these radiation doses, including the development of appropriate and updated metabolic parameters and environmental transfer parameters for the important radionuclides. These techniques for dose calculation and the parameters used in them should be reassessed periodically.

o If ongoing work at candidate bedded salt sites should show that these sites are unsuitable, several other known bedded salt deposits with favorable characteristics should be investigated.

o The U.S. Department of Energy should institute a more deliberate overall technical review of its program on geologic disposal. This technical review should be done on a continuing and extended basis, with full technical input representing the technical breadth of the program and with emphasis on relating program efforts to the goal of developing a repository with predictable satisfactory performance. The program management should develop better technical coordination between the many different contractors, it should broaden the charter and expertise of its review groups to ensure adequate consideration of interdisciplinary problems, and it should develop a greater systems analysis content in its management and review.

o The current fragmentation of the waste-disposal program among several federal agencies should be reduced.

o Retrievability should be reassessed. An extensive exploratory program at the repository level, carried out prior to any waste emplacement, should be considered as an alternative to ensure that unexpected geologic features that could subsequently render a site unsuitable will not be encountered.

o A cost-benefit analysis and system optimization should be performed to evaluate effects of the emplacement distance between waste packages, the storage time before emplacement of packages, the concentration of radionuclides in the waste form, and the use of an overpack resistant to corrosion.

2

THE CHARGE TO THE PANEL

During the next few years, federal agencies will enter a period in which critical decisions will need to be made with respect to the disposal of long-lived, high-level radioactive wastes--decisions that must be founded on more meaningful technological information and criteria than exist at present. Much of the work to date on geologic isolation of such waste has attempted to optimize one or more of the elements in a series of largely decoupled barriers against the release of potentially hazardous radionuclides into the biosphere. However, results from such an approach are of only limited value in decision making, and they do not provide a basis for development of a sound rationale for selecting individual technological elements of a geologic isolation system.

As technology has advanced and design concepts have become increasingly sophisticated, more and more options for selection of the various waste-isolation system elements have become available to improve the safety margins of geologic isolation and to reduce the probability of release of radionuclides to the biosphere. At the same time there has been a growing recognition that the elements and subsystems of any proposed geologic isolation system cannot meaningfully be analyzed or optimized individually but must be considered in the context of the complete ensemble. This panel was charged to review the status of the alternative technologies available, evaluate the need for and possible performance benefits from these technologies as potential system elements, and identify appropriate technical criteria for choosing among them to optimize the overall performance of a geologic repository.

Some related major concerns were deemed to lie outside the study's purview. Among them were the following:

- proliferation of nuclear weapons
- whether spent fuel should be reprocessed
- the need for and future of nuclear power
- the economics of alternative nuclear fuel cycles
- management of defense nuclear wastes
- biological effects of low-level radiation
- what risks and radiation exposures are, or should be, acceptable to the public
- waste management alternatives other than deep geologic isolation

- hazards from the surface storage of spent fuel
- transportation of nuclear wastes

Other considerations were identified that would have an impact on the ultimate performance of a waste-isolation system and that the study would therefore need to address:

- the choice of the nuclear fuel cycle (e.g., no reprocessing, reprocessing with recycling)
- the time for decay of radionuclides prior to their emplacement in a geologic repository
- the selection of the physical and chemical forms of the waste materials and canisters
- the choice of the geologic media
- the selection of design temperatures for the wastes and surrounding rock in the repository
- the use of multiple engineered barriers to the mobilization and transport of radionuclides

Evaluation of the performance of a repository requires definition of one or more indexes of performance. One such index to be considered was the estimated amount of and radiation exposure from each of the important hazardous radionuclides that might eventually be transported from the waste repository to the biosphere as a function of time. Likely and possible properties of the components of the isolation system and the possible variability and uncertainty in these properties were seen to be important inputs to these overall predictions of long-term radionuclide releases, as was an assessment of the uncertainties in such long-term estimates.

The types of wastes to be considered from the commercial nuclear fuel cycle include unreprocessed discharged fuel, high-level reprocessing wastes, and transuranic wastes, each in a different solid form and amount and contained in a specified canister material. In addition to the fission products and transuranic elements in high-level and transuranic wastes, the radionuclides iodine-129, carbon-14, and krypton-85 are to be considered. These will be present in unreprocessed fuel, or they may be recovered separately in fuel reprocessing. Each of these many different waste forms gives rise to a different potential for possible contamination of the biosphere.

The importance of different kinds of technological barriers varies with the waste sources. Discharged fuel and high-level reprocessing wastes, both with significant content of fission products, require some barrier to prevent or protect against the possible release of strontium-90 and other potentially toxic fission products, especially during the first few hundred years after emplacement. Relevant barriers to environmental release include the waste form and canister, possible overpack material, suitably low groundwater velocity, sorption by the geologic medium, and distance to the nearest entry into the biosphere.

The endurance of the waste form and canister as possible barriers for these heat-generating wastes depends on their material and chemical properties, the radiation level and the cooling time prior to emplacement in the repository, the amount of waste per canister, the choice of temperature for the wastes and surrounding rock, the chances and time for water intrusion, and the chemical properties of the groundwater. It is difficult to assess the importance of one of these properties alone without relating it to the performance of the geologic isolation system as a whole. Review of the status of each of the alternative technologies and the evaluation of the possible benefit of each of them to the overall performance of the geologic isolation system during the first few hundred years after emplacement were to constitute one focus of the study.

The possibility of longer-term (more than 1,000 years) contamination of the environment from the longer-lived species in these wastes, as well as the possibility of longer-term intrusion of groundwater and deterioration of the waste material, may place additional requirements on the natural and technological barriers that are to prevent or retard the transport of radionuclides from the wastes to the biosphere. A different set of radionuclides becomes more important after the first thousand years--the transuranics and their daughters, the longer-lived fission products technetium-99 and iodine-129, and longer-lived activated species such as carbon-14. Although little heat is generated by decay in this period, the earlier time-dependent temperature history of these wastes and their possible earlier, thermally induced interaction with moisture and rock may affect their longer-term release and transport through the geologic medium. The extent to which the waste form and its earlier history can be and need be controlled as one of the barriers to the longer-term migration of radionuclides was to be reviewed. Also to be considered was the possible efficacy of and need for overpack materials, such as those designed to sorb particular radioactive species that might be released from the wastes or others designed to control the chemical environment or amount of moisture in the neighborhood of the waste containers.

Similarly, other waste sources and waste forms and the system of design variables and barriers needed to provide successful containment of these wastes within the isolation system were also to be considered. Each waste source will have its own particular needs in terms of choice of barriers, preemplacement cooling time, and repository environment.

For operational purposes, the panel formulated the following questions to be answered during the study:

o What criteria are appropriate for use in assessing the long-term performance of the geologic waste-isolation system?

o What is the likely and expected long-term performance of the geologic waste-isolation system?

o What properties of the components of the waste-isolation system are sufficient to meet the criteria for performance?

o What uncertainties arise in the prediction of long-term performance, and to what extent can we rely on present knowledge in predicting this future performance?

o What is the state of technology of waste packages and other engineered barriers? Is sufficient performance available with present technologies? What benefits can be reasonably expected from alternatives proposed or under development, and on what time scale?

o Are the federal programs reasonably designed to achieve the necessary and sufficient results when needed?

o To what extent are technologies being developed for defense wastes applicable to possible future commercial reprocessing wastes?

In effect, these questions framed the mission of the two-year effort, which the panel undertook through eight specific tasks:

1. Review of existing isolation system studies.
2. Review of waste sources and characteristics.
3. Review of technologies for waste forms and canisters.
4. Review of technologies for engineered barriers.
5. Development of geologic-hydrogeologic-geochemical parameters for systems analysis.
6. Review of criteria for systems analysis.
7. Performance analysis of the geologic isolation system.
8. Overall evaluation of technological alternatives.

The anticipated end result was to be a report that would benefit the program planning for geologic waste isolation, provide one of the tools for decision making by agencies of the federal government, and provide a basis for developing meaningful technological criteria related to the expected and necessary performance of the waste-isolation system.

3

THE GEOLOGIC WASTE-DISPOSAL SYSTEM

The waste-disposal system under consideration in this study is a deep geologic repository for spent fuel or reprocessed waste from commercial nuclear power reactors. The system comprises the waste form and the balance of the waste package, the geologic repository for waste emplacement, and the surrounding geologic environment.

The objective of the geologic waste-disposal system is to protect future humans from the radioactive waste. This is accomplished principally by isolating as much as possible of the waste from the environment. Radioactive waste is to be disposed deep underground under conditions such that almost all of the radioactive material disappears by radioactive decay while underground, and such that the amount of radioactive material ever reaching the biosphere is so small as to present no unacceptable hazard to humans. Geologic isolation is to be achieved by placing suitably prepared packages of solid radioactive waste into cavities mined in deep underground rock. The waste-isolation system must guard, among other things, against eventual dissolution of the radioactive material into groundwater and the transport of this contaminated groundwater to the biosphere. Dissolution and groundwater transport may be the most important potential pathway for some portion of the buried radioactive material eventually to reach humans, and it is the pathway emphasized in this study.

The features of the geologic waste-isolation system that protect against waste dissolution and hydrogeologic transport are as follows:

o A repository host rock can be selected that has no flowing groundwater, nor is expected to have any for many thousands of years, so that the rate of waste dissolution can be made small or zero. Little standing and no flowing groundwater is present in bedded or domal salt (standing water in the form of brine pockets may exist in salt formations), and very little is expected in unsaturated tuff above the water table. Alternatively, a repository host rock can be selected that has only a small flow rate of groundwater, so that the rate of dissolution of waste is small and so that the time required for groundwater to flow to the boundaries of the repository host rock is sufficient to allow radioactive decay of many of the radionuclides that do become dissolved in the groundwater. Low flow rates can be found in basalt and granite formations and in saturated tuff. Bedded and domal salt, tuff, basalt, and granite are the repository host rocks considered in this study.

o The waste package can be designed to delay the exposure of the radioactive solid waste form to groundwater and to decrease the rate of dissolution of radionuclides when and if the waste form is so exposed.

o The waste form itself can be designed to have high mechanical integrity, low bulk solubility, low surface area exposed to water, and low solid-phase diffusion rates for radionuclides of concern and thus limit the release of radionuclides when and if the waste form becomes exposed to groundwater. The dissolution rate of many of the radionuclides may be further limited by the low solubility of their chemically stable species in groundwater.

o The site can be selected so that the time for contaminated groundwater to travel from the repository host rock to the biosphere is long enough for many of the potentially hazardous dissolved radionuclides to decay before reaching the biosphere.

o Additional time delays for radioactive decay result from the sorption of most radionuclides on the minerals in the repository host rock and, importantly, in the media between the repository host rock and the biosphere.

o Bore holes, access shafts, and mined openings for exploring and developing the underground repository can be plugged and sealed to reduce the pathways for groundwater to reach the waste and for contaminated groundwater to flow toward the biosphere.

o The repository site can be selected to reduce the probability of human intrusion.

The site, its natural features, and the engineered components of the isolation system are the key elements of the geologic waste-isolation system.

Because complete isolation of radionuclides in a geologic repository is impossible, the geologic waste-disposal system must protect future humans from the relatively small amounts of radionuclides that will ultimately be released to the environment. Natural features that contribute to this protection are the following:

o The concentration of radionuclides in the groundwater, and potential radiation doses from these radionuclides, can be further reduced by dispersion and by diluting interflows of aquifers in the region between the repository host rock and the biosphere.

o If the contaminated groundwater discharges into large volumes of surface water, the concentration of radionuclides in the surface water and radiation doses therefrom can be low as a result of dilution by the flowing surface water. Dispersion during groundwater transport can lengthen the time periods over which discharges to surface water occur and can reduce the concentrations and radiation doses from radionuclides in surface water.

The geologic waste-isolation system analyzed most extensively in this study is the system to isolate waste resulting from reprocessing uranium fuel discharged from commercial light-water reactors. The repositories analyzed are designed to accommodate unreprocessed spent fuel or wastes resulting from reprocessing fuel 165 days after it is discharged. The waste is to be stored 10 years before emplacement in a repository. However, most spent fuel to be reprocessed during the next several decades will have been discharged from the reactors for much longer periods, ranging up to 30 to 40 years. The high-level and transuranic wastes are assumed to be calcined and incorporated into a borosilicate-glass matrix, stored for 10 years, and then emplaced into underground cavities mined in either basalt, granite, salt, or tuff. Waste forms that may be alternatives to borosilicate glass are also considered. Emphasis is given to the prediction of the long-term performance of the geologic waste-isolation system. Where possible, hydrologic, geologic, and geochemical properties and repository designs for specific sites now under consideration are used in the evaluation and assessment of system performance.

The geologic waste-isolation system also includes those operations, carried out before waste is emplaced in a repository, that prepare the waste packages for emplacement and the other operations that affect the radioactive content and heat generated by the waste when it is later emplaced in the underground repository. The operations of waste solidification and of waste-package preparation, transport, and emplacement must not themselves add radioactive contaminants to the environment in amounts that would negate the benefit from geologic isolation of the resulting waste.

Because decay heat generated by high-level waste in an underground repository can affect the rock and waste temperatures and their system performance, the length of time that high-level waste is stored before underground emplacement is an important system parameter. Consequently, above-ground preemplacement storage of high-level waste can be considered a part of the waste-disposal system. Repositories are being designed on the basis of above-ground storage for at least 10 years after discharge from the reactor. An additional 25 years of above-ground storage would about halve the heat generation rate and temperature rise in the repository. Varying the distance between waste packages in the repository and varying the amount of radioactive material within each waste package are alternative means of adjusting the temperatures in the repositories. The technological need for such temperature controls is considered in the present study.

The selection of the reactor fuel cycle can have an important effect on the radionuclide content of the wastes for geologic disposal and must therefore be considered an important decision element of the waste-isolation system. The wastes already being generated by today's reactors must be accommodated, whether by treating spent fuel as waste or by separating the waste in fuel reprocessing plants. Wastes generated by reprocessing spent fuel from the present light-water power reactors contain less plutonium and uranium than if unreprocessed spent fuel is chosen as the waste to be emplaced, and the reprocessing waste can be incorporated into solid forms and packages better designed for isolation performance.

Reprocessing wastes from possible future nuclear fuel cycles that involve recycling plutonium in light-water reactors or fast-breeder reactors will contain greater quantities of long-lived radioactive actinides than do the wastes from reprocessing uranium fuel from light-water reactors, and there will be different quantities of some fission products and activated species, such as carbon-14. Similarly, there can be significant differences in the amounts of some of the long-lived radionuclides in the waste generated by fueling reactors with thorium and enriched uranium, as in the power reactor at Fort Saint Vrain, Colorado.

The following chapters deal with the state of scientific knowledge and technology of each of the important elements of the geologic waste-isolation system. The subjects considered are the radioactivity properties of the wastes (Chapter 4); waste forms and packages (Chapter 5); mined repositories (Chapter 6); geologic, hydrologic, and geochemical properties of sites with different host rocks (Chapter 7); criteria for overall performance of the waste-isolation system (Chapter 8); evaluation of overall system performance (Chapter 9); and natural analogs relevant to geologic disposal (Chapter 10). Conclusions and recommendations resulting from these evaluations are presented in the Executive Summary (Chapter 1).

4

WASTE CHARACTERISTICS

4.1. INTRODUCTION

Materials that are being considered for emplacement in geologic repositories are commercial spent fuel, high-level waste (HLW), and transuranic (TRU) waste. Fuel assembly structure material (frequently referred to as cladding) is a particular TRU waste that can be characterized separately, and concentrates of iodine-129, carbon-14, krypton-85, and tritium are also potential candidates for geologic disposal. Because of their volatile nature, these latter four species can be removed and treated separately from other wastes. Finally, there is a special burden of material from the Three Mile Island Nuclear Station that is candidate material for disposal in a repository as a consequence of the March 1979 accident at that installation. All of these materials are derived both from the commercial nuclear generation of electricity and from the national defense program; however, spent fuel becomes a waste form only in the event that it is not reprocessed for recovery of its uranium and plutonium content.

Proposed regulations governing the disposal of spent fuel and high-level waste in geologic repositories appear in the Code of Federal Regulations (10 CFR 60). Environmental Protection Agency (EPA) standards that limit the potential magnitude of krypton-85 and iodine-129 releases from nuclear fuel cycle facilities have been published (U.S. Environmental Protection Agency 1976), and standards for carbon-14 and tritium are under consideration.

Defense wastes that have accumulated over the past 36 years at government sites greatly exceed in volume those from the commercial sector, but their radioactivity is significantly less than that of the commercial spent fuel now in inventory (Table 4-1). Defense high-level waste is stored as alkaline liquids, sludges, salt cake, and capsules of separated strontium and cesium at Hanford; as alkaline solutions, sludges, and salt cake at the Savannah River Plant; and as acid solutions and granular calcine at the Idaho Chemical Processing Plant. Prior to 1970, defense TRU waste was emplaced in shallow land burial, but since that time it has been stored retrievably at the surface. The projections of defense waste in Table 4-1 are based on the assumption that present waste management practices will continue through the end of this century.

TABLE 4-1 Current and Projected Characteristics of Key Nuclear Waste Materials

	End of Calendar Year 1980			End of Calendar Year 2000		
Category of Material	Volume (m^3)	Activity (MBq)	Thermal Power (MW)	Volume (m^3)	Activity (MBq)	Thermal Power (MW)
Defense						
HLW	291,000	4.8×10^{10}	3.4	335,000	5.9×10^{10}	4.8
TRU						
Buried	273,000	1.5×10^{7}	0.007	273,000	1.1×10^{7}	0.007
Stored	61,000	4.1×10^{7}	0.022	142,000	1.1×10^{8}	0.065
Commercial						
Spent fuel	3,100[a] (6,700 MgHM)	3.7×10^{11}	39	31,600[a] (72,000 MgHM)	2.0×10^{12}	190
Reprocessing wastes[b]						
HLW	2,200	1.4×10^{9}	0.1	29,400[c]	1.0×10^{12}	110
Cladding				18,700[d]	1.3×10^{10}	2.6
TRU					2.9×10^{7}	0.2
^{129}I					7.0×10^{4}	0
^{14}C					1.5×10^{6}	0
^{85}Kr					1.2×10^{10}	0.5
^{3}H					1.2×10^{9}	0

NOTE: 1 megabecquerel (MBq) = 2.7×10^{-5} curie (Ci); 1 curie = 3.7×10^{4} megabecquerels.

[a] Based on nominal overall dimensions of the fuel assemblies. MgHM = megagrams of heavy metal.

[b] Based on reprocessing in the year 2001 the 72,000 MgHM that are projected to be discharged through the year 2000.

[c] High-level waste (HLW) is stored as 0.378 m^3 of solution per MgHM reprocessed.

[d] Volume of uncompacted cladding and structural material is 0.18 m^3/MgHM and 0.38 m^3/MgHM for PWR and BWR fuel, respectively.

[e] Volume of TRU waste is 68 m^3 per metric ton of plutonium processed. Waste from fabrication of plutonium-bearing fuel is not included.

SOURCE: Adapted from data presented in U.S. Department of Energy (1981) and Croff and Alexander (1980).

Most of the commercial spent fuel is stored at the nuclear power stations; however, about 487 megagrams of heavy metal (MgHM) are presently stored at the Nuclear Fuel Services (NFS) plant in New York and at the Midwest Fuel Recovery Plant in Illinois. ("Heavy metal" denotes the sum of uranium and transuranium elements that are present.) In addition, there are about 5 MgHM of diversified spent fuels from numerous Department of Energy (DOE) and commercial research and test reactors in storage at the Savannah River Plant and the Idaho Chemical Processing Plant (U.S. Department of Energy 1981). The commercial high-level waste is in storage as 2,150 m^3 of alkaline solution and sludge and 45 m^3 of acid solution at NFS. The remaining commercially generated reprocessing wastes have been emplaced with large volumes of low-level wastes at the NFS burial grounds or have otherwise been released to the environment. Projections of commercial fuel and waste are based on the installed U.S. nuclear capacity increasing from 53.8 gigawatts of electric power (GWe) in 1980 to about 180 GWe in the year 2000, and on all of the accumulated spent fuel being reprocessed in the year 2001.

Although the characteristics of commercial spent fuel and wastes are emphasized in this chapter, the basic nuclear waste technology in use today was developed within defense programs, and during the past five years government-sponsored research and development have focused on the disposal of defense wastes and commercial spent fuel. Defense wastes are similar to the anticipated commercial wastes in many respects, but significant differences, when they occur, are discussed briefly within the chapter.

4.2. NUCLEAR FUEL CYCLE ALTERNATIVES

Although many fuel cycle alternatives have been evaluated from considerations of nuclear proliferation and safeguards, it is apparent that the light-water reactor (LWR), the high-temperature gas-cooled reactor (HTGR), and the liquid-metal-cooled fast breeder reactor (LMFBR) are the most technically viable options in the United States for the next few decades. The LWR and LMFBR are based on the use of uranium and recycled plutonium fuels, while the HTGR uses thorium and uranium-233 (Figure 4-1). The once-through LWR fuel cycle with slightly enriched UO_2 fuel represents the present U.S. situation. After being discharged from the reactor, the fuel may be stored in surface facilities for 10 to 50 years and then transported to a repository for disposal. For this fuel cycle the spent fuel constitutes the only waste destined for disposal in a repository.

The other fuel cycle alternatives are based on reprocessing the spent fuel to recover the uranium, thorium, and plutonium, which may then be recycled in fresh fuel to the reactors. The reprocessing and recycle fuel fabrication steps give rise to the variety of wastes shown in Figure 4-1. The wastes from different fuel cycles are very similar in their nuclear, chemical, and physical characteristics. The principal exceptions are in the HTGR fuel cycle where SiC hulls are residues from reprocessing rather than metallic cladding, and about four times as much carbon-14 is generated per gigawatt(electric)-year as in the LWR and LMFBR (Davis 1979). Also, the TRU waste from the HTGR fuel cycle would contain principally uranium-232 and uranium-233 and their decay daughters rather than the transuranic elements. Commercial fuel that may be reprocessed over the remainder of this century will be almost entirely uranium-235-enriched fuel from LWRs, and the LMFBR may be an important source of nuclear power thereafter. Since the HTGR currently has a lower development priority in the United States, the wastes from this fuel cycle are not further discussed.

4.3. SPENT FUEL AS A WASTE FORM

4.3.1. Characteristics of LWR Fuel Assemblies

LWR fuel assemblies (Blomeke et al. 1978) are composite units of fuel pins in a geometric cluster held together by end pieces and a number of grid spacers. Although boiling-water reactor (BWR) and pressurized-

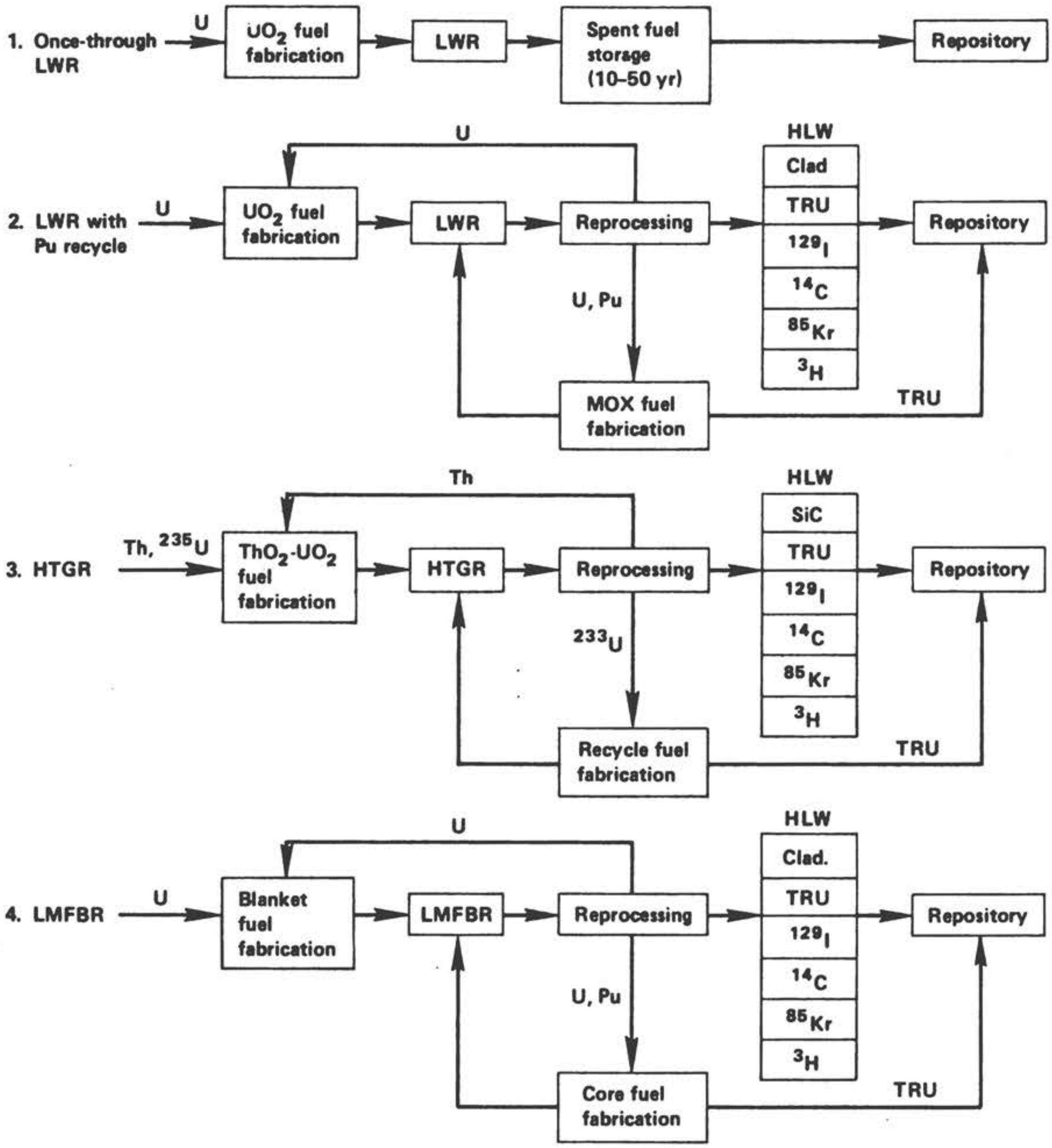

FIGURE 4-1 Sources of wastes from nuclear fuel cycles.

water reactor (PWR) fuel assemblies differ significantly, the basic components of each are the fuel pins, which are long sections of zircaloy tubing filled with ceramic pellets of uranium dioxide or mixed uranium-plutonium dioxide. Physical characteristics of typical fuel assemblies are given in Table 4-2. When considering spent fuel assemblies as a waste form, two relevant characteristics are overall size and weight. A typical BWR assembly has a 13.9 x 13.9 cm cross section, an overall length of 447 cm, and a weight of 275 kg. A typical PWR fuel assembly has a 21.4 x 21.4 cm cross section, an overall length of 406 cm, and a weight of 658 kg. Approximately 175 assemblies are discharged each year by a 1,000-MWe BWR and about 60 assemblies are discharged annually from a 1,000-MWe PWR.

Pertinent irradiation parameters of enriched-uranium LWR fuels are summarized in Table 4-3. An assembly is irradiated in a BWR (PWR), producing an average of 4.75 (17.3) MW of thermal power. After the equivalent of 1,062 (880) full-power days of irradiation, it is discharged. At this time, it contains uranium with a uranium-235 enrichment of 0.68 (0.84) wt% and 1.57 (4.32) kg of plutonium. The spent fuel also contains fission products, activation products, and isotopes of neptunium and transplutonium elements.

Calculations to predict the relevant characteristics of spent BWR and PWR fuel assemblies were performed with the ORIGEN 2 computer code using the input data of Tables 4-2 and 4-3 (Croff and Alexander 1980). Two relevant characteristics of spent fuels are the thermal power and the radioactivity as a function of decay time, and the three major groups of fuel constituents are the structural materials (cladding, grid spacers, etc.), the actinides, and the fission products.

The variations of the thermal power and the radioactivity of spent BWR and PWR fuel assemblies are shown in Figures 4-2 and 4-3, respectively. The fuel assembly structural materials are negligible

TABLE 4-2 Physical Characteristics of Typical Unirradiated LWR Fuel Assemblies

	BWR	PWR
Overall assembly length (m)	4.470	4.059
Cross section (cm)	13.9 × 13.9	21.4 × 21.4
Fuel pin length (m)	4.406	3.851
Active fuel height (m)	3.759	3.658
Fuel pin o.d. (cm)	1.252	0.950
Fuel pin array	8 × 8	17 × 17
Fuel pins/assembly	63	264
Assembly total weight (kg)	275.7	657.9
Uranium/assembly (kg)	183.3	461.4
Uranium dioxide/assembly (kg)	208.0	523.4
Zircaloy/assembly (kg)	56.9[a]	108.4[b]
Hardware/assembly (kg)	9.77[c]	26.1[d]

[a] Includes zircaloy fuel pin spacers.

[b] Includes zircaloy control rod guide thimbles.

[c] Includes stainless steel tie plates and Inconel springs.

[d] Includes stainless steel nozzles and Inconel-718 grids.

TABLE 4-3 Typical Irradiation Parameters of LWR Fuels

Parameter	BWR	PWR
Uranium/assembly (kg)		
Initial	183.3	461.4
Discharge	176.3	440.7
Enrichment (wt% ^{235}U)		
Initial	2.75	3.20
Discharge	0.69	0.84
Plutonium/assembly at discharge (kg)	1.57	4.32
Average power (MWth/assembly)	4.75	17.3
Average specific power (KWth/kg initial uranium)	25.9	37.5
Average discharge burnup MWthd/Mg initial uranium)	27,500	33,000
Irradiation time (full-power days)	1,062	880

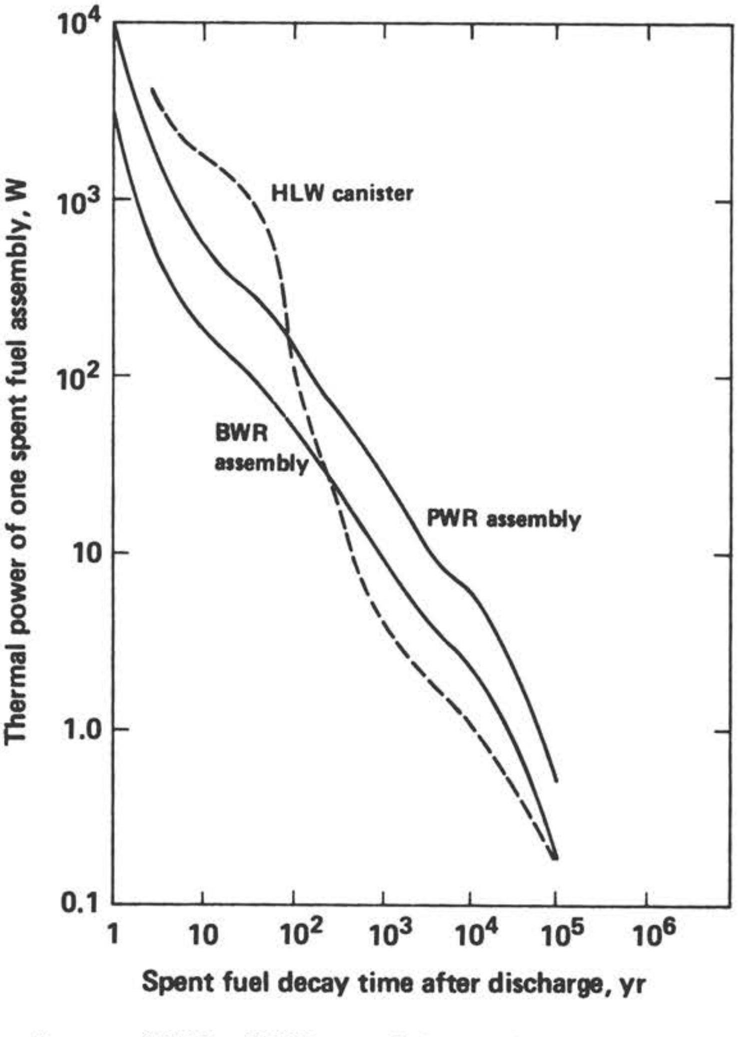

FIGURE 4-2 Thermal power of BWR and PWR spent fuel assemblies and of a canister of high-level waste.

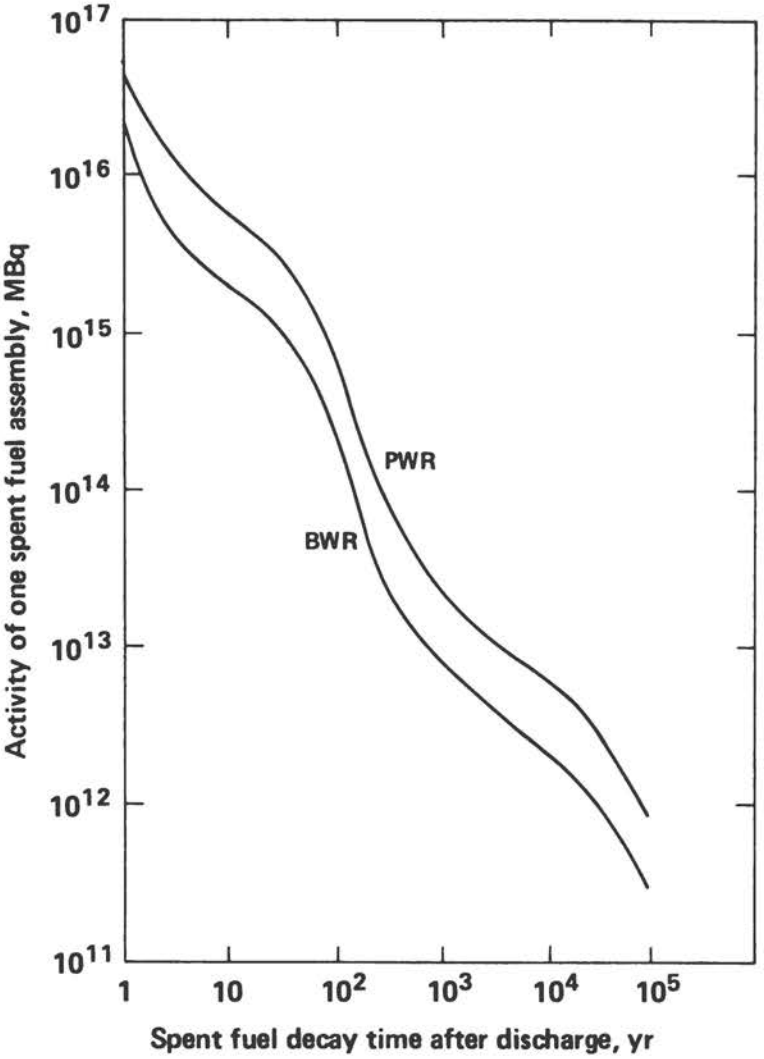

FIGURE 4-3 Activities of spent BWR and PWR fuel assemblies.

contributors to the thermal power, activity, and radiotoxicity of the assemblies at all decay times. The fission products dominate all three characteristics at decay times of less than 100 years, while the actinides dominate at decay times greater than 300 years. At decay times between 100 and 300 years, both the fission products and actinides contribute substantially to the totals.

The mass and radioactivity of nuclides in a PWR fuel assembly that are of significance in assessments of geologic isolation are given in Table 4-4.

Water dilution volume (WDV) is frequently used to identify and compare the major contributors to the radiotoxicity of various waste materials. For this purpose, the WDV is usually defined as the volume of water required to dilute the radionuclides to concentrations specified by the U.S. Nuclear Regulatory Commission (1982, Appendix B,

TABLE 4-4 Significant Radionuclides in One PWR Spent Fuel Assembly

	Time After Discharge					
	10 yr		100 yr		1,000 yr	
Nuclide	g	MBq	g	MBq	g	MBq
^{14}C	1.6×10^{-1}	2.5×10^{4}	1.6×10^{-1}	2.5×10^{4}	1.4×10^{-1}	2.3×10^{4}
^{79}Se	2.7	7.0×10^{3}	2.7	7.0×10^{3}	2.7	6.9×10^{3}
^{90}Sr	2.0×10^{2}	1.0×10^{9}	2.3×10^{1}	1.1×10^{8}	~ 0	~ 0
^{99}Tc	3.6×10^{2}	2.3×10^{5}	3.6×10^{2}	2.3×10^{5}	3.6×10^{2}	2.3×10^{5}
^{126}Sn	1.3×10^{1}	1.3×10^{4}	1.3×10^{1}	1.3×10^{4}	1.3×10^{1}	1.3×10^{4}
^{129}I	8.3×10^{1}	5.3×10^{2}	8.3×10^{1}	5.3×10^{2}	8.3×10^{1}	5.3×10^{2}
^{135}Cs	1.4×10^{2}	6.3×10^{3}	1.4×10^{2}	6.3×10^{3}	1.4×10^{2}	6.3×10^{3}
^{137}Cs	4.4×10^{2}	1.4×10^{9}	5.5×10^{1}	1.8×10^{8}	~ 0	~ 0
^{226}Ra	1.6×10^{-7}	5.4×10^{-3}	1.2×10^{-5}	4.2×10^{-1}	1.4×10^{-3}	4.9×10^{1}
^{234}U	8.8×10^{1}	2.0×10^{4}	1.2×10^{2}	2.7×10^{4}	1.5×10^{2}	3.3×10^{4}
^{238}U	4.4×10^{5}	5.4×10^{3}	4.4×10^{5}	5.4×10^{3}	4.4×10^{5}	5.4×10^{3}
^{237}Np	2.1×10^{2}	5.4×10^{3}	2.7×10^{2}	7.2×10^{3}	6.6×10^{2}	1.7×10^{4}
^{238}Pu	6.0×10^{1}	3.7×10^{7}	3.0×10^{1}	1.8×10^{7}	2.6×10^{2}	1.8×10^{4}
^{239}Pu	2.3×10^{3}	5.3×10^{6}	2.3×10^{3}	5.3×10^{6}	2.3×10^{3}	5.1×10^{6}
^{240}Pu	1.1×10^{3}	8.7×10^{6}	1.1×10^{3}	8.7×10^{6}	1.0×10^{3}	7.9×10^{6}
^{241}Pu	3.5×10^{2}	1.3×10^{9}	4.6	1.8×10^{7}	8.6×10^{-5}	3.6×10^{2}
^{242}Pu	2.1×10^{2}	3.1×10^{4}	2.1×10^{2}	3.1×10^{4}	2.1×10^{2}	3.1×10^{4}
^{241}Am	2.3×10^{2}	2.9×10^{7}	5.0×10^{2}	6.4×10^{7}	1.2×10^{2}	1.5×10^{7}
^{243}Am	4.0×10^{1}	2.8×10^{5}	3.9×10^{1}	2.8×10^{5}	3.6×10^{1}	2.6×10^{5}
^{245}Cm	5.6×10^{-2}	3.5×10^{2}	5.5×10^{-2}	3.5×10^{2}	5.1×10^{-2}	3.3×10^{2}
^{246}Cm	4.3×10^{-2}	4.9×10^{2}	4.3×10^{-2}	4.9×10^{2}	3.7×10^{-2}	4.2×10^{2}

SOURCE: Alexander et al. (1977).

Table II) as being the maximum acceptable in drinking water for unrestricted use. The International Commission on Radiological Protection (ICRP) has recently published new values for radiation protection guidance for occupational exposure that include significant changes for several isotopes that are important in considerations of waste isolation (International Commission on Radiological Protection 1979). Notably, the permissible concentration of strontium-90 was increased by a factor of 13, and that of its daughter yttrium-90 by a factor of 3; the concentration of radium-226 was increased by a factor of 8; and concentrations for technetium-99 and iodine-129 were increased by factors of 2.5 and 11, respectively. On the other hand, the permissible concentrations of neptunium-237, americium-241, plutonium-239, and plutonium-240 were reduced by factors of 340, 25, and 6, respectively (Croff 1981). The WDV of 1 Mg of spent PWR fuel showing the major contributors as a function of decay time and based on the new ICRP-recommended values is given in Figure 4-4 (Croff et al. 1982). As a measure of the impact of these new values, a comparison of the WDV of this fuel with that of the uranium ore required to fabricate the fuel is presented in Figure 4-5 for both the values recommended by the U.S. Nuclear Regulatory Commission (1982) and those recommended by ICRP (Croff et al. 1982). The use of ICRP-30 (International Commission on Radiological Protection 1979) results in a significantly lower total WDV during the first 100 years because of the lower WDV for strontium-90; however, it is substantially higher thereafter because of the higher WDVs of the several transuranic isotopes and the reduced WDV of

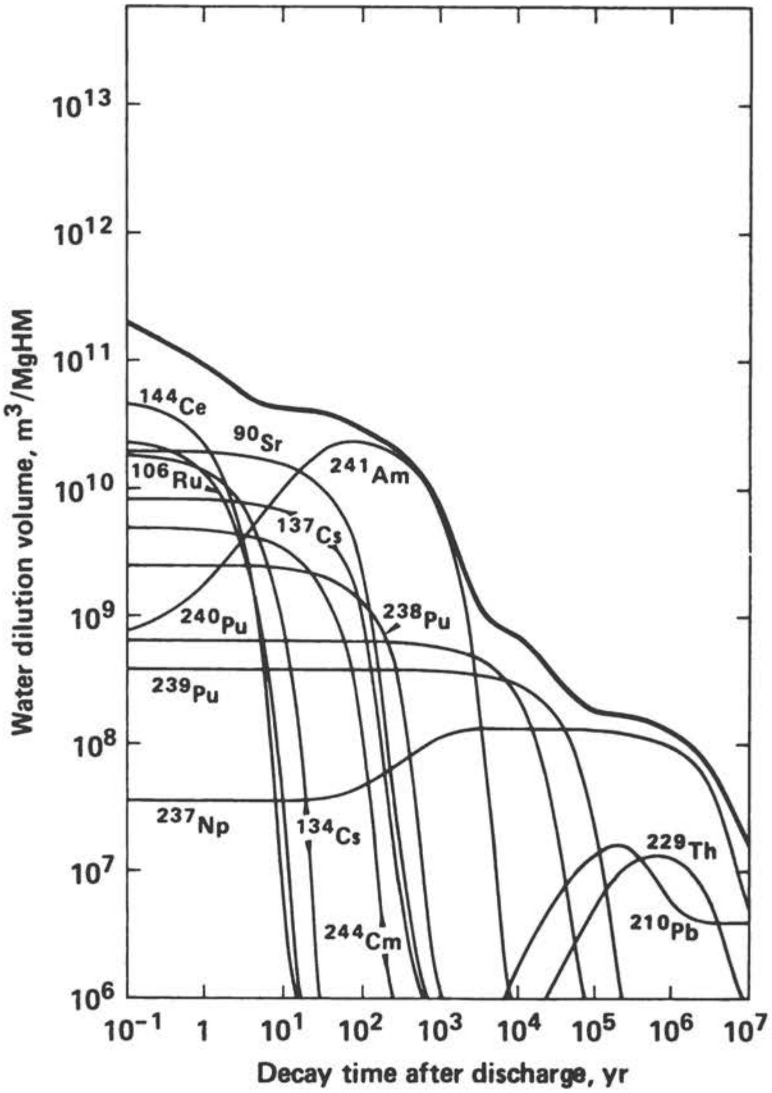

FIGURE 4-4 Water dilution volume of PWR spent fuel.

radium-226 that is present in uranium ores. The total WDV for spent fuel does not fall to the level of its parent ore until about 3 million years (rather than the approximately 7,000 years based on U.S. Nuclear Regulatory Commission (1982) values).

4.3.2. Projections of Spent Fuel to Be Discharged

Projections of spent fuel to be discharged from LWRs in the United States through the year 2000 are given in Table 4-5. The reference growth rate of nuclear power in the United States currently projects

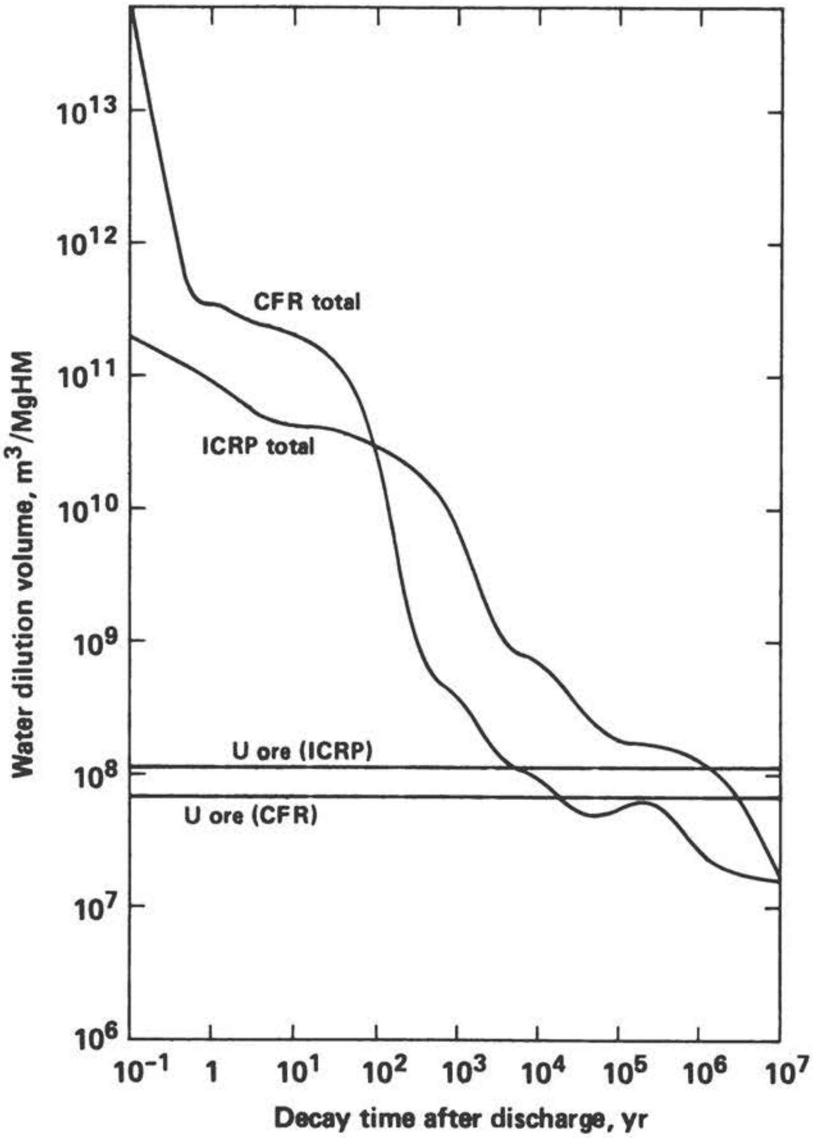

FIGURE 4-5 Water dilution volumes of PWR spent fuel and its parent uranium ore.

about 180 GWe of installed capacity in the year 2000. Two-thirds of this capacity will be PWRs and one-third BWRs. If no spent fuel is reprocessed, storage will be needed for about 150,000 BWR fuel assemblies and 100,000 PWR fuel assemblies in the year 2000, resulting in a total of 250,000 fuel elements containing about 72,000 Mg of uranium and representing about 1,800 GWe-yr of electricity. If this fuel were packaged for geologic disposal as one PWR assembly or two BWR assemblies in canisters about 0.3 m in diameter by 4.6 m high, about 178,000 canisters would be required. This is the approximate capacity of a conceptual repository.

TABLE 4-5 Projected Accumulation of Spent Fuel

	Calendar Year Ending			
Type of Fuel	1980	1985	1990	2000
Boiling water reactor				
Number of assemblies	16,000	35,000	65,000	150,000
Mg uranium	2,800	6,200	12,000	28,000
Pressurized water reactor				
Number of assemblies	9,100	22,000	46,000	100,000
Mg uranium	3,900	9,200	20,000	44,000
Total				
Number of assemblies	25,200	57,000	110,000	250,000
Mg uranium	6,700	15,800	32,000	72,000

NOTE: Based on installed nuclear capacities of 53.8, 137.5, and 179.7 GWe at the end of calendar years 1980, 1990, and 2000, respectively.

SOURCE: U.S. Department of Energy (1981).

4.4. HIGH-LEVEL WASTE

4.4.1. Commercial High-Level Waste

Although high-level waste is defined in 10 CFR 60 as "(1) irradiated reactor fuel, (2) liquid wastes resulting from the operation of the first cycle solvent extraction system, or equivalent, and the concentrated wastes from subsequent solvent extraction cycles, or equivalent, in a facility for reprocessing irradiated reactor fuels, and (3) solids into which such liquid wastes have been converted," the term as used in this chapter applies only to parts (2) and (3) of the definition. The composition of the high-level waste is strongly dependent on the details of the chemical and operating flowsheets of the reprocessing plant. As generated, it is a nitric acid solution of about 5 m^3/MgHM, typically containing more than 99 percent of the nonvolatile fission products, essentially none of the tritium and carbon-14, none of the noble gases, about 0.1 percent of the iodine, about 0.5 percent of the uranium and plutonium, and virtually all of the neptunium, americium, and curium in the spent fuel. Gadolinium may be added during reprocessing as a neutron poison, and the high-level waste from reprocessing LMFBR fuel may also contain about 0.7 percent of the activated stainless steel in the fuel assembly that was dissolved with the fuel. This waste is evaporated to a volume of about 0.4 m^3/MgHM and, under 10 CFR 50, Appendix F, may be stored for as long as 5 years before being converted to a solid form. Characteristics of typical high-level liquid waste from reprocessing PWR and LMFBR fuels are summarized in Tables A-1 and A-2 of Appendix A. The radionuclide compositions are calculated on the assumption that the fuels are reprocessed 160 days after discharge from LRWs and 90 days after discharge from LMFBRs. Actually, if commercial reprocessing is resumed as early as 1990, the LWR fuel will have been stored, on average, for about 15 years and an additional 10 to 20 years will elapse before short-decayed LWR fuel is routinely reprocessed.

The only commercial high-level waste presently on hand is that in storage at the NFS plant in West Valley, New York. Approximately 2,150 m^3 of this waste were neutralized and are stored as an alkaline supernate and precipitated sludge. In addition, 45 m^3 are stored as acid solution. The characteristics of these wastes are summarized in Tables A-3 and A-4 of Appendix A.

If the 29,400 m^3 of high-level liquid waste that are projected in Table 4-1 for the year 2000 were converted to a borosilicate glass and put into canisters 0.3 m in diameter by 3 m high, about 34,000 of these canisters would result and would represent the nominal capacity of a repository. Each canister would contain the high-level waste from reprocessing 2.1 MgHM and would typically have thermal characteristics as shown in Figure 4-2. The isotopes of major interest to repository risk analysis present in one such reference canister are characterized in Table 4-6. Details of the design and performance characteristics of these waste packages are discussed in Chapter 5.

The WDV of high-level waste from reprocessing PWR spent fuel based on ICRP-recommended values is given in Figure 4-6 (Croff 1981), and comparisons with its parent ore using both ICRP and 10 CFR 20 values are presented in Figure 4-7. Note that the use of the new ICRP values (see Section 4.3 above) increases the time required for the high-level waste

TABLE 4-6 Significant Radionuclides in One Canister of HLW

	Time After Reprocessing[a]					
	10 yr		100 yr		1,000 yr	
Nuclide	g	MBq	g	MBq	g	MBq
^{14}C[b]	3.4×10^{-2}	1.2×10^{2}	3.4×10^{-1}	1.2×10^{2}	2.9×10^{-1}	1.1×10^{2}
^{79}Se	1.2×10^{1}	3.2×10^{4}	1.2×10^{1}	3.2×10^{4}	1.2×10^{1}	3.1×10^{4}
^{90}Sr	8.8×10^{2}	4.4×10^{9}	1.0×10^{2}	5.2×10^{8}	~ 0	~ 0
^{99}Tc	1.6×10^{3}	1.0×10^{6}	1.6×10^{3}	1.0×10^{6}	1.6×10^{3}	1.0×10^{6}
^{126}Sn	5.8×10^{1}	6.0×10^{4}	5.8×10^{1}	6.0×10^{4}	5.7×10^{1}	6.0×10^{4}
^{129}I[b]	3.8×10^{-1}	2.5	3.8×10^{-1}	2.5	3.8×10^{-1}	2.5
^{135}Cs	2.4×10^{3}	2.7×10^{4}	2.4×10^{3}	2.7×10^{4}	2.4×10^{3}	2.7×10^{4}
^{137}Cs	2.0×10^{3}	6.4×10^{9}	2.5×10^{2}	8.0×10^{8}	~ 0	~ 0
^{226}Ra	2.9×10^{-7}	1.1×10^{-2}	3.1×10^{-6}	1.1×10^{-1}	1.6×10^{-4}	5.8
^{234}U	2.9	6.6×10^{2}	9.3	2.2×10^{3}	1.6×10^{1}	3.7×10^{3}
^{238}U	9.9×10^{3}	1.2×10^{2}	9.9×10^{3}	1.2×10^{2}	9.9×10^{3}	1.2×10^{2}
^{237}Np	9.3×10^{2}	2.4×10^{4}	9.5×10^{2}	2.5×10^{4}	1.0×10^{3}	2.7×10^{4}
^{238}Pu	1.3×10^{1}	8.1×10^{6}	6.4	4.1×10^{6}	1.4×10^{-2}	9.0×10^{3}
^{239}Pu	5.3×10^{1}	1.2×10^{5}	5.5×10^{1}	1.3×10^{5}	6.8×10^{1}	1.6×10^{5}
^{240}Pu	3.6×10^{1}	3.1×10^{5}	6.1×10^{1}	5.2×10^{5}	5.6×10^{1}	4.8×10^{5}
^{241}Pu	7.8	3.0×10^{7}	1.0×10^{-1}	3.9×10^{5}	3.9×10^{-4}	1.5×10^{3}
^{242}Pu	4.8	6.8×10^{2}	4.9	6.9×10^{2}	5.0	7.1×10^{2}
^{241}Am	1.2×10^{2}	1.5×10^{7}	1.1×10^{2}	1.4×10^{7}	2.6×10^{1}	3.3×10^{6}
^{243}Am	1.8×10^{2}	1.3×10^{6}	1.8×10^{2}	1.3×10^{6}	1.6×10^{2}	1.2×10^{6}
^{245}Cm	2.5×10^{-1}	1.6×10^{3}	2.5×10^{-1}	1.6×10^{3}	2.3×10^{-1}	1.5×10^{3}
^{246}Cm	2.0×10^{-1}	2.2×10^{3}	1.9×10^{-1}	2.2×10^{3}	1.7×10^{-1}	1.9×10^{3}

NOTE: The canister is 0.3 m in diameter by 3 m high and contains the solidified waste from reprocessing 2.1 MgHM of PWR spent fuel.

[a] The fuel is reprocessed 160 days after discharge from the reactor.

[b] The high-level waste contains 0.1 percent of the carbon and iodine in the spent fuel.

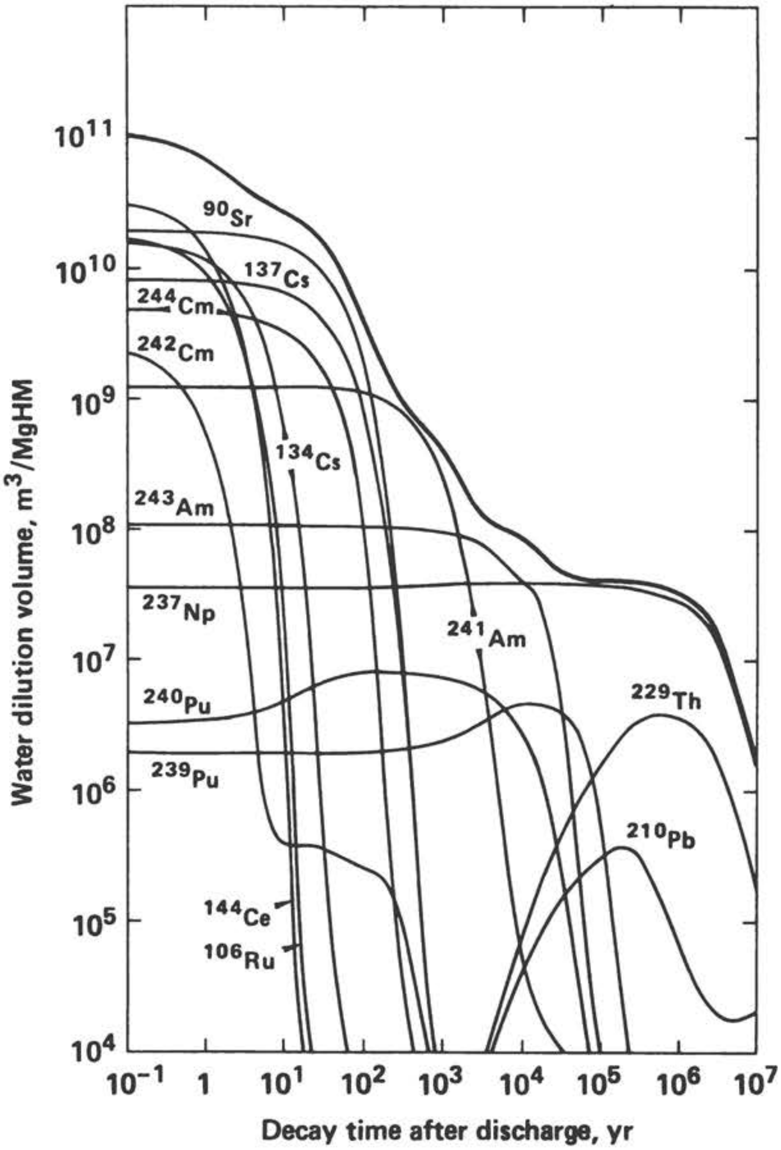

FIGURE 4-6 Water dilution volume of PWR high-level waste.

and ore to reach equal toxicities from about 400 years to about 20,000 years. Based on these new ICRP values, the effect of reprocessing is to reduce by two orders of magnitude the time at which the water dilution volume of the waste equals that of the ore.

Examination of data similar to those of Figures 4-6 and 4-7 suggests that the long-term (1,000 years) hazard of radioactive waste placed in a geologic repository could be reduced by separating the most significant long-lived radionuclides and transmuting them to stable products by bombardment with neutrons. A comprehensive study of this approach (Croff et al. 1980) concluded that, while the concept was technically

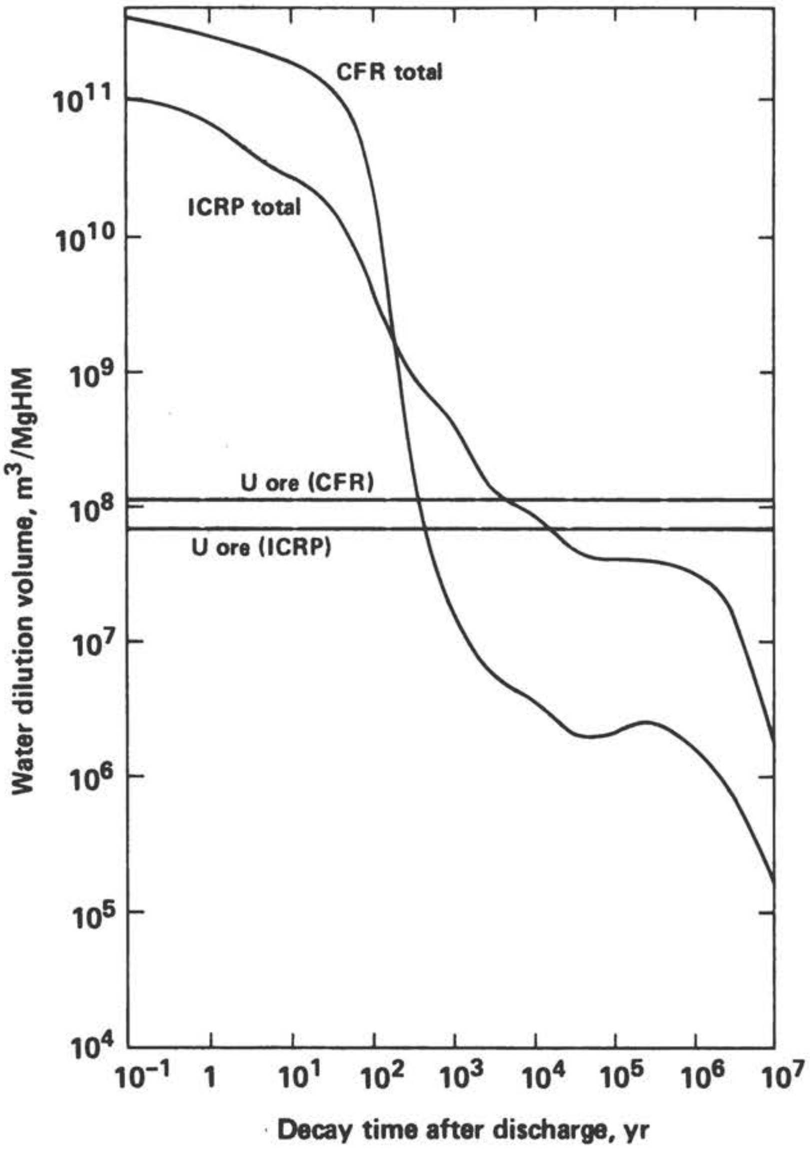

FIGURE 4-7 Water dilution volumes of PWR high-level waste and its parent uranium ore.

feasible, an analysis of its costs, risks, and benefits showed that it was not cost effective. The cost of separating and transmuting the actinide elements from fuel cycle wastes was estimated to be $9.2 million/GWe-yr. The short-term radiological risk was increased by 0.003 (health effect)/GWe-yr, and the expected long-term benefit (i.e., incremental risk reduction from a repository) was found to be 0.06 (health effect)/GWe-yr integrated over 1 million years. The latter is only about 0.001 percent of the health effects expected from natural background radiation, and the cost is $153 million per health effect saved.

4.4.2. Defense High-Level Waste

Defense and commercial high-level wastes are similar in that both are residues of aqueous-organic solvent extraction processes and contain the same spectrum of fission products and major actinide elements. Nevertheless, differences in the nature of the fuels as well as in reprocessing plant operating practices have resulted in some important dissimilarities between the high-level liquid waste now on hand and that expected from future reprocessing of commercial fuel. Defense fuels have much lower radiation exposures than commercial fuels, and hence the waste contains lower concentrations of fission products and transuranic elements. Defense waste also contains substantial amounts of inert chemicals (e.g., sodium, aluminum, zirconium, and iron) and typically has been neutralized for storage in carbon steel tanks, while commercial high-level waste will be a relatively pure solution of fission products and actinide elements in HNO_3. Thus, solidificaton of defense high-level waste will result in solids having lower concentrations of radionuclides and heat generation rates one to two orders of magnitude less than those typical of commercial waste.

Defense high-level wastes at Hanford, the Savannah River Plant, and the Idaho Chemical Processing Plant differ because of differences in characteristics of the fuels that have been reprocessed at the respective sites and in plant operating practices over the past 20 to 40 years. Using Savannah River as an example, 117,000 m^3 of high-level waste are projected to be on hand at the end of the year 2000 in the form of liquid, salt cake, and sludge (U.S. Department of Energy 1981). If this were converted into a borosilicate glass using a presently anticipated process, it could be encapsulated in about 10,000 canisters 0.61 m in diameter by 3 m high. A typical canister would have a heat generation rate of 270 W and contain 73 g of plutonium.

4.5. CLADDING WASTE

Cladding waste consists of the zircaloy cladding, from which the fuel has been dissolved, and the other fuel assembly structural materials such as stainless steel and Inconel. It is sheared into 5- to 8-cm-long fragments and is highly radioactive as a result of the activation products and residual fuel materials that are present. The uncompacted volumes (assuming 80 percent voids) are about 0.18 and 0.38 m^3/Mg of PWR and BWR fuel reprocessed, and 0.85 m^3/Mg of LMFBR fuel. This waste is generated at the same time as the high-level waste, i.e., 160 days after fuel discharge for LWRs and 90 days after discharge for LMFBRs. The primary constituent is the activated cladding and structural metals (zircaloy, etc., for LWRs and stainless steel for LMFBRs), but it may also contain 0.05 percent of the nonvolatile fission products and actinides that remain after dissolution of the oxide fuels and perhaps 30 percent of the tritium that is captured in the zircaloy cladding of LWR fuel. Characteristics of cladding wastes are given in Tables A-5 and A-6 of Appendix A.

It is possible that this waste may be packaged without additional treatment such as compaction or melting. If so, it would likely be packaged in canisters similar to those proposed for defense high-level waste. A canister 0.61 m in diameter by 3 m high would contain the cladding from about 5 MgHM of PWR fuel or 2.3 MgHM of BWR fuel. About 21,000 of these canisters would result from reprocessing the 72,000 MgHM that are projected for the year 2000. A typical canister would contain 1,400 kg of cladding waste with 1.7 kg (5×10^6 megabecquerels, where 1 becquerel (Bq) = 2.7×10^{-11} curie (Ci)) of actinide elements and would generate 120 W of thermal power.

4.6. TRANSURANIC WASTE

Transuranic waste, as defined by the U.S. Department of Energy (1982), is material that is contaminated with alpha-emitting radionuclides of half-lives greater than 20 years to a level greater than 100 nCi/g or 3,700 Bq/g. The ingrowth of TRU daughter products must be factored into the calculation of this control value. Other isotopes of uranium and plutonium, as well as isotopes of americium and curium, can also contribute to the alpha activity in TRU wastes. TRU waste is generated at reprocessing plants and at mixed uranium-plutonium dioxide fuel fabrication plants. It consists of a wide assortment of solid materials, including items made of cellulosics, ceramics, and metals as well as salts and sludges that arise in the treatment of liquid waste streams and filters from cleanup of off-gas. The transuranium element content ranges from trace amounts to as much as 100 g/m^3 and averages about 10 g/m^3. The densities of the uncompacted wastes vary from about 0.03 g/ml to as much as 3 g/ml. From one-half to two-thirds of this waste (by volume) is combustible and can be reduced via incineration by factors of about 50 and 20 in volume and weight, respectively. About one-half to three-fourths of the waste (by volume) can be reduced in volume by factors of 2 to 20 through compaction. Several cubic meters are generated per metric ton of heavy metal reprocessed or refabricated, about a third of which must be handled remotely because of fission-product contamination or external radiation from some of the TRU isotopes.

It is expected that, after volume reduction and/or fixation, TRU waste will be packaged in a variety of containers such as 55-gal (0.21 m^3) steel drums and rectangular steel boxes as large as 2.2 m long by 1.4 m wide by 1.4 m high. A cubic meter of this waste might typically contain about 100 g of TRU elements (mostly plutonium) and have a heat generation rate of about 1 W. Properties of 1 g of plutonium typical of that expected in commercial TRU waste are given in Table A-7 of Appendix A.

4.7. IODINE

Iodine is a semivolatile fission product that, because of its complex chemical properties and its high biological significance, has always required special attention to ensure adequate safety in its management. About 0.01 g of iodine isotopes is formed per megawatt(thermal)-day, and the isotope of greatest concern during reactor operation and fuel reprocessing is 8.05-d iodine-131. However, the species of consequence to longer-term waste management is 1.6×10^7-yr iodine-129, which comprises about 75 percent of the weight of the fission-product iodine in spent fuel at the time of reprocessing.

Research and development work aimed at reducing iodine releases from fuel reprocessing plants shows promise of removing at least 99.9 percent of the iodine from the other fuel constituents by volatilization at the head-end of the process and then trapping iodine by absorption in aqueous nitric acid solutions and/or sorption on zeolites. The final form into which the iodine may be processed for packaging, shipment, and disposal has not been defined. There is no known method to immobilize iodine-129 for a substantial part of its half-life; hence, the objective must be to prepare a material with an iodine-129 release rate sufficiently low as to not give rise to a radiological hazard after disposal. Among the immobilization forms considered, 10 percent $Ba(IO_3)_2$ in concrete has been the most extensively studied (Clark et al. 1975, Rogers et al. 1980). One cubic meter of this composite would contain about 72 kg (4.7×10^5 MBq) of iodine-129 derived from 400 MgHM of spent fuel.

4.8. CARBON-14

Carbon-14 (with a half-life of 5,730 years) is produced in oxide-fueled reactors principally by an (n,p) reaction with nitrogen-14 impurity in the fuels, but also as the product of an (n,α) reaction with oxygen-17. It will be released as CO_2 during nitric acid dissolution of the fuel and can be separated from the off-gases by reaction with either $Ca(OH)_2$ or $Ba(OH)_2 \cdot 8H_2O$ (Haag 1981). Although values of nitrogen impurity ranging from 1 to 100 ppm have been observed in oxide fuels, the typical concentration is 25 ppm. This would result in about 2×10^4 MBq of carbon-14 per MgHM in spent oxide fuels from LWRs and LMFBRs. In addition, approximately 3.7×10^4 MBq of carbon-14 would be present in the cladding wastes from these reactors (Tables A-5 and A-6).

The carbon-14 in the oxide fuels would exist as either CO_2 or low molecular weight hydrocarbons that could be oxidized to CO_2 for fixation. About 4 liters of $BaCO_3$ would be produced per MgHM reprocessed, and although this material appears to be suitable for mixing with cement, no studies are known to have been made to determine performance data of such mixtures.

4.9. KRYPTON AND TRITIUM

The noble-gas fission products consist principally of stable and short-lived isotopes of krypton and xenon. Typically, from 4 to 6 kg are released during the reprocessing of each MgHM from LWRs and LMFBRs, and while xenon comprises about 90 percent of the weight of the mixture, the only significant radioisotope remaining after 150 days' decay is 10.8-yr krypton-85 (about 3×10^8 MBq/MgHM). The U.S. Environmental

Protection Agency (1976) standard 40 CFR 190 will require recovery of krypton-85 from commercial power fuels irradiated after 1982. Methods for removal of the noble gases by adsorption in fluorocarbons and sorption on zeolites are under development, and final waste forms being studied are zeolites and ion implantation on nickel-lanthanum alloys. Packaging requirements have not been specified.

Between 400 and 1,300 Ci of tritium (with a half-life of 12.33 years) are generated per megagram of spent fuel. From 20 to 40 percent of this material is associated with the zircaloy cladding in LWR fuels, whereas greater than 90 percent will diffuse through the stainless steel cladding of LMFBR fuels during reactor operation. Methods for its separation and retention are being studied, but it is too early to define the nature of its final form.

It seems likely that, because of their relatively short half-lives, stabilized forms of krypton-85 and tritium will be stored at shallow to intermediate depths in dry wells rather than in deep geologic repositories, as is being considered for the other wastes described above.

4.10. THREE MILE ISLAND RESIDUES

The March 28, 1979, accident at the Three Mile Island Nuclear Station, Unit 2, created an as yet undefined quantity of material that will probably be sent to a geologic repository. These residues have not all been identified, much less characterized; however, the fuel and ion exchange resins from the submerged demineralizer system are prime candidates. The reactor core consists of 177 PWR fuel assemblies containing 82 Mg of uranium. The total decay power and radioactivity of this fuel 2-1/2 years after the accident have been calculated to be 0.04 MW and 4×10^{11} MBq (England and Wilson 1980). There are also about 3,000 m^3 of containment building water containing 1.8×10^{10} MBq of cesium-137 and 5.5×10^8 MBq of strontium-90 that is being processed through zeolite beds for sorption of the radioisotopes. It is expected that 8 to 12 canisters, each containing 0.23 m^3 of zeolite and 2×10^9 MBq of cesium and strontium, will result. Finally, 10 to 12 filter units contaminated with TRU elements have been tentatively identified for disposal in a repository.

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5

THE WASTE PACKAGE

5.1. INTRODUCTION

5.1.1. Definition of Waste Package

The waste package refers to the material, including radioactive waste, inert components, and their containers, that is to be placed deep in the ground for the purpose of effective isolation of radiation from the human environment. The object of the design of a complete waste package and its associated repository is the achievement of an acceptably small release of radioactive substances from the total system. Waste forms are combinations of radioactive waste materials with inert solids, such as glass, ceramics, or concretes. Waste forms are encased in metal canisters, and in some waste packages an additional metal envelope, called the overpack, protects the canister from exposure to groundwater while the repository is hot.

5.1.2. Role of the Waste Package

The waste package or components of it may be needed for several functions.

Above-Ground Storage

The waste form and its surrounding canisters may be kept in above-ground storage for several years or decades, depending on the age of the waste at the time it is formed and encased in the canister and on the heat load requirements of the repository design. There are a number of waste forms discussed in this chapter that meet the requirements of surface storage and cooling. Reasonable physical integrity of the form is necessary so that fine particulates are not formed that could be dispersed if the canister were breached. The forms considered in Section 5.4 meet this criterion.

Transport and Emplacement

Similar demands are placed on the waste form and canister for transporting the waste from the reprocessing unit to the surface storage facility and subsequently to the repository site for assembling the complete waste package for permanent disposal. At this time of emplacement, the waste form and canister are combined with other components to form a waste package. The properties of several candidate waste packages are such that we have high confidence that the assembly and emplacement can be effected safely.

Isolation of Radionuclides from the Biosphere During the Thermal Period

The waste package may be required to limit the release of soluble radionuclides, mainly fission products, to groundwater during the several-hundred-year period required for these nuclides to decay to acceptable levels of radioactivity. If the environmental temperature is kept sufficiently low, below, say, 100°C, borosilicate glass can adequately limit release. The Nuclear Regulatory Commission (NRC) has proposed that further security be attained through a corrosion-resistant overpack with a design life of about 1,000 years or more under repository conditions. Protection against release of shorter-lived, high-solubility radionuclides is provided through viable repository designs for which groundwater travel times to the biosphere exceed 1,000 years.

Longer-Term Isolation of Radionuclides from the Biosphere

We do not have assurances that the waste package will effectively prevent the release of long-lived radionuclides at times exceeding 1,000 years. However, the waste package need not perform this function if other components of the waste-isolation system can be relied on for containment. First, it is shown in Chapter 9 that our best assessment of the solubility of long-lived radionuclides is sufficiently low to limit release to acceptable levels. Second, repository sites can be selected with water transport properties such that release will be limited to acceptable levels. And third, repository sites can be selected such that their groundwater discharge will provide adequate dilution to keep individual-dose exposures within acceptable limits.

Retrieval of Waste from a Repository

While waste packages can be designed to facilitate retrieval of waste from a repository if postemplacement data indicate retrieval is necessary, this additional requirement is costly and may detract from more important design features. It is not presently an NRC requirement, and we recommend against retrievability as a waste-package design feature.

Overall Functions

The waste package may provide serendipitous benefits, but these should not be the criteria by which this system element is evaluated. Thus, long-term limiting of radionuclide release, after 1,000 years, and retrievability are desirable but not necessary waste-package features if we can depend on geologic isolation, low solubility, and limited transport for long-term containment. However, a waste package qualitatively better than guaranteed by any of the existing packages could give a level of confidence in the waste package such that this of itself would provide adequate long-term isolation of radionuclides. This would make available many more sites for disposal, with less demanding site requirements. The panel recommends waste packages containing borosilicate glass as the first choice for further testing and for repository planning, and continued backup studies on waste packages containing other waste forms are also recommended.

5.1.3. Chapter Overview

This chapter includes a description of different waste forms that have been proposed, i.e., combinations of radioactive materials with inert solids such as glass. This is followed by an analysis of the data and theories that are available for estimating rates of leakage of radioactivity into surrounding rock after many years. An overall assessment of borosilicate glass appears in Section 5.8.

The choice of metals for the canister and overpack is discussed in Section 5.9. Since the waste package is required to achieve a safe low level of radioactivity added to the environment over many years, the chapter includes a short summary of proposed criteria for waste-package performance. The current waste-package designs are reviewed in Section 5.10, followed by a review of the proposed functions and expected performance of the backfill material that may constitute the outer layer of the emplaced waste package. Forms and packages for transuranic wastes and for separated radionuclides are reviewed in Sections 5.12 and 5.13.

5.1.4. General Description of Waste Package

The waste package includes the solid waste form that contains the radioactive material, either as a mixture, a solid solution, or a chemical compound, and a canister containing the solidified waste. In packages for heat-generating wastes, the canister may be surrounded by an overpack, a corrosion-resistant barrier to delay exposing the waste form to groundwater during the period of repository heating. In some designs the overpack is thick enough to provide shielding from gamma radiation emitted from the waste form, thereby simplifying placement of waste packages in a repository. The space between a waste package and the surrounding host rock of a repository is expected to be filled with a backfill material. This may consist of highly compressed pieces of bentonite or other clay materials that expand when wet with groundwater to form a low-permeability envelope around the inner components of the waste package. In a salt repository, crushed salt may suffice as a backfill. The waste package may also include internal structural supports to protect the contents from possible crushing forces from the surrounding rock and groundwater.

Waste-package designs vary with the waste to be contained. Waste packages considered in this study are for the following different wastes:

o high-level waste from fuel reprocessing
o unreprocessed spent fuel
o transuranic waste
o radioactive gaseous wastes (carbon-14, iodine-129, tritium, krypton-85)

Possible wastes from decommissioning of reactors and fuel cycle facilities are not considered in this study because they will be predominantly low-level waste. Those that are not low-level waste will fall within one of the categories listed above.

5.2. FUNCTIONAL AND PERFORMANCE CRITERIA FOR WASTE PACKAGES

5.2.1. Criteria by Federal Agencies and Contractors

The expectations of the Office of Nuclear Waste Isolation (ONWI 1981b) as to the function and performance of the possible waste-package components are given in Table 5-1. A possible configuration of a waste package with all of these components is shown in Figure 5-1. In earlier

TABLE 5-1 Function and Performance Expectations of Waste-Package Components

Component	Function and Performance Expectation
Waste form	Immobilize the radioactive materials; minimize adverse interactions with other package components; retain radiolytic gases produced within the waste form; resist effects of mechanical impacts during handling; maintain chemical and physical stability in terms of radioisotope retention over long time period
Stabilizer	Maintain the physical and chemical stability of the spent fuel waste form; enhance heat transfer; provide structural support; minimize adverse interactions with other package component; control criticality
Canister	Serve as primary container of the waste form during interim storage and transportation; resist mechanical, thermal, and radiological impact; limit the release of radioactive gases or particulates from the waste form
Overpack	Serve as a high-integrity protective, physical barrier between the canistered waste form and groundwater throughout operations, retrieval, and long-term isolation
Sleeve[a]	Facilitate the removal of the waste canister/overpack from the emplacement hole; protect the canister/overpack from rupture by host rock creep or backfill expansion
Backfill[b]	Prevent flowing groundwater from contacting the waste canister/overpack; provide sorption capacity for radionuclides contained within the waste; contain chemical agents to minimize chemical attack on the canister/overpack and decrease radionuclide solubility; serve as a plastic stress-adjustment medium

[a] The sleeve incorporated in earlier waste packages was designed to facilitate retrieval.

[b] The original ONWI description has been modified as suggested by J. Kircher (Office of Nuclear Waste Isolation, personal communication, 1982).

SOURCE: Office of Nuclear Waste Isolation (1981b).

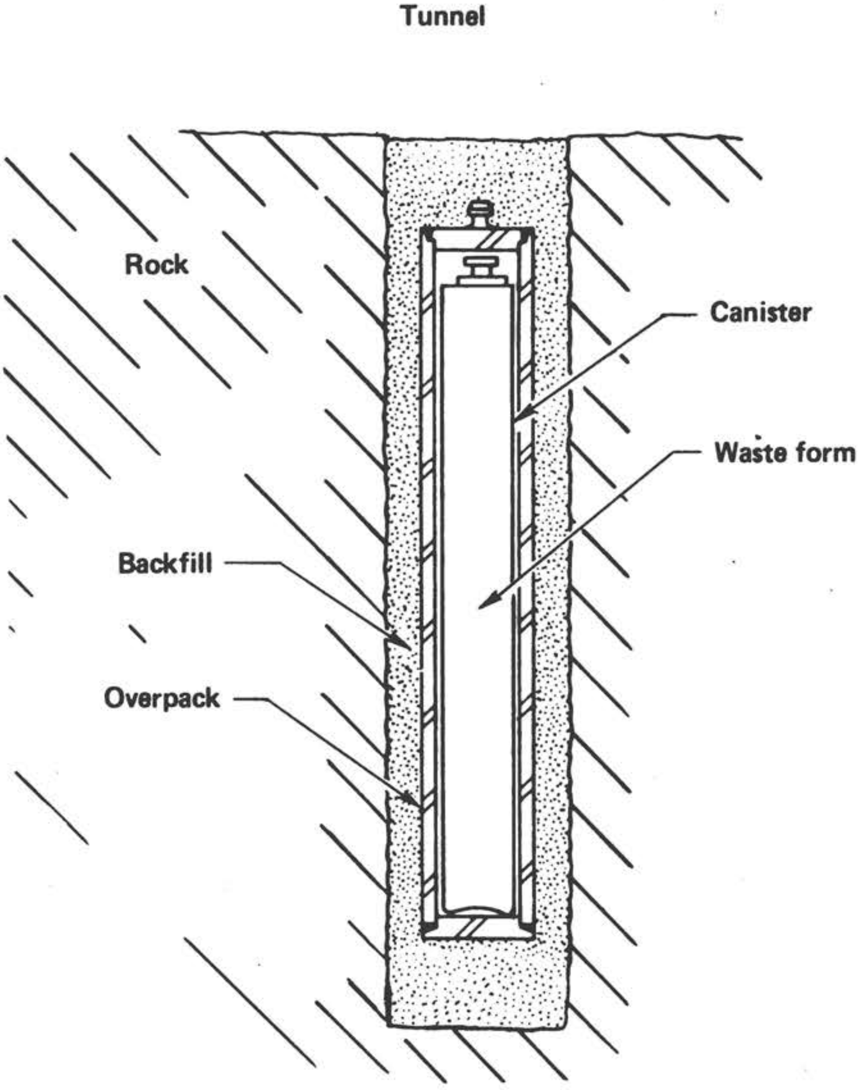


FIGURE 5-1 Waste package schematic.

designs the sleeve was added to facilitate retrievability, but this function is not found in current designs. For wastes that do not generate appreciable decay heat, some of the components such as the overpack may be eliminated.

The functional requirements in Table 5-1 were selected by ONWI to meet tentative performance criteria specified by the U.S. Nuclear Regulatory Commission (1982) in its proposed regulations 10 CFR 60. The NRC staff's proposed numerical criteria (U.S. Nuclear Regulatory Commission 1982) for the waste package are as follows:

o No release of radionuclides from the waste package for 1,000 years after the repository is sealed.

o A fractional release rate of less than 10^{-5}/yr of the radionuclide inventory within the waste package after 1,000 years, but excluding any radionuclide released at a rate less than 0.1 percent of the calculated total annual release at 1,000 years.

In this chapter we are concerned with the state of technology of the waste package and its components and with the predicted performance of the waste package under repository conditions. Rather than adopt NRC's proposed numerical criteria as objectives for waste-package performance, we have evaluated the performance that can reasonably be expected from the waste-package technologies under development. In Chapter 9 we analyze the overall performance of geologic repositories containing such waste packages. It is the overall performance criterion of 10^{-4} Sv/yr dose to the individual, adopted for this study (Chapter 8) that is the measure of the suitability of the waste-package technology in conjunction with the rest of the waste-isolation system.

The long-term performance of the geologic repository is the emphasis of this study. The waste-package components that can contribute most clearly to long-term performance and for which information is sufficiently available for evaluation are the waste form, the overpack, and the backfill. These are considered individually in succeeding parts of this chapter, followed by a summary of the state of the technology of overall waste-package design.

5.2.2. System Requirements for the Waste Package and Components

Repositories are being designed for high-level wastes that have been aged for 10 years or more since the spent fuel was discharged from reactors. Early emplacements in a geologic repository will likely be derived from the large accumulation of spent fuel already aged for several decades, and less temperature-induced stresses will be placed on these early waste packages. When reprocessing becomes available, and after the backlog of aged spent fuel has been reprocessed, the high-level waste may be formed and stored in canisters as early as 5 or 6 months after discharge from the reactor. During subsequent above-ground storage for 10 or more years, it is important that the canister not deteriorate and allow contained radionuclides to escape. Additional above-ground storage may be required if it is found necessary to reduce the heat generation rate as one of the means of lowering the repository temperature, thereby placing further demands on continued integrity of the waste canister.

It is likely that the waste package will not be assembled until it is ready to be loaded into the repository and that the backfill will not be added until the package is emplaced in the host rock. After emplacing high-level waste packages, the temperatures of the waste and the surrounding rock rise rapidly, as shown in Chapter 6 for the 10-year-old wastes. The near-rock temperature peaks in about 10 years at values well above the boiling point of water. In a repository below the water table and in rock normally saturated with groundwater, such as basalt, granite, and saturated tuff, groundwater will slowly seep back into the repository. In the basalt repository a section of the repository that has been filled with waste, backfilled, and sealed is estimated to resaturate with groundwater within about a decade (D. J. Brown, Rockwell Hanford Operations, personal communication to T. H. Pigford, 1982) and a repository in tuff within perhaps 20 years. Boiling during the process of resaturation can be expected.

When the first liquid water contacts portions of the multicomponent waste package, the package will be locally cooled, and considerable thermal stresses are likely to develop. The transition between dry storage and resaturation may be the period of most severe stress on the waste package and its components. Following resaturation there will be a period of readjustment of the groundwater chemistry, from the air-exposed oxidizing conditions to the more reducing conditions characteristic of the normal geochemistry of the deep groundwater. The waste package must survive this chemical transition. Waste packages emplaced in salt or unsaturated tuff will probably not be subjected to these resaturation-induced stresses.

It is more important that waste packages survive stresses intact if they are designed with overpacks to prevent leakage of groundwater to the waste canister.

The retrievability requirement proposed by NRC in 1981 does not appear in NRC's latest proposed criteria for waste packages. The earlier designs of retrievable packages by DOE contractors have given way to plans for backfilling emplacement enclosures soon after waste has been emplaced, with the expectation that retrievability can be accomplished if necessary by remining the emplacement area.

5.3. FACTORS AFFECTING WASTE-FORM PERFORMANCE

5.3.1. Movement of Radionuclides to the Waste-Form Surface

To be brought into contact with leachant, radionuclides must either be exposed as the waste form dissolves or they must diffuse to a surface of the waste form. The first mechanism is important for any waste form that dissolves more rapidly than the majority of the long-lived waste products. Because bulk diffusion in glassy and crystalline ceramics is very slow, a waste form of very low solubility could, in principle, limit release rates to levels well below the performance required in our study. A possible problem, not yet addressed by experiments, is that bulk diffusion in ceramics can be increased by several orders of magnitude if there are defective atomic lattices--e.g., anion vacancies, which are almost certain to exist or to be created by alpha damage. In ceramics with very small grains, increased diffusion along grain boundaries may control the release rate.

Surface diffusion is a still more rapid phenomenon that allows diffusing material to move along surfaces of grains and cracks. This has not been studied as a mechanism for the transport of radionuclides to outer surfaces but would be effective in proportion to the degree of internal cracking.

5.3.2. Entry of Radionuclides into Groundwater

At or near the external waste-form surface the radionuclide will enter the groundwater by one of several mechanisms. The rate constants for release and dissolution can be interpreted as the fractional release rate, i.e., the fraction of the waste-package inventory of a given constituent that is released from the waste form per unit time. The fractional dissolution rate is expressed similarly in terms of the rate of dissolution. For congruent dissolution, i.e., if all constituents within a waste form dissolve when the waste-form matrix dissolves, the fractional dissolution rates are equal for all constituents.

The radionuclides exposed to groundwater when the waste matrix dissolves or restructures do not necessarily dissolve, because precipitates of low-solubility constituents can form on the surface of the waste form. For these low-solubility species the release from the waste form may be congruent, but their dissolution in the leachant is not. When immobilized precipitates form, the low-solubility species enter the groundwater at lower fractional rates than the waste matrix.

The process by which released radionuclides reach the groundwater outside the waste package is more complicated when the outer layers of the waste package are first penetrated by groundwater, through leaks and cracks. However, as is shown in Chapter 9, a more important consideration may be the longer-term exposure to groundwater, after the outer metallic layers of the waste package may be presumed to have completely failed. This allows a simpler identification of the mechanisms by which radionuclides in the liquid at the surface of the waste form undergo mass transfer into the slowly moving groundwater in the rock. If a backfill is present, it can be assumed in the present discussion to have a porosity and groundwater motion similar to that of the surrounding rock. The effects of different backfill properties on mass transfer are discussed in Section 5.7.4.

Dissolved radionuclides transport through the liquid at the waste-form surface, or at the inner surface of the backfill, by molecular diffusion through the liquid-filled pores. As the dissolved radionuclides progress outward, they are carried away by the convective flow of groundwater.

Polymers and colloids can form in liquids containing low-solubility materials, especially some of the actinides such as plutonium (Benedict et al. 1981). The presence of colloids containing radionuclides provides a means for greater concentrations of radionuclides in the liquid than could occur from true solution. These colloids can undergo slow diffusion into the groundwater, and it is conceivable that they could be abraded from the solid surface by shear forces from groundwater motion. Radionuclides could also enter the liquid as particulates if the precipitate layer does not adhere to the waste-form surface and is carried by the moving groundwater. Data on the formation of colloids in laboratory tests with borosilicate glass are discussed in Section 5.6.7. The effect of these release mechanisms on the rate of release of radionuclides to groundwater from borosilicate glass is discussed in more detail in Section 5.7.4.

Not only can colloids affect the rate at which radionuclides enter the moving groundwater, but they can also affect migration through the geologic media. The precipitation (McVay and Buckwalter 1982), sorption (Allard 1982a,b), trapping (Avogadro et al. 1981), and convection (Eichholz and Craft 1980) of radionuclides in colloids must be considered along with the advective transport of dissolved radionuclides (cf. Section 5.6.7).

5.4. WASTE FORMS FOR HIGH-LEVEL REPROCESSING WASTE

5.4.1. Summary of High-Level Waste Forms

A number of materials for high-level waste from fuel reprocessing have been proposed, made, and evaluated. These are shown in Table 5-2. By far the largest effort has been given to borosilicate glass, which incorporates various radionuclides into its molecular structure as the glass is melted with fission-product and actinide oxides. Waste elements can become part of the crystal structures of various titanates, zirconates, or other host ceramic crystalline materials by sintering mixtures of their fine powders or dried sols. Either glass or crystalline ceramics can be made in large shapes with low surface-to-volume ratios or mixed as powders or lumps in various matrix materials, such as metals or cements.

At the request of the Department of Energy, a committee of materials scientists gave a mid-1981 ranking of alternative waste forms for defense wastes based on least risk, engineering practicality, and leach tests carried out for 28 days at 90°C (Alternative Waste Form Peer Review Panel 1981; cf. Section 5.5.1 and Tables 5-4 and 5-5). Borosilicate glass ranked first, although the committee noted that differences in overall leaching performance were small.

It is premature to select waste-form materials on the basis of such rankings. The effects of higher temperatures and the effects of realistic repository conditions could alter the rankings. Further, a number of the alternative waste-form materials have had little study. Later, in Sections 5.6 and 5.7, we point out that for most of the important long-lived radionuclides the laboratory leach data that have been used in these rankings have little relevance to the release of these radionuclides in a geologic repository.

5.4.2. Waste-Form Manufacture

The reliability, safety, and cost of process and facilities for manufacturing waste forms are important. Isolation of radionuclides from the environment must include controlling environmental releases during processing as well as during the eventual decommissioning of the processing plants. There is little or no experience in long-term operation of high-temperature processes for radioactive materials, such as glass melting or firing waste ceramics, wherein the environmental release of radionuclides must be kept near zero. Shorter-term successful experience on borosilicate-glass melters at full-production

TABLE 5-2 Waste Forms for High-Level Waste

Waste-Form Material	Shapes	Sizes	Some Advantages	Some Disadvantages
Borosilicate glass, melted	Large monoliths	Up to ~½ m dia × 3 m length	Most experience; R&D; can dissolve most of the waste	Metastable material; attacked in hydrothermal conditions
	Marble	~ few cm		
Sintered porous high-silica glass and inert tube collapsed around form	Large monoliths, discs, and other smaller shapes	Up to ~½ m dia × 3 m length	Outer inert shell; less soluble glass	More complex process
Concrete (FUETAP); other cement materials	Large castings	~m	Lower-temperature process; cements have most field experience	Poorer leach resistance at low temperatures
Calcines; sintered waste	Pellets	~½ to 1 cm	Simple; intermediate-temperature process	Poorer leach resistance
Clay ceramics; sintered clay	Extruded rods, bricks	~ tens of cm	Simple; intermediate-temperature process	Poorer leach resistance
SYNROC (primarily titanates); hot pressed, sintered, tailored ceramics	Large shapes, rods, discs, pellets	~ tens of cm	Probably greater leach resistance at higher temperature	Higher-temperature process; more complex
Glass ceramic	Large monoliths	Up to ~½ m dia × 3 m length	Potential for a more stable-form material	Relatively little known; possible soluble phases
Sol-gel ceramics, sintered	Particles, small spheres	~mm	Lower temperature; simpler process	Relatively little R&D; higher surface area form shapes
Metal matrix; inert metals containing marbles, pellets, or powders	Large castings	~m	Convenient package; can use small-form shapes	When particulates are exposed, higher surface area; less waste in form
Concrete or ceramic matrices	Large castings	~m	Convenient package; can use small-form shapes	When particulates are exposed, higher surface area; less waste in form
Multibarrier materials PyC, SiC, Al_2O_3 coatings Metal coatings	Particles	~mm	Probably lowest release rate; early resistance to leaching	Complex processes; higher temperatures

SOURCE: General literature reading by J. R. Johnson.

scale has been obtained with simulated waste and with radioactive waste both in the United States and abroad.

Although some test specimens of metal-matrix waste forms have been fabricated, further programs for backup development of alternative waste forms should consider the incorporation of waste-oxide fines in selected metals using powder metallurgy techniques. Such processing may be no more complex than that of pouring molten glass into metal canisters.

5.4.3. Physical Properties

Some typical physical properties are shown in Table 5-3 for integral samples of waste-form materials. Full-scale waste forms will likely be cracked and otherwise flawed, and the physical properties will be correspondingly downgraded.

TABLE 5-3 Some Typical Physical Properties of Various Waste-Form Materials

Waste Form	Waste Content (wt%)	Density (g/cm^3)	Tensile Strength (MPa)	Compressive Strength (MPa)	Thermal Conductivity (W/m-°C)	Linear Thermal Expansion (10^{-5}/°C)
Borosilicate glass	29	3.0	50	250	~1	~1
High-silica glass	30	2.8	[50]	[200]	[~1]	~0.5
Ceramic titanates (SYNROC)	20	4.35	75	800	~3	~1
Tailored ceramics	65	4.4	[70]	[500]	[2-3]	[~1]
Concrete (FUETAP)	20	2	–	30	~0.6	~1

NOTE: Data are for integral samples. Brackets indicate estimates where no data were found.

5.4.4. Homogeneity and Integrity

The homogeneity and integrity of the waste form will depend on the materials and the processes by which it is made. Assurance of homogeneity of the waste form is important to minimize the presence of more soluble phases in the waste form. Crystalline phases within the host material may provide regions of internal stress. Large glass forms may devitrify to the extent of a few weight percent near the surface and more than 10 percent in the center. Crystalline ceramic forms are found to have accommodated waste elements into their stable phases according to the process time and temperature and the initial degree of homogeneity of the input materials. Few ceramics are equilibrium materials systems, defined by composition and temperature. Glass, in particular, is a metastable material subject to transformation.

Large brittle ceramic forms processed at high temperatures will develop thermal stresses on cooling, which may result in cracking. It is estimated that the minimum internal surface area would be twice the outer surface if all measures were taken to reduce cracking, and up to 40-fold greater if no precautions were taken. However, penetration of water and leaching via such internal cracks show little effect on leach rate as long as the shards remain closely packed so that the leachant must penetrate via tight cracks. No information is reported on surface migration of radionuclides via such cracks.

If the rates of leaching of waste-form constituents in a repository are controlled by diffusion of low-solubility species through the groundwater adjacent to the waste package, as is discussed in Section 5.9.4, the existence of internal cracks will not affect the dissolution rate of those species.

In experiments on half-scale zinc-borosilicate-glass waste cast in stainless steel canisters, Martin (1980) found cracking similar to that reported above, with an internal surface area about tenfold greater than the external area. He found a large interior tensile stress due to the earlier cooling of the outer glass, and he calculated a tensile stress of 250 MPa in the steel canister.

The combined effects of residual stress, radiation damage, internal temperature gradients, and inhomogeneities will cause further cracking over long periods of time. Future tests of large-scale glass forms should include exposure of glass with the canister removed, simulating the long-term conditions in the repository.

Thermal stress damage in concrete is not reported in the literature available to the panel. SYNROC discs of about 20 cm in diameter and 10 cm thick can be made free of apparent cracks. No data were found for larger forms.

Glass devitrification, involving the nucleation and growth of crystals, will create new phases more or less soluble than the original glass, will generate internal stresses, and can increase the surface area within the waste form that could be eventually exposed to water. Devitrification is reported to cause at most a tenfold increase in leach rate in laboratory tests, and usually less than a fivefold increase at temperatures below 90°C. Most of the expected devitrification occurs when the glass is cast and cooled.

Experiments indicate that both the well-cured concretes and crystalline refractory ceramics, e.g., SYNROC, should be thermodynamically stable at temperatures below 200°C. It is not known what may result from the combined effects of heat, radiation, and temperature, e.g., metamictization of large crystal phases to small submicron crystals. As noted in Section 5.3.1, this may increase intergranular diffusion rates or expose more soluble phases.

In summary, ceramic waste forms containing useful loadings of fission products will be cracked from thermal stress. Additional cracking is likely. A tenfold increase in leach rate due to long-term devitrification seems well established from laboratory leach tests. No other significant effects of inhomogeneities on leaching have yet been identified from laboratory experiments. The extent to which inhomogeneities affect performance in a repository depends on whether or not the reaction rate of the waste-form surface with water is the rate-controlling process for dissolution and release of radionuclides to groundwater.

5.4.5. Radiation Effects on Waste-Form Properties

Radiation damage to the waste form has been investigated principally for glass materials. Limited information is available for concrete and crystalline ceramics.

Mendel et al. (1981) summarize radiation damage to waste forms resulting in changes in leach rates, form volume, microstructure, phases, stored energy, and physical properties. In the first several hundred years in a repository, there will be intense beta and gamma radiation from fission-product decay. Thereafter the principal radiation will be alphas from actinide decay. Atomic displacements from beta and gamma radiation effects on the waste form will be relatively unimportant. Radiation may decompose any organic matter that may be present.

Alpha-induced density changes in glasses range between ±1 percent for exposures up to about 3 x 10^{18} alphas/cm^3, where the effect seems to saturate (Roberts et al. 1981). Crystalline ceramics show about 1 to 2 percent decrease. SYNROC B, when exposed to a time-integrated fast-neutron flux of 1.3 x 10^{20} neutrons/cm^2 showed a density decrease of about 5 percent (Reeve 1981). This is calculated to be equivalent to about 2 x 10^{19} alphas/cm^2, or about 10^5 years of a high-level waste in a repository.

Stored energy in actinide-doped glasses and in a few crystalline materials occurs primarily by atom displacements. Doses of about 10^{18} alphas/cm^3 have been found to produce stored energy ranging from 50 to 400 J/g, where the process appears to saturate. Because the stored energy could be released only if the waste form were to be heated to 300°C, release of stored energy in a repository is not a problem. In a repository this stored energy can accumulate only after the form is relatively cool.

The possible importance of radiation damage to waste-form materials is related to the transport mechanisms cited in Section 5.3.1. Changes in volume that lead to cracking, or metamictization that changes the grain-boundary internal surface area and creates defect structures, can affect the diffusion of material to the surfaces where leaching occurs.

Some observers have found metamictization of crystalline materials on exposure to alpha doses greater than about 10^{18} alphas/cm^3. The crystals of ceramic waste-form materials such as SYNROC become amorphous. Cubic materials, e.g., UO_2, do not show such amorphization. This phenomenon also occurs with some of the devitrified crystals in glass forms and has been observed to be associated with microcracking (Weber et al. 1979).

Laboratory tests indicate that radiation exposure does not affect the leach rate of glass (Plodinec et al. 1982), but the leach rate of crystalline materials is affected. As will be discussed later, alpha, beta, and gamma radiation can affect the chemical activity of the leachant in a repository.

Helium gas from alpha decay can build up in the canister, but the pressure will be negligible. Hydrogen and oxygen can be generated by radiolysis in cement waste-form materials. Although recombination is likely, the availability of such hydrogen to a titanium alloy canister or overpack could be detrimental and should be investigated if cement waste forms are used.

Hebel et al. (1978) have suggested that the chemical changes associated with transmutations by radioactive decay, particularly the decays of cesium to barium and strontium to zirconium, could affect the long-term durability of the waste form. However, recent tests (Gray 1982) on borosilicate glass loaded with short-lived cesium show no effect of extensive cesium decay on leachability.

In summary, in studies on borosilicate glass, damage caused by self-irradiation results in only minor changes in the observed dissolution rate and in density changes of the order of 1 percent (Bibler 1981, McVay et al. 1981). Stored energy and helium buildup are concluded to have little or no adverse effect on waste-package performance.

5.5. LABORATORY LEACH DATA FOR HIGH-LEVEL WASTE FORMS

5.5.1. Introduction

There have been extensive laboratory tests on the rates of dissolution or "leaching" of waste-form materials in deionized water and in simulated groundwater. Most tests have been performed in the range of 25°C to 90°C, but data at temperatures up to 350°C and higher have been also reported (Altenhein et al. 1982, Malow 1982, Wicks and Wallace 1982, Cousens et al. 1982, Westsik and Peters 1981, McCarthy et al. 1979, Westsik and Turcotte 1978). The 90°C data summarized by the 1981 peer review committee (Alternative Waste Form Peer Review Panel 1981), with additions by J. L. Crandall (Savannah River Laboratory, personal communication to T. H. Pigford, 1981) for the present study, appear in Tables 5-4 and 5-5.

There have been attempts to use such laboratory data to predict the long-term performance of waste forms in a repository (Hench et al. 1980, Macedo et al. 1982, Altenhein et al. 1982, Wicks and Wallace 1982). Other studies have pointed out that the mechanisms that control dissolution in the long-term environment of a repository may be different from those that control in the laboratory tests, and one must be cautious in extrapolating the available data to repository design. Few of the studies have identified the species in the leachant with regard to ions, colloids, or complexes, and there have been only limited investigations of the chemical effects on leaching due to other components in the waste package. Many leaching experiments have included the effects of groundwater geochemistry, although few have simulated the lack of oxygen in deep groundwaters (McVay et al. 1981). None of the leaching experiments has included the resistances to dissolution that can result from the slow diffusion of dissolved radionuclides into the groundwater of a repository. No data are reported on the effect of expected thermal-period temperature gradients in leaching.

5.5.2. Laboratory Leach Data

In typical laboratory experiments, a small sample of a waste-form material is exposed to a leachant liquid in a vial kept at constant

TABLE 5-4 Alternative Waste-Form Leach Data for MCC-1 Static Leach Test, 28 Days, 90°C, Deionized H_2O

	Normalized Leach Rate ($g/m^2/d$)			Surface-to-Mass Ratio for Waste Form in Canister[a]	Fractional Release Rate[b] (yr^{-1})		
Waste Form	Cs	Sr	Actinide	(cm^2/g)	Cs	Sr	Actinide
Borosilicate glass	1.39	<0.04	<0.02 (Nd)	0.032	1.6×10^{-3}	$<4.6 \times 10^{-5}$	2.4×10^{-5} (Nd)
High-silica glass	0.10[c]	0.08	0.005 (U)	0.026	9.4×10^{-5}	7.6×10^{-5}	4.7×10^{-6} (U)
SYNROC	0.10	0.34	0.02 (U)	0.018	6.5×10^{-5}	2.2×10^{-4}	1.3×10^{-5} (U)
Tailored ceramics	<0.10	<0.035	0.007 (U)	0.018	$<6.5 \times 10^{-5}$	2.3×10^{-5}	4.6×10^{-5} (U)
Concrete (FUETAP)	20.0[d]	0.47	0.20 (Pu)[e]	0.036	2.6×10^{-2}	6.2×10^{-4}	2.6×10^{-4} (Pu)
Coated sol-gel particles[f]	$<10^{-3}$	$<10^{-5}$	$<10^{-5}$ (U)	1.03[g]	$<3.8 \times 10^{-5}$	$<3.8 \times 10^{-7}$	$<3.8 \times 10^{-7}$ (U)
Glass marbles in Pb matrix	0.04	<0.02	–	0.51[h]	7.4×10^{-4}	$<3.7 \times 10^{-4}$	–

[a] In order to determine the surface-to-mass ratio, the geometric surface area of a waste-form monolith 2 ft in diameter and 8 ft high enclosed within a 2 ft by 10 ft canister (i.e., a canister 80 percent full) was assumed. Increased surface area due to cracking was not considered. This assumption was made in order to provide calculated fractional release rates. In reality, the specific surface area of the forms may vary depending upon (1) the extent of cracking and (2) if the forms take the shape of a single monolith or several small monoliths stacked in a single canister. The mass of the monolith was calculated as the product of the density of the form multiplied by the volume of a 2 ft by 10 ft canister 80 percent full (i.e., a 2 ft × 8 ft monolith).

[b] The fractional release rate is calculated as the product of the leach rate and the surface-to-mass ratio.

[c] Na leach data to simulate cesium.

[d] Cesium added as coarse zeolite.

[e] Pu leach rate data, modified International Atomic Energy Agency test, 28 days at 25°C.

[f] Leach rate calculated for seven days at 90°C in deionized H_2O.

[g] In order to determine the surface-to-mass ratio for a canister of coated sol-gel particles, a 2 ft diameter by 10 ft high canister 80 percent full of coated particles was assumed. It was also assumed that 40 percent of the volume occupied by the coated particles was interstitial void space. The total surface area was calculated as the product of the number of coated particles (1.54×10^8) in the canister multiplied by the average surface area for a single coated particle (1.21×10^{-2} cm^2). The total mass was calculated as the product of the number of coated particles in the canister multiplied by the average mass of a single coated particle (1.17×10^{-2} g).

[h] In order to determine a representative value for the surface-to-mass ratio for the metal matrix form, a 2 ft by 10 ft canister 80 percent full was assumed. It was also assumed that glass occupied 64 percent of the volume; Pb, 36 percent. The surface-to-mass ratio was calculated using the geometric surface area of a 2 ft by 8 ft monolith and the total mass of glass and Pb. This value was averaged with the surface-to-mass ratio calculated using only the geometric surface area of the glass marbles and the mass of the glass marbles. The average of these two values was used to represent the surface-to-mass ratio for the cast metal matrix form.

SOURCE: J. L. Crandall, Savannah River Laboratory (personal communication to T. H. Pigford, June 1981).

TABLE 5-5 Static Leach Test Data for Silicate H_2O and Brine at 90°C for 28 Days

	Silicate H_2O						Brine					
	Leach Rate (g/m^2-d)			Fractional Release Rate (yr^{-1})			Leach Rate (g/m^2-d)			Fractional Release Rate (yr^{-1})		
Waste Form	Cs	Sr	Actinide	Cs	Sr	Actinide	Cs	Sr	Actinide	Cs	Sr	Actinide
Borosilicate glass	0.83	<0.04	<0.02 (Nd)	9.7×10^{-4}	4.6×10^{-5}	2.4×10^{-5}	0.24	<0.04	<0.02 (Nd)	2.8×10^{-4}	4.6×10^{-5}	2.4×10^{-5}
High-silica glass[a]												
SYNROC	0.11	0.06	<0.017 (U)	7.2×10^{-5}	3.9×10^{-5}	1.1×10^{-5}	–	<0.25	<0.42 (U)	–	$<1.64 \times 10^{-4}$	$<2.7 \times 10^{-4}$
Tailored ceramics[a]												
Concrete (FUETAP)	16.0	0.45[b]	–	2.1×10^{-2}	5.9×10^{-4}	–						
Coated sol-gel particles[a]												
Glass marbles in Pb matrix[a]												

NOTE: Only leach rate and fractional release data are shown. All other data are the same as those in Table 5-4. Data are from MCC-1 static tests.

[a] Not available.

[b] Fourteen-day leach period.

SOURCE: J. L. Crandall, Savannah River Laboratory (personal communication to T. H. Pigford, June 1981).

temperature. The liquid can be kept continuously in contact with the solid sample during the entire test or can be replaced intermittently to avoid the accumulation of dissolved materials in the solutions. Such accumulation can cause a significant retardation of leaching as saturation is approached.

The results are typically reported as the mass of each element of interest entering the solution over a given time interval, divided by the surface area and by the weight fraction of the element originally present in the sample, called the "normalized leach rate." Emphasis has been given to data from static leach tests carried out for 28 days at 90°C using deionized water, referred to as MCC-1 experiments, such as the data summarized in Tables 5-4 and 5-5. More recent data for SYNROC from Oversby and Ringwood (1982) are summarized in Table 5-6.

The data in Tables 5-4, 5-5, and 5-6 for fractional rates for waste forms were calculated by assuming that the rate of dissolution of each element in a waste form, per unit outer surface area of the form, is the same as deduced from the laboratory experiments. Note that cesium, the most soluble substance of those listed, leaches from glass about 35 times faster than strontium. If its rate were to remain constant, most of the cesium would be lost from the waste form itself in fewer than a thousand years.

TABLE 5-6 Summary of New Data for Leaching of SYNROC

Element	Total Waste Content[a] (wt%)	Maximum Time of Test (d)	Temperature (°C)	Experimental Dissolution Rate[b] (g/cm²-d)	Fractional Dissolution Rate[c] (yr⁻¹)
Cs	9	145	95	1.3×10^{-8}	1.5×10^{-7}
	9	60	200	1.2×10^{-7}	1.4×10^{-6}
	16	104	95	4.0×10^{-8}	4.7×10^{-7}
	20	84	95	1.8×10^{-7}	2.1×10^{-6}
	20	64	200	2.4×10^{-7}	2.8×10^{-6}
Sr	9	104	95	1.4×10^{-8}	1.6×10^{-7}
	9	47	200	2.9×10^{-7}	3.4×10^{-6}
	20	47	200	8.8×10^{-7}	1.0×10^{-5}
U	9	17	95	4.3×10^{-11}	4.9×10^{-10}
	9	23	200	2.5×10^{-9}	2.9×10^{-8}
	20	27	200	4.4×10^{-9}	5.1×10^{-8}
Nd	9	19	95	7.4×10^{-10}	8.7×10^{-9}
	9	19	200	4.2×10^{-9}	4.9×10^{-9}

NOTE: The experimental rates listed are those observed at the longest exposure times used in the measurements. In some cases these were roughly constant, but the rates of change could not be established clearly. They were "normalized" by the authors by dividing each observed rate by the weight of each element in the waste form.

[a] Simulated high-level waste, based on weight of fission-product and actinide oxides in waste.

[b] The averaged normalized rates for the principal constituents of the three compounds composing the host synthetic rock were about 7×10^{-7} g/cm²-d for barium and calcium, 1×10^{-9} g/cm²-d for titanium, and 3×10^{-9} g/cm²-d for zirconium, respectively.

[c] The fractional leach rates per year were calculated for cylinders having the dimensions given in Table 5-3. The rates in a repository were assumed proportional to the internal surface area of the canister.

SOURCE: Data of Oversby and Ringwood (1982) at the greatest times of exposure to deionized water.

From the data in Tables 5-4 and 5-5 it is apparent that dissolution is not congruent among the elements listed. The greatest fractional dissolution rate occurs for cesium in all cases, except for SYNROC, in which strontium dissolves at the greater rate. The smallest values of the rate constants occur for uranium and plutonium and for the rare earth fission product neodymium. An explanation for the incongruent dissolution of borosilicate glass is that all constituents are released congruently from the dissolution of the glass, but the low-solubility constituents cannot dissolve at that rate and precipitate as part of a surface layer (Wicks and Wallace 1982, Malow 1982, McVay et al. 1981).

The laboratory leach rates of many elements also depend on the ratio of the sample surface area to the leachant volume (S/V), because this ratio affects the approach to solution saturation of the dissolved constituents.

The latest normalized leach rates for SYNROC, shown in Table 5-6, indicate that the SYNROC values for cesium and strontium in Table 5-4 may be too great. According to Oversby and Ringwood (1982), their new data indicate that SYNROC is about 3,000 times more resistant to leaching than borosilicate glass. Although these new measurements may have several defects, due to the nature of the experiment, one must assume that synthetic minerals or other crystalline ceramic waste forms of some kind may be very resistant to the escape of radionuclides. This could be especially interesting for cesium and other elements that are comparatively soluble in water. It could be important for the low-solubility radionuclides if the even lower dissolution rates predicted in Section 5.7.4 for waste forms in repositories cannot be validated or if a redundant back-up to the low-solubility releases is desired.

As discussed in Section 9.11, synthetic mineral-waste forms could be useful, especially if there were repository sites with limited protection other than from the waste form itself, because of short water travel times through surrounding rock or because of limited dilution of escaping groundwater. This line of investigation should be pursued.

5.6. INTERACTION OF BOROSILICATE GLASS WITH WATER

5.6.1. Chemical Reactions

The basic chemical reactions that occur when water reacts with glass are shown in Figure 5-2 (Wicks 1982b). Initially, sodium ions in the glass leave the solid when hydrogen or hydronium ions from the solution exchange with them. In a static laboratory experiment this increases the pH of the liquid as hydroxyl ions are formed, resulting in a leachant whose hydroxyl ions react with the silicon-oxygen network of the glass at its surface, releasing some of the silicon to the solution as hydrated species. The constituents of the glass, including the radionuclides, are also released. There is left a porous gel-like layer on the glass surface.

Because fission-product cesium is also an alkali element, it would be expected to be released more rapidly from glass in the early stages of dissolution, like sodium.

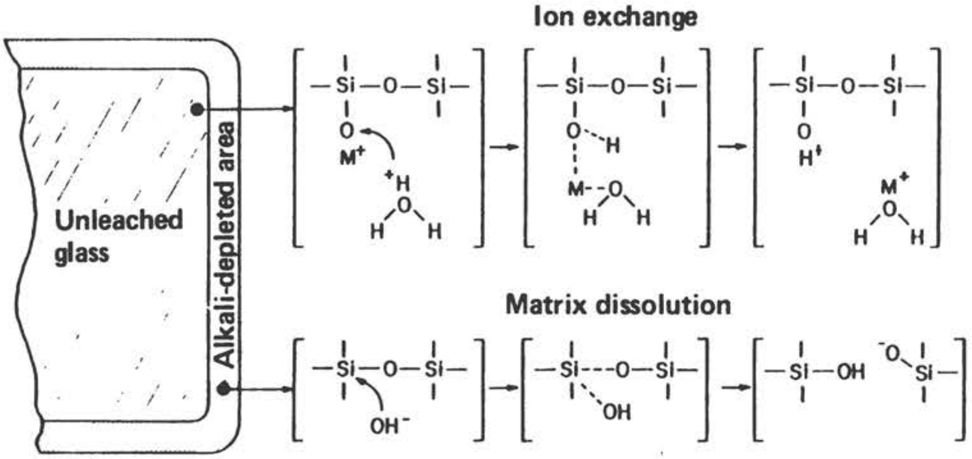


FIGURE 5-2 Chemical reactions of glass with water. Source: Courtesy of G.G. Wicks.

5.6.2. Effect of pH

The initial rate of penetration of hydrogen ions into borosilicate glass is determined by the diffusion of water species, e.g., H_3O^+, the depth of penetration being proportional to the square root of time. Soon, within hours or days (Mendel et al. 1981), the release of alkali from the waste becomes linear with time, and is thought thereafter to be controlled by the rate of release of silicon from the waste. Ion exchange is the predominant process of silicon release of mobile species, e.g., alkali and alkaline-earth elements, in the pH range from 4.5 to 9.5 and at both higher and lower pH matrix dissolution of the dealkalized layer controls, as illustrated in Figure 5-3 (Plodinec et al. 1982). The midrange of pH insensitivity spans the expected pH range of repository groundwaters. The local chemistry immediately surrounding the waste form may generate a pH outside this range. In closed systems, as in the static laboratory experiments, the dissolution of the dealkalized layer can be accelerated by a sufficient increase in leachant alkalinity or by acidic reactions promoted by some salt brines and by radiolysis.

5.6.3. Effect of Dissolved Constituents in the Leachant

The buildup of additional dissolved constituents in the leachant in a closed system can also affect dissolution rates. As the low-solubility constituents approach saturation, their dissolution rates decrease. This has led to correlations of laboratory data in terms of the ratio of the sample surface area to leachant volume, the higher values of this ratio resulting in lower dissolution rates of silicon and other low-solubility constituents as saturation is approached.

Accumulation of dissolved constituents and reaction products in cracks results in lower leach rates from interior surfaces than from exposed surfaces (Perez and Westsik 1981), a manifestation of the high interior surface area and rapid saturation of interior liquid by silica

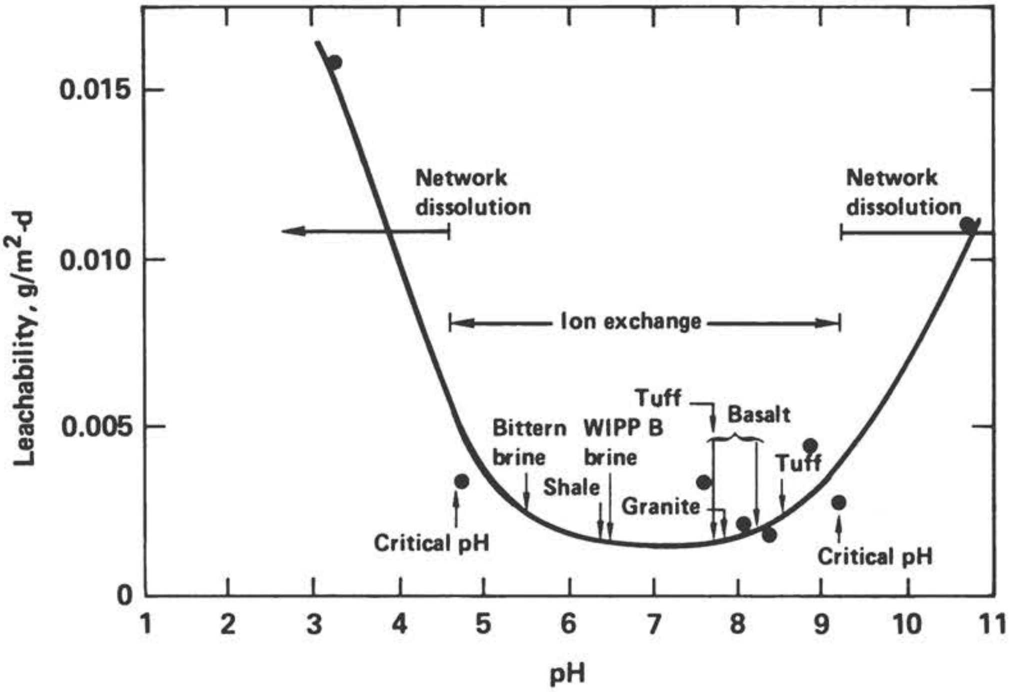


FIGURE 5-3 Effect of pH on leach rate of silicon from borosilicate glass, based on 5-day static tests at 23°C with S/V = 0.1 cm^{-1}. WIPP refers to the Waste Isolation Pilot Plant. Source: Plodinec et al. (1982).

(McVay 1982). Groundwater that already contains appreciable quantities of dissolved silica, such as the basalt groundwater at Hanford, lowers the dissolution rate (Braithwaite 1980). The onset of saturation effects can be modified by the presence of other species in solution; for instance, sodium in natural waters increases the solubility of quartz. A small increase in leach rate was found when iron container material was present, although colloidal precipitation (cf. Section 5.6.7) removed silica from solution (McVay and Buckwalter 1982). In the presence of bentonite backfill, sodium is released more rapidly and is incorporated in the backfill. The possible effects of dissolved nitrogen and oxygen are discussed in Sections 5.6.8 and 5.6.9.

5.6.4. Congruent Release and Incongruent Dissolution

Based on laboratory experiments spanning the temperature range from 25°C to 250°C, it is generally concluded or assumed (Altenhein et al. 1982, Grambow 1982, Malow 1982) that, after the initial alkali transient, borosilicate-glass constituents are released congruently from the sample, but the low solubility of many of the constituents, including the actinides, prevents them from going into solution as fast as they are released from the glass matrix. Thus, while release from the sample is congruent, dissolution is not.

The less-soluble species form precipitates near the outside of the silicon-depleted layer, which may contain compounds such as ferric hydroxide, manganese dioxide, and calcium silicate. Correlations discussed in Section 5.6.10 suggest that either these surface layers or localized precipitates on the glass surface reduce the dissolution rates in laboratory experiments. Increasing pH usually affects the solubilities of the transition metals, rare earths, and actinides, which may explain the pH dependence of dissolution rates of these species in laboratory experiments.

The time-dependent cumulative releases of individual elements from borosilicate glass in static experiments are shown in Figure 5-4 (McVay et al. 1981). Only sodium, boron, and molybdenum dissolve at a constant rate. For other elements, dissolution rates decrease with time because of the approach to saturation in the leachant. Later negative rates, i.e., net removal from solution, are attributed to precipitation resulting from changing pH and solution concentrations.

5.6.5. Surface Layers

Hypotheses on the formation and structure of surface layers formed during glass leaching, and their effects on leaching, have been advanced by Simmons et al. (1982), Roy (1982), Hench et al. (1980), Altenhein et al. (1982), Clark et al. (1982), Grambow (1982), McVay (Pacific Northwest Laboratory, personal communication to J. R. Johnson, 1982), Wicks and Wallace (1982), and Malow (1982). Clark et al. (1982) suggest that the alkali ions preferentially dissolving from the glass surface leave vacancies in the glass-silica network. The dealkalized layer becomes dehydrated and thicker with time. Release of some ions is inhibited by trapping as they become hydrated, resulting in a crust that is partially crystalline. Hydrate formation, particularly of divalent and trivalent species, is consistent with observations (Roy 1982) that phase-equilibria data predict that, except for the tetravalent oxides and some of their compounds, the oxides will react with water to form these less-soluble hydroxylated phases.

Reported differences in the rates of formation and composition of layers may be due to differences in sample composition.

Three principal issues related to these surface layers are their stability, their effect on laboratory dissolution rate, and their effect on dissolution rates in a repository. The latter two are considered in Sections 5.6.10 and 5.7.

The composition profiles and stability of surface layers on borosilicate glass have been studied at temperatures from 25°C to 200°C. The layer seems to become thicker with time. Malow (1982), in work done at the Pacific Northwest Laboratory (PNL), reports that layers formed in brine leachants up to 200°C peeled off after short leaching times, but longer leaching periods yielded denser surface layers that could not be removed. Similar stability at all temperatures studied is reported by Altenhein et al. (1982), Wicks et al. (1982a,b), and Wicks and Wallace (1982). Simmons et al. (1982), Malow (1982), Wicks et al. (1982a,b), Wicks and Wallace (1982), and Mendel et al. (1981) found some peeling and spalling on cycling from wet to dry conditions.

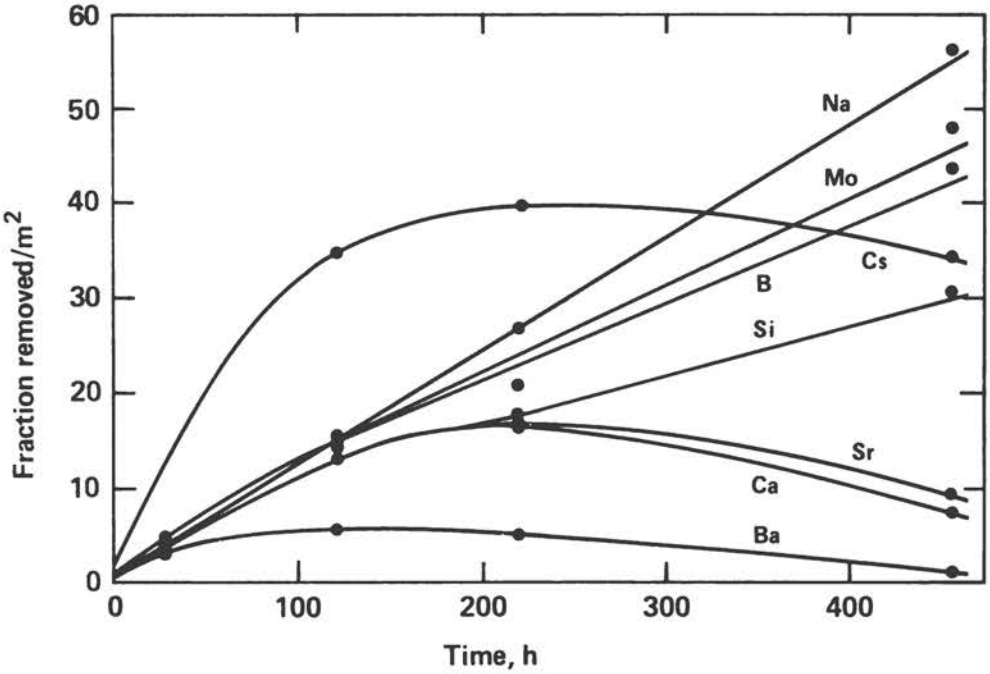


FIGURE 5-4 Fraction of original elemental concentration removed by leaching in distilled water per unit surface area of sample of PNL-76-68 glass at 89°C with a surface area to solution volume ratio of 10 m^{-1}. Source: McVay et al. (1981).

5.6.6. Structural Alteration of Glass by Water Vapor

Borosilicate glass undergoes hydration reactions with water at an increasing rate as temperature is increased. This is evidenced in part by the early formation of "crusts" on the glass. When borosilicate glass was exposed to water vapor, crusts formed within 3 to 7 days, varying from 0.03 mm thick at 150°C to 3 mm at 350°C (Bates et al. 1982). The possible effects of these crusts and surface layers on dissolution rate are discussed in Section 5.6.10. This rapid hydration suggests that over a long time and especially at high temperatures, glass may become completely altered structurally, even though it has not dissolved. If so, the present laboratory leach data that include only a relatively small degree of such alteration at the sample surface may not be indicative of longer-term releases.

If the physical integrity and surface area of the glass are important in determining the dissolution rate of radionuclides in a repository, as they are in laboratory experiments, then long-term structural alterations can become significant. Conversely, if the dissolution rate of radionuclides is determined by the rate of diffusional-convective transport of low-solubility species into the surrounding groundwater, then long-term structural alterations may be unimportant.

5.6.7. Colloid Formation

Many of the low-solubility species in radioactive wastes can form colloids or become part of other colloidal substances in the leachant. The possible effect of colloids on release of radionuclides to the groundwater has been mentioned in Section 5.3.2. McVay and Buckwalter (1982) found, in studying the effect of a ductile iron container on glass leaching, that iron-silicate colloids were formed, silicon was rapidly removed from solution, and many other elements were found incorporated in colloidal precipitates. No data on actinides or radionuclides were reported, but the authors suggest that radionuclides could become trapped in these colloidal precipitates.

Colloids and polymeric phases incorporating a radionuclide species can affect the amount of that species in solution at saturation (Rai and Swanson 1980).

Formation of actinide colloids was studied by Avogadro et al. (1981), who flowed water over powdered borosilicate glass and then through a column of sand to simulate porous rock. Actinides in water entering the column were found to be principally in colloidal and suspended form, but most (98 to 99 percent) were trapped in the sand column. Actinides in the trapped colloids were eluted with carbonate solution. Whether the concentrations in water leaving the borosilicate glass were greater or less than what would be predicted from leaching experiments was not determined. A similar experiment using a dense sample of borosilicate glass with a well-characterized surface area and without particulates would be more relevant to performance in a repository.

5.6.8. Radiolysis

Radiolysis of leachant solution by alpha, beta, or gamma radiation can change the chemistry of the aqueous solution and affect the leaching of glass. Gamma radiolysis is likely to be more important during the thermal period, and alpha radiolysis can be important during the longer term at ambient temperature. Different radiolytic effects are expected with alpha particles.

The increase in silicon leach rate due to gamma radiolysis is greater at lower temperatures. When air is excluded, there is no measurable effect on leach rate at 90°C for gamma dose rates of 2.4×10^4 Gy/hr (2.4×10^6 rad/hr) (McVay and Pederson 1981). However, actinide release may be affected by the resulting oxidizing environment (McVay et al. 1981).

If the water contains nitrogen, as in the early period of repository resaturation, nitric acid is formed as a radiolysis product and has increased leach rate by an order of magnitude in static laboratory experiments (McVay and Pederson 1981).

From studies of gamma irradiation of basalt groundwater at 90°C, McVay (1982) found the same leach rate of cesium and boron from borosilicate glass as in deionized water. Oxalic acid was generated at about 80 ppm at 90°C, and less at lower temperatures. Oxalic acid is sometimes used to keep colloids from flocculating, and it forms low-solubility compounds with some actinides. Its effect on waste-form performance is unknown.

Rai et al. (1980) found nitric acid formation and increased plutonium solubility in alpha irradiation of air-saturated water, so radiolysis can affect the solubility-limited dissolution rate of such species if air or dissolved nitrogen is present.

5.6.9. Effect of Oxidation Potential

Less information is available on the effects of oxidation potential on leach rates. The oxidation potential of groundwater, usually expressed as Eh, can decrease with depth, depending on geochemical conditions (see Chapter 7). At proposed repository depths, leaching may take place under reducing conditions, where the solubilities of the actinides and technetium are relatively small (see Table 7-1) and for which lower leach rates would be expected. However, essentially all laboratory leach tests on glass have been obtained in air-saturated solutions. One measurement in water degassed to about 1 ppm oxygen showed a general decrease in the leach rate of actinides (McVay et al. 1981). It appears that the present laboratory leach data are conservatively high. Waters in hard rock repositories have generally been reported to be reducing, but Winograd and Robertson (1982) report dissolved oxygen in such waters and caution that the assumption of reducing conditions in all deep aquifers is not necessarily correct.

5.6.10. Correlations of Laboratory Leach Data

Earlier leach-rate data on borosilicate glass showed an initially high rate that decreased with time in a manner characteristic of diffusion-controlled release, followed by a later constant rate. The validity of the data on the early diffusion-controlled release has been challenged (G. L. McVay, Pacific Northwest Laboratory, personal communication to T. H. Pigford, 1981). A more important consideration is the longer-term trend in leach rates. Several independent studies of the trends in laboratory data taken over a period of about one year show that there is a transition from a constant rate of release of soluble species to a rate that decreases with time (Altenhein et al. 1982, Kuhn et al. 1982, Westsik and Peters 1981, Wicks and Wallace 1982). Although silicon dissolution rate should decrease with time in a static experiment as the silicon concentration in the leachant approaches saturation, a decreasing rate is still inferred after the saturation effect is accounted for. However, no decrease in leach rate with time is reported for flowing experiments, carried out over a period of 18 months (Macedo et al. 1982).

Correlations found separately by Wicks and Wallace (1982) and Altenhein et al. (1982) suggest that the later decrease in dissolution rate in static experiments is due to a diffusional resistance across a surface layer of precipitates that increases in thickness linearly with time. When diffusion through this layer is controlling and when saturation effects are excluded, the rate of dissolution is predicted to decrease inversely as the square root of the time. Both groups find that the laboratory leach rates, obtained for time periods up to about one year of leaching, decrease even more rapidly with time than predicted by these correlations. The correlations by Wicks and Wallace have been based largely on leach data for defense waste in borosilicate glass in brine leachants, and the correlations by Altenhein et al. have been based on commercial borosilicate glass in various leachants at temperatures up to 200°C.

Kuhn et al. (1982) correlated leach-rate data (Adams 1979, Strachan 1982, Westsik and Peters 1981) for commercial borosilicate-glass waste in various leachants at temperatures from 50°C to 250°C, obtained for leach times up to about one year. The correlation was based primarily on boron leach data, so no saturation effects were expected because of the high solubility of boron. They found that, at a given temperature, the leach rates are initially constant, and after about 20 days vary inversely as the square root of time, except for leach rates at 250°C, which decrease more rapidly with time. Because these later leach rates at several temperatures below 250°C all follow the same function of time, it was concluded that diffusion through a thickening surface layer could not account for the observed decreases. The data were correlated by postulating an unidentified, congruently released constituent that resorbs on a portion of the sample surface and blocks further release from that part of the surface, suggesting a temperature-dependent Langmuir sorption isotherm.

At most, each of these correlations is empirical. Rate and sorption constants are adjustable parameters and have not been justified as being physically reasonable. Regardless of the mechanisms postulated to explain the correlations, it seems confirmed that the leach rate of the solid sample in static experiments decreases approximately as the square root of time for time periods up to about a year.

The low apparent dissolution rates from nepheline-glass waste buried in soil for 15 years have suggested a qualitative explanation on the basis of a surface layer, supported if necessary by the surrounding soil, that provided a diffusional resistance to leaching (Merritt 1976).

Altenhein et al. used their surface layer/diffusion correlation to predict long-term time-dependent leach rates of borosilicate glass in a salt repository initially at 200°C. It was assumed that a relatively large volume of brine surrounds each waste canister, consistent with an assumed $S/V = 0.01\ \text{cm}^{-1}$. The rate of glass dissolution was calculated to become negligible, i.e., essentially zero, after a few hundred years if the rate decreases with time as described above. In another calculation a yearly detachment and reformation of the surface layer was assumed, so that after each detachment the higher leach rates associated with a fresh surface were resumed. Again, it was estimated that the net release rate will become negligible after a few hundred years.

It is possible that such extrapolations based on laboratory leach data, with little credit for saturation effects, represent a conservative upper limit to the release rates in a repository. However, the laboratory data cover such a short exposure time that the extrapolations are necessarily of uncertain validity, even as upper-limit estimates. These empirical correlations provide a reasonable basis for interpolation and for predicting dissolution rates at times not far removed from the time scale of the experiment. However, there is little basis for extrapolating to the thousands and hundreds of thousands of years important in the performance analysis of repositories.

Further, the analyses that take into account the approach to silicon saturation correlate the data on the release and dissolution of silicon from the glass. The applicability of such correlations to the rate of dissolution of low-solubility radionuclides, that dissolve at different rates and that are found to reside in different positions in the precipitate layers, is questionable.

There remains the possibility of longer-term structural alterations of the glass and its effect on laboratory-type leach rates.

We do not adopt such extrapolations in the present study, but instead use the dissolution rates predicted in Section 5.7.4 for our repository performance analysis in Chapter 9.

5.7. PREDICTION OF WASTE-FORM DISSOLUTION RATES IN A REPOSITORY

5.7.1. The Phenomena Controlling Dissolution Kinetics

The estimates in Tables 5-4, 5-5, and 5-6 of the fractional dissolution rates for a waste package in a repository may be upper limits to the actual dissolution rates, because they are based on the assumption that in a repository the rate of dissolution per unit area of waste-form outer surface will be the same as the rate of dissolution per unit area in laboratory experiments. In the laboratory the dissolution rates are largely controlled by the kinetics of solid-water chemical interaction, by leachant composition, by surface precipitates, and possibly by diffusion rate within the surface layers. However, in a repository there is additional resistance to dissolution owing to the slow diffusion and convection of dissolved low-solubility species into the slowly moving liquid in the surrounding porous medium. If the rates of transport into the groundwater are less rapid than the rates of release of radioactivity due to the solid-liquid interaction, then the diffusion kinetics in the liquid boundary layer will control the net rate of dissolution. The rates of diffusive-convective transport into the groundwater adjacent to the waste are considered in Section 5.7.4 and are compared with the rates of solid-water interactions deduced from laboratory experiments on borosilicate glass.

5.7.2. Extrapolations Based on the Ratio of Waste Surface-to-Water Volume

In correlating data from static laboratory leach experiments, it has been recognized that the buildup of corrosion products within the leachant in static experiments can affect the rate of dissolution of the waste and its components. As discussed earlier (Section 5.6.3) the ratio of sample surface to volume of static leachant (S/V) has been introduced as a correlating parameter (Pederson et al. 1982, Wicks and Wallace 1982). Various proposals have been made to extrapolate the laboratory leach rate data to repository conditions by adopting the laboratory data taken at the same S/V ratio that is presumed to exist for the waste package in a repository (e.g., Macedo et al. 1982, Yen-Bower et al. 1979, Plodinec et al. 1982). Some simulation of a borehole emplacement in salt, if flooded with brine from an unexpectedly large amount of thermally migrated inclusion, is conceivable, although the extrapolation is uncertain. However, in a wet-rock repository there is no meaning to an equivalent volume of groundwater in contact with the surface area of each waste package.

Some artificial value of the equivalent groundwater volume, dependent on diffusivity and groundwater flow, could be calculated from a more mechanistic theory of the processes that control dissolution in the repository, where the corrosion products are carried away slowly but continuously by the moving groundwater. If such a mechanistic theory were available, however, there would be no need to rely on the artificial concept of an equivalent water volume. One possible approach toward a mechanistic theory for the dissolution rate of low-solubility species is discussed in Section 5.7.4.

Although the study of glass waste forms and their interaction with water is worthwhile, the present types of laboratory leaching investigations seem unlikely to yield more than the properties of glass and not a design procedure in which the properties of repository rock affecting waste performance are taken into account. In a wet-rock repository the waste is dissolved by groundwater flowing slowly in the porous or fractured rock surrounding the waste. Under these conditions the rates of diffusion and convective transport of dissolved species away from the waste surface into the surrounding groundwater may be slow enough for the low-solubility species that they limit the net rate of dissolution from the waste.

In the existing laboratory leaching experiments, whether static or slowly flowing, natural convection over the weeks and months of the experiment results in negligible or nonexistent concentration profiles within the leachant, even though the experiment is at constant temperature. For that reason the laboratory leaching experiments measure only the solid-liquid kinetics. Assuming that these are the controlling kinetics in a repository environment is not warranted. However, laboratory studies of interactive phenomena are important to the development of a technique for predicting waste-package performance in a repository, such as experiments that help define under repository conditions the chemical environment and the chemical species that control solubility and colloid formation.

5.7.3. Solubility-Limited Bulk Flow of Radionuclides

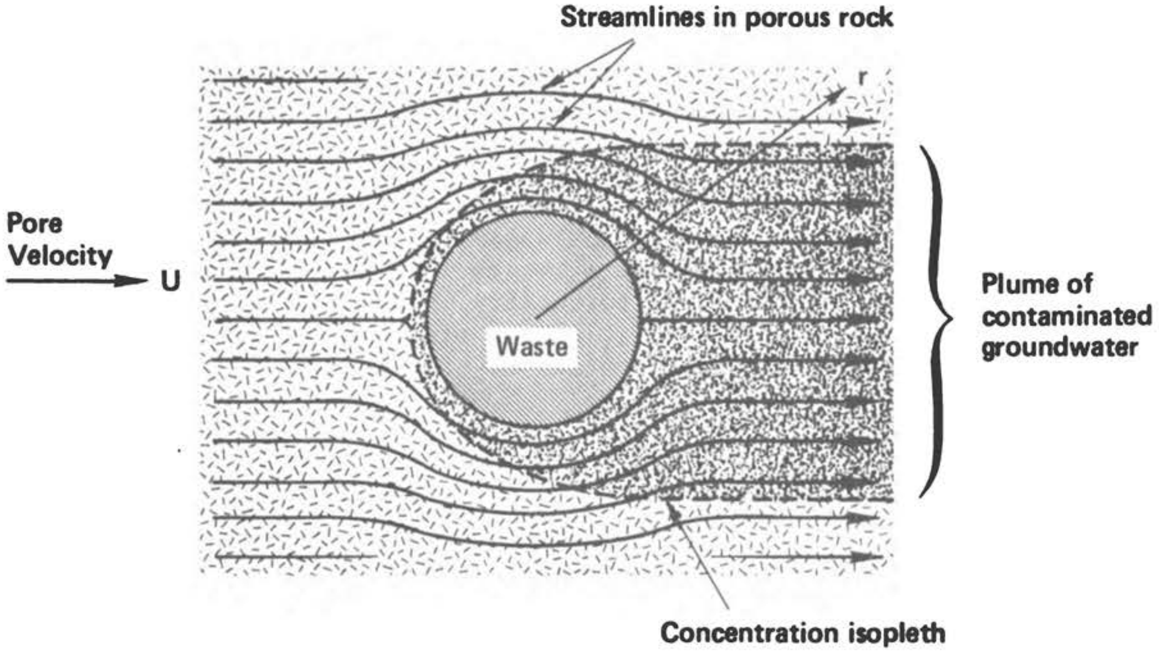
The slow rates of diffusion of dissolved substances through liquids and the very slow movement of groundwater across the surface of a buried waste canister suggest the possibility that, whatever the rates of release of radioactive elements from waste-form materials in laboratory tests may be, there will be some upper limit to the rates of transport of radionuclides from the repository toward the environment. One wholly unrealistic limit can be found by an elementary computation (Wood 1980, Wood and Rai 1981). If the entire flow of groundwater through the waste-emplacement zone of a repository can somehow become saturated with radioelements, then the maximum rates of transport of those elements and their isotopes can be given by the product of the solubility of the stable compounds of the element (cf. Table 7-1) and the volumetric flow rate of groundwater. Assuming that suspended solids and colloids cannot penetrate the porous or fractured rock, this product must be an upper limit to the rate of release of a radionuclide from the repository, regardless of the total quantity of radioactivity stored, the measured release rates, or the exposed surface area of the waste material.

5.7.4. Mass Transport Through the Liquid Boundary Layer

The analyses by Chambré et al. (1982a,b,c) predict the rate at which radionuclides in liquid at the surface of a waste package can be transported into groundwater in the surrounding porous medium. The calculation assumes that the concentration of each elemental species in the water at the surface of the waste package is at a maximum value given by the solubility of that species. This assumption was suggested from the laboratory experiments by Rai and Strickert (1980), who observed that the equilibrium concentrations of low-solubility actinides in static leachant in contact with borosilicate glass were equal to the solubilities of the stable compounds of those species in the leachant, and by the many observations (Altenhein et al. 1982, Malow 1982, Wicks et al. 1982b) of actinides and other low-solubility constituents in precipitates formed on borosilicate-glass surfaces during laboratory leaching experiments.

Assuming that the waste form is in contact with porous rock or with backfill of essentially the same porosity and permeability as the rock, the profiles of groundwater flow, of the concentration of a dissolved radioelement, and of the resulting plume of contaminated water flowing past the waste form are illustrated in Figure 5-5. Diffusion and convection of the individual dissolved constituents through this concentration boundary layer are analyzed to predict the limiting dissolution rate of low-solubility radionuclides. Effects of gaps, different backfill properties, and colloids are discussed later.

In the ensuing discussion, "release rate" refers to the total rate at which a chemical element is transported from the waste package into the surrounding groundwater. The theories are applicable to both dissolved and colloidal species if the appropriate boundary concentrations and diffusion coefficients are known. When the "release rate" theory is applied only to a dissolved species, the "dissolution rate" is predicted.



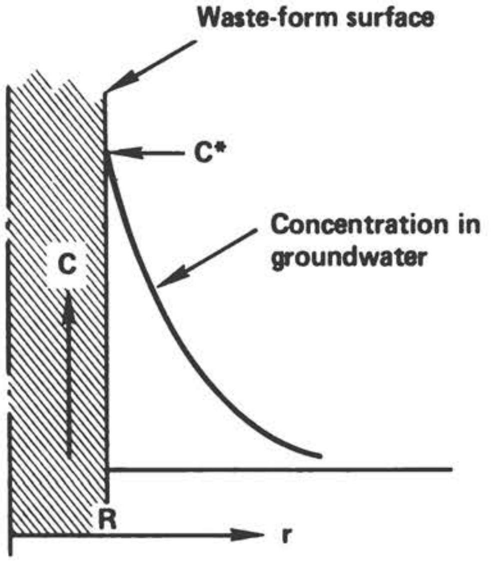


FIGURE 5-5 Velocity and concentration profiles for groundwater flowing across a waste cylinder.

Release Rate Controlled by Diffusion and Convection

The diffusion-and-flow calculation makes use of the known distribution of groundwater velocities around a semi-infinite cylinder through the pores of the surrounding rock, called Darcy flow. The velocity distribution is mathematically equivalent to the potential flow of theoretical hydrodynamics. The rate of diffusion through such a fluid is assumed to be represented by Fick's law, using a diffusion coefficient inside the rock pores equal to that in a free liquid and conservatively disregarding the tortuosity of the pores.

The fractional release rate f_j of the elemental species j and its isotopes is calculated at steady state to be

$$f_j = \frac{8N_j^* D_j^{1/2} \varepsilon U^{1/2} (1 + R/L)}{(\pi R)^{3/2} n_j}, \quad UR/D_j > 4, \tag{1}$$

where N_j^* is the concentration in the groundwater at the waste surface, D_j is the liquid diffusion coefficient, L is the length of the waste cylinder, R is its diameter, U is the pore velocity of the groundwater before it comes near the waste, ε is the porosity of the surrounding rock, and n_j is the bulk density (g/cm^3) of elemental species j in the waste. To predict solubility-limited dissolution rate, N_j^* is chosen as the solubility.

Table 5-7 gives values of the solubility of silica and the solubilities in water of radioelements in borosilicate glass. The solubilities are those estimated by K. B. Krauskopf (Stanford University, personal communication to T. H. Pigford, 1982) for the stable compounds of the elements in typical groundwater (cf. Chapter 7, Table 7-1). Also listed in Table 5-7 are the bulk densities and the calculated fractional release rates for a typical waste form of glass exposed to groundwater at an approach velocity of 1 m/yr in a rock of 1 percent equivalent porosity. The assumed diffusion coefficient of 10^{-5} cm^2/s is typical for an electrolyte in water. It conservatively neglects the effect of tortuosity (Neretnieks 1980a), which in granite can result in more than a 1,000-fold reduction in D_j (Bradbury et al. 1982) and more than a 30-fold reduction in the estimated fractional dissolution rates.

Release Rate Controlled by Diffusion

When the conditions are such that UR/D_j is much less than four, Equation (1) must be replaced by an equation developed (Chambré 1982c) for a finite waste package in the limiting case of zero velocity:

$$f_j = \beta \varepsilon D_j N_j^*/n_j, \text{ as } U \to 0, \tag{2}$$

where β is a geometrical factor that can be calculated from the waste-form dimensions. For a sphere, $\beta = 3/R^2$; for a prolate spheroid that approximates the cylindrical waste form of dimensions given in

TABLE 5-7 Calculated and Observed Fractional Release Rates for Borosilicate Waste Glass

Constituent	Initial Waste Concentration (g/cm^3)	Solubility[a] (g/cm^3)	Fractional Release Rate (yr^{-1})	
			Calculated[b]	Observed[c]
SiO_2	1.6	5×10^{-5}	1.1×10^{-6}	1.6×10^{-3}
Tc	1.92×10^{-3}	1×10^{-9}	2×10^{-8}	
U	1.22×10^{-2}	2×10^{-9}	8×10^{-9}	1.5×10^{-6}
Np	1.92×10^{-3}	1×10^{-11}	2×10^{-10}	6.6×10^{-4}
Pu	1.15×10^{-4}	1×10^{-9}	4×10^{-7}	2.6×10^{-5}
Am	3.56×10^{-4}	1×10^{-10}	1×10^{-8}	2.7×10^{-6}

NOTE: Conditions assumed for theoretical computations: radius of cylinder = 0.152 m; length of cylinder = 2.46 m; diffusion coefficient of all substances in water = 10^{-5} cm^2/s; porosity of rock = 0.01; groundwater pore velocity = 1 m/yr; density of glass = 3.0 g/cm^3; temperature = 20° C; quantities of fission-product and actinide oxides in waste initially are those from 460 kg of uranium fuel.

[a] Recommended by K. B. Krauskopf (Stanford University, unpublished data, 1982) at 20°C for mildly reducing conditions (Eh = -0.1 to -0.2 V); see Table 7-1. The solubility for SiO_2 is for amorphous silica from Fournier and Rowe (1977).

[b] The calculation assumes that the groundwater is free of all substances listed. The value shown is therefore an upper limit. Calculated from Equation (1).

[c] Based on data of McVay et al. (1981) for IAEA static leach test of borosilicate glass with periodic replacement of leachant. Original data are adjusted for the surface area of the canister on the assumption that release rates are proportional to the surface area exposed.

SOURCE: This table is similar to one in Chambré et al. (1982b).

Table 5-7, $\beta = 37\ m^{-2}$. Using the properties listed in Table 5-7, the limiting low-velocity fractional release rate for SiO_2 at 20°C is calculated to be 3.5×10^{-7}/yr at steady state, about a fourth the release rate calculated at 1 m/yr.

Measurements of the product ϵD_j for nonsorbing species in granite indicate values about 100-fold less than those used in the present calculation (Skagius and Neretnieks 1982).

Comparison with Laboratory Leach Data

Table 5-7 gives values of the fractional dissolution rates for silica and for various radioactive elements, calculated from experimental values of leach rates reported from IAEA-type laboratory experiments in which leachant is periodically replaced. Note that, for substances having limited solubility, the values of f_j computed from Equation (1) are smaller then several values derived from these laboratory leach tests. One must conclude, therefore, that for slightly soluble species in waste that has been embedded in a repository rock and is surrounded by Darcy flow, the slow diffusion and slow movement of the liquid around the waste containers may be more significant in controlling the net rate of dissolution than the rate at which the substances inside the waste material reach the surface of the waste form. If the solubility is very small, the rate of escape into groundwater will be determined primarily by the properties of the porous rock and the velocity of groundwater; if the solubility is sufficiently large, the kinetics of the interaction between the solid waste constituents and water may dominate.

The Effective Boundary Layer Thickness

The volume of water around a waste package has little significance because the dissolved constituents will be found in a liquid layer which streams past the package and extends for a very great distance downstream. Near the package the effective thickness of the liquid boundary layer that presents the resistance to diffusive-convective mass transfer is of the order of

$$(\pi D_j \epsilon R/U)^{1/2} = 1.2 \text{ cm},$$

for a pore velocity of 1 m/yr and using the numerical values given at the top of Table 5-7. Material leached from the waste does not accumulate in a closed volume of liquid surrounding the waste form as it does in the static laboratory experiments; it is carried away continuously at a volumetric rate that depends on both the diffusion rate and the water velocity. The chemical conditions surrounding the waste form are not necessarily those determined simply by the accumulation of leach products, as they are in the laboratory leach rate experiments. In the repository, depending on the access of groundwater to the waste-form surface, the near-field chemical conditions will also be determined by the flow rate of groundwater and by the chemical environment produced by materials through which the groundwater has passed.

Application to Repositories

Predicted solubility-limited fractional dissolution rates in Table 5-7 for the low-solubility species are much smaller than the fractional release rates estimated from laboratory leaching experiments wherein the concentration boundary layer resistance is either negligible or absent. This means that for a waste form in the repository environment, surrounded by groundwater in a porous rock, the rate of dissolution for these low-solubility species will be controlled by the rate of transport into the liquid boundary layer adjacent to the waste solid. For most important radionuclides in reprocessing waste, except cesium, the kinetics of the solid-liquid interactions are rapid enough that the concentration of the dissolved species in the liquid at the waste surface will be near or only a little below the solubility limits for these species. Because the kinetics of the solid-liquid interactions are not controlling for these low-solubility species, the laboratory leach rate data are irrelevant and are not useful for estimating the dissolution of low-solubility species under repository conditions.

From our analyses in Section 9.10.4 for wastes from fuel reprocessing, the very soluble cesium-135 is the only radionuclide important to long-term repository performance whose dissolution rate from glass may not be affected by solubility-limited dissolution, provided that cesium can dissolve faster than the waste matrix itself. Recent data (G. L. McVay, Pacific Northwest Laboratory, personal communication to J. R. Johnson, 1982) indicate that even cesium may have exhibited a solubility-limit effect in static leaching experiments on borosilicate glass for leaching times of the order of a year. If the dissolution rate of cesium-135 is not important to overall repository performance, as is suggested in Section 9.10.4, then the rate of dissolution or degradation of borosilicate glass itself would affect repository performance only to the extent that a redundant backup resistance to dissolution may be needed.

The higher repository temperatures during the thermal period can change the solubilities and increase the diffusion coefficients. These effects on diffusive-convective dissolution rates are considered quantitatively in Section 9.13.

Dependence on Waste-Form Material Properties

The waste-form material and its alteration products can affect the chemical forms and solubilities of the released and dissolved species. However, if the chemical species of radionuclides released to groundwater are the same as assumed in the calculations for Table 5-7, then the fractional dissolution rates of Table 5-7 would apply to any other waste form of the same surface area and waste loading, unless the waste matrix itself is so insoluble that its fractional release rate is less than that calculated for the solubility-limited radionuclides. This does not occur for borosilicate glass, nor is it predicted for SYNROC, but waste forms more insoluble than any of those now tested are still possible. The potential for such low-solubility waste forms is discussed further in Section 9.11.

Because the kinetics of the solid-liquid interactions in borosilicate glass are rapid enough for the low-solubility species to be at or near their solubility limits in the adjacent liquid of the repository environment, interior cracks in the waste solid, devitrification, and other such mechanisms that could increase the rate of the solid-liquid interaction would not be expected to affect the rate limited by liquid diffusion and convection.

The chemical and material properties of the waste form itself can affect the formation of stable precipitate layers (cf. Section 5.6.5) and colloids (cf. Section 5.6.7), which can affect the applicability of the solubility-limited diffusion-convection analysis.

Effects of Solubility Uncertainties and Colloids

The above analyses of the solubility-limited dissolution rates are useful for predicting the long-term release of radionuclides in a repository if the solubilities are known and if there are no other mechanisms that can release and transmit radioactivity to the biosphere. The solubilities of many of the species are sensitively affected by the local oxidizing-reducing conditions of the groundwater and the host rock, the acidity or alkalinity, the temperature, and the presence of complexing species such as anions and some organics. Some indications of these effects are given in Tables 7-1 and 9-5, and the effects of solubility uncertainties on repository performance are discussed in Section 9.20. It is important that the solubilities be known reliably for such analyses to be used as a basis for repository design.

For such low-solubility species it is possible that the rate of release to groundwater can be increased if colloids and suspended precipitates form at or near the waste surface (cf. Section 5.6.7). It may be possible for such suspended matter to be transported to the biosphere by groundwater as it flows through the porous or fractured media, in addition to the material transported in solution. If colloidal particles form, they may transport at the water velocity if the particles are smaller than the rock pores and if they are not sorbed (Allard 1982a,b). Otherwise, colloidal particles may be temporarily or permanently trapped in the pores (Avogadro et al. 1981, Eichholz and Craft 1980).

Conversely, if the reported solubilities include an appreciable amount of species such as colloids, there will be less in solution, and there will be a lower rate of diffusive transport into the concentration boundary layer than estimated in Table 5-7. The effective diffusion coefficients of colloids in liquids are very low, about 5×10^{-7} cm^2/s for colloids of size 5×10^{-6} cm (Thomason et al. 1982), even without tortuosity corrections (cf. Section 5.7.4). Therefore, because the diffusion coefficient of a colloid is small compared with that of solutes, only the solute fraction of the "solubility" will diffuse appreciably into the groundwater.

In view of the potential importance of the formation and transport of suspended matter containing low-solubility radioelements, this issue should be resolved.

Effect of Backfill and Effect of Other Geometries

Equations (1) and (2) assume that the waste form is in contact with porous rock containing groundwater. If there is a liquid-filled annular space between the waste form and the rock or backfill, as could occur by long-term corrosion of the waste-pack structure and waste form, the solubility-limited rate of mass transfer into the rock or backfill would remain the same, if there is not net convective flow through the annular space. The effect of a net convective flow within the annulus has not been analyzed. A low-permeability backfill (cf. Section 5.11.4) between the waste and the rock would remove this uncertainty, and this may become one of the most needed functions of backfill. For radionuclides with half-lives of a few thousand years or more, the dissolution rate would not be affected by sorption in the backfill and in the surrounding rock. A few centimeters of backfill of much higher porosity than the rock will not appreciably alter the result if convective flow through the backfill is not important.

The analyses that led to Equations (1) and (2) assumed that groundwater flows perpendicularly to the cylindrical axis of a waste package. Other flow directions are possible in a repository. Chambré et al. (1982a) have considered flow parallel to the axis and have shown that the predicted rates are not significantly different from those given by the above equations.

The Need for Validation Experiments

Although the predictive techniques of Equations (1) and (2) contain no unknown constants to be empirically determined, the techniques do need validation by carefully designed laboratory experiments. In planning such experiments, the time to reach the steady state conditions is important. The time-dependent solubility-limited dissolution rates have been derived by Chambré et al. (1982a). For a nonsorbing species whose decay during movement through the concentration boundary layer can be neglected, the "equilibration time" for the release rate to come within 1 percent of the steady state diffusion-controlled rate of Equation (2) is 310 years for the parameters given in Table 5-7. The time is much shorter, of the order of a few years, when the flow velocity is great enough to warrant use of Equation (1). For a sorbing species, with a retardation coefficient greater than unity, the equilibration time is proportional to the retardation coefficient of that species in the rock. Although these equilibration times are short compared to the times of interest for storage, they are long enough to be important in designing experiments to test the predictions of these boundary layer controlled rates.

The steady state conditions of Equations (1) and (2) can be reached within reasonable experimental times by choosing parameters different from those expected in a repository, and this is a reasonable approach for testing the predictive technique for mass transfer in liquids in porous media. Validation under simulated repository conditions may have to depend on careful tests of the time-dependent dissolution rates predicted from the more general theory that led to Equations (1) and (2).

Similar Approaches in Other Countries

A qualitatively similar approach toward the estimation of dissolution rates in a repository has been developed by Neretnieks (1978, 1982), who considered a waste package, including backfill, intersected by planar fractures in the surrounding rock. By estimating equivalent steady state transport resistances for the layers in the waste package and in the concentration boundary layer outside the package and assuming solution saturation at the waste-form surface, a rate of steady state release into groundwater flowing through the planar fractures is estimated. No laboratory data for waste-form leaching enter into Neretnieks' calculations.

Hughes et al. (1981) have also recognized the need to estimate dissolution rates in a repository as limited by diffusion in the surrounding groundwater, rather than attempting to extrapolate from laboratory leach data. They used the laboratory data to set an upper limit for the repository rate and computed a lower limit by assuming that the waste package is a sphere surrounded by motionless groundwater. They concluded that under all cases of practical importance it is groundwater access and solubility that limit the rate of solution of glass. Using their assumed properties of rock, glass contained in a canister of the size we have used for computations would last about 800,000 years. Their estimated rate of dissolution of silica is about a third that given in Table 5-7.

5.8. OVERALL EVALUATION OF BOROSILICATE GLASS AND ALTERNATIVE WASTE FORMS

Our overall evaluation of borosilicate glass (Sections 5.8.1 to 5.8.8) and our consideration of the need for continuing research and development on alternative waste forms (Section 5.9) are based on the foregoing sections of this chapter and the performance analyses discussed in Chapter 9.

5.8.1. Choice of the Principal Waste Form for Further Testing and for Repository Design

Borosilicate glass is the appropriate choice for further testing and for use in current repository designs. It is in the most advanced stage of waste-form development. Glass incorporating radioactive high-level waste has been manufactured in full-scale glass melters in the United States and abroad. It is capable of incorporating useful inventories of fission-product and actinide waste. Although glass may not be the ideal waste-form material, its specific properties are not critical and it will likely be adequate to meet our overall criterion for repository performance, based on the performance analyses in Chapter 9.

5.8.2. The Need for Further Testing

Although borosilicate glass is the most developed of all of the waste forms, there are uncertainties about its performance in a repository that need to be better understood before glass waste would be acceptable for emplacement in a repository. A high-level waste package will be acceptable for emplacement when its performance in a repository can be adequately predicted and when the predicted repository performance meets the overall performance criterion. There is not yet a validated technique for predicting the performance of borosilicate glass--or of any other waste form--in a repository. The predictions of waste-form performance for Chapter 9, based on solubility-limited dissolution (cf. Section 5.7.4.), depend little on properties of the waste form itself, but many of the uncertainties in these predictions can be affected by the waste material. Whatever technique the U.S. Department of Energy

(DOE) adopts for predicting waste-form and waste-package performance must be carefully validated before any waste form and waste package can be considered acceptable. This includes the testing of borosilicate glass itself under conditions devised to yield data for predicting performance in repositories. Until such testing is completed and validation obtained, borosilicate glass cannot be considered sufficiently proved to be acceptable for emplacement in a geologic repository.

5.8.3. Release of Radionuclides from Borosilicate Glass in a Repository at Ambient Temperature

Laboratory leaching tests show that some radionuclides, e.g., cesium-135 (cf. Table 5-4) and neptunium-237 (cf. Table 5-7), can be released from borosilicate glass more rapidly than would be desired in a repository. However, the rates of release from a borosilicate-glass waste package in a repository are estimated to be much lower than the laboratory rates because the rates of diffusion into slowly flowing groundwater may be limited by the small ambient-temperature solubilities of the long-lived radionuclides in the glass. On this basis, suitable long-term repository performance (cf. Chapter 9) would be achieved, in part, by the low solubilities of the individual radionuclides. These solubilities can be affected by borosilicate glass and by its alteration products. In this sense a waste form such as borosilicate glass can contribute to repository performance, even though the rates of release to the repository may not be determined by the interaction kinetics and retention properties of the glass itself. As discussed in Chapter 9, there are more uncertainties about the performance of a repository in tuff, and a waste package that releases radionuclides to tuff groundwater less rapidly than predicted in this study may be desirable.

5.8.4. Release of Radionuclides from Borosilicate Glass During the Period of Repository Heating

Borosilicate glass reacts more rapidly with hot groundwater, and there will be more rapid alterations and deterioration of its physical integrity. However, solubility-limited diffusion from the waste-package surface into the surrounding groundwater may control the rate of release of radionuclides during the thermal period and may not be affected by structural alterations in the glass. There are not sufficient data on the temperature-dependent solubilities of the important radionuclides to estimate their release rates during the thermal period, but if their temperature-affected release rates are similar to the rate predicted in Chapter 9 for silicon, then the repository performance may still meet the overall performance criterion even if borosilicate glass is exposed during the thermal period. There are more uncertainties in the predicted performance of borosilicate glass during the thermal period than in the later ambient period.

Uncertainties in the performance of borosilicate glass when exposed to leaching solutions at high temperatures may require that glass high-level waste be protected from groundwater by a corrosion-resistant overpack when repository rock is at temperatures greater than about 100°C (cf. Section 5.8.6).

5.8.5. Uncertainties in the Predictions of Waste-Form Performance

The present uncertainties in calculated radionuclide release rates from the waste package are more directly related to the quantitative aspects of the solubility-limited diffusion-convection predictions (cf. Section 5.7.4). Although the predictions in Chapter 9 indicate adequate performance of waste packages containing borosilicate glass in most of the repository concepts studied (cf. Sections 9.6 through 9.10), there is no experimental proof of these release rates, and experiments are needed. If it is found from properly designed laboratory tests that the release rates are much greater than now predicted, then a much better understanding of the release properties of borosilicate glass itself will be needed. This will include better understanding of the changes in the physical structure of glass that are expected in the long term at ambient temperature and that will occur more rapidly if glass is exposed to groundwater during the period of repository heating. New experiments and tests will then be necessary to develop and validate a means of predicting the performance of glass in repositories.

The present predictions of release rate do not take into account interactive phenomena such as the formation of colloids and other waterborne radionuclide particulates and their effect on release rate from the waste package. Experiments show that such particles are not readily transported through porous materials such as sand or porous rock, although the data are fragmentary. Further experiments and analyses are needed to indicate to what extent colloidal formation could increase the rate of release of radionuclides from a waste package in a repository.

5.8.6. The Need for a Corrosion-Resistant Overpack

The analyses in Chapter 9 suggest that it may not be necessary to protect borosilicate glass from hot groundwater, but present estimates of radionuclide release during thermal-period exposure have many uncertainties, particularly at the higher temperatures generated by 10-year-old waste. If further experiments and analyses indicate that it is unacceptable to expose such short-cooled glass waste to groundwater during the thermal period, and if it is confirmed that borosilicate glass is adequate during the ambient period, the glass should be protected by a corrosion-resistant overpack, or the temperature should be lowered by reducing the waste loading in each waste package, by longer preemplacement storage, by removing cesium and strontium during reprocessing, and by greater spacing between emplaced waste packages.

5.8.7. When Must Uncertainties be Resolved?

There is considerable time to resolve the uncertainties in the performance of borosilicate waste packages during the thermal period. The first geologic repositories in the United States will be initially loaded with waste that has already cooled for many decades prior to emplacement. The temperatures encountered by these wastes will be much lower than for the 10-year-old waste. A large backlog of long-cooled discharged fuel is waiting to be reprocessed, and there will be several decades before a commitment must be made for the even later emplacement of 10-year-old waste in a repository. There is time to establish the validity of the solubility-limited release predictions and, if necessary, to clear up the uncertainties in the behavior of borosilicate glass at higher temperature. There is time to assess the need for an overpack or for any of the alternatives for reducing the temperature. A systems analysis should be performed to evaluate the effects of the system parameters that affect the thermal-period performance of glass. The plans for repositories should include identification of expected temperatures during the early and later periods of waste emplacement, and there should be developed a corresponding schedule of necessary decisions on options for the 10-year-old waste that may eventually be emplaced.

It is more important now to resolve the uncertainties in the long-term performance of borosilicate glass under ambient conditions. As has been discussed in Chapter 9, radionuclides released during the post-thermal period may be the most important contributors to individual radiation doses, and there are fewer alternatives for solving the post-thermal performance problem than there are for the thermal period. Emphasis should be given to the development of an adequate technique for predicting performance of waste packages in a repository and to the validation of that technique and its prediction.

5.8.8. Additional Information Needed on Borosilicate Glass

The DOE program for borosilicate glass should be redirected toward developing information necessary for adequate prediction of the long-term release of radionuclides from glass waste packages in a repository. One or more methods that appear promising for such prediction should be selected, and development and testing should be focused toward proving the methods and providing the necessary data to make the predictions. If the solubility-limited diffusion-convection analysis of Section 5.7.4 is selected as one of the methods for testing and proof, the following are needed:

o Measure in laboratory experiments the rates of diffusive-convective mass transfer of solutes from cylindrical and spherical surfaces to groundwater in simulated porous and fractured media, with and without liquid annuli and backfill. Determine the effective coefficients for steady state diffusion and confirm with field measurements.

o Measure the temperature-dependent solubilities of radionuclide solutes in the aqueous environment that is expected to exist at the outer surface of borosilicate-glass waste in a repository. The measurements should include the effects of other materials that will coexist with and may interact with glass in the repository environment.

o Determine under simulated repository conditions the mobility of radionuclide colloids and particulates formed on the surface of borosilicate glass and in backfill and the extent to which they can be released into groundwater in surrounding porous and fractured media.

o If colloids and particulates are found to be sufficiently mobile to be released appreciably to flowing groundwater, determine the amounts of the radionuclide colloids and particulates expected to be formed and released, including the effects of temperatures, temperature gradients, and exposure to liquids and vapor expected during the period of repository heating. Determine the effective release and near-field transport mechanisms of the radionuclide colloids and particulates and develop methods of predicting the long-term rates of such releases. Experiments should then also be initiated to determine the parameters that affect the far-field transport of radionuclides released in colloids and particulates.

o Identify those repository situations wherein appreciable air in groundwater can be expected, determine which radiation (gamma or alpha) is expected to be dominant when air or nitrogen is present, and determine the effect of radiolysis on the radionuclide solubilities.

o If the waste package includes an overpack, perform failure-mode tests and analyses on the effects on overpack leak tightness due to mechanical, thermal, and chemical stresses.

o Perform laboratory and field tests to confirm the predictability of the long-term release of radionuclides from glass waste packages.

It will also be useful to continue general and fundamental research on borosilicate glass, to improve the understanding of the phenomena that are important to its performance in repositories. Even here, priorities should be given to studies of those phenomena expected to control or significantly affect long-term radionuclide releases to groundwater in a repository.

5.8.9. Continuing Research and Development on Alternative Waste Forms

It appears that there are at present no substantial development programs within the Department of Energy that are concerned primarily with alternative waste forms. A continuing program should be supported for the following reasons and including the following objectives.

To provide against the contingency that release rates or uncertainties in the range of such rates prove to be considerably higher than presently estimated, alternative waste forms should be developed to the point where their merits can be more realistically evaluated than is presently possible. Such waste forms offer the possibility of releasing contained radionuclides at the qualitatively lower rates that are predicted when release is controlled by diffusion within a solid.

Furthermore, if the low release rates estimated by the theory of solubility-limited diffusion-convection are verified by appropriate experiments, then comparably low rates may be expected from some other waste-form candidates that have heretofore been considered to be relatively leachable. A more realistic assessment of leachability may affect some of the presently prescribed processes for fabricating alternative waste forms.

Even though present waste forms may be demonstrated to be satisfactory, the magnitude of the national program for geologic disposal, its extended time scale of development, design, and implementation, and the potential advantages of better waste packages justify a continuing backup technical program to develop better understanding of the main elements of geologic disposal and to provide even better waste-package alternatives that could be used in later stages of waste emplacement. Desirable options include waste forms that can accommodate higher loadings of radioactive waste per package and operate reliably at the resultant higher temperatures, that allow greater emplacement density in the repository, that provide smaller surface-to-inventory ratios, and that offer the low solubilities and low release rates that can theoretically result when such rates are controlled by solid diffusion.

5.9. CHOICE OF METALS FOR CANISTERS AND OVERPACKS

In some waste-package designs the waste form in its metal canister is surrounded by an "overpack" metal container, whose purpose is to prevent the waste form from contacting groundwater during the period of repository heating, i.e., several hundred years. In this section we discuss the data relevant to the selection of metals and alloys for use in canisters and overpacks.

5.9.1. General Corrosion Considerations

Corrosion mechanisms of interest here include selective leaching of elements out of an alloy, general corrosion (gradual removal of material from the surface or formation of a surface oxide that flakes or spalls off), pitting corrosion, crevice corrosion, stress corrosion cracking, and corrosion fatigue.

General corrosion is typified by the rusting of steel or cast iron, whereby iron is converted to the oxide or hydroxide and then spalls off due to thermal or mechanical stress, impact, or abrasion. Such corrosion is expected for canisters and overpacks of cast steel or cast iron.

Pitting is associated with formation of local electrolytic cells between alloy matrix and secondary phases, typically carbides in steel or intermetallics in aluminum, etc. Pitting is more severe at grain boundaries than within grains. Carbides in stainless steel often lead to pitting and may advance to trenching of entire grain structures in severe cases. This does not take place, however, in homogenous alloys.

Crevice corrosion is a preferential, local attack in areas that have been damaged mechanically, such as a residual crack in a weld or the area around a rivet or washer, or around surface defects where a local electrolyte can change as corrosion proceeds. The attack is rapid but quite local.

Stress corrosion cracking, as the name implies, is the combined result of an aggressive atmosphere and high local stresses. The swelling of the waste form, for example, could introduce high surface stresses in the canister and cause stress corrosion cracking. Welding, which leads to locked-in residual stress, often causes stress corrosion cracking unless it is done properly or annealed.

Once induced, a crack pattern, often intergranular, progresses fairly broadly and spreads into a large number of cracks in a branch pattern. Catastrophic failure is the result. Fatigue or cyclic stressing due to thermal or mechanical stress often leads to stress corrosion cracking.

5.9.2. Overpack and Canister Alloys

In limited tests, several titanium alloys have been reported to have excellent corrosion resistance in salt, brine, and deionized water. Ticode 12 in particular, a simple alloy of titanium, 0.8 percent nickel, and 0.3 percent molybdenum, has performed extremely well, as was expected (Molecke et al. 1981). Ticode 12 is a tough and ductile alloy with excellent corrosion resistance to water, brine, and salt even at 250°C. Other titanium alloys could be selected to enhance other properties if desired. Recent work (G. L. McVay, Pacific Northwest Laboratory, personal communication to J. R. Johnson, 1982) shows generation of hydrogen from hot brines (90°C) sufficient to hydride 5 to 10 percent of the original alloy in 50-day tests, resulting in embrittlement.

In the experiments by Molecke et al. (1981) on Ticode 12 some hydrogen uptake occurred from a galvanic corrosion source. Current work with radiolysis as a hydrogen source will examine this potential problem further.

Titanium alloys could be used as the metal matrix for encapsulation of calcined wastes; titanium alloys can as readily be used as canisters. Titanium is probably unsuitable for pouring and solidifying molten glass because of the reactivity of titanium at the temperatures required.

Certain types of alloys are preferred if canisters are to be used to contain molten borosilicate glass. For example, 20Cr-25Ni stainless steel, type 317L (an Fe-16Cr-12Ni-4Mo-low C alloy); Hastelloy X (22Cr-18.5Fe-9Mo-0.1C-bal Ni); and C-276 (a nickel-chromium alloy containing high molybdenum) are all excellent candidates, especially for salt and brine exposure. Prepared by rapid-solidification powder-metallurgy techniques, corrosion and mechanical properties should be superior to those of current ingot-based alloys.

The high-level waste disposal program in Sweden (cf. Section 5.10.5) contemplates lead-titanium overpacks for reprocessing waste and thick copper overpacks for spent fuel.

The final choice of metals for the overpack, if an overpack is required, may be determined by results of future experiments in a properly simulated repository environment, wherein the overpack is subjected to reactions with canister metals, to reactions with the repository groundwater, or to mass transfer phenomena. It is important that such experiments be carried out if an overpack is to be depended on in the waste-package design.

5.9.3. Cast-Steel and Cast-Iron Overpacks

Heavy sections of cast steel and cast irons have been suggested by several sources. Thick steel or iron cylinders can provide sufficient attenuation of gamma radiation from the enclosed radioactive waste to enable direct handling of the waste package during emplacement, and they can provide structural support to resist external loading in the repository. Data supporting the slow corrosion of these materials come from the observation that old cannon have survived exposure to salt water on ocean bottoms for more than 100 or 200 years with limited corrosion. However, these same materials rust and spall severely when exposed to air. In sufficiently heavy sections they would be expected to survive for a long time when fully immersed in nonaerated water but would be in difficulty if partially immersed. Effects of steam exposure for decades in a hot repository during resaturation could be severe. The presence of metallic iron may enhance corrosion of glass waste forms, via colloid precipitation, as shown by McVay and Buckwalter (1982) (cf. Section 5.6.3).

With heavy castings of steel or iron for canisters and/or overpacks, there would be major problems in welding to produce leak-proof joints. The large casting would present shipping, moving, and handling problems. Finally, large castings of steel or iron free of significant porosity, cracks, or other defects are difficult to make.

5.9.4. Overpack Summary

From the test data currently available it is clear that the best results by far have been obtained with titanium alloys, particularly Ticode 12. Depending on the nature of the storage environment, it is possible that cupro-nickel alloys may be useful too. They would be cheaper than titanium alloys, would be resistant to corrosion in brine, and would be adaptable to the manufacturing techniques used in the production of total waste packages.

5.10. DESIGNS OF PACKAGES FOR HIGH-LEVEL WASTE

The waste package must be tailored not only to each waste type but also to the specific rock medium in which it is to be emplaced. The U.S.

Department of Energy and its contractors are developing conceptual designs of waste packages to meet the evolving regulatory criteria for disposal of spent fuel (SF) high-level waste, commercial high-level waste (CHLW) from reprocessing fuel discharged from light-water reactors, defense high-level waste (DHLW), and transuranic (TRU) waste in three rock types: salt, basalt, and tuff. The expected repository conditions are described in Chapter 6. The present waste-package designs are based on using borosilicate glass for the defense and commercial reprocessing wastes and compacted fuel pins for spent fuel waste. The principal specifications adopted by Westinghouse Electric Corporation (1981) for these design studies appear in Table 5-8. The waste-package designs are most advanced for salt and basalt.

5.10.1. Waste Packages for a Salt Repository

Westinghouse Electric Corporation (1981) has developed conceptual waste-package designs for two alternative methods of emplacement:

1. Emplacement in vertical boreholes in the floor of a mined repository.
2. Horizontal emplacement on the floor of the mine using a package self-shielded by a thick overpack.

With the borehole package, all operations require shielding or remotely controlled equipment, whereas the self-shielded package allows most of the handling and emplacement operations to be performed with little or no additional biological shielding. In some designs of the borehole concept, the overpack is a thin corrosion-resistant barrier made of titanium alloy (Ticode 12). The self-shielded approach relies on a thick iron or steel overpack to serve both as a corrosion barrier and as a radiation shield. Designs for defense high-level waste packages are illustrated in Figures 5-6 and 5-7. The conceptual packages for commercial spent fuel and reprocessing waste are similar to those for defense high-level waste, differing only in the physical dimensions of their components. A summary of the principal features of each reference package is presented in Table 5-8.

The packages are designed for long-term containment of commercial or defense reprocessing waste that has been processed into borosilicate glass sealed in 304L stainless steel canisters, and for spent fuel that has been disassembled into fuel pins, with pins from a number of fuel assemblies placed in a single package. In the case of borehole packages, a relatively thick overpack reinforcement fabricated of carbon steel is used to resist the 16.2-MPa lithostatic pressure from the salt. Corrosion protection may be provided by a 0.25-cm-thick shell of Ticode 12 around the overpack reinforcement. Although stabilizers or fillers in the annular gaps were considered for enhanced heat transfer and rigidity, they were not incorporated because of fabrication problems. Although crushed salt was used to backfill the top of the emplacement hole, no additional backfill around the package was planned because of the small quantity of brine that would accumulate from the

TABLE 5-8 Features of Reference Waste-Package Designs for Salt

	Borehole Package			Self-Shielded Package		
Feature	DHLW	CHLW	SF (PWR)	DHLW	CHLW	SF (PWR)
Waste form/canister/stabilizer						
Waste form	glass	glass	compacted pins	glass	glass	compacted pins
Waste weight (Mg)	1.47	0.6	2.77	1.47	0.6	3.69
Canister diameter (m)	0.61	0.32	–	0.61	0.32	–
Canister length (m)	3.0	3.0	–	3.0	3.0	–
Power when emplaced (kW)	0.42	2.2	3.3	0.42	2.2	4.4
Stabilizer	none	none	none	none	none	none
Overpack						
Overpack o.d. (m)	0.83	0.475	0.60	1.25	1.16	1.4 (square)
Reinforcement thickness (m)	0.095	0.057	0.08	0.31	0.405	0.47
Ticode 12 thickness (cm)	0.25	0.25	0.25	none	none	none
Overpack length (m)	3.34	3.38	0.42	3.9	4.0	5.5
Package weight (Mg)	8.8	3.2	9.5	31.7	28.8	76.9
Backfill	none	none	none	none	none	none
Emplacement thermal loading (W/m²)	13.8	37.5	12	15.2	37.5	18.8

SOURCE: Westinghouse Electric Corporation (1981).

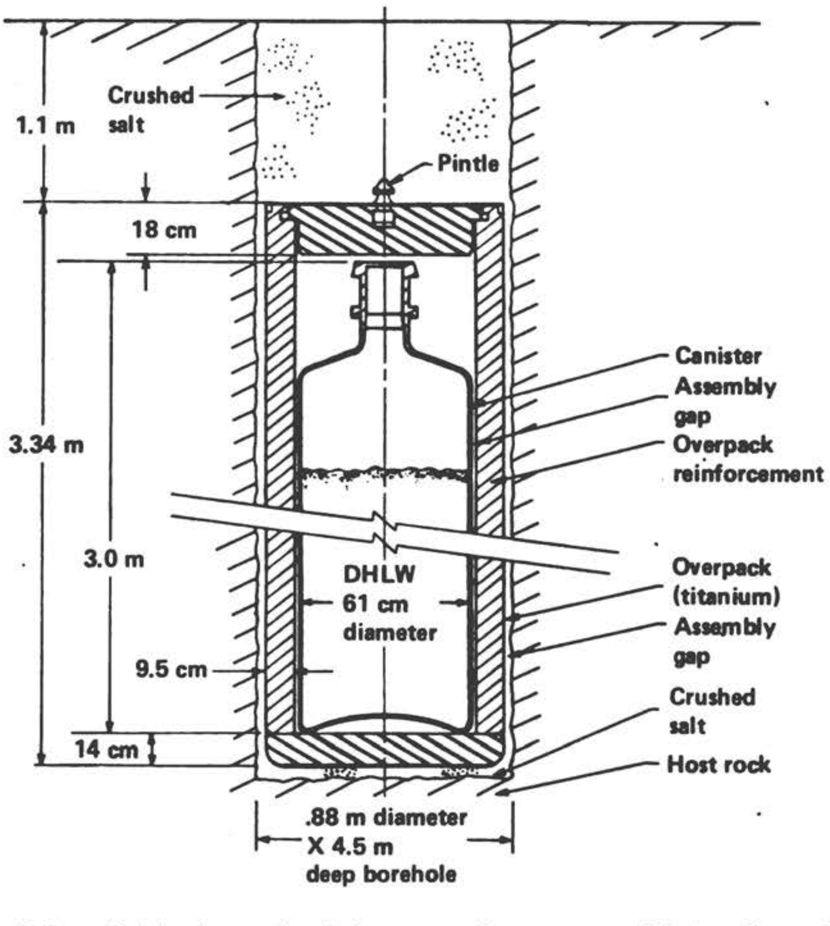


FIGURE 5-6 Defense high-level waste borehole waste package conceptual design. Source: Westinghouse Electric Corporation (1981).

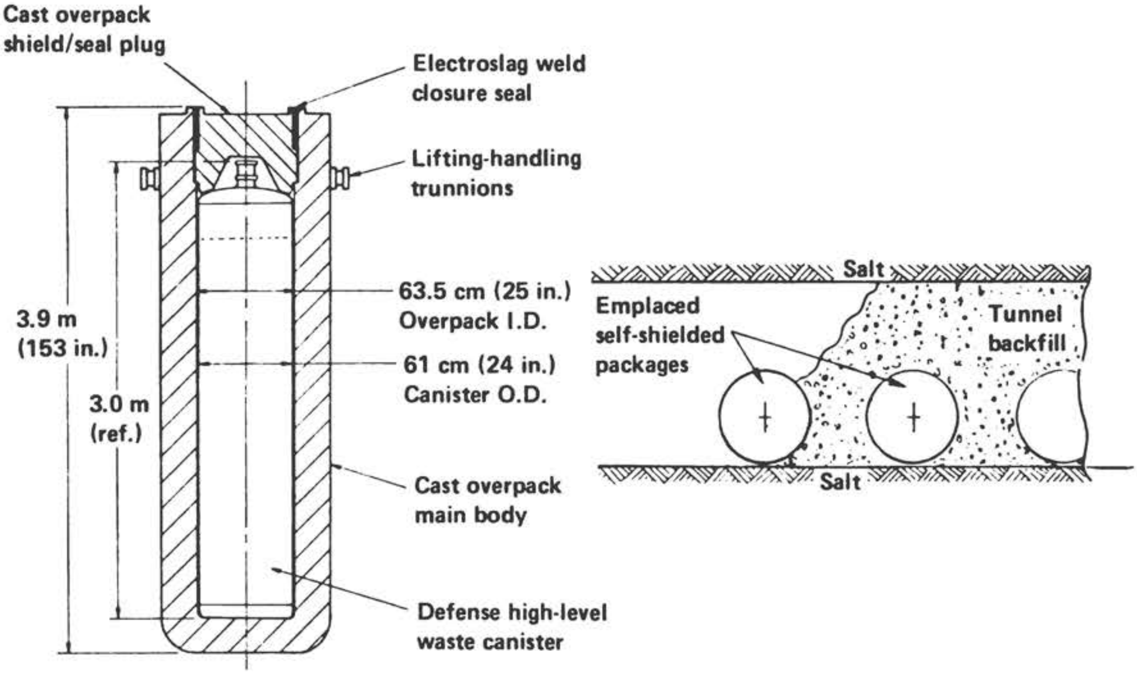


FIGURE 5-7 Defense high-level waste self-shielded waste package conceptual design. Source: Westinghouse Electric Corporation (1981).

thermally induced migration of brine inclusions in the salt. Retrievability is to be aided by including a removable top portion of the overpack.

The principal component of the self-shielded package concept is the overpack, a thick-walled cast steel or cast iron closed vessel that serves as a corrosion barrier, provides protection from internal pressure and from lithostatic loading, and attenuates radiation to levels in the range of 5×10^{-4} to 10^{-3} Sv/hr at the outer surface. These features offer potential advantages for horizontal storage and contact handling during emplacement and retrieval, but they are to some extent countered by the size and mass of each package.

Westinghouse found that the calculated performance of all of the above designs met the criteria and requirements specified by DOE. Alternate designs were developed that approach the thermal limits more closely, but ultimate optimization of the packages is to be done later in conjunction with constraints of fabrication and overall repository design.

5.10.2. Waste Packages for a Basalt Repository

The Basalt Waste Isolation Project (BWIP) at Hanford is now considering horizontal boreholes, with several packages in each borehole. A sketch of a defense high-level waste package for basalt is shown in Figure 5-8

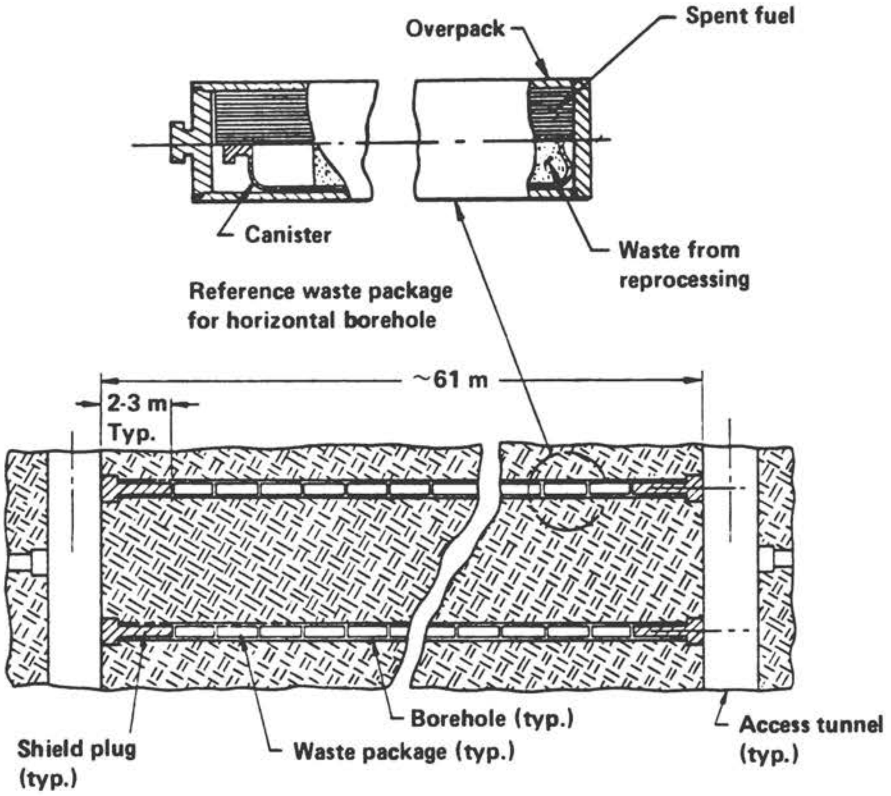


FIGURE 5-8 Borehole type package for horizontal emplacement in basalt. Source: Westinghouse (1982a).

(Westinghouse Electric Corporation 1982a). Self-shielded concepts were also considered, but no analysis of their performance has been provided. Each borehole is to be closed with a shield plug. To meet the waste-form temperature limit for spent fuel, only half as many fuel pins (792 versus 1,584) are contained in each spent fuel package as in the specifications listed in Table 5-8. The canisters for defense and commercial reprocessing waste are to be fabricated of 304L stainless steel, and those for spent fuel are of carbon steel. In all cases the canisters are used only to contain the wastes during handling and shipping and are not assumed to aid in meeting waste-package containment requirements after emplacement. Fillers or stabilizers are not used.

5.10.3. Waste Packages for a Tuff Repository

Preliminary conceptual designs of waste packages for emplacement in tuff (Westinghouse Electric Corporation 1982b) are similar to those described above for a salt repository, but stabilizers and backfills, other than crushed tuff, are not used.

5.10.4. Evaluation of the Performance of Waste-Package Designs

The present waste-package design studies (Westinghouse Electric Corporation 1981, 1982a,b) reflect engineering analysis of the mechanical and structural integrity of the waste package when subjected to stresses in fabrication, shipping, and emplacement and under the thermal, hydraulic, and lithostatic stresses imposed by the heated repository. The overpack materials are selected and specified on the basis of their ability to resist corrosion for 1,000 years. Radiation shielding is also considered.

The present design studies apparently do not yet include analyses of the ability of the waste package, or of the waste-package and associated backfill, to meet some release rate requirement, such as the numerical criterion of less than 10^{-5}/yr after 1,000 years that has been proposed by NRC. As noted in Chapters 8 and 9, the panel has reservations about both the need and the sufficiency of the NRC criterion when applied uniformly to all of the radionuclides identified as important by EPA and NRC, and the panel has reservations about the ability of the present waste-package designs to meet this proposed criterion for some of these radionuclides. Comments on the ability of backfill to help meet the NRC's proposed release rate criterion appear in Section 5.11.4. However, based on the performance analyses in Chapter 9 and the assumptions therein, these waste-package designs seem adequate to meet the overall performance criterion of 10^{-4} Sv/yr individual dose adopted for the purpose of this study.

Regardless of whether or not the NRC's presently proposed release rate criterion is adopted, it is important that the performance of the waste package and backfill to aid in achieving some overall performance criterion of the geologic waste-disposal system be carefully analyzed as part of the process of designing the waste package.

We do not yet find any substantive analyses of the failure modes of the waste packages, including considerations of the local stresses and "metalline" corrosion that seem possible when a hot waste package, in an initially dry environment, is subjected to the influx of groundwater during water intrusion and resaturation after the emplacement region is sealed.

5.10.5. Waste Packages for a Granite Repository in Sweden

A comprehensive study of waste packages for disposal in granite was done in Sweden to fulfill the Swedish government's requirement for a demonstration of how "absolutely safe" disposal of high-level waste can be effected. The designs for spent fuel and reprocessing waste were reviewed within Sweden and internationally (National Research Council 1980) and were found to meet the government's requirements. The following description of the package designs is from a summary by Ahlstrom (1979).

The waste packages were designed for borehole emplacement, with waste loading per package less than half that in the U.S. designs. It was assumed that the wastes were stored at the surface for 40 years, rather than 10 years as in the U.S. designs, to reduce radiation levels and heat generation rates prior to emplacement. The maximum temperature rise of the granite is predicted to be about 50°C, and the maximum waste-form surface temperature for reprocessing waste is specified to be 85°C or less (R. Pusch, University of Lulea, personal communication to T. H. Pigford, 1982).

The overpack for the reprocessing waste is a 10-cm-thick lead vessel sheathed with 6 mm of titanium alloy. The lead is to reduce the radiation level and the radiolysis of the groundwater, and the titanium is to provide a corrosion life of at least 10,000 years in anoxic groundwater. If the titanium sheath fails, penetration of the lead by corrosion is estimated to require about 4,500 years. Bentonite and sand is to be used to backfill the emplacement boreholes (R. Pusch, University of Lulea, personal communication to T. H. Pigford, 1982).

Disassembled pins of spent fuel are placed directly in a thick copper overpack that provides radiation shielding and corrosion resistance, estimated to last more than 1 million years. The space around the fuel pins is filled with lead for internal stability and to enhance heat transfer. Blocks of highly compacted bentonite backfill the emplacement holes. The compacted bentonite has a very low conductivity for water (2×10^{-14} m/s). Its swelling pressure of about 5 MPa when wet is expected to seal any minor cracks in the backfill and in the rock around the package. The repository rooms and tunnels are to be backfilled with a mixture of 20 percent bentonite and 80 percent sand.

5.11. BACKFILL AS A BARRIER IN THE WASTE-ISOLATION SYSTEM

5.11.1. Backfill Functions

Most of the repository designs show backfill material packed between the outer metallic layers of the waste package and the surrounding rock. In the terminology of the U.S. Nuclear Regulatory Commission (1982), discrete backfill is considered to be an integral part of the waste package, and the waste-package criteria proposed by NRC (cf. Section 5.2.1) are intended to include the backfill. Several possible functions of the backfill material have been suggested.

- o Assist or enhance the ability of the waste package to isolate radioactive wastes.
- o Function independently as a barrier to radionuclide migration should failure of the waste-package occur.
- o Serve as a plastic stress-adjustment medium to avoid localized stresses on the remainder of the waste package that could otherwise result from direct mechanical contact with surrounding rock.
- o Enhance heat transfer from the waste form to the surrounding rock.

The programs of DOE and NRC seem to emphasize the first two funtions above, but we find some uncertainty as to what can be reasonably expected from the backfill material in controlling the release of radionuclides from the waste package.

Candidate backfill materials include bentonite, montmorillonite, illite, and other smectite clays; zeolites; crushed host-rock materials such as basalt, granite, salt, or tuff; ferrous-iron minerals to aid in promoting low-solubility reducing conditions; quartz sand for heat transfer; and others.

R. Pusch (University of Lulea, personal communication to T. H. Pigford, 1982) reports that the backfill properties of zeolites are inferior to those of smectite clays with regard to hydraulic conductivity, plastic behavior and self-healing, and resistance against cementation and embrittlement.

Not only must the backfill survive the thermal period of repository heating, but it must also not add significantly to the temperature of the inner components of the waste package.

Reports and publications on backfill that were available to the panel do not contain analyses of the need or effectiveness of backfill in distributing the loads from the surrounding media to avoid stress concentrations on the inner components of the waste package. We are not able to evaluate the importance of backfill to fulfill this function.

5.11.2. Temperature Rise Across Backfill

One of the earliest proposals for using bentonite backfill as a barrier was made in Sweden (Jacobson and Pusch 1977). Highly compacted bentonite was proposed for borehole emplacement of unreprocessed fuel rods. For a package of reprocessing waste, the greater heat generation rate per package led to a modified backfill composed of 80 to 90 percent quartz, with the balance bentonite, to improve thermal conductivity. Sweden's program contemplates 40 years of preemplacement storage to keep the local rock temperatures well below 100°C. Different problems arise when applying this concept to the U.S. designs, wherein the repositories are to be capable of accommodating 10-year-old waste.

A backfill of 30.5 cm of bentonite and sand, with a thermal conductivity of 0.75 W/m°C, is specified in recent designs (Westingouse Electric Corporation 1981, 1982a,b) of waste packages for spent fuel and reprocessing waste for a salt repository. Calculations by Claiborne (1982) show that this thickness of backfill results in a temperature rise across the backfill of about 120°C for a typical 10-year-old waste package generating 2.16 kW at the time of emplacement, as shown in Figure 5-9. By comparison, the calculated temperature rise from the rock to the 0.3-m waste canister, in the absence of backfill and with no water in the annular space, would be less than 100°C, showing that for such thickness the backfill will not aid heat transfer but instead will act as an insulator.

Other values of the backfill thermal conductivity are possible, depending on the materials used and the extent of water intrusion. High-density blocks of pure bentonite with a thermal conductivity greater than 1 W/m°C have been formed under high isostatic pressure (Ahlstrom 1980), although the effective conductivity is expected to be lower after emplacement. Temperature rises comparable to those shown in Figure 5-9 can be expected for backfill of similar conductivity in other

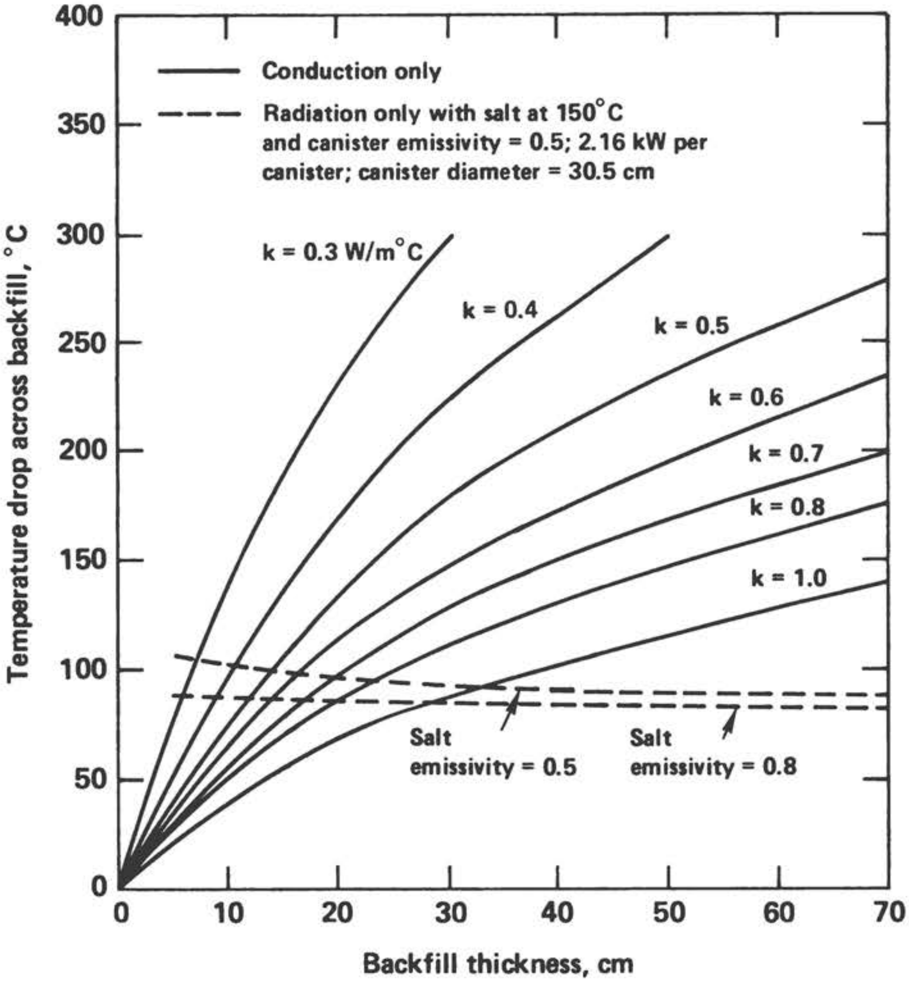


FIGURE 5-9 Temperature drop across backfill as a function of thickness and thermal conductivity. Source: Claiborne (1982).

rock media, but the temperature drop across the annulus in the absence of backfill will be small after the repository is resaturated. Clearly, the use of thick backfill in the U.S. designs is bought with a penalty of significantly higher temperatures of the inner components of the waste package.

A backfill such as bentonite cannot keep the inner components of the waste package dry once a wet-rock repository has resaturated. Assuming that the waste package employs a corrosion-resistant overpack, as proposed by NRC to protect the inner waste form from contacting water during the thermal period, a thick backfill will result in the overpack's contacting moisture at considerably higher temperatures, with corresponding increase in corrosion rate. If the overpack is not used, then the corresponding temperature rise across the backfill is applied to the waste canister in contact with water, and subsequently to the exposed waste form within the canister (cf. Section 9.13).

5.11.3. Sorption Retardation in Backfill

Arguments for backfill, especially in thick sections, have been based on the calculated benefits from sorption of radionuclides in the backfill material. Sorption can retard the rate of transport through the backfill and delay the appearance of sorbing radionuclides at the outer surface of the backfill. In this sense, backfill sorption adds to waste-package containment for that period of time, after the beginning of waste-form dissolution, for dissolved radionuclides to diffuse through the backfill and be released to the surrounding rock and groundwater. Nowak (1979) applied the time-dependent equations of diffusional transport through the backfill, in the absence of radioactive decay, and estimated a "breakthrough" time of 10^4 years for a backfill thickness of 0.3 m, assuming a retardation coefficient* of 4×10^4. The breakthrough time is proportional to the retardation coefficient, so for no backfill sorption, i.e., a retardation coefficient of unity, the breakthrough time would be as short as 3 months. A threefold increase in backfill thickness to 0.9 m results in a tenfold increase in the backfill containment time.

Calculations similar to those of Nowak have been made by Ahn et al. (1982b) for backfills of synthetic minerals such as zeolites or titanates, of assumed thicknesses from 0.3 to 60 m. The results show that a 1-m-thick backfill could provide total containment for a period of up to 1,000 years, based on the ambient-temperature properties assumed in the calculation.

Whether a thick, sorbing backfill can be justified on the grounds of delaying the release of sorbing radionuclides to the surrounding rock and groundwater is uncertain. A time delay of a few hundred to a few thousand years in the release to groundwater does not reduce the calculated radiation doses to the individual from long-term hydrogeologic transport (cf. Section 9.8), assuming constant fractional release rates. The temperature-affected release rates that would result if waste solids were exposed to groundwater at the higher rock temperatures during the thermal period may not significantly increase the calculated radiation doses to the individual (cf. Section 9.13). This challenges NRC's justification for its proposed 1,000-year

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*The retardation coefficient is defined as the ratio of groundwater pore velocity to the velocity at which the radionuclide concentration moves through the porous sorbing material. In the absence of groundwater motion, it can be thought of as the total amount of a radionuclide in a small representative sample of solid plus pore liquid to the amount in the liquid portion of that sample. For local chemical equilibrium of the radionuclide distributed between the liquid and porous solid, the retardation coefficient K is given by:

$$K = 1 + \rho K_d (1 - \varepsilon)/\varepsilon,$$

where ε is the porosity, ρ is the solid density, and K_d is the distribution constant, expressed in cubic centimeters per gram.

corrosion-resistant overpack. However, if it is assumed that NRC's proposed 1,000-year containment is to be fulfilled, redundant containment for most--but not all--of the radionuclides could be obtained by thick, sorbing backfill, but the seeming redundancy would be compromised by the much higher temperatures of the overpack and the waste solid, and the time at elevated temperature would be greater than without a thick backfill.

5.11.4. Effect of Backfill on Radionuclide Release Rate

The second claimed feature of backfill is to control and reduce the rate of release of radionuclides from the dissolving waste form to the disturbed zone outside the backfill. From their calculations of transient diffusion of long-lived radionuclides through sorbing backfill, Ahn et al. (1982a) conclude that the backfill alone can meet a fractional release rate requirement of 10^{-5}/yr (cf. Section 5.2.1) for zeolite backfill greater than 3 m thick. Dayal et al. (1982) conclude that the 10^{-5}/yr release rate criterion will be met by zeolite backfill of 1 to 3 m thick. These results, together with those of Nowak, are quoted by the U.S. Nuclear Regulatory Commission (1982) in support of its conclusions that a multibarrier waste package including such backfill can meet its release rate criteria for all important radionuclides.

Apart from the obvious temperature problems of such thick backfill around heat-generating waste, these calculations do not show that the sorbing backfill can appreciably affect the release rate of long-lived radionuclides after breakthrough has occurred. After moisture has penetrated the backfill and caused corrosion of the waste form and dissolution of radionuclides, and after the time delay for these dissolved radionuclides to diffuse to the outer surfaces of the backfill, there will exist a very long period of essentially constant rate of dissolution from the waste form and diffusion through the backfill. For example, adopting Ahn's estimate of 1,000 years for the breakthrough time for a 1-m zeolite backfill, fractional release rates of the order of 10^{-5}/yr or less will result in dissolution times of the order of 10^5 years, long enough for quasi-steady-state diffusional transport through the backfill. For those radionuclides whose concentrations at the inner surface of the backfill are at the solubility limits, as seems likely for most of the important long-lived radionuclides in borosilicate glass and other waste forms, and if the repository temperature has relaxed to the ambient condition, true steady state diffusion will occur for each radionuclide species after its concentration band has penetrated the backfill.

The effect of sorption disappears when steady state is reached. The "effective diffusion coefficient" adopted by Nowak (1979) and Ahn et al. (1982a,b) is the diffusion coefficient in the pore liquid divided by the retardation coefficient. It is applicable to the transient analysis of breakthrough time, but not to steady state diffusion. Thus, for those radionuclides whose half-lives are long enough to survive the breakthrough times calculated by Nowak (1979) and Ahn et al. (1982a,b), the release rate is neither determined by nor affected by the backfill sorptive properties they considered. If one were interested in the average release rate to the outer rock, averaged over a time that includes the time for first penetration through the backfill, then the fractional release rate results quoted by Ahn et al. (1982a,b) and Dayal et al. (1982) would be relevant. However, that is not the definition of release rate adopted by NRC or adopted in this study. The predicted reduction in release rate from backfill sorption is not correct.

However, backfill can aid in obtaining a low release rate by preventing net convective flow of groundwater in the annulus between the other components of the waste package and the borehole and by maintaining a low-porosity region from the inner surface of the rock to the surface of the waste-package components (cf. Section 5.7.4). For these functions the backfill need be only as thick as the gap between borehole and waste-package components necessary for emplacement. The thinner backfill results in more reasonable waste-package temperatures.

An annulus between the waste form and backfill can later develop as the waste form corrodes. An attractive backfill candidate for preventing net convective flow through this annulus is a smectite-rich clay, which excludes water flow under the low regional hydraulic gradients expected during the ambient period (R. Pusch, University of Lulea, personal communication to T. H. Pigford, 1982).

Our conclusions concerning the function of the backfill are consistent with the calculations by Neretnieks (1982), who finds that backfill does not present an appreciable resistance to the long-term release of radionuclides that have dissolved from the waste form.

Chambré et al. (1982a,b,c) have analyzed the time-dependent diffusion of radionuclides through backfill layers. When radioactive decay is included in the analysis, backfill sorption can affect the steady state release rate from the waste package. However, the sorption effect is important only for relatively short-lived radionuclides. Sorption saturation of backfill can considerably reduce the breakthrough time for sorbing radionuclides.

The delays in release possible from a thick, sorbing backfill do not necessarily reduce the inventory of some of the radionuclides. Calculations by Pigford et al. (1980) show that a thick, sorbing backfill provides a delay that will increase the concentration of the decay daughters thorium-230 and radium-226 within the backfill layer, thereby increasing the later release rate of radium-226 to the surrounding rock. Consequently, a thick, sorbing backfill could make it difficult to meeting NRC's proposed release rate limit for radium-226 (cf. Section 8.6.7).

5.11.5. Chemical Effects

It is also necessary to consider the chemical effects of the backfill as a component in the repository materials system. The groundwater chemistry can change through ion exchange, e.g., potassium in the groundwater for sodium in a bentonitic backfill. As reported in Section 5.6, sodium leached from glass waste forms is taken up by the bentonite backfill and the leach rate is increased. Release of other ions could be affected in a similar way.

In addition to bentonite, other backfill materials have been proposed or studied. Alkaline earth oxides are shown to provide a more durable water shield for the waste form. Once water penetrates, however, the calciferous backfill can have a negative effect because it can raise the pH of the leachant.

Bentonites will lose some or all of their expected sorption properties if they are overheated in the thermal period of the repository. Bentonites can also be a source of alumino-silicate colloids that can intermix with radionuclides released from the waste form (see Section 5.6.7).

It is important that the performance of backfill be evaluated realistically as a component of the waste-isolation system.

5.12. TRANSURANIC WASTE

Transuranic wastes are currently defined by DOE (U.S. Department of Energy 1982) as relatively dilute wastes containing greater than 3,700 Bq (10^{-7} Ci) of alpha emitters (i.e., transuranics) per gram of material. These wastes exist as waste organics, paper, metals, etc., containing the transuranic radionuclides, usually in very small amounts. The large volume of these dilute wastes must be reduced by incineration and physical/chemical decontamination of contaminated parts. The ashes, salts, and sludges can then be incorporated in waste-form materials described for high-level waste or in bitumen.

Experiments by Bida et al. (1980) indicate that concrete, borosilicate glass, or bitumen may not be adequate for concentrated transuranic wastes. Radiolysis may be severe in concrete, except for low-water types such as FUETAP. Leach test experiments at the Pacific Northwest Laboratory on borosilicate glass containing large concentrations of transuranics indicate fractional leach rates greater than 10^{-4}/yr in brine and deionized water at 90°C. It is likely that both glass and concrete materials can be substantially improved for transuranic waste forms.

If the loadings of transuranics in glass are made similar to those now considered for high-level waste, or if solubility limits govern, then glass or FUETAP concrete may be suitable form materials for transuranic wastes. The releases of actinides from transuranic glass waste should be no greater than from high-level glass waste, if the extra chemical diluents in the present transuranic glass waste samples are removed.

During the long-term operation of a repository at ambient temperature, radionuclides released from transuranic wastes are comparable in importance to those released from high-level waste, and the waste forms must be of comparable integrity and performance for both wastes. In Chapter 9 it is shown that when radionuclide release is controlled by waste-form dissolution rate, two important contributors to radiation dose in the environment are radium-226 and its decay daughter lead-210, resulting from the decay and transport of uranium-234, uranium-238, plutonium-238, americium-242m, and curium-242 in the waste (cf. Figure 9-2). Uranium-238 controls the radiation dose for water travel times much greater than about 10,000 years, and the other precursors control for shorter travel times.

In Table 5-9 are listed the amounts of precursors of radium-226 and lead-210 in high-level and transuranic wastes from reprocessing 10^5 Mg of uranium fuel from light-water reactors, based on data in Chapter 4. The potential amount of radium-226 and lead-210 that can be formed from uranium-234, plutonium-238, americium 242m, and curium-242 in the waste is only threefold greater in high-level waste than in transuranic waste. For repositories with longer water travel times, the potential amount of radium-226 and lead-210 formed and released from transuranic waste is about twice as great as that from high-level waste. Therefore, comparable emphasis must be given to controlling the long-term releases in both high-level and transuranic wastes. However, because the transuranic waste does not generate heat, it can be loaded into a cooler region of the repository and need not have whatever thermal protection is provided for the high-level waste form. If borosilicate glass is adopted as a high-level waste form, it is also a logical candidate for transuranic wastes.

5.13. SEPARATED RADIONUCLIDE WASTES

The panel reviewed the possible waste forms and packages for krypton-85, carbon-14, iodine-129, and tritium, which could be separated and recovered in fuel reprocessing. For the purpose of this study, it was assumed that these separated wastes will be disposed of in a geologic repository.

5.13.1. Krypton-85

A regulation of the U.S. Environmental Protection Agency requires that U.S. commercial fuel reprocessing plants recover krypton-85 from fuel that is irradiated after 1982. Release will be limited to 1.8×10^{15} Bq/GWe-yr, so that more than 80 percent of the krypton-85 in reprocessed fuel must be recovered and stored. The recovered krypton-85 can be stored in pressurized cylinders, or it can be immobilized by ion implantation in metal matrices, e.g., nickel-lanthanum alloys, or by trapping the gas in pores of zeolites or porous glass sintered in an atmosphere of krypton-85. Ion implantation is the currently proposed method to be scaled up to a pilot plant.

Early experimental results indicate that krypton-85 will be well contained by any of these processes. Possible limitations include (1) pressure cylinders must be stored in the low-temperature region of a repository; (2) losses of krypton-85 from zeolites or glass are estimated to be smaller than 10^{-4} after 10 years at 300°C, but processing at high temperatures (700°C to 900°C) and high pressure (ca. 100 MPa) is required; and (3) the metal matrix for ion implantation has been recently developed only on a small scale.

It appears practical to recover and store krypton-85. The required storage time is of the order of a century, a relatively short time for consideration in geologic isolation. Sea disposal of bottles of krypton-85 is an alternative that should be considered. At present, however, the London Convention on sea disposal of radioactive waste allows only solid waste to be disposed of in the ocean (Benedict et al. 1981).

5.13.2. Carbon-14

Carbon-14 appears in the reprocessing off-gases as CO_2, which can be fixed as $BaCO_3$ by reacting with solid or slurried barium hydroxide (Holladay and Haag 1979). Alternatively, the CO_2 can be collected on molecular sieves prior to the $Ba(OH)_2$ reaction. The $BaCO_3$ is then mixed with concrete to make the final waste form.

No leach data are available on carbon-14 waste forms, but substantial degradation would be expected in about 10^4 years, inferred from data for cement forms containing iodine-129. Analyses in Section 9.7 indicate that carbon-14 releases within this time period may result in individual radiation doses near or exceeding the panel's overall performance criterion for some repositories. In Section 9.10.2 we discuss calculations of the dissolution performance of a possible waste package for carbon-14, consisting of cold-compacted or granular calcium carbonate (N. J. Grant, Massachusetts Institute of Technology, personal communication to T. H. Pigford, 1982), with sufficiently low overall surface-to-inventory ratio to reduce the dissolution rate.

Sea disposal of carbon-14 in a solid form is an alternative that should be considered.

5.13.3. Iodine-129

Fission-product iodine is recovered in fuel reprocessing by absorption or adsorption from the gases evolved when fuel is chopped and dissolved. The iodine can be later distilled off and permanently fixed on silver-loaded adsorbents, such as zeolites, silica, or alumina (Benedict et al. 1981, Thomason et al. 1982). Alternate waste forms for iodine-129 that have been considered in the U.S. program include Portland cement, glasses, sodalite, organic materials, and zeolites. The most leach-resistant forms are cements containing silver iodide or iodates of barium, calcium, strontium, or mercury. Data indicate leach times of a few hundred to a few thousand years for iodates in cement to possibly 10^5 years for silver iodide in cement or in sodalite (Burger et al. 1981). The longest of these leach times is less than 1 percent of the half-life of iodine-129.

Thermal instability of iodine compounds limits the use of high-temperature processes in making waste forms. Hydrolysis in cement forms of the iodates or their possible reduction in the reducing waters of geologic repositories both lead to more soluble iodine compounds.

Because of the long half-life of iodine-129--1.7 x 10^7 years--and because iodine released from waste forms is usually assumed not to be appreciably sorbed and retarded as it undergoes hydrogeologic transport through the media surrounding the repository, it will be difficult to prevent most of the iodine-129 from reaching the biosphere, where it must then be diluted by flowing surface water to avoid appreciable radiation doses to future humans. However, a calculation discussed in Section 9.10.3 indicates that by incorporating silver iodide into waste packages with sufficiently low ratio of surface area to iodine inventory, and assuming that dissolution rate is controlled by solubility and diffusive-convective transport (see Section 5.7.4), an appreciable fraction of the iodine-129 may be retained in the waste form long enough for radioactive decay. However, the effects of ammonium and chloride ions on the solubility of silver iodide introduce serious uncertainties.

If dilution turns out to be the most effective long-term process for protecting future humans from released iodine-129, then sea disposal is an alternate that should be considered.

5.13.4. Tritium

Although fission-product tritium is now released to the atmosphere at reprocessing plants, technology is being developed for tritium recovery in such plants (Benedict et al. 1981). However, the process is complex and expensive. Because the required storage time for tritium to decay is about a century, storage as containers of tritiated water could be considered. Alternatively, any solid that contains firmly bound water may be suitable as a solid waste form for tritiated water, including drying agents such as silica gel, molecular sieves, and calcium sulfate, as well as cement and organic polymers. Most experience is with cement.

Although concrete is a monolithic solid, it is quite porous. In contact with water, about a third of the tritium will be released in the first month, mainly by isotopic exchange. The release may be retarded by coating the cement. Because of the relatively high leachability, cemented tritiated water may have to be stored in gas-tight cylinders.

In view of the relatively short half-life of tritium and of the enormous isotopic dilution possible with natural hydrogen, sea disposal is an alternative for dealing with tritium.

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6

CONCEPTUAL DESIGN OF REPOSITORIES

6.1. INTRODUCTION

The major factors and design features affecting repository systems performance, which are discussed in this chapter, are as follows:

- proposed repository layouts, including shafts
- proposed methods of excavation and stabilization and the design basis
- proposed shaft excavation and sealing procedure
- mechanical factors influencing the extent of the disturbed zone around repository openings
- influence of temperature on short- and long-term performance
  - --proposed maximum temperature and areal thermal loading criteria for different rock types and their bases
  - --potential for thermal cracking and increased permeability in the rock and rock mass surrounding the repository
- retrievability in hard rock and salt
- subsidence/uplift criteria
- restrictions due to tectonic environments

6.2. REPOSITORY LAYOUT

Conceptual (or "preconceptual") designs have been developed in the United States for excavated underground repositories in salt (both domal and bedded) and in basalt. Repositories in other candidate rock masses, such as granite and competent tuff, are expected, in principle, to be similar in design to the one proposed for basalt.

As an example of a conceptual repository layout, Figures 6-1 through 6-5 illustrate the main features of the preconceptual design developed by KE/PB (a joint venture of Kaiser Engineers and Parsons, Brinckerhoff, Quade and Douglas) for a repository in basalt (Schmidt 1981). The repository would be developed as a set of storage panels, each dimensioned for one year of waste receipt. Each panel has six storage rooms in the form of tunnels that are connected by safety crosscuts, as shown in Figure 6-3. The tunnels are widely spaced (37 m center to center) so that the possible influence of one storage tunnel on the

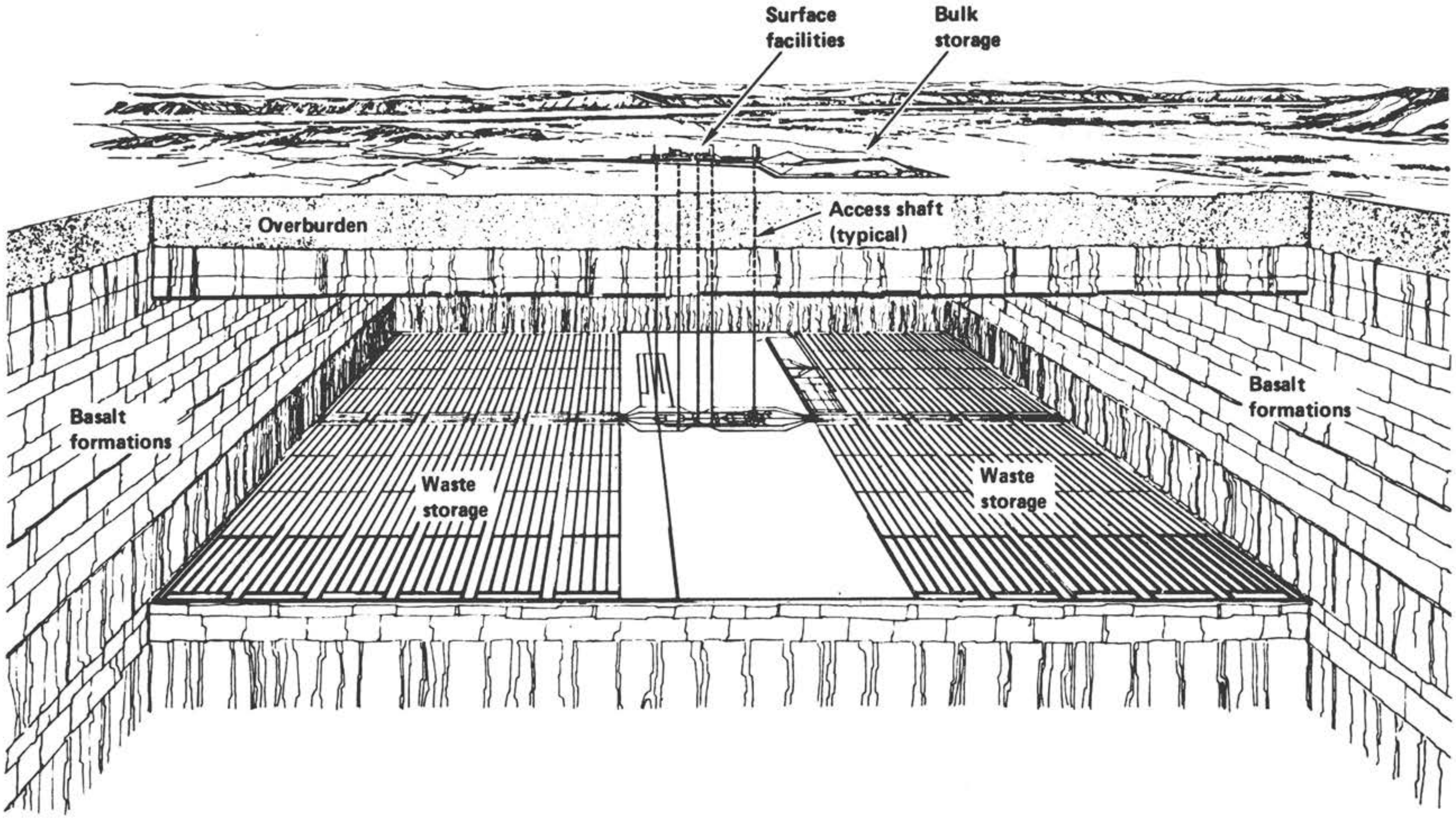


FIGURE 6-1 Cutaway perspective of a conceptual repository in basalt. Source: Schmidt (1981).

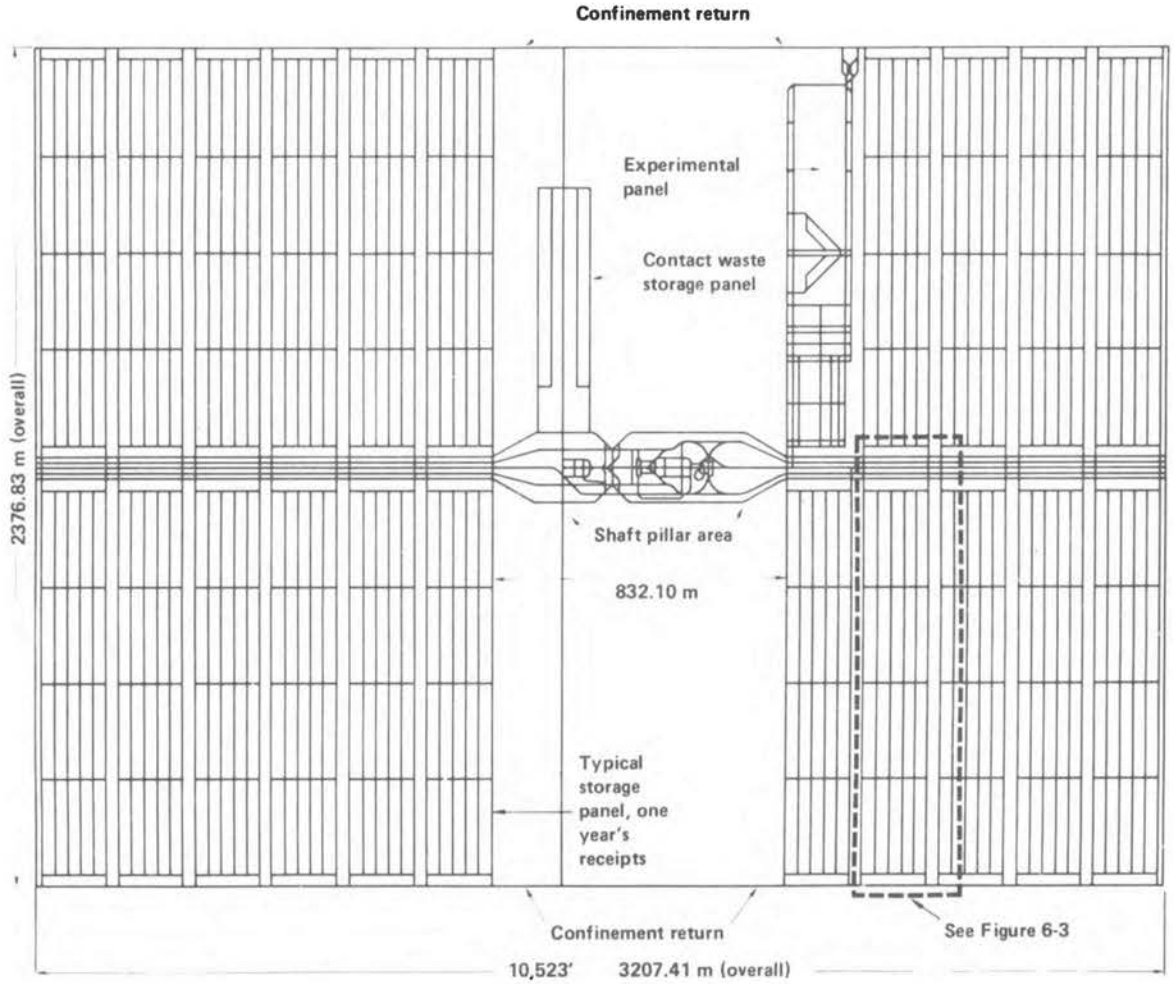


FIGURE 6-2 Underground facilities layout of a conceptual design for a repository in basalt. Source: Schmidt (1981).

stability of adjacent tunnels is minimized through the use of wide pillars. This design approach is important for all rock types, but particularly so for repositories in salt, as emphasized in the Waste Isolation Pilot Plant (WIPP) Progress Report (National Research Council 1981a).

Figure 6-5 shows the rather complex layout of the shaft pillar at repository depth. This complexity is mainly caused by the need for different openings to transport waste versus muck, men, and materials and to ensure a separation of the ventilation of openings under development (mine supply air and mine exhaust air) and ventilation of openings containing waste (confinement supply air and confinement exhaust air). In principle, this has to be attained regardless of geological host medium.

It should be noted that the complexity in the shaft pillar area at repository depth is not a cause for concern in terms of construction unless the ground conditions are quite adverse with regard to excavation

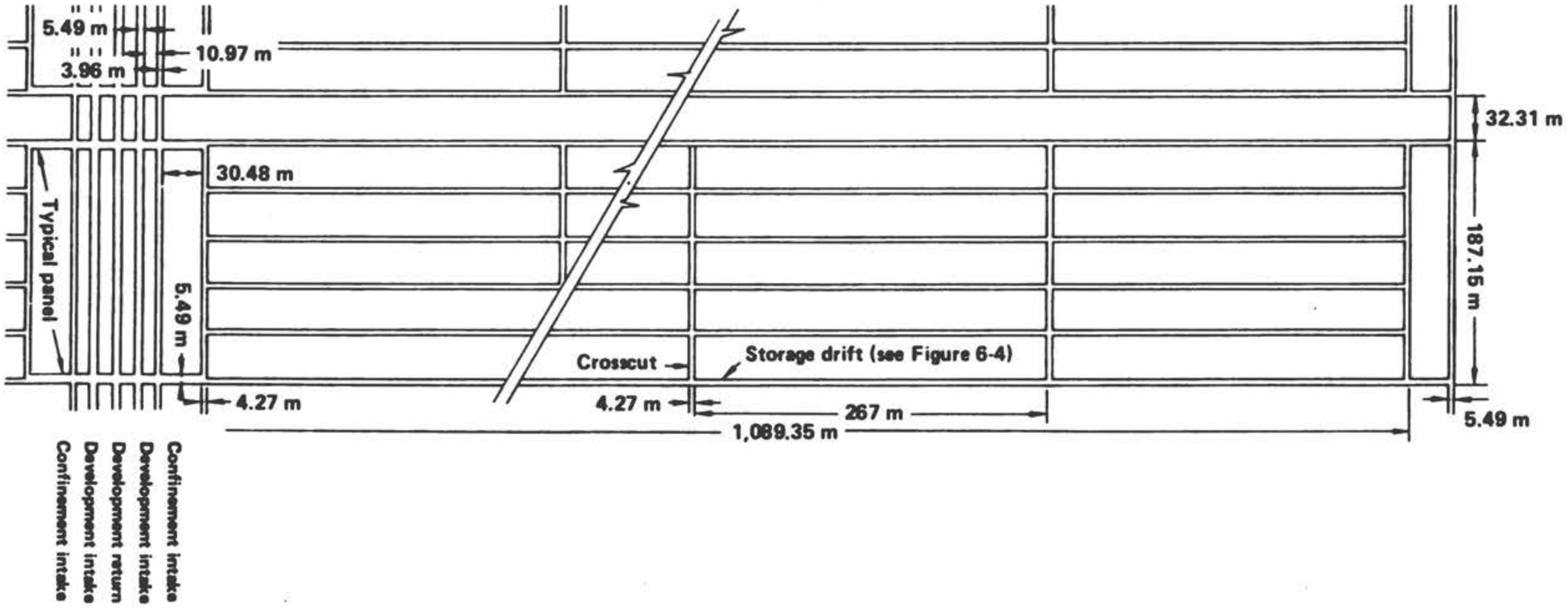


FIGURE 6-3 Typical storage panel in a conceptual design for a repository in basalt. Source: Schmidt (1981).

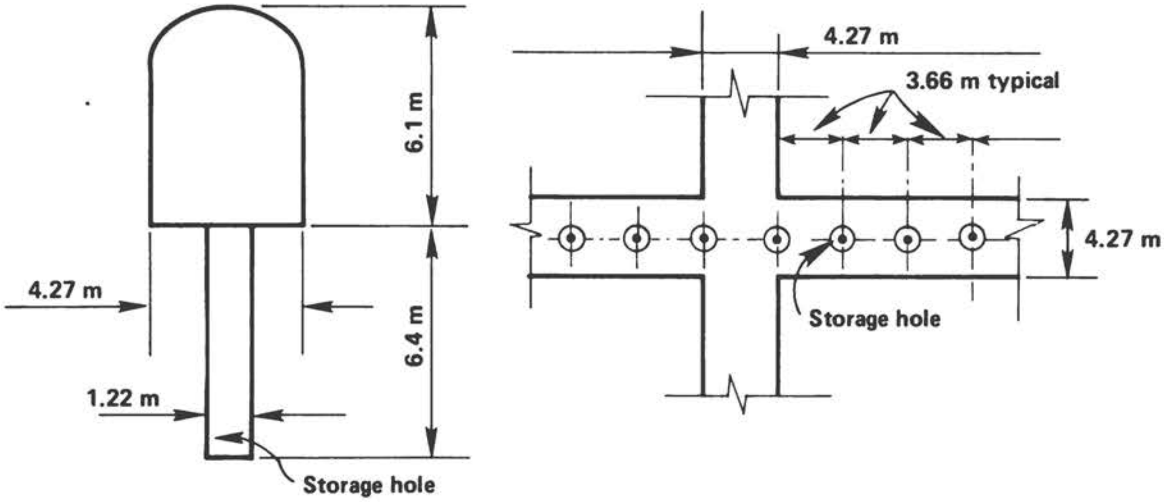


FIGURE 6-4 Room cross sections and plan in a conceptual design for a repository in basalt.
Source: Schmidt (1981).

and stabilization. Complex systems of underground openings have been successfully excavated. For example, several hundred underground hydroelectric power plants have been constructed, often involving complex geometries in the area adjacent to the power house. Many underground mines also have very complex geometries.

Repository layouts do not vary significantly as a function of rock types. For example, the perspective view of the proposed Hanford repository in basalt (Figures 6-1 and 6-5) is very similar to that shown for a bedded salt repository (Figure 6-6). One notable difference is the position of the shaft area or pillar. For one design the shafts are offset from the main storage rooms by approximately 610 m, as opposed to a central location for the other design. This difference does not appear to be dictated by basic rock properties, but it reflects different design efforts.

6.2.1. Excavation and Stabilization

The method of excavation will influence the extent of rock mass disturbance around the underground openings. Every effort should be made to minimize this disturbance, as it is detrimental to the permeability of the rock nearest to the opening. Mechanical excavation will minimize this disturbance as compared with conventional methods of excavation (i.e., drill and blast). Mechanical excavators have long been employed in salt mines. Thus, no evidence has been presented that new excavation technologies are needed for repositories in salt.

Full-face tunnel-boring machines (TBMs) have been successfully used for excavation in sedimentary rock (e.g., in Chicago, Illinois), regional metamorphic rock (e.g., in Washington, D.C.), and in some igneous rock such as granite (e.g., Kerckhoff 2 Hydro Project, California).

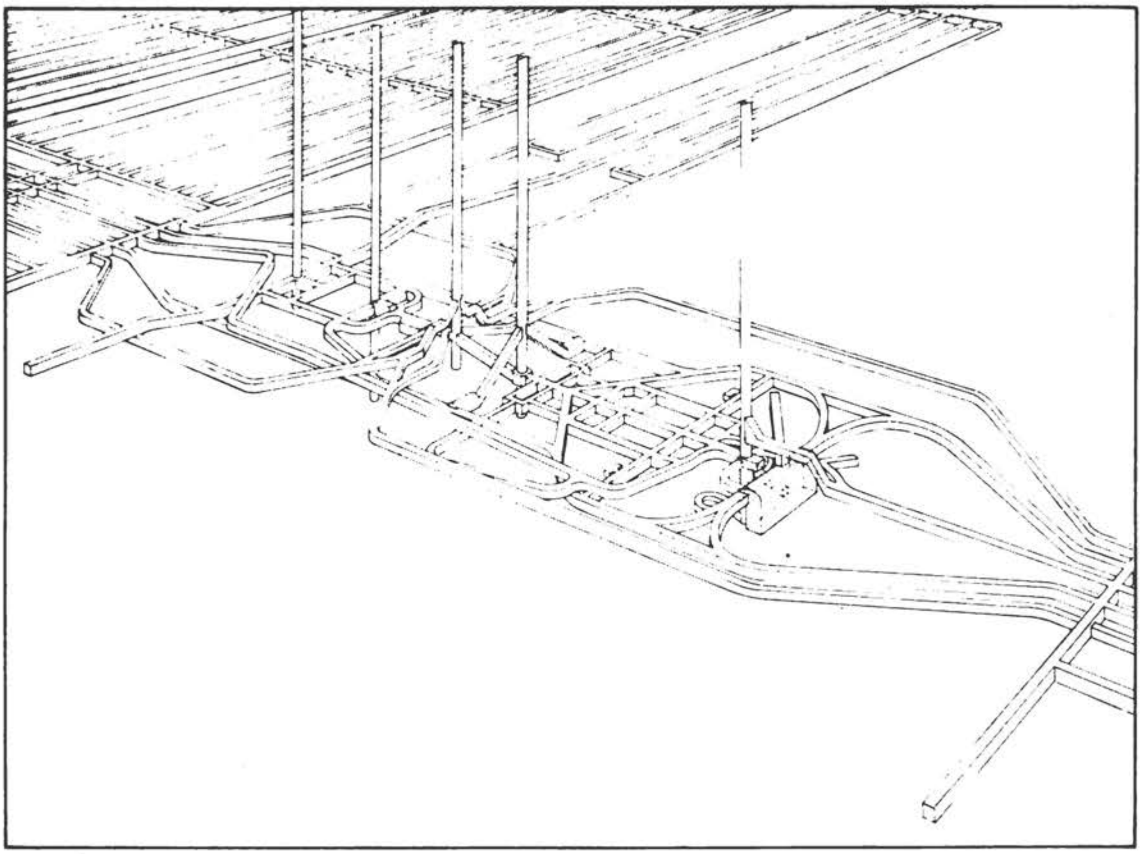


FIGURE 6-5 Shaft pillar arrangement in a conceptual design for a repository in basalt. Source: Schmidt (1981).

The preconceptual design for basalt is premised on the use of drilling and blasting for repository development. It has been assessed, at least at this stage, that the candidate rock type is too hard or strong for an economic employment of TBMs. The rapid improvement in TBM cutter technology in recent years indicates that the use of TBMs could become a viable option for basalt. Certainly, it seems appropriate that TBMs be used in excavation of critical tunnels that are to be plugged, such as the tunnels between the shaft complex and the repository proper (see Figure 6-5).

In terms of needed stabilization measures, one can again draw on existing technology as used in civil and mining projects. The measures at hand include fully grouted rock bolts, with or without wire mesh, unreinforced or reinforced shotcrete, steel sets embedded in shotcrete and/or concrete, and concrete lining. The sinking of shafts through aquifers can be carried out using freezing (temporary stabilization) or grouting, as proposed for the site at Hanford. Albeit very costly, these methods are technically sound. During the study of the Hanford site characteristics, the panel became aware of the rather extensive discing that has been observed in the cores from deep drill holes (Myers and Price 1979). Discing is the failure of rock core during drilling to

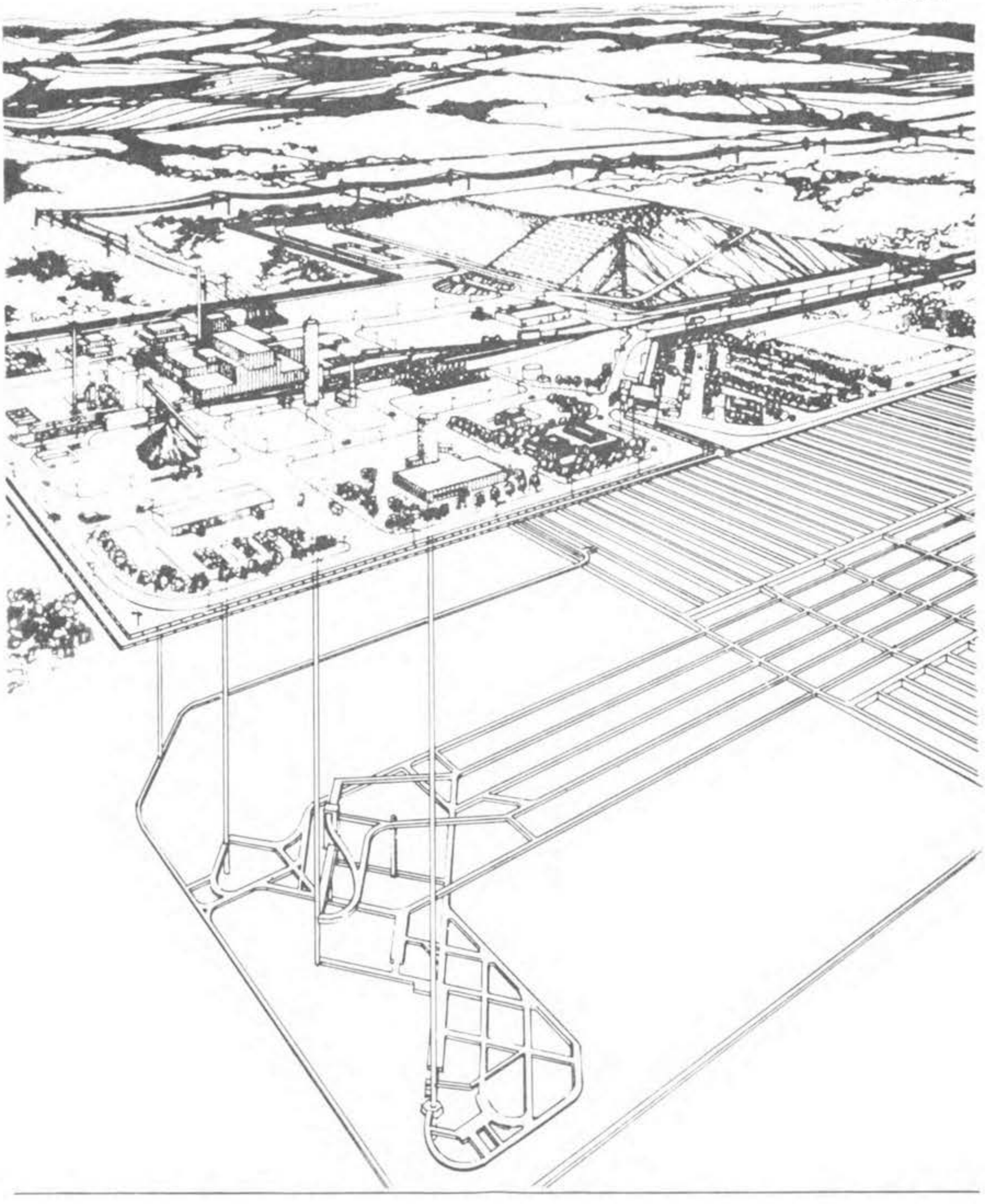


FIGURE 6-6 Perspective view of an NWTS conceptual bedded-salt repository. Source: Ritchie et al. (1979).

form nearly flat discs or chips from what would otherwise be more continuous lengths of core. It often occurs in brittle, strong rock where the in-situ stress field is relatively large. Discing is generally recognized as a warning of possible difficult tunneling conditions, e.g., rock bursting or slabbing. More important than the construction-related problems of overstressed rock near tunnel openings is the adverse influence of rock failure on the extent of the disturbed zone around these openings.

A summary of the results of a preliminary evaluation of the discing phenomenon at Hanford conducted by Rockwell is given in Table 6-1. This evaluation does not show a positive correlation with drilling effects, nor does the discing appear to be related to localized rock structural conditions, to interflow structures in the basalt, or to petrography. Thus the phenomenon is most likely caused by high in-situ stresses. Figure 6-7 shows a core with some moderate discing; Figure 6-8 shows a core with intensive discing. As previously indicated, discing in nonlaminated, brittle rock is ascribed to excessive and anisotropic stresses relative to the rock strength (Obert and Stephenson 1965). At Hanford the regional tectonics indicate a high compressive thrust in approximately the north-south direction. Hence, the horizontal stress in this direction may well be the cause of the discing.

It is not known what the behavior of jointed basalt will be during excavation under such in-situ stress conditions (and it will not be known until excavations and in-situ experiments are carried out at the proposed repository depth). It is likely, however, that some rock burst phenomena could ensue, having a significant influence on design and construction of a repository, as well as on the extent of the disturbed rock zone around tunnels and shafts.

It is the understanding of the panel that the cause and design consequences of the core discing are now being addressed. We note, however, that the phenomenon has been known for some time (Myers and Price 1979).

TABLE 6-1 Data on Core Discing Phenomenon

Factor	Information Study Results
Mineral lineation	No relationship with any mineral lineation
Drilling method	
Bit pressure	Insufficient data
Mud pressure	Insufficient data
RPM	Insufficient data
Bit changes	No effect
Bit types	No effect
Vibration	No effect
Localized phenomenon	None, occurs virtually in every deep hole
Relationships with interflow structure	No discing in flow top; no appreciable difference in entablature and colonnade in Umtanum
Petrography	No correlation in Umtanum; insufficient information in other flows
Mechanical property	No data on disced cores due to problem with samples

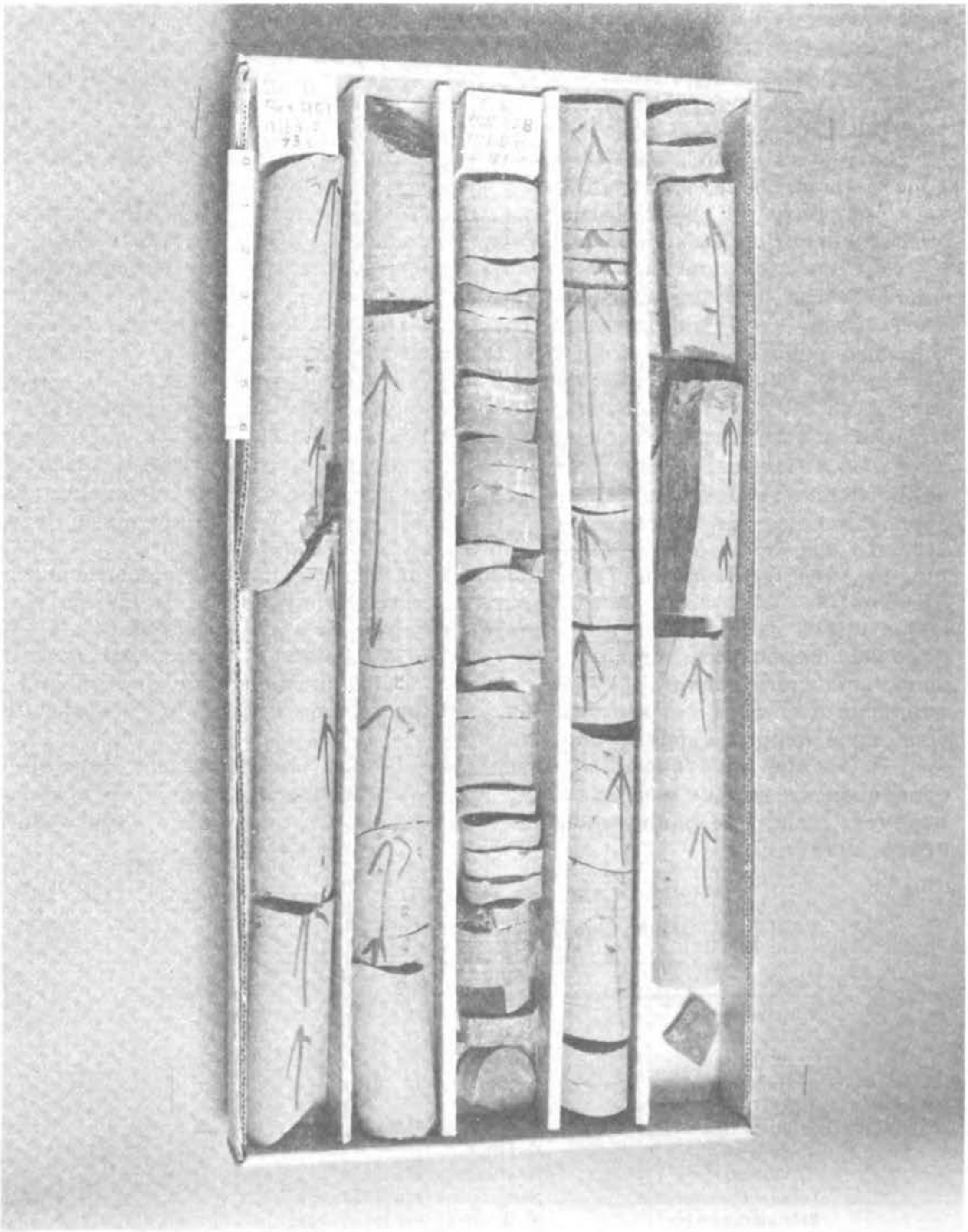


FIGURE 6-7 Basalt core from Hanford–moderate discing. Source: Courtesy of Rockwell International.

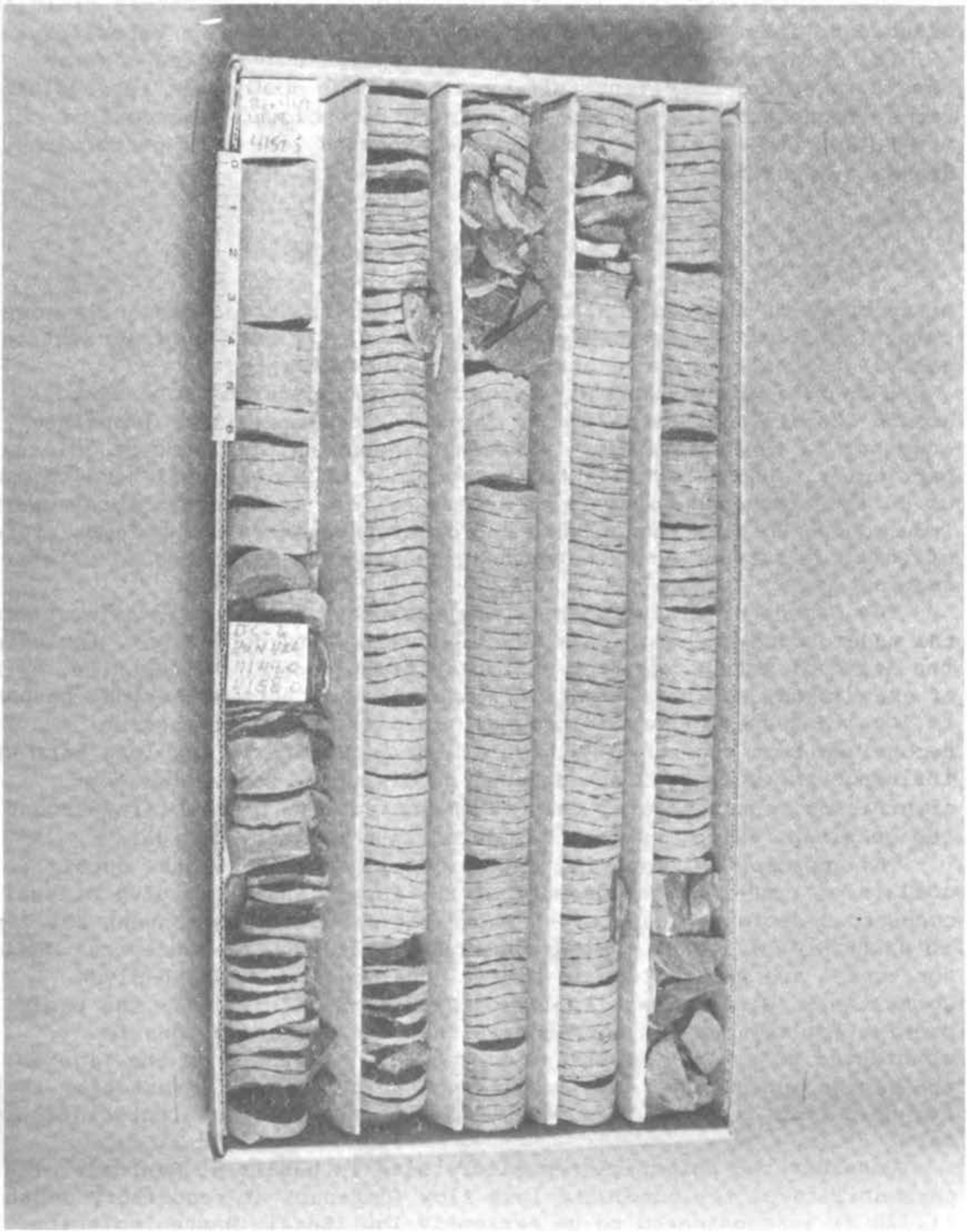


FIGURE 6-8 Basalt core from Hanford–intense discing. Source: Courtesy of Rockwell International.

Some of the consequences of a high and anisotropic stress field are alluded to later in terms of the long-term behavior of a repository.

The development of repositories in candidate geologies such as basalt, granite, competent tuff, and bedded or domed salt is feasible in terms of present construction and mining technology. The cost of excavation and stabilization, and the expected long-term performance, are site specific.

6.3. DISTURBED ROCK ZONE

Disturbance of the rock mass is ascribed to (1) stress concentrations resulting from the presence of the openings, (2) method of excavation, and (3) thermally induced stresses and displacements.

A major issue at hand with regard to the disturbed zone of rock around tunnels and shafts is the increase in permeability immediately adjacent to these openings. Existing discontinuities such as joints, particularly those parallel to the openings boundary, will tend to open up during excavation because of stress relief, and new fractures can be induced due to blasting effects and/or overstressed rock. Temperature effects can further exacerbate the situation, as described in a later section.

The damage or disturbance to the rock mass immediately adjacent to the underground openings is a function of in-situ conditions, including the state of stress and rock characteristics; opening geometries, method of excavation, and stabilization measures; and thermomechanical response to local and areal thermal loading. In particular, it must be recognized that the superposition of these factors in the long term will influence the extent of the damaged or disturbed zone. The most significant consequence of this is an increase in permeability around the openings, which must be considered in repository design.

The approach to resolving this problem has been to make cutoff collars or tunnel bulkheads part of the tunnel and shaft plug or seal conceptual designs. Figure 6-9 shows the essential components for seals in salt (Kelsall 1981). Figure 6-10 and 6-11 show preconceptual designs for tunnel and shaft plugs in basalt developed by Woodward-Clyde Consultants (Basalt Waste Isolation Project Staff 1981a). The basic premise for this design approach is that the disturbed zone is considered to be part of the seal or plug. As seen from the figures, the design approach for sealing systems in salt is different from that for hard rock, in that it must accommodate the creep characteristics of salt.

Consider the potential repository site in basalt at Hanford. The permeability of the candidate lava flow (Umtanum) at repository depth (1,128 m) is considered to be extremely low (Basalt Waste Isolation Project Staff 1981a). (It should be noted however, that this assumption is based on incomplete data at this time (Basalt Waste Isolation Project Staff 1981b).) The existing joints are quite frequent (3 to 10 per meter of core), although their permeability is low due to clay fillings or coatings. However carefully and expensively the collars are

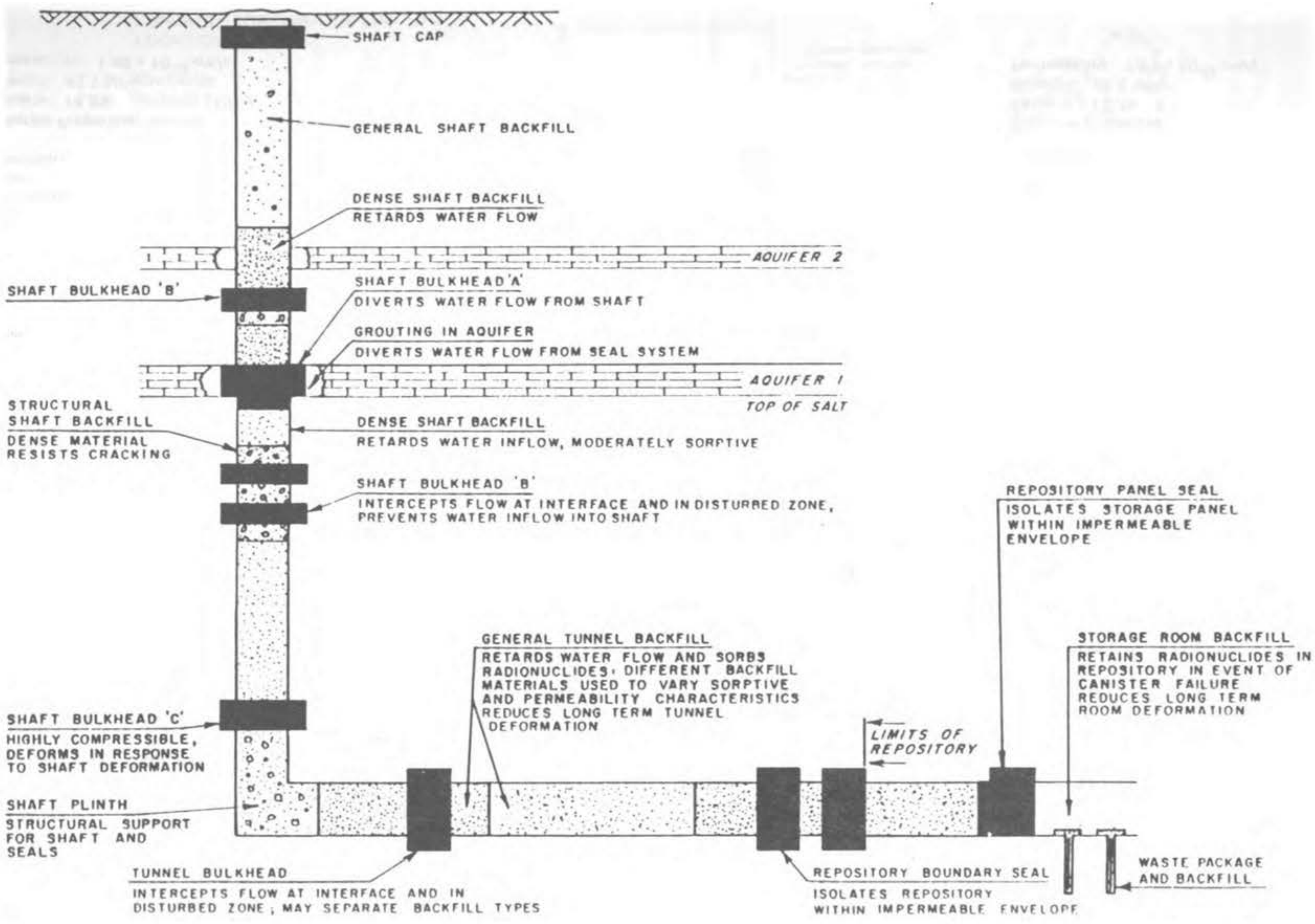
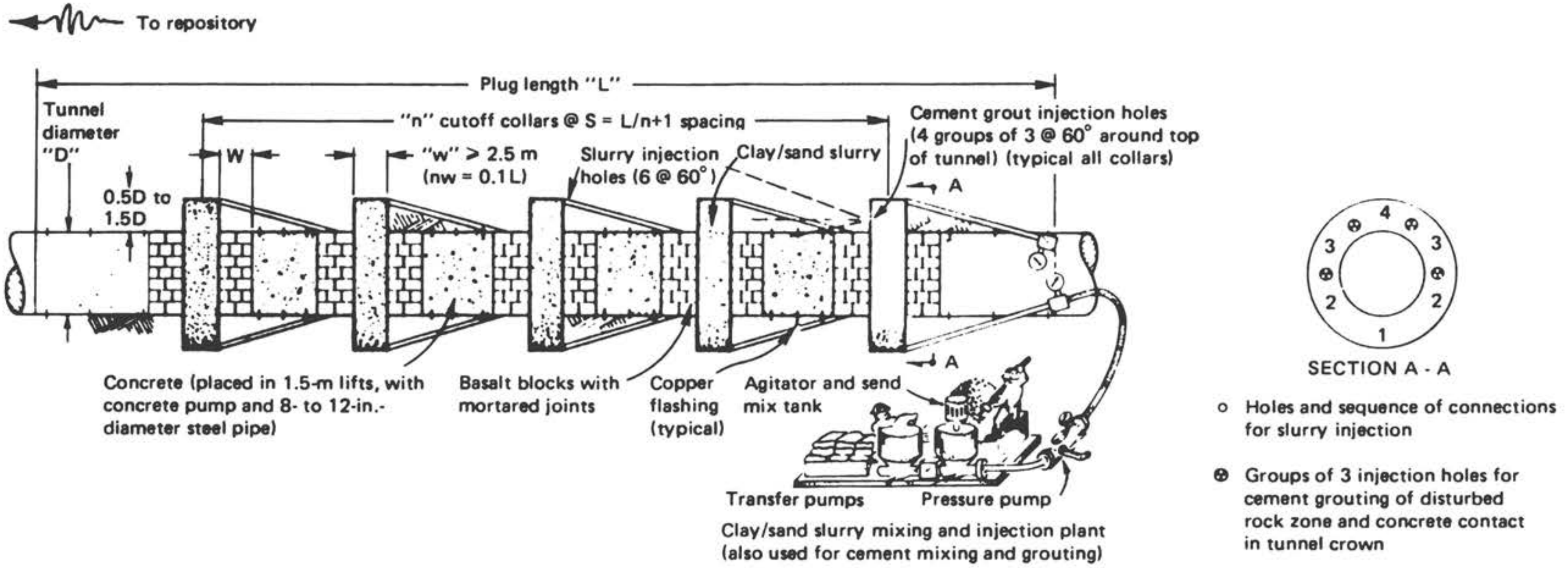


FIGURE 6-9 Essential components for shaft and tunnel seals in salt. Source: Courtesy of D'Appolonia Consulting Engineers.



CONCRETE FOR HIGH-TEMPERATURE LOCATION, T > 100° C

Mix Design
Portland Type V
Silica flour
Glaciofluvial sand
¾-in. gravel
Water
Plastiment

Concrete Properties
Porosity: 14.8%
Strength: 42.1 MPa
Permeability: 1.49 x 10^{-9} cm/s

BASALT BLOCKS WITH MORTARED JOINTS

Basalt Blocks
Size: 0.2 m x 0.2 m x 0.1 m
Weight: 10 kg
Porosity: 1.24%
Strength: 300 MPa
Permeability: 10^{-9} cm/s

CLAY SAND SLURRY

Mix Design
Shurgel
Glaciofluvial sand
Water

Slurry Properties
Unit weight: 1,340 kg/m^3
Porosity: 80%
Permeability: Future testing

CONCRETE FOR LOW-TEMPERATURE LOCATION, T < 100° C

Mix Design
Portland Type V
Lassenite
Glaciofluvial sand
¾-in. gravel
Water
Plastiment

Concrete Properties
Porosity: 13.3%
Strength: 36.5 MPa
Permeability: 3.67 x 10^{-9} cm/s

FIGURE 6-10 Preconceptual design for tunnels in basalt. Source: Basalt Waste Isolation Project Staff (1981a).

CONCRETE FOR LOW-TEMPERATURE LOCATION, T ≤ 100°C

Mix Design
Portland Type V
Lassenite
Glaciofluvial sand
¾-in. gravel
Water
Plastiment

Concrete Properties
Porosity: 13.3%
Strength: 36.5 MPa
Permeability: 3.67×10^{-9} cm/s

CLAY SAND MIX

Clay: Wyoming bentonite
Sand: Glaciofluvial
Clay sand ratio: 1
Max. dry density: 1,983
Optimum water content: 14.2%
Void ratio: 0.45
Swell pressure: 0.9 MPa
Estimated permeability less than 10^{-9} cm/s to be determined by future testing

CONCRETE FOR HIGH-TEMPERATURE LOCATION, T ≥ 100°C

Mix Design
Portland Type V
Silica flour
Glaciofluvial sand
¾-in. gravel
Water
Plastiment

Concrete Properties
Porosity: 14.8%
Slump: 11 cm
Strength: 42.1 MPa
Permeability: 1.49×10^{-9} cm/s

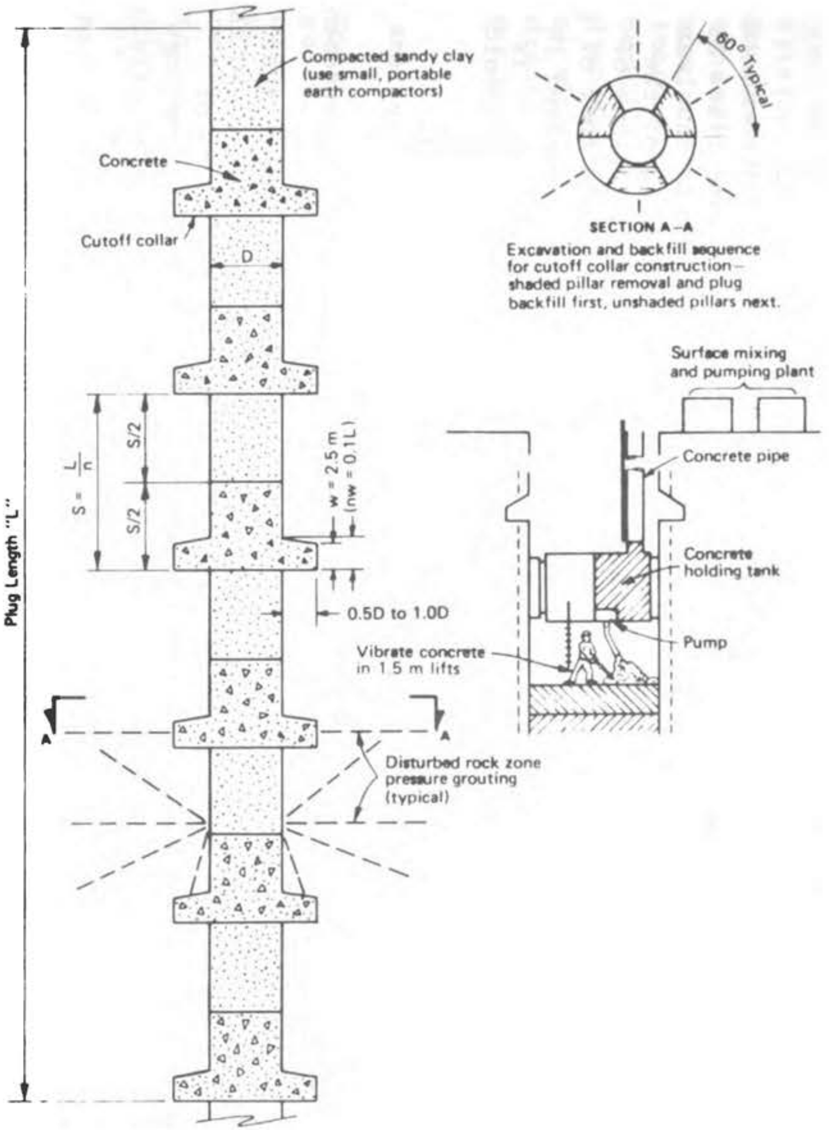


FIGURE 6-11 Preconceptual design of shaft plugs in basalt. Source: Basalt Waste Isolation Project Staff (1981a).

excavated, new joints will open up due to stress relief. Hence, the collars may retard shunt flow somewhat but not cut it off.

While grouting may also retard flow within the damaged zone, it will not achieve a permeability as low as the rock mass prior to excavation, if the hydraulic conductivity was initially less than 10^{-8} to 10^{-9} cm/s (10^{-10} to 10^{-11} m/s). This applies for all rock types.

It appears that a more realistic approach to fluid containment would be to develop a plugging scheme along, for example, the same lines as used in plugs in hydroelectrical power plant projects. This involves thick-walled concrete linings, followed by pressure grouting of the disturbed zone, and could well be used with intermediate sections of sorptive backfill. If the ensuing plug permeability exceeds that of the original in-situ permeability, the consequences should be assessed in terms of overall repository system performance. It seems that it is fundamentally questionable to have a plug design criterion that could exclude an otherwise acceptable site simply because the in-situ permeability is too low to be reestablished and field verified.

The evaluation of the performance of sealing and plugging systems and the extent of the disturbed zone around underground openings requires carefully controlled in-situ field experiments and measurements carried out under proposed repository conditions within the site-specific rock mass under consideration. These experiments must consider the appropriate geometry and size, initial conditions, and the thermomechanical/hydrochemical regime for accurate predictions of repository performance. Design criteria for the underground openings and the sealing and plugging systems should be verifiable in the field and should be compatible with practical construction or mining procedures.

6.4. OPERATIONAL STABILITY, RETRIEVABILITY, AND LONG-TERM PERFORMANCE--THERMOMECHANICAL ASPECTS

The extent to which temperature affects the performance of a waste repository largely depends on the host media or rock type and the maximum rock temperature attained. Important rock mechanics aspects of performance are (1) the possible increase in the near-field permeability due to thermal cracking and (2) the general time-dependent nature of rock mass properties as a function of temperature. In this context, media under consideration are generally divided into two categories: (1) hard rocks, i.e., basalt, competent tuff, and granite, and (2) rock salt, both domal and bedded. This division is based on the markedly differing physical properties of hard rocks and salt and the consequent markedly different thermomechanical and thermochemical response to changes in temperature and pressure or stress.

The temperature within the rock surrounding the repository depends on the in-situ temperature at repository depth and on the local and average areal thermal loading, as well as on the thermomechanical rock mass properties. The stress within the rock mass depends on the in-situ stress state, the geometry of the openings, and the induced thermal stress (a function of the local and areal thermal loading and rock mass properties).

Major questions concerning safety during repository operation, the ability to retrieve waste, and the long-term performance with respect to isolation are directly or indirectly related to opening stability. Instability, as defined in terms of major rock falls or excessive opening closure, would obviously hinder repository operation and retrievability. Long-term performance (postoperation or decommission) focuses on the aspect of containment or isolation. In the long term the consequence of instability would increase the extent of the disturbed zone around the repository and, hence, the extent of the zone of increased permeability of the rock mass. Even if the repository is backfilled immediately after waste emplacement, the extent of the disturbed zone could increase with time, as the low stiffness of the backfill relative to the undisturbed rock mass will allow for continuing deformations. Again, the consequences of instability and mechanisms are different for the two rock categories, salt and hard rock.

6.4.1. Repository Operation and Retrievability

The duration of repository operation will be on the order of 40 to 90 years, and proposals have been made to allow for retrieval of waste for up to 50 years after waste emplacement operations are completed (U.S. Nuclear Regulatory Commission 1980). Retrievability allows for the removal of canisters from the repository if the long-term performance of the repository systems is found to be unacceptable prior to sealing (U.S. Department of Energy 1980). A retrieval period does not preclude the early backfill of repository rooms; it means only that methods for handling the hot backfill and removing the canister intact are included as a part of the system design.

The concept of stability with respect to repository operation and retrievability, as applied to the rock mass within the immediate vicinity of the underground opening, is difficult to define accurately and even more difficult to assess. Factors of safety, usually derived as the ratio of rock mass strength to induced stress, although useful as an indicator of rock condition, can at best be crudely related to actual stability. Furthermore, the evaluation of stability depends on the magnitude of failures tolerated and on the use of ground support, rock reinforcement, and/or backfill to mitigate the effects of ground failure.

Hard Rock Repositories

For openings situated in hard rock masses, the criteria for stability consider the size and extent of fallouts of loosened material. Because the deformed rock mass within the immediate vicinity of the opening can undergo considerable loss of strength with shear deformation or opening closure (a cataclastic process resulting in an increase in the volume of the rock mass due to displacements along existing discontinuities and formation of new fractures with the opening of voids), this region of material can be susceptible to collapse.

The extent of the loosened zone and potential size of the fallout indicate the magnitude of the instability. Localized slabbing of overstressed rock and minor fallouts of loosened rock blocks are commonly controlled by scaling or a system of light reinforcement. Major fallouts, involving more than a cubic meter of materials (total volume), are likely to result in excessive support requirements and disturbance in the surrounding rock mass and should be avoided.

Predicting the stability of underground openings generally proceeds along one of two lines. A limit equilibrium analysis of potential fallout wedges or blocks of rock can be carried out, or alternately a continuum model employing a rock mass strength failure criterion can be used. Neither of these models accurately predicts the total response of the rock mass. Nearest the opening, within the disturbed zone, the limit analysis is more appropriate, while farther from the opening the continuum approach is reasonable.

For the analysis of underground opening in basalt at the Hanford site, a simple linear thermoelastic continuum model with an empirical strength criteria for the rock mass was employed (Schmidt 1981). This approach is, in principle, very conservative. Stresses predicted within the immediate vicinity of the opening are too high, as no consideration was given to the inelastic deformations that are bound to occur. Furthermore, the beneficial effects of ground support cannot be accurately modeled with the approach used, resulting in an erroneous conclusion concerning the effectiveness of support for improved stabilization (Schmidt 1981).

Given this overall conservative approach, a repository design in basalt has been proposed incorporating a local areal thermal loading of 13 W/m^2. (This is essentially the same loading specified in the Interim Reference Repository Conditions for a Nuclear Waste Repository in Basalt (Reference Repository Conditions Interface Working Group 1981c), 12.3 W/m^2.) Unfortunately, this design assumes a very favorable uniform initial stress state. As previously discussed, the average horizontal in-situ stress may be several times the overburden pressure. This, coupled with the unfavorable shape of the repository rooms, that is, relatively high sides to opening width, will lead to considerably higher stresses than have already been predicted. If the tunnel openings are reanalyzed using the same method of analysis (Schmidt 1981), assuming less favorable conditions (i.e., horizontal in-situ stress is equal to several times the overburden pressure), it will be necessary to reduce the thermal loading if the same margin of stability is to be achieved, all other factors remaining constant. Appropriate tunnel opening design can reduce but not necessarily eliminate the adverse effects of unfavorable in-situ stress conditions.

Determination of whether or not the areal thermal loading is too high or too low for some arbitrarily prescribed margin of opening stability cannot be accurately evaluated with numerical models, due to the complexity of geologic materials. Ignoring the economic considerations, very high thermal loadings are possible if the openings are fully supported and/or reinforced. The upper-bound limit of areal thermal loadings at which instability adversely affects repository operation or retrievability, however, should not be the major consideration of repository opening design. Other more critical factors, such as the long-term performance involving rock mass disturbance in the near and far fields, should dominate the design process.

Unfortunately, the mechanisms of rock mass disturbance are complex and poorly understood. Furthermore, the processes are highly dependent on site-specific conditions (i.e., rock mass characteristics, size and shape of openings, repository depth, method of excavation and support, state of stress). Although methods by which to minimize disturbance are known--such as the use of mechanical excavators or controlled blasting techniques; early placement of efficient support/reinforcement; proper opening size, shape, and arrangement; and limiting the areal thermal loading--the quantitative effects of any one factor can be accurately evaluated only through in-situ field testing. Resolution of this important issue will require detailed in-situ experiments carried out under the appropriate site-specific conditions.

It is unlikely that repository openings can be constructed without disturbance of the rock mass nearest the opening, irrespective of the method of careful excavation employed or other techniques to reduce disturbance. Disturbance associated with stress concentrations upon excavation of the openings cannot be practically avoided. Consequently, the existence of the disturbed zone should be included in the design with the following ramifications: (1) the development of a zone of increased permeability surrounding the openings and (2) the need for required stabilization measures (i.e., a support and/or rock reinforcement system) to ensure operational safety and retrievability. As previously indicated, evaluating the extent of this zone for design will require detailed field experiments.

The above discussion is largely directed at the known or assumed conditions for proposed repository openings located in the basalt rock mass at the Hanford site. Other possible hard rock repository sites, as yet undetermined, located within more massive formations with more favorable initial stress conditions may not require the design compromises described above; and the conservative approach currently employed for the design of the Hanford repository openings would not prove overly restrictive. The need for detailed in-situ field experiments for investigation of the disturbed zone, however, is important for all proposed repository sites, irrespective of the rock types or rock mass characteristics.

Temperature distributions for a reference repository in basalt with a local and thermal load of 12.3 W/m^2, a depth of 1,100 m, and an initial temperature of 50°C are given in Figures 6-12 through 6-15 (Reference Repository Conditions Interface Working Group 1981c). Based on a maximum design spent fuel cladding temperature of 300°C (Basalt Waste Isolation Project Staff 1981), the maximum temperature of the rock nearest the canister is roughly 200°C and occurs 11 years after emplacement. The exact maximum temperature of the rock will depend on the composition and in-situ properties of the overpack and backfill actually employed around the waste package.

Gross thermal spalling of basalt is reported not to be a problem, as the rock is considered essentially nonspallable (Thirumalai 1970). The maximum temperature for basalt is limited by mineralogical phase changes that take place in excess of 500°C, hence the maximum design

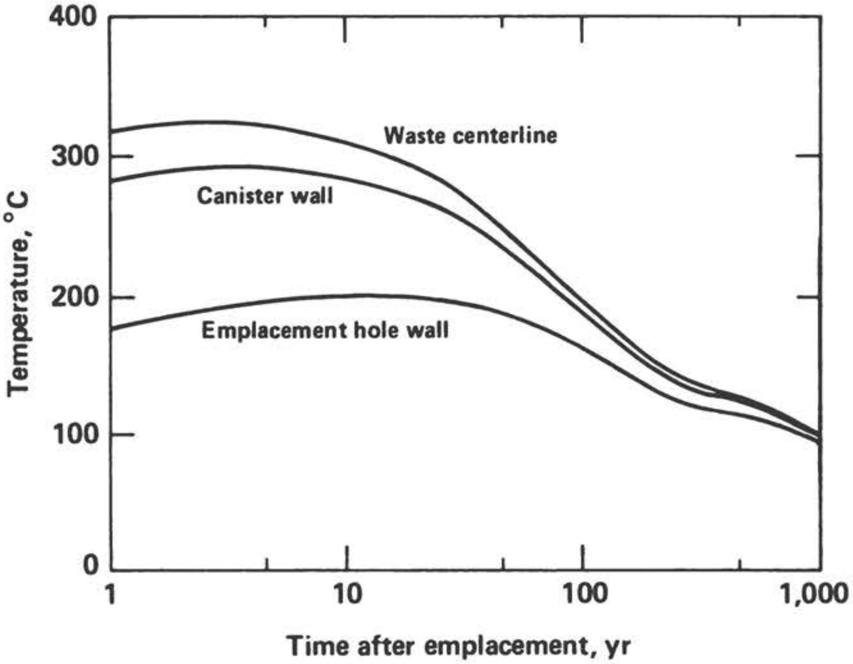


FIGURE 6-12 Reference spent fuel repository maximum temperatures in basalt. Source: Reference Repository Conditions Interface Working Groups (1981b).

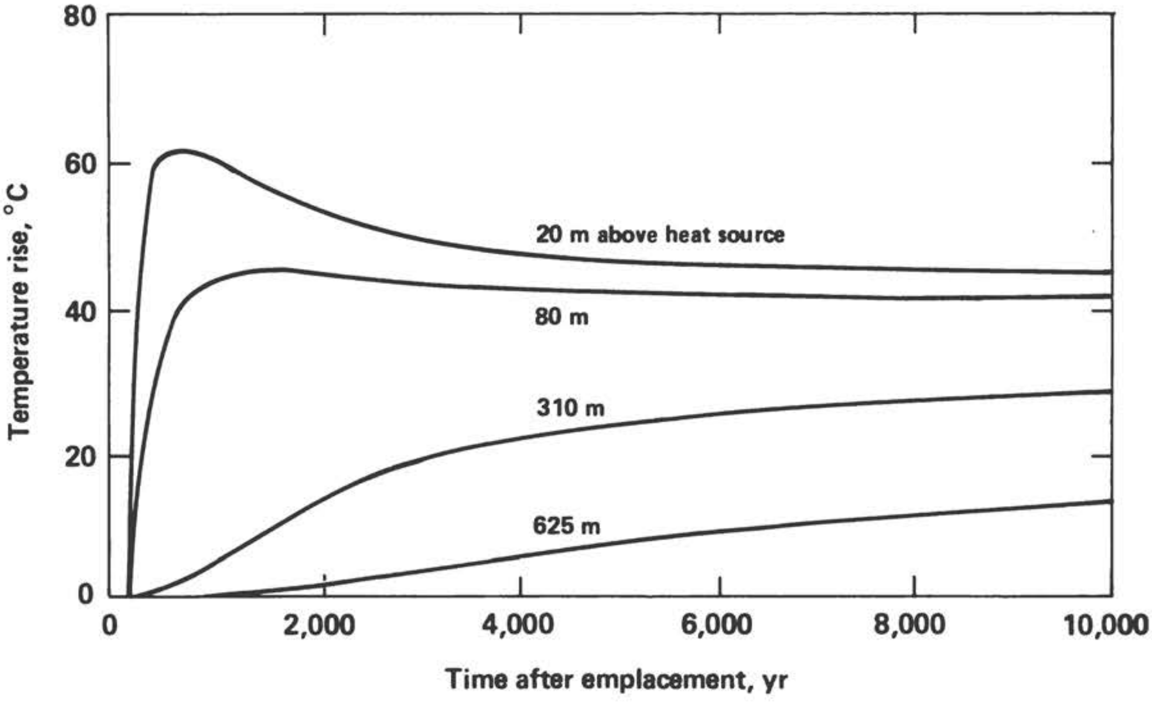


FIGURE 6-13 Far-field temperature effects for an areal thermal load of 12.3 W/m² in basalt. Source: Reference Repository Conditions Interface Working Groups (1981b).

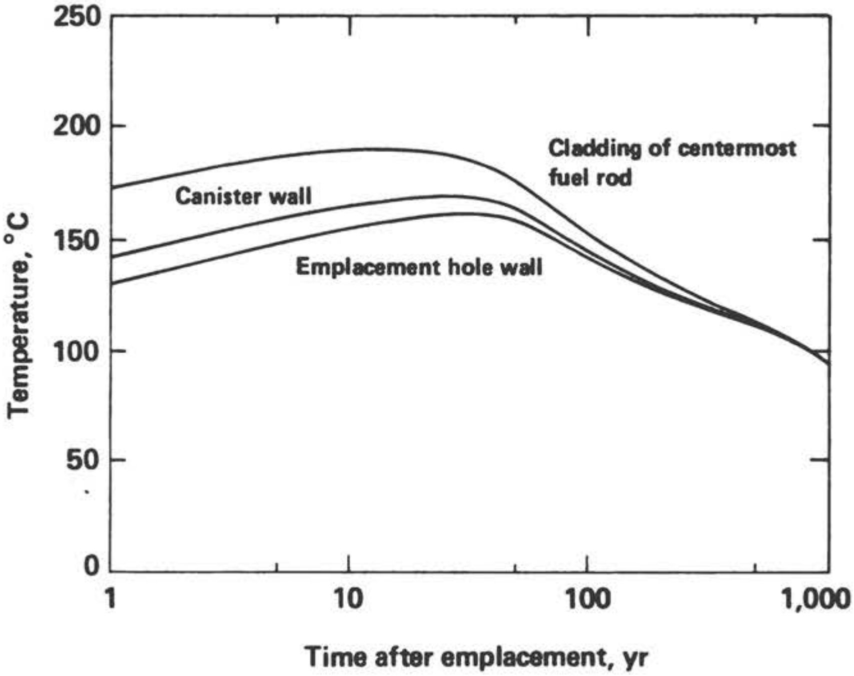


FIGURE 6-14 Alternate spent fuel repository maximum temperatures in basalt. Source: Reference Repository Conditions Interface Working Groups (1981b).

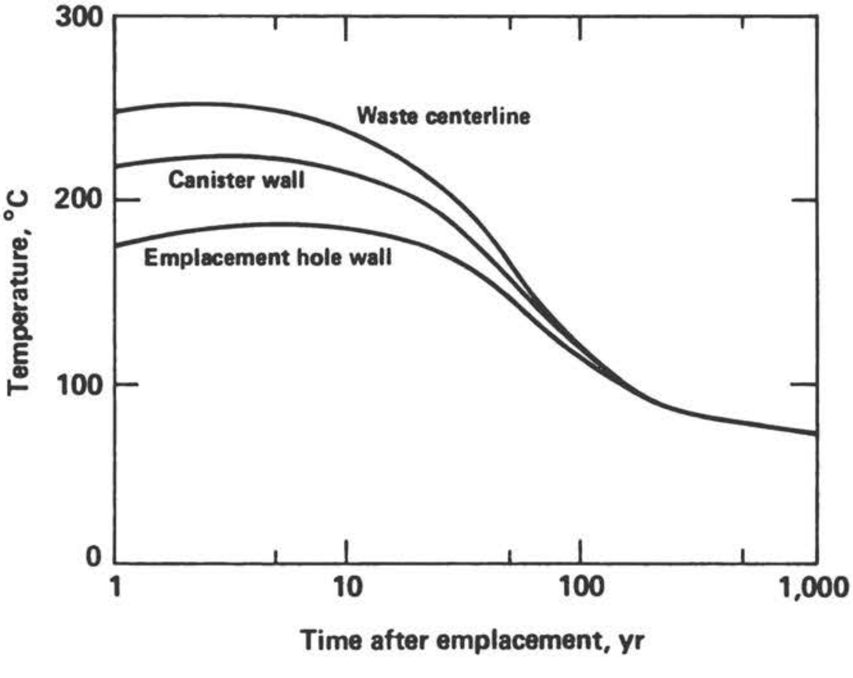


FIGURE 6-15 Alternate commercial high-level waste repository maximum temperatures in basalt. Source: Reference Repository Conditions Interface Working Groups (1981b).

temperature of 500°C (Kaiser Engineers/Parsons Brinckerhoff 1980). It is important to emphasize that other, more subtle, detrimental effects of temperature due to thermal cracking are noted at much lower temperatures, on the order of 200°C. This is discussed in the section on long-term performance.

As there are no proposed sites for a repository in granite, examples from generic tests are provided to illustrate the range of temperature limits associated with thermal spalling or decrepitation. Heating experiments in granite at Cornwall and at Stripa revealed decrepitation at 300°C or higher (Hood 1979); however, thermal spalling in gneiss, a crystalline rock similar to granite, has been reported for temperatures less than 110°C (Gray 1966).

The effects of temperature in tuff are complicated by the relatively high water content of some materials and possible mineralogical phase changes involving changes in both sorptive capacity and volume. Work to better define the temperature limits in tuffs is presently being carried out at Sandia National Laboratories (Lynch 1982). Preliminary findings indicate that the structural integrity of nonwelded tuffs can be adversely affected by contraction on drying if sufficient amounts of the contracting phase are present. The temperature of dehydration depends on many factors, including fluid pressure, which is itself dependent on the interaction of rock mass permeability, heating rate, and pressure release path length. Some drying can also occur from repository exposure to room-temperature air.

Thermal expansion tests on unconfined samples of both zeolitized and devitrified tuffs reveal a net contraction upon cooling, which suggests that in-situ rock mass permeability could increase and thermal conductivity decrease during cooling of the repository. Devitrified welded tuffs containing cristobalite were also shown to undergo a significant volume expansion due to phase inversion on heating to temperatures of 150°C to 200°C. A considerable effort will be required to establish the influence of dehydration and phase changes on temperature limits and the suitability of tuffs as a repository medium.

Despite the uncertainties indicated above and the difficulty of performing accurate calculations for situations in which appreciable tuff dehydration is predicted, interim reference repository conditions have been given (Reference Repository Conditions Interface Working Group 1981a). For spent fuel and commercial high-level waste, the maximum rock temperature was calculated at 165°C to 185°C and 140°C to 225°C, respectively. The range of temperatures indicated for each waste type corresponds to the cases of (1) assumed evaporation of all water inside the 100°C isotherm and (2) no evaporation.

With proper stabilization methods, repository openings in hard rock should remain stable without backfill during the operation period. Opening stability should not be a limiting factor in the design of hard rock repositories with the option for retrievability over periods considered in proposed repository design criteria (National Research Council 1981b).

Salt Repositories

Stability during the operational phase is largely a question of tolerable room closure. This includes floor heave and possible spalling of room walls and roof falls. Based on mining experience, the stability of unsupported mined openings in ductile materials, such as salt and potash, cannot be ensured for closures in excess of 15 percent (Office of Nuclear Waste Isolation 1981). If the rooms were required to remain open during a retrieval period, this would have a major impact on the design of the repository, in particular, the allowable areal thermal loading that would restrict closure to acceptable levels (Ritchie et al. 1979). Alternately, backfilling the repository rooms as soon as possible, then reopening by mining before retrieval, minimizes the closure problem and has several advantages for improved long-term isolation (considered in the next section). Remining the hot salt, if it should ever be necessary, would be extremely expensive; furthermore, compliance with mining regulations as required by the U.S. Nuclear Regulatory Commission in 10 CFR 60 would be difficult.

Because salt responds in a relatively ductile manner when subjected to the temperatures and stresses typical for a repository environment, the inelastic time-dependent creep deformations are large, essentially obscuring the elastic response. Creep deformations resulting in room closure are a function of areal thermal loading, repository geometry, repository depth, and material properties (although salt properties do not vary markedly as compared to hard rocks). In one study, a thermoelastic/viscoelastic analysis revealed that of these three parameters (thermal loading with spent fuel, 30 to 45 KW/acre; geometry, pillar height/width of 1/5 to 1/3; and depth, 300 to 800 m), repository depth had the most influence on total room closure (Wagner 1980).

Using the constitutive model for salt obtained for the WIPP site, it is possible to generalize the effect of depth on room closure (Munson 1979). Given that the creep strain is roughly proportioned to the applied stress to the fifth power, doubling the repository depth, say from 300 to 600 m or 400 to 800 m, has the effect of increasing the rate of closure by a factor of 32. Thus, for a specified thermal loading, depth becomes a major parameter.

With a consideration to limiting creep closure for improved stability, reducing repository depth has a major impact. In this regard the Reference Repository Conditions Interface Working Group's (1981a) 600-m specification on repository depth is unnecessarily restrictive. Selection of repository depth should be a part of the site-specific design process with the constraint of a minimum depth at which isolation from the surface is compromised. This minimum depth has been specified at 300 m (U.S. Nuclear Regulatory Commission 1980).

Accuracy of the predictions for creep closure of repository openings largely depends on the validity of the constitutive models employed. Although there has been considerable effort in this area, there is a lack of information on the mechanisms and the creep parameters that operate at the temperatures and pressures anticipated for the repository environment (Munson 1979). Generally, the constitutive models cannot handle complex thermomechanical histories, are developed only for one dimension, and are largely empirical (Senseny 1981). The use of empirically derived models is not necessarily undesirable; however, there must be sufficient data at appropriate temperature and stress state obtained over a sufficient duration of time to ensure the validity of the creep parameters. At present, this information, particularly long-term creep data, is not available. Structural or numerical codes that employ constitutive models are, however, in a relatively advanced state for repository design (Morgan et al. 1981).

Other problems requiring attention that affect the modeling methodology include the evaluation of brittle failure or fracture in salt, particularly under low confining pressure, tertiary creep, also involving fracture, and the validity of modeling salt as an incompressible material.

Most thermoelastic/viscoelastic and viscoplastic analyses model the salt as an incompressible material (i.e., no volume change associated with inelastic deformation). Although this is appropriate for general modeling of the repository, the prediction of opening closure is not accurate for large strains. Near-isovolumetric ductile behavior predominates at low deviator stress when the confining pressure and/or temperature is relatively high; however, some volume increase is observed even at a temperature of 200°C and confining pressure of 20.7 MPa. At lower pressures and temperatures, the volumetric strain is a major proportion of the indicated shear strain, indicating dominant cataclastic processes with significant dilatancy (Wawersik and Harmum 1980). It has been suggested, however, that a large portion of the measured dilatancy is related to method of testing and that larger samples reveal less dilatancy. Nevertheless, the brittle phenomenon associated with this process is important for stability considerations.

Because the salt nearest the opening is under no confinement, it is susceptible to deterioration at large strains. This material can loosen, adversely affecting the stability of the opening. Analyses employing isovolumetric constitutive models will underestimate the magnitude of room closure at large strains and cannot be used to accurately predict the deformation at which fallouts are likely to occur. For predictions with improved accuracy, a model incorporating progressive failure of the salt is required and has been proposed (Matalucci and Hunter 1981).

The maximum temperature to which the salt can be subjected is 250°C to 400°C, at which point decrepitation occurs (Bradshaw et al. 1968, Bradshaw and McClain 1971, Gevantman 1980). Decrepitation is undesirable, as it reduces the thermal conductivity of the salt nearest the waste package, which may lead to higher canister temperatures (Russell 1978). Possible mechanisms for fracturing are (1) unequal thermal expansion along the crystal axis, (2) chemical reactions such as combustion of organic materials, and (3) the buildup of pressure in brine inclusions (Bradshaw et al. 1968). The rate of heating has little effect on the temperature at which decrepitation takes place.

Maximum temperatures for the salt nearest the canister, as predicted for the interim reference repository, are considerably less than the lower-bound temperature at which decrepitation was observed (Reference Repository Conditions Interface Working Group 1981a). Maximum temperatures are 140°C with spent fuel and 160°C with commercial high-level waste, given a local areal thermal loading of 25 W/m^2 and an initial temperature of 34°C. Disposal of defense high-level waste with an areal thermal load of 11.6 W/m^2 results in a maximum temperature of only 80°C. Because of potential (far-field) limitations imposed by excessive uplift and failure of overlying rock formations, the average areal loading with spent fuel is recommended not to exceed 15 W/m^2 (Russell 1978). However, as indicated by Russell (1978), "this limit must be reevaluated for each specific site in order to assess the effects of rock mass movement on the hydrological regime and long-term safety." Thermal loadings are limited by maximum near-field temperature for commercial high-level waste and by spatial considerations for defense high-level waste.

Temperature histories for the three different waste forms at various positions above a 600-m-deep repository are given in Figures 6-16 through 6-18.

6.4.2. Long-Term Performance

Predictions of the long-term performance of a repository depend on a fundamental understanding of the processes governing the response of the surrounding rock mass to the induced perturbations of excavation, heating, and cooling. Many of these processes influencing the physical properties, and hence the isolation potential, of the rock mass increase nonlinearly with temperature and, more important, are time dependent and irreversible. In general, very little is known about the time-dependent processes in the inelastic deformation of hard rocks as compared with rock salt (Handin 1980a,b). Because of the relatively complicated rock mass reponse upon construction, operation, and decommission of an underground repository, and because the processes governing this response are incompletely understood, the repository design must necessarily be conservative in all aspects influencing long-term isolation.

Hard Rock Repositories

For repositories in hard rock, a major question of long-term performance is related to prediction of the rock mass permeability or, specifically, the transmissivity of the fracture system. Because the intact permeability of the major hard rock types under consideration for repository siting is low compared with the apparent permeability of the fracture system, the discontinuities dominate water transport. For example, through-going fractures in dense crystalline rock such as granite (Westerly) and gabbro reveal an apparent permeability 6 to 10 orders of magnitude greater than the intact permeability (i.e. of the solid rock) when compared at the same effective stress state (Trimmer et al. 1980). Accordingly, the thermomechanical and thermochemical responses of the fracture system are of primary importance.

Investigations of the thermomechanical and thermochemical processes

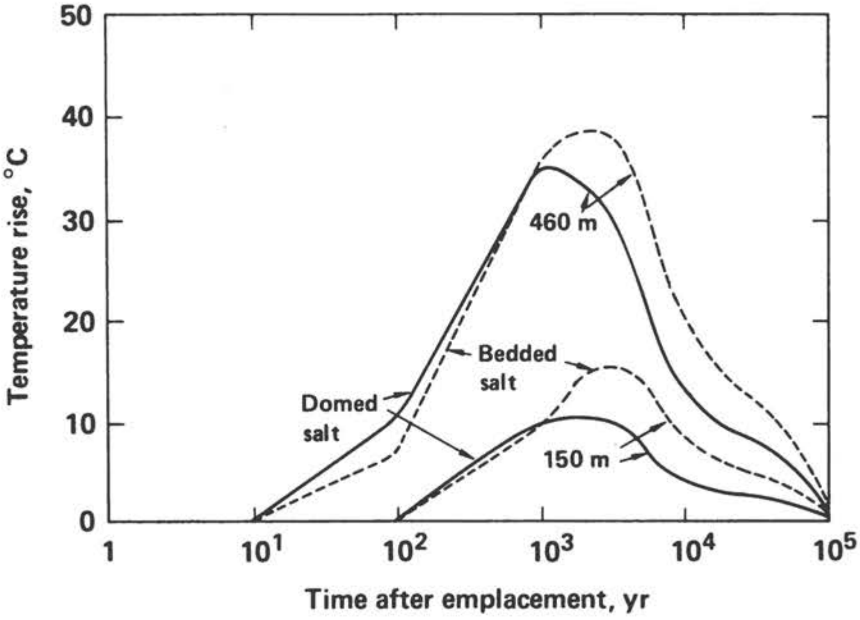


FIGURE 6-16 Transient thermal response at several points above a spent fuel repository with an areal thermal loading of 15 W/m² located 600 m deep in salt. Source: Reference Repository Conditions Interface Working Groups (1981a).

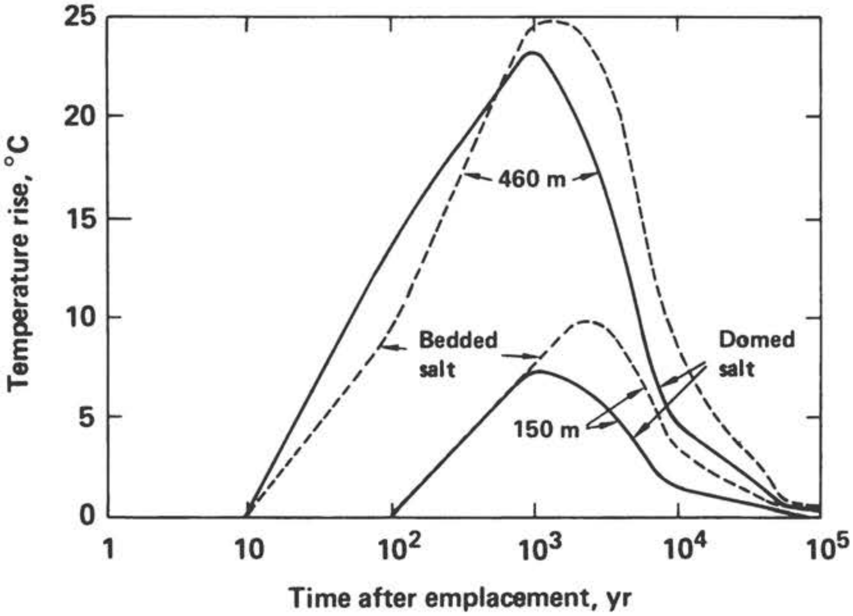


FIGURE 6-17 Transient thermal response at several points above a commercial high-level waste repository with an areal thermal loading of 25 W/m² located 600 m deep in salt. Source: Reference Repository Conditions Interface Working Groups (1981b).

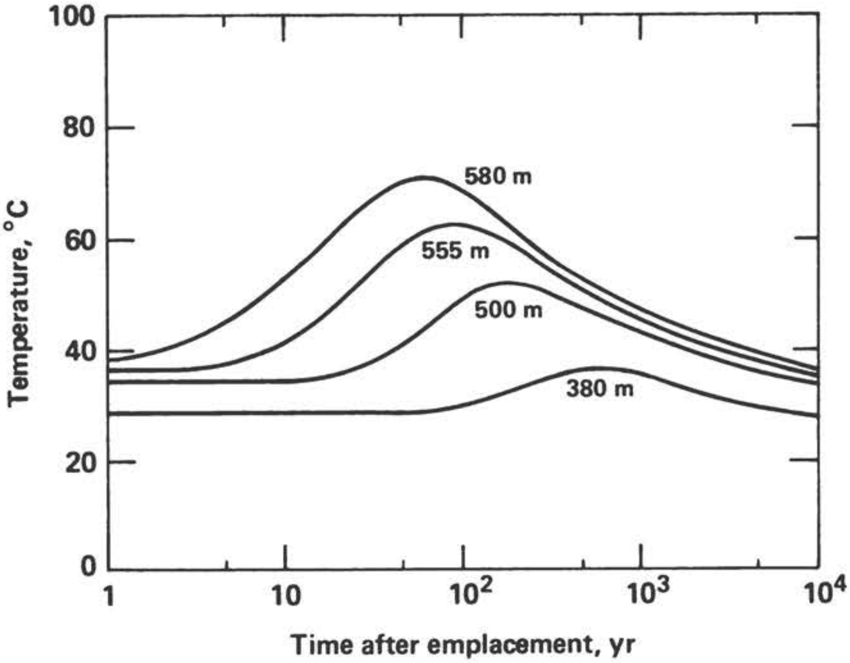


FIGURE 6-18 Transient thermal response at several points above a defense high-level waste repository with an areal thermal loading of 11.6 W/m^2 located 600 m deep in dome salt. Source: Reference Repository Conditions Interface Working Groups (1981a).

influencing rock mass permeability are at an early stage of development, largely due to the difficulties of studying rock masses as opposed to intact rock specimens. About the only properties that are determinable with reasonable certainty are thermal conductivity, specific heat, and density needed for prediction of the temperature field for specific heat sources. Several investigators have concluded that test data from intact specimens are adequate for thermal modeling of the rock mass (Handin 1980a, Chan and Javandel 1980, U.S. Department of Energy 1980).

Most studies of thermal phenomena have concentrated on intact samples. Investigations of thermal expansion and thermal cracking, important for predicting the changes in stress, displacement, and permeability due to heating, reveal property dependence on crack porosity, heating rate, and previous maximum temperature as well as mineralogical composition, preferred crystal orientation (Richter and Simmons 1974) and, possibly, grain size. Generally, thermal cracking is implied from the difference between the thermal expansion measured and the average value calculated for the constituent minerals. The cracking is an irreversible process that results in an increased volume and hence a potential increase in permeability.

From the uniform heating of unconfined igneous rocks (i.e., granite, gabbro, and diabase), Richter and Simmons (1974) found a threshold of increased crack production for heating rates greater than 2°C/min and differential thermal strains larger than 3×10^{-4} between component crystals. The rate at which thermal cracking proceeds with temperature appears to be dependent on the rock type. Simmons and Cooper (1978) found the rate of increase in new crack volume to increase exponentially with temperature for tests on unconfined Westerly granite, while Friedman et al. (1979) only observed a linear increase with temperature for Charcoal granodiorite.

Another indirect indicator of thermal cracking is the irreversible reduction in elastic modulus measured with increased temperature. Employing models for anisotropic single-phase ceramics, McLaren and Tichell (1981) estimated a crack surface area increase of 10^2 to 10^3 m^2/m^3 of granite when heated to 200°C. The effect of confining pressure was not considered.

Using the same type model, it appears that uniform heating of intact basalt from the Hanford site causes little thermal cracking up to temperatures of 200°C (minimal change in elastic modulus, Figure 6-19). The influence of temperature on basalt rock mass response, however, has yet to be determined. Of major concern in this respect is the influence of heat on the joint filling materials, in particular zeolites and clay minerals.

When considering the behavior of the rock at some distance from the repository openings, the effect of confining pressure on thermal cracking is important. Most experimental results are consistent with cracks tending to open with increasing temperature and to close for increasing confining pressure. Inferred permeability changes for Westerly granite up to 300°C and confining pressure up to 55 MPa reveal that confinement does not overcome the destructive effect of temperature for this particular rock (see Figure 6-20). Although confinement does not eliminate the effect of thermal cracking on modulus, thermal expansion, and permeability, it does reduce the adverse effect on rock strength (Bauer and Johnson 1979).

The influence of pressure and (uniform) temperature on permeability when measured directly indicates variable results, largely because of complications introduced by the reactions between the pore fluid and rock minerals or the geochemistry. Short-term experiments with distilled water on Westerly granite and two adamellites (quartz monzonite) suggest a reduction in permeability with temperature in the range of 100°C to 150°C at constant effective confining pressure of 35 MPa (Potter 1978). Above 150°C, thermal cracking resulted in an increase in permeability. Because of the relatively short duration of the experiment, the reactions of the water with the rock were minimized.

Experiments on Westerly granite, lasting up to 17 days, illustrate the influence of geochemistry on permeability (Summers et al. 1978). For a temperature range of 100°C to 400°C, a constant confining pressure of 50 MPa, deviator stress of 0 to 350 MPa, and a pressure gradiant of 27.4 MPa, initially the permeability increased with temperature by a factor of 10 to 100 times over the permeability at 25°C as the result of thermal cracking. With time, however, the permeability decreased as the outlet flow channels became plugged with dissolved quartz and plagioclase. After ten days the flow rates from samples at 300°C were less than those at lower temperatures, and the flow from the sample at 400°C was not measurable.

As illustrated above, water-rock interactions can result in a permeability decrease by the formation of secondary minerals; however,

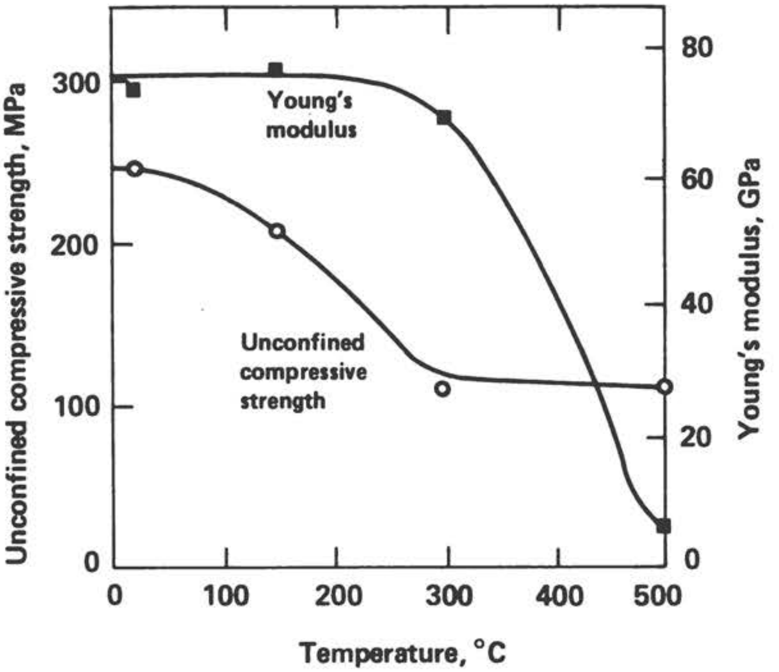


FIGURE 6-19 Strength and modulus of basalt as a function of temperature. Source: Schmidt (1981).

dissolution can also lead to an overall permeability increase. For example, after five days the samples of adamellite at 200°C (described above) revealed an increase in permeability due to dissolution of quartz (Potter 1978). Actual response of a particular intact rock within a proposed repository environment is obviously very site specific.

The long-term response of a rock mass (i.e., intact rock with discontinuities) to heating and cooling, including geochemical considerations, has not been studied in detail. Short-term field experiments, however, have been carried out and provide some indication of expected behavior. These include permeability tests in rock heated with injected water and by installed electrical heaters, as well as groundwater inflow measurements during heater experiments. Generally, the test results indicate that thermally induced or applied compressive normal stresses tend to reduce hydraulic conductivity while shear displacements increase conductivity. Conditions under which irreversible damage or increased permeability may occur upon heating and subsequent cooling have not been established.

Experiments in granite at Stripa included permeability tests with water injected at 10°C and at 35°C. Measurements indicated a 50 percent reduction in hydraulic conductivity for the 25°C temperature increase, despite the reduction in water viscosity at the higher temperature (Lundstrom and Stille 1978). Similarly, measurements of groundwater flow into heater and nearby instrumentation holes indicated a general decrease in inflow and permeability as a result of heating, after an initial period in which the flow increased. This initial flow

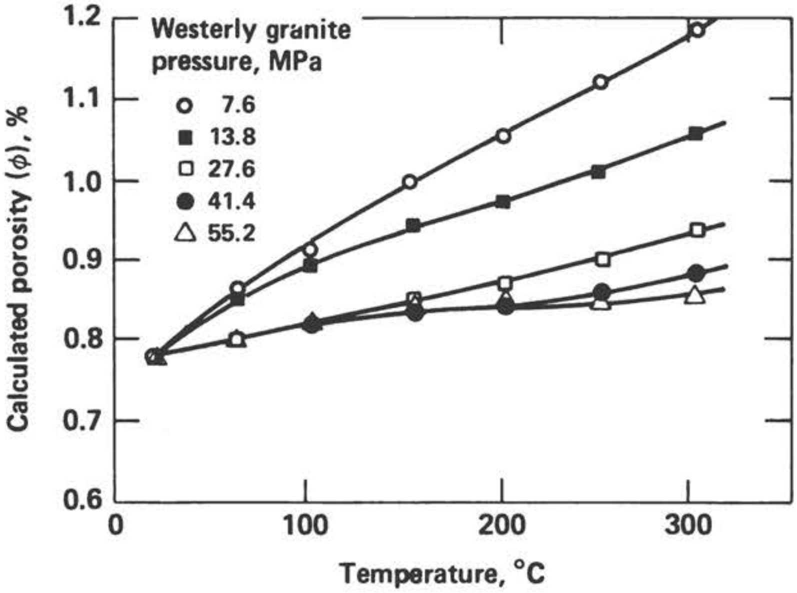


FIGURE 6-20a Calculated porosity versus temperature in Westerly granite for different confining pressures. Source: Heard and Page (1981).

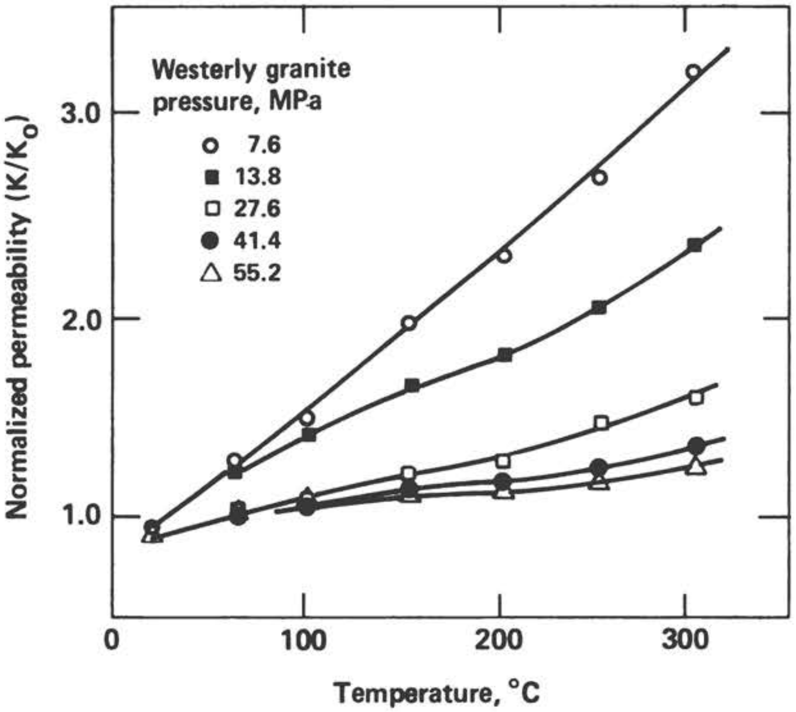


FIGURE 6-20b Normalized permeability (calculated) versus temperature in Westerly granite for different confining pressures. Source: Heard and Page (1981).

increase on heater activation was ascribed to the finite source of water squeezed from storage in joint cracks on compression and closure of these cracks (Nelson and Rachiele 1982). These test results did not provide information on possible irreversible changes in permeability after cooling.

Tests on a mineralized joint in a heated 8 m^3 block of gneiss revealed a fourfold reduction in hydraulic conductivity for an applied normal stress of 6.9 MPa at ambient temperature, 12°C (Voegele et al. 1981). Repeating the test at a higher temperature, 74°C, resulted in a 30-fold reduction in conductivity. The difference in reduction at ambient and elevated temperatures was most likely due to the improved matching of joint surfaces at the higher temperature (being closer to the temperature at which the joint was formed). A small induced shear displacement of 0.25 mm caused sufficient dilation to increase permeability by a few percent; however, the boundary condition of the block test did not allow for larger shear displacements.

Tests with model joints under low confining or normal stress indicate significant increases in permeability for larger shear displacements. Permeability was found to increase by as much as a factor of 10 for the first 2 mm of shear displacement and a similar amount for an additional 4 mm of displacement (Maini 1971).

For a given shear displacement, the influence of higher normal stresses appropriate for a repository environment would undoubtedly result in a smaller increase in permeability than indicated above. The potential importance of shear strain on rock mass permeability, however, is clearly indicated. The rock mass nearest the repository openings, because it is subjected to potentially large shear strains as a result of excavation and heating, is susceptible to disturbance and increased permeability. The extent or depth of the potential disturbed zone is very site specific.

Because of the large number of variables involved in the evaluation of rock or rock mass permeability, it is essential that detailed laboratory and field studies concentrate on site-specific rock types subjected to the appropriate repository environment, especially when considering rock-water interactions. Generic studies, while of general interest, are limited in their applicability for future repository design.

Crack growth in hard rocks, aside from being a thermally activated process, is a rate-controlled process (i.e., time dependent). The rate of crack growth depends on the rate at which corrosive agents can decrease the strength of the rock at the crack tip (Carter et al. 1981). This process, called stress corrosion cracking, is markedly influenced by the presence of polar pore fluids, such as water. Confining pressure tends to inihibit crack growth due to (1) an increase in the energy barrier for continued crack propagation, (2) closing of extensile cracks, thereby reducing the migration rate of corrosive fluids and gases to the crack tip, and (3) reduction of the rate of crack linking (Krang 1980).

At low homologous temperature, i.e., ratio of sample temperature to melt temperature less than 0.1, the permanent strain associated with crack growth in most silicate rocks takes place at stresses greater than half the unconfined compressive strength (National Research Council 1981b). Only at higher ratios of applied deviator stress to compressive strength, after a critical crack density has been achieved, will the onset of the tertiary creep ultimately result in fracture instability or creep rupture. Studies with Barre granite show that the applied deviator stress is approximately proportional to the logarithm of time to reach fracture instability (Krang 1979).

Although the majority of the rock surrounding a repository will not exceed relatively low homologous temperatures or a stress of half the compressive strength, the behavior of rock masses (including discontinuities) has not as yet been studied. If appropriate support measures are employed, the likelihood of opening instability due to creep rupture is remote; however, the major question is again the predicted change in rock mass permeability with time, as implied from time-dependent crack growth in creep experiments.

In summary, unless the temperature of the rock within the immediate vicinity of the repository is kept relatively low, between 100°C and 200°C, depending on the rock type, thermal cracking of intact rock is possible. Induced compressive normal stresses will tend to reduce the hydraulic conductivity of joints while shear displacements can increase conductivity. Whether or not this results in an increase in long-term permeability of the rock mass strongly depends on the specific geochemistry of the site. Very little information on the thermomechanical or thermochemical response of rock masses is available.

To reduce the uncertainty when predicting the long-term performance of a repository, the choices appear to be (1) to keep the temperatures relatively low to preserve the integrity of rock mass and/or (2) to design for a zone of increased permeability within the immediate vicinity of the repository rooms and shafts. Selection of appropriate temperatures and the evaluation of the extent of the disturbed zone are site specific and will require a detailed program of investigation emphasizing field experiments carried out under conditions approximate for the repository environment.

However, there needs to be a better definition of the importance of rock integrity and permeability, as affected by mining and heating, to the long-term performance of a repository. These are likely to be near-field effects, mainly in the emplacement rock itself. The theory in Section 5.7 describing the long-term rate of dissolution of waste-package constituents indicates that the dissolution rate may be only weakly affected by water flow rate within the emplacement zone. The performance analyses in Chapter 9 indicate that a key performance parameter is the total water travel time from the waste to the environment. The data in Tables 9-4 and 9-7 indicate that, for some repositories, the total water travel time is little affected by the flow properties of the emplacement rock. In such cases the time for groundwater to travel from the waste to the edge of the emplacement rock is small compared with the travel time in the surrounding media. This does not seem to be true for unsaturated tuff, where the travel time in the unsaturated zone seems to be controlling, and the effects on the emplacement rock would therefore become more important.

Salt Repositories

Long-term performance of a repository in salt is not complicated by many of the factors previously described for openings in hard rock. Intact salt has a very low permeability (estimated to be on the order of 2×10^{-16} m/s; Cloninger et al. 1980), and it is largely free of natural fractures. Discontinuities associated with bedded salt formations, such as clay seams, are also relatively impermeable. The ductile behavior of salt, particularly at elevated temperature, while a problem for short-term stability considerations, is of benefit to the long-term performance. Fractures within the salt tend to heal at low to moderate temperature and pressure (Costin and Wawersik 1980). After backfilling of the repository, fractures within the salt nearest the openings, resulting from large creep strains at little or no confinement, will tend to close and heal with compaction of the backfill.

The total effects of salt creep on long-term isolation are dependent on the interaction of several factors. In general, the greater the amount of creep closure allowed, the greater the subsidence and disturbance to the surrounding rock mass, including more brittle overlying formations other than salt. Conditions that promote creep closure, such as increased repository depth or applied stress and higher temperatures due to increased areal thermal loading, have a beneficial effect on encapsulation (the sealing of the repository through creep deformation) and creep healing of fractures. Undoubtedly, to maximize isolation and minimize overall disturbance, the total closure of the repository rooms will have to be limited. The amount, however, depends on the depth and geometry of the repository openings, the areal thermal loading, the properties of the salt and surrounding formations, and the timing of the placement and density of the backfill. Establishing these limits for specific sites is of major importance (National Research Council 1981a,b) and relies heavily on the use of predictive numerical modeling.

Placement of backfill within the repository rooms as soon as possible is one method to minimize disturbance. The feasibility of this, including the design for retrieval by remining, has yet to be demonstrated, although there do not appear to be any major obstacles.

6.5. SUBSIDENCE/UPLIFT

The integrity of formations surrounding the repository that restrict groundwater flow, the aquitards and/or aquicludes, must be preserved. Design criteria to ensure integrity should be established for site-specific conditions. Thermomechanical codes incorporating realistic strength failure models (Callahan and Ratigan 1978, Callahan 1981), including a no-tensile stress criterion (Monsees et al. 1981), are employed to establish the limits of rock mass stress or strain and the total allowable subsidence/uplift. A general subsidence/uplift limit expressed in terms of so many feet, although possibly a useful guideline, is not an appropriate design criterion, as it does not consider site-specific parameters.

The calculated change in stresses and displacements within these units is largely due to the thermal loading, as the contribution from excavation of the openings is generally very small due to the low extraction ratios proposed. An exception to this may occur for repositories in salt in which creep closure of the openings is excessive. Possible irreversible disturbance of the surrounding formations must be considered in establishing the magnitude of allowable room closure. Accuracy of the required thermomechanical calculations will markedly depend on the in-situ rock mass properties, in particular the thermal expansion and modulus of deformation.

6.6. TECTONIC ENVIRONMENT

Repositories must be located to avoid centers of recent igneous activity (within the Quaternary period) and areas of recent faulting that may have an adverse impact on the long-term performance of the repository (Office of Nuclear Waste Isolation 1980). Similarly, measured rates of natural surface subsidence or uplift must not adversely influence repository performance. Standard geotechnical/geological site investigation techniques are appropriate for satisfying the above criterion.

Acceptable design approaches for ground motions associated with a maximum credible earthquake are available. Compared with the underground workings, surface facilities are generally subjected to stronger ground motions due to the attenuation of specific components of the earthquake waves with depth.

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7

GEOLOGIC, HYDROLOGIC, AND GEOCHEMICAL PROPERTIES OF GEOLOGIC WASTE-DISPOSAL SYSTEMS

7.1. INTRODUCTION

Two main pathways are available for radionuclides in the waste to reach the environment after the repository has been sealed. One is by dissolution and hydrologic transport. This pathway is emphasized in this chapter on the different proposed geologic environments. The second is by any one of several possible but unexpected events such as human intrusion, volcanism, or other failures.

The natural barriers to hydrologic transport of radionuclides are mainly:

- o Low solubilities of the waste form and of key radionuclides such that the rate of release of these radionuclides to the environment is low; solubility of each radionuclide is affected by its geochemical properties and geochemistry of associated waters.
- o Sufficient sorption of key radionuclides by rocks in the repository and in the surrounding media; sorbabilities are controlled mainly by the geochemistry of each element, by minerals such as clays and zeolites in the rocks, and by the properties of associated pore waters, including pH, Eh, and dissolved species.
- o Lack of moving groundwater in the repository host rock, or a sufficiently long time for water to travel from the waste to the environment.

Other physical and chemical properties of the repository environment affect the migration of radionuclides. Thermal conductivity of the rocks affects repository temperatures, along with the type and age of the waste, as well as the waste loading. Temperature changes affect local flow pathways, permeabilities, and velocities as well as groundwater chemistry. Brine inclusions in salt can migrate toward the hot waste, thus affecting reaction rates. The geologic medium also affects the features and problems of repository construction, waste emplacement, and sealing.

Specific sites that have been studied extensively include Hanford, Washington (thick basaltic lava flows); the Waste Isolation Pilot Plant (WIPP) site, New Mexico (bedded salt deposits); and the Nevada Test Site (NTS) (rhyolitic volcanic tuffs). Other rocks and hybrid environments of two or more rock types have characteristics that may be even more favorable for repositories, but these have not yet been studied in much detail and are considered here as "generic." Detailed evaluations of bedded salt deposits, granitoids, the basaltic lava flows of the Basalt Waste Isolation Project (BWIP), and a hybrid type of granitoid rocks overlain by off-dipping sedimentary rocks with a regional aquifer are considered in detail in background papers that are to be published separately. Summary evaluations in this chapter emphasize the favorable and unfavorable aspects of each site (or generic type) for long-term waste isolation. Questions given special attention are:

o What is the present state of knowledge concerning the expected isolation performance of the natural barriers provided by each specific site or generic type?

o What are the principal uncertainties in this knowledge, particularly as they affect long-term prediction of isolation performance?

o What is our evaluation of the adequacy of programs under way to obtain the needed information?

Summary Table 7-1 lists present best estimates of sorption properties and solubilities of the principal radionuclides as affected by the geochemical environments of the main rock types being considered as hosts for repositories. Section 7.3 is a summary of the principal hydrological parameters for different candidate rock types (principally used in Chapter 9). Section 7.4 is a summary of the generic characteristics of candidate host-rock types. This is followed by more specific evaluations of specific sites or of generic types for those with inadequate specific data. The chapter concludes with a discussion of the data and evaluations that led to Table 7-1.

7.2. SUMMARY OF SORPTION PROPERTIES AND SOLUBILITIES OF RADIONUCLIDES

At the request of the panel, K. B. Krauskopf, Stanford University, has compiled a table of solubilities and sorption retardation factors for various radionuclides in the principal candidate rock types (Table 7-1). The sources of data and bases for selecting these values appear in Section 7.10. The retardation factor K is the ratio of pore velocity of groundwater to the net velocity of transport of a dissolved contaminant, assuming local chemical equilibrium of the contaminant dissolved in the liquid and sorbed on the rock. The boldfaced values of retardation factors are those that Krauskopf considers to be suitably conservative for the purpose of predicting the performance of conceptual repositories in these media (Chapter 9).

The sorption data for the media surrounding a salt repository take into account the expected salinity of groundwater that may intrude into a salt repository and carry dissolved radionuclides through the surrounding media and finally to the environment.

TABLE 7-1 Solubilities and Retardation Factors of Some Hazardous Radioactive Elements

	Solubility (log ppm)					Retardation Factor (1 + 10K_d)				
		Reducing: Eh = -0.2		Oxidizing: Eh = +0.2						
Element	Most Probable	pH = 9	pH = 6	pH = 9	pH = 6	Granite	Basalt	Tuff	Clay, Soil, Shale	Salt
						5	5	5	5	20
Se	-3 (?)	–	–	–	–	**50**	**50**	**50**	**50**	**200**
						200	200	200	200	1,000
						10	50	20	50	1
Sr	high	-0.2	high	-0.2	high	**200**	**200**	**200**	**200**	**10**
						2,000	2,000	10,000	5,000	100
						500	500	500	500	300
Zr	-4	-4	-6	-4	-6	**5,000**	**5,000**	**5,000**	**5,000**	**1,000**
						30,000	10,000	10,000	50,000	5,000
						1		1	1	1
Tc	-3	-10	high	high	high	**5**	**5**	**5**	**5**	**5**
						40	100	100	20	20
						100	100	200	200	10
Sn	-3 (?)	-4	-4	-4	-4	**1,000**	**1,000**	**1,000**	**1,000**	**100**
						5,000	5,000	5,000	5,000	1,000
						10	10	10	10	5
Sb	-3 (?)	–	–	–	–	**100**	**100**	**100**	**100**	**50**
						1,000	1,000	1,000	1,000	500
						1	1	1	1	1
I	high	high	high	high	high	**1**	**1**	**1**	**1**	**1**
						1	50	1	1	1
						100	100	60	200	1
Cs	high	high	high	high	high	**1,000**	**1,000**	**500**	**1,000**	**10**
						10,000	10,000	10,000	20,000	2,000
						10	20	20	20	5
Pb	-1	-1	0	-1	0	**50**	**50**	**50**	**50**	**20**
						200	500	500	500	100
						50	50	50	50	5
Ra	-2	-3	-1	3	-1	**500**	**500**	**500**	**500**	**50**
						5,000	5,000	5,000	5,000	500
						500	500	500	500	300
Th	-3	-4	-4	-4	-4	**5,000**	**5,000**	**5,000**	**5,000**	**1,000**
						10,000	10,000	10,000	50,000	5,000
						10	20	5	50	10
U	-3	-3	-5	high	high	**50**	**50**	**40**	**200**	**20**
						500	1,000	200	5,000	60
						10	10	10	10	10
Np	-3	-4	-4	-2	-1	**100**	**100**	**100**	**100**	**50**
						500	500	500	400	300
						10	100	50	500	10
Pu	-3	-5	-4	-5	-3	**200**	**500**	**200**	**1,000**	**200**
						5,000	5,000	5,000	20,000	10,000
						500	60	300	200	300
Am	-4 (?)	-8	-5	-8	-5	**3,000**	**500**	**1,000**	**800**	**1,000**
						50,000	50,000	50,000	50,000	5,000
						200	100	100	200	200
Cm	-3 (?)	–	–	–	–	**2,000**	**500**	**500**	**2,000**	**1,000**
						10,000	10,000	10,000	20,000	3,000

NOTE: See Section 7.10 for comments, explanation, and bibliographic references. Boldfaced values of retardation factors are those that K. B. Krauskopf considers to be suitably conservative for predicting the performance of conceptual repositories (Chapter 9).

SOURCE: Compiled by K. B. Krauskopf, Stanford University.

7.3. SUMMARY OF HYDROLOGIC PROPERTIES OF CANDIDATE ROCK TYPES

A summary of the hydrologic properties of selected candidate host rocks and of the surrounding media that could affect the hydrogeologic transport of radionuclides to the environment appears in Table 9-4. Reported values were derived from a variety of sources and by a variety of individuals using different measurement techniques. Often, only a few boreholes were available for permeability tests, with only eight, for example, at the Yucca Mountain tuff site (see Section 7.8). These data are preliminary until more extensive testing is completed, using old and new boreholes specifically designed for aquifer tests. For example, in evaporite basins (see Section 7.5), rocks may be so poorly permeable that months to years are required for shut-in pressures to equilibrate. Yields may be so low that specially designed pumps are needed to maintain constant pumping stresses at rates low enough to prevent premature, complete borehole dewatering. Similarly, shut-in pressures within nested piezometers (stacked observation wells, each completed within a single aquifer of a multiple sequence) may recover so slowly that recovery tests require one or more years to readjust. Water level data also must be adjusted for density differences when computing equivalent freshwater hydraulic heads.

Groundwater digital models to simulate groundwater flow require potentiometric surface data for verification and calibration before radionuclide transport predictions can be made. Parameter-estimating routines used in groundwater modeling must be tied to as many "fixed" or known values as possible to improve reliability. Typically, water level configuration data are used to match observed data against predicted data, and trial and error is used to adjust input data such as effective porosity, hydraulic conductivity, and boundary conditions until a satisfactory match has been obtained.

Values of hydraulic parameters from a given rock mass vary with the method of testing. For example, Bair (1980) and Bair and Parizek (1981) found that conductivity data obtained from rock cores typically are lowest when compared with other methods, largely because of the dependence on small rock samples that ignore contributions by fractures, rock partings, etc. Packer tests and other in-situ single-well tests provide higher values that include the effects of secondary openings. Multiple-well tests, where one serves as the pumped well and one or more other wells are used to observe water level responses, provide still higher conductivity values because they involve larger masses of rock. Finally, sensitivity-test-derived data from modeling forecasts may show still higher values. A spread in hydraulic conductivity as high as five orders of magnitude was observed by Bair (1980) and Bair and Parizek (1981) for folded and faulted Pennsylvanian coal-bearing rocks in an anthracite field in Pennsylvania. J. P. Bredehoeft (U.S. Geological Survey, personal communication to D. E. White, 1981) reports a spread of nearly seven orders of magnitude for shale-confining beds above the Dakota Sandstone of South Dakota. Similar relationships are to be expected for host rocks under study as repositories.

The main point here is that a range in values will exist for a rock mass, depending on where drill holes are placed and how they are tested. An average permeability or porosity value does not correctly characterize a rock mass. Rather, the best estimate of the spatial distribution of these values is needed for near-field and far-field groundwater modeling studies in nuclide migration predictions. Also, site-specific rather than generic data are needed because local variability in rock masses will dominate over homogeneity and isotropy.

One important structural feature, zones of fracture concentration, can greatly add to the permeability of brittle rocks, such as anhydrite, shale, sandstone, basalt, granite, and tuff. These nearly vertical zones of multiple fractures may be only a few meters wide, as revealed by fracture traces, or a few meters to perhaps 4 km wide, as revealed by lineaments of various lengths. Their importance in water velocity determinations is summarized in Table 7-2, as determined for granite. No similar data exist for basalt at Hanford, for tuff at Yucca Mountain, for dolomites within the Rustler Formation (Culebra and Magenta dolomite aquifers) at WIPP, or for other brittle rocks of the Palo Duro Basin of west Texas. Such structures may increase hydraulic conductivities of brittle rock by three to seven orders of magnitude (or even higher for soluble dolomite that tends to dissolve in a differential manner along zones of fracture concentration to produce karstlike channels).

Intentional location of exploratory holes on such structures requires site-specific data. At Yucca Mountain, for example, such structures might not only increase flow velocities between the proposed repository and Fortymile Wash, but also provide a more direct line of transport along one or more zones of multiple fracture. At Hanford, such structures, if present, may enhance vertical hydraulic

TABLE 7-2 Hydraulic Properties of Different Hydrogeologic Structures for the Reference Repository Site Area in Granite at Depths of 1 m and 500 m

Description	Width (m)	Conductivity (m/s)	Porosity	Velocity (Unit Gradient) (m/s)	Water Travel Time for 1 km Under 0.001 Gradient (yr)
Rock mass					
1 m depth		10^{-7}	3×10^{-3}	3.33×10^{-5}	951
500 m depth		10^{-10}	10^{-5}	1×10^{-5}	3,170
First-order fracture zones (tension)					
1 m depth	50	10^{-5}	3×10^{-2}	3.33×10^{-4}	95
500 m depth		10^{-8}	10^{-4}	10^{-4}	317
Second-order fracture zones (tension)					
1 m depth	10	10^{-5}	3×10^{-2}	3.33×10^{-4}	95
500 m depth		10^{-8}	10^{-4}	10^{-4}	317
Second-order fracture zones (shear)					
1 m depth	20	5×10^{-7}	5×10^{-3}	1×10^{-4}	317
500 m depth		5×10^{-10}	1.7×10^{-5}	3×10^{-5}	1,057
Second-order fracture zones (compression)					
1 m depth	5	10^{-6}	5×10^{-3}	2×10^{-4}	159
500 m depth		10^{-9}	1.7×10^{-5}	6×10^{-5}	529

conductivities between the repository horizon and permeable interflow zones. At WIPP these structures can promote differential dissolution of salt from concentrated flow of groundwater within brittle anhydrite and dolomite as well as differential subsidence of dolomite, thereby increasing its permeability.

For these various reasons, the hydraulic parameters listed in Table 9-4 must be regarded as useful for preliminary studies. An ongoing hydrogeologic exploration and monitoring effort is needed for each proposed repository site on both near-field and far-field scales. Also, state of the art in-situ experiments are needed, such as the ventilator experiments being conducted at Stripa, to evaluate more adequately the hydrologic characteristics of large masses of rock. Similar field-scale experiments also are needed to characterize dispersion coefficients of repository host rocks and adjacent strata and to define the nature and significance of their anisotropy and response to thermal stresses. The significance of secondary rock fractures on sorption of key nuclides also must be further investigated. The more highly fractured rocks revealed by fracture traces and lineaments may have higher sorption characteristics than the dense adjacent rock masses because of differences in surface area and abundance of nuclide-sorbing secondary minerals and alteration products.

7.4. GENERIC CHARACTERISTICS OF CANDIDATE HOST-ROCK TYPES

Each rock type has certain "generic" advantages and disadvantages, but the reader is reminded that no repository can be evaluated without site-specific hydrogeologic, hydrochemical, and structural data.

7.4.1. Bedded Salt

Of potential repository rocks, bedded rock salt has been the most thoroughly studied for the longest time. Favorable properties normally to be expected include high thermal conductivity (which minimizes temperatures for a given waste loading); very low permeability (in the absence of hydrologic discontinuities); no moving groundwater for hydrogeologic transport of radionuclides; abundant availability in thick widespread masses, with extensive lateral homogeneity; plasticity that permits tight closure and self-sealing at repository depths; and low cost of mining.

Bedded salt deposits are never pure sodium chloride. They contain variable proportions of other saline and rock-silicate minerals, which tend to maintain roughly uniform proportions parallel to original layers of deposition but differ greatly in earlier and later deposited layers. Water contents are very low in massive salt (ca. one percent or less) but are generally higher in interbeds containing other minerals and in cross-cutting breccia pipes and other discontinuities. The sorptive capacity of salt is the lowest of all the candidate rock types, but interbeds higher in silicate minerals (cf. Table 7-1) have higher capacities. The thermal conductivity of pure salt is the highest of all candidate rocks, about threefold higher than that of granitoids.

Salt is highly soluble in water; hence, salt is always associated with saline waters (saturated or nearly saturated chloride brines) that are highly corrosive to metals, especially at high temperatures. Many metals with low solubilities in dilute waters (less than 0.1 percent dissolved matter) are highly soluble in thermal brines as metal-chloride complexes. Some salts and associated brines are attractive sources of common salt, potassium, bromine, and other constituents. Most sedimentary basins containing salt deposits are also attractive targets for nearby oil and gas at shallow and great depths. Because of their high salinities, the waters of salt deposits are not normally attractive for most domestic and industrial uses. However, salt deposits may be overlain by aquifers containing potable water.

The plasticity of salt, increasing at higher temperatures and higher lithostatic pressures, can be an advantage in tending to heal fractures and excavated openings. However, it will create some problems in maintaining open spaces over time intervals required to emplace waste and to backfill emplacement rooms. It will also create especially difficult problems in keeping emplacement rooms open for decades, if direct access for retrieval is required. If present recovery requirements are relaxed or eliminated, the plastic behavior of salt has great advantages in promoting self-sealing and isolation. The thermal expansion of salt is nearly threefold greater than that of other potential repository rocks. Thus, with thermal loading, vertical uplift and induced stresses in more brittle surrounding rocks may become important.

The site for the Waste Isolation Pilot Plant in New Mexico has been the most thoroughly studied. It is being developed as a repository for defense transuranic waste and is not a candidate for a commercial high-level waste repository.

Issues of concern include assessment of the significance of karst processes and landform development on anisotropic permeability in overlying aquifers; identification, genesis, and significance of breccia pipes; potential for strata-bound dissolution within the salt section; brine migration; potential occurrence, origin, and significance of pressurized brine reservoirs; resource denial; and the potential for human intrusion, both accidental--during search for resources associated with bedded salt--and intentional.

7.4.2. Salt Domes

Salt domes are large masses that have been forced upward through overlying rocks by the plastic flow of thick bedded salts initially at greater depths, pressures, and temperatures. The more important domes of interest are restricted to the Gulf Coastal region of the United States. Large volumes of domal salt may have uniform properties, but their tops, borders, and any internal discontinuities are nonuniform. Domal salt deposits are similar in many respects to bedded salts but are isolated in their areal distribution to pluglike bodies with somewhat variable margins and diameters. Internally, domes tend to be structurally more complex (in places almost "homogenized"), but not on their tops and borders, where extensive dissolution has generally occurred. Also, adjacent strata penetrated by domes tend to be faulted and folded in a complex manner, making their physical continuity and effects on water flow more difficult to characterize. This makes the prediction of radionuclide transport less certain. Fluid inclusions in domal salt tend to be smaller and lower in total volume than those in bedded salt that has not been subjected to as much pressure and compactive force. However, because of the very high thermal conductivity of salt, heat from depth is focused upward through salt domes. Thus, temperatures are generally higher in the upper parts of buried domes, and water contents are much lower relative to bedded salts and other rocks at similar depths. Up-warped and faulted sedimentary strata on the margins and tops of salt domes are especially attractive for oil and gas exploration, and their hydrology is more complicated and uncertain. Domes are also attractive as a source of nearly pure rock salt, as sites of deposits of elemental sulfur formed by reduction of calcium sulfate on the dome margins, and as sites for underground excavations for storing oil, gas, and other fluids.

Domes are limited in number and easy to locate, making them more likely targets for future human intrusion. Because they penetrate various water-yielding sedimentary rocks to great depths, they are subject to dissolution near their margins and may be surrounded by both potable water at shallow depths and more mineralized groundwater at greater depths. Their locations near the Gulf Coast are favorable in that any minor amounts of radionuclides transported in highly saline groundwater must be greatly diluted at shallow levels before becoming potable or will be discharged directly into the sea for ultimate dilution. Despite the dynamic piercement origin of salt domes, various lines of evidence document their present structural stability. Long-term future changes in sea level may alter details of groundwater flow systems operating in enclosing coastal plain aquifers by increasing or decreasing path lengths and water velocities, but the consequences of such possible changes can be adequately bounded.

7.4.3. Granitoids*

Granitoids and similar crystalline igneous and metamorphic rocks are the most abundant rocks of the earth's subareal upper crust. True granites constitute only a small percentage of the physically similar rocks included here as granitoids. Granitoids underlie much of the United States near the surface and at shallow depths, especially in tectonically stable areas and in the cores of many mountain ranges.

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*Details appear in a separate background paper on granitoids by D. E. White (in press).

These rocks have some outstanding attributes for waste repositories because they are generally strong, structurally and chemically stable, low in initial porosity and permeability (but with superposed faults and fractures), and often nearly homogeneous in three dimensions over distances of hundred of meters. Repositories are planned in these rocks in Canada and Sweden, and granitoids are leading candidates in Great Britain and other countries, in part because of the scarcity of other favorable candidate rocks. Study of these rocks in the United States has not been vigorous, largely because of early focus on other rock types, but also because of skepticism that low overall permeability and long groundwater flow paths can be identified.

The water contents of granitoid rocks are low, commonly 1 to 2 percent, and are concentrated mainly in fractures and hydrous silicate minerals. Permeable conduits are largely confined to discontinuities, i.e., fractures, zones of fracture concentrations, faults, and breccia zones (Table 7-2). The pattern and location of these discontinuities can be mapped in surface outcrops, underground openings, boreholes, and in part by geophysical methods but are difficult to extrapolate with confidence to unexposed portions of the rock mass. Nearly vertical zones of fracture concentration are betrayed by lineaments and fracture traces observed on aerial photographs, by satellite imagery, by geophysical surveying techniques, and by alteration minerals. Depending upon the size of these zones of fracture concentrations and their tensional, shear, or compressional origins, hydraulic conductivity data obtained in Sweden were found to range from as high as 10^{-5} to 10^{-8} m/s to as low as 10^{-7} to 10^{-10} m/s, with intervening less fractured masses ranging below 10^{-10} m/s. More site-specific data are still lacking in the United States, but values from 10^{-14} to 5×10^{-9} have been reported in the literature for unfractured metamorphic and igneous rocks and 10^{-8} to 5×10^{-3} for fractured igneous and metamorphic rocks (Freeze and Cherry 1979). In fractures, flow is controlled largely by overall aperture width, the flow varying with the cube of the width. Unfortunately, aperture width varies locally and is difficult to measure reliably in a rock mass, being best inferred from flow tests.

Primary minerals of granitoids, especially quartz and feldspars, when fresh are low in sorption capacity for radionuclides. However, clays and other products of alteration of primary silicate minerals are generally present, especially in and adjacent to fractures (which accounts for some of the high values in Table 7-1). These suggest higher initial permeability by their presence and should provide modest sorption capacities, regardless of net changes in permeability. Typical shallow granitoid waters are dilute, slightly oxidizing, and nearly neutral in pH. Deep slowly moving waters are likely also to be neutral, slightly reducing, and in some localities moderately to highly saline, perhaps having migrated from other environments.

Of all potential repository rocks, granitoids are most likely to be homogenous in vertical as well as horizontal extensions, with some consequent advantages but some disadvantages. Laboratory values of permeability are notably unreliable because the natural fractures are not adequately represented in cohesive laboratory samples. In-situ values at particular sites are not predictable within a factor of 10^5 (Brace 1980) and permeability may not decrease in a predictable manner within measured depths. A crustal average is about 10 millidarcies (1 darcy = 9.61 x 10^{-4} cm/s or 3.03 x 10^2 m/yr). Of all repository candidates, vertical permeability in granitoid rocks is most likely to equal or exceed horizontal permeability because lithostatic load tends to close fractures with large horizontal dimensions.

The near-surface waters of most granitoid rocks consist of seepage from rain or snow that has reacted slightly with nearly insoluble rock-silicate minerals. However, some pore waters at depth are saline and gravitationally stable, perhaps having migrated from other environments, such as from marine sediments with saline pore waters. At depths near 1,000 m, salinities and water compositions will be unfavorable enough to discourage accidental human intrusion during the search for adequate water supply.

Granitoid rocks that underlie most of the tectonically stable areas of the United States are more than 0.6 billion years old, but crystalline rocks in many tectonically active areas, especially in the western United States, are commonly tens to hundreds of millions of years old. Some granitoids have mineral deposits near their intrusive margins and, less frequently, internally. Economically attractive groundwater tends to be concentrated at shallow depths; hence, deep exploration for potable water is highly unlikely. Granite is exposed in and near some coastal areas and on islands, so that any escaping radionuclides could be diluted directly in the ocean. Large masses also occur in northern regions of the United States that were previously glaciated and are highly likely (probability of 1) to be subjected to renewed glaciation within 100,000 to 200,000 years. Probable consequences of renewed glaciation on the performance of granite repositories have not been adequately addressed.

An issue of some concern for granitoids of the eastern United States is deep geophysical data suggesting thrust-fault displacement of granitoids over now-concealed Paleozoic sedimentary rocks that may contain hydrocarbon resources. Deep geophysical surveys and test drilling are necessary to test this possibility.

The thermal conductivity of granitoid rocks is intermediate among repository candidates--lower than that of salt and anhydrite ($CaSO_4$) but higher than that of shale, volcanic tuffs, basalt, water-unsaturated alluvium, and most rocks above the water table.

7.4.4. Basalt

Basalt lava flows may occur as thick accumulations, i.e., "flood basalts," especially in Washington, Oregon, and Idaho. Many individual flows are at least 10 m thick, and one is 150 m thick. Initially, all had permeable tops and bottoms brecciated by flow movement, and many flows are bounded by interflow sediments of high permeability. With time and flow of pore water, commonly mildly thermal, basalt flows and their interbeds generally become less permeable, i.e, self-sealed, from deposition of secondary minerals, especially clays and zeolites, as described for rhyolitic rocks (Keith et al. 1978). However, worldwide experience has shown that basalts, especially young basalts, are generally more favorable for developing water supplies--much more so than granitoid rocks and shales. A major reason for considering basalt for repositories is its abundance in federal land near Hanford, Washington, and the Idaho National Engineering Laboratory (INEL) and not its overall favorable characteristics.

Basalt, because of its strength and interlocking of fracture blocks, is generally favorable for maintaining unsupported mined openings. Its secondary clay minerals and zeolites have high sorption capacities, as shown in Table 7-1, thus providing a potential for inhibiting migration of many radionuclides. Another major favorable chemical characteristic of most basalt is its strong reducing capacity, i.e., low Eh, of deep pore waters, buffered by ferrous iron that is more abundant than in other candidate repository hosts. Most radionuclides are least soluble in reducing environments, as indicated in Table 7-1. Thick basalt accumulations are unlikely sources for mineral and energy resources, but concealed resources may exist in underlying rocks.

Secondary joints and vertical zones of fracture concentrations and faults that provide vertical communication between aquifers located in interflow zones are difficult to locate and will require inclined drilling, development of mine openings, and specially designed aquifer tests.

7.4.5. Rhyolite Tuffs

Rhyolite tuffs, which are explosively erupted volcanic rocks high in silica, have some favorable characteristics for repositories. Some ash-flow tuffs were so thick and hot when erupted (600°C to 1,000°C) that their siliceous glass fragments deformed plastically, forming dense "welded" tuff. Other ash flows and "air-fall" tuffs retained their initial high porosity, but initial glass has commonly altered at low temperatures to zeolites and clays with high sorption capacities. Such minerals are commonly absent in welded tuffs of nearly identical chemical composition, which devitrified from natural glass at high temperature to more stable silica and silicate minerals. Silicic tuffs are generally low in iron content, most of which has been oxidized to ferric iron. Thus, their pore waters are likely to be dilute, oxidized, and unfavorable for long-term retention of uranium and other radionuclides of low solubility in reducing environments (cf. Table 7-1). However, they may have offsetting advantages in the high sorption capacities of associated amorphous manganese and iron oxides, clays, and zeolites.

Tuffs are relatively homogeneous in their original horizontal dimensions but are generally very heterogeneous vertically, with each erupted layer differing in porosity, permeability, strength, and extent of devitrification and sorption capacities. Also, permeable water-sorted "volcaniclastic" sedimentary rocks commonly form interbeds above or below uniform tuffs, and individual units have been faulted and fractured in response to tectonic activity. Thermal conductivities of all tuffs are relatively low, especially when porous but water unsaturated. Most silicic tuffs are sufficiently strong after welding, devitrification, and cementation to maintain stable mined openings. Silicic tuffs frequently occur in the western United States in regions favorable for mineral resources. Oil and gas seldom occur in the tuffs but may occur in underlying sedimentary rocks.

Where present in fault block mountains, as at the Nevada Test Site, tuff may be surrounded by highly permeable sand and gravel deposits containing groundwater of favorable quality. Water flow within unsaturated and saturated welded and unwelded tuff, cut by zones of fracture concentration, joints, and faults, will be difficult to characterize in detail; hence, estimated water travel times to adjacent alluvial aquifers are likely to vary by 10^3 to 10^4 years. Permeability and porosity values of these secondary rock openings might be better characterized by testing with inclined borings that cut steeply dipping structures missed by vertical drill holes.

7.4.6. Water-Unsaturated Alluvium Over a Regional Aquifer

Water-unsaturated alluvium underlain by a regional aquifer is a combination with some outstanding properties for a repository. The water-unsaturated zone in some arid parts of the Great Basin is as much as 600 m deep below ground (Winograd 1981). The downward rate of percolation of surface precipitation is very low in the present semiarid climate and probably can be engineered to low vertical flow rates by appropriate use of clays of low permeability. Clays and zeolites are abundant in alluvium of the Nevada Test Site, providing natural sorbants for most radionuclides (Table 7-1). Altered volcanic rocks within and below the unsaturated zone are also potential sorbants. Some basins are underlain by deep regional permeable carbonate aquifers characterized by long travel times to the biosphere, times that would delay the release of radionuclides to the environment and provide more opportunity for dispersion and dilution. None of these groundwater flow systems discharges to significant surface streams.

Young alluvium is highly porous, commonly 30 percent or more, and is uncemented and unable to maintain unsupported underground openings. Old alluvium commonly is cemented enough to maintain unsupported openings (Winograd 1981).

Disadvantages include the low thermal conductivities of water-unsaturated materials relative to their saturated equivalents; differences increase roughly in proportion to porosity. Also, pore vapor pressure is nearly atmospheric, seriously limiting permissible thermal loading of the repository because clays and zeolites lose much water near 100°C at atmospheric pressure. Water-unsaturated alluvium is unlikely to contain mineral resources, but underlying rocks are attractive targets for concealed mineral resources in many western areas. Also, the saturated zone, if not too deep, is a potential source of potable water and would rise closer to the surface during pluvial periods. An explosion crater from the Sedan nuclear test in Nevada alluvium has been proposed for a shallow repository (Winograd 1981) that might be especially attractive for bulky low-level waste. This type of repository is not considered further in this report.

7.4.7. Granitoid and Metamorphic Rocks Under Regional Sedimentary Aquifers

Granitoid and metamorphic rocks overlain by regional sedimentary aquifers are important variants of the granitoids discussed in Section 7.4.3, as suggested by Bredehoeft and Maini (1981). This hybrid combination uses the most favorable characteristics of two different rock groups while avoiding the major disadvantages of each. The sedimentary blanket must contain at least one permeable aquifer whose flow characteristics are either known or can be determined and modeled by conventional theory and technology. In many locations the migration path in the aquifer is long, its flow rate to the biosphere is very low, its pressure gradient is low and predictable, and clays and other minerals in the aquifer serve as radionuclide sorbants, as indicated in Table 7-1.

The underlying crystalline rocks are generally competent and favorable for repository construction. Even where crystalline rocks contain diverse permeable fractures, faults, and breccia zones, their overall mass permeability is less than that of a permeable regional aquifer; pressure gradients in the crystalline rocks consequently should be low, and flow rates nearly zero. Also, in many places the deep waters of both sedimentary and crystalline rocks are saline, high in density, and gravitationally stable relative to the shallow dilute waters. Moreover, with extremely low flushing rates, the deep waters are generally reducing through reactions with ferrous minerals and/or hydrocarbons, thereby ensuring low solubilities for most radionuclides. Thus, many characteristics of this hybrid environment provide multiple natural barriers to impede or prevent the escape of radionuclides. The assumptions upon which the model is based must be tested by drilling. Careful analysis of hydraulic heads and water compositions with depth will confirm or deny their validity.

Crystalline rocks are very unfavorable for oil and gas, and large masses of low mineral potential can be delineated from drill hole and geophysical data. The saline pore waters of most deep environments, generally 500 to 1,000 m deep or more, are unfavorable for present domestic and industrial purposes. Many parts of the United States are likely to contain some favorable sites with this hybrid environment.

In time, radionuclides located in a coastal site that might escape these rocks might seep slowly into the ocean for dilution and possible incorporation at low levels into the food chain. The consequences of heavy groundwater pumpage from overlying coastal aquifers or of long-term changes in sea level on repository performance can be adequately bounded.

7.5. AN EVALUATION OF SALT

7.5.1. Geologic Features of Bedded Salt

Bedded salt deposits of Permian age underlie much of southeast New Mexico, West Texas, southeast Utah, and southwest Colorado. No salt deposits are pure sodium chloride; some cyclic layering is characteristic, consisting of nearly pure salt along with varying proportions of sulfates, carbonates, or clastic silicate minerals that are largely clays. Cyclic spacing of beds is commonly on the order of tenths of meters to several meters, but in places almost pure homogeneous beds of sodium chloride are many meters thick.

Pure salt contains no water, but natural bedded salts always contain some water in hydrated minerals such as gypsum and clays and as fluid inclusions in salt crystals. The total water content is about 0.5 percent. The possible effects of these fluid inclusions on repository performance are discussed in Section 7.5.2.

The most intensive studies of bedded salts have involved the site of the Waste Isolation Pilot Plant in the Delaware Basin of southeast New Mexico. This project is designed for geologic disposal of transuranic radioactive waste from the defense program and not for high-level waste. Plans include, however, the temporary placement of some high-level wastes for experimental purposes. This site is now in its validation stage, with two shafts already sunk to the proposed repository horizon in the Salado Formation at a depth of about 650 m. Tunnels from these shafts will provide locations for in-situ experiments and demonstrations. Geologic characteristics of this site are described in a Sandia National Laboratories report (Powers et al. 1978), which provides insight into the properties of bedded salt that may be developed at other locations for commercial high-level waste. The areas under active consideration for a commercial-waste repository are in West Texas and Utah.

Thick bedded salt deposits have many well-known favorable attributes as repositories, as summarized in Sections 7.4.1 and 7.4.2. Among the undesirable characteristics of bedded salt are high solubility, plasticity, corrosive nature of associated brines, irregular distribution of potentially permeable cross-formational breccia pipes, and possible occurrence of pressurized brine reservoirs. Also, beds of nearly pure halite are interbedded with strata containing other evaporite minerals and silicate-bearing sediments of variable water content. These more brittle beds may contain water-yielding fractures still of uncertain significance to repository performance.

Permian salt deposits, in spite of their high solubility, have persisted for 200 million years where buried deep enough or in favorable central parts of basins that are sufficiently isolated from actively circulating groundwater. However, these salt deposits are undergoing dissolution wherever exposed to flowing salt-unsaturated water on their tops, margins, and bottoms. Much is now known about dissolution loci and rates of present dissolution.

Extensive bedded salts occur northeast of the WIPP site in the Anadarko, Palo Duro, and Dalhart basins of West Texas and in the Paradox

Basin of southeastern Utah. In many places the shallow bedded salts have been dissolved, commonly with recognizable dissolution fronts that extend downward and laterally into the margins of the deeper salt-rich basins. These dissolution fronts are commonly marked on the surface by topographic escarpments, breccia pipes,* collapse sinks, salt pans, seeps, and saline springs; the latter are direct manifestations of active subsurface salt dissolution. The solute load in streams draining the main topographic basin of eastern New Mexico and the Texas panhandle has been computed, expressed as mean annual solute loads and also as annual rates of horizontal and vertical dissolution. A steady eastward decline of heads in the deep brine systems of eastern New Mexico and the Texas panhandle suggests that regional hydraulic connections permit flows between outcropping recharge areas in New Mexico and discharge areas in north-central Texas and western Oklahoma. Path lengths for groundwater flow within this regional system are as much as 160 to 260 km. Reliable travel times are not yet available.

In the Dalhart and Anadarko basins, salt is limited in areal extent because of modern and ancient dissolution. The San Andres Formation of the Palo Duro Basin contains the thickest relatively pure salt beds that exist at depths of 300 to 900 m over large areas. Dissolution has occurred at depths of 150 to 330 m where San Andres salt is shallow, and to depths as great as 1,200 m where the dissolution front is deep.

The Permian bedded salts contain potash deposits, typical only of end-stage chloride evaporites, primarily in the Delaware Basin of southeast New Mexico, 15 to 30 km north to northwest of the WIPP site. No significant concentrations of potash salts are yet known in several of the salt basins of West Texas.

The process of selecting specific repository sites in the bedded salts of West Texas or Utah is now under way. In the selection process, special attention should be given to structural simplicity, adequate depth, apparent absence of objectionable dissolution features in large areas, and low potential for potash and hydrocarbon resources. Sites near escarpments, known zones of collapse features, and flowing saline waters obviously should be avoided. Areas of structural complexity and deformed bedded salts are clearly inferior to broad areas of horizontal or gently dipping beds at depths near or somewhat greater than 0.5 km.

Drilling near the WIPP site, as well as in potash mines north of the site, have revealed puzzling occurrences of saturated brine pockets, some of which are geopressured, show a variety of chemical and isotopic compositions, and have flowed for many days or weeks. No similar occurrences are yet known in several of the West Texas basins.

Oil and gas do not normally occur in commercial quantities in bedded salt deposits but frequently are present in underlying sedimentary rocks as well as on the margins and caps of salt domes and diapirs. The study of candidate repository sites must include careful assessment of whole sedimentary basins, selecting those parts with favorable characteristics

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*Features referred to as breccia pipes in West Texas appear to be smaller and less deep seated than known breccia pipes near the WIPP site.

but that are also least favorable for resources of fluid hydrocarbons, potash, coal, and groundwater. For this assessment, all available geologic and geophysical data, along with recorded past drilling and production, are obviously essential. In general, groundwater associated with bedded salts is unfavorable for domestic and industrial use, especially where less saline waters are available. However, poor quality water may be pumped from shallow aquifers for stock use, and these aquifers may contain water of more favorable quality during future pluvial periods.

A summary and evaluation of the geologic features of potential domal salt sites appear in the reports by Harwell et al. (1981) and by Martinez et al. (1978, 1979).

7.5.2. Migration of Brine Inclusions

Bedded and domal salt deposits contain small amounts of brine inclusions, located within the salt crystals and at grain boundaries. These inclusions can migrate through the salt under the influence of gradients in the chemical potential in the surrounding salt, such as the gradients in temperature due to repository heating. Some can migrate up the thermal gradient, collect in the space around the waste package, and cause corrosion. Additional migration may occur through microfractures in the salt, under the influence of local gradients in pressure. There has also been some concern about the possibility that the moving brine inclusions can contribute to the transport of radionuclides away from the waste package. The status of current knowledge on these issues of brine migration, resulting from extensive studies during the past 15 years, has been summarized by Jenks and Claiborne (1981) and more recently by Shefelbine (1982).

Inclusions in bedded salt crystals range in size from less than 1 mm to a few millimeters; most of the volume is contained within inclusions greater than about 0.5 mm. The inclusion content is less than about 0.5 percent by weight for samples from the Salado Formation of southeast New Mexico and less than about 0.25 percent for samples from the bedded salt of Lyons, Kansas. Data indicate smaller amounts of inclusions in domal salt than in bedded salt. Contents of less than 0.03 percent have been reported for samples from the Rayburn and Vacherie domes in Louisiana, and less than 0.2 percent for samples from the Avery Island domes, also in Louisiana.

In bedded salt the brine in inclusions is rich in $MgCl_2$ (about $2.44\underline{M}$), is saturated with NaCl (less than $2.0\underline{M}$) and $CaSO_4$ (about $0.03\underline{M}$), and contains appreciable amounts of Br^- (about $0.05\underline{M}$) and some K^+. In domal salt the brine seems to be mainly NaCl. The $MgCl_2$ in bedded salt inclusions is generally more corrosive than pure NaCl brine and must be considered in evaluating waste-package performance.

When a thermal gradient is imposed on salt containing all-liquid brine inclusions, the salt dissolves at the inclusion's higher-temperature surfaces and crystallizes from the inclusion liquid at the lower-temperature surfaces, and the inclusions migrate toward the region of higher temperature. An inclusion can migrate across a grain boundary, or it can move along the boundaries and through connected pores and/or microcracks. Inclusions that contain a gaseous phase can, under some conditions, move away from regions of high temperature. Based on the theories (Anthony and Cline 1972, Jenks 1979, Geguzin et al. 1975, Olander et al. 1981) that explain laboratory data for migration of brine inclusions in crystals, and based on the long-term stability of natural inclusions in the presence of geothermal gradients and gravity, it has been predicted that there will be no migration of brine inclusions in the region of lower-temperature gradients that will occur some distance from the waste package (Pigford 1982).

The predictive techniques of Jenks (1979) seem adequate for estimating the amount of migration of brine into the waste cavity during the first year or two after emplacement and for making conservative estimates for longer times. More precise predictions of longer-term rates and accumulation of brine in the waste cavity will depend on the presently uncertain effective permeability of the rock salt adjacent to the emplacement hole. Assuming that the migration rates predicted for the first two years are constant for the next 100 years, and assuming a repository in bedded salt with a temperature history similar to that shown in Chapter 6, Jenks and Claiborne (1981) estimate that about 25 liters of brine will accumulate in the waste cavity for a commercial high-level waste package, 8 liters for a spent-fuel package, and 1 liter for a package of defense high-level waste. For a repository in domal salt with 0.03 weight percent inclusions, the estimated 100-year accumulation is about 2.5 liters for commercial high-level waste, with correspondingly smaller amounts for spent fuel and defense waste. These estimates are likely to be conservatively high.

To avoid excessive accumulation of brine in the waste cavity, Jenks and Claiborne (1981) suggest the following considerations in site selection and in repository and waste-package design:

1. Locate the emplacement holes within the salt formation where there is a minimum amount of brine that could conceivably migrate from the salt into the emplacement hole.
2. Employ designs and practices such that the walls of the emplacement hole are maintained under compressive stresses. In order to accomplish this it may be necessary to employ canister dimensions and backfill materials that will minimize closure movement of the salt walls.
3. Avoid impurities in the rock salt that could yield water upon thermal or radiolytic decomposition and thus contribute to the amount of water that might migrate into the emplacement hole. Design the emplacement so that the maximum temperature of the salt walls does not exceed about 250°C in order to avoid possible decrepitation of salt, with release of trapped water.
4. Design so that the maximum pressure within the emplacement hole remains less than the compressive stresses on the salt walls, to reduce the development of microcracks within the salt.
5. Use a dessicant-barrier material as part of the backfill, with the general objective of absorbing water, shielding the metallic components from brine and corrosive vapors, and possibly sorbing radionuclides. Dessicants such as CaO or MgO in combination with sand

or crushed salt are estimated to absorb more than the predicted accumulation of brine.

Although the predictive techniques seem adequate for estimating brine release rate during the early times when the transport mechanisms are those characterized in the laboratory studies, more accurate predictions of longer-term releases will require longer-term in-situ experiments and better knowledge of the effects of radiation damage, radiolysis, and the effects of local stresses and reconsolidation of the salt on the effective permeability for migration from grain boundaries to the waste cavity. The importance of better predictions of longer-term release depends considerably on the design features of the repository and the waste package, e.g., the selection of the host salt, the waste loading and thermal gradients, the presence of a corrosion-resistant overpack, and the use of dessicants.

If an overpack is to be relied upon, and for emplacement in bedded salt, it is important to characterize the chemical composition and corrosion properties of the brine that can be released to the waste cavity. The process of migration through salt of increasing temperature can be expected to result in a composition of released brine that can be significantly different from that of the natural inclusions.

Because net transport of brine inclusions is into the waste cavity, outward migration being stopped by the low thermal gradient, the postulated transport of radionuclides via the migration of brine inclusions does not appear to be an important issue.

7.6. AN EVALUATION OF GRANITOID ROCKS

The principal challenges for granitoid repositories are:

o Can large volumes of low permeability be found at depths near 1 km?

o Are unsealed deep fractures sufficiently abundant and permeable to provide rapid escape to the environment? Can these larger fractures be identified and sealed effectively at reasonable cost and effort?

o Do old, nearly immobile saline brines exist at depth in many granitoid masses, sufficiently high in salinity and density so that even when moderately heated they will be gravitationally stable relative to shallow dilute waters?

Recent data from Canada and the United States (Frape and Fritz 1982; Fritz and Frape, in press) suggest that deep pore waters in stable areas of Precambrian rocks commonly have densities higher than sea water (ca. 1.025 to greater than 1.1 g/cm^3). Moreover, their chemical and isotopic signatures differ from all known presently forming brines; some and perhaps most of these brines are deficient in $^{18}O/^{16}O$ relative to deuterium and hydrogen and plot on the low-^{18}O side of meteoric waters. This is in striking contrast to their nearest equivalents in Paleozoic and younger rocks, suggesting that such waters are not forming now (or require long times to form). This implies great age, probably greater than 10^6 years, exceedingly low flow rates, and great gravitational stability. An additional advantage is their low indicated solubilities of SiO_2 (approximately 10 percent of dilute waters), possibly because of their low activities of H_2O. Siting of a repository below such a brine interface could impede or prevent thermal convection from repository heating and would also discourage human intrusion in the search for useful water supplies.

High-salinity brines are characteristic of adjacent deep sedimentary basins at depths commonly as little as 1 km (White et al. 1963, Graf et al. 1966). At favorable times through the geologic past, these dense sedimentary brines may have flowed into and displaced any less dense granitoid waters accessible through faults and fractures; once emplaced, these dense brines are displaced by shallower dilute meteoric water only by slow flushing and upward diffusion.

Granitoid rocks are not highly reactive to water-rock interaction; some of these rocks are moderately oxidized, but most are sufficiently high in ferrous to ferric ratios to be mildly reducing (cf. Table 7-1). With long contact and low flow rates, free oxygen of the water combines with the ferrous iron of the rocks. However, the reducing capacity of granitoids is generally much less than that of basaltic rocks. Granitoid rocks generally buffer associated pore waters to pH between 6 and 9 (White et al. 1963), generally nearer a pH of 6.

Sorption capacities of primary minerals of granitoids are low, but clay minerals and fine-grained micas are generally abundant enough to be effective sorbants of most radionuclides (Table 7-1).

A problem in constructing a repository in granitoid rocks is the effective sealing of vertical shafts. Construction-related vertical permeability may augment natural permeability through steep-dipping fractures, thus making the site unsuitable if the disturbed zone is extensive and cannot be sealed. One alternative is to construct the shafts off center with respect to the repository, preferably up the regional hydraulic gradient toward the recharge area. The vertical permeability directly above the repository, if otherwise suitably low, is not adversely affected, and the off-center disturbed volume is exposed only to normal low temperatures rather than to repository heating and potential convection.

A second alternative is to site repositories beyond the limits of outcropping granitoids, below thickening wedges of younger sedimentary cover. This hybrid alternative is considered separately in Section 7.9.

A subject of some concern is the potential for human intrusion. Water quality is unfavorable for water supplies, oil and gas rarely occur, and monotonous geophysical properties can help to discourage search for mineral resources.

7.7. AN EVALUATION OF BASALT LAVA FLOWS OF THE PASCO BASIN, WASHINGTON*

7.7.1. Geology and Tectonism

The Pasco Basin of south-central Washington is underlain by a thick series of three formations of basaltic lava flows (Figure 7-1), decreasing downward in permeability and in proportion of interbedded sediments. One of the candidate horizons, the Umtanum flow, is in the lowest (oldest) of the three formations, existing everywhere beneath the Hanford site. Near the axis of the Cold Creek syncline (Figure 7-1), the Umtanum flow is 65 ± 5 m thick and lies at a depth of 1,110 m below the ground surface (D. J. Brown, Rockwell Hanford Operations, personal communication to T. H. Pigford, 1982). Its rather uniform dense central zone, specifically proposed for the repository, is 47 ± 5 m thick and consists of fracture-bound columns that are commonly hexagonal and tend to be interlocking. The precise reference repository location has not yet been determined.

The region has been and probably still is undergoing north-south horizontal compression, warping the thick competent flows into sharply folded and faulted anticlines separated by broad open synclines. Strong horizontal compressive stress, probably at least twice the vertical stress, is the likely cause of a phenomenon called core discing (Chapter 6) because of the appearance of individual rock discs in cylindrical drill cores. Where most intensively developed, each disc is about 0.5 cm thick and appears like a uniformly warped poker chip. The most intense discing is found in thick lava flows at depths of 800 m and more. While core discing is an indicator of high in-situ stress, with the potential for rock bursting from the surfaces of mined openings, studies indicate that for the measured horizontal-to-vertical stress ratio in the Umtanum, and at the expected repository depth, rock bursting is not expected (D. J. Brown, Rockwell Hanford Operations, personal communication to T. H. Pigford, 1982).

The most critical geologic problems related to the repository host rock are:

- The present state of stress, with the potential problems of rock-burst phenomena discussed above and in Chapter 6. The effect of repository heating on the glass matrix of the Umtanum flow may also adversely affect the mechanical properties.
- The possible lack of lateral homogeneity of the Umtanum flow. Although the Umtanum is relatively thick for a basalt flow, initial and structural inhomogeneities may result in exposure of permeable flow boundaries during repository construction.
- The abundance and tightness of faults and fractures at the repository location and depth.

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*Details are found in a separate background paper on basalt lava flows by D. E. White (in press).

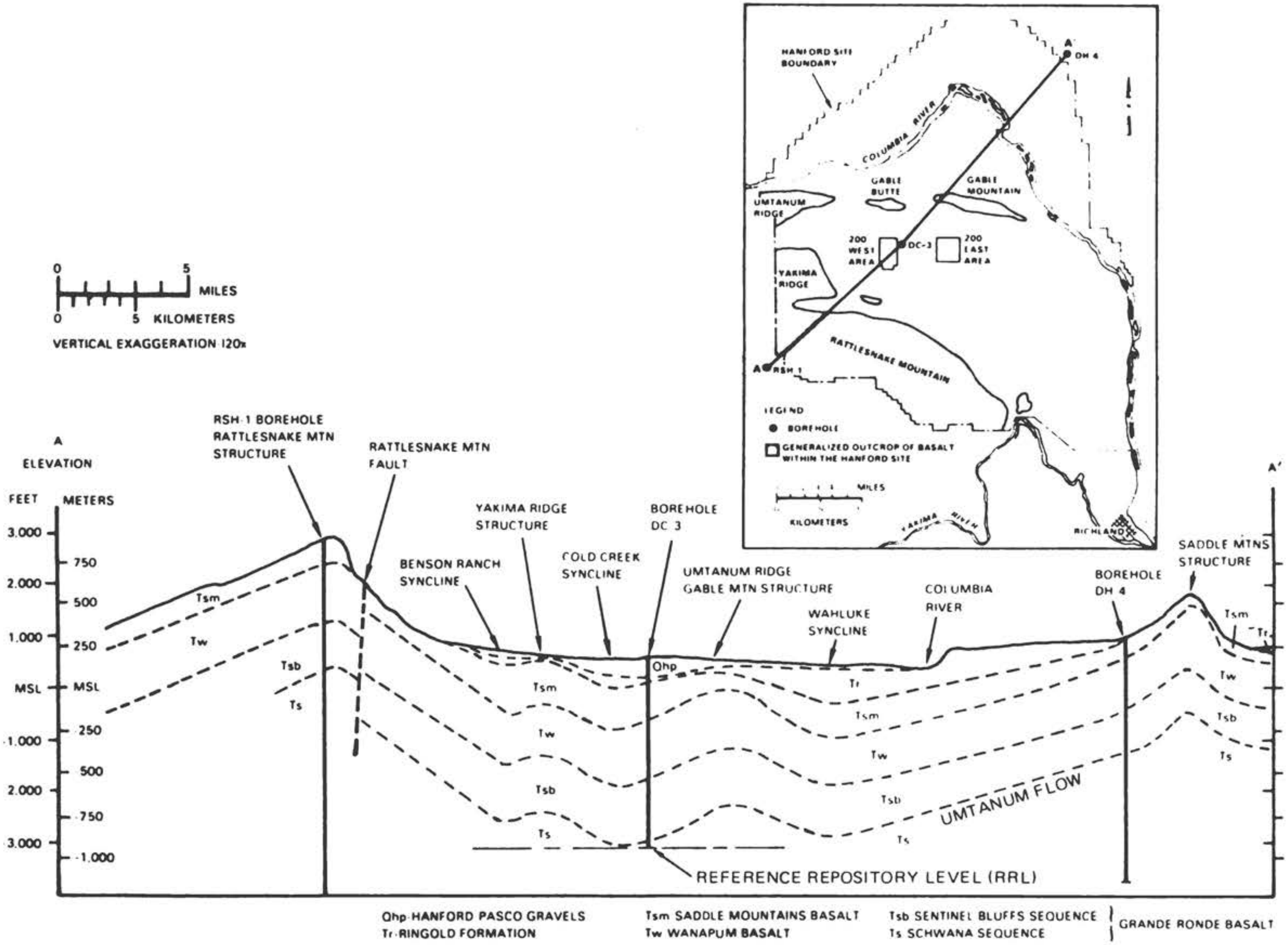


FIGURE 7-1 Generalized geologic cross section of the Pasco Basin, Washington. Source: Basalt Waste Isolation Project Staff (1981a).

o A geothermal temperature gradient of about 42°C/km, considerably above the continental average, resulting in an ambient repository temperature of 57°C or higher at the 1.1-km depth of the reference repository location (RRL).

Because of these problems the Umtanum host rock considered for the BWIP project is physically much less favorable than some other repository rock types discussed in this report. There remains uncertainty in the response of the Umtanum basalt to repository excavation, particularly with regard to the possibility of rock fractures from excavation communicating directly with adjacent permeable aquifers.

7.7.2. Hydrology

Hydrologically, the Columbia River basalts of the Pasco Basin are, in general, less permeable with depth. The central zones of Grande Ronde lava flows have hydraulic conductivities of 10^{-10} to 10^{-12} m/s; brecciated flow tops, i.e., interflows, typically range from 10^{-5} to 10^{-8} m/s, or about one to two orders of magnitude lower than in interflows and interbeds of the shallower basalts. Hydrologic and hydrochemical evidence, some of which appears in Figures 7-2, 7-3, 7-4, and 7-5, supports the concept of significant vertical upflow in the general area favored by Rockwell for the repository. Note that flow is most rapid from the reference repository level to the east and southeast (Figure 7-2), but salinities are highest near RRL (Figure 7-5). This is most reasonably explained by upflow of deep saline water near RRL. Table 7-3 and Figure 7-4 show the "normal" compositional differences. Distances to discharge areas along the Columbia River probably range from 10 to 35 km (the range from RRL to drill hole DC-6 and to Richland with a mean distance of about 20 km) along the Columbia River, according to most estimates. Somewhat greater estimates, 60 to 80 km, have been made by hydrologists.

Flow rates from recharge to discharge can be approximated from data on ages of deep waters from well DC-15 (Figure 7-4). These "apparent" ages, based on carbon-14, may be too old due to loss of early upstream carbonates precipitated as recharge waters reacted with rocks and pH increased; ages may also be too old due to cross-formational upflow of deeper water low or lacking in carbon-14. The apparent ages may also be too young from downward mixing with younger waters. In spite of these uncertainties, a relatively long age of 10,000 years or more for the Grande Ronde pore waters seems most probable, not only because of the apparent ages of DC-15 waters, but also from chemical evidence (Figure 7-4). Both the sulfate content and changes in deuterium-to-hydrogen ratio of Grande Ronde waters indicate their relative isolation, with less cross-formational flow than in the younger basalts. If an average water travel time of 10,000 years is assumed from RRL to the mean of the most probable zone of discharge (a distance of about 20 km), the indicated pore velocity is on the order of 2 m/yr, probably ranging from less than 0.4 to 4 m/yr.

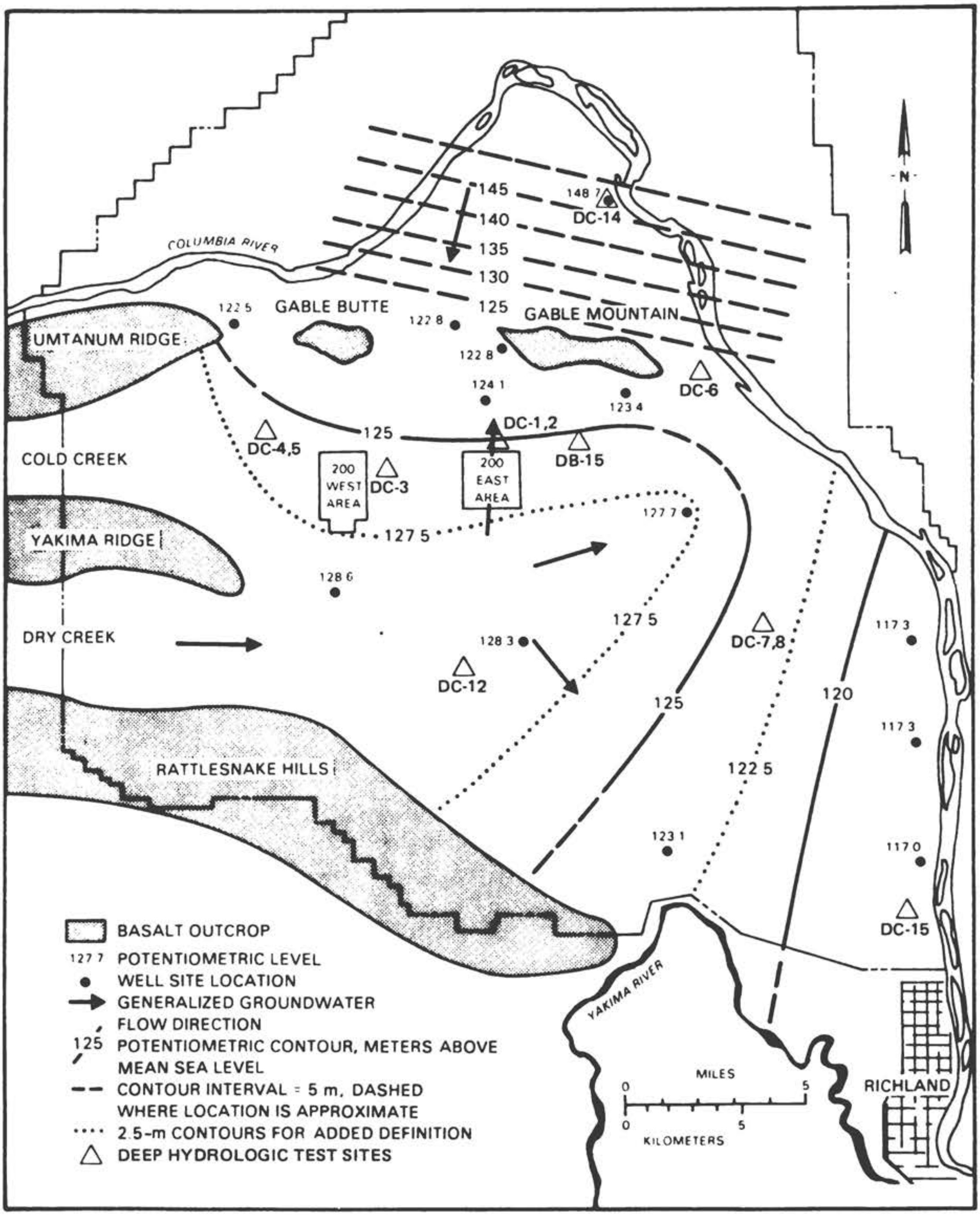


FIGURE 7-2 Potentiometric map and inferred flow direction of groundwater within the Mabton Interbed, one of the major aquifers of the Saddle Mountains basalt. This map also shows the locations of deep hydrologic test sites and the general area of the reference repository level. Source: Basalt Waste Isolation Project Staff (1981a).

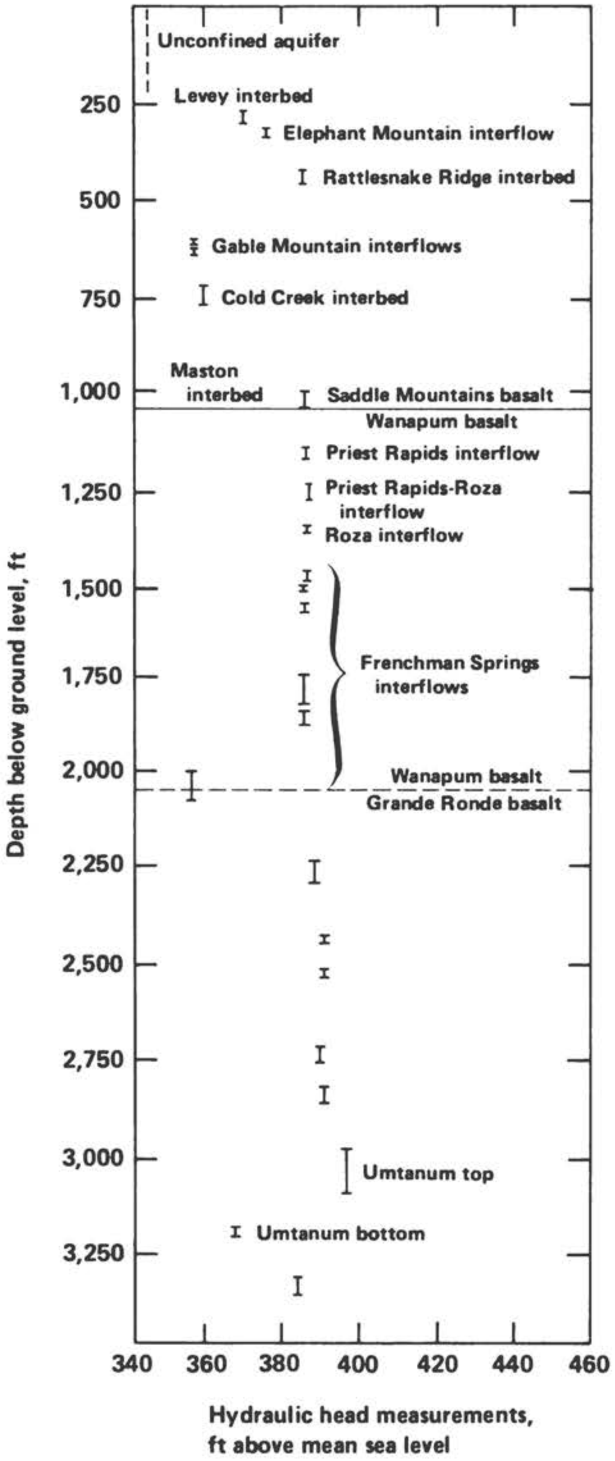


FIGURE 7-3 Hydraulic head measurements within the Columbia River basalt in borehole DC-15. Source: Basalt Waste Isolation Project Staff (1981b).

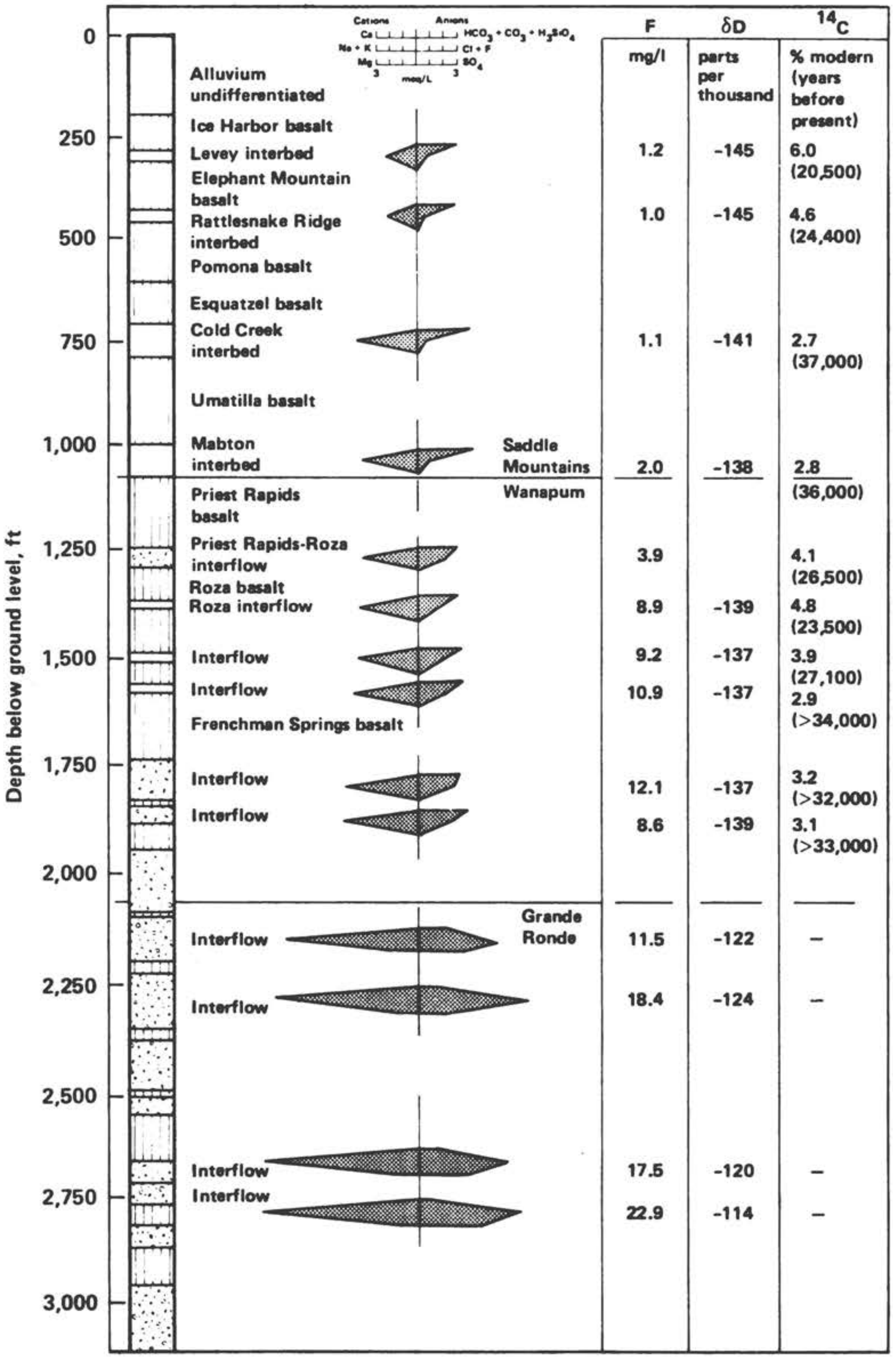


FIGURE 7-4 Selected hydrochemistry for borehole DC-15. Dashes indicate carbon source too small to obtain reasonable age date. Source: Basalt Waste Isolation Project Staff (1981b).

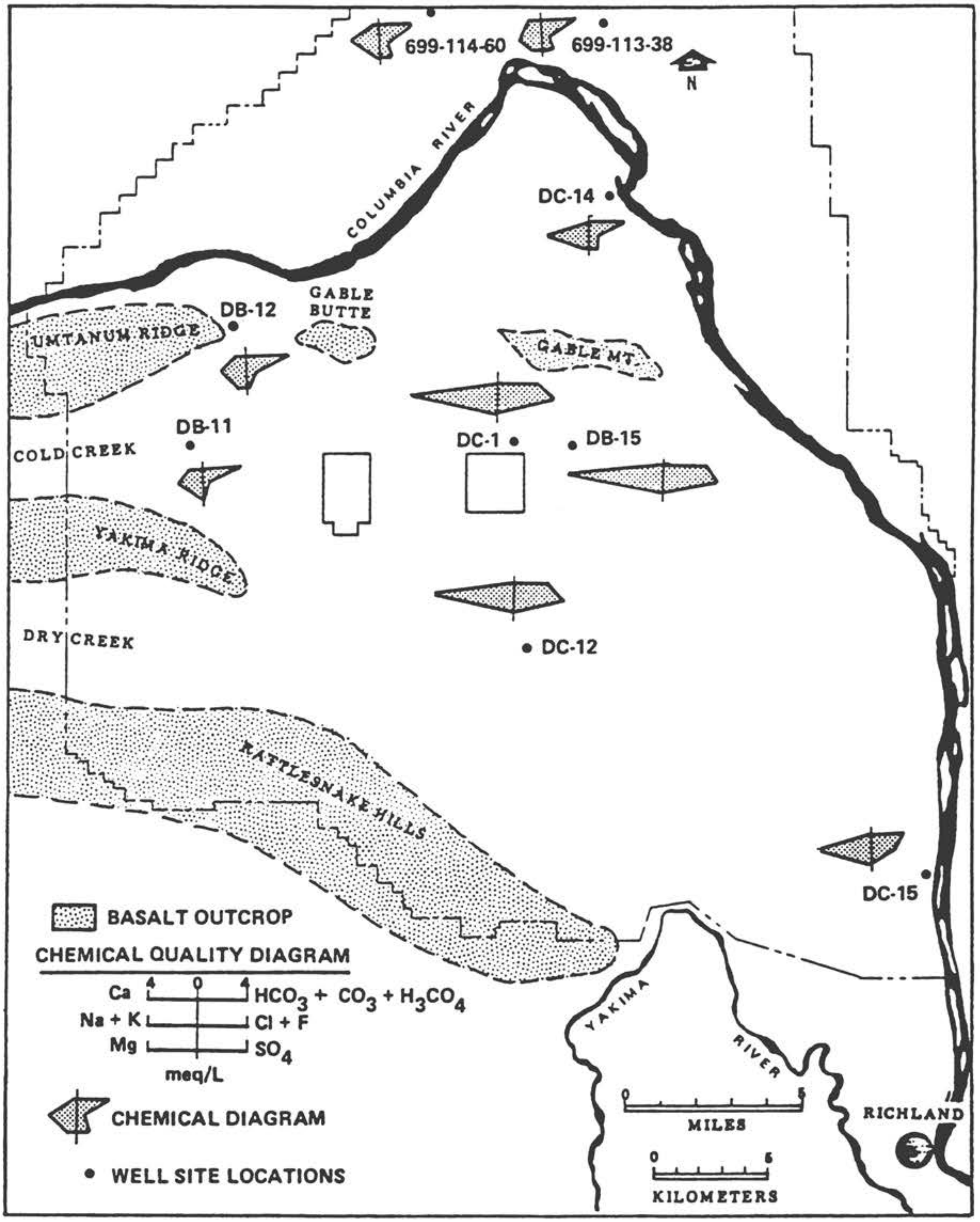


FIGURE 7-5 Hydrochemical facies map for Priest Rapids groundwater, uppermost Wanapum formation. Source: Basalt Waste Isolation Project Staff (1981b).

TABLE 7-3 Mean Compositions of Groundwaters in Pasco Basin Basalts, in Parts per Million

	Saddle Mountain	Wanapum	Grande Ronde
Na^+	58	96	257
K^+	11	14	6.5
Ca^{+2}	14	3.4	2.4
Mg^{+2}	4.2	0.8	0.04
HCO_3^-	180	128	57
CO_3^{-2}	4.2	18	21
Cl^-	12	43	169
SO_4^{-2}	15	11	125
F^-	1.3	8	30
OH^-	0.08	0.27	0.98
SiO_2 (total)[a]	63	62	109
pH	8.2	9.3	9.7

[a] All silica species converted to SiO_2.

SOURCE: Rockwell International (personal communication to T. H. Pigford, 1982).

Hydrologic data demonstrate great complexities in flow patterns that cannot be projected reliably to the reference repository location or to points of discharge. Rockwell's physical and chemical data cannot be analyzed briefly here; the interested reader can consult the background paper on basalt lava flows by D. E. White. However, horizontal flow in interbedded sediments and flow top breccias is clearly dominant, with permeabilities generally decreasing downward. Major hydrologic inadequacies and uncertainties are:

o There are no data on vertical permeabilities through faults and fractures, which constitute the normal main channels of vertical flow through volcanic lava flows (Basalt Waste Isolation Project Staff 1981c).

o Locations of discharge of deep groundwaters are not yet established (Basalt Waste Isolation Project Staff 1981c). Opinions differ greatly, indicating the need for detailed studies including chemical and thermal observations in shallow wells along the Columbia River.

o Changes in hydraulic head with depth in individual wells and between wells are irregular (Basalt Waste Isolation Project Staff 1981c) and as yet unpredictable but are in themselves evidence for a dynamic flow system requiring continuing recharge and discharge.

o Hydraulic heads decrease in several wells at depths below the Umtanum flow, implying possible escape paths through permeable aquifers below RRL (Basalt Waste Isolation Project Staff 1981d); all relationships below the Umtanum are inadequately known.

o Future melting of polar ice caps, with consequent rise in sea level close to the altitude of the Columbia River Gorge, raises questions of time and extent of possible flooding due to downstream changes. These could also occur unpredictably from volcanic damming of the lower Columbia River.

o Renewed glaciation and catastropic flooding, similar to those of the Columbia River 12,000 and more years ago, raise questions of times and extents from upstream changes.

The indicated complexities in flow patterns may never be decipherable in detail, but they are consistent with low flow rates, in part cross formational and with extensive upward dispersive mixing.

7.7.3. Chemistry of the Rocks

Chemically, the Umtanum flow and associated basaltic rocks are characterized by a large dominance of ferrous over ferric iron. This essentially guarantees that all groundwaters long in contact with the basalts have lost their dissolved atmospheric oxygen and thus are moderately to highly reducing. Primary minerals are dominated by the common rock silicates. The thick central part of the Umtanum contains as much as 70 percent of the total rock as undevitrified glass, high in ferrous iron (Basalt Waste Isolation Project Staff 1981b). This glass, as well as other minerals, should maintain a reducing environment in pore waters. The minerals and glass are converted along fractures to clay minerals and zeolites that probably have high sorption capacities.

Critical chemical questions are:

o Will abundant ferrous iron in the rock actually stabilize most radionuclides in the low-solubility forms considered most probable in Table 7-1?

o Will clays, zeolites, and other minerals provide sorption capacities sufficient to inhibit the escape of most of the soluble radionuclides, as indicated by the "basalt" column in Table 7-1?

o Will repository heating increase the devitrification rate of Umtanum glass matrix, with adverse chemical and hydrologic effects?

Although we cannot yet be confident of positive answers to the first two questions, the outlook is favorable for most radionuclides. Increased devitrification is likely to produce more clays and zeolites and a greater contact area for sorption in and near the repository; also, these minerals increase in volume with respect to the glass. These effects may actually be favorable in decreasing permeability.

7.7.4. Chemistry of the Waters

Most water samples from drill holes in the Saddle Mountains Formation, the youngest and uppermost of the three basaltic formations, are dilute and dominated by sodium and bicarbonate (Table 7-3). The underlying Wanapum Formation waters are less dilute and its Priest Rapids upper member increases in salinity eastward, especially near RRL, as its dominant anion changes from bicarbonate to chloride (Figure 7-5). This seems best explained by significant cross-formational upflow of deeper, more saline waters, perhaps mainly on faults and fractures near RRL.

Grande Ronde waters are the highest in salinity of the three basalt formations (Figure 7-4 and Table 7-3), especially in sodium, chloride, and sulfate, and are too high in sodium, chloride, and fluoride (4 to 12 ppm) for most current domestic and agricultural purposes.

The overall downward increase in salinity and in apparent age of waters since recharge is consistent with longer travel times and higher proportions of rock to water with depth. These data are also consistent with considerably greater vertical permeability than yet recognized by Rockwell. Especially serious are the indications of significant cross-formational upflow in the general areas favored for the reference repository location discussed above (Section 7.7.2). These indications are illustrated in Figure 7-5 and are considered in detail in the background paper on basalt lava flows by D. E. White (in press), based especially on chemical and physical evidence in hydrostratigraphic charts of deep test wells (Basalt Waste Isolation Project Staff 1981d). Of the five available deep stratigraphic charts, three (DC-14, DC-6, and DC-12) have significant chemical contrasts with depth that differ from the "normal" DC-15 (Figure 7-4), and only DB-15 is similar. The exceptions are:

- DC-14, where shallow potentiometric levels are anomalously high and "Saddle Mountains type" water extends down to the base of the Wanapum Formation.
- DC-6, where no waters from the two upper formations were analyzed, but the Grande Ronde waters are highest in salinity near its top but only half as high below the Umtanum.
- DC-12, where "Wanapum type" water extends down at least 300 m into the underlying Grande Ronde basalts. Unfortunately, no water samples seem to have been collected from the deeper part of this hole.

Decreasing pressure gradients deep in DC-15 (Figure 7-3) and a qualitative decrease in DC-5 below the Umtanum were discussed in Section 7.7.2. These data all demonstrate complexities in flow patterns that are not yet understood and cannot be reliably modeled. However, the total evidence suggests that water travel times from RRL to the environment are long enough now (an assumed 10,000 years) to permit limited increases in vertical flow, and that natural barriers will inhibit radioactive waste migration. Extensive subsurface dispersion plus great dilution in the Columbia River provide additional features that aid in reducing the radiation doses to future individuals from contaminated surface water.

The waters of BWIP are especially notable for their high pH values (greater than 9, Table 7-3) and low Eh values (from abundant ferrous iron), in combination with adsorbing clay minerals in fractures and on borders of open cavities. Inspection of Table 7-1 indicates that most radionuclides have low mobilities in this environment. Technetium, one of the more hazardous in most environments, has its lowest solubility in basalt (Table 7-1), lower than in any other repository rock considered here. The elements most difficult to control are iodine and possibly selenium.

7.7.5. Changes Related to Repository Construction

Additional geological issues related to repository construction are:

- The upper and lower contacts of the central Umtanum layer will not be strictly horizontal because of initial flow and topographic irregularities and because of secondary folding and faulting (Figure 7-1, but with many detailed variations). Man-made openings will probably need to be centered within this zone to avoid penetration into flow borders, some of which may be highly permeable. Also, permeable faults that offset the contacts probably occur, although their location cannot yet be identified because of the wide spacing between drill holes.
- Shaft sinking will encounter moderately high temperatures and locally very high permeabilities.
- Because of the high geothermal gradient, repository temperatures will be at least as high as 57°C and even higher if significant local cross-formational upflow is occurring, as indicated in Sections 7.7.2 and 7.7.4. These are far too high for working conditions and will require extensive cooling.
- Some faults and fractures with modest to high vertical permeability probably will be encountered, especially near broken anticlines.
- Permeable aquifers with relatively short travel to the environment may exist below the Umtanum; no data yet eliminate this possibility.

The local physical hydrology will evolve through four main stages during and after construction: initial dewatering, active period of waste emplacement, resaturation after closure, and long-lived changes after hydrologic reintegration into the regional flow system. Corresponding changes will also occur in groundwater geochemistry. The background paper on basalt lava flows by D. E. White (in press) provides a more detailed analysis.

Data are presently inadequate for a full evaluation of the implications of core discing. If the discing is caused by strongly unbalanced horizontal stresses and, consequently, increased pore pressures, rock-burst phenomena could be a serious problem during and after repository construction. During two years of panel deliberations, Rockwell has been urged to study this potential problem intensively by recognized experts in stress measurements, but definitive data are not yet available.

Inhomogeneities of the Umtanum central zone from initial flow irregularities and later tectonic changes are not yet known in enough detail to predict potential mining and drainage problems.

7.8. AN EVALUATION OF TUFF AT THE NEVADA TEST SITE, NEVADA

7.8.1. Introduction

The candidate repository site at Yucca Mountain lies along the west boundary of the southern part of the Nevada Test Site and extends westward, lying partly on the Nellis Air Force Bombing Range and partly on federal land controlled by the Bureau of Land Management (BLM) (Figure 7-6).

The region encompassing Yucca Mountain (Figure 7-7) is part of the geologically complex south-central Great Basin. Tertiary volcanism deposited more than 2,000 m of rhyolite and quartz latite tuff, lavas, and associated sedimentary rocks and resulted in numerous volcano-tectonic collapse features called calderas. Calderas, some buried by volcanic rocks, lie north and west of Yucca Mountain; one may lie directly beneath (Figure 7-6). Quaternary deposits consist of as much as 800 m of valley fill and some basalt flows and cones.

The geologic structure of the Nevada Test Site is complex. The pre-Tertiary marine sedimentary rocks were intensely deformed during late Mesozoic and perhaps early Tertiary time by folding, easterly directed thrusting, and strike-slip faulting. Resulting uplift and erosion produced a mountainous terrain upon which the Tertiary volcanic rocks were deposited. Volcanism was accompanied by large-scale block faulting, which produced the characteristic Basin and Range topography. These normal faults resulted from extensional stresses that persist to the present. Yucca Mountain is a fault block gently tilted 3°E to 6°E produced by this faulting (Figure 7-8).

The rocks of the NTS region have been grouped into ten hydrogeologic units (Winograd and Thordarson 1975) consisting of six aquifers and four aquitards. The most widespread aquifers are the lower carbonate aquifer in lower Paleozoic rocks and the valley-fill aquifer in Quaternary deposits. The other aquifers have limited occurrence within the saturated zone; they consist of the upper carbonate aquifer of upper Paleozoic rocks and local aquifers in zones of bedded tuff, welded tuff, and lava flows.

Throughout much of the area the water table lies at great depth--150 to 600 m (Figure 7-8). The complex structure of the region and the wide lateral and vertical range in permeability of the volcanic and associated sedimentary rocks (where deep enough to be below the water table) strongly influence the regional and local movement of groundwater (Winograd and Thordarson 1975).

7.8.2. Origin and Features of Ash-Flow Tuffs

Ash-flow tuffs are an important component of the Tertiary volcanic strata. They are the potential host rocks for most of the candidate repository horizons at Yucca Mountain. Ash-flow tuffs are interpreted as having been emplaced by an avalanche or flow of hot gas-laden ash, the heat of which in many places caused softening of pumice chunks and glass shards, and the weight of which caused collapse and flattening of the softened fragments, which then welded together into compact sheetlike masses having similarities to both lavas and tuff. The collapsed pumice fragments are bent or draped around rock fragments and crystals. These oriented flattened fragments provide a banding analogous to bedding and are used to determine the postemplacement tilting of fault blocks.

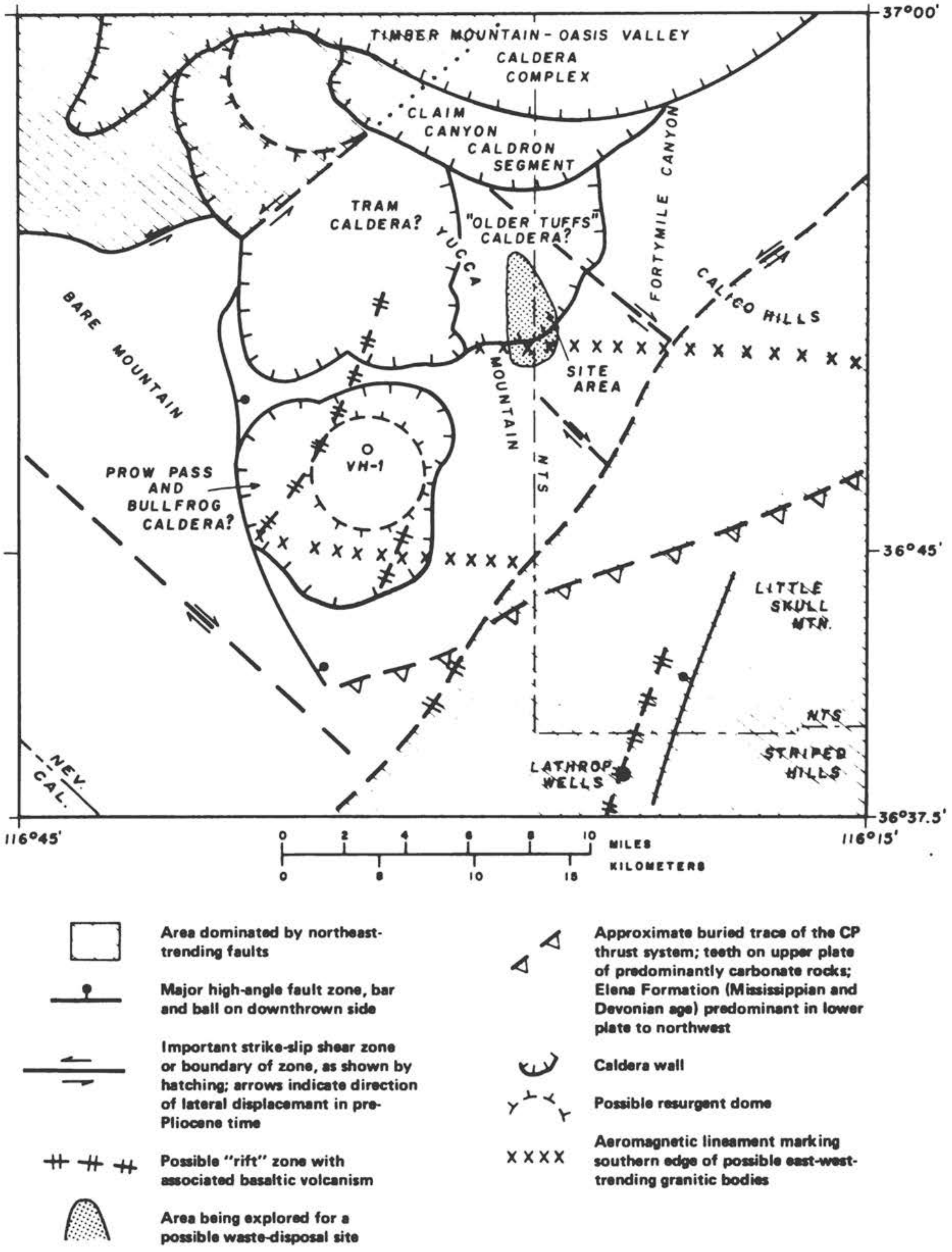


FIGURE 7-6 Diagram of the Yucca Mountain region, showing known and inferred volcanic calderas, major thrust and strike-slip faults, and basalt rift zones. Source: Carr (1982).

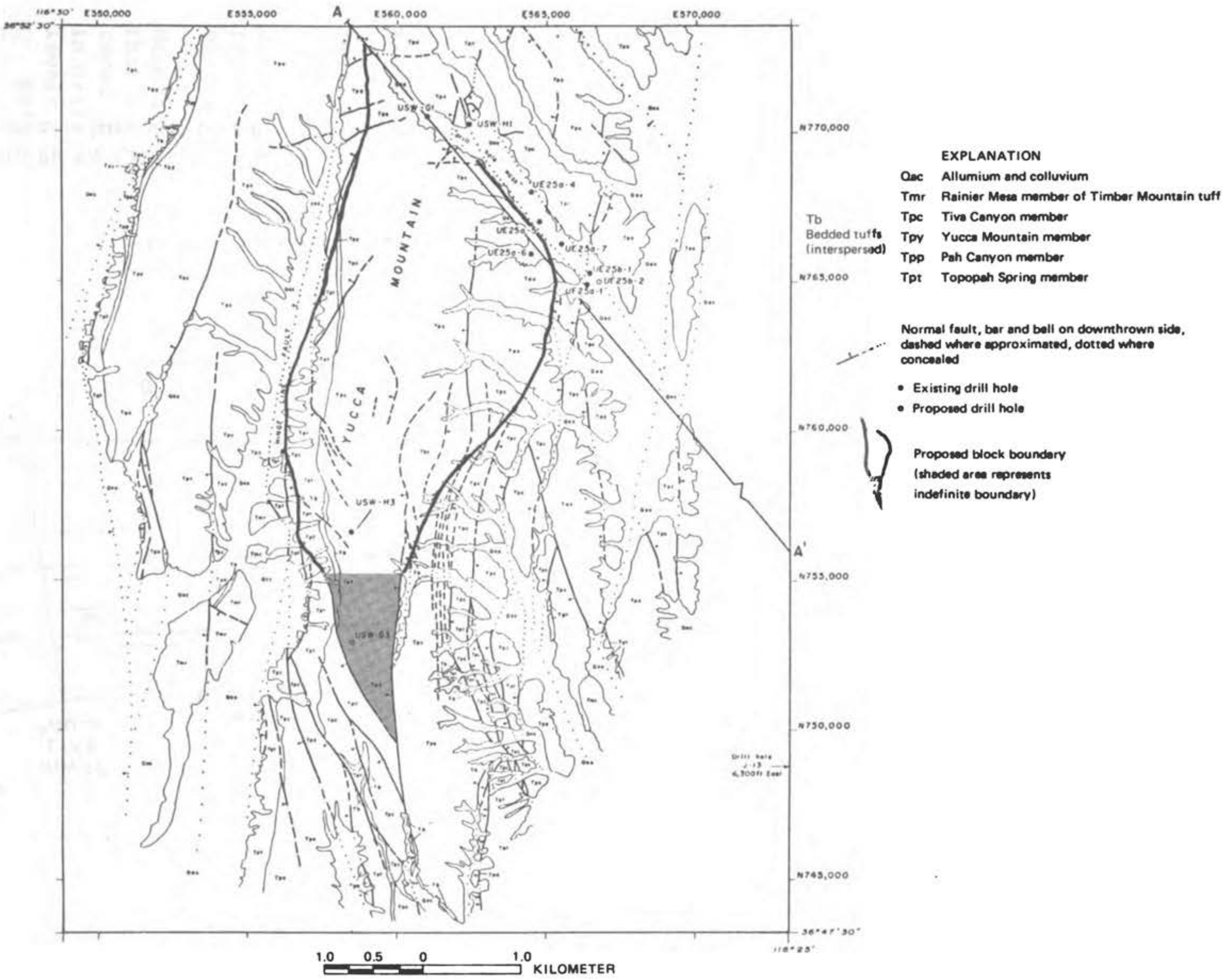
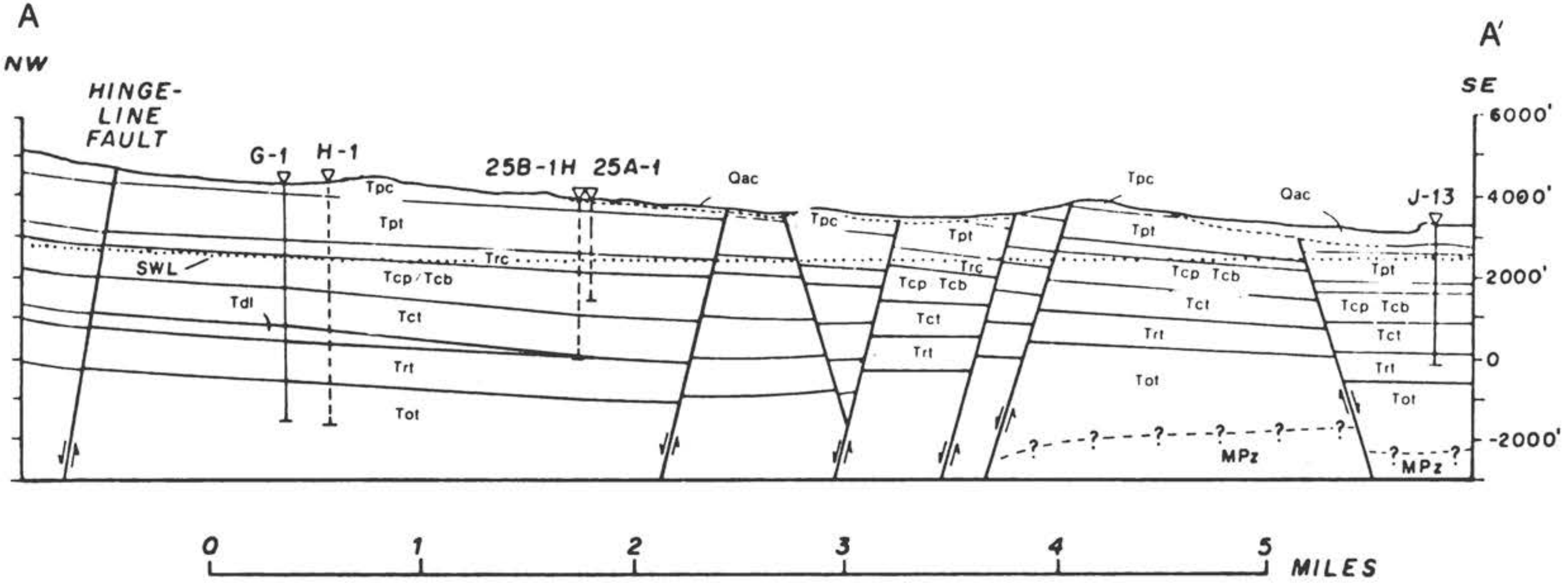


FIGURE 7-7 Generalized geologic map of Yucca Mountains. Source: Courtesy of G. Dixon, U.S. Geological Survey.



Qac	Alluvium	Tct	Tram unit
Tpc	Tiva Canyon member	Tdl	Dacite flow breccia
Tpt	Topopah Spring member	Trt	Lithic-rich tuff
Trc	Bedded tuff of Calico Hills	Tot	Tertiary older tuffs
Tcp/Tcb	Prow Pass and Bullfrog members	MPz	Mesozoic and Paleozoic rocks

FIGURE 7-8 Cross-section through northern part of Yucca Mountain (section A-A′ in Figure 7-7), west-central part of Nevada Test Site, Nevada. Dotted line labelled SWL is static water level. Source: Courtesy of G. Dixon, U.S. Geological Survey.

Although some ash flows were emplaced at temperatures too low for welding, others were hot enough to become welded and are properly called welded ash-flow tuff, or welded tuff, in which all glassy shards have coalesced, with loss of most pore space, commonly forming a dense black glass called vitrophyre. This densely welded material tends to be well jointed and generally lies between zones of partial welding that are relatively free of joints, grading upward and downward into unwelded regions. In addition to these vertical variations, there are comparable lateral variations in thicknesses of the zones and in the degree of compaction and welding. Upon cooling, large parts of the ash-flow tuff crystallize by devitrification, vapor-phase crystallization, and condensation of vapor-phase minerals in pore spaces remaining in partially welded and unwelded tuffs.

These many variations bear on the hydrologic, chemical, and mechanical behaviour of ash-flow tuffs. They include (1) primary features such as thickness and lateral extent of zones having different degrees of compaction and welding (which control porosity) and of size, type, and distribution of included blocks and (2) secondary features (at least partly related to the primary ones) such as distribution and amount of devitrification, vapor-phase crystallization, cooling joints, and subsequent tectonic joints.

7.8.3. Water Flow in the Saturated Zone

Data from eight drill holes in Yucca Mountain provide the basis for preliminary conclusions about the hydrology (U.S. Department of Energy 1982b). Groundwater beneath Yucca Mountain flows southeasterly and then southerly (Figure 7-9), probably to discharge points at Alkali Flat in the Amargosa Desert or Furnace Creek Ranch in Death Valley, 50 to 60 km away. Flow is controlled primarily by structural features. Recharge water is mostly from precipitation that falls north of the area, perhaps with minor recharge at Yucca Mountain. Hydraulic gradients are low in the eastern part of the Yucca Mountain block but steepen sharply in the western and northwestern part (Figure 7-9), suggesting greatly reduced permeability there. A major fault that marks the west edge of the repository block probably acts as a groundwater barrier.

Within the saturated zone, most permeability is due to fractures in devitrified welded tuff. Testing is under way to determine whether the fractures are primary cooling joints controlled by the zones of welding or are of subsequent tectonic origin, with regional significance.

The apparent lack of Paleozoic carbonate rocks beneath Yucca Mountain and the general absence of a vertical head gradient suggest that the deep carbonate aquifer does not influence flow in the volcanic rocks (U.S. Department of Energy 1982a). However, preliminary testing in drill hole H-1 (Figure 7-8) suggests some downward flow in the deeper levels (Dixon 1981).

Estimates of groundwater travel times within the saturated zone from Yucca Mountain to the J-12 well in Fortymile Wash about 10 km away (Figure 7-9) were made by two groups independently. The results are 10^2 to 10^5 years (Dove et al. 1982) and 2×10^3 years (Wilson 1982a). Uncertainties arise from imprecision in data such as on the

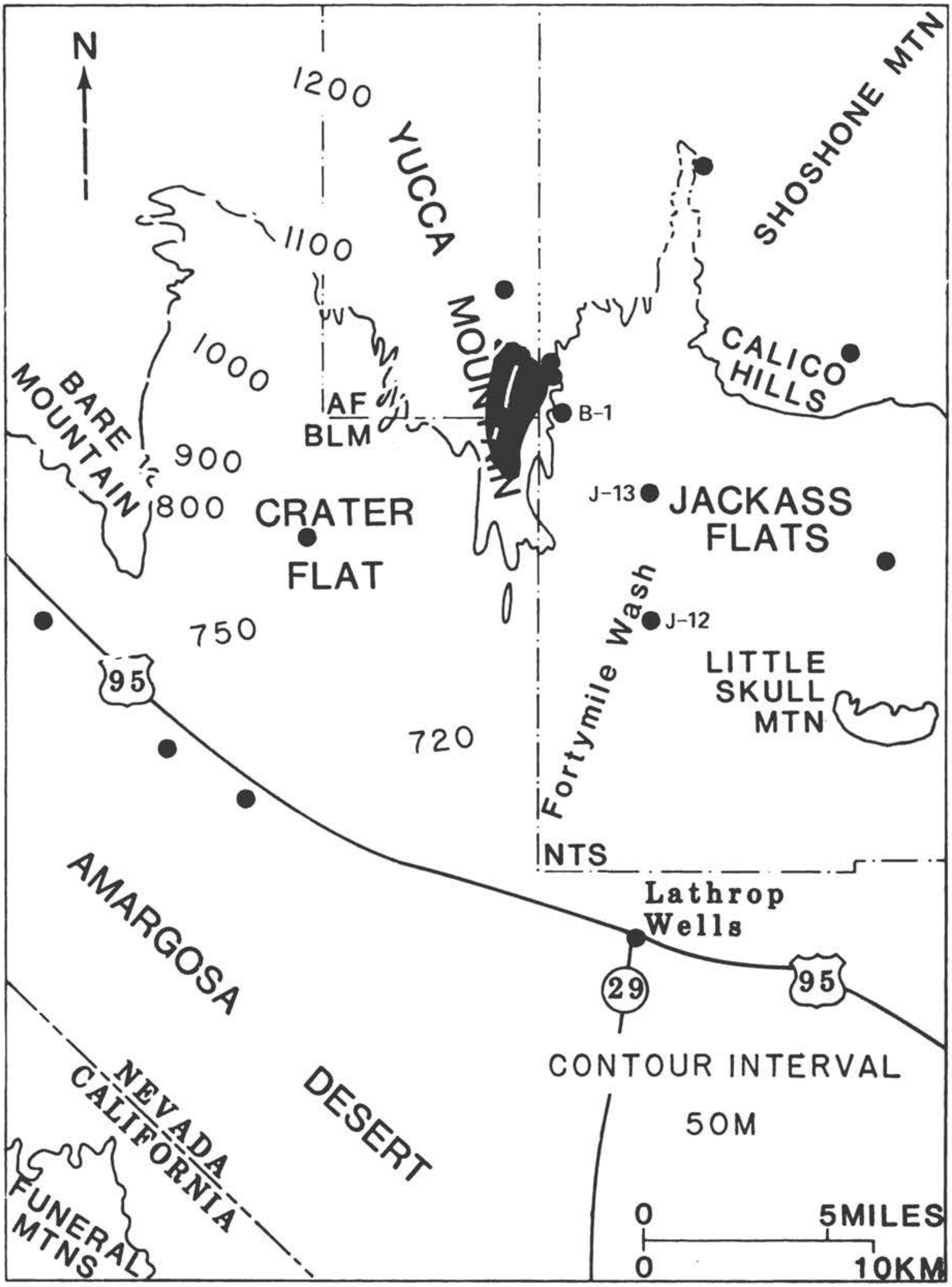


FIGURE 7-9 Altitude of water table, in meters above sea level. Also shows locations of significant wells (B-1, J-12, and J-13) used in calculating water travel times. Source: Courtesy of G. L. Dixon and W. E. Willson, U.S. Geological Survey.

potentiometric surface, the wide variation from the means of hydrologic properties used, the lack of specific data for the effective porosity of fractures in jointed tuff, and the necessity of approximating the porosities and proportions of tuff and alluvium along the flow path. In addition, the straight-line distance was assumed to be equal to the flow path, but the actual flow path may not be the shortest. In addition, the flow lines would have some vertical component, not used in the calculation, thereby lengthening the flow line and travel time (Wilson 1982a,b).

Lathrop Wells, 13.5 km south of J-12 (Figure 7-9) and presumably along the same flow line, is the nearest place to Yucca Mountain where water is consistently pumped at the present time. Calculation of the flow time from the repository block to Lathrop Wells indicates a travel time of about 4.3 x 10^3 years (Wilson 1982a,b). The resulting average groundwater velocity is 5 to 6 m/yr.

7.8.4. Water Flow in the Unsaturated Zone

One of the attractive repository horizons in the unsaturated zone of Yucca Mountain is in devitrified densely welded tuff of the Topopah Spring member of the Paintbrush tuff (Figures 7-8 and 7-10). At the proposed site for an exploratory shaft, this zone contains at least 30 m of rock with apparently suitable thermomechanical properties. This zone lies at a depth of about 375 m, and is about 130 m above the water table (Figure 7-10). Another potential candidate horizon consists of zeolitized tuff of Calico Hills, also above the water table. Two candidates in devitrified welded tuffs of the Tram and Bullfrog members of the Crater Flat tuff are both below the water table.

Yucca Mountain is unique in being the only site in the national program where the water table is exceedingly deep (about 470 to 700 m) and options exist for candidates in bedrocks in the unsaturated zone (i.e., above the water table) as well as in the saturated zone. This has important consequences for the hydrologic isolation of waste.

Groundwater flow provides the most likely means of transporting radionuclides from a repository to the accessible environment. A repository constructed in the unsaturated zone has opportunities for isolating radionuclides more readily than one constructed below the water table. Not only would the hydraulic path be longer, but slow downward percolation of groundwater in the unsaturated zone would contribute very little mass of water that could leach waste materials. Perhaps a self-draining repository can be designed with no flux for leaching.

The concept of a repository in the unsaturated zone has just recently been proposed and is based only on broad general considerations. Hence, site-specific studies of the unsaturated zone are just beginning, and very little information exists. Generic hydrologic modeling has been done on porous flow in the unsaturated zone. However, the principal flow in volcanic rocks in the unsaturated zone at Yucca Mountain appears to be fracture flow, especially in the

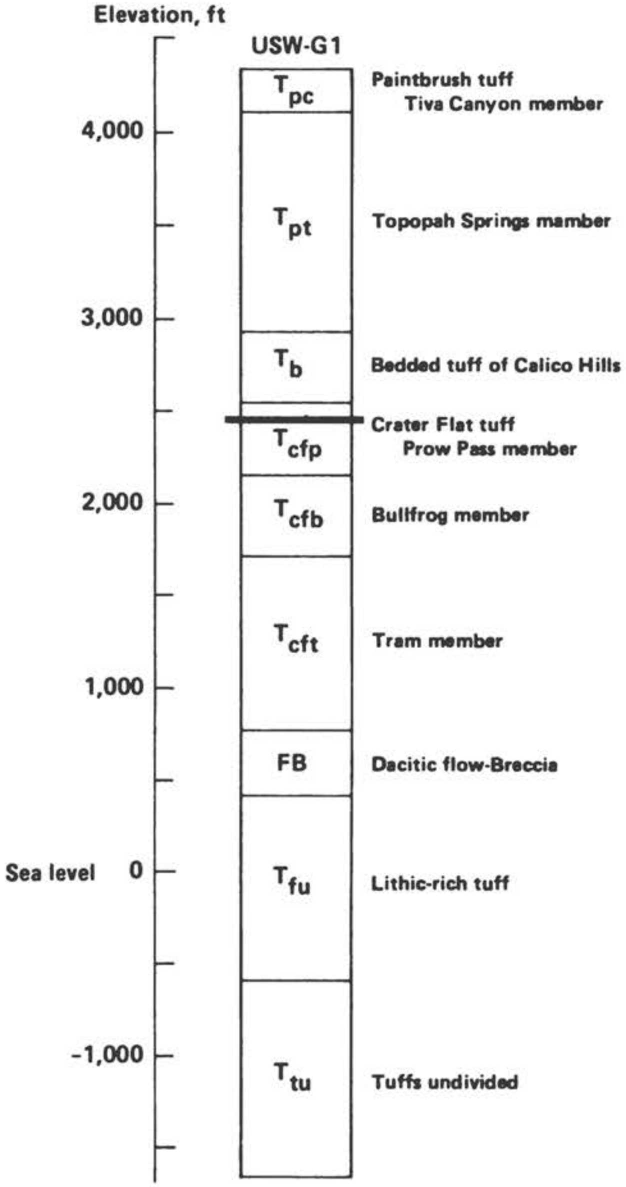


FIGURE 7-10 Stratigraphic column from drill hole USW-G1 in Yucca Mountain. The heavy black line is the static water level in this hole. Source: Bish (1981).

welded tuffs. Before meaningful performance assessments can be made, it must be determined what physical processes govern the flow of water through fractured (jointed) tuff in the unsaturated zone and how these processes are measured.

Investigations planned or under way include studies to determine the potential release mechanisms and the nature and rate of radionuclide transport in the unsaturated zone by both porous and fracture flow. Nevertheless, some approximations and calculations by Peters and

Johnstone (1982) for the rate of flow and water travel times for an assumed repository in unsaturated tuff have been supplied to the panel by L. D. Tyler (Sandia National Laboratories, personal communication to T. H. Pigford, 1982). Emphasis was placed on an unsaturated horizon in the Topopah Spring member of the Paintbrush tuff. The estimates were based on the following assumptions:

1. Recharge to the unsaturated zone is entirely by infiltration.
2. Annual rainfall is 15 cm.
3. Recharge to and through the unsaturated zone is by infiltration, assumed to be a maximum of 2 percent of total precipitation, based on data from Pahute Mesa (Blankennagel and Weir 1973), which is relatively moist and cool compared with Yucca Mountain.
4. Flow is gravity controlled and is assumed to be vertically downward. Newly added water is in contact with old water and flows only in those parts of the rock that already contain water (i.e., only in the pore fraction that is saturated and does not contain air).

Thus, the superficial (Darcy) velocity of water passing through the plane of emplacement in the unsaturated zone and progressing into the saturated zone will be about 3×10^{-3} m/yr. Because water moves through only part of the total volume of rock, depending on the effective porosity, the pore velocity will be higher.

To estimate water travel time through the unsaturated region, the stratigraphic section from the candidate repository horizon in the Topopah Spring member down to the water table was divided into six horizontal layers, or zones, based on the porosity and degree of saturation, as shown in Table 7-4. Assuming that the water flowing in each zone is entirely due to that resulting from infiltration into the upper zone, the average pore velocity in each zone is then estimated by dividing 3×10^{-3} m/yr by the product of porosity and fractional saturation for each zone. The water travel time for each zone is estimated as the zone thickness divided by the average pore velocity, yielding the results shown in Table 7-4. Summing the water travel times

TABLE 7-4 Properties of Zones Located Above the Static Water Level in Topopah Spring Tuff

Zone	Bulk Density (g/cm^3)	Porosity	Fraction Saturation	Thickness (m)	Average Pore Velocity (m/yr)	Water Travel Time (yr)
1	2.24	0.12	0.8	44	0.031	1.4×10^3
2	1.79	0.25	1.0	34	0.012	2.8×10^3
3	1.60	0.33	1.0	104	0.0091	1.1×10^4
4	1.87	0.25	1.0	20	0.012	1.7×10^3
5	1.67	0.30	1.0	19	0.010	1.9×10^3
6	2.01	0.22	1.0	25	0.014	1.8×10^3
Total						2.1×10^4

SOURCE: L. D. Tyler (Sandia National Laboratories, personal communication to T. H. Pigford, 1982).

for all zones yields a total water travel time to the water table of about 2.1 x 10^4 years. To estimate the total time for water to travel from the unsaturated zone in Lathrop Wells, the travel time of 2.1 x 10^4 years is added to the travel time of 4 x 10^3 years from the saturated zone to Lathrop Wells, resulting in a total travel time of about 2.5 x 10^4 years.

I. J. Winograd (U.S. Geological Survey, personal communication to D. E. White, 1983) suggests that those travel times may be too long for flow in the unsaturated zone; the velocities could be as much as two orders of magnitude greater if the flow proceeds along through fractures in the dense Topopah Spring Formation.

Among the additional features of unsaturated tuff for a repository, as suggested by Winograd and considered by Roseboom (in press, in a summary of this environment made available too late to affect other chapters of this report), are:

o Concern for longevity of borehole or shaft seals is minimal, because there is no aquifer cross flow. Moreover, it is actually desirable that an unsaturated-zone repository drain away the small recharge, providing minimal contact time of water and waste.

o Concern for natural fractures, or for thermally or mechanically increased fracture transmissivity, is minimal for the same reasons listed above.

o Concern for the effect of Holocene faulting on permeable pathways is also minimal, for the same reasons.

o Retrieval would be easier because of no need for dewatering.

o The small vadose flow reaching the water table is likely to be diluted by several orders of magnitude during flow in the aquifer.

o Simple engineering measures, such as the placing of canisters in sloping gravel-lined holes in the drift walls, can reduce the amount of recharge that contacts the waste. In addition, circulation of air to remove radiogenic heat during the early thermal period (Roseboom, in press) can evaporate much of the water that reaches the repository level. Any zeolitic water driven off during the thermal period is said by Winograd to have little or no significance. Circulation of air can reduce the temperatures; the zeolitic crystal structures can eventually resaturate without change of sorption properties. After the thermal period has subsided, natural air convection through the repository could replace forced circulation (but at the cost of continuing evidence for the repository's existence, increasing the possibility of future human intrusion).

7.8.5. Physical and Chemical Properties

Data from ongoing research indicate that there is a wide range in physical and chemical properties of the tuffs and associated sedimentary rocks. The high covariance of these properties with processes critical to performance assessment makes it difficult to model the processes and to assess the performance. For example:

- Strength is largely dependent on porosity.
- Presence of water (a function of porosity) weakens the rock under a thermal load because of increase in pore pressure, sensitizes tuff to time-temperature effects, and may influence creep behavior.
- Nonwelded zeolitized tuff dehydrates and contracts on heating, and, when saturated, strength is highly temperature dependent, decreasing 30 to 40 percent between 23°C and 200°C.
- Strength of devitrified welded tuff shows no temperature dependence.
- In the prevailing bicarbonate groundwater, clinoptilolite reacts at temperatures near 100°C to form analcime, with resultant release of water, reduction in volume, and reduced sorptive properties (Nevada Nuclear Waste Storage Investigations Project Staff 1982a). However, in Yellowstone Park's thermal areas, this reaction occurs only above 75°C, where SiO_2 in glass, opal, and cristobalite has already undergone slow conversion to quartz (Keith et al. 1978).
- Dehydration of zeolites and clay minerals at temperatures as low as 100°C also results in release of water, reduction of volume, production of minerals with less favorable sorption properties, and reduced rock strength.

Any water released from thermally induced mineral reactions might condense in cooler places in the waste-emplacement hole but is much more likely to evaporate into water-unsaturated circulating air.

Calculations (Erickson 1981, Helfferich 1962) and experiments (Rosen 1952) on the transport of nuclides by fracture flow have shown that, in addition to sorption on the fracture walls, diffusion into the matrix is an important retardation mechanism. The porosity of tuffs at Yucca Mountain ranges from 0.05 to 0.50 (Johnstone and Wolfsberg 1980). High porosity and low matrix permeability are highly favorable properties because diffusion into the rock matrix is enhanced. Below the water table the high porosity represents a large volume of water that is essentially stationary because the matrix permeability is so low. Radionuclides can be stored in this pore water via the diffusion mechanism, which also applies to nonsorbing elements (Nevada Nuclear Waste Storage Investigations Project Staff 1982a). Field tests by the U.S. Geological Survey are planned to obtain quantitative information on this pore diffusion and related nuclide transport in fractured tuff. Current emphasis on modeling flow and diffusion in porous fractured media is being shifted by the U.S. Geological Survey from the saturated to the unsaturated zone.

One major chemical disadvantage of ash-flow tuffs relative to most other candidate repository types is their water compositions. The panel has been given no analyses of groundwaters from Yucca Mountain, but with recharge derived entirely from precipitation on the mountain, initial waters are assumed to be very dilute and saturated with atmospheric O_2 and CO_2. Such waters typically have pH values of 5 to 6, perhaps rising to pH 7 to 8 in transit underground. Silicic ash-flow tuffs have little buffering capacity for O_2, being initially low in Fe^{+2} or other reduced constituents such as S^{-2}, and these are oxidized during high-temperature eruption. In the absence of better data, the column in Table 7-1 with pH 6 and Eh +0.2 is suggested as the best available guide to the solubilities of radionuclides in this environment. In general, their solubilities are higher than in other candidate host environments. This could be a distinct disadvantage for a water-saturated environment but has little relevance if the slight recharge can be excluded from direct contact with the waste, as discussed above.

7.8.6. Volcanism

Because Cenozoic volcanic rocks make up Yucca Mountain and dominate the terrain for miles in all directions, a comprehensive investigation was made to determine the likelihood of renewed volcanism in this area (Crowe and Carr 1980). The annual probability of volcanic activity that could disrupt a repository located within a 15-mile radius of Yucca Mountain was concluded to be on the order of 1 in 800 million (10^{-8} to 10^{-9}).

During the past 6 to 8 million years there has been a low rate of volcanic (basaltic) activity in the region, at a rate of 10^{-3} to 10^{-9} volcanic events per square kilometer per year. These consisted of Strombolian eruptions that formed cinder cones and short lava flows fed by dikes.

A preliminary consequence analysis (Crowe and Carr 1980) concluded that the disruptive effects of direct contact of waste with magma would be very limited. However, the analyses made to date ignore the secondary effects of disruption of wall rocks. These secondary effects include disruption of groundwater and changes in the stress field, which may be very important and will be included in subsequent analyses.

7.8.7. Changes in Climate

Interpretation (Van Devender and Spaulding 1979) of assemblages of isotopically dated fossilized plants in pack rat middens from various altitudes in southern Nevada indicates that during the glacial maximum the climate was only 6°C to 9°C cooler and precipitation was less than 25 percent higher than at present (Nevada Nuclear Waste Storage Investigations Project Staff 1982a). Other studies (Winograd and Doty 1980) indicate that the water table was no more than about 50 m higher than at present levels and that the indicated 50-m difference may be partly due to tectonic uplift of the region and not solely to pluvial climatic conditions. The U.S. Geological Survey concludes (U.S. Department of Energy 1982a) that similar changes in the future would not significantly affect repository performance; however, more information is needed on the relations between increased precipitation, recharge, and hydraulic gradients.

7.8.8. Faults

The Yucca Mountain candidate repository site is a fault-bounded block (Figure 7-7). Limits of the site were selected on the basis of the positions of faults or fault zones that are presumed to be hydrologic boundaries not lying within the candidate repository block. Geologic, isotopic, and geomorphic studies suggest absence of recent fault movement. The youngest possible age of movement on the major fault that bounds the west edge of the block is greater than 20,000 years, as determined by the isotopic age of unbroken calcrete that seals the fault. Actual age is probably greater than several hundred thousand years, as based on morphology of the scarp and age of the alluvium affected (Szabo et al. 1981).

Unbroken calcite fillings of fractures in the fault zone along the eastern edge of the block (in drill hole VE25a-1) have an apparent uranium-series age of more than 400,000 years (Szabo et al. 1981). Thus, a comparable age limit exists for the youngest faulting on both sides of the block.

7.8.9. Seismicity

The principal seismic concerns are the maximum accelerations the repository site will be subjected to and the frequency of such accelerations. The temporary support facilities and equipment for emplacing waste are the structures most susceptible to seismically induced damage from some threshold level of ground motion.

Estimates of probable peak acceleration in the NTS region (Rogers et al. 1977) are that more than half of the NTS (including most of the southwest quadrant) is susceptible to surface motion of at least 0.5 g with a frequency of about 2,500 years, that sites within a few kilometers of major faults might be subject to 0.7 g if the fault ruptures along its entire length, and that the frequency for the 0.7 g events is on the order of 15,000 years.

Residual issues are that all active faults may not be known and that the frequency of major earthquakes in the vicinity has not been accurately established.

Yucca Mountain is remarkably free of recorded earthquake epicenters, even though parts of the surrounding area are very active (Rogers et al. 1981). As an example, of the 356 earthquakes located in the region during 1980, none occurred closer than 12 km to Yucca Mountain.

7.8.10. Summary

Each of the four candidate repository horizons has certain advantages and potential disadvantages in terms of performance assessment. Briefly, they are as follows (in part from Nevada Nuclear Waste Storage Investigation Project Staff 1982b):

Topopah Spring Member of the Paintbrush Tuff

Advantages

- Situated in the unsaturated zone some 230 m above the water table, resulting in limited flux.
- Thick unit of devitrified welded tuff (approximately 350 m thick).
- High matrix thermal conductivity of about 1.6 W/m°C.
- Highest mechanical strength (100 MPa unconfined uniaxial compressive strength).
- Rock relatively free of zeolites or clay minerals, with a positive linear thermal expansion and a minimal release of water during heating.
- Transport barrier to downward flux of nuclides, supplied by about 115 m of highly zeolitized rocks lying between this unit and the water table.
- The shallowest of the four potential horizons, providing easiest access, lowest thermal background, and least overburden pressure.

Disadvantages

- Highly jointed or fractured rock, making vertical hydraulic permeability probably high and gas permeability high.
- Abundant lithopysae in some zones, resulting in reduced strength and potentially increased lateral permeability (though the latter has minimum consequence in the unsaturated zone).
- Limited knowledge of lateral variability of properties.
- Limited knowledge of hydrologic mechanisms and flow of water in the unsaturated zone, especially in fractured tuff.

Tuffaceous Beds of Calico Hills

Advantages

- Limited water flux, since throughout most of the repository block these strata lie within the unsaturated zone from 30 to 150 m above the water table in USW G-1 (Figure 7-10).
- Highly zeolitized, so highly sorptive and probably low in permeability.
- Low frequency of fractures compared with the other horizons in welded tuff.
- Easily mined by use of continuous mining machines.

Disadvantages

- Repository space with less lateral extent than for other candidate horizons due to unit's thinning southward within the Yucca Mountain block.

- Lower part below the water table in places.
- Mineralogically quite variable and rich in zeolites and clay minerals, resulting in complex and variable thermal expansion and contraction, depending on relative effects of heating and dehydration.
- Lowest thermal conductivity of all four horizons (about 0.9 W/m°C).
- Lowest strength of all four horizons (about 35 MPa unconfined uniaxial compressive strength), implying potential creep of underground openings.
- Possible support of perched groundwater zones above the water table by zeolitized nonwelded tuffs.

Bullfrog Member of the Crater Flat Tuff

Advantages

- Devitrified welded tuff.
- Relatively free of zeolites and clay minerals, giving a positive linear thermal expansion and making it simpler to model than the bedded tuffs of Calico Hills.
- High matrix thermal conductivity (about 2 W/m°C).
- High mechanical strength (about 50 MPa unconfined uniaxial compressive strength).

Disadvantages

- Potential repository zone relatively thin (about 130 m in USW G-1).
- High variability over short distances suggested by very limited hydrologic data available at present.
- Mining costs higher than for the two horizons above because of greater depth.
- Within the saturated zone, about 160 m below the water table.

Tram Member of the Crater Flat Tuff

Advantages

- Devitrified welded tuff.
- Relatively free of zeolites and clay minerals, giving a positive linear thermal expansion and making it simpler to model than the bedded tuffs of Calico Hills.
- Highest matrix thermal conductivity of the four horizons (about 2.2 W/m°C).
- Greatest unconfined uniaxial compressive strength of the four horizons (about 72 MPa).

Disadvantages

o Potential repository zone relatively thin (about 130 m in USW G-1).

o High variability over short distances suggested by very limited hydrologic data available at present.

o Considerable lateral variability of rock types.

o Deepest of all four horizons (more than 800 m), making it most expensive to mine and possibly requiring enhanced support of workings.

o Within the saturated zone, about 300 m below the water table.

7.9. AN EVALUATION OF GRANITOID ROCKS OVERLAIN BY A REGIONAL SEDIMENTARY AQUIFER*

The hybrid combination of granitoid rocks, including igneous and metamorphic rocks, overlain by a regional sedimentary aquifer, takes advantage of the strengths of two kinds of rocks while avoiding the major problems of each (Bredehoeft and Maini 1981). The generic characteristics of this combination have been summarized in Section 7.4.7. Potentially favorable repository sites for this combination of rocks are in coastal areas and around the margins of interior regions of structurally up-domed crystalline rocks. Groundwater flow in the sedimentary rocks can be investigated and modeled by conventional, tested methods. The flow system operates as an active barrier, such that long migration paths, low pressure gradients, and very low flow rates can be ensured.

Past approaches toward identifying rock types suitable for geologic isolation have emphasized the simplicity of a single rock type, and the features of this hybrid rock environment seem to have been given little weight. Each of these single rock types, such as granitoid alone, has major uncertainties, such as location, character, and irregular distribution of flow channels, that are exceedingly difficult to predict or to model with confidence. However, an overlying sedimentary aquifer, because of its permeability, will bypass any complex system of flow channels in the deeper granitoids, so the latter need not be fully known in detail. The coastal plains of Maryland (Bredehoeft and Maini 1981) are an example of the principles involved, but comparable interior areas of crystalline rocks are surrounded by sedimentary rocks that include permeable aquifers. As one other specific example, the low-permeability bedrocks underlying the Tertiary sediments of the Savannah River Plant in South Carolina have been found unsuitable for hydrologic reasons, but the nearby region to the east is overlapped by sedimentary rocks that may contain a favorable short-circuiting sedimentary aquifer.

In Maryland, pre-Mesozoic granitoid and metamorphic "basement" rocks are exposed 50 km west of Chesapeake Bay and, in succession, are

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*Details appear in a separate background paper on granitoid rocks overlain by a regional sedimentary aquifer by D. E. White (in press).

overlain eastward by thickening wedges of younger sedimentary rocks (Figure 7-11). These include permeable sandstone aquifers much higher in permeability than associated shales and underlying basement rocks. The latter crystallized, or recrystallized, at temperatures of 300°C or more, eliminating nearly all primary porosity and permeability; their upper contact with the sedimentary rocks dips seaward at about 1°. Most basement rocks are uniform in only one or two directions, generally changing to other crystalline rocks without major change in properties, except as modified by faults and fractures.

The tectonic environment is relatively stable; seismic activity is low except for rare major earthquakes, not yet well understood but being studied intensively; and young volcanism is absent. A temperature of 40°C is a reasonable maximum at 1 km depth. Sites of minimal potential for natural resources can be identified through geophysical surveys and test drilling. The crystalline repository host rocks are especially unfavorable for fossil fuel resources. Data are not yet adequate for specific repository siting.

The most permeable aquifers are likely to be Upper Cretaceous sandstones underlain by less permeable shales and other sediments. The permeable aquifers control the regional flow patterns in the underlying rocks because the short-circuiting pressure gradients of the aquifers are so low. Thus, flow rates in the basement rocks are even lower because of longer flow paths and, consequently, even lower pressure gradients. In addition, a gradational zone between dilute pore waters and underlying saline waters of higher density occurs about 600 m under the coastline, presumably shallowing eastward and eventually grading upward into saline seawater (Figure 7-11). Circulation rates should decrease downward and seaward, impeded by the density of the saline waters. Flushing of original seawater in overlying marine sedimentary rocks has progressed only to about 600 m in depth because of extremely low rates of flushing and upward diffusion of the saline water. A maximum depth of circulation is controlled by topographic relief at the land surface.

Regional flow patterns and hydraulic gradients are illustrated in Figure 7-11 and in a separate background paper by D. E. White (in press). A 10-fold contrast in hydraulic conductivity was assumed by Bredehoeft and Maini (1981), but actual contrasts are more likely to be 100-fold or more, according to these authors.

When sea level lowers during the next glaciation, the discharge may not necessarily be into saline water (Winograd 1983). During extended periods of glaciation, the saline water may be flushed from the aquifer. This problem relates to the great need for better understanding of the nature and origin of the salinity change. Is it highly responsive to changes in sea level, or is the gradational zone of density stratification at a depth of 600 m below present coastline actually relict from earlier glaciations? Does the saline pore water communicate directly with offshore seawater, or is this "evolved connate water"? Detailed study, chemical analyses, and water age-dating data (with appropriate concern for mixtures) should resolve these issues (White 1965, Davis and Bentley 1982).

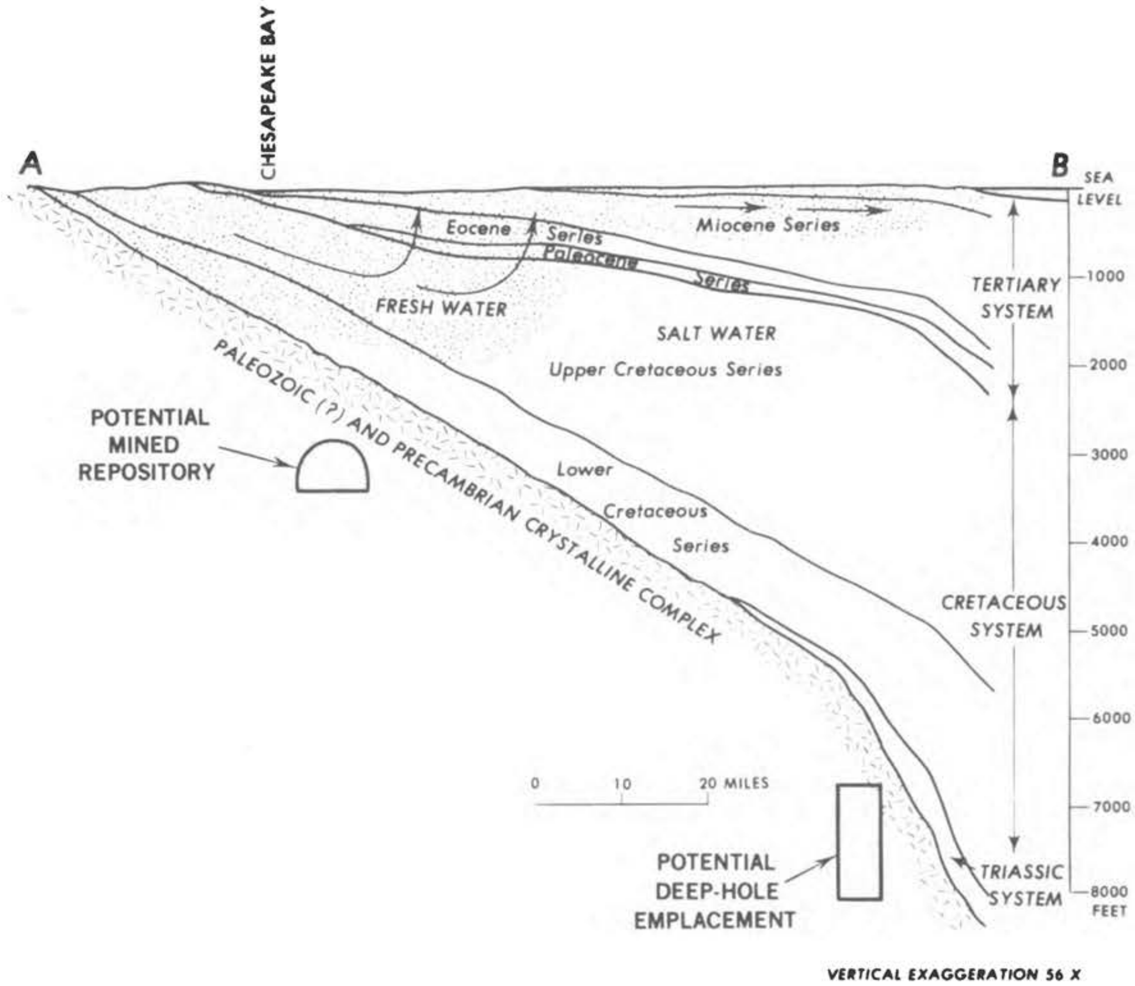

FIGURE 7-11 Section of coastal Maryland, illustrating a granitoid repository overlain by sedimentary rocks containing an aquifer (Upper Cretaceous series). Source: Bredehoeft and Maini (1981).

In this hybrid combination, the repository gains from construction in hard, stable crystalline rocks that require minimal engineered support, while the sedimentary aquifer imposes low flow rates and pressure gradients in potential flow channels in the crystalline rocks. A need for detailed knowledge of fractures and flow channels in the granitoids is thereby circumvented, in contrast to the granitoids of Section 7.6.

The whole-rock chemistry differs with specific rocks, especially for the sedimentary rock cover, but is unlikely to be highly significant. Many crystalline rocks and all carbonaceous sediments have sufficient reduced iron (Fe^{+2}) and reactive hydrocarbons to remove free oxygen from associated pore waters if flow rates are low enough, thereby resulting in low Eh. Unaltered crystalline rocks are likely to have only modest sorption capacities for radionuclides, but feldspars and other minerals may be partly altered to clay minerals. Sedimentary clays, but rarely zeolites, are characteristic of the overlying fine-grained marine rocks.

Recharging waters are initially dilute and saturated with atmospheric oxygen. With long travel times and deep circulation, and with head provided by landward recharge above sea level, oxygen reacts with ferrous iron, sulfides, and organic matter. Near the base of circulation, the quality of water changes as oxygen is removed and saline constituents are added by diffusion and mixing (Figure 7-11), becoming slightly to moderately reducing and unfavorable for most radionuclide transport. The shallow low-salinity waters are generally dominated by sodium, bicarbonate, and sulfate but have little chloride. With depth and higher salinity, water compositions change, increasing greatly in chloride, bicarbonate, and sodium (White et al. 1963, Tables 12 and 13).

Ages and flow rates of waters from a recharge area to a hybrid-type repository must be determined for specific coastal candidate sites. Very low flow rates of the deeper, more saline waters are indicated by their physical and chemical environments. The waters will generally be nearly neutral, with pH of 6.5 to 8, and slightly to moderately reducing.

No data are yet available for specific coastal sites outside Maryland. We foresee no major hydrochemical problems. Waters at repository depths will be brackish or saline and thus unattractive for most human use. Flow rates are likely to be so low that actual properties may have little significance for radionuclide transport.

Construction of the repository through onshore vertical shafts will, as usual, threaten its integrity through imperfect sealing. With radiogenic heating and thermal expansion, some fractures are opened or are newly formed and permeabilities may generally increase. However, the overlying regional aquifer and any intervening low-permeability rocks will buffer changes that do occur. J. D. Bredehoeft (U.S. Geological Survey, personal communication to D. E. White, 1981) has suggested that repository shafts be sunk "upstream," i.e., landward from a coastal site, up the hydraulic gradient from the repository, with access being provided by horizontal workings from the shaft. Rocks disturbed by construction, imperfect shaft sealing, and thermally induced permeability increases are localized landward from the repository. These rocks remain cold and unaffected by radiogenic heating, and their low convection potential remains unchanged. Undisturbed rocks above the repository are heated by conduction, probably without serious consequences because of inhibited convection. Any changes that do occur, such as locally increased permeability from conductive heating and thermal expansion, are buffered by the overlying regional aquifer. Changes in groundwater chemistry are also unlikely to be significant.

The dense population of most eastern coastal localities presents geographic and environmental problems that can be minimized but not ignored. Sites can be found that have low population densities and are also unlikely to contain mineral resources, i.e., where the depths of sediments are too shallow for oil and gas and where the underlying crystalline rocks are geophysically monotonous and unpromising for mineral resources. Because the brackish and saline pore fluids are presently unattractive for normal uses and better-quality water supplies are available elsewhere at shallower depths, the likelihood of accidental future human intrusion in search for water is very low. South of Maryland the coastal plain extends farther inland, with consequences for depth of sedimentary rocks, presence of aquifers, salinity distribution with depth, and potential resources that must be evaluated.

Data are adequate to emphasize the marked advantages of a crystalline rock repository overlain by a regional aquifer, but detailed studies of potential sites are essential.

Although this example is mainly for the many potential coastal sites, favorable combinations of crystalline rocks overlain by regional aquifers also exist on the flanks of many interior bedrock structural highs. Some of these have significant potential for organic fuel resources and mineral deposits, which must be evaluated.

7.10. EVALUATION OF DATA FOR SORPTION AND SOLUBILITIES

7.10.1. Introduction

Table 7-1 is a summary of data available in the published literature on the solubilities and retardation factors of important radionuclides in the major proposed geologic media. An attempt is made to supply a single number that can safely be used for the solubility and retardation factor of each nuclide in each medium under the most probable repository conditions--pH between 6 and 9 and Eh between 0.2 and -0.2 V. For solubilities these are the numbers in the column headed "Most Probable," and for retardation factors they are the bold-faced numbers in each group of three for each element. Single numbers for elements with more than one oxidation state (especially technetium, uranium, neptunium, plutonium, and tin) must be used with caution, because both solubilities and retardation factors for these elements are very sensitive to slight changes in Eh. To give an idea of possible variations, the table includes additional numbers for solubility under different assumed repository conditions and for extremes of reported retardation factors. The single numbers in all cases are chosen to be conservative but not absurdly so.

In selecting data for more detailed performance analyses of actual repositories, it is important that the local conditions of pH and Eh be known. Actual measurements of dissolved oxygen in groundwater are needed. In dense rocks in particular, the whole-rock mineralogy is not necessarily an indicator of the redox conditions of water moving through fractures.

7.10.2. Solubilities

The numbers in the "reducing" and "oxidizing" columns of Table 7-1 are in part taken from published experimental work and in part calculated from thermochemical data. The "most probable" values (for pH 6 to 9 and Eh 0.2 to -0.2 V) are estimated from these limits. Because thermochemical data for some elements are uncertain and because solubility is influenced by many factors (notably grain size, degree of hydration, and presence of complex-forming ligands in groundwater), the estimated values are higher than, or at least as high as, the numbers given for comparable conditions in the "reducing" column for pH 9. A good deal of subjective judgment is necessarily involved in choosing the most probable values. The designation "high" means that solubility is greater than 10 ppb and hence could not be an effective control of radionuclide concentration.

For some elements, solubilities are simply not known, either from thermochemical data or from experiments, and the numbers given are guesses based on chemical similarities (tin, antimony, selenium, and curium). For technetium and neptunium, it should be emphasized that solubilities would be considerably greater than the "most probable" value under only slightly more oxidizing conditions. Data for americium are especially perplexing: earlier work suggests solubilities on the order of 1 ppm, hence not controlling, but the paper by Rai et al. (1981a) at the Pacific Northwest Laboratory (PNL) gives good evidence for the numbers listed in Table 7-1.

7.10.3. Retardation Factors

Most of the retardation factors (K) in Table 7-1 are calculated from measured values of the distribution coefficient (K_d) by:

$$K = 1 + \rho K_d (1 - \varepsilon)/\varepsilon,$$

where ε is the porosity, i.e., the water volume divided by the volume of solid plus water, ρ is the density of the solid phase, and K_d is the distribution coefficient expressed in cubic centimeters per gram. A value of 10 is assumed for $\rho(1-\varepsilon)/\varepsilon$, which seems safely conservative because rock densities are seldom less than 2 g/cm^3 and porosities are seldom greater than 20 percent.

Recorded K_d values and retardation factors for most elements cover an enormous range, reflecting differences in experimental conditions and techniques of measurement. Choosing reasonably conservative values requires subjective judgment not only about the numbers themselves but about quality of the experimental work, particularly the relevance of the experimental conditions to the probable conditions in a repository. For each nuclide and each geologic medium, Table 7-1 gives three values: the first is a rock-bottom conservative figure, chosen from the lowest experimental figures reported and almost certainly lower than would be expected in nature; the second is a reasonably conservative estimate for most expectable repository conditions; and the third is a number picked from higher values in the literature that seem possible but a little high for conservative use. The range in each group of three reflects the fact that retardation factors cannot be specified more closely than about an order of magnitude, because their measurement is difficult and because they are influenced by many environmental factors.

The retardation numbers for tuff in Table 7-1 include all varieties of this rock (fresh, devitrified, zeolitized, etc.) but for the sake of conservatism are weighted toward the less-sorptive types. In the "clay, soil, shale" column, experimental values are lumped that seem to depend largely on sorption by clay minerals; such numbers may well be applicable to weathered or altered granite and basalt with clay-lined fractures. The column for salt does not refer to sorption on rock salt itself (which is very small) but to sorption on ordinary rocks near a salt repository where groundwater presumably contains much dissolved NaCl. These numbers are particularly uncertain because they obviously depend on the concentration of salt as well as other variables.

In measuring K_d values some authors have not been careful to ensure that the original concentration of sorbate was less than its solubility limit, so that the K_d values may be inflated by the amount precipitated rather than sorbed. This may be responsible for some of the very high values in the literature for plutonium, americium, and curium, and perhaps also for thorium, zirconium, and tin. From the practical standpoint of radionuclide movement, the questionable K_d values are probably not important, because the combination of insolubility and at least reasonably high retardation would in any event keep concentrations of these elements below MPC values.

Some apparent anomalies in Table 7-1 have no clear explanation, except that they generally involve results from different laboratories that used somewhat different materials or techniques. Why, for example, should tuff sorb uranium less well than granite or shale? Why should basalt be less effective for americium and curium than granite? Why should sorption of americium, curium, and selenium be apparently just as strong or stronger from brines than from dilute groundwater, whereas for most elements the sorption from brines is much less? Such questions probably have little actual significance because they may refer to different kinds of starting materials or different experimental techniques.

Sorption data are very scanty for curium, radium, lead, tin, selenium, and antimony, and the numbers for these elements are little better than guesses.

7.10.4. Notes on Individual Elements

Strontium (Sr)

Solubility Strontium forms two somewhat insoluble compounds with common constituents of groundwater, $SrSO_4$ and $SrCO_3$. Neither would hold the concentration anywhere near 10 ppb under repository conditions. At pH 9, calculation indicates that $SrCO_3$ in water with HCO_3^- at $0.002\underline{M}$ would give about 0.6 ppm of Sr^{2+} to the solution; this result agrees with a more sophisticated calculation of Muller et al. (1981). No complexes are known that would influence solubility.

Retardation Strontium is a favorite element for sorption experiments; many values are recorded in the literature, and the values have an enormous range: 16 to 3,000 for granite; 160 to 11,000 for basalt; 20 to 200,000 for tuff; 100 to 400,000 for clay; and 0.2 to 40,000 for rocks in concentrated brine. The number 200 picked for all the rocks in dilute groundwater is a conservative average for experiments that use simulated repository conditions and seem to be well done. No reason is evident for the very high reported numbers; some may represent formation of slightly soluble compounds rather than sorption. There seems general agreement that sorption from brine is much less than from dilute solution. The ultraconservative minimum values record common experience that crushed very fresh rock is not a good sorbent for strontium; probably sorption in ordinary granite and basalt is enhanced by the usual slight alteration of feldspars to sericite or clay minerals.

Cesium (Cs)

Solubility Cesium normally forms no insoluble compounds with common anions, but it can be trapped in analcime, a sodium zeolite that accommodates some cesium if available during crystallization (Keith et al., in press).

Retardation Like strontium, cesium has been very extensively studied, and ranges of reported values are equally large. There seems general agreement that cesium is strongly sorbed on common rocks from dilute solutions but not from brines. Sorption is especially strong on rocks that have layer-silicate minerals (micas and clay minerals). When strontium and cesium are compared, much greater sorption of the latter is commonly noted. The somewhat smaller figures for tuff are simply from averaging experimental data, and the differences almost certainly have no real significance.

Technetium (Tc)

Solubility Technetium is very soluble in groundwater as TcO_4^- under oxidizing conditions, but it precipitates as the very insoluble TcO_2 when conditions are reducing. Unfortunately, thermochemical data for technetium are not well enough known to set the boundary between these two forms precisely. The numbers in Table 7-1 are from Bondietti and Francis (1979); the enormous difference in solubility between pH 6 and pH 9 is a consequence of the large coefficient of H^+ in the equations

$$TcO_2 + 4H^+ = Tc^{4+} + 2H_2O$$

and

$$TcO_2 + 2H_2O = TcO_4^- + 3e^- + 4H^+.$$

Muller et al. (1981) describe Bondietti and Francis's estimate as "not well enough supported to be acceptable," and simply say the solubility of reduced technetium is "certainly low." Smith et al. (1981) choose a value of 1 ppb, based on Bondietti and Francis's experimental result and old thermochemical data given by Pourbaix (1966). The National Waste Terminal Storage (NWTS) Program Staff (U.S. Department of Energy 1981), in response to panel questions, gives much lower calculated results, but then says, "They can hardly be considered realistic." The figure of 1 ppb seems safely conservative, provided conditions remain fairly reducing and slightly alkaline; but the solubility increases rapidly with slight increases in either acidity or redox potential.

Retardation There is general agreement that TcO_4^-, the probable form of technetium under even slightly oxidizing conditions, is very slightly sorbed. Johnstone and Wolfsberg (1980), whose work seems very good, give experimental K_d values for tuff ranging from 0.2 to 2.0 ml/g for experiments in air, and values from 8 to 118 ml/g for experiments in N_2. Other quoted K_d values range up to 1,000 ml/g (e.g., Muller et al. 1981), but most are below 20. Muller et al. note that the form of technetium under reducing conditions is unknown but is probably Tc^{2+} or $Tc(OH)_2^+$; since these are cations, they should show some sorption. For slightly reducing conditions, at least slight retardation (factor of 5 in Table 7-1) seems probable, but a factor greater than 100 is not justified for any of the rocks.

Iodine (I)

Solubility Iodine would be present under reducing conditions as I^- and under oxidizing conditions as I_2 or IO_3^- (depending on pH). All of these are soluble under any imaginable repository conditions.

Retardation There is wide agreement that iodine shows little or no retardation. Cloninger and Cole (1981), however, report an experimental value of 5 ml/g for K_d in basalt.

Uranium (U)

Solubility Uranium is soluble under oxidizing conditions (Eh greater than +0.1 V) in the form of UO_2^{2+}. If much CO_3^{2-} is present (not likely under repository conditions), it is soluble even in reducing conditions as low as Eh = -0.1 V, as a carbonate complex. Phosphate and fluoride complexes are stable enough to increase solubility appreciably if these ligands are present in amounts only slightly greater than those in ordinary groundwater. Deju (1981c) records experimental solubilities between 2×10^{-3} and 2 ppm for basalt groundwater at pH 9 to 10 and Eh -360 to -410 mV. He notes that the theoretical solubility under these conditions is less than 2 x 10^{-4} ppm. Calculations done by the NWTS Program Staff (U.S. Department of Energy 1981) in response to panel questions give solubilities for reducing conditions in different kinds of groundwater ranging from $10^{-2.6}$ to 10^{-6} ppm; Heckman and Donich (1979) give less than 10^{-10}; Wood and Rai (1981) suggest 10^{-3} at pH 9 and 10^{-5} at pH 6; Muller et al. (1981) calculate $10^{-6.3}$ for the solubility of crystalline UO_2 at pH 6 but note that the figure would be greater by more than five orders of magnitude for amorphous UO_2. Smith et al. (1981) adopt a value of 10^{-3} ppm after reviewing several sources. The figure of 10^{-3} ppm seems safe, but solubility is strongly dependent on Eh, complexing agents, and crystallinity of the solid.

Retardation Deju (1981a) gives experimental K_d values for fresh basalt from 6 to 170 for reducing conditions and 2 to 70 under oxidizing conditions, then notes that the numbers are much higher (30 to 1,200) for clay and zeolites in cracks in the basalt (through which groundwater would presumably travel). Wolfsberg et al. (1981) cite experimental values for devitrified tuff ranging from 0.5 to 14 ml/g and for zeolitized tuff from 2.3 to 57 ml/g. Other figures for retardation range up to 5,000 (still higher if organic matter is present), but most are under 500. There seems general agreement that sorption is less from brine than from dilute solution. The relatively low figures for tuff in Table 7-1 are based on Wolfsberg's work.

Neptunium (Np)

Solubility Neptunium, like uranium, is soluble (as NpO_2^+ and NpO_2^{2+}) under oxidizing conditions but forms an insoluble dioxide under reducing conditions. The transition is not well defined by available thermochemical data. For an Eh of 0.2 V, Rai et al. (1982) give 0.1 ppm at pH 6 and 0.01 at pH 8; the diagrams of Simon and Orlowski (1980), on the other hand, suggest 10^{-3} under these conditions and about twice that value if CO_2 is present. Muller et al. (1981) calculate $10^{-7.3}$ ppm at pH 6 under reducing conditions, and a thousand times smaller at pH 8. Deju (1981c) suggests $10^{-5.7}$ at pH 7; Smith et al. (1981) adopt 10^{-3}. The figure of 10^{-3} ppm seems safe, but the solubility, like that of uranium, is strongly influenced by Eh, complexing agents, and crystallinity of solid NpO_2.

Retardation Allard et al. (1980) report K_d values for minerals separated from granite and basalt in the range of 30 to 500 ml/g under oxidizing conditions. Muller et al. (1981) note that neptunium probably exists chiefly as NpO_2^+, a singly charged and weakly sorbed complex, and estimate K_d values between 0 and 100. Wolfsberg et al. (1981) give experimental K_d values of 6 ml/g on devitrified tuff and 20 ml/g on zeolitized tuff, both under oxidizing conditions. Cloninger and Cole (1981) choose retardation factors of 50 for basalt and 670 for granite; Burkholder et al. (1975) use 100 for K_d. Smith et al. (1981) adopt 100 for a retardation factor. Foley et al. (1981) give retardation factors of 50 for basalt, 666 for granite, 460 for shale, and 230 for sorption from brine. Data summarized by Moody (1981) result in retardation factors ranging from 500 for granite to 1,500 for tuff and basalt under reducing conditions, and from 10 to 100 under oxidizing conditions. The data are confusing and the numbers in Table 7-1 are little more than guesses.

Plutonium (Pu)

Solubility Like neptunium and uranium, plutonium is soluble under oxidizing conditions (as PuO_2^{2+} in acid solution and hydroxy complexes at neutral and high pH) and precipitates as the dioxide under reducing conditions. The stability field of the dioxide is larger than that for uranium and neptunium, so plutonium is generally less soluble. Calculated solubilities are very low, generally below 10^{-7} ppm for all reasonable repository conditions, but experiments suggest higher values. Some convincing experiments by Rai and Ryan (1981) give solubilities of PuO_2 at pH 7 of 10^{-3} ppm and of $Pu(OH)_4$ 30 times higher; both values decrease by a factor of 10 per unit increase in pH. Extrapolating experimental results on borosilicate glass, Wood and Rai (1981) estimate solubilities of 10^{-3} and 10^{-4} ppm for oxidizing and reducing conditions at pH 6 and a solubility of 10^{-5} for both conditions at pH 9. Smith et al. (1981) adopt 10^{-3} ppm. Muller et al. (1981) estimate a high solubility (as Pu^{3+}) under reducing conditions at pH 6 and $10^{-3.7}$ ppm at pH 8. A figure of 10^{-3} ppm seems safe, with the usual cautions about complexes, Eh, and crystallinity.

Retardation The numerous studies of plutonium sorption are unanimous in suggesting high values, even from strong brines. Very few numbers for retardation in the literature are below 200, and high values range up to 900,000. The numbers in Table 7-1 are conservative averages from experimental work that looks reliable. Many authors note that the numbers may be influenced by complexing, especially with organic ligands.

Americium (Am)

Solubility There is great confusion about the solubility of americium, presumably because the basic thermochemical data are poor. It is generally agreed that the only stable oxidation state in solution is +3 and that higher oxidation states are strong oxidizing agents; whether AmO_2 is sufficiently insoluble to exist as a solid in repository environments, despite its oxidizing properties, seems to be a matter of debate. Apps et al. (1977) do not even mention AmO_2 in their compilation of thermochemical data, noting only that the stability field of AmO_2^+ is far outside of terrestrial conditions. Data summarized by Moody (1981) show AmO_2 as the stable solid under low-Eh conditions and AmO_2OH at high Eh. Wood and Rai (1981) maintain that "it is expected that Am(III) solid and solution species will be stable throughout the environmental pH range. Eh should not, therefore, be an important factor for Am solubility." Early estimates of americium solubility are high: Smith et al. (1981) make it 50 ppm; Foley et al. (1981) suggest $10^{-2.7}$ ppm; and Muller et al. (1981) describe americium as "relatively soluble" but give no definite value. The NWTS values seem absurd, and the authors themselves describe them as "highly suspect."

Recent work leaves the solubility of americium still ambiguous. Edelstein et al. (1983) measure the concentration of americium remaining in solution after precipitation from a 0.1<u>M</u> $NaClO_4$ solution at pH over the range 5 to 10, and they report values of about 0.002 ppm at pH 9, 0.2 ppm at pH 8, and greater than 10 ppm at pH 7 and lower. They describe these numbers as "an upper limit to the solubility of amorphous $Am(OH)_3$," although the precipitated solid was not analyzed. Much lower figures were obtained by Rai et al. (1981a) in solutions formed by shaking contaminated sediments at Hanford with 0.0015<u>M</u> $CaCl_2$: $10^{-4.7}$ ppm at pH 6 and $10^{-7.7}$ ppm at pH 9. They were unable to identify the solid compound of americium, but they conclude that it would resemble a solid that might be precipitated under repository conditions. Similar solubilities are recorded by Deju (1981) and by Wood and Rai (1981) but are probably based on the same work. Because Rai et al. used conditions more closely resembling a repository situation, their numbers seem preferable to those of Edelstein et al. for present purposes, and 10^{-4} ppm seems a reasonably conservative guess for maximum solubility. The concentration that might be obtained by rapidly neutralizing an acidic americium solution, however, would doubtless be much higher.

<u>Retardation</u> There seems a general consensus that americium is strongly sorbed by all common rocks at pH values expected in a repository environment, even from fairly concentrated brines. The lowest number recorded is a retardation factor of 60 for basalt (Cloninger and Cole 1981); high values range up to 500,000. With regard to brines, Muller et al. (1981) deviate from the majority opinion by suggesting K_d values in the range of 1 to 100; but at least some of the work they quote was done at pH 1 to 4, where the sorption is indeed low. Recent experimental work giving high values is reported by Johnstone and Wolfsberg (1980), Beall et al. (1980), Cloninger and Cole (1981), and Allard et al. (1980). No reason is evident why basalt should be less sorptive than granite, especially for an element that is not influenced by Eh, so Cloninger and Cole's low figure needs checking. As for the other actinides, the solubility and retardation of americium may be markedly influenced by ligands that form complexes with Am^{3+}.

Curium (Cm)

Solubility Published laboratory data for curium are nonexistent, and guesses are based on similarity to americium. Muller et al. (1981) suggest high solubilities for both elements; most others guess on the low side. The 1 ppb listed in Table 7-1 is a pure guess, and the question mark beside it is important.

Retardation As is true for americium, reported values for curium are uniformly high, but dependable experimental data are scarce.

Radium (Ra)

Solubility Like the other alkaline-earth metals, radium forms a moderately insoluble sulfate and carbonate. The sulfate is about four orders of magnitude less soluble than $SrSO_4$ but still would hold the concentration of Ra^{2+} only to about $10^{-6.4}\underline{M}$ or 0.1 ppm (for assumed $SO_4^{2-} = 10^{-4}\underline{M}$). The numbers for pH 9 in Table 7-1 are based on the dubious assumption that the carbonate is less soluble to about the same extent, but no recorded value was found. In any event, solubility will not be an effective control for radium.

Retardation Scant experimental data suggest stronger sorption than for Sr^{2+}, and this agrees with the general tendency for increasing sorption with increasing atomic weight in the first two columns of the periodic table. The numbers in Table 7-1 are not based on firm evidence.

Lead (Pb)

Solubility Lead in a repository environment would have the single valence 2+ and could precipitate as the moderately insoluble $PbSO_4$ or $PbCO_3$. Numbers in Table 7-1 are calculated for these compounds, on the assumptions that

$$SO_4^{2-} = 10^{-4}\underline{M}$$

and

$$HCO_3^{-} = 0.002\underline{M}.$$

In a more sophisticated calculation taking account of complexes, Hem (1976) arrives at 2 ppm for pH 6 and 0.2 ppm for pH 9. Thus, lead concentrations are not effectively controlled by solubility. This might not be true, however, for dilute solutions containing sulfide ion, because lead forms the very insoluble PbS (solubility product $10^{-27.5}$).

Retardation The same set of numbers reported by Moody (1981) and the NWTS Program Staff (U.S. Department of Energy 1981) is probably based on experiments at PNL. They suggest modest sorption, less in salt water than in fresh. The difference between numbers for basalt and granite is puzzling.

Thorium (Th)

Solubility The dominant form of thorium in solution at pH values above 5, according to Ames and Rai (1981), is $Th(OH)_4^o$; for its concentration in equilibrium with ThO_2 they give $10^{-4.3}$ ppm. The same number is given by Bondietti and Francis (1979). A higher number, $10^{-1.2}$ ppm, is given by Muller et al. (1981), but whether this refers to crystalline ThO_2 or a hydrated form is not clear. In river water, Heckman et al. (1979) report the same number, $10^{-1.2}$; but in a river draining a thorium deposit in Brazil, M. Eisenbud (Institute of Environmental Medicine, NYU Medical Center, personal communication, 1982) finds only $10^{-3.7}$ ppm. The tabulated number, 1 ppb, seems safely conservative, although complexing or colloid formation might give higher values locally.

Retardation Reported retardation factors are uniformly high, ranging from 500 (for brine solutions) to 1,000,000 (for clay), but data are scanty.

Zirconium (Zr)

Solubility Data for zirconium are poor. Ames and Rai (1978) suggest less than 10^{-9} ppm at pH 3.5 to 8.0 and 10^{-7} ppm at pH 9; Muller et al. (1981) calculate $10^{-5.7}$ ppm at pH 6 and $10^{-4.3}$ at pH 8; Smith et al. (1981) conservatively guess 10^{-3} ppm. A value of 10^{-4} ppm seems safely conservative, but it is not well supported.

Retardation As for thorium, reported values for zirconium are high and the quantity of data is small.

Tin (Sn)

Solubility The most stable solid compound of tin is the very insoluble dioxide SnO_2. This can dissolve in alkaline solution to form $Sn(OH)_6^{2-}$, or the more stable fluoride complex if appreciable F^- is present. Under the reducing conditions of many repositories, however, the tin would probably be reduced to Sn^{2+} (or some complex thereof). Heckman et al. (1979) calculate a total solubility of about $10^{-12.4}$ ppm at pH 6.8 and $10^{-9.7}$ ppm at pH 8.2, but measure $10^{-3.7}$ ppm in river water. Muller et al. (1981)

suggest 10^{-4} ppm for the range pH 6 to 8. From these meager data, a guess of 1 ppb is a conservative figure, but it rests on shaky evidence.

Retardation Moody (1981) and the NWTS Program Staff (U.S. Department of Energy 1981) record similar numbers, evidently derived from the same study at PNL, in which the range of retardation factors for reducing conditions in various rocks is 100 to 500, and for oxidizing conditions 500 to 5,000 (except for 10 for brine in a reducing environment). Others give higher numbers: Cloninger and Cole (1981), 1,000 to 3,300; Heckman et al. (1979) 1,000; the Nevada Nuclear Waste Storage Investigation Project Staff (1982a), 2,000 to 100,000. Numbers in Table 7-1 are largely guesses.

Selenium (Se)

Solubility A reasonable guess is that native selenium would be the only control on solubility in a reducing environment, but thermochemical data are inadequate for meaningful calculations. The figure of 1 ppb in Table 7-1 is little more than a guess.

Retardation Moody (1981) and the NWTS Program Staff record similar numbers, evidently from the same study at PNL. Numbers are from 10 to 50 under oxidizing conditions and from 30 to 200 under reducing conditions, with a maximum of 1,000 in brine. Cloninger and Cole (1981) give a range of 24 to 150; the NWTS Program Staff (U.S. Department of Energy 1981) gives 70 to 140 for tuff. Deju (1981b) gives K_d values ranging from 0.6 to 8.0 for basalt, increasing to 15 to 21 if pyrite is present. The restricted range gives some confidence in their validity.

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8

OVERALL PERFORMANCE CRITERION FOR GEOLOGIC WASTE DISPOSAL

8.1. INTRODUCTION AND SUMMARY

In previous chapters we have described and evaluated the state of knowledge of the components of the geologic waste-disposal system. In Chapter 9 we present the calculated performance of the overall system of these components in terms of expected and possible long-term releases of radionuclides in the environment and radiation doses therefrom. To assess whether or not this predicted long-term performance is adequate, an overall performance criterion is needed.

No overall performance criterion for geologic waste disposal has yet been adopted by the federal agencies. The Environmental Protection Agency (EPA) has been working for several years to develop such a standard. The EPA staff has written several internal drafts and supporting technical reports, and a proposed standard has recently been issued for public review. This begins the formal process for promulgating an EPA regulation that would adopt some overall performance criterion as its standard. The Nuclear Regulatory Commission (NRC) has issued both draft and proposed final regulations, which contain detailed numerical criteria for individual components and are intended by NRC to implement the proposed EPA standard.

As is explained later, the panel finds technical flaws in EPA's derivation and justification of its proposed standard. In reviewing NRC's proposed regulations, we conclude that they are premature in that they purport to implement an overall EPA standard not yet issued and not yet subjected to the review process wherein the bases and merits of the standard can be fully examined. We also find flaws in NRC's technical basis for some of its numerical criteria. Similar reservations concerning NRC's numerical criteria have been noted by the National Research Council's Board on Radioactive Waste Management (Wilson and Krauskopf 1981). Consequently, we are reluctant to adopt the EPA and NRC approaches for selecting an overall criterion for this study.

The Department of Energy (DOE) has not adopted, to our knowledge, an interim overall criterion for evaluating waste-isolation performance, although authors of studies by DOE contractors have assumed values of the individual-dose rate as criteria for comparing with calculations of doses from radionuclide migration and release.

Therefore, the panel found it necessary to adopt its own performance criterion for the purposes of this study. We conclude that the most meaningful and useful form of the criterion is the annual or lifetime radiation dose to an individual exposed at some future time to radionuclides released to the environment from a geologic repository. We have adopted as our criterion an annual radiation dose of 10^{-4} Sv* to an individual, averaged over his lifetime, calculated at all future times. This dose criterion is to apply to estimated average annual doses resulting from events that have a high probability of eventually occurring.

When the predicted radiation exposure is due to some unexpected release of radioactivity, i.e., a release probability of less than unity, then the predicted dose should be weighted by the probability before comparing with this performance criterion. This is qualitatively similar to the present NRC-EPA practice of allowing greater individual radiation exposures for suitably improbable accidental releases of radioactivity.

One reason for adopting the individual-dose criterion is that the individual dose from released radionuclides can be expressed as some fraction of the radiation dose that each individual receives from background radiation. Another reason is the considerable precedence for individual-dose criteria in radiation safety, as is reviewed in Section 8.2. In Section 8.3 we discuss why a criterion based on radiation exposures as low as reasonably achievable is not meaningful at this time. In Section 8.4 we discuss the features of an alternate approach, in terms of a criterion based upon population risk or collective population dose. In Section 8.5 we review the EPA proposed population-risk-based standard, and we discuss our reasons for not adopting such a performance criterion or the activity release limits derived therefrom.

In Section 8.6 we review the numerical criteria proposed by the NRC to implement the EPA's proposed standard, and we discuss the reasons for not adopting the NRC staff's numerical criteria for the purpose of this study. In Section 8.7 we discuss the compatibility of geologic isolation with the criterion of suitably low radiation dose to future individuals.

8.2. PRECEDENTS FOR AN INDIVIDUAL-DOSE CRITERION

8.2.1. Considerations in Specifying an Individual-Dose Criterion

Two arguments for an individual-dose criterion are (1) individual dose can be more meaningfully predicted than can a population dose and (2) even if a population-dose criterion is adopted, an individual-dose criterion would necessarily also be included to prevent undue individual exposure.

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*1 sievert (Sv) = 100 rem.

A consideration in selecting a value for an individual-dose criterion for geologic disposal is that the value of the individual dose be low enough that the number of individuals potentially exposed is not a major consideration. We examined several existing radiation standards developed for protection of the public from radionuclides released to the environment. Natural background radiation exposure was also considered in selecting a value of the individual dose criterion.

8.2.2. Recommendations by the International Commission on Radiological Protection

The International Commission on Radiological Protection (ICRP) concludes that a lifetime exposure to an individual member of the public of 10^{-3} Sv/yr corresponds to an acceptable level of risk, based on a total stochastic risk factor of about 1.4×10^{-2}/Sv (International Commission on Radiological Protection 1977). The commission recommends a limit of 5×10^{-3} Sv for the exposure to an individual in any one year, and it concludes that this would result in an average dose rate equivalent of less than 5×10^{-4} Sv/yr to members of the public. These exposures are considered to be in addition to exposure from background radiation.

8.2.3. Federal Radiation Council Guidance

In 1960 the U.S. Federal Radiation Council (FRC) issued guidance for exposure to members of the public from federally sponsored or licensed activities. The basic radiation protection guide (RPG) is 5×10^{-3} Sv/yr to any individual (the maximally exposed individual). The FRC also stated that the average annual dose to a particular exposed group should not exceed 1.7×10^{-3} Sv/yr (U.S. Federal Radiation Council 1960). These RPGs are also implied as the basis for most of the present limits in the U.S. NRC (1982a, Appendix B) regulation concerning concentrations of radionuclides in effluents from licensed facilities.

8.2.4. The Environmental Protection Agency's Fuel Cycle Standard

The EPA published an environmental protection standard for the uranium fuel cycle that contains a dose equivalent limit of 2.5×10^{-4} Sv/yr to any individual member of the public (U.S. Environmental Protection Agency 1976b). This standard has been incorporated by the NRC in its regulations for nuclear fuel cycle licensees (U.S. Nuclear Regulatory Commission 1982a) and has been proposed by the NRC for inclusion in its rules on commercial low-level (U.S. Nuclear Regulatory Commission 1981d) and high-level (U.S. Nuclear Regulatory Commission 1981c) waste disposal. In the EPA high-level waste standard, it is proposed to include the dose limits for application to the above-ground preemplacement operations of a high-level repository (U.S. Environmental Protection Agency 1982).

8.2.5. Other Regulatory Standards

A number of other regulations have adopted radiation dose or dose rate to the individual as the radiation safety criterion, with values varying from below 10^{-4} Sv/yr up to 5×10^{-3} Sv/yr. Some of these regulations were developed for situations wherein the radionuclides are already present in the uncontrolled environment. They include:

- 5×10^{-5} Sv/yr, uncontrolled release of biomedical wastes containing tritium and carbon-14 (U.S. Nuclear Regulatory Commission 1981b)
- 10^{-4} Sv/yr decommissioning of nuclear facilities (U.S. Nuclear Regulatory Commission 1981a)
- 2.5×10^{-4} Sv/yr, equivalent whole-body dose to an adult from public water supply from radium (U.S. Environmental Protection Agency 1976a)
- 4.3×10^{-4} Sv/yr, whole-body exposure for home construction on phosphate lands (U.S. Environmental Protection Agency 1979)
- 9×10^{-4} Sv/yr, gamma dose from disposal of uranium wastes (U.S. Nuclear Regulatory Commission 1981f)

8.2.6. Natural Background Radiation

According to the National Council on Radiation Protection and Measurements (NCRP), whole-body irradiation from natural sources results in dose rates to individuals in the United States varying from 0.7×10^{-3} Sv/yr to about 2×10^{-3} Sv/yr, depending on location (National Council on Radiation Protection and Measurements 1975). This includes penetrating radiation from cosmic rays, from naturally occurring radionuclides in the atmosphere and in the earth's crust and from radionuclides in the human body. The average individual whole-body dose rate from these sources in the United States is about 10^{-3} Sv/yr.

The variation of background radiation dose with location is due to several factors. Cosmic radiation intensity increases with altitude; the average whole-body dose rate from cosmic rays at an elevation of 1 mile (1.6 km) is approximately twice the sea-level dose rate of about 3×10^{-4} Sv/yr (National Council on Radiation Protection and Measurements 1975). Variations in the concentration of naturally occurring radionuclides in the earth's crust also affect the doses received. Regional whole-body dose equivalent rates from terrestrial sources range from about 2.3×10^{-4} Sv/yr in the Atlantic and Gulf coastal plain to about 9×10^{-4} Sv/yr in the Colorado plateau. The above-average dose rates in the Colorado plateau region of the United

States are due to extensive deposits of uranium (National Council on Radiation Protection and Measurements 1975).

Internal doses vary considerably as a result of variations in radioactivity concentrations in drinking water and air due to natural terrestrial sources. In some communities, elevated levels of radionuclides in the uranium-238 decay series (primarily radium-226 and daughters) in domestic water supplies result in dose rates to organs such as bone surfaces that are several times above the average U.S. bone dose rate of about 1.2×10^{-3} Sv/yr (National Council on Radiation Protection and Measurements 1975). Exposure to respiratory tissues, and to a lesser extent to other internal organs, results from the inhalation of naturally occurring radioactive gases, primarily radon-222 produced from the decay of radium-226. This is more of a problem inside buildings constructed on soil or fill materials containing natural uranium or thorium. The inhalation dose varies with several factors such as building ventilation rates, and lung doses that are several times the U.S. average lung dose of about 2×10^{-3} Sv/yr can occur (National Council on Radiation Protection and Measurements 1975). The organ doses given here have been adjusted upward by a factor of two to account for the increase in the quality factor for alpha radiation from the value of 10 used in the 1975 NCRP report to the current recommended value of 20 (International Commission on Radiological Protection 1977).

8.2.7. Use of Individual-Dose Criteria in Other Countries

It is evident from publications and project reports that the lifetime dose commitment to future individuals, or the average lifetime dose rate to individuals, is being calculated in other countries to assess the performance of geologic isolation systems (e.g., Wuschke et al. 1981). Limits to individual dose have been adopted by the Swiss regulatory authorities as safety protection goals (Nuclear Energy Agency 1980). In a draft report by the International Atomic Energy Agency (1982) for criteria for underground disposal, the principal numerical radiological protection criterion is specified in terms of the radiation dose to future individuals.

8.2.8. Summary and Discussion

The average lifetime dose rate to an individual of 10^{-4} Sv/yr selected as the criterion for this study is about 10 percent of the average annual effective whole-body dose equivalent from all sources of external background radiation in the United States. It is also less than half of the current average annual dose commitment from the ingestion of naturally occurring radionuclides. There is some parallel to future doses due to radionuclides released from a geologic repository, because ingestion is predicted to be the most important potential mode of exposure. The individual-dose criterion of 10^{-4} Sv/yr is well within the range of variations in the natural background dose rate.

The dose standards and criteria now applied by EPA and NRC for regulatory purposes suggest that a dose equivalent of 2.5 x 10^{-4} Sv/yr to the average individual in a particular exposed population group or to the maximally exposed individual is considered to be sufficient protection for public health and safety. The panel has seen several regulations or proposed regulations that use this value. We have also seen a regulation that uses 10^{-4} Sv/yr and one that uses 0.5 x 10^{-4} Sv/yr.

The values of 2.5 x 10^{-4} Sv/yr are applied to radionuclides that are, in many instances, identical with some of the more important radionuclides of concern in a geologic repository for high-level waste. Examples are the application of 2.5 x 10^{-4} Sv/yr to ground disposal of low-level radioactive wastes (U.S. Nuclear Regulatory Commission 1981e) and radium in drinking water (U.S. Environmental Protection Agency 1976a). Other examples are the 10^{-4} Sv/yr limit for decommissioning nuclear facilities, together with the bone-dose equivalent exposure limits of 3 x 10^{-5} Sv/yr for uranium and transuranic wastes.

The ICRP has concluded that an annual whole-body equivalent dose of 10^{-3} Sv/yr represents an acceptable level of risk such that no limit need be placed on the size of the exposed population (International Commission on Radiological Protection 1977). This is tenfold greater than the value selected by the panel.

Finally, several regulations or guides use 5 x 10^{-3} Sv/yr as an acceptable limit for exposure to individual members of the public, including the proposed NRC rule on low-level waste disposal.

These observations suggest that the panel's individual-dose criterion of 10^{-4} Sv/yr provides an adequate margin, of about two orders of magnitude, below a dose that could be reasonable cause for concern.

8.2.9. Application of the Panel's Performance Criterion

The panel wishes to make clear that the individual-dose criterion of 10^{-4} Sv/yr is not intended as an upper limit of radiation exposure. It is simply a goal against which a particular repository system performance can be compared. The dose value of 10^{-4} Sv/yr is sufficiently low so as to provide reasonable assurance that no member of the public will be exposed to a radiation risk greater than that experienced and permitted from natural sources in day-to-day life. Other, higher limits (International Commission on Radiological Protection 1979) should be used to evaluate the upper levels of exposure estimated from the uncertainties inherent to the parameters used for calculating system performance.

8.3. AS LOW AS REASONABLY ACHIEVABLE

The panel has made no evaluation as to whether the individual dose rate criterion of 10^{-4} Sv/yr is as low as is reasonably achievable, referred to by the U.S. Nuclear Regulatory Commission (1982a) as ALARA. According to the International Commission on Radiological Protection (1973), determining whether radiation exposure from a proposed activity is as low as reasonably achievable requires, among other things, an assessment of the costs of protective measures weighed against the expected benefits from the activity. Present data on the design, costs, and expected performance for geologic waste disposal are not sufficient for any meaningful analysis of the costs of incremental reductions in radiation dose. As will be seen in the ensuing discussion of this chapter and in Chapter 9, there are large uncertainties in estimating the radiation dose, particularly if it is the population dose usually considered in ALARA calculations, and there are large uncertainties in the technological performance of a geologic disposal system. It is not possible to make any meaningful determination at this time of what radiation doses and dose rates would be as low as reasonably achievable.

8.4. FEATURES OF INDIVIDUAL-RISK AND POPULATION-RISK CRITERIA

A central issue is whether to select for the present study an overall performance criterion based on the radiation dose, or risk to future individuals, or on the integrated radiation dose, or risk to future populations. There are important features to each.

A population-risk criterion, if implementable, could avoid undue risk to a large number of people and over many generations. A given risk to an individual may be reasonable if it does not exceed some specified level of acceptable risk. Radiation exposures that result in risks not far below the specified acceptable limits for individuals may be unacceptable if large numbers and many generations of people are similarly exposed. Some geologic repositories for radioactive waste are predicted to release small quantities of radionuclides over long periods of time, and there are mechanisms by which this released radioactivity can reach even distant populations. This argues for serious consideration of a population-risk-based criterion for geologic repositories.

If such a population-risk limit could be meaningfully implemented, it would be reasonable to accept an allowable dose to a few individuals that is greater than the average lifetime individual dose to the population. However, a population-risk criterion alone, as is proposed by EPA for geologic repositories, can allow intolerable risk to a few individuals, as is shown in the ensuing analyses in this chapter and in Chapter 9.

The practical differences between an individual-risk criterion and a population-risk criterion can be illustrated for a geologic repository for which the expected risk results from the long-term groundwater transport of a small portion of the radioactive inventory to the environment. Future humans can use that contaminated water for drinking and for growing food. A future maximally exposed individual is conservatively assumed to be one who obtains his lifetime intake of potable water from this contaminated source and his lifetime intake of food from edible species grown in or by this contaminated water. This lifetime radiation dose can be estimated from estimated concentrations of released radionuclides in water and from food chain calculations. By assuming that risk is proportional to accumulated dose, the calculated individual dose is translated to an estimated risk to that individual. This assumption is the linear hypothesis that has been adopted by the Committee on the Biological Effects of Ionizing Radiation (BEIR)(National Research Council 1980) for estimating stochastic risks that might be associated with the lifetime exposure of a population of individuals to radiation at a dose equivalent rate of about 10^{-2} Sv/yr or greater, provided the exposures are well below the range of acute exposures where the effects become nonstochastic. Adoption of this linear hypothesis for all levels of lifetime exposure that could result from a geologic repository is a fundamental assumption in EPA's proposed standard (U.S. Environmental Protection Agency 1982).

There are many uncertainties in such a calculation of maximum individual dose, including uncertainties in the prediction of waste dissolution and hydrogeologic transport. The dose calculation assumes that future individuals will have dietary habits similar to those at present. It assumes that they will drink about the same amount of water, will eat the same amount and kind of food, and will respond in the same way to ingested radionuclides. It assumes that agricultural practices and uptake of radionuclides in food chains will be the same as at present. The result of a criterion based on individual dose is to limit the maximum concentration of individual radionuclides in water contaminated by future radioactive releases from a repository. This maximum concentration can be controlled by choosing radioactive waste material with suitably small rates of dissolution, by selecting repository sites with suitably long travel times for contaminated groundwater to reach the environment, and by selecting repository sites with natural processes that dilute the concentration of released radionuclides.

Once a population-risk criterion has been specified, to determine compliance, such as the maximum number of health effects from the entire exposed population over some specified number of generations, it will be necessary, in principle, to integrate the individual radiation exposures and risks therefrom over extremely large numbers of people and over many generations. Some of these individuals may be the maximally exposed individuals described above who live in the vicinity of the repository site; others will live elsewhere but will consume some of the food products grown from the contaminated groundwater. A large number of these individuals will receive extremely small radiation doses.

To estimate each individual risk, it is necessary to know the relation between individual dose and risk for individual doses ranging from the minuscule to the maximum described above. No proved or recommended relation exists over this dose range (cf. Section 8.5.7). However, if one assumes, as does EPA, that the linear hypothesis is valid for all values of the incremental collective (i.e., accumulated) dose to the many different exposed individuals, then the estimate of population risk translates to an estimate of the total radiation dose summed over all of the exposed people for many generations.

A meaningful estimate of the total intergenerational population dose requires detailed estimates of the time-dependent number of individuals who may be living in the vicinity of the repository site and the extent to which their water and food are derived from the contaminated water, and it requires estimates of the extent to which this contaminated water is used to grow food for shipment to other parts of the world. It requires estimates of the time-dependent numbers, geographical distribution, and eating habits of the future populations who may eat food grown by the contaminated water. Clearly, the direct estimate of population dose and risk involves far more uncertainties than does the estimate of the dose and risk to the maximally exposed individual.

If the population-risk criterion is translated by the regulator into a limit on the amount of radionuclides released from a repository, as has been attempted by EPA (1982), then the derived release limit is subject to all of the uncertainties described above for determining compliance with a population limit, and the uncertainties are compounded by the need to assume how radioactivity released from presently unspecified repository sites will result in radiation doses to future humans. These problems inherent in EPA's derivation of release limits to achieve a specified population-dose criterion are discussed in Section 8.5.

The implementation of a population-risk criterion, or of a release-limit criterion derived therefrom, could result in a different dependency of risk on the concentration of radionuclides in the contaminated water. For example, if a low radionuclide concentration results because of dilution in water at the repository site, the dose to the maximally exposed individual is reduced, but there may be a larger number of future individuals who use the contaminated water. If it can be assumed that, within a given generation, the number of people using the contaminated water or ingesting food grown from the contaminated water is proportional to the volume or flow rate of the contaminated water, then a population-dose limit for that generation would result in a limit on the amount of radionuclides released to the environment during a human lifetime. It would not limit the concentration of these radionuclides in contaminated water.

If an intergenerational population-dose or risk criterion is to be applied, and if parameters assumed above to be constant over a human lifetime are now assumed to be constant over the many tens and hundreds and thousands of years found to be important in estimating individual doses, then again a population-dose limit would not limit the concentration of radionuclides in contaminated water but would limit the total amount released over the long time period of many generations.

Application of a population-dose criterion can result in views as to the adequacy of a repository site that can be quite different from those resulting from an individual-dose criterion. Large differences in estimated doses to maximally exposed individuals for different repository sites are shown in Sections 9.7 and 9.10, where the different individual doses result mainly from differences in water flow rates. The contrast between different sites is less apparent if a population-dose criterion is applied, without considering doses to individuals.

In principle, dose criteria that specify both the dose to the maximally exposed individual and the intergenerational dose to future populations should be considered. However, the latter should be adopted as a numerical criterion only if it can be reasonably implemented without requiring the accumulation of unrealistic assumptions that would undermine its validity. If this proves to be impossible, a semiquantitative or qualitative criterion may still serve the purpose of limiting the integrated risk over many generations to a tolerable level. This would help emphasize that compliance with an individual-risk criterion should be achieved to a maximum reasonable extent by containment and decay within the geologic disposal system.

In past regulatory practice (e.g., 10 CFR 20, Sect. 106(e)), it has frequently been assumed that the choice of a suitably conservative individual-dose criterion, such as that adopted in this study, would result in acceptably low population doses. The same principle may apply to geologic repositories, although this assumption would require some detailed study.

8.5. THE EPA PROPOSED STANDARD

8.5.1. Introduction

The proposed EPA standard has undergone an evolutionary process. Draft number 12 was provided to the panel in September 1980. Meetings and extensive written communications have been held with members of the EPA staff to discuss their technical work that forms the basis of the proposed standard. Several drafts have been provided to the panel during the course of the study, and the proposed standard was released for public comment in late 1982 (U.S. Environmental Protection Agency 1982). Our technical evaluation is based on these drafts, on discussions and communications with the EPA staff, and on EPA draft technical reports (C. B. Smith et al. 1981, J. M. Smith et al. 1981) provided to the panel for the purpose of this review. Additional draft reports by the EPA staff are understood to be pertinent to this review, but they were not provided during the panel study. These include an individual-dose assessment report, an environmental impact statement, and other identified reports (D. J. Egan, Environmental Protection Agency, personal communication to T. H. Pigford, 1981). The review summarized herein is based in part on a more detailed technical analysis by Pigford and Mann (1982) carried out to support the panel's study.

A fundamental premise of the proposed EPA standard is that there should be no more than 1,000 fatalities ("health effects") in the next 10,000 years resulting from radioactive releases from a full-scale geologic repository, i.e., a repository containing the radioactive inventory from 100,000 Mg of uranium fuel from light-water reactors. EPA proposes to achieve that objective by prescribing quantitative limits on the amounts of radioactivity from various radionuclides that could be released to the "accessible environment" over a period of 10,000 years.

8.5.2. EPA Estimates of Population Intake of Released Radioactivity from Worldwide Averages

The EPA approach has confronted--or avoided--the problem of estimating future population doses from a geologic repository by developing world average statistics on food and water consumption and predicting from those what fraction of the radioactivity released to the environment from a repository will eventually be ingested by future humans over an assumed time period of 10,000 years. The EPA adopts world averages for the use of flowing surface water for drinking and in irrigating crops, growing marine life, etc. It then extrapolates these world averages over 10,000 years by assuming a constant future world population of 10^{10} people consuming the available surface water and food, about threefold greater than the present world population.

From these extrapolated world averages and population assumptions, EPA estimates what fractions of the world river flow are used for drinking and for growing food, and then assumes that these fractions apply to the use of contaminated surface water and groundwater near any site that is chosen for a geologic repository. This applies regardless of repository location, except where the underground formation does not provide a practical source of water. Next, they estimate the retention of radionuclides in soil as a result of irrigation by contaminated water, and they estimate the uptake of these radionuclides into plants and then into animals and humans. They then assume that these soil retention and plant uptake characteristics are the same, regardless of where a repository is located. Finally, EPA assumes that the retention of radionuclides by humans eating the crops and animals nurtured by the contaminated water will be the same for all population groups that may consume this food, now and for the next 10,000 years.

8.5.3. EPA Calculation of Population-Dose-Based Radioactivity Release Limits

Based upon averages of the uptake of various chemical elements from water by soil, plants, animals, and ultimately by humans, EPA calculates the fraction of released radionuclides that will eventually be ingested by humans. From its own calculations of radiation dose per unit amount of radioactivity ingested, and by adopting the linear hypothesis relating risk to collective dose, they then estimate the total number of health effects that would result from a given amount of radioactivity released from the repository, regardless of concentrations of released radionuclides in the contaminated water and regardless of repository location. Then they calculate population-based radioactivity release limits for individual radionuclides, such that any one radionuclide species, if released alone and at any time during the 10,000 years, would result in the limit of 1,000 health effects. When several radionuclide species are released, the sum over all species of the ratio of the amount released to the release limit must not exceed unity.

The EPA technique of calculating release limits is illustrated in Table 8-1. The total number of health effects per unit activity

TABLE 8-1 Activity Release Limits for 10,000 Years, as Proposed by EPA Standard

Radionuclide	Health Effects per Becquerel Released to a River[a]	Release Limit for 1,000 Health Effects	
		Becquerels[b]	Curies[c]
^{14}C	1.24×10^{-12}	8.1×10^{14}	2×10^{4}
^{90}Sr	3.27×10^{-12}	3.0×10^{14}	8×10^{3}
^{99}Tc	7.70×10^{-15}	1.3×10^{17}	1×10^{6}
^{126}Sn	3.24×10^{-12}	3.0×10^{14}	8×10^{3}
^{129}I	2.92×10^{-13}	3.4×10^{15}	9×10^{4}
^{135}Cs	1.03×10^{-13}	9.7×10^{15}	2×10^{5}
^{137}Cs	5.35×10^{-13}	1.8×10^{15}	5×10^{4}
^{226}Ra	8.92×10^{-11}	1.1×10^{13}	3×10^{2}
^{237}Np	1.61×10^{-11}	6.7×10^{13}	2×10^{3}
^{238}Pu	6.19×10^{-13}	1.6×10^{14}	4×10^{4}
^{239}Pu	1.87×10^{-12}	5.3×10^{14}	1×10^{4}
^{240}Pu	1.76×10^{-12}	5.7×10^{14}	1×10^{4}
^{242}Pu	1.83×10^{-12}	5.5×10^{14}	1×10^{4}
^{241}Am	1.94×10^{-11}	5.1×10^{13}	1×10^{3}
^{243}Am	7.24×10^{-11}	1.4×10^{13}	4×10^{2}

NOTE: Data are based on a repository containing radioactive waste from 10^5 Mg of uranium fuel from light-water reactors. Each release limit applies if no other radionuclide is released.

[a] Derived from data of J. M. Smith et al. (1981).

[b] 1,000 divided by health effects per becquerel released to river.

[c] U.S. Environmental Protection Agency (1982); U.S. Nuclear Regulatory Commission (1982c).

released to river water, derived from EPA data for worldwide averages as described above, is divided into the EPA goal of 1,000 health effects to obtain the release limits (Pigford 1981).

The EPA draft states that these population-risk-based release limits are to be applied to those "reasonably foreseeable" processes and events that could result in radioactivity releases, such as human intrusion. The panel has not evaluated the possibilities and consequences of human intrusion into a repository. However, we expect that human intrusion is not certain to occur, and it is therefore less likely than the long-term dissolution of radioactive waste in a repository with flowing groundwater, followed by the long-term transport of dissolved radionuclides to the environment. It is this hydrogeologic transport that is the focus of the present study, and the EPA release estimates are later discussed with regard to that release mechanism.

The EPA also recognizes that moderate releases of radioactivity could result from much less likely events, such as fault movements or other disruptive geologic events, and it proposes that the release limits for these "very unlikely releases" shall be less than 10 times the quantities shown in Table 8-1.

The EPA designates as "extremely unlikely" very large releases that would result only from the intrusion of volcanos or impacts by huge meteorites, but it identifies no release limits for these events.

We note that, in its rationale for proposed technical criteria to implement the release limits in the EPA's proposed standard, the NRC treats water intrusion into a salt repository as a "reasonably foreseeable" event and considers the resulting waste dissolution and hydrogeologic transport to the EPA-defined accessible environment in relation to the Table 8-1 release limits. No justification has been given for assigning such likelihood to this disruptive event for a salt repository, and we do not agree that hydrogeologic transport from a repository in natural salt is as likely as hydrogeologic transport from a repository in rock expected to contain flowing groundwater, such as granite, basalt, or saturated tuff.

8.5.4. Radioactive Releases to the Accessible Environment

The EPA proposed standard states that these release limits apply to releases to the accessible environment, defined as the atmosphere or a surface or underground location more than 10 km from the emplaced waste. Although the EPA release limits were derived from the hypothesized use of river water, the limits are evidently intended to apply to whatever contaminated water occurs at the 10-km location. With this definition, EPA intends to protect aquifers that might become significant sources of water in the future, regardless of whether they are now being used as water supplies. EPA states that specific exceptions can be made for underground formations that are impractical sources of water, for example, because of depth or low productivity of water.

Evidently there would be no requirement, in complying with the EPA proposed standard, to consider releases to the actual environment or to seek sites with greater distances from the waste to the actual environment wherein humans may be expected to use the contaminated water.

8.5.5. Compliance Would Be Reduced to a Simple Calculational Exercise

The EPA proposed standard contains no requirement for estimating radiation dose to the individual or any dose criterion relating thereto. It requires no prediction of radioactive releases and radiation doses for time periods greater than 10,000 years.

To the repository designer, the proposed standard may seem attractive. It seems to fulfill the objectives of a population-based criterion, yet it requires no data or calculations concerning the numbers, location, and eating habits of future populations affected by the repository being designed. It avoids the more difficult problem of making accurate predictions of releases over time periods of millions of years. It requires relatively little hydrologic information concerning the repository site, because there would be no need to calculate the concentrations of individual radionuclides in groundwater and surface water and individual radiation doses therefrom. It requires no knowledge of the need or practicability of using contaminated water for irrigation in the vicinity of the site, and it requires no knowledge of the retention of radionuclides by the actual soil that is to be irrigated or of the uptake of radionuclides by the actual food crops, if any, that may be indigenous to that location. It requires neither calculations of radiation dose nor investigation of uncertainties thereof.

Not only would the EPA proposed standard simplify the calculations for compliance, it would seem to have relatively little effect upon site selection. For any repository location, the same fraction of released radionuclides would be assumed to be ingested by humans as for any other location. Large existing populations and expected future populations in the vicinity of the site would evidently not enter the calculations of compliance. The technical basis of the standard, as described above, assumes that it makes no difference whether the released activity is distributed over a large number of people or whether it is all ingested by a small number of people; in either case EPA expects the population dose to be the same. According to the EPA assumptions, if the activity were released to a rapidly flowing river, the activity concentration in the river water would be small but the number of people using the water would be large, with the same result as if the activity were released to a small surface stream in high concentrations.

From its definition of the accessible environment, the EPA seems to have given no consideration to the wide variations in possible and likely use of potentially contaminated water at the 10-km location, whether it is readily obtained surface water or underground, whether it is potable or saline, and whether it is in an arid region where groundwater use is likely. It is clear that EPA is saying that, regardless of repository location, radionuclides released from a repository would enter water used in precisely the same way by future people as river water is used by the present worldwide average population, adjusted only by the assumed threefold increase in worldwide population.

No consideration seems to have been given to the possible and likely new technologies that may markedly change the ways in which water is used by humans. Although humans have been drinking water and growing food for a very long time, the techniques for obtaining and using water for these purposes have changed drastically during recent times. The EPA extrapolation of present world-average quantities to 10,000 years is based upon but a brief period in history. However, the uncertainties in this extrapolation would evidently not need to be considered in predicting future radiation exposures for a repository if the EPA proposed standard were adopted.

If a repository is sited where released radionuclides can discharge into nonflowing surface water, the EPA limits assume that such nonflowing contaminated water is used in the same way as the worldwide average use of river water. No consideration seems to have been given to the possible reuse of surface water and to the possible accumulation of released radionuclides in a nonflowing system, nor would such consideration evidently be required of the repository designer.

The EPA release limits can result, in some cases, in expected health effects for real candidate sites that are far lower than in the health effect goal basic to the EPA calculation, and in other cases population exposures and numbers of health effects much greater than the EPA objective seem likely. For example, EPA assumes in its calculations that one-half of the contaminated water will be used for irrigation and 0.02 percent will be used for drinking. The possible and likely variability of these use fractions among real sites is obvious. This likely variability in achieving the stated public health objective does not meet the test of a useful and meaningful standard.

We conclude that the EPA has not proposed a useful or meaningful way of obtaining its goal of limiting population risks from a geologic repository. A redeeming feature of the proposed standard is that it makes for simple calculations of compliance, but that does not justify its adoption.

8.5.6. Individual Doses Implied by the EPA Standard

From the hydrologic and individual-dose parameters presented later in Tables 9-1 and 9-4 of Chapter 9, we estimate that the EPA activity release limit for technetium-99 could result in an individual dose rate as low as 3×10^{-8} Sv/yr if that amount of technetium-99 were discharged uniformly over a period of 8,000 years into the Columbia River flowing at 1.1×10^{11} m^3/yr. This would be the average annual whole-body dose to an individual who drinks only the contaminated river water and whose entire food supply is grown from the contaminated water. It is a radiation exposure about five orders of magnitude below background radiation.

An even lower individual dose can be inferred from the EPA calculations. These calculations are not concerned with whether or not all the water and food consumed by an individual is that derived from the contaminated river. Recognizing that food grown from the contaminated water can be shipped worldwide, the EPA averages for the projected world use of river water allow for the possible worldwide distribution of a fraction of the released radionuclides as ingestible products. From the data provided by the EPA staff (J. M. Smith et al. 1981), we estimate that their release limit for technetium-99, if discharged uniformly over 8,000 years, would result in an average annual dose to the individuals in the world population as low as 6×10^{-9} Sv/yr.

If the water at EPA's accessible environment happens to be contaminated water flowing underground at a rate as low as 10^3 m^3/yr (cf. Table 9-4), the EPA release limit for neptunium-237 could result in an individual dose rate as high as 100 Sv/yr. This annual dose is high enough to introduce the possibility of acute, short-term radiation effects that are not included in the linear risk-dose relation adopted by EPA. No requirement to calculate such individual radiation doses appears in the EPA proposed standard or in the NRC (1982c) final regulation that is intended to implement that proposed standard.

8.5.7. The Use of the Collective-Dose Linear Hypothesis for Low Individual Exposures

Not only can the EPA release limits result in individual doses so high as to cause possible short-term health effects as well as the longer-term fatal cancers implied by EPA's dose-risk proportionality, but the very low possible individual doses, distributed over so many people, can raise further questions about the validity of the EPA assumption. Conversion factors developed by the EPA relate collective dose to the number of expected fatal cancers from radiation exposure to several organs of the body and to first-generation genetic effects from radiation exposure to the reproductive organs (J. M. Smith et al. 1981). These factors were derived on the "linear hypothesis" assumption of a constant proportionality between collective dose and number of health effects.

The EPA use of the linear hypothesis in relating population dose to health effects is of questionable validity when it is applied to collective doses obtained, in effect, by summing very small individual doses over a very large number of people. The National Academy of Sciences Committee on the Biological Effects of Ionizing Radiation (BEIR) (National Research Council 1980) reviewed the available data on the possible dose-response relationships at low doses to humans. The committee pointed out that many difficulties and uncertainties remain in the estimation of risks from low doses, and the committee was not willing to make quantitative estimates of the risk that might be associated with the lifetime exposure to a population of individuals exposed to a radiation at a dose equivalent rate of less than 10^{-2} Sv/yr. Yet, in developing its proposed population-risk-based standard, the EPA is applying the risk-dose proportionality to individual doses delivered at essentially constant rates that can be as much as five orders of magnitude below the BEIR Committee's own low-rate limit. This introduces further uncertainty as to the validity of the EPA-derived release limits as a means of achieving the population-risk objective stated by EPA.

8.5.8. EPA 10,000-Year Time Limit

The EPA staff justifies its assumption that the calculations of future radioactivity releases and radiation doses therefrom can be limited to a time period of 10,000 years on the following grounds:

- Releases that will have occurred by this time will not be deceptively low.
- It is more difficult to calculate reliably the releases over longer times.
- A disposal system capable of meeting the EPA requirements for 10,000 years will continue to protect people and the environment beyond 10,000 years.

o Radioactivity in the buried wastes will have decreased by 10,000 years to a hazard level about the same as from an equivalent amount of unmined uranium ore.

It is characteristic of geologic repositories that radionuclides released by groundwater transport are expected to be important over time periods much greater than 10,000 years. Although uncertainties in estimating future releases are greater for the longer time periods, there is no discontinuity in predictive reliability at and beyond 10,000 years. There are data that allow prediction of releases at greater times, and there are methods for estimating uncertainties at times extending well beyond 10,000 years.

In selecting the 10,000-year time limit, EPA concluded that for a shorter time limit the amount of radionuclides reaching the accessible environment would be "deceptively low." The inference that significant releases will have occurred within the first 10,000 years is contradicted by NRC's (1982c) regulation requiring that the fractional rate of release of radionuclides from the waste packages be no greater than 10^{-5}/yr, beginning no sooner than 1,000 years after emplacement. Assuming this NRC criterion, and assuming NRC's required minimum water travel time of 1,000 years from the waste to the accessible environment, only about 8 percent of a long-lived nonsorbing radionuclide could be released before EPA's 10,000-year time limit. Only 7 percent of the carbon-14 could be released. Based upon the recommended retardation coefficients in Table 7-1, only 4 percent of the technetium-99 and none of the other radionuclides would reach the accessible environment within EPA's time limit. Yet, as can be deduced from calculations presented in Chapter 9 (cf. Figure 9-1), most of the releases and time-integrated radiation dose would occur, for the NRC parameters, in the time interval of about 2×10^4 to 10^6 years, even for an assumed constant population. This interval would be excluded from consideration by the EPA proposed standard.

As shown in Chapters 7 and 9, water travel times of 10,000 years and greater are expected for many candidate repository sites, resulting in zero release of even nonsorbing radionuclides during the first 10,000 years. Important releases continue for millions of years. Clearly, the EPA time limit of 10,000 years for release to the environment makes for simple calculations of compliance, but it does not provide a long enough time to focus on the expected and likely releases of radionuclides and health effects therefrom.

A waste-disposal system capable of meeting EPA's 10,000-year release limits is not necessarily capable of continuing to protect people and the environment beyond 10,000 years. In Section 8.6.5 we show that NRC's proposed numerical criteria are sufficient, together with reasonable retardation coefficients, to meet the EPA release limits, in part because they delay releases at the 10-km location beyond the 10,000-year deadline. For most of the important radionuclides, these delays are not effective when releases beyond 10,000 years are considered. Releases of radium-226 during each subsequent 10,000-year period will increase and finally exceed the EPA release limit.

As shown in Figure 9-1 of Chapter 9, an entirely different set of radionuclides appears at the 10-km location after the first 10,000 years. The calculated population dose from these later radionuclides is greater than for the first 10,000 years, and these later radionuclides place different chemical and physical demands on the disposal system than those released during the first 10,000 years. Therefore, designing for performance during the first 10,000 years does not ensure adequate performance for subsequent periods.

Given the necessary increase in time period over which releases should be calculated, the uncertainties in EPA's population-dose technique become even greater. To fulfill the EPA objective of limiting future health effects, we must now predict numbers, locations, and habits of future populations over time periods of millions of years. In the absence of any meaningful approach toward such calculations of the future population dose from a geologic repository, we return instead to the individual-dose criterion that has been adopted for this study.

In arriving at the level of population risk that would be permitted by its proposed 10,000-year standard, EPA has observed that (1) most of the benefits derived in the process of waste production fall upon the current generation while most of the risks fall upon future generations and (2) that a criterion of "intergenerational equity" would require no risk to be passed on to future generations, but this criterion could not be met by foreseeable disposal technologies. This would seem to argue against selecting such a short time period for estimating future releases from a repository.

Quite a different point of view on the importance of long-term radiation doses received by future generations has been articulated by others (Parker 1981). In commenting on individual doses calculated for time periods of millions of years, it was suggested that a given radiation dose to a future individual should not be given the same weight as the same dose to a person 50 years from now. This would seem to argue for a relatively short time period for estimating future releases and doses therefrom.

These value judgments on our responsibility to future generations, or lack thereof, are beyond the expertise of this panel and are outside the scope of this study.

8.5.9. Technical Parameters Adopted by EPA

Apart from the questionable validity of the EPA approach in setting its 10,000-year release limit, we are uncertain about the validity of many of the technical data that have been adopted by EPA for its calculations. We have examined the data used by EPA to obtain the whole-body and organ doses per unit activity ingested for various radionuclides. In Appendix C these results are compared with estimates from other sources. The data from different sources vary by more than two orders of magnitude for some radionuclides. In comparing results from calculations of radionuclide uptake and accumulation in the food chain pathways, we infer similar large variations among the estimates from different sources, although EPA has not supplied sufficient data concerning its calculations of water use and plant and animal uptake for us to compare specifically its calculations with those at Pacific Northwest Laboratory (PNL) and at other places. There is no evidence of any such comparison having been made by EPA or by any of the other government agencies and their contractors. We are left with the conclusion that there may be large uncertainties in these calculations of radiation doses from contaminated water, that the magnitudes of the uncertainties are unknown, and that too little effort has been made to determine and evaluate these uncertainties.

8.5.10. EPA Evaluation of Best Available Technology

The EPA (1982) proposed standard states that the proposal is based on ". . . the best technology that can reasonably be achieved given current scientific, technical and fiscal capabilities." However, as described above, we are able to derive the activity release limits of the EPA proposed standard on the basis of the risk goal and data from world averages, with no reference to the technology for geologic waste isolation. Even so, the proposed standard is considered by the EPA staff to be "technology-based, not risk-based" (Goldin 1981) and the standard represents EPA's best estimates of how well a repository can be expected to perform without necessarily considering the risks that may develop from the releases (D. J. Egan, Environmental Protection Agency, personal communication to T. H. Pigford, 1981).

That EPA has made a reasonable and realistic assessment of the performance of available waste-isolation technology is questionable. DOE and its PNL and Office of Nuclear Waste Isolation (ONWI) contractors have commented specifically on many of the parameters adopted by EPA in its calculations of repository performance, pointing out that in many cases the EPA-adopted parameters are overly conservative (Heath 1981). In its analysis of conceptual repositories, EPA did not include calculations of the possible release of radium-226 and lead-210 to the accessible environment. Calculations made for the panel by Fujita et al. (1982) and Sato et al. (1982) show that, even adopting the parameters and the mixed-flow repository model assumed by EPA, these radionuclides are likely to be important contributors to the cumulative health effects.

The calculations for the panel have also considered release times well beyond the 10,000-year cutoff adopted by EPA (cf. Figure 9-1). EPA's 10,000-year cutoff gives a distorted and incorrect indication of the sensitivity of total releases and health effects to important parameters of the repository and site, such as the dissolution rate of the waste material, the effect of solubility limits, the effect of retardation constants, and the effect of water travel time. Thus, the panel cannot conclude that the EPA standard is based on an adequate assessment of the performance of available technology for geologic disposal of radioactive waste.

8.5.11. A Derived Release Limit Should Not Be Separated from Its Basis

As has been noted above, the EPA proposed release limits are not in themselves a safety goal for geologic waste disposal; rather they are quantities derived from the more fundamental safety goal of 1,000 health effects in 10,000 years. We have noted many uncertainties and assumptions of technical data used in deriving these release limits, and we are concerned not only that such release limits might be adopted but that they might also be adopted without full and continuing recognition of the uncertainties on which they are based.

It can be reasonably expected that new and better technical knowledge will change our view on what are the proper parameters to use in the derivation of such limits, just as we have learned within the very recent years that the biological risks from ingestion of radionuclides important in geologic waste disposal are considerably greater for some radionuclides and considerably less for others than heretofore expected. An overall performance criterion stated in terms of the desired goal, such as a maximum risk or radiation dose, requires continued attention to the changing technical knowledge of the factors that affect the magnitude of the risk or dose, but a derived release limit in terms of so much activity over a specified time limit neither invites nor requires such attention. This is another reservation we have about EPA's proposed standard, which only states release limits but does not give adequate recognition in the standard to the many assumptions embodied in their derivation.

The purpose of such a technical criterion in regulations should be to help achieve public health and safety. When sufficient and necessary, such a technical criterion can be useful as long as its technical bases remain sound. Regardless of how well founded and formulated such a technical criterion may be at the time promulgated, it is likely that increased technical knowledge will warrant change.

If the technical purpose and foundation of the original criterion is not clear and sound, and if the criterion is adopted without adequate reference to its basis, then the technical framework for later review and challenge of the need and adequacy of such a technical criterion has been lost. The technical criterion then becomes viewed as a set of arbitrary numbers sacred within themselves, rather than as a representation of necessary technical functions and performance that a criterion, such as a set of release limits, is intended to achieve. To the extent that some such derived technical criterion may be finally adopted, adequate descriptions of its technical purpose, foundation, and assumptions should be included.

8.5.12. Summary

Because of the problems of making any meaningful estimates of numbers, locations, and eating habits of future populations, because of the many uncertainties in EPA's derivation of release limits to achieve its objective of population risk, because of the lack of justification of the EPA 10,000-year time limit for consideration of future releases of radionuclides to the environment, and because the population-dose-based release limits can allow individual radiation exposures greater than what we consider to be reasonable, we do not adopt population dose or activity release limits as an overall performance criterion for our study.

8.6. THE NRC PROPOSED REGULATION TO IMPLEMENT THE EPA PROPOSED STANDARD

8.6.1. The Numerical Criteria

In July 1981 the NRC issued its draft proposed regulation 10 CFR 60 (U.S. Nuclear Regulatory Commission 1981c) for geologic repositories, stating that the regulation was intended to implement the EPA standard. The NRC draft included several numerical criteria for the performance of individual components of the waste-isolation system, including:

o No release of radionuclides from the waste package for 1,000 years after the repository is sealed.

o A fractional release rate of less than 10^{-5}/yr of the radionuclide inventory within the waste package after 1,000 years, but excluding any radionuclide released at a rate less than 0.1 percent of the calculated total annual release at 1,000 years.

o A minimum water travel time from the repository to the environment of 1,000 years.

No time limit is specified for the required release rate performance of the waste package.

8.6.2. Review by the Board on Radioactive Waste Management

At the request of the National Research Council's Board on Radioactive Waste Management, members of the Waste Isolation System Panel prepared a summary of the technical features of the draft 10 CFR 60 regulation, based on technical reviews and analyses then carried out by the panel. The resulting appraisal by the Board on Radioactive Waste Management (Wilson and Krauskopf 1981) of the NRC's draft regulation includes the following conclusions regarding NRC's proposed numerical criteria:

o NRC has not presented adequate evidence that these numerical criteria can "support a finding of no unreasonable risk to the health and safety of the public."

o NRC has not shown that these numerical criteria are either necessary or sufficient to meet the "EPA Standard."

o It has not been shown that adoption of the numerical criteria will simplify the licensing process.

o No attempt has been made to demonstrate the technical validity of the proposed criteria.

o NRC has not shown how the proposed numerical criteria for the waste package can be verified.

The board recommended ". . . that precise numerical criteria for major elements of the repository system be eliminated." The working paper that presented the technical basis for these conclusions was issued separately (Pigford 1981).

8.6.3. NRC's Proposed Final Rule

In July 1982 the NRC made public its proposed final 10 CFR 60 rule (U.S. Nuclear Regulatory Commission 1982c), accompanied by a new rationale (U.S. Nuclear Regulatory Commission 1982b). The numerical criteria proposed in NRC's first draft remain, but the NRC now reserves discretion so that it may require DOE to comply instead with "such other values as the Commission may approve or specify." At the time of this writing, the proposed final rule has not been adopted by the NRC commissioners.

The NRC document states that the final rule is to implement the activity release limits that appear in the EPA proposed standard, and it further states that the release limits are to apply at EPA's specified accessible environment, 10 km from the emplaced waste. These are the release limits discussed in Section 8.5.3, calculated to limit the population risk to 1,000 health effects over 10,000 years.

Our review of this proposed final rule is based in part upon a more detailed analysis by Pigford (1982), carried out to support the panel's study.

8.6.4. Problems of Estimating Population Distribution

In Section 8.5 we discussed the problems of estimating numbers, locations, and eating and drinking habits of future populations, a problem that must be confronted when a population-dose-based criterion is selected, as is the EPA's release-limit criterion that NRC proposes to implement. NRC seems also concerned with uncertainties when dealing with future populations. They acknowledge that such demographic factors could be of concern to the extent that they could affect the probability or consequences of releases associated with unanticipated processes or events, and they conclude (U.S. Nuclear Regulatory Commission 1982b) that ". . . it makes little sense to attempt to limit such consequences by means of a population-related siting criterion, since long-range demographic forecasts are so inherently speculative and unreliable. . . ." However, with regard to the anticipated long-term dissolution of waste and the transport of radionuclides to the environment, the process considered by NRC in its discussion of compliance with EPA's population-based release limits, NRC strangely concludes (U.S. Nuclear Regulatory Commission 1982b) that "population distribution over the long term is immaterial if the geologic repository operates as anticipated." With this we cannot agree. If one is seeking to obtain a limit to the population risk, then population distribution is an essential ingredient, however difficult it may be to predict. Evidently, the NRC is taking instead the narrow view that review and regulation for safety to future populations have been simplified to the exercise of calculating compliance with the EPA release limits at a prespecified distance of 10 km from the waste. This exercise does indeed avoid the problem of population distributions because in deriving the limits EPA has assumed that the fraction of released radionuclides reaching humans will be the same regardless of where the repository is located.

8.6.5. Statistical Analysis of Repository Performance

To support and justify NRC's proposed numerical criteria, their earlier rationale referred only to predictions of repository performance in terms of limiting the radiation dose to future individuals, with no demonstrated relevance to the population-risk objective or release limits proposed by EPA. NRC now quotes recent analyses carried out for them by Pepping et al. (1982). From these new calculations of activity releases over 10,000 years, NRC concludes that for basalt and salt its numerical criterion of a release rate of 10^{-5}/yr or less from the underground facility is necessary and sufficient for basalt and salt repositories, but it concludes that the performance of a repository in tuff could be significantly improved by waste-package release rates tenfold lower than the NRC's numerical criterion.

It is difficult to understand and evaluate the calculated results presented in NRC's rationale because NRC presents only the statistical variations in overall results, for an enormous spread in assumed parameters and with assumed statistical distributions of these parameters. The results demonstrate a statistical distribution of "proximity to the EPA release limits" for assumed statistical inputs. The calculated performance from likely and expected parameters is not identified, and the input data and results are not compared with what can be deduced from the many calculations by others.

We can learn more from examining which nuclides present the greatest problem in achieving EPA's release limits and examining the uncertainties in the parameters that affect these critical species. Adopting the retardation coefficients in Table 7-1 and assuming NRC's numerical criteria, we find that the EPA release limits will be met for all radionuclides within the 10,000-year time limit. For iodine-129, carbon-14, and technetium-99 the release limits are met, within EPA's 10,000-year time limit, by the waste-package release rate, with no requirement for retardation or for a finite water travel time. For all other radionuclides the release limits are met, within the 10,000-year time limit, by the range of retardation coefficients selected in Table 7-1 and by the 1,000-year water travel time. For these radionuclides the waste-package release limit, if achievable, provides redundant protection. Consequently, we conclude that, with these conservatively estimated retardation coefficients, the NRC numerical criteria are sufficient to ensure compliance with the EPA standard. The questions of whether or not they are necessary, reasonable, achievable, and verifiable are dealt with later.

We do not agree with NRC that there is less retardation for uranium in welded tuff than in the media surrounding a salt repository, and because of that we do not reach NRC's finding that welded tuff requires a much lower waste-package release rate than do the other media.

In applying the EPA release limits of Table 8-1 to a salt repository, NRC is assuming that the intrusion of flowing groundwater into a salt repository is "reasonably foreseeable" and is not associated with "fault movements or other disruptive events." With this we cannot agree. We expect groundwater to intrude into basalt, granite, or saturated tuff, and we understand that it is the resulting hydrogeologic transport, as well as other expected events, that is the subject of the Table 8-1 release limits calculated by the EPA staff. If the intrusion of groundwater into natural salt were equally probable, the salt would not be there. EPA's analyses indicate low expectation of water intrusion into natural salt.

8.6.6. NRC's Proposed 1,000-Year Containment by the Waste Package

We further point out in Chapter 9 that our calculations show no benefit of a 1,000-year waste package in achieving our overall safety criterion if the basic dissolution rate constants are assumed to be time independent. This conclusion is confirmed by the calculations of Burkholder (1982) and Chu et al. (1982).

When estimating compliance with EPA's proposed release limits, we do find an advantage of the 1,000-year waste package in that it causes a reduction of 12 percent in the time available for waste dissolution during EPA's 10,000-year time period. Although this feature of the 1,000-year package may aid compliance, it has little to do with ultimate public health and safety.

In Chapter 9 we also estimate the effect of exposing waste forms to hot repository water during the 1,000-year period. It is to avoid the uncertainties of calculations of performance during the first 1,000 years that was the NRC staff's principal argument for the 1,000-year package. Although the calculations are preliminary and are based on incomplete data, we conclude that waste-package performance without a 1,000-year corrosion-resistant overpack can be subjected to technical analysis and experimental verification, and we estimate that exposing waste forms at higher temperature does not necessarily result in unsatisfactory performance. We also note that such analysis and tests will probably be necessary regardless of NRC's 1,000-year containment criterion, because statistical imperfections in manufacturing are very likely to result in containment failure for some waste packages.

8.6.7. The Release Rate Limit for the Waste Package

NRC's Calculation of Effectiveness

The NRC justifies the effectiveness of its 10^{-5}/yr fractional release limit for the waste package by calculating the maximum amount of each radionuclide that could be released from spent fuel waste packages in a full-scale repository and comparing that amount with the EPA release limit at the 10-km accessible environment. The results are shown in Table 8-2.

The NRC's estimate of the carbon-14 inventory is 100-fold lower than that quoted in Chapter 4 of this study. Adopting the 1,000-year inventory of 5.1 x 10^{15} Bq calculated from the data in Chapter 4, and adopting NRC's method of calculation for the maximum possible releases from the waste package over the next 9,000 years, the cumulative release of carbon-14 from the waste package would be within 62 percent of the EPA release limit to the 10-km accessible environment.

For those radionuclides whose ratios in Table 8-2 are greater than unity, geologic transport over a distance of 10 km must either result in sufficient attenuation by radioactive decay or it must delay arrival until after EPA's 10,000-year time limit. Either mechanism can result in compliance with the EPA proposed standard. If we were interested in how much of this radioactivity released from the waste package in 10,000 years could actually reach the accessible environment, we would find that for a 1,000-year water transport time, and for the retardation coefficients adopted for this study (Table 7-1), americium-241, americium-243, plutonium-239, and plutonium-240 would be easily attenuated by decay. Plutonium-242 would be marginally attenuated, and neptunium-237 would be essentially unattenuated and would exceed the EPA proposed limit when released to the 10-km accessible environment.

The NRC calculation on radium-226 is misleading. Because its half-life is only 1,602 years, essentially none of the radium-226 leaving the waste package could ever reach the 10-km accessible environment unless it were assumed not to be retarded by sorption. Instead, radium-226 appears at considerable distances from the waste package because of the migration of its parents uranium-234, uranium-238, and thorium-230. The NRC release rate criterion would allow about 5 x 10^{14} Bq of uranium-234 to be released from the waste packages during the 9,000-year release period. If this reaches the accessible environment, it could bring with it about 10^{14} Bq of radium-226, exceeding the EPA limit in Table 8-1 by a factor of about 5,000.

Effect of the EPA 10,000-Year Time Limit

However, the EPA release limits at the 10-km accessible environment will actually not be exceeded by neptunium-237 and radium-226 because of the more limiting--although nonphysical--barrier built into the EPA standard--the 10,000-year time limit. For neptunium-237 and uranium to reach the 10-km accessible environment within NRC's minimum water

TABLE 8-2 Effect of Applying NRC's 10^{-5}/yr Waste-Package Release Limit

Radionuclide	Repository Inventory[a] (Bq)	Ratio of NRC Allowable Waste-Package Release[b] to EPA Accessible Environment Release[c]
^{14}C	5.0×10^{13} [d]	6×10^{-2} [d]
^{90}Sr	5.6×10^{9}	0
^{99}Tc	5.2×10^{16}	1.3×10^{-1}
^{126}Sn	2.1×10^{15}	6.2×10^{-1}
^{129}I	1.4×10^{14}	4×10^{-3}
^{135}Cs	8.2×10^{14}	1×10^{-2}
^{137}Cs	3.7×10^{10}	0
^{226}Ra	1.0×10^{13}	9×10^{-2}
^{237}Np	3.7×10^{15}	4.5
^{239}Pu	1.2×10^{18}	2.9×10^{2}
^{240}Pu	1.6×10^{18}	4×10^{2}
^{242}Pu	6.3×10^{15}	1.5
^{241}Am	3.4×10^{18}	3×10^{3}
^{243}Am	5.8×10^{16}	3.5×10^{2}
Total	6.3×10^{18}	

[a] Unreprocessed spent fuel, 10^5 mg of uranium, 1,000 years after discharge, as calculated by U.S. Nuclear Regulatory Commission (1982b).

[b] Released during 9,000 years at 10^{-5}/yr of the 1,000-year inventory.

[c] Derived by EPA for 1,000 health effects in 10,000 years (Table 8-1), specified at 10 km from disturbed zone.

[d] U.S. Nuclear Regulatory Commission (1982b). Estimate for ^{14}C is too low by a factor of 100. (cf. Chapter 4 and Table 9-3.)

transport time of 1,000 years, the retardation coefficients for these nuclides must be less than 9, and even then only the differential amount released from the waste package at the beginning of the first year of dissolution will cross the boundary. Even though such low retardation coefficients can be inferred as possible (cf. Table 7-1), they seem unlikely.

The important point here is that EPA's 10,000-year time limit, evidently adopted in NRC's rationale, makes compliance rather easy. This we do not support because, as we have discussed in Section 8.5.8, we see no valid justification for this time limit. This illustrates how the 10,000-year time limit can markedly affect the technical interpretation of performance and compliance. The EPA release limits and the NRC numerical criteria reduce compliance to a simple calculational contest of what, if anything, can keep the more important radionuclides from reaching the 10-km location within the 10,000-year deadline. Designs that may succeed in 10,000-year compliance are not necessarily the same as designs that promote the greatest amount of waste isolation. The NRC-EPA calculational approach may seem to simplify licensing, but we do not understand how such an exercise can support the finding, required in licensing, that there be no unreasonable risk to the health and safety of the public.

Thus, we agree that the NRC numerical criteria are sufficient to achieve the EPA release limits within the EPA time limit. However, we do not find that these numerical criteria are sufficient to obtain, by themselves, a reasonable degree of public health and safety. Assuming that the 10-km location, with its 1,000-year water travel time, occurs underground, and assuming water withdrawal for human use, our calculations in Chapter 9 show that the expected individual dose rates may be of the order of 1 Sv/yr, a thousand times greater than background and 10^4 times greater than the individual-dose criterion adopted for the present study. Similar individual doses in groundwater can be inferred from the earlier calculations of Cloninger et al. (1980, Cloninger and Cole 1981) that were referred to in NRC's rationale for its earlier draft regulation. Yet consideration of individual dose rates is not required in the EPA's proposed draft standard nor in NRC's proposed rule.

Although EPA's 10,000-year time limit might seem to rule out such individual dose rates, in the same way that it has ruled out appreciable cumulative releases as discussed above, even the carbon-14 arriving within EPA's time limit can give rise to individual dose rates in the range of 1 Sv/yr.

EPA argues that its application of release limits at the 10-km accessible environment protects aquifers that might become significant sources of water in the future, regardless of whether or not they are now being used as water supplies. With that objective, our calculations of individual dose rates indicate that the EPA and NRC numerical limits and criteria are not sufficient to achieve this objective of protecting aquifers because, as shown in Chapter 9, the groundwater at the 10-km location will likely be too contaminated for human use. In our evaluation of predicted repository performance, as discussed in Chapter 9, we do not adopt EPA's view that all potentially productive underground aquifers 10 km from emplaced waste need such protection. Rather, we suggest that the need for protecting underground aquifers near a repository should depend upon whether or not future human use of these aquifers is likely, determined in part by the regional characteristics of the site.

Inflexibility of NRC's Release Criterion

In Chapter 9 we consider the effect of various fractional release rates on the predicted individual dose rate for various suggested and conceptual repository sites. We find that a fractional release rate of 10^{-5}/yr is adequate for some radionuclides and not for others. This is also demonstrated by NRC's calculation in Table 8-2. Further, in Chapter 5 we review dissolution rates inferred from laboratory data and predicted for the conditions in a geologic repository, and we find that for a given waste form quite different dissolution rates are expected for different radioelements. Therefore, it is both unnecessary and unrealistic for the NRC staff to prescribe a single fractional dissolution rate that must apply to essentially all of the 1,000-year inventory of radionuclides.

NRC's across-the-board specification of a release rate limit of 10^{-5}/yr for all radionuclides, above the 0.1 percent minimum, is not only unnecessary but it is also likely impossible to achieve. For example, NRC's data used to calculate the release ratios in Table 8-2 show a 1,000-year inventory of radium-226 of 10^{13} Bq for unreprocessed spent fuel. As shown later, radium-226 will likely not be excluded by NRC's 0.1 percent cutoff. The allowable release rate of radium-226 from the waste package at 1,000 years, and at all times thereafter, would be 10^8 Bq/yr. However, as a result of decay of its precursors, there will be about 4.5×10^{14} Bq of radium-226 within the waste package at 10,000 years. The maximum allowable fractional release rate of radium-226 at this time would then be 2.2×10^{-7}/yr. Radium is not solubility limited, and data in Chapter 5 indicate that such a low release rate for radium may not be obtainable by any of the technologies quoted by NRC.

Further, in Chapter 9 we point out that the expected low solubility and large inventory of uranium can result in an extremely low fractional dissolution rate for uranium, but that the presence of so much uranium precipitate in the waste package can result in a relatively large fractional dissolution rate of its second decay daughter, radium-226, which is not expected to be solubility limited.

Yet, as has been pointed out earlier, release of radium-226 directly from the waste package contributes nothing to the releases of radium-226 to the environment. Because of the 1,602-year half-life and decay of radium-226, it is only the radium that is created during the transport of released uranium and thorium that can appear at any appreciable distance from the waste. Therefore, NRC's technical criterion for waste-package release rate is, in the case of radium-226, unnecessary and probably not achievable. This is also true for other radionuclides.

NRC's 0.1 Percent Threshold

The NRC proposes that its 10^{-5}/yr release rate criterion not apply to any radionuclide that is released at a rate less than 0.1 percent of the calculated total annual release at 1,000 years following permanent closure of the repository. From the assumption of congruent release of all radionuclides at the maximum fractional rate of 10^{-5}/yr at 1,000 years, resulting in the NRC estimates in Table 8-2, any radionuclide whose 1,000-year inventory is less than 0.1 percent of the total 1,000-year inventory would seem to be excluded. On this basis, the only nuclides of further concern to NRC would be technetium-99, plutonium-239, 240, and 242; and americium-241 and 243. Some DOE contractors have evidently adopted this interpretation.

By this rationale, NRC requires that the waste package perform as specified on the high-activity radionuclides. However, high activity is not the proper criterion of importance in terms of release from the waste package. As we show in our performance calculations in Chapter 9, and as has been shown in the many calculations by others who have considered the issues of repository performance with regard to specific radionuclides, plutonium-239, plutonium-240, americium-241, and americium-243 are readily attenuated by transport delays and decay and are not important contributors to radiation dose. NRC will have excluded from consideration the radionuclides that are the important contributors. For their assumption of congruent dissolution and 1,000-year water travel time, the radionuclides that present greater danger to people than those of concern in NRC's waste-package release specification are calculated in Chapter 9 to be neptunium-237, carbon-14, selenium-79, lead-210, and, depending on the degree of dispersion during groundwater transport, cesium-135. The NRC waste-package criteria can result in focusing on the wrong species.

Noting that some DOE contractors are interpreting the NRC staff's 0.1 percent exclusion as described above, a different interpretation is more reasonable. Although NRC seems to assume congruent dissolution in its rationale, its rule does not require that assumption. As discussed in Chapter 5, congruent dissolution is neither observed nor predicted. Our best estimates of the 1,000-year fractional dissolution rates of various radionuclides are given in Tables 5-6 and 9-4 for reprocessing waste. These fractional release rates for plutonium will be even lower for the unreprocessed spent fuel assumed in the NRC calculation of Table 8-2 because of the higher plutonium inventory. Applying the fractional release rates of Table 5-6, we estimate a total first-year release at 1,000 years of about 10^{12} Bq, about 100-fold less than the NRC estimate. Now if the small-inventory radionuclides in Table 8-2 are released at NRC's 10^{-5}/yr limit, their releases will exceed the 0.1 percent limit specified by NRC. This is why radium-226 is likely to fall above the 0.1 percent limit, creating the unnecessary technological challenge of suppressing radium release that has been discussed above.

From the above interpretation, cesium-135 must still be considered in determining compliance with NRC's proposed release limit. As discussed in Chapter 5, we have seen no data that imply that the possible waste forms and waste packages suggested by NRC could achieve a fractional release rate as low as 10^{-5}/yr for cesium. Either NRC is adopting the simpler interpretation, that would exclude the more important radionuclides in determining compliance with its waste-package performance requirement, or it has misinterpreted existing data when it concludes that the required performance can probably be achieved and verified.

We have seen no data or analyses of the release of carbon-14 from unreprocessed spent fuel. Carbon-14 is not likely to be solubility limited, and any inference that its release rate from spent fuel waste can meet the NRC criterion is speculative.

If some specification of fractional dissolution rates were necessary or useful at this time, then the specification should be based on necessary performance and should vary from one waste constituent to another. However, such specification is neither necessary nor useful. Instead, we recommend that a clear and meaningful overall performance standard be adopted and that the necessary performance of each component of the waste isolation to meet that standard be calculated, using the best available data on expected performance and realistic data on the repository site being considered. That is not the approach that has led to NRC's proposed numerical criteria.

8.6.8. Individual Dose Rates Allowed by the NRC Numerical Criteria

We note that NRC's rationale for its numerical criteria deals only with their effect in achieving the EPA 10,000-year release limits. Whereas NRC concludes that in many cases these numerical criteria will yield releases within the EPA limits, NRC has failed to comment on the individual doses that could result from some of these parameters that it has adopted. From our calculations in Chapter 9 of individual doses, we can easily interpolate and find that NRC's numerical criteria can result in individual doses during the 10,000-year period that exceed any expected individual dose rates now allowed for NRC-licensed facilities. In departing from the individual-dose analyses referenced in its 1981 justification for its technical criteria, NRC is neglecting an important aspect of the protection of public health and safety.

Although the NRC proposed final rule identifies EPA's proposed standard as the standard that the regulation is to implement, the NRC technical criteria are not confined to the time period of the EPA release limits. NRC has not stated that its proposed 10^{-5}/yr release rate for all significant radionuclides is to apply only during the period from 1,000 to 10,000 years. Taking the proposed rule literally, the release rate would seem to apply to all times after 1,000 years. The longer the time period that the release rate criterion must apply, the more difficult it will be to achieve, in part because of the continued growth of decay daughters that are not present initially in the waste. However, even though there are increasing difficulties in predicting and achieving performance at later times, our calculated long-term performance results presented in Chapter 9 show that releases from the waste package and from the repository after 10,000 years must be considered.

8.6.9. Verification of Waste-Package Performance

Verification of the expected release rate performance, whether at 1,000 to 10,000 years after emplacement, or whether at millions of years later, will be difficult. The predicted performance depends much on the local chemistry of the groundwater and on the long-term failure modes of several of the components of the waste package, and it requires a validated theory of long-term release rate that does not now exist. Until the uncertainty of prediction and verification of waste-package performance is better understood, specification of a fractional release rate as a numerical criterion to be complied with has little meaning.

Prediction of waste-package performance is likely to be more difficult than reflected in NRC's rationale. For reasons discussed in Chapter 5, we do not agree with the present extrapolations of laboratory dissolution data quoted by NRC. We also note some apparent confusion by NRC with regard to the effect of backfill material on the release rate. In technical support of its belief that its 10^{-5}/yr release rate is achievable, NRC quotes estimated time delays for radionuclides to penetrate the backfill material that might surround the waste. One quoted study calculates dissolution rates averaged over several thousand years, including the time during which no radionuclides have penetrated the backfill. If NRC intends that its release rate criterion be anything other than a performance standard that must be met at every instant, it should so state. To assess the ability of a backfill material to help attain NRC's required release rate, the rate of release of long-lived radionuclides from the waste package during the eventual steady state diffusion through the backfill should be considered, i.e., during the time when backfill sorption has little effect on the release rate.

We do not consider the NRC staff's review of the waste-form technology and its expected and possible performance in a repository to be adequate to support their proposed release rate criterion. The NRC evaluation is not based on a predictive technique with a clear or valid technical basis. To verify that waste packages comply with such performance criteria in the long-term future, a proven and reliable predictive technique must be available. Such verification has not been addressed in the proposed rule, and the predictive techniques used by NRC do not seem capable of verification.

8.6.10. Summary

The NRC's numerical criteria for 1,000-year containment and an across-the-board fractional release rate of 10^{-5}/yr from the waste package after 1,000 years are of questionable importance to long-term safety and are proposed without a technically valid basis and with invalid assumptions of existing technology. If such numerical criteria were adopted, compliance could probably not be verified. It would be more appropriate for NRC to state the considerations that may help guide DOE in its development and proof of the waste package as one of the possible barriers that may aid in meeting a reasonable overall safety criterion. One of the important considerations is for DOE to continue work on developing a means of predicting the long-term performance of waste packages.

NRC has not yet addressed the question of how either its numerical criteria or the EPA release limits can "support a finding of no unreasonable risk to the health and safety of the public," the objective stated in NRC's first release of the proposed rule.

Because of the foregoing, we do not adopt the NRC technical criteria as indicators of satisfactory performance of the waste-isolation technology. Instead, we adopt the overall performance criterion in terms of average annual radiation dose to an individual, and in Chapter 9 we evaluate the expected and possible performance of the waste-isolation system in terms of meeting this overall criterion.

8.7. IS GEOLOGIC ISOLATION INCOMPATIBLE WITH AN INDIVIDUAL-DOSE CRITERION?

It has been suggested that adopting an individual-dose criterion for geologic waste isolation can lead to approaches for protecting future individuals from released radioactivity that may be incompatible with achieving waste isolation (M. J. Bell, Nuclear Regulatory Commission, personal communication to T. H. Pigford, 1982). As an example, the magnitude of individual dose from ingestion depends on the concentration of radionuclides in water, so one way of reducing individual doses is to dilute released radionuclides with large quantities of water. By contrast, a stated goal of geologic isolation is to prevent radionuclides from reaching the environment, so that techniques of dilution are unnecessary. The problem occurs because there is no complete isolation of all radionuclides in geologic systems. Geologic repositories in natural salt come the closest because there is no flowing groundwater in salt to carry radionuclides to the environment. Isolation in natural salt would seem to be complete but for the possible intrusion into the salt repository by people or by a major natural diversion of flowing water, however improbable it may be. Therefore, we must deal with the reality of incomplete isolation, and the public must be protected from those radionuclides that eventually do reach the environment.

We have already pointed out in Section 8.4 that although limiting the total releases of radioactivity to the environment, as in the EPA and NRC proposals, might seem the most direct and practical approach toward achieving isolation, the proposed standard with its 10,000-year release limits does little toward limiting the releases that will later occur. Further, our calculations summarized in Chapter 9 show that even though the EPA 10,000-year release limits are achieved by the NRC numerical criteria, the radionuclides released to the groundwater that EPA and NRC intend to protect are likely to reach concentrations far greater than would be allowed in potable water. We show that radionuclides released to surface waters can be easily diluted by rapidly flowing rivers to concentrations far below the levels that would result even in the low individual-dose criterion of 10^{-4} Sv/yr adopted for this study. We also show that for surface water flowing at a relatively low rate, the released radionuclides may not be diluted enough to result in sufficiently low radiation doses to individuals. Although these individual doses are ignored in the proposals by EPA and NRC, protecting the health and safety of future individuals is not to be ignored.

What this means is that geologic isolation is a worthwhile objective, but it should not become a slogan that obscures the fact that even though most of the radionuclides will be successfully isolated long enough to disappear by decay, some will be released. Protection of individuals and populations from those released radionuclides is a realistic, legitimate, and nontrivial issue. If hazards to individuals from released radionuclides can be reduced by taking advantage of dilution, as by rapidly flowing rivers, then repository sites that need and have that capability for dilution should be given full credit.

8.8. REFERENCES

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9

PERFORMANCE ANALYSIS OF THE GEOLOGIC WASTE-DISPOSAL SYSTEM

9.1. INTRODUCTION

This chapter presents the results of analyses of the long-term performance of the geologic waste-disposal system. Here we draw upon data and evaluations in Chapters 4 through 8 concerning the performance of individual components and barriers of the waste-disposal system. We present predictions of the long-term releases of radionuclides to the environment and the resulting radiation doses to humans, calculated for the purpose of this study by Pigford et al. (1982a), using data supplied by the panel. These predicted radiation doses are compared to the performance criterion that has been discussed in Chapter 8, and we use these calculations to identify the performance features and limitations of each of the isolation mechanisms in the geologic system.

Emphasis is placed here on (1) the expected long-term performance of the conceptual geologic repositories in different rock media and (2) the effect of uncertainties on that long-term performance. Estimates of expected performance should be based on the most probable values of the repository parameters, and it is these most probable values that have been sought for this study. This is a more meaningful approach than that of adopting only "most conservative" parameters in the predictive calculations, with no knowledge of the degree of conservatism of the final results. However, even though our purpose is to present predictions of likely performance of repositories, the considerable uncertainties in input data, especially in the sorption properties, have led to a selection of properties that may, in many instances, be on the conservative side. Therefore, the present predictions of future radiation doses should be viewed as the panel's "best estimates," within the time and scope limitations of this study.

In this chapter we first review, in Section 9.2, the measures of overall performance that are to be calculated. We review in Section 9.3 the techniques of calculating radiation dose, in Section 9.4 the selection of hydrologic and other data for calculations; and in Section 9.5 the method of calculating space-dependent/time-dependent radionuclide concentrations and radiation doses due to hydrogeologic transport.

In Section 9.6 we present a typical calculation of the time-dependent concentration of selected radionuclides reaching an assumed 10-km distance from the waste. This illustrates how the maximum individual doses are obtained for each assumed location, or for each selected water travel time from the waste.

In Section 9.7 we present and discuss the maximum individual doses in groundwater and surface water, as a function of water travel time, for repositories in basalt, granite, salt, and tuff, in the absence of dispersion. The effect of a time delay before dissolution is discussed in Section 9.8, and the effect of dispersion on the maximum doses is shown in Section 9.9.

In Section 9.10 we show the effect of the solubility-limited dissolution rates, calculated as discussed in Chapter 5, on the individual doses for the various repository rocks. These calculations represent our best estimate of repository performance as a result of waste dissolution and hydrogeologic transport, calculated for repositories loaded with reprocessing waste. The importance of the cesium dissolution rate and the design of waste packages for separated carbon-14 and iodine-129 are discussed. Similar calculations for a basalt repository loaded with unreprocessed spent fuel are presented in Section 9.12.

The effect of waste dissolution during the period of repository heating is considered in Section 9.13.

The ensuing Sections 9.14 through 9.22 deal with the effect of surface storage before emplacement, the effects of different waste compositions from other reactor fuel cycles, the effects of uncertainties in solubilities and retardation coefficients, and the effect on radionuclide transport in fractured media due to pore diffusion.

9.2. MEASURES OF OVERALL PERFORMANCE

The fundamental measure of overall performance of a geologic repository in isolating radionuclides is the radiation dose to humans that could result from the release of radionuclides to the environment, expressed in sieverts (Sv).

In this study we estimate the lifetime-average radiation dose rates to the individual resulting from waste dissolution and hydrogeologic transport to the biosphere at any future time after the geologic repository is loaded with waste and is sealed. The resulting individual dose rates calculated for a repository in wet rock, i.e., a rock normally containing some moving groundwater, are then compared with the individual dose rate criterion of 10^{-4} Sv/yr adopted for this study (cf. Chapter 8). This dose rate criterion is about 10 percent of the average annual dose to an individual from background radiation at sea level.

We also estimate the radiation dose rates that could result from unexpected or less probable events, such as water intrusion into a repository constructed in salt, followed by waste dissolution and groundwater transport of radionuclides to the biosphere. We also estimate, for all the different rock media, the radiation doses that could result from human use of contaminated underground water before it has reached the surface. These events are either improbable or expected, depending on the site characteristics. The predicted radiation doses therefrom are to be weighted by their probabilities before comparing them with the performance criterion of 10^{-4} Sv/yr adopted in this study for expected events. There is not sufficient information on probabilities of these "unexpected" events for probabilistic weighting to be included in the present study.

9.3. CALCULATION OF RADIATION DOSE

Emphasis is placed here on radiation doses from human ingestion of radionuclides that have become dissolved in groundwater and are transported to the biosphere. The eventual release of some amount of radionuclides to the biosphere, however small, is considered likely to happen over a long enough time scale for repositories constructed in rock that normally contains groundwater. For most radionuclides the radiation doses from external radiation exposure and from inhalation are of lesser consequence.

Because the long-term release of radionuclides varies considerably over long periods of time, the radiation doses are time dependent. However, within an individual lifetime the rate of release of radionuclides and rate of exposure to an individual will be essentially constant, so the dose to an individual is calculated on the basis of a lifetime radiation dose, i.e., "dose commitment," from radionuclides ingested continuously at constant rate over that lifetime.

When contaminated groundwater reaches a surface aquifer, humans can ingest the radionuclides by drinking the contaminated water and by eating food grown by water containing the radionuclides. In these analyses we assume that an individual obtains his total lifetime intake of potable water from water contaminated with the released radionuclides and that his lifetime consumption of food is obtained from food crops nurtured by the contaminated water. After estimating the time-dependent concentrations of radionuclides in surface water, we obtain the lifetime (70-year) dose to the maximally exposed individual by using the pathway and intake calculations of Napier et al. (1980), whose radiation dose factors are derived from the 1959 report of the International Commission on Radiological Protection. We then adjust the calculated doses for some of the critical radionuclides by using new techniques and metabolic data for calculating dose presented in a recent report of the International Commission on Radiological Protection (1980), using correction factors calculated by Runkle and Soldat (1982) that reduce the calculated dose from radium-226 by a factor of 90 and increase the calculated dose from lead-210 and neptunium-237 by factors of 4.4 and 200, respectively. ICRP-30 (International Commission on Radiological Protection 1979, 1980) corrections on other radionuclides, of lesser importance in the dose calculations, are not included in this study. The calculated 70-year doses to the individual are then divided by 70 to obtain an average yearly dose to the individual.

The dose conversion factors derived for this study are listed in Table 9-1. A breakdown of the doses from different pathways for

TABLE 9-1 Average Lifetime Dose Rate per Unit Concentration of Radionuclides in Water

Radionuclide	Half-Life (yr)	Average Dose Rate per Unit Concentration ($Sv\text{-}m^3/Bq\text{-}yr$)	Ratio of Drinking Water Dose to Total Dose
^{14}C	5.73×10^{3}	9.21×10^{-7}	1.22×10^{-4}
^{79}Se	6.5×10^{4}	1.60×10^{-7}	4.27×10^{-4}
^{93}Zr	1.5×10^{6}	4.83×10^{-13}	3.0×10^{-1}
^{99}Tc	2.12×10^{5}	7.03×10^{-10}	1.0×10^{-2}
^{126}Sn	1×10^{5}	2.77×10^{-8}	1.2×10^{-2}
^{129}I	1.7×10^{7}	2.04×10^{-8}	7.07×10^{-2}
^{135}Cs	3×10^{6}	5.26×10^{-8}	2.67×10^{-2}
^{210}Pb	2.1×10^{1}	7.68×10^{-6} [a]	5.08×10^{-2}
^{225}Ra	4.05×10^{-2}	1.40×10^{-6}	1.16×10^{-1}
^{226}Ra	1.60×10^{3}	2.40×10^{-6} [b]	1.07×10^{-1}
^{229}Th	7.34×10^{3}	5.56×10^{-7}	6.84×10^{-2}
^{230}Th	8×10^{4}	8.03×10^{-8}	6.91×10^{-2}
^{233}U	1.62×10^{5}	3.80×10^{-8}	1.92×10^{-1}
^{234}U	2.47×10^{5}	3.80×10^{-8}	1.88×10^{-1}
^{238}U	4.51×10^{9}	2.91×10^{-8}	2.15×10^{-1}
^{237}Np	2.14×10^{6}	1.29×10^{-5} [c]	8.41×10^{-2}
^{239}Pu	2.44×10^{4}	9.80×10^{-9}	1.94×10^{-1}
^{240}Pu	6.58×10^{3}	9.80×10^{-9}	1.94×10^{-1}
^{242}Pu	3.79×10^{5}	9.50×10^{-9}	1.85×10^{-1}
^{241}Am	4.58×10^{2}	1.26×10^{-7}	4.3×10^{-2}
^{243}Am	7.95×10^{3}	1.23×10^{-7}	4.29×10^{-2}

[a] Increased by a factor of 4.4 above data of B. A. Napier (Battelle Pacific Northwest Laboratory, personal communication to T. H. Pigford, January 22, 1982) to allow for ICRP-30 corrections (Runkle and Soldat, 1982).

[b] Reduced by factor of 90 below data of B. A. Napier (Battelle Pacific Northwest Laboratory, personal communication to T. H. Pigford, 1982) to allow for ICRP-30 corrections (Runkle and Soldat, 1982).

[c] Increased by a factor of 200 above data of B. A. Napier (Battelle Pacific Northwest Laboratory, personal communication to T. H. Pigford, 1982) to allow for ICRP-30 corrections (Runkle and Soldat, 1982).

SOURCE: Derived from data of B. A. Napier (Battelle Pacific Northwest Laboratory, personal communication to T. H. Pigford, January 22, 1982) for 70-year lifetime dose commitment, 70-year intake of water and of food.

carbon-14, radium-226, and neptunium-237 is shown in Table 9-2. Although we have not evaluated the validity of the data that enter the pathway calculations, we have compared the dose conversion factors derived as described above with similar conversion factors derived by Zach and Iverson (1979) and Zach and Mayoh (1980) for the Canadian program on nuclear fuel waste management (Wuschke et al. 1981). The comparison, given in Table B-1 of Appendix B, shows that for similar assumptions there is good agreement between the dose conversion factors from these two studies.

To concentrate on the uncertainties in the dose calculations, B. Mann (consultant to the panel, personal communication, 1982) has compared data used by different agencies and groups on the calculated whole-body dose equivalent per unit amount of activity ingested, for those radionuclides important in geologic disposal. This compilation, which excludes the pathway parameters, appears in Appendix C. The data indicate orders of magnitude variation from one source to another in the calculated dose for some nuclides. It is apparent that the dose-intake calculation can introduce considerable uncertainty in the dose

TABLE 9-2 Relative Pathway Contributions to Individual Ingestion Dose from Contaminated Surface Water

	Average Yearly Dose (Sv/yr for unit: Bq/m^3 in water)		
Pathway	^{14}C	^{226}Ra [a]	^{237}Np [b]
Terrestrial food pathways			
Leafy vegetables	1.76×10^{-8}	1.79×10^{-7}	7.60×10^{-7}
Root vegetables	1.45×10^{-7}	9.43×10^{-8}	4.98×10^{-7}
Other above-ground vegetables	2.63×10^{-8}	6.50×10^{-8}	2.92×10^{-7}
Orchard fruit	9.21×10^{-8}	8.78×10^{-8}	4.38×10^{-7}
Grain	3.66×10^{-7}	1.79×10^{-7}	7.90×10^{-7}
Eggs	3.36×10^{-8}	2.11×10^{-13}	9.06×10^{-11}
Milk	9.21×10^{-8}	2.11×10^{-8}	1.14×10^{-9}
Beef	7.75×10^{-8}	5.52×10^{-9}	1.26×10^{-7}
Pork	1.76×10^{-8}	4.88×10^{-10}	2.20×10^{-8}
Poultry	3.22×10^{-8}	7.16×10^{-12}	1.29×10^{-10}
Total	9.00×10^{-7}	6.22×10^{-7}	2.90×10^{-6}
Aquatic pathways			
Fish	7.02×10^{-9}	2.43×10^{-7}	2.14×10^{-7}
Mollusks	1.40×10^{-8}	1.25×10^{-6}	8.58×10^{-6}
Drinking water	1.13×10^{-10}	2.56×10^{-7}	1.08×10^{-6}
Total[c]	2.19×10^{-8}	1.79×10^{-6}	9.94×10^{-6}
Total, aquatic plus terrestrial	9.21×10^{-7}	2.40×10^{-6}	1.29×10^{-5}

[a] Reduced by factor of 90 below data of B. A. Napier (Battelle Pacific Northwest Laboratory, personal communication to T. H. Pigford, 1982) to allow for ICRP-30 corrections (Runkle and Soldat, 1982).

[b] Increased by factor of 200 above data of B. A. Napier (Battelle Pacific Northwest Laboratory, personal communication to T. H. Pigford, 1982) to allow for ICRP-30 corrections (Runkle and Soldat, 1982).

[c] Aquatic pathways total includes contribution from external exposure due to swimming, boating, and shoreline occupancy. For ^{14}C and ^{226}Ra this is less than 0.3 percent of the aquatic total. For ^{237}Np it contributes 6.72×10^{-6} Sv/yr, or about 68 percent to the aquatic total.

SOURCE: Data of B. A. Napier (Battelle Pacific Northwest Laboratory, personal communication to T. H. Pigford, 1982) for 70-year lifetime dose commitment, 70-year intake.

predictions for a geologic repository. Further uncertainties can be expected for the pathway calculations. We find very little in the way of such comparative evaluations of dose factors or of environmental transport calculations in the project literature. The federal agencies should initiate a comprehensive evaluation of the data and methods used in these pathway and dose-response calculations.

For expected events, such as long-term waste dissolution and groundwater transport in wet-rock repositories, the calculated dose rate to the individual is compared to the individual dose criterion of 10^{-4} Sv/yr adopted for this study (cf. Chapter 8).

In estimating the dose rates to future individuals using the contaminated groundwater from a repository, we have adopted, for simplicity, the dose conversion factors of Table 9-1. If the flow of contaminated water in the underground aquifer is small, it may be unrealistic to assume that this groundwater is used not only for drinking but also to grow crops, and the resulting estimated radiation doses may be greater than could actually occur. The data in Table 9-2 show that drinking water contributes 0.5 percent to the total dose for carbon-14, 14 percent for radium-226, and 11 percent for neptunium-237. The fraction of tne total dose contributed by drinking water, based on Napier's calculations, is listed in Table 9-1 for each of the radionuclides. These fractions should be applied to the calculated doses from contaminated groundwater, presented in later sections of this chapter, if it is concluded that drinking is the only likely future human use of the contaminated groundwater.

9.4. HYDROLOGIC, GEOCHEMICAL, AND FUEL CYCLE DATA ADOPTED FOR PERFORMANCE CALCULATIONS

For this performance analysis we have adopted the conceptual repository designs in salt, granite, basalt, and tuff, each designed to contain high-level radioactive wastes, transuranic wastes, and other special long-lived wastes from the irradiation of 10^5 Mg of uranium fuel in light-water reactors. Our reference case is based on waste that would result from reprocessing fuel 160 days after discharge from the reactor and storing the resulting waste for 10 years prior to emplacement in the repository. We also consider the effects of the following changes in fuel cycle operations:

- Emplace unreprocessed fuel in the repository after storing it for 10 years after reactor discharge.
- Store the discharged fuel for realistically longer periods prior to reprocessing.
- Store the reprocessing waste for longer periods prior to emplacement in the repository.
- Reprocess fuel from light-water and fast-breeder reactors that have been loaded with uranium and recycled plutonium.
- Reprocess uranium-thorium fuel.

Radionuclide inventories for the principal radionuclides in the radioactive waste for the reference case are derived from the data in Table 9-3. It is assumed that the repository contains the high-level and transuranic waste from reprocessing 10^5 MgHM, i.e., 10^5 Mg of uranium in fuel before irradiation, in fuel discharged from pressurized-water reactors. Reprocessing is assumed to occur 160 days after reactor discharge, and the wastes are assumed to be emplaced 10 years after discharge. It is assumed that 0.5 percent of the uranium and plutonium in the discharge fuel appears in the reprocessed high-level waste and 0.5 percent appears in the transuranic wastes. It is assumed that all radioiodine, krypton-85, carbon-14, and tritium in discharge fuel are recovered during reprocessing and appear in waste packages in the repository.

Hydrologic properties adopted for the calculations are the flow rates of surface water and of potentially contaminated groundwater shown in Table 9-4. The values of solubilities used in these calculations are the "most probable values" given in Table 7-1, and the values of retardation coefficients are the "suitably conservative values" prepared for this study by K. B. Krauskopf (Stanford University, personal communication, 1982) and listed in Table 7-1.

TABLE 9-3 Radionuclide Inventories in Wastes from Fuel Reprocessing

Radionuclide	Half-Life (yr)	Inventory (Bq)
^{14}C	5.73×10^{3}	5.73×10^{15}
^{79}Se	$\leqslant 6.5 \times 10^{4}$	1.46×10^{15}
^{90}Sr	2.77×10^{1}	2.68×10^{20}
^{93}Zr	1.5×10^{6}	6.68×10^{15}
^{99}Tc	2.12×10^{5}	4.84×10^{16}
^{126}Sn	1×10^{5}	2.88×10^{15}
^{129}I	1.7×10^{7}	1.16×10^{14}
^{135}Cs	3×10^{6}	1.28×10^{15}
^{234}U	2.47×10^{5}	4.16×10^{13}
^{238}U	4.51×10^{9}	1.16×10^{13}
^{237}Np	2.14×10^{6}	1.16×10^{15}
^{238}Pu	8.6×10^{1}	4.26×10^{16}
^{239}Pu	2.44×10^{4}	1.16×10^{16}
^{240}Pu	6.58×10^{3}	1.95×10^{16}
^{241}Pu	1.32×10^{1}	4.57×10^{18}
^{242}Pu	3.79×10^{5}	6.50×10^{13}
^{241}Am	4.58×10^{2}	7.03×10^{17}
^{243}Am	7.95×10^{3}	6.31×10^{16}
^{242}Cm	4.46×10^{-1}	7.29×10^{19}
^{244}Cm	1.76×10^{1}	5.55×10^{18}
^{245}Cm	9.3×10^{3}	7.68×10^{13}
^{246}Cm	5.5×10^{3}	1.06×10^{14}

NOTE: Data represent total repository waste from 10^5 Mg uranium in light-water reactor fuel. Assumes 0.5 percent of uranium plus plutonium becomes high-level waste and 0.5 percent of uranium plus plutonium becomes transuranic waste. Calculated for 160 days after reactor discharge (cf. Chapter 4).

To obtain sufficient data for the technical systems analysis required in the panel's charter, hydrologic data for specific sites were used, when available. When site-specific data were not available, data for conceptual generic sites were adopted.

The volumetric flow rate of contaminated groundwater is the sum of the estimated volumetric flow rates of contaminated water leaving the repository proper, later diluted by interflows from aquifers in the surrounding media. This effective flow rate of contaminated groundwater is expected to increase with distance from the repository, corresponding to increasing dilution of dissolved radionuclides. However, no such distance-dependent data on the flow rates of potentially contaminated groundwater are available from the U.S. Department of Energy (DOE) and the U.S. Geological Survey (USGS) sources for the generic or proposed repository sites, so a constant value of this flow rate is adopted for each repository for this study. A constant flow rate of contaminated groundwater for each repository was adopted in the generic repository studies by Cloninger et al. (1980), Cloninger and Cole (1981), and Burkholder (1982).

Because no groundwater exists within natural salt, the contaminated flow from a salt repository is assumed to result from a major diversion of an aquifer in surrounding nonsalt strata, which then dissolves and flows through the salt, dissolves waste, and joins an aquifer that then flows eventually to the surface. The contaminated water thus reaching

TABLE 9-4 Hydrologic Parameters for Reference Repository Sites

	Repository Rock					Tuff[e]	
	Basalt[a]		Bedded[b] Salt	Domal[c] Salt	Granite[d]	Saturated	Unsaturated
Location	Hanford, Wash.		Permian and Paradox basins	Generic	Generic	Nevada Test Site	
Flow rate through repository (m^3/yr)	17		40 to 400				
Flow rate of contaminated aquifer (m^3/yr)	3.2×10^4 (at 10^4 yr)		1.3×10^3	5.2×10^5			
Flow rate of surface water (m^3/yr)	1.1×10^{11}		3.2×10^8	2.8×10^9	3.8×10^9		
Path lengths, repository to biosphere (km)	Rockwell estimates	PNL[f] estimates				6 (to well J13) 25 (to Lathrop Wells)	7 (to well J13) 26 (to Lathrop Wells)
Minimum	60	12	10		1		
Maximum	80	16	10^3		10^2		
Mean	70	14	10^2	10^2	10		
Water travel time to biosphere (yr)						1.2×10^3 (to well J13) 4.3×10^3 (to Lathrop Wells)	2.2×10^4 (to well J13) 2.5×10^4 (to Lathrop Wells)
Minimum	1.3×10^4	1.3×10^4	1×10^4		10^3		
Maximum	1×10^6	1.7×10^4	1×10^6		$>10^6$		
Mean	1.9×10^5	1.5×10^4	1×10^5	4×10^4	10^6		
Average pore velocity							
Emplacement rock (m/yr)	0.0032						0.03
Surrounding media (m/yr)	0.5	0.9	1	2		5.7	5.7
Effective porosity in aquifer	10^{-3} to 5×10^{-2}	1.8×10^{-1}	0.10	10^{-5} to 3×10^{-2}		0.18 (to Lathrop Wells)	

[a] D. J. Brown and R. A. Deju (Rockwell Hanford Operations, personal communications to T. H. Pigford, 1982).

[b] S. Goldsmith (Office of Nuclear Waste Isolation, personal communication to J. O. Neff, 1982), except flow rate of contaminated aquifer is from G. E. Raines (Office of Nuclear Waste Isolation, personal communication to T. H. Pigford, 1982). Data assume a salt repository breached by flowing water.

[c] Bechtel Group (1981). Data assume a salt repository breached by flowing water.

[d] Data from Chapter 7, except flow rate of contaminated aquifer is from Cloninger and Cole (1981).

[e] L. D. Tyler (Sandia National Laboratories, personal communication to T. H. Pigford, 1982). See Figure 7-9 for well locations.

[f] Dove (1982), Dove et al. (1982).

the accessible environment will be too saline to be potable or to be used directly for irrigation. The resulting salinity of the aquifer flow will affect the retardation constants for certain radionuclides, particularly cesium.

Also shown in Table 9-4 are volumetric flow rates of water in rivers that may be typical of flowing surface water into which the contaminated groundwater discharges. Dilution by the river flow further reduces the concentrations and resulting individual radiation doses. Relying here on effective groundwater travel times from the repository to the river, as supplied by the indicated sources of these data, our estimated radiation doses from river water are not affected by uncertainties in the effective volumetric flow rate of contaminated groundwater.

The data in Table 9-4 on water travel time from the repository to the accessible environment are not used as input for the long-term release calculations. Instead we adopt the average water transport time as a variable parameter. From these calculations we can determine the effects of the different estimates of water travel times quoted in Table 9-4.

There are many uncertainties in the present hydrogeologic data, and some quantitative estimates of the effects of these uncertainties on estimated long-term performance are discussed in Section 9.19. Additional uncertainties are introduced when we assume that these hydrogeologic properties will remain constant over the long times for which these calculations are made. The possibility that there can be major changes in these geologic features in the future has not been ignored in our considerations. For example, streams can be diverted by tectonism, and glaciation might reduce their flow. However, in the absence of any estimates or other guidance of the possible directions, magnitudes, and probabilities of such future changes, the panel has based its performance analyses on the present hydrogeologic data and on the present estimates of uncertainties in these data. Such estimates based on present data provide the best available guide to long-term performance and are useful in understanding the extent to which these hydrogeologic features are important. A study of the possible long-term changes in the geologic features that affect repository performance would be a major benefit to the planning and assessment of geologic repositories for radioactive waste.

9.5. METHOD OF CALCULATING RADIONUCLIDE TRANSPORT

The concentration of each radionuclide in a migrating decay chain was calculated as a function of distance from the waste packages and as a function of time since the beginning of waste dissolution using the exact analytical solutions for one-dimensional advective transport in a porous medium with dispersion, derived by Harada et al. (1980). At any given distance from the waste packages, expressed either in a lineal distance or in terms of a groundwater travel time, the time-dependent concentration is then searched to obtain the temporal maximum concentration for that location. It is assumed that radionuclides discharged to a river mix quickly with the river flow at the discharge location. The maximum concentration of each radionuclide is multiplied by the dose conversion factor from Table 9-1 to obtain the maximum dose rate to the individual.

These analytical equations are solved with computer programs used as benchmark tests for an international workshop on methods for predicting radionuclide migration in geologic media (Andersson 1982). Possible corrections due to multidimensional dispersion, two-dimensional flow pathways, and nonequilibrium transport were examined using the calculational techniques of Pigford et al. (1980).

We assume first that waste dissolution begins shortly after emplacement, neglecting time delays for resaturation of repositories below the water table and neglecting protection of the waste from a possible overpack corrosion barrier. For the initial calculations we assume congruent dissolution of the waste and its contained radionuclides such that the dissolution rate is specified by a rate constant, the fractional rate of dissolution, which is here assumed to be the same for all radionuclides. We will later show the effects of dissolution rate constants calculated for solubility-limited dissolution, as discussed in Chapter 5. It is the later graphs (Figures 9-9 through 9-12), calculated on the basis of solubility-limited dissolution and dispersion, that represent our best estimate of the performance of the geologic repositories considered herein.

Calculations of dose rates based on the assumption of congruent dissolution are presented here as reference, and for comparison with predictions from the many other published calculations that have assumed congruent dissolution (Cloninger et al. 1980, Cloninger and Cole 1981, Burkholder 1982). After reviewing the results for congruent dissolution, the importance of later corrections for solubility-limited dissolution will be more readily apparent.

9.6. TIME-DEPENDENT RADIATION DOSES FROM GROUNDWATER TRANSPORT

Calculated radiation dose to the individual, from different radionuclides as a function of time after emplacement, is shown in Figure 9-1 for a hypothetical basalt repository loaded with reprocessing waste, with an arbitrarily assumed water travel time from waste to the environment of 1,000 years, corresponding to a water travel distance of 1 km at an assumed pore velocity of 1 m/yr. Dissolution of waste is assumed to begin at the time of emplacement, and a constant congruent fractional dissolution rate of all radionuclides of 10^{-4}/yr, based on the initial inventory, is assumed. The radiation doses shown in Figure 9-1 are only relative doses. The actual radiation doses depend in magnitude on the volumetric flow rate of water carrying the released radionuclides. That is dealt with later.

The first groundwater carrying dissolved carbon-14 and iodine-129 will take an average time of 1,000 years to carry these nonsorbing radionuclides to the 1-km location assumed in this calculation, but because of dispersion some of the contaminated water will arrive earlier, causing the rounded edges of the fronts of the release bands. The release of carbon-14 and iodine-129 continues for a time given by

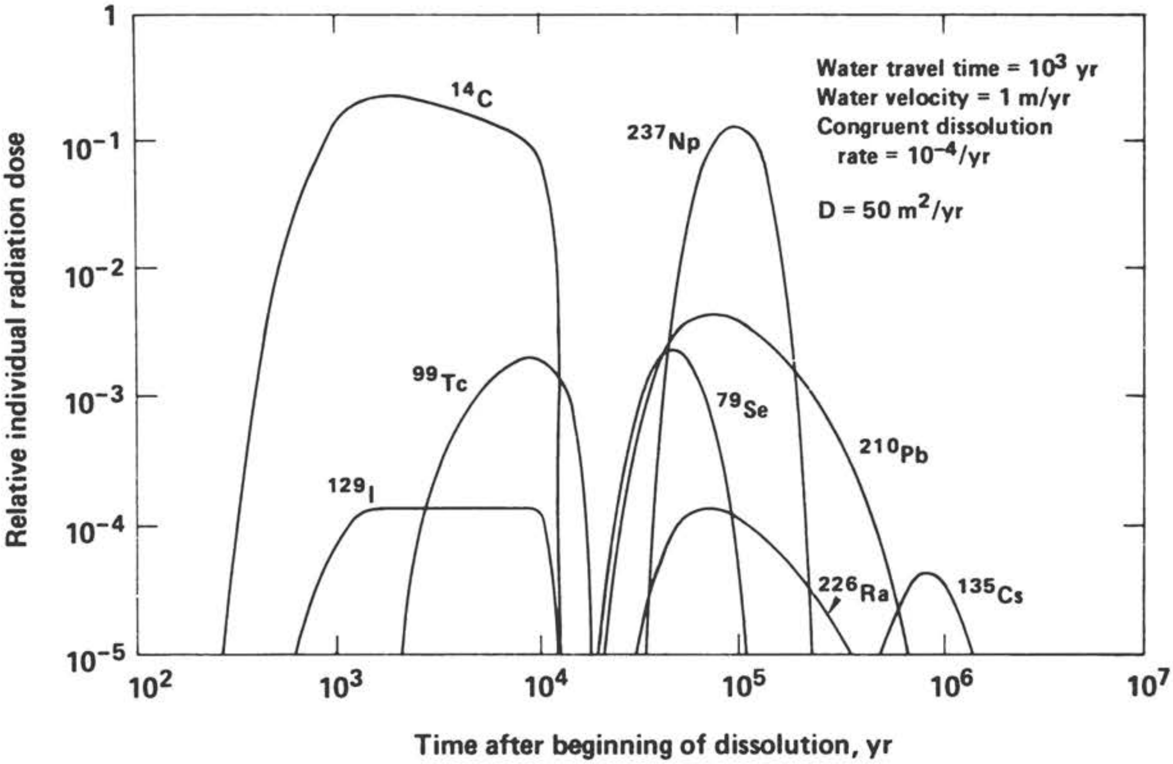

FIGURE 9-1 Relative individual radiation dose as a function of time: reprocessing waste from 10^5 Mg uranium fuel in basalt, congruent dissolution.

the dissolution time, here assumed to be 10,000 years, the reciprocal of the fractional release rate. After 11,000 years, and neglecting some delayed transport due to dispersion, all of the nonsorbing radionuclides originally in the waste will have either decayed or will have discharged into the environment.

Because of its long-half life of 17 million years, and for waste-dissolution times of the order of 10,000 years, all of the iodine-129 originally in the waste will discharge to the environment, and this will hold for water travel times orders of magnitude greater than that assumed for Figure 9-1. Even though some of the iodine may be ingested into humans for brief human retention periods, it ultimately enters the global iodine pool, primarily contained in the ocean, where it is further diluted, resides, and accumulates until it finally decays (Kocher 1981). Technetium-99, with a half-life of 2.12 x 10^5 years, suffers much the same fate, assuming the same waste dissolution time and negligible sorption.

Dispersion causes a greater rounding and attenuation of the later arriving radionuclides, such as technetium-99 and neptunium-237.

Neptunium-237 does not appear appreciably in the environment until a time given by the product of the water transport time and its retardation constant of 100 (cf. Table 7-1); i.e., 10^5 years. Because of its long half-life of 2.14 million years and because of the relatively low retardation constant (K = 100) for neptunium adopted for this study, neptunium-237 is predicted to reach the 1-km environment in sufficient concentrations to result in a relative radiation dose greater than that from any other actinide. About 97 percent of the neptunium-237 in the waste would reach the environment from the hypothetical repository illustrated here, and most of this would ultimately reach the ocean. As we shall see later (cf. Figure 9-9), for the water travel time of 1.9×10^5 years estimated by Rockwell Hanford Operations (RHO) (D. J. Brown and R. A. Deju, personal communications to T. H. Pigford, 1982) for the Basalt Waste Isolation Project (BWIP), most of the neptunium-237 would be removed by decay before it reaches the environment. Alternatively, adopting Rockwell Hanford's estimate of 5.6×10^5 for the retardation coefficient for neptunium, neptunium-237 would be removed by decay with water travel times as short as a few hundred years.

Adopting retardation coefficients of 500 for plutonium and americium (cf. Table 7-1), these elements will not appear in the assumed environment until about 5×10^5 years, long enough for all of their important radioisotopes to decay.

For the assumed 1,000-year water travel time of Figure 9-1, 1,602-year radium-226 appears in the environment after 50,000 years, as a result of the transport of 2.47×10^5/yr uranium-234, most of which is formed by the earlier decay of americium-242m, curium-242, and plutonium-238 in the waste, according to the decay chain shown in Figure 9-2. The release band of radium-226 is relatively broad, and its discharge to the environment occurs over a long period of time because of the simultaneous migration of the long-lived parents uranium-234 and thorium-230. For much longer water travel times, radium-226 continues to be discharged to the environment as a result of the transport of 4.51-billion-year uranium-238 and its daughters, but its rate of discharge is much smaller than that resulting from migration of the transient uranium-234.

The discharge of radium-226 is accompanied by the discharge of its decay daughter, 21-year lead-210. Although radium-226 and lead-210 will migrate through geologic media at activity concentrations in constant ratio given by the inverse ratio of their retardation coefficients (Harada et al. 1980), these two different radionuclides can follow different food chain pathways once they reach the environment. They are treated separately in the dose calculations.

Many other radionuclides that must be considered in analyzing the performance of a geologic repository are shown in the later graphs, but they are omitted from Figure 9-1 for simplicity.

9.7. EFFECT OF WATER TRAVEL TIME ON RADIATION DOSES, WITH CONGRUENT DISSOLUTION

9.7.1. Introduction

Because the peak concentrations of released radionuclides occur over many human lifetimes, as illustrated in Figure 9-1, we are interested in the peaks of the calculated releases when comparing with our overall

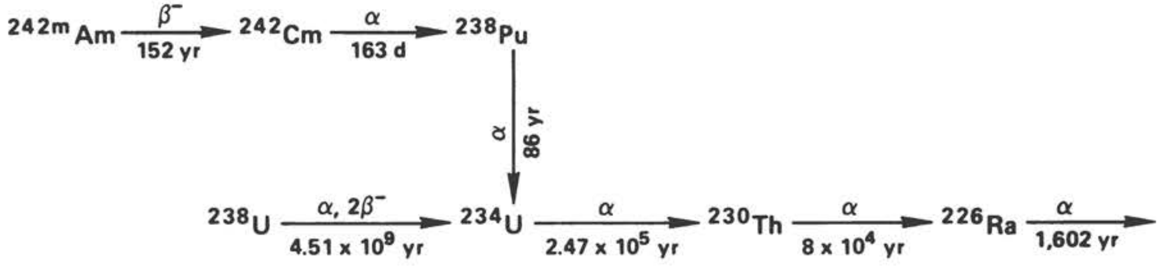

FIGURE 9-2 Decay chains leading to radium-226

performance goal expressed as an average lifetime dose rate to an individual. We will first examine the peak radiation doses calculated for congruent dissolution of the waste and neglect dispersion, so as to identify the effect of radionuclide inventories, radioactive decay, water travel time, and sorption delays on the isolation process. Corrections for dispersion are then introduced (Section 9.9), and the effect of more realistic dissolution rates is considered (Section 9.10). Dissolution is assumed to begin shortly after the repository is completely filled with waste from fuel reprocessing, and all waste is assumed to undergo dissolution at the same time and rate. The effect of a delay in the beginning of dissolution is considered (Section 9.8), as are the effects of the earlier higher temperatures on the dissolution rate (Section 9.13).

9.7.2. Basalt

The peak doses from the more important radionuclides, calculated for various water travel times, are shown in Figure 9-3 for a basalt repository. The groundwater flow is derived from data supplied by R. A. Deju and D. J. Brown (Rockwell Hanford Operations, personal communications to T. H. Pigford, 1982) for BWIP, and the river flow rate is that for the Columbia River, into which groundwater from the potential BWIP site is predicted eventually to discharge. For assumed groundwater travel times to the Columbia River between about 10 and 1,000 years, the principal contributor to the individual dose is neptunium-237. The calculated dose rate of 1.6 x 10^{-5} Sv/yr from river water is sixfold below the performance criterion of 10^{-4} Sv/yr adopted for this study. Carbon-14, assumed not to be retarded by sorption, is not attenuated appreciably by decay during this time period and contributes 4 x 10^{-6} Sv/yr to the individual dose.

For the average groundwater travel time to the Columbia River of 1.9 x 10^5 years estimated by BWIP (R. A. Deju, Rockwell Hanford Operations, personal communication to T. H. Pigford, 1982), carbon-14 and most of the other radionuclides are attenuated by decay before reaching the environment. For this groundwater travel time the maximum dose to the individual is due to neptunium-237 and lead-210. The latter reaches the biosphere by the migration of uranium-238 and its decay

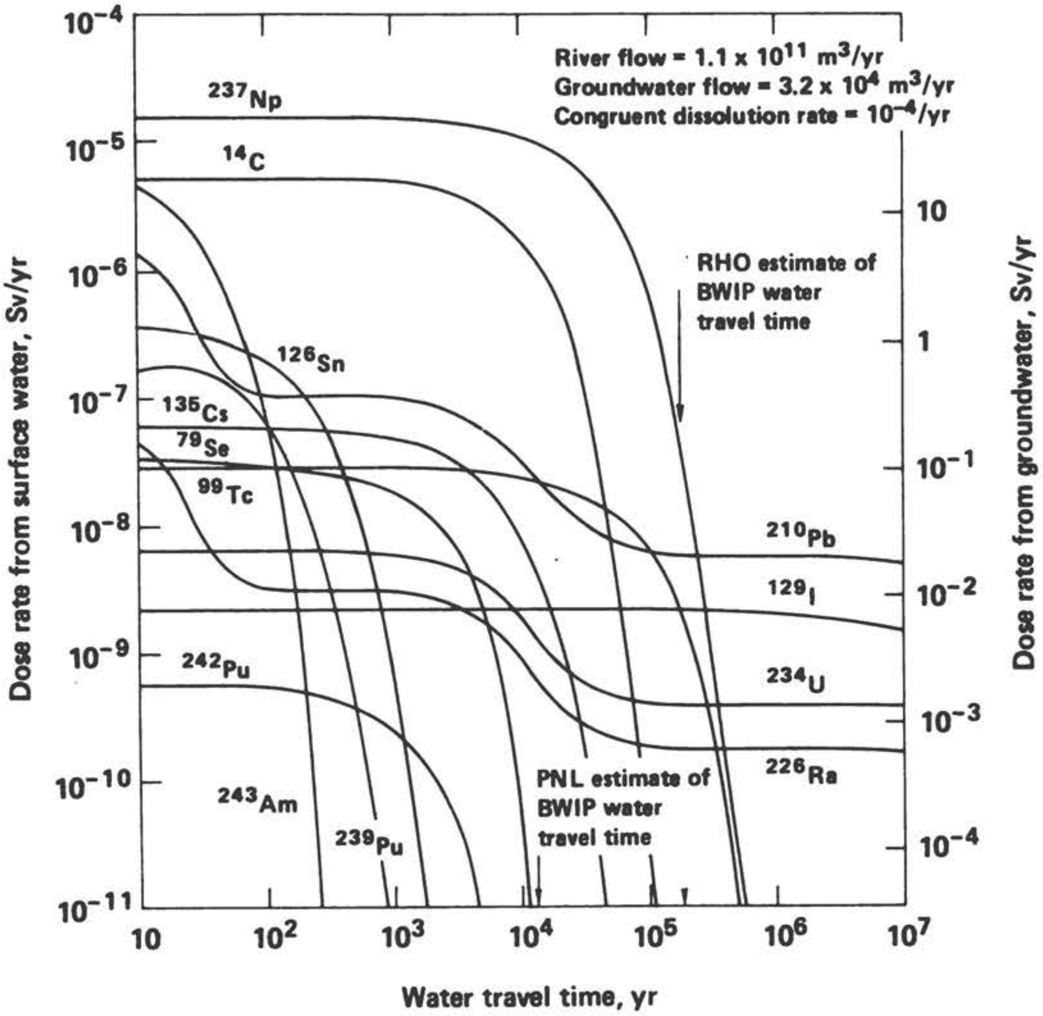

FIGURE 9-3 Individual radiation dose as a function of water travel time in basalt: reprocessing waste from 10^5 Mg uranium fuel, congruent dissolution, no dispersion.

daughters. At longer water travel times the lead-210 dose dominates, at a level of 6×10^{-9} Sv/yr, over four orders of magnitude below the performance criterion of 10^{-4} Sv/yr.

For the average groundwater travel time to the Columbia River of 1.5×10^4 years estimated by Pacific Northwest Laboratory (PNL) (Dove 1982, Dove et al. 1982), the maximum dose to the individual is about 10^{-5} Sv/yr from neptunium-237, tenfold below the performance criterion of 10^{-4} Sv/yr.

Because of the relatively low flow rates of groundwater, the dose rates to individuals, who theoretically use this contaminated groundwater for drinking and for irrigation, are all far above the performance goal of 10^{-4} Sv/yr. In calculating the doses from using groundwater, a constant groundwater flow rate suggested by R. A. Deju (Rockwell Hanford Operations, personal communication to T. H. Pigford, 1982) has been assumed. This is the effective value after the groundwater has traveled for 10^4 years from the repository. The effective groundwater flow rate increases with distance from the repository, and hence increases with water travel time, due to mixing with additional groundwater as the contaminated water moves through the media away from the repository. If sufficient data were available to include this effect, the dose rates from groundwater would decrease more rapidly with increasing groundwater travel time than shown in Figure 9-3.

9.7.3. Granite

Because site-specific data for a repository in granite are not being obtained in the United States (S. Goldsmith, Office of Nuclear Waste Isolation, personal communication to J. O. Neff, 1982), we adopt here the generic average properties for the purpose of this study, as listed in Table 9-4. The calculated individual doses for a granite site as a function of water travel time are shown in Figure 9-4. The effect of water travel time is similar to that calculated for basalt because the probable retardation constants listed in Table 7-1 for basalt and granite are equal for all of the important elements except plutonium and americium. The smaller retardation constant for plutonium in granite allows the plutonium to migrate 2.5 times farther, in distance measured by water travel time, than in the case of basalt. Even so, the plutonium isotopes do not contribute importantly to the radiation dose. The retardation constant for americium is sixfold greater in granite than in basalt. In neither case is americium important in terms of hydrologic transport of radionuclides to the biosphere.

The main difference between the calculations for granite and basalt results from the different flow rates adopted for these calculations. The river flow rate of 3.8×10^9 m^3/yr adopted for the granite site is based on an assumption that appeared in the U.S. Department of Energy's (1980) Final Environmental Impact Statement, and it is also the value adopted by Cloninger and Cole (1981) and by Burkholder (1982) for their calculations of generic granite and basalt repositories. This flow rate is 28 times less than the flow rate of the Columbia River, adopted here for the basalt repository. Consequently, the calculated doses in surface water for the granite repository are 28 times greater than for the basalt repository.

The groundwater flow rate assumed in Figure 9-4 for the granite repository is also that assumed by Cloninger and Cole (1981) and Burkholder (1982). It is threefold greater than the groundwater flow rate quoted for the BWIP site (R. A. Deju and D. J. Brown, Rockwell Hanford Operations, personal communications to T. H. Pigford, 1982), so the calculated doses from the assumed use of groundwater for the granite site are correspondingly lower than for the basalt site.

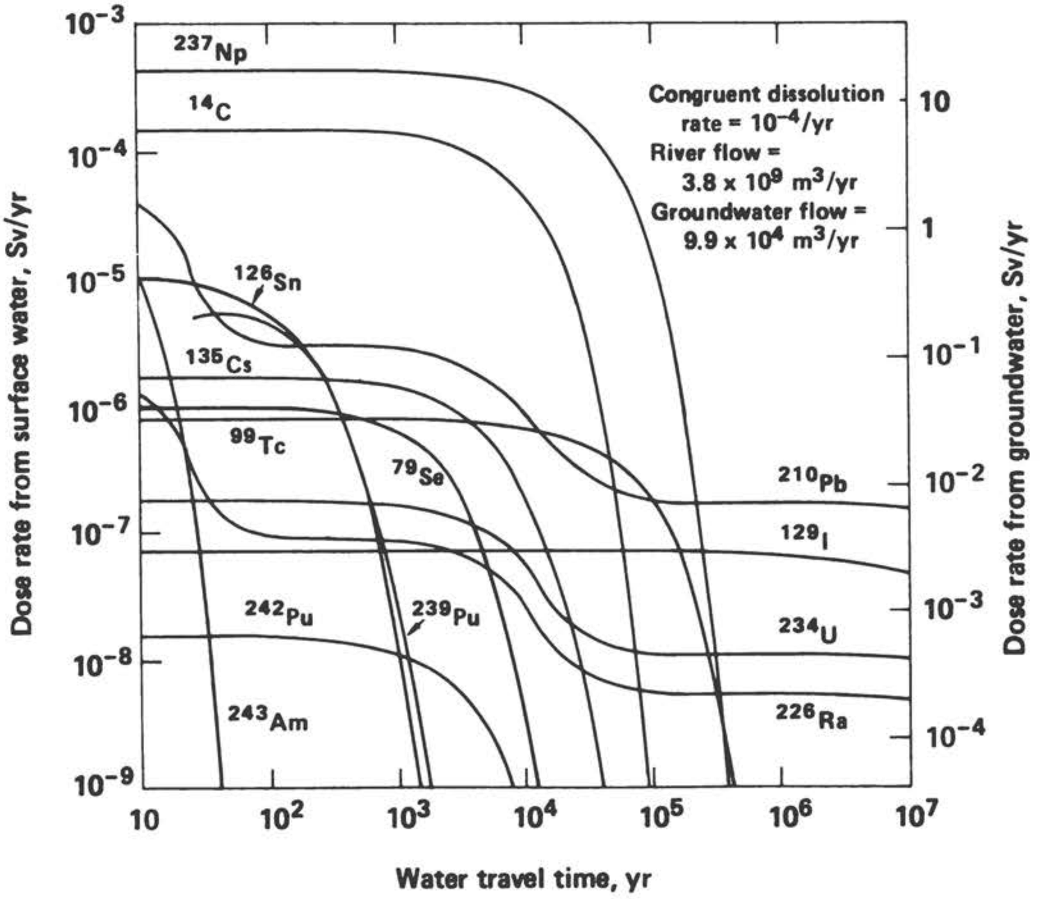

FIGURE 9-4 Individual radiation dose as a function of water travel time in granite: reprocessing waste from 10^5 Mg uranium fuel, congruent dissolution, no dispersion.

9.7.4. Salt

The calculated dose rates for a hypothetical salt repository are shown in Figure 9-5 as a function of water travel time for congruent dissolution. Here, for the purpose of comparing with other host sites, we assume that dissolution begins soon after emplacement. Because there is no groundwater in salt, dissolution by groundwater flow may never occur, or if it does occur by unexpected intrusion of an aquifer from adjacent media, it may be delayed for a much longer time than assumed here.

Here the predicted dose for cesium-135 persists with little attenuation for much longer water travel times than in the case of basalt or granite. This is a consequence of the 100-fold lower retardation constant for cesium in media through which saline water is flowing. The lower ratio of the radium retardation constant to that of uranium increases the dose from radium-226 from a salt repository. Neptunium-237 travels twice as far from the salt repository, measured by water travel time, than from repositories in basalt and granite.

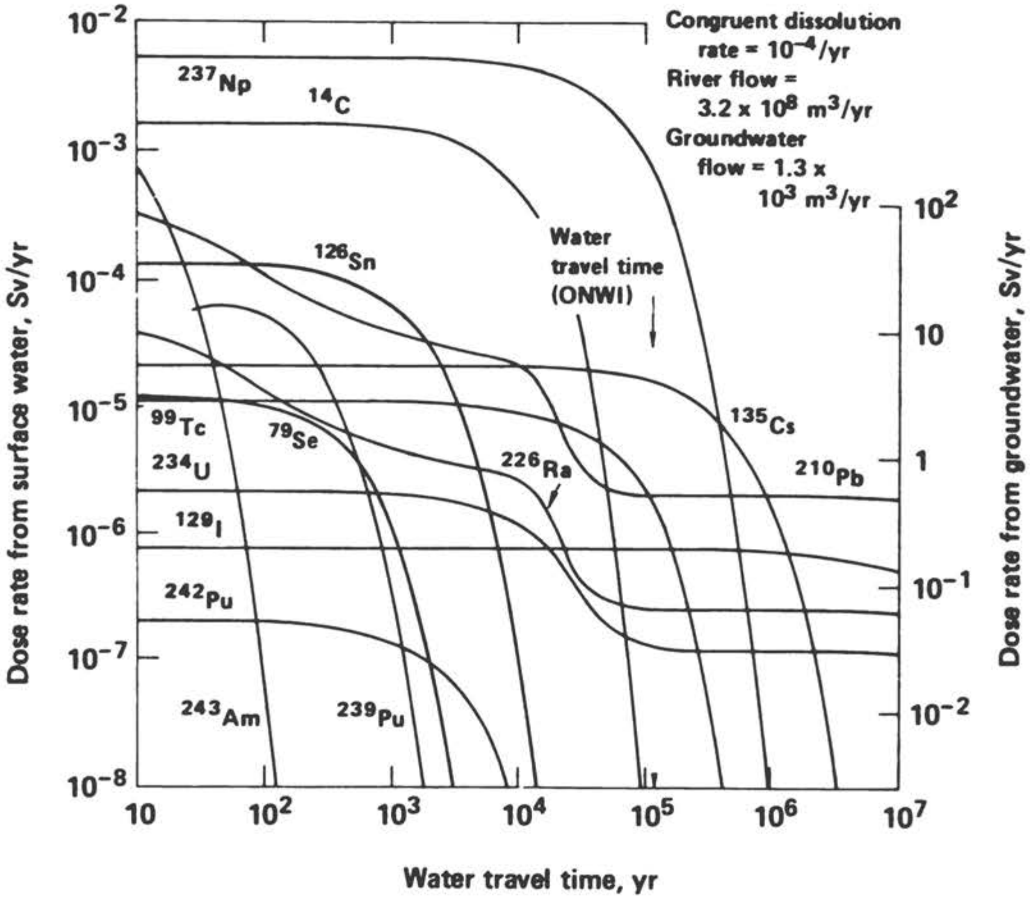

FIGURE 9-5 Individual radiation dose as a function of water travel time in salt: reprocessing waste from 10^5 Mg uranium fuel, congruent dissolution, no dispersion.

The volumetric flow rate of surface water for the hypothetical salt repository is 3.2×10^8 m^3/yr as recommended by S. Goldsmith (Office of Nuclear Waste Isolation, personal communication to J. O. Neff, 1982) for repositories constructed in the bedded salt in the Permian or Paradox basins. The flow rate of potentially contaminated groundwater of 1.3×10^3/yr is estimated from data supplied by G. E. Raines (Office of Nuclear Waste Isolation, personal communication to T. H. Pigford, 1982), from flow data for the Permian Basin. The river flow is 340 times less than the Columbia River flow associated with the basalt site, resulting in a correspondingly greater dose rate in surface water.

The dose rates in surface water for a site in domal salt would be ninefold less than those for bedded salt in Figure 9-5, because of the quoted greater surface water flow for the domal salt site. The hydrologic data for domal salt are for a site not now under consideration by DOE, as data for domal salt sites under actual consideration are not now available (S. Goldsmith, Office of Nuclear Waste Isolation, personal communication to J. O. Neff, 1982).

For radionuclides to undergo hydrogeologic transport from a salt site, water must intrude into the repository. This is a less-probable event than in the wet-rock repositories, so we would expect that the applicable radiation dose performance criterion for hydrogeologic transport from a salt site would be greater than that adopted for the other repository media.

9.7.5. Tuff

The calculated individual dose rates from contaminated groundwater from a tuff repository are shown in Figure 9-6 as a function of water travel time. Here the doses are affected by water travel time in much the same way quantitatively as in basalt and granite, because the retardation constants in these three media are almost the same for the important radionuclides.

The area being considered for a possible repository in tuff is the Nevada Test Site. There are four possible sites under consideration, two in saturated tuff, below the water table, and two in unsaturated tuff, above the water table. To the accuracy of these calculations, Figure 9-6 applies for either location.

Data do not now exist on the rates of flow of groundwater that could be contaminated by radionuclides from a tuff repository. The calculations of individual dose rate for a tuff repository assume, arbitrarily, that the groundwater flow rate is the same as that adopted by Cloninger and Cole (1981) and by Burkholder (1982) for other generic repositories.

Because there is no flowing surface water into which potentially contaminated groundwater from the tuff site can discharge, the calculations for the tuff site are limited to the doses from contaminated groundwater.

We have then sought to determine the pathways by which potentially contaminated groundwater from the potential tuff site could reach the surface or in other ways be used by humans. Data supplied by L. D. Tyler (Sandia National Laboratories, personal communications to T. H. Pigford, 1982) summarized in Table 9-4, refer to the map shown in Figure 7-9. A possible repository in saturated tuff is at location B-1 on this map. To the southeast is an operating well, J-13, and to the south of that is another operating well, J-12. We are informed that these two wells furnish potable and service water for the Nevada Test Site. The distances to each of these wells from the possible repository site are 6.4 km for well J-13 and 11.1 km for well J-12, with estimated water travel times from the assumed repository site of 1,200 and 2,200 years. We assume that these wells could be plugged, if necessary, if a repository were to be constructed on the tuff site and if it were determined that there could be a reasonable chance of contaminating the well water by radionuclides from the repository.

Farther south is a well at Lathrop Wells, at a distance of 24.6 km from the possible repository site, with an estimated water travel time of about 4,300 years. The potentiometric profiles indicate that groundwater may travel in a southerly direction from the field of

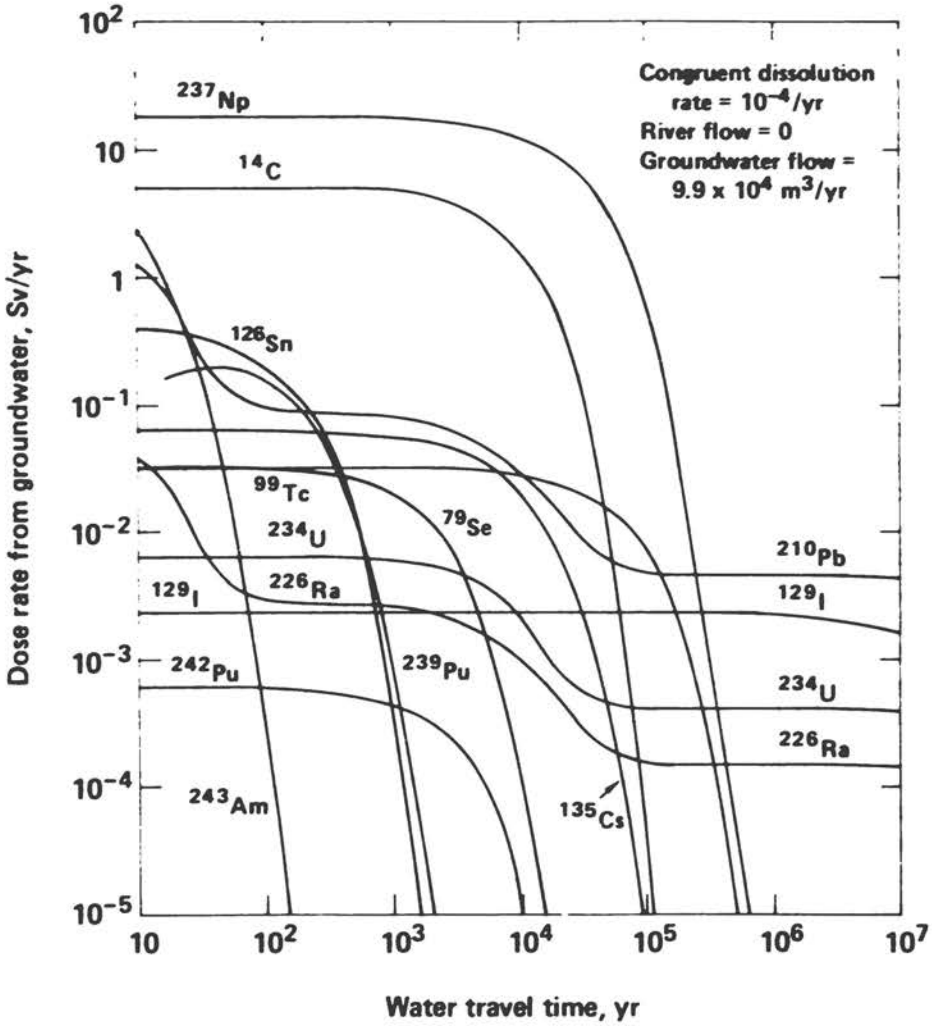

FIGURE 9-6 Individual radiation dose as a function of water travel time in tuff: reprocessing waste from 10^5 Mg uranium fuel, congruent dissolution, no dispersion.

possible repository locations, so these wells may be on the route of groundwater potentially contaminated by waste. The groundwater eventually reaches the Amargosa Desert, where some groundwater is used for irrigation, and it may even flow as far as California. We have received no further information concerning the flow pathways of groundwater from a possible tuff site. Evidently, no further information is now available concerning the possible use of that water by humans.

We have been informed by L. D. Tyler (Sandia National Laboratories, personal communication to T. H. Pigford, 1982) that emphasis has shifted to the unsaturated tuff for a possible repository location. Preliminary data suggest that there is some recharge of about 2 percent of the 15-cm annual rainfall into the unsaturated zone by infiltration, corresponding to a local pore velocity in the repository of about 3 cm/yr. The estimated travel time for water to flow from the waste-emplacement zone to the water table is about 2.1 x 10^4 years. This is to be added to the travel times quoted above for a repository in saturated tuff, resulting in a total water travel time to Lathrop Wells of about 2.5 x 10^4 years, about sixfold greater than for a repository in saturated tuff. This is long enough for a 20-fold attenuation in the peak dose of carbon-14 in the groundwater, but the neptunium-237 dose for congruent dissolution remains many orders of magnitude greater than the individual dose limit. As we shall see later (Section 9.10.8), the expected low solubility limit of neptunium and other radionuclides will result in groundwater doses from a tuff repository that are much closer to the individual dose criterion of 10^{-4} Sv/yr.

The tuff site is difficult to evaluate in terms of calculated doses from use of water by humans. Although our later calculations with solubility-limited dissolution and dispersion will reduce the estimated doses in groundwater, it seems that any normal and continuing use of the potentially contaminated groundwater from a site in saturated or unsaturated tuff could present a problem, if the flow rates of contaminated groundwater are anywhere near those adopted in this study. The absence of flowing surface water in this region presents a greater incentive to use groundwater than for sites in less arid regions. For the long-term future considered here, the present wells are not significant as definite locations of future use of potentially contaminated groundwater, but they are significant in that they show some likelihood of future human use of groundwater in this general area. Therefore, we attach greater significance to the calculated groundwater doses for the tuff site than for sites in less arid regions. Further discussion of the performance of a repository in tuff appears in Section 9.10.8.

9.8. EFFECT OF TIME DELAY BEFORE DISSOLUTION

Assuming that all radionuclides undergo congruent dissolution at the same constant fractional release rate, the effect of a time delay before the beginning of dissolution can be deduced directly from the decay equations. A 1,000-year delay decreases the carbon-14 dose by 11 percent. It considerably decreases the dose from strontium-90 and americium-241, which are important only in the very near field. It increases the near-field dose from plutonium-239, neptunium-237, and uranium-234 by allowing time for the decay of their precursors. All of these decay effects are relatively small.

No important effect of delay on dose rate, with constant dissolution rate, would be expected until the delay is of a time comparable to the half-lives of the radionuclides, or of their precursors, that are the important contributors to the radiation doses in the biosphere. Such delays are likely in natural salt, wherein continued dissolution can begin only after the unlikely intrusion of flowing water into the salt.

In Section 9.13 we analyze the thermal effects on dissolution rates and radiation doses if dissolution begins during the period of repository heating.

9.9. EFFECT OF DISPERSION

The locally nonuniform distribution of water velocities through pores and fractures can act dispersively on the concentration gradients at the edges of the moving plumes of dissolved radionuclides, resulting in broadening of the concentration bands and some reduction in concentrations. The most important effect on calculations of the rate of discharge of radionuclides to river water, and on the maximum individual dose therefrom, is that resulting from dispersion in the direction of flow, i.e., axial dispersion. Little is known about the proper value of the dispersion coefficients for the media considered here, but a value of 50 m^2/yr for the axial dispersion coefficient is assumed for the purpose of this study, based largely on data from dye-tracer tests at the BWIP site (D. J. Brown, Rockwell Hanford Operations, personal communications to T. H. Pigford, 1982).

Dispersion corrections to the maximum dose rates from carbon-14, cesium-135, iodine-129, neptunium-237, and radium-226 for a basalt repository are shown in Figure 9-7. As would be expected, dispersion reduces the maximum dose as the radionuclide travel time increases. Attenuation by dispersion is greater when the radionuclide travel time is much greater than the dissolution time of the waste. The actual travel time for nonsorbing iodine-129 is the same as the water travel time, and dispersion does not attenuate the maximum concentration of iodine-129 band until about 10^5 years. Cesium-135 is strongly sorbed, with a retardation constant of 1,000 for basalt, so for a water travel time as short as 10^3 years the cesium travel time is long enough for dispersion to be appreciable. Even though radium-226 and its parents are appreciably retarded by sorption, dispersion corrections are relatively small because of the very wide band of radium and its parent thorium-230 distributed through the geologic medium. Any mechanism that increases the bandwidth, such as increasing the time period over which radionuclides are released from the waste, can reduce the effects of axial dispersion.

In later calculations for solubility-limited dissolution, dispersion is less important because of the resulting long times over which solubility-limited radionuclides are released. Dispersion is most important in reducing the peak doses from cesium-135, which is assumed not to be solubility limited.

For the dissolution rate assumed for Figure 9-7, the maximum concentration and dose rate from nonsorbing carbon-14 will not undergo appreciable dispersion attenuation during the time period of a few tens of thousands of years that carbon-14 can exist.

Transverse dispersion is an important near-field effect, but transverse dispersion during far-field transport does not appreciably affect the maximum concentrations and doses from radionuclides discharged to the river. However, the concentrations of radionuclides in groundwater are affected by transverse and axial dispersion and by interflow dilution, the effect becoming more important as the lifetime of the contaminant plume increases. These effects of progressively increasing transverse dispersion and interflow dilution are not shown in the graphs presented here for the predicted doses from groundwater, but they should be considered in more detailed analyses of groundwater

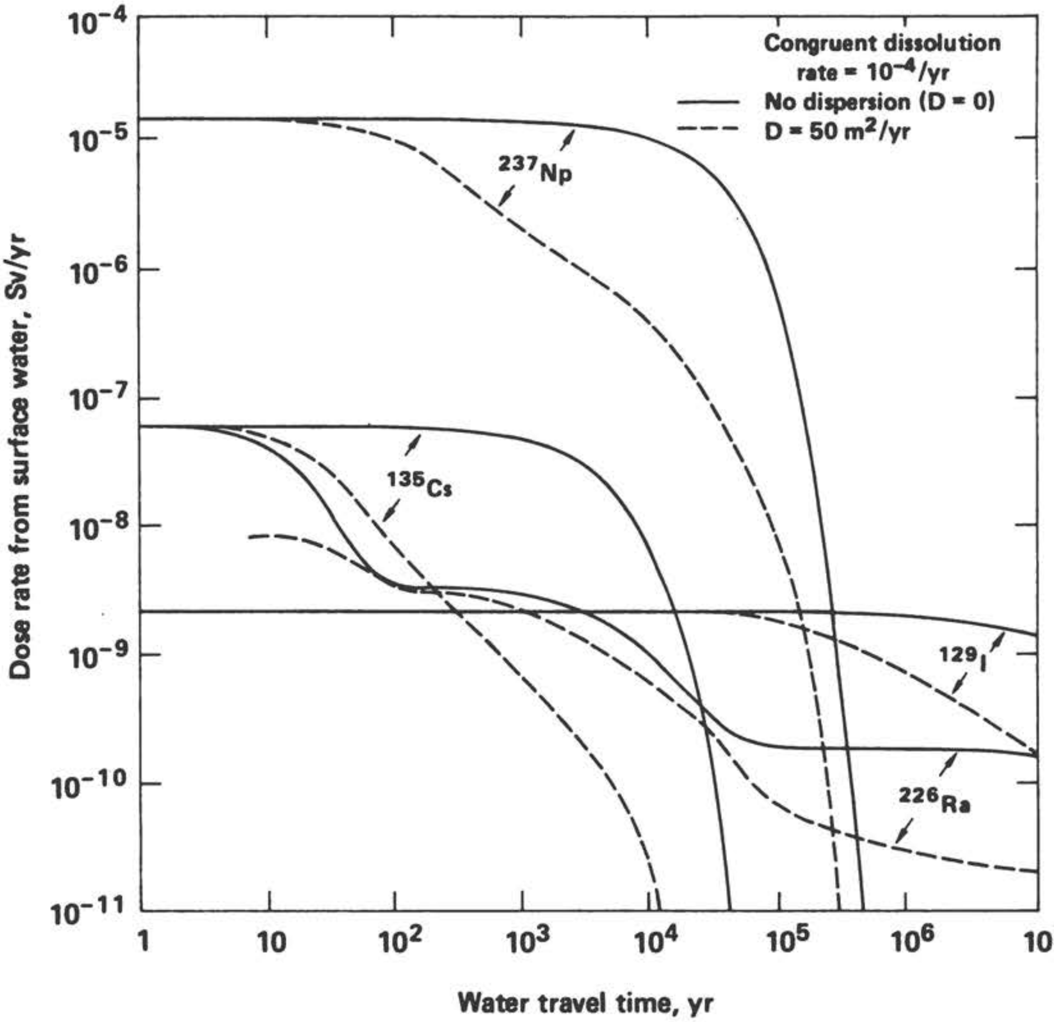

FIGURE 9-7 Individual radiation dose as a function of water travel time in basalt, showing the effect of dispersion: reprocessing waste from 10^5 Mg uranium fuel, congruent dissolution.

contamination. For each repository site, the groundwater concentrations and radiation doses therefrom are calculated for a single effective flow rate of contaminated groundwater specified for that site. The groundwater flow rate of 3.2×10^4 m^3/yr for the BWIP site was estimated for a 10,000-year-old plume from the repository (D. J. Brown, Rockwell Hanford Operations, personal communications to T. H. Pigford, 1982), and this value was used as a constant for all groundwater travel times. If more information were available on the volume extent of the contaminated plume as a function of water travel time, such data would be used in these calculations and would predict a more rapidly decreasing concentration in groundwater with increasing travel time than is shown in the present graphs. Such information should be developed by DOE for its candidate sites.

Groundwater flowing past an individual package of dissolving waste creates a plume of contaminated water downstream of each waste package. Transverse dispersion soon merges the plumes from the many waste packages into a broad plume of essentially uniform concentration (Pigford et al. 1980, Chambré et al. 1982a) that may later become diluted with merging interflows through contacts after it leaves the host rock of the repository. This local near-field dispersion does not affect the maximum far-field doses calculated in this study.

The EPA staff (C. B. Smith et al. 1981), Wood (1980), and Wood and Rai (1981) treat solubility-limited transport as originating with a volume flow of groundwater uniformly saturated with each solubility-limited radionuclide as the groundwater flows across the plane of waste emplacement. This is an unrealistic and unnecessarily conservative assumption in that it neglects the important resistance in the concentration boundary layer adjacent to the waste surface, and it assumes instantaneous diffusive and dispersive transport across the flow cross section. Although useful for upper bound estimates of solubility-limited transport, such estimates do not reflect a valid physical model of waste dissolution and transport. This example illustrates that EPA's analysis of generic repositories, which is part of EPA's basis for its proposed standard, does not necessarily represent the performance of available technology for geologic isolation.

9.10. EFFECT OF WATER TRAVEL TIME ON RADIATION DOSES, SOLUBILITY-LIMITED DISSOLUTION

9.10.1. Solubility-Limited Dissolution Rates

It is not necessary to assume that all radionuclides dissolve congruently with the same fractional release rate, as is usually assumed in other analyses of repository performance (Cloninger et al. 1980, Cloninger and Cole 1981, Burkholder 1982). As has been pointed out in Chapter 5, congruent dissolution of most of the radionuclides in the waste is not expected to occur. Many of the important radioelements are so insoluble that they cannot go into solution as rapidly as the waste matrix itself.

In Chapter 5 (cf. Section 5.7.4) there is summarized a theoretical prediction of the long-term rate of dissolution of a discrete waste solid exposed to groundwater in a porous medium (Chambré et al. 1982a,b,c). The theory predicts a time-dependent fractional rate of dissolution of the waste matrix and of individual radioelements, as affected by the solubility in groundwater of each constituent being dissolved, the diffusion coefficient in the groundwater, the velocity of the groundwater flowing past the waste package, the effective porosity of the surrounding medium, the dimensions of the overall waste form, and the amount of that constituent in the undissolved solid waste. Constituents with suitably low solubility in the groundwater, such as neptunium, plutonium, and americium, can have fractional dissolution rates less than that of the waste matrix. When the waste matrix dissolves, precipitates of insoluble compounds of these radioelements would be expected to form, and it is the rate of dissolution of these low-solubility precipitates that is predicted by the theory utilized here.

In our calculations discussed in Section 9.7 for congruent dissolution at a given fractional dissolution rate, the rate of dissolution of an individual radionuclide is given by the Bateman equation for batch decay (Harada et al. 1980). For the purpose of estimating peak lifetime radiation doses, it is not necessary to resolve the advective transport equations to account for the more complicated time-dependent release resulting from the application of Equations (1) and (2) in Chapter 5. The peak concentrations and radiation doses from these radionuclides can be reasonably estimated by applying Equations (1) and (2) to the waste form as it first begins to dissolve.

In Table 9-5 we summarize the solubility-limited fractional dissolution rates for silica and insoluble elements in a borosilicate-glass waste form, calculated for a typical waste cylinder at 20°C exposed to groundwater flowing at a pore velocity of 1 m/yr. In applying these data to calculate the performance of conceptual repositories, we first neglect the effects of higher temperatures if waste is exposed to groundwater during the thermal period. The possible magnitude of the temperature effect is estimated in Section 9.13. We assume here that the groundwater pore velocity in all repository host rocks is constant at 1 m/yr. This water velocity is larger than the expected value for basalt, even during the thermal period. This may also be true for other repository rocks. As we have shown in Chapter 5, a limiting fractional dissolution rate for the case of zero velocity is only about a fourth of the predicted values at 1 m/yr, so the present predictions at 1 m/yr are very close to the limiting low-velocity dissolution rate at 20°C. Correcting to the ambient temperature of basalt of 57°C, the release rate predictions are expected to be too low for basalt by a factor of two. Such small corrections in the predicted dissolution rate are not yet justified because of larger uncertainties in the solubilities and because the theory is yet to be tested by experiment.

9.10.2. Carbon-14 Waste

Carbon-14 is not a constituent within the packages of high-level reprocessing waste, because it is separated in fuel reprocessing. However, assuming that carbon-14 is recovered separately and is emplaced in a geologic repository, we can estimate the solubility-limited dissolution rate of a hypothetical waste package containing carbon-14. It is assumed here that all of the carbon-14 recovered from reprocessing the entire 10^5 Mg of uranium fuel, whose waste represents the full loading of a repository, is accumulated and incorporated into a single waste package. The total inventory of carbon-14 is 19.8 kg. We assume that the carbon-14 is in the form of calcium carbonate, with a total mass of 144 kg. The heat generation rate within this special carbon-14 package is about 40 W, far less than for a single high-level waste package. Therefore, the carbon-14 package can be assumed to operate at the ambient temperature.

TABLE 9-5 Calculated Fractional Release Rates for Borosilicate-Glass Waste at 20°C

Constituent	Initial Waste Concentration (g/cm^3)	Solubility (g/cm^3)	Fractional Release Rate (yr^{-1})
SiO_2	1.6	5×10^{-5} [a]	1.1×10^{-6}
Tc	1.92×10^{-3}	1×10^{-9}	2×10^{-8}
U	1.22×10^{-2}	1×10^{-9}	4×10^{-9}
Np	1.92×10^{-3}	1×10^{-9}	2×10^{-8}
Pu	1.15×10^{-4}	1×10^{-9}	4×10^{-7}
Am	3.56×10^{-4}	1×10^{-10}	1×10^{-8}
Se	1.40×10^{-4}	1×10^{-9}	3×10^{-7}
Sn	9.40×10^{-5}	1×10^{-9}	5×10^{-7}

NOTE: Waste cylinder: radius = 0.152 m; length = 2.46 m; fission-product and actinide oxides from 460 kg of uranium in fuel; groundwater pore velocity = 1 m/yr.

[a] For amorphous SiO_2 (Fournier and Rowe, 1977). Other solubilities are from Table 7-1.

The solubility of calcium carbonate at 25°C is 1.4×10^{-5} g/cm^3 (Seidell 1965). Assuming that the carbon-14 waste package is of the same overall dimensions as the high-level waste package assumed in Table 9-6, we estimate a solubility-limited fractional release rate of carbon-14 of 4×10^{-7}/yr. This reduces the individual dose rate from carbon-14 by a factor of 500 below that shown in Figures 9-3 through 9-6 for congruent dissolution.

Although we have considered in this study separated carbon-14 as a possible candidate for a geologic repository, we suggest that alternatives be considered, such as ocean disposal.

9.10.3. Iodine-129 Waste

We assume that the iodine recovered in fuel reprocessing is to be emplaced in a geologic repository and that the iodine is converted to a suitably insoluble compound loaded into canisters of the same dimensions as assumed for high-level waste. Although no chemical or physical form

TABLE 9-6 Effect of Temperature on the Solubility and Liquid Diffusion Coefficient for Silica

Temperature (°C)	Solubility[a] (g/cm^3)	Diffusion Coefficient[b] (cm^2/s)
20	5.0×10^{-5}	1×10^{-5}
50	8.8×10^{-5}	2.0×10^{-5}
100	1.7×10^{-4}	4.5×10^{-5}
150	2.8×10^{-4}	7.9×10^{-5}
200	4.2×10^{-4}	1.2×10^{-4}
250	5.8×10^{-4}	1.6×10^{-4}

[a] Fournier and Rowe (1977).

[b] Estimated from the Einstein equation: $D\mu/T$ = constant; D = diffusion coefficient; μ = viscosity; T = absolute temperature.

of the iodine waste has been specified, we assume here that iodine is in the form of silver iodide, with a density of 5.6 g/cm^3. For an estimated solubility of the contained iodine of 1 x 10^{-8} g/cm^3 (Seidell 1965), and correcting for dilution by the stable iodine-127 in the waste, the estimated solubility-limited dissolution rate of iodine is 8 x 10^{-11}/yr, resulting in an individual dose rate from iodine in river water and groundwater that is 1.2 x 10^{-6} times that calculated in Figures 9-3 through 9-6 for congruent dissolution. The fractional dissolution rate of the silver iodide waste is now low enough that most of the iodine-129 decays before it is released from the waste package. Here decay affects only the total release of radioiodine, but it does not appreciably affect the maximum individual dose from iodine.

The total mass of iodine in each waste package would be 560 kg, with a heat generation rate of a fraction of a watt, so there is no self-heating. About 40 waste packages would contain all of the 22.4 Mg of total iodine recovered from the 10^5 Mg of uranium fuel, and 18.8 Mg of silver would be required. Assuming the May 1982 cost of industrial silver of $0.17/g, the total cost of silver to combine with the iodine would be $3.3 million.

At the above calculations assume, perhaps unrealistically, that the groundwater contains no excess halide or ammonium ions that easily complex and solubilize silver iodide. Therefore, the calculations only illustrate how choosing a waste form of low surface-to-volume ratio can reduce the dissolution rate. The reliability of the dissolution rate estimates for silver iodide is very uncertain.

Although we have considered in this study separated iodine-129 as a possible candidate for a geologic repository, we suggest that alternatives such as ocean disposal be considered before committing separated radioiodine to geologic disposal.

9.10.4. Cesium Dissolution Rate

All of the important radionuclides in reprocessing wastes other than cesium-135 can be limited in their dissolution rates because of the low solubilities of their compounds. We can only estimate the range of dissolution rates of cesium, because the solubilities of its usual compounds in water are too great to limit its dissolution rate.

A possible exception to the usual conclusion that cesium will not be solubility limited has been pointed out by G. Jacobs (Rockwell Hanford Operations, personal communication, 1982). If waste in a basalt repository is exposed to groundwater during the thermal period, at temperatures as high as 200°C, cesium can react with basalt minerals to form low-solubility pollucite. The inferred solubility of the reacted cesium of about 10^{-6} g Cs/cm^3 at this temperature is too high to appreciably reduce the cesium dissolution rate. Pollucite formation would evidently not occur if the waste is exposed to groundwater at the lower temperatures following the thermal period. However, because of uncertainties in the cesium dissolution rate during thermal-period exposure, pollucite formation should be considered as a means of narrowing the range of possible dissolution rates of cesium during the thermal period.

A lower limit to the dissolution rate of cesium would be the dissolution rate of the waste matrix, estimated in Table 9-5 to be 1.1×10^{-6}/yr, if cesium dissolves congruently with the silica matrix. However, some laboratory experiments indicate that cesium can diffuse preferentially through the waste solid. Although cesium diffusion is evident in the laboratory tests, there is no reliable basis for predicting whether or not diffusion-controlled dissolution of cesium persists for the times of interest in a repository. Adopting laboratory data on cesium dissolution from borosilicate glass, J. L. Crandall (Savannah River Laboratory, personal communication to T. H. Pigford, 1981) quotes a fractional release rate of 1.6×10^{-3}/yr, based on static experiments at 90°C. From the diffusion parameters suggested by the data of Godbee and Joy (1974) we extrapolate to times of a few hundred years after the beginning of dissolution, and we estimate a steady state fractional dissolution rate of cesium of 7×10^{-3}/yr for the waste-form dimensions given in Table 9-5. If the waste form were the ceramic alternative SYNROC, instead of borosilicate glass, data reviewed in Chapter 5 indicate that the cesium fractional release rate may be as low as about 10^{-7}/yr.

In Figure 9-8 we show the effect on radiation doses from cesium-135 in a basalt repository that would result from the range of dissolution rates of cesium, from 1.1×10^{-6}/yr to 1.6×10^{-3}/yr. If dispersion is negligible, then the effect of uncertainties in cesium dissolution rate will be large and important, and cesium-135 could be the main contributor to radiation dose for water travel times up to 10^5 years. However, with an axial dispersion coefficient as large as 50 m^2/yr, cesium-135 is important only within the near field of the repository. The three dispersion curves of Figure 9-8 coalesce at water travel times greater than about 10^3 years. This illustrates the important conclusion, recently demonstrated by Sato et al. (1982), that the far-field peak concentration of a radionuclide migrating under the influence of dispersion can be independent of the dissolution rate of that radionuclide, provided that the migration time is much longer than the dissolution time. The greater near-field concentration resulting from a higher dissolution rate is more rapidly attenuated by dispersion because of the relatively short time period over which the release occurs, and the resulting far-field concentration becomes identical to that from a lower dissolution rate.

Consequently, we can conclude that with the expected dispersion accompanying hydrogeologic transport, the actual dissolution rate of cesium from the waste may be relatively unimportant for repository sites wherein cesium retardation is as great as that adopted here for basalt. This insensitivity of cesium dose to cesium dissolution rate is important for repositories in basalt, granite, or tuff because of the relatively high retardation coefficients and consequently high dispersion during the water travel times of interest. It is not as useful for more weakly sorbing radionuclides, and it will not be as useful for cesium released from a salt repository, where sorption and dispersion of cesium are not as great as in basalt, granite, and tuff.

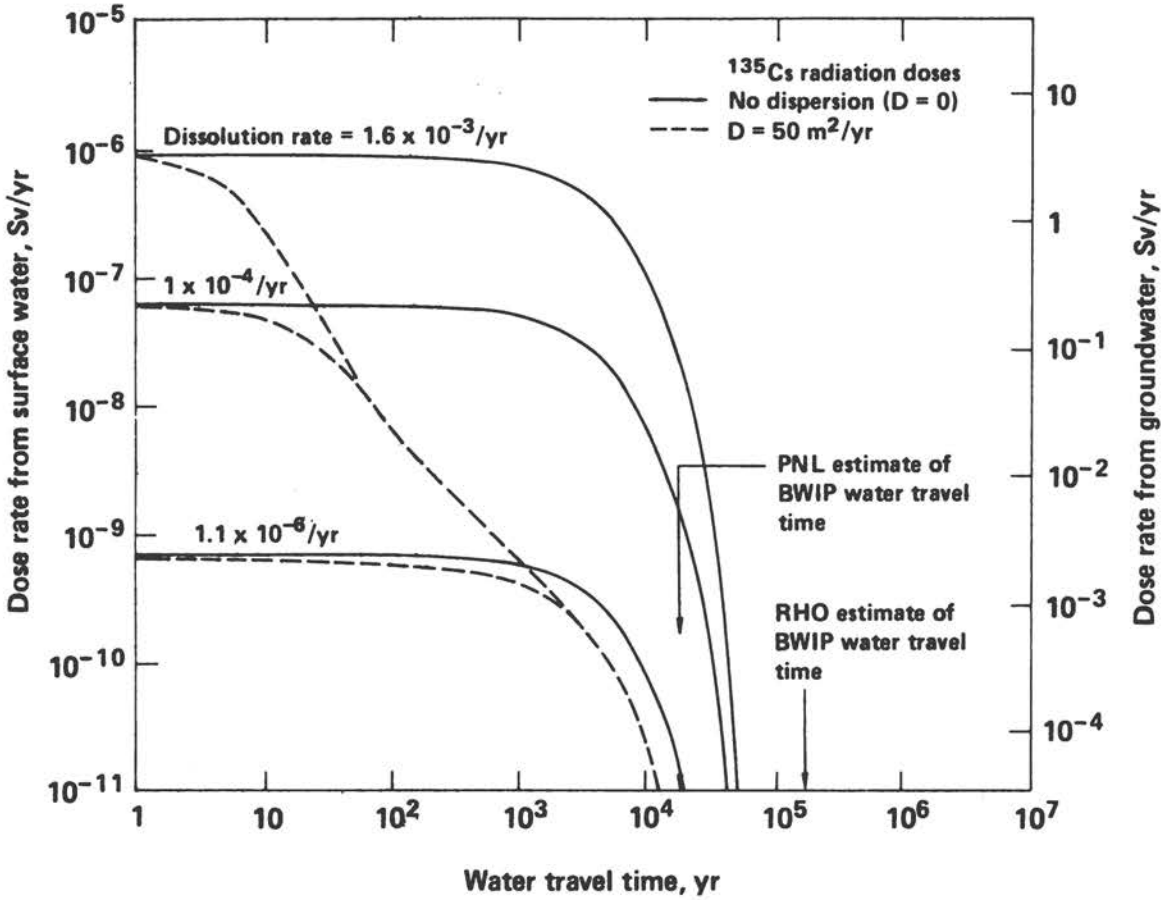

FIGURE 9-8 Individual dose from cesium-135 as a function of water travel time in basalt, showing the effect of dissolution rate and dispersion: reprocessing waste from 10^5 Mg uranium fuel.

9.10.5. Basalt

The fractional release rates are applied individually to the radionuclides to calculate the peak individual doses for a Hanford basalt repository. The results shown in Figure 9-9 are calculated for solubility-limited dissolution of all radionuclides except cesium-135. The effects of axial dispersion, with an assumed dispersion coefficient of 50 m^2/yr, are including in these calculations of doses from repositories with solubility-limited dissolution.

Selenium-79, technetium-99, and tin-126 are no longer principal contributors to the dose. The solubility-limited dissolution rate of uranium reduces the radiation dose from uranium-234, but its most important effect is on the radiation dose from the decay daughter radium-226. Radium-226 and its precursor thorium-230 will not be present in sufficient mass for their solubilities to be limiting, but the low dissolution rate of the chain mother, uranium, means that the only way for appreciable concentrations and doses from radium-226 to

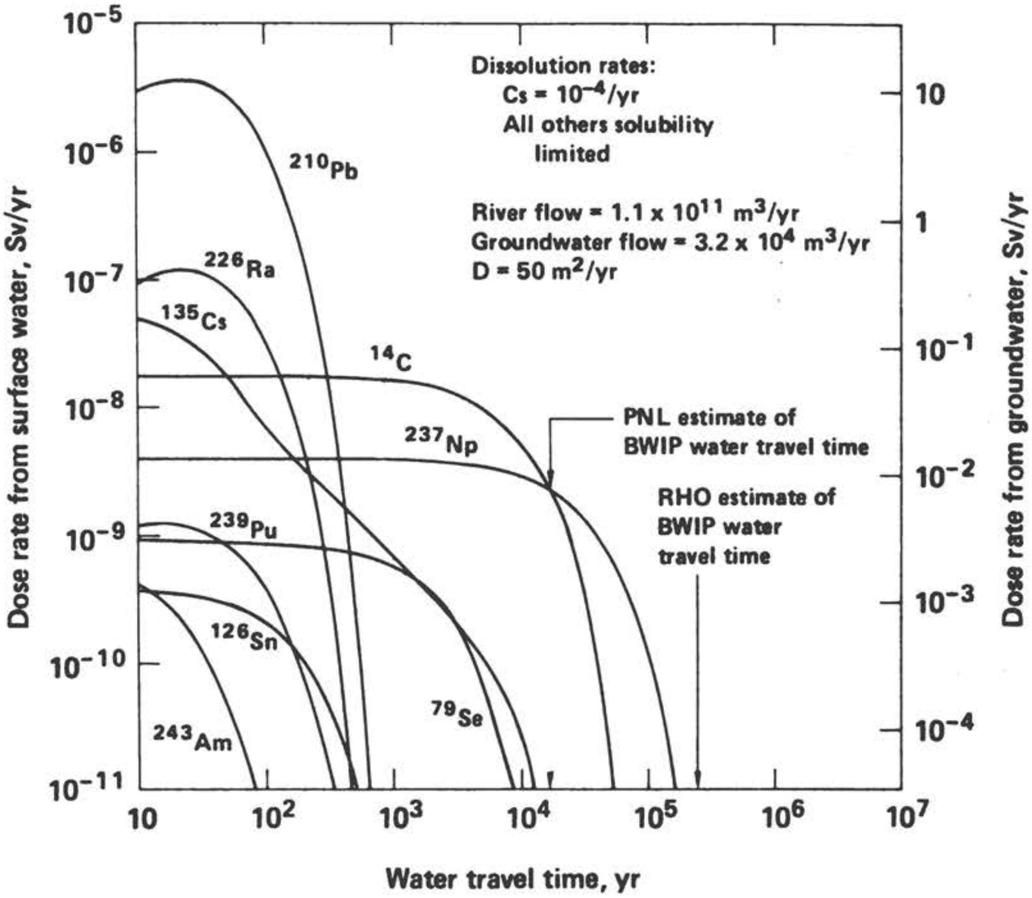

FIGURE 9-9 Individual radiation dose as a function of water travel time in basalt: reprocessing waste from 10^5 Mg uranium fuel, solubility-limited dissolution.

appear in the environment is by transport of 80,000-year thorium-230 directly from the waste. This is possible only for relatively short water travel times, of the order of a few hundred years. Consequently, when uranium is solubility limited, radium-226 is a principal contributor to the individual dose only for water travel times of a few hundred years and less, as shown in Figure 9-9.

Based on these predictions, and based on the estimate by the BWIP project of a water travel time to the environment of 1.5×10^5 years, the maximum individual dose in river water will be about 10^{-11} Sv/yr, a factor of 10 million below the radiation dose criterion of 10^{-4} Sv/yr adopted for this study.

For the average groundwater travel time to the Columbia River of 1.5×10^4 years estimated by Pacific Northwest Laboratory (Dove 1982, Dove et al. 1982), the maximum dose to the individual will be about 3×10^{-9} Sv/yr, over four orders of magnitude below our individual dose criterion.

Figure 9-9 represents what we consider to be a reasonable but preliminary estimate of the possible performance of a basalt repository. It indicates an approach toward analyzing repository performance that takes into account the technical performance of the isolation of waste package, geology, and hydrology with regard to each of the important radionuclides. It suggests many technical features that will require verification through experiment and further analysis. Many features of the present analysis may conservatively predict greater doses than will actually occur. There are also many uncertainties and other phenomena, not taken into account in the present analysis, that must be determined before the margin of uncertainty in the predicted dose can be determined. Such analysis is beyond the scope of this study. However, it appears that the large margin between the calculated dose in river water and the goal of 10^{-4} Sv/yr may sufficiently allow for these uncertainties.

9.10.6. Granite

The solubility-limited dissolution rates estimated in Table 9-4 are adopted here for a generic granite repository, and the predicted doses are shown in Figure 9-10. Because the controlling radionuclides have the same estimated retardation constants for granite as for basalt, the calculated doses for granite behave in the same way as for basalt, but the magnitude of doses in the surface water for the granite site are about 30-fold higher, because of the lower flow rate of river water assumed for the granite site. If the water travel time to the environment for the granite site is about 1,000 years, the maximum predicted radiation dose is from carbon-14. If the water travel time is as long as 10^5 years, a maximum dose rate of about 3×10^{-9} Sv/yr occurs in the surface water for the granite site, a factor of 3×10^4 below the radiation dose criterion.

9.10.7. Salt

The solubility-limited dissolution rates estimated in Table 9-5 are adopted for a generic repository in bedded salt, although it might be expected that the geochemical environment in the media adjacent to a salt repository could result in somewhat greater solubilities and dissolution rates than for basalt and granite. Also, there is some uncertainty about the applicability of the solubility-limited diffusion-convection analysis (cf. Section 5.7) to predict the dissolution of radioactive waste in a salt repository when water intrudes. Our analysis conservatively assumes that water intrusion, from some unexpected event, exposes all of the waste packages to moving groundwater within the first few thousand years after waste emplacement. It assumes that the result of water intrusion is to surround all of the waste with brine moving slowly through a rubble of undissolved salt.

The estimated radiation doses for a salt repository are shown in Figure 9-11. Here cesium-135 dominates the dose for water travel times

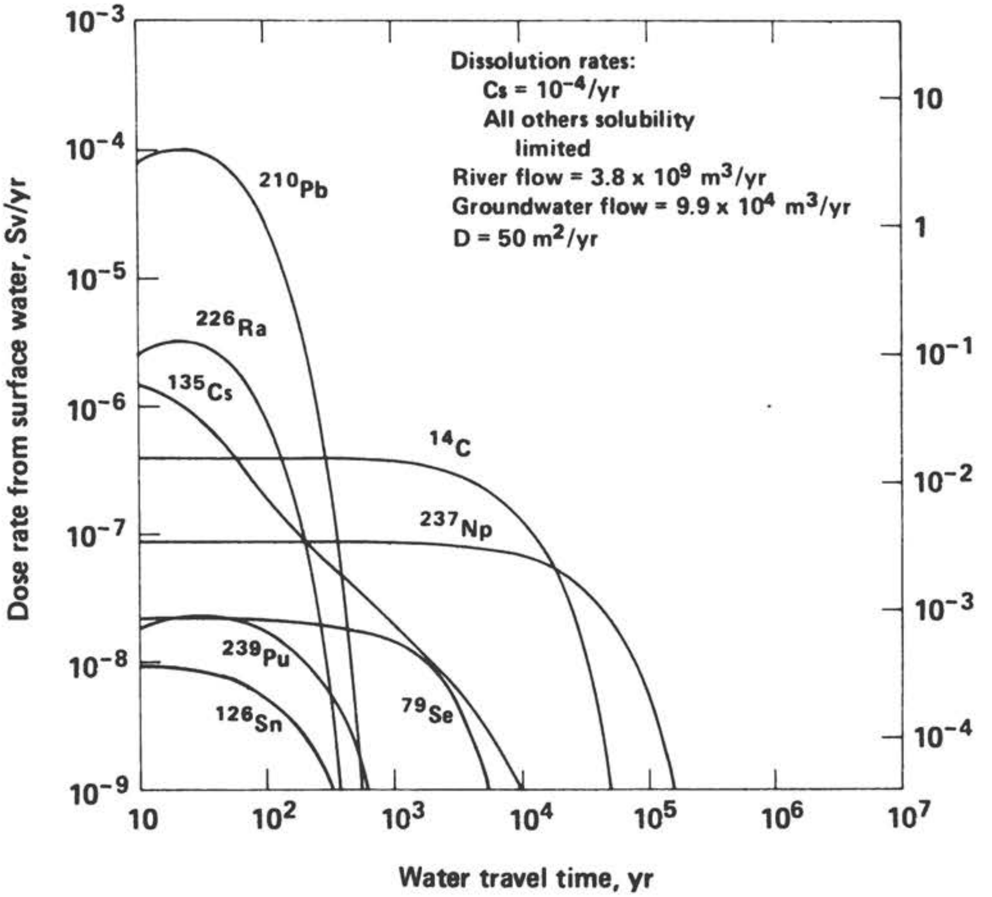

FIGURE 9-10 Individual radiation dose as a function of water travel time in granite: reprocessing waste from 10^5 Mg uranium fuel, solubility-limited dissolution.

greater than 10^5 years. It is apparent that determination of the appropriate dissolution rate for cesium-135 is of special importance for a salt repository. For the cesium retardation coefficient adopted here, cesium dispersion is not sufficient to result in the insensitivity of predicted doses to the dissolution rate of cesium.

The hydrologic parameters adopted for calculating the data in Figure 9-11 are those listed in Table 9-4 for a generic repository in bedded salt. As noted in Section 9.7.4, these estimates can be translated to estimates for a generic repository in domal salt by applying the hydrologic data for domal salt in Table 9-4.

Water intrusion into salt and the resulting hydrogeologic transport of radionuclides is not an expected event, as discussed in Section 9.7.4.

9.10.8. Tuff

Adopting the solubility-limited dissolution rates of Table 9-5, the calculated doses for a tuff repository at the Nevada Test Site are shown

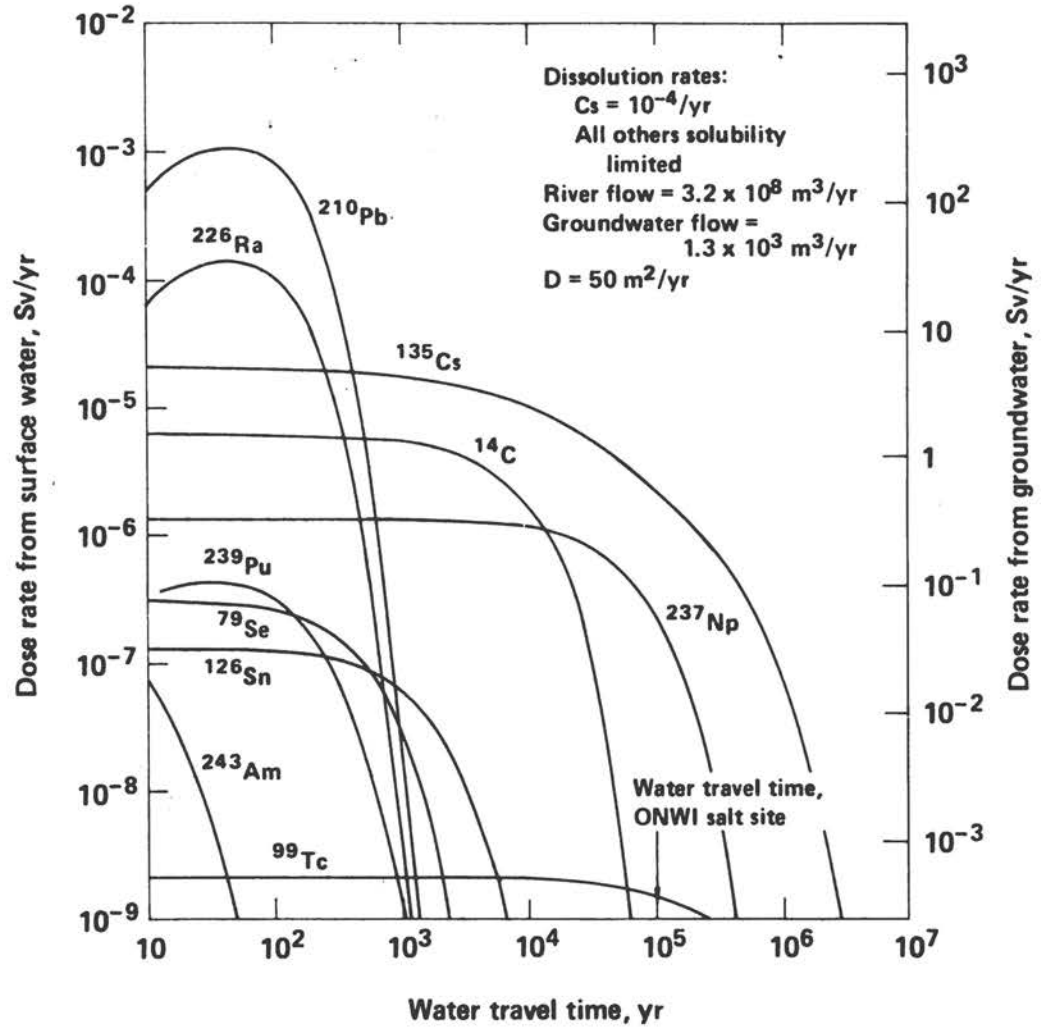

FIGURE 9-11 Individual radiation dose as a function of water travel time in salt: reprocessing waste from 10^5 Mg uranium fuel, solubility-limited dissolution.

in Figure 9-12. The relative values of the doses from individual radionuclides vary with water travel time in the same way as for the basalt and granite repositories. We have no estimates of groundwater flow rate for a tuff site, and it appears that there is no surface water into which potentially contaminated groundwater can discharge. Attention must be focused on possible use of the potentially contaminated groundwater by humans, for potable water and irrigation.

Tentatively adopting 4,300 years as the possible water travel time from a repository in saturated tuff to an off-site well, the radiation dose will be dominated by carbon-14 or, depending on its dispersion coefficient, by cesium-135. Adopting 2.1×10^4 years as the possible water travel time to the same off-site location from a repository in unsaturated tuff, the carbon-14 dose is attenuated so that it is

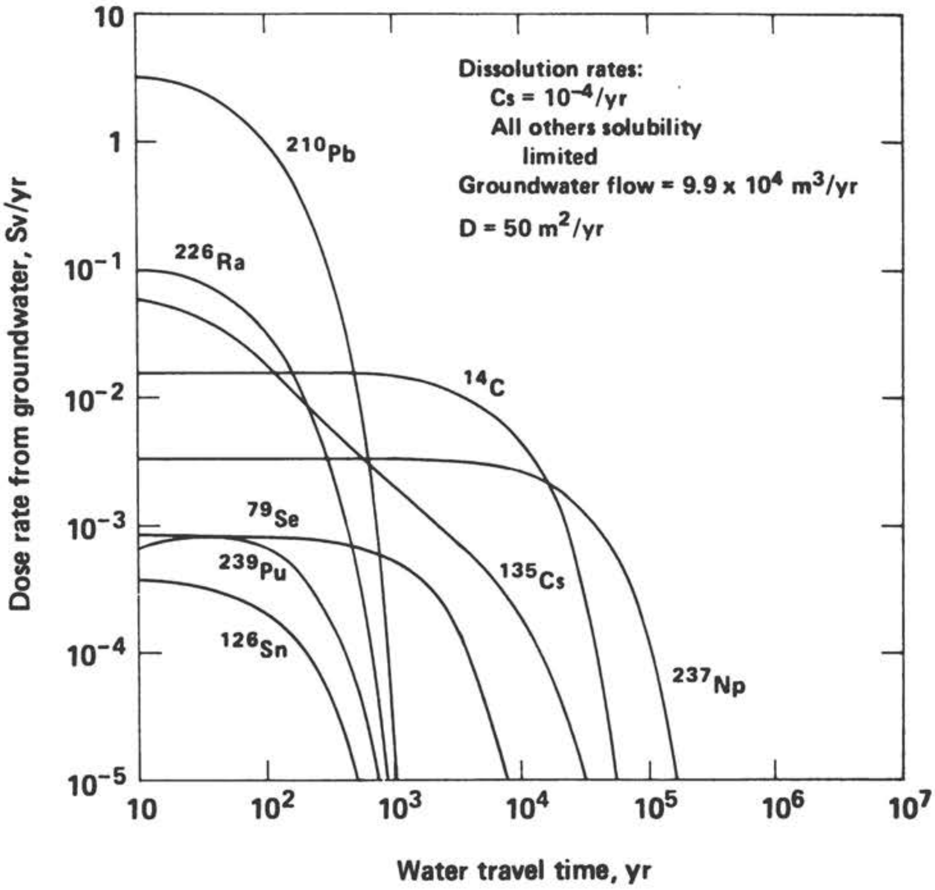

FIGURE 9-12 Individual radiation dose as a function of water travel time in tuff: reprocessing waste from 10^5 Mg uranium fuel, solubility-limited dissolution.

comparable with the predicted dose from neptunium-237. If the groundwater flow rate is within the range of flow rates considered in this study, the individual dose from carbon-14 in groundwater would be between one and two orders of magnitude greater than the performance criterion adopted for this study. The probability of future use of this contaminated groundwater must be taken into account, but the general arid area of the Nevada tuff site suggests that the potentially contaminated groundwater is likely to be used.

This performance analysis of a tuff repository illustrates one of the differences between adopting a performance criterion in terms of an individual dose or in terms of a population dose or radioactivity release limit, the latter appearing in EPA's proposed standard. Our calculations indicate that this possible tuff repository would meet the EPA 10,000-year release limits (cf. Table 8-1) at the 10-km location, assuming the numerical criteria proposed by the NRC staff (cf. Section 8.6), and assuming that those numerical criteria can be met by all of the radionuclides with releases above NRC's 0.1 percent threshold. Yet, with the parameters adopted for the calculations herein, a tuff repository may not meet the individual dose rate criterion. The EPA's proposed standard and the NRC's proposed numerical criteria are not concerned with the effects of water flow rates, nor are they concerned with concentrations and individual doses from radionuclides in groundwater and surface water.

Roseboom (in press) has made an overall performance analysis of a waste repository in unsaturated Nevada tuff, although his hydrologic data do not include the more recent data and estimates supplied by the USGS-DOE-Sandia team (L. D. Tyler, Sandia National Laboratories, personal communications to T. H. Pigford, 1982) for the purpose of the present study. The overall performance criterion adopted by Roseboom is the fractional release rate of radionuclides from waste into groundwater, and he adopted the limit of 10^{-5}/yr that appears in NRC's proposed regulation 10 CFR 60. On the basis of his estimates of the flow rate through the emplacement zone and his estimates of solubilities, Roseboom concludes that the NRC release rate criterion will be met and that the repository performance will be adequate. He does not consider the problems of meeting the NRC release rate criterion for some of the individual radionuclides, as discussed in Section 8.6.7, nor does he consider the radiation doses that could result if there were human use of the contaminated groundwater.

These different approaches and conclusions in the evaluation of a repository in Nevada tuff illustrate the need for a clearly defined and technically substantiated overall performance criterion for a geologic repository.

9.11. EFFECT OF WASTE-FORM DISSOLUTION RATES

As indicated in previous sections, most waste radionuclides that dissolve incongruently are less soluble than glass. Cesium is a possible exception due to its unusual mobility and solubility. The effect of low-solubility waste forms on radionuclide release rates is to decrease the number of radionuclides that may dissolve more slowly than the host until, in the limit, all waste products will be released congruently or diffuse out and dissolve faster than the host. This limiting condition probably occurs at waste-form dissolution rates around 10^{-9}/yr or 10^{-10}/yr (see Section 10.2).

If we choose a rate of 10^{-7}/yr as an intermediate and probably achievable rate for a variety of waste forms, the peak concentrations, calculated without solubility limits, for uranium, neptunium, plutonium, and tin should be reduced proportionately, i.e., by three orders of magnitude compared with the concentrations calculated at a rate of 10^{-4}/yr. Dose due to selenium may not be proportionately lowered, because of the difference in ionic size compared to the previously mentioned elements. The concentrations of radium-226 and lead-210 will be sharply reduced by low-solubility waste forms even though they are relatively unaffected initially by solubility limits. Their peak concentrations should be reduced by orders of magnitude as compared with those calculated without inclusion of solubility limits. The behavior of these elements is complex, for reasons that have been discussed earlier. However, when uranium and americium are retained in a low-solubility waste form, lead and radium should be similarly retained unless they diffuse out at rates greater than the 10^{-7}/yr rate assumed for the host.

If the waste form is assumed to have no effect on the diffusion of cesium and it dissolves incongruently at a rate as high as 10^{-4}/yr, it can become the principal contributor to the peak radiation dose at very short water travel times. If material transported by groundwater at an assumed travel time of 10 years or greater is diluted with 4 x 10^7 m^3/yr of additional surface water, the performance criterion will not be exceeded. This volume is small compared with the surface water flows assumed here for basalt, granite, and salt.

The potential advantages of a waste form that dissolves at a rate of 10^{-7}/yr are as follows:

- Because primary dependence for containment is on the waste form itself, validation of performance will depend largely on the measurement within the laboratory and in situ of the release rates of radionuclides from the waste form, together with a characterization of uncertainties in the nearby hydrology and the geochemical factors that can affect the integrity and dissolution rate of the waste form itself.
- The need to evaluate solubility limits for each of the important radionuclides is limited to the extent that they are used to provide additional barriers.
- The necessary dimensions of the controlled area can be smaller than is presently contemplated.
- The dilution factor necessary to ensure compliance with the performance criterion is reduced, especially for short water travel times.
- The number of sites that can serve as suitable repositories should increase.
- The overall degree of assurance of satisfactory long-term performance would be increased by provision of an additional resistance to dissolution.

Low-solubility waste forms have not yet been developed to the same extent as glass, and relatively little research has been carried out on dissolution rates. The crystalline structures of zirconates and titanates are particularly accommodating for a variety of waste products of interest. Other promising waste-form candidates are also mentioned in Chapter 5. In view of the potential benefits of such forms, we recommend continued, vigorous research on promising options.

9.12. RADIATION DOSES FOR A BASALT REPOSITORY LOADED WITH UNREPROCESSED SPENT FUEL

The calculated radiation doses for a basalt repository completely loaded with unreprocessed spent fuel are shown in Figure 9-13 for assumed congruent dissolution at a rate of 10^{-4}/yr and neglecting dispersion.

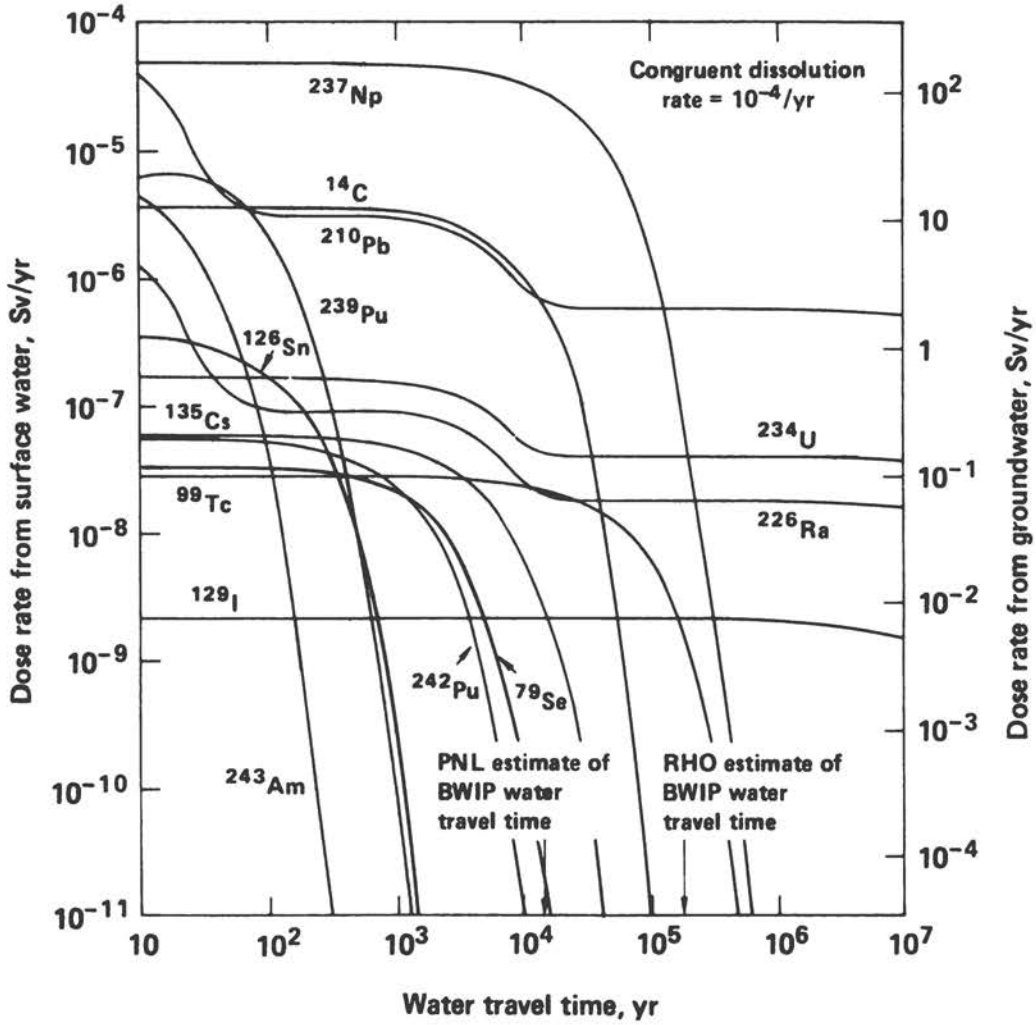

FIGURE 9-13 Individual radiation dose as a function of water travel time in basalt: unreprocessed spent fuel from 10^5 Mg uranium fuel, congruent dissolution, no dispersion.

In comparison with similar data in Figure 9-3 for a basalt repository loaded with reprocessing waste, several differences are apparent:

o The maximum dose rate from neptunium-237 is increased threefold because all of the plutonium-241 in the spent fuel is available to decay to americium-241 and thence to neptunium-237. Neptunium-237 remains the most important radionuclide for water travel times up to several hundred thousand years.

o The maximum dose rate from plutonium-239 increases 37-fold because all of the plutonium-239 in spent fuel is loaded into the repository. For reprocessing waste most of the plutonium-239 reaching the groundwater results from the decay of americium-243 initially in the waste. Plutonium-239 is important only in the near field for water travel times of a few hundred years or less.

o The maximum near-field dose from radium-226 increases fourfold because of the presence of additional uranium-234 and plutonium-238 in spent fuel. The maximum far-field dose from radium-226 increases 100-fold because of the greater amount of uranium-238 in spent fuel.

In estimating the solubility-limited dissolution rates for spent fuel, we assume that the solubilities for the stable species in groundwater are those listed in Table 9-5. For simplicity we neglect the larger waste surface area per unit mass of contained fission products for a spent fuel waste package as compared with the waste package containing reprocessing high-level waste. The consequence is that the fractional dissolution rate for spent fuel decreases for those solubility-limited radionuclides with greater initial inventory, but the resulting mass dissolution rate of the parent radionuclide is the same as for reprocessing waste. The predicted doses are shown in Figure 9-14. Because of solubility limits the calculated doses from neptunium-237 are the same for spent fuel as shown in Figure 9-9 for reprocessing waste, and this is also true for selenium-79, americium-241, and the other solubility-limited species within the high-level waste package.

Because uranium is solubility limited, radium-226 migrates appreciably from the repository only as a result of the migration of thorium-230, which is not at a high enough concentration to be solubility limited. Consequently, the higher initial uranium-234 of the spent fuel creates fourfold more thorium-230 and fourfold greater concentrations of radium-226 for water travel times less than about a thousand years.

Assuming that the release rate of cesium is the same for spent fuel as for reprocessing waste, the dose rate from cesium-135 will be the same. Laboratory data suggest a greater dissolution rate for cesium in spent fuel, but cesium dispersion, as illustrated in Figure 9-8, should make the cesium dose rate for spent fuel insensitive to release rate for water travel times greater than a few hundred years.

Because spent fuel is not reprocessed, each high-level waste package contains its proportion of the carbon-14 and iodine-129 in the repository. The ratio of package surface area to the inventory of each of these two radionuclides in each package results in no solubility limits on the release of these radionuclides. Their dissolution rates are likely to be determined by the kinetics of dissolution and restructuring of the UO_2 fuel. Assuming a fractional release rate of 10^{-4}/yr for carbon-14 and iodine-129, we predict that carbon-14 will be a relatively large contributor to the dose rates for groundwater travel times less than about 10^5 years, and iodine-129 will be the main contributor to radiation doses at longer times. Disposing of spent fuel as waste not only precludes the option of designing solubility-limited waste packages for carbon-14 and iodine-129, but it also precludes the options of nongeologic disposal of these radionuclides, as mentioned in Sections 9.10.2 and 9.10.3.

The differences between spent fuel and reprocessing waste, as illustrated here for a basalt repository, will apply as well to the other repository media.

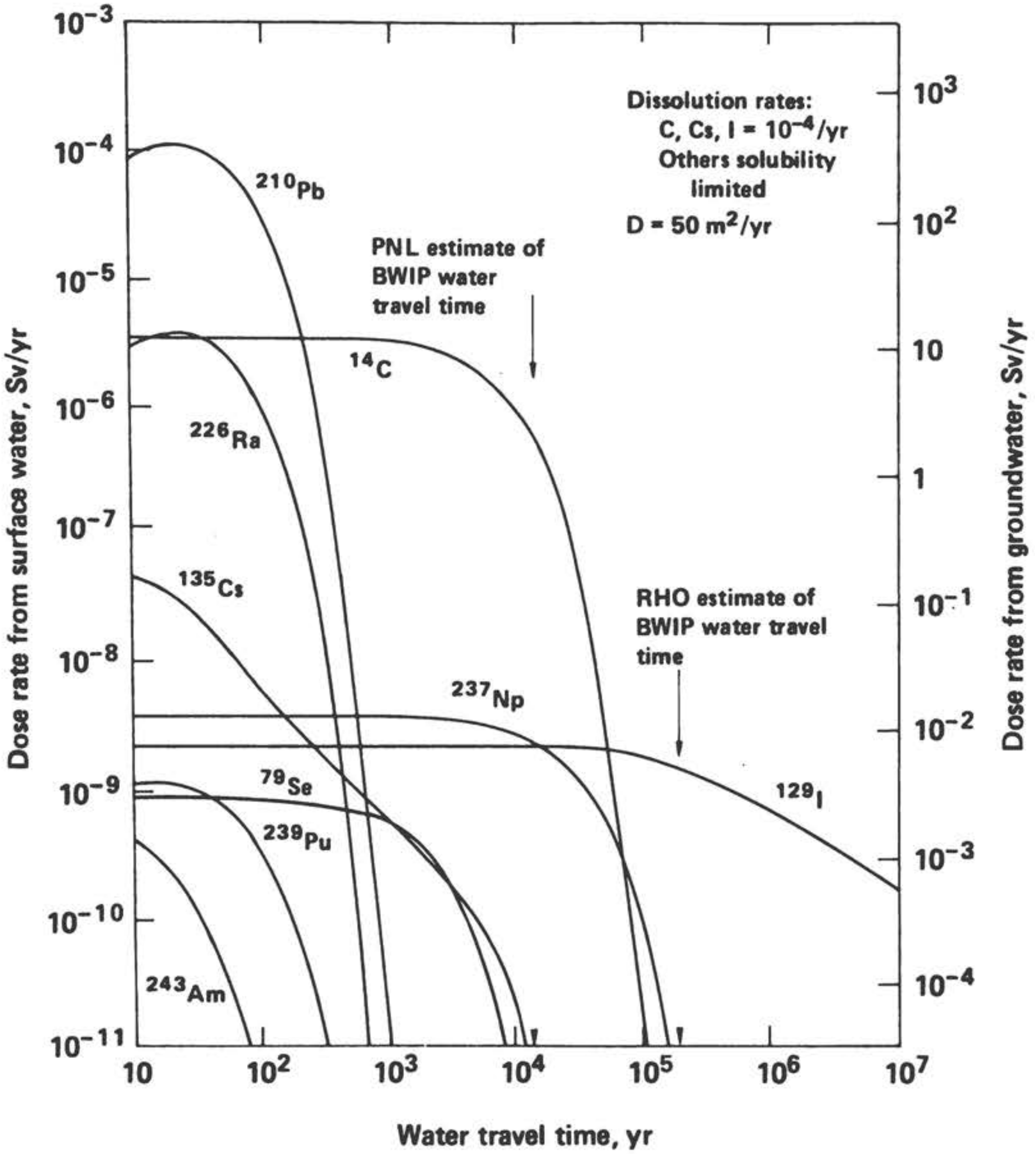

FIGURE 9-14 Individual radiation dose as a function of water travel time for unreprocessed spent fuel from 10^5 Mg uranium fuel, solubility-limited dissolution.

For the reasons outlined above, the reprocessing of spent fuel allows preparation of waste forms that will improve repository performance.

9.13. EFFECT OF REPOSITORY HEATING ON SOLUBILITY-LIMITED DISSOLUTION RATES AND RADIATION DOSES

Even if a corrosion-resistant overpack is included in the waste package to protect the waste forms from exposure to groundwater during the thermal period, we must still assume that some waste forms will be exposed because of statistically expected failures of some small fraction of the waste packages. A much greater number of waste packages can be exposed during the thermal period if the overpack is not used. The panel has sought data from the federal agencies and their contractors on the effect of repository heating on radionuclide release to the groundwater and resulting radiation doses. Although the U.S. Nuclear Regulatory Commission (1981, 1982) has concluded that temperature effects are important and should be avoided by a 1,000-year overpack, no calculations or experiments were available to justify these conclusions. In the absence of any data or analyses from these or other sources, we have made our own estimates of the effect of higher-temperature exposure on dissolution rates and repository performance.

When the repository temperatures are higher than ambient, solubility-limited dissolution rates can be increased because the higher temperatures increase both the solubilities and the diffusion coefficient in the liquid boundary layer, and because the increased local velocities of groundwater from thermal convection flow can decrease the boundary layer thickness. The effects of temperature on the solubility of silica and on the diffusion coefficient in the liquid are shown in Table 9-6.

Temperatures affecting solubilities during the thermal period are the time-dependent rock temperatures at the emplacement-hole surface, as shown in Figures 6-12 through 6-18. The panel has sought data on the effect of rock heating on flow rates and local velocities of groundwater. Only the BWIP basalt project has supplied analyses of the thermal convective flows though a repository (D. J. Brown, Rockwell Hanford Operations, personal communication to T. H. Pigford, 1982). The results, in terms of time-dependent temperatures (Altenhofen 1981), potential gradients, pore velocities, and volumetric flow rates through the repository rock, are shown in Table 9-7. In calculating these flow data, it was assumed that the entire repository is loaded with 10-year-old spent fuel and that it is saturated with water immediately after closure. In calculating the maximum rock temperatures for reprocessing waste, it was assumed that the wastes were emplaced in the repository 15 years after reactor discharge.

The data show that during the thermal period the thermally induced potentiometric gradient far outweighs the ambient gradient, and the local pore velocities in the host rock increase from the ambient value of 3.2×10^{-3} m/yr to as high as 0.64 m/yr. The time for water to travel to the edge of the host rock decreases from 6,600 years to 33 years. Both of these host-rock travel times are small compared with the total travel time to the river, estimated to be 1.9×10^5 years without repository heating and 2.0×10^5 years with heating. Heating is calculated (D. J. Brown, Rockwell Hanford Operations, personal communication to T. H. Pigford, 1982) to increase the water travel time in the surrounding media because buoyancy in the region above the repository forces the exterior flow into strata of lower permeability.

The remaining data in Table 9-7 were calculated by the panel. The ambient groundwater velocity of 3×10^{-3} m/yr is considerably less

TABLE 9-7 Calculated Fractional Release Rate During Thermal Period for Basalt

	Time After Waste Emplacement					
	Ambient, Prior to Repository Construction, or $>5 \times 10^3$ Years	5 Years	100 Years	500 Years	1,000 Years	5,000 Years
Maximum rock temperature (°C)						
Spent fuel	57	185	160	135	120	90
Waste from fuel reprocessing	57	250	130	90	85	80
Potential gradient within repository	0.001	0.06	0.05	0.04	0.03	0.02
Pore velocity (m/yr)	0.0032	0.64	0.41	0.28	0.20	0.09
Water travel time to repository host rock boundary (yr)[a]	3.1×10^3	1.6×10^1	2.4×10^1	3.6×10^1	5.0	1.1×10^2
Water travel time to river (yr)	1.9×10^5		2.0×10^5			
Volumetric flow rate through repository (m^3/yr)	1.7×10^1	3.3×10^3	2.1×10^3	1.4×10^3	1.0×10^3	4.9×10^2
Volumetric flow rate past waste packages (m^3/yr)	1.2	2.3×10^1	1.5×10^2	1.0×10^2	7.2×10^1	3.4×10^1
Fractional release rate of SiO_2[b] (yr^{-1})	1.7×10^{-6}	7.4×10^{-5}	1.0×10^{-5}	4.1×10^{-6}	3.6×10^{-6}	3.2×10^{-6}
Fractional release rate at indicated temperature relative to fractional release rate at ambient temperature	1	44	6.2	2.4	2.1	1.9

[a] Calculated for 10 m from waste emplacement to top of emplacement rock.

[b] Neglects presence of dissolved SiO_2 in BWIP groundwater. Controlled by solubility and diffusion, estimated from Equation (2) of Chapter 5.

SOURCE: Hydrologic data and temperatures supplied by D. J. Brown (Rockwell Hanford Operations, personal communication to T. H. Pigford, 1982) and R. A. Deju (Rockwell Hanford Operations, personal communication to T. H. Pigford, 1982).

than the value of 1 m/yr assumed in calculating the solubility-limited dissolution rates listed in Table 9-5. At this low velocity, Equation (1) in Chapter 5 for the dissolution rate with simultaneous diffusion and convection is not valid (as explained in Chapter 5). For a better estimate we must use Equation (2) for diffusion-controlled transport through the concentration boundary layer. An even better estimate would result from the use of the transient solution (Chambré et al. 1982c) for mass transfer during the thermal period, because there may not be sufficient time to establish the steady state conditions assumed in Equation (2). However, such calculations are beyond the scope of the present study. Because the ambient temperature of basalt is greater than that assumed in Table 9-5, the resulting fractional release rate of silica is about 50 percent greater than that estimated in Table 9-5. For the purpose of illustration, we will assume that the heats of solution of the solubility-limited radioelements are the same as for silica. If so, the solubility-limited release rates for ambient basalt adopted for the dose calculations of Figure 9-8 are about 50 percent too low.

Similarly, we can estimate the fractional release rates for silica in the basalt repository at the different times during repository heating, with the results shown in Table 9-7. From these results, exposure of the waste form to groundwater during the thermal period at the maximum 5-year temperature of 250°C could result in a silica dissolution rate about 40 times greater than the rate at the ambient temperature of 57°C, or about 70 times greater than the dissolution rate at 20°C listed in Table 9-5. After 100 years, as the rock temperature at the emplacement hole decreases, the silica dissolution rate decreases to about sixfold greater than the ambient value.

As a first approximation, we assume that the same proportional increase applies to all of the low-solubility radioelements, with the consequence that exposure of all the waste during the thermal period could cause a 40-fold increase in the calculated near-field doses, where the main effect is the greater dissolution rate resulting from greater solubilities and diffusion rates. For the low water velocities in the basalt host rock, thermally affected flow velocities have little effect on the dissolution rate, because at these low velocities transport through the liquid boundary layer is controlled by molecular diffusion, and transport through the boundary layer by convection is relatively unimportant.

The effect of repository heating on the doses from radionuclides reaching surface water is not so great because of dispersion during transport. If the temperature were to cause a 40-fold greater dissolution rate for a few hundred years, there would be a 40-fold increase in the concentration of each radionuclide at the leading edge of its chromatographic band in the field near the repository. This is a rather short-duration increase in the near-field concentration for the time periods of hydrogeologic transport. Because it occurs during a time interval much smaller than the radionuclide travel time to the biosphere, the concentration spike will almost completely disappear because of dispersion by the time it reaches the biosphere. For example, a spike in the concentration of neptunium-237 lasting for a few hundred years would be attenuated to a few percent of the initial amplitude of the concentration spike after a water travel time of 10^5 years, assuming a dispersion constant of 50 m^2/yr (Sato et al. 1982). Thus, what can be an appreciable concentration increase in the near field of the repository may result in a rather small effect when it reaches the biosphere.

The above calculations have been based on the temperature-dependent solubility of amorphous silica. Langmuir's (1978) compilation of thermodynamic data on solid and aqueous uranium species as a function of temperature, Eh, and pH provides a basis for estimating the solubility of uranium at the elevated temperatures during the period of repository heating. The expected dominant species will be either UO_2 in silica-free groundwater or $USiO_4$ in the the silica-rich basalt groundwater. Using these data together with estimates of the temperature-dependent alkalinity (pH) of the basalt system (M. K. Smith et al. 1980), Wood (1980) estimates that the effective solubility of uranium in BWIP groundwater will actually decrease as the temperature increases from 65°C to 150°C, as shown in Figure 9-15. Wood and Rai (1981) suggest that the temperature dependence of uranium solubility deduced from Langmuir's compilation may be representative of the other actinide species. If so, the dissolution rates of the actinides will not change as much with temperature as estimated above for silica.

Goodwin (1980) in Wuschke et al. (1981) has estimated the solubilities of uranium in granite groundwater at 25°C, 100°C, and 150°C, with the oxidation potential determined by the magnetite-hematite equilibrium at a pH of 7. His estimates, shown in Figure 9-15, indicate a fivefold increase in solubility from 25°C to 100°C, and a small decrease from 100°C to 150°C, again indicating that the temperature effect on uranium dissolution rate may not be as great as has been predicted for silica.

These calculations suggest that the performance of the repository if waste is exposed during the thermal period is amenable to calculation, provided data are obtained on temperature-dependent solubilities. They show that the effect of thermally induced flow on the dissolution rate may not be important in a repository wherein the flow velocities are normally quite low. They show that appreciable concentration increases can occur in the near field of the repository during the thermal period, but that for long water transport times to the biosphere the concentration increases may be relatively small. Although there are many uncertainties in these estimates, it may be easier and more cost effective to clear up these uncertainties by careful experiments and analyses, especially on temperature-dependent solubilities, than to provide the 1,000-year corrosion-resistant overpack proposed by the U.S. Nuclear Regulatory Commission (1981, 1982) to reduce the uncertainties in estimating repository performance during the thermal period.

9.14. EFFECT OF STORAGE TIME BEFORE EMPLACEMENT

The calculated rock temperatures in Chapter 6 and in Table 9-7 are based on the assumption that waste is emplaced in the repository within 10 to

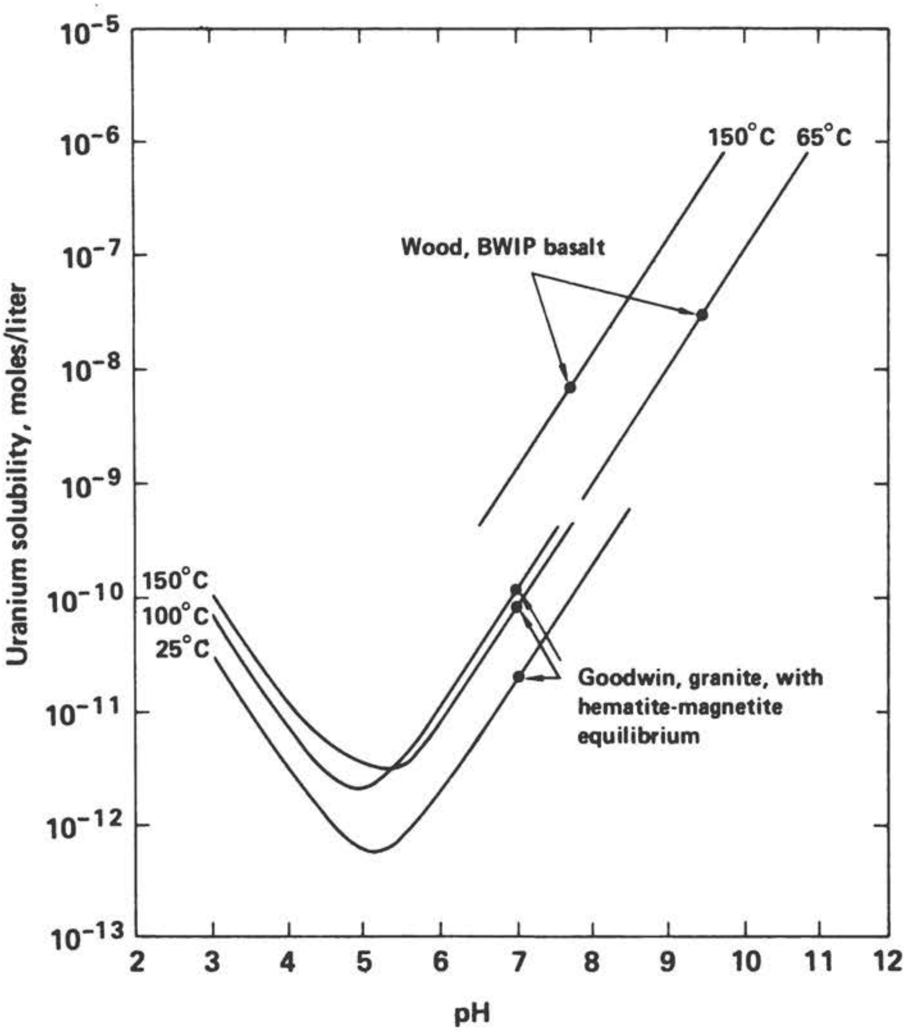

FIGURE 9-15 Uranium solubility as a function of pH and temperature. Sources: Wood (1980), Goodwin (1980).

15 years after reactor discharge. These are about the shortest preemplacement cooling times contemplated for the steady state fuel cycle. Because of the accumulating inventory of unreprocessed spent fuel at reactors, the high-level waste initially emplaced in a repository will have cooled for decades, and the temperatures will be much lower than those listed in Chapter 6 and in Table 9-7. After some decades of repository loading, it is possible that the wastes available for emplacement will be of the 10- to 15-year age assumed in these temperature calculations. If more detailed analysis and experiment indicate that lower temperatures are then warranted, additional preemplacement cooling is a reasonable possibility.

An additional 25 years of cooling will halve the heat generation rate, and it will almost halve the temperature rise in the repository, depending on the change of thermal conductivity of the rock with temperature. This could reduce the temporal maximum rock temperatures to well under 200°C for basalt and under 100°C in salt, with the other rock media falling within this range. The heat generation rate in reprocessing waste can be reduced 20-fold by storing for a few hundred years.

This study has raised several questions concerning the possible effects of temperature on the long-term performance of a geologic repository. These include physical deterioration of the waste, possible increases in the corrosion rate of the waste-package container and the waste form, increases in the flow velocity near the waste, and changes in the geochemical conditions of the groundwater. Nearer-term temperature effects of practical importance are thermally induced stresses in the rock, thermal stresses in the waste package during resaturation, and possible retrievability of the waste. All of these issues need to be explored to verify the present designs and to assess the need for technological fixes, such as NRC's proposed 1,000-year container. From the analyses that we review herein, there is no clear indication that the repository performance at these temperatures will necessarily be unsatisfactory. However, extended preemplacement cooling is a significant alternative that will be available to the designer, and even later to the repository operator, if reduction in temperature is found to be necessary.

9.15. EFFECT OF PLUTONIUM RECYCLE TO LIGHT-WATER AND BREEDER REACTORS

Recycling plutonium will significantly increase the amounts of americium and curium isotopes in the discharge fuel and in the high-level wastes from reprocessing. Additional transuranic wastes, mainly containing plutonium isotopes, will be created when fabricating fuel of recycled plutonium. We have estimated the amounts of these isotopes relative to the amounts in waste from reprocessing first-cycle light-water reactor fuel.

The most important differences are in the sources of radium-226, formed by the decay chain of Figure 9-2, and its more toxic daughter lead-210. The radium-226 and lead-210 from these transient sources, not including the longer-lived uranium-238, will appear in the environment for water travel times of about 10,000 years or less for congruent dissolution (cf. Figures 9-3 through 9-6) and 1,000 years or less for solubility-limited dissolution (cf. Figures 9-9 through 9-12). During these periods the doses from radium-226 and lead-210 are estimated to be about threefold greater for plutonium recycle fuel than for nonrecycle fuel. Americium-243 will be about sixfold greater and americium-241 about threefold greater for the plutonium recycle fuel in light-water reactors. Carbon-14 will be slightly less for plutonium recycle. None of these differences are large enough to be significant in repository performance analysis.

If the high-level and transuranic wastes are derived from the reprocessing and recycle of fast-breeder fuel, the increase in actinides will be similar to, but less than, the increase described above for recycle in light-water reactors.

9.16. EFFECT OF URANIUM-THORIUM FUELING

In the uranium-thorium fuel cycle, reactors are fueled with thorium and enriched uranium-235, as in the high-temperature gas-cooled reactor at Fort Saint Vrain, Colorado. If this fuel cycle is closed by reprocessing, the recovered uranium will be recycled to fuel fabrication and the remaining actinides will appear in the high-level and transuranic waste. This fuel cycle produces about twice the amount of neptunium-237 but much less americium, curium, and long-lived plutonium than do the uranium fuel cycles (Benedict et al. 1981). However, because all of the plutonium-238 in the discharge fuel is expected to appear in the high-level waste, and because of the relatively large amount of uranium-234 resulting from neutron capture in bred uranium-233, the amounts of these transient sources of radium-226 and lead-210 that can be formed are about 20-fold greater than in the uranium fuel cycle with fuel reprocessing, assuming the same amount of electrical energy generation. The transient radium-226 and lead-210 will be about equal in magnitude to the amounts of these radionuclides in unreprocessed uranium fuel. The water travel times that can result in relatively important doses from radium-226 and lead-210 from these transient sources are discussed in Section 9.15.

9.17. CARBON-14 WASTE FROM GRAPHITE FUEL

In graphite reactors, carbon-14 is produced by neutron absorption in carbon-13 and in nitrogen impurities in the graphite. In the high-temperature gas-cooled reactor (HTGR), most of the reactor graphite is in the fuel elements, and the carbon-14 must be recovered when discharge fuel is reprocessed. The recovered carbon-14 will be highly diluted with the carbon-12 from the graphite fuel, so the volumes of the carbon-14 waste will be relatively large. The amount of carbon-14 in these wastes per unit of electrical energy produced is estimated to be from 3 to 10 times greater than in the wastes from reprocessing light-water reactor fuel (Nuclear Energy Agency 1980, Benedict et al. 1981). The highly dilute carbon-14 from HTGR reprocessing would not be amenable to loading in a small number of waste packages to reduce the surface-to-inventory ratio and the solubility-limited dissolution rate, as is discussed in Section 9.10.2 for the more concentrated carbon-14 from light-water reactors. Thus, a repository hypothetically loaded with HTGR waste, from the same amount of energy production as from 10^5 Mg of uranium fuel, could result in individual dose rates from carbon-14 about 3 to 10 times greater than shown in Figures 9-3 through 9-6 for congruent dissolution, assuming a fractional dissolution rate for carbon-14 of 10^{-4}/yr.

9.18. KRYPTON-85 AND TRITIUM WASTE

If unreprocessed discharge fuel is disposed of in a geologic repository, it will carry with it about 2×10^{16} Bq of krypton-85 if the repository is fully loaded with 10-year-old discharged fuel. If the discharged fuel is first reprocessed, the U.S. Environmental Protection Agency requires the recovery of krypton-85 from commercial fuel reprocessing plants, and it can be assumed that the recovered krypton-85 would be stored in the repository, perhaps in cylinders of compressed gas. Considering the actual age of the fuel to be reprocessed, much of this krypton decay will have occurred before emplacement in a repository. This is more of an operational issue rather than one of long-term confinement. It is not clear that a geologic repository offers any special advantage for the storage of recovered krypton.

Similarly, the amount of fission-product tritium in 10-year-old waste from 10^5 Mg of uranium fuel will be about 1.5×10^{18} Bq. There is presently no requirement that tritium be recovered as a waste from fuel reprocessing, but tritium recovery appears in some conceptual flowsheets for future reprocessing plants. If recovered, the tritium might typically be in the form of tritiated water absorbed on a molecular sieve or on anhydrous $CaSO_4$ (Benedict et al. 1981). Because tritium's half-life is only 12.3 years, the storage of separated tritium does not necessarily require its disposal in a geologic repository.

9.19. EFFECT OF VARIATIONS AND UNCERTAINTIES IN WATER TRAVEL TIME

The calculated doses shown in Figures 9-8 through 9-12 show that radiation doses are relatively insensitive to water travel time for water travel times in the range of a few hundred to a few thousand years for basalt, granite, and tuff and up to about 10^5 years for a salt site. The greatest reduction in dose occurs when water travel time is increased to about 10^5 years for basalt, granite, and tuff and to about 10^6 years for a salt site. For the basalt and salt sites the expected water travel times range from about 10^4 to 10^6 years, with means at about 1.9×10^5 years estimated by Rockwell Hanford Operations (D. J. Brown and R. A. Deju, personal communications to T. H. Pigford, 1982) and 1.5×10^4 years estimated by Dove et al. (1982). In the one-dimensional calculations used in this study, it has been assumed that there is a single travel time for water to reach the environment after contacting the waste form. In reality, there is a spectrum of travel times for the contaminated groundwater, distributed over the many different pathways that finally lead to the biosphere. If the spread in water travel times is greater than the time for all of a given radionuclide to be dissolved, then the spread of water travel times can lengthen the time over which the radionuclide will be discharged to surface water and can decrease the maximum concentration of the radionuclide in the surface water. On the other hand, if the spread of water travel times is smaller than the time constant for waste dissolution, then the spread of water travel times may not appreciably affect the peak concentrations of radionuclides in surface water.

If we adopt the range of travel times in Table 9-4 as indicating the spread over the different flow pathways, then this spread would be expected to reduce the peak concentration of cesium-135 in surface water, because the dissolution time constant, or leach time, of cesium may be as short as a few thousand years. However, the spread would not be expected to significantly affect the peak concentrations of the solubility-limited radionuclides because their estimated leach times are of the order of millions to billions of years.

The effect of the spread of water travel time should be calculated for the individual radionuclides when adequate data on the hydrology of specific sites become available.

As discussed in Section 9.4, further uncertainties in these estimates of long-term performance result from the assumption that hydrogeologic properties of the repository sites will not change markedly over the time periods considered in these calculations.

9.20. EFFECT OF UNCERTAINTIES IN SOLUBILITIES

The analyses presented herein demonstrate that, for a waste-form matrix dissolution rate of 10^{-4}/yr, solubility limits for all important radionuclides, other than cesium-135, may be the most important of the physical phenomena that limit radiation doses from repositories. For a waste incorporating separated iodine-129, it is only the solubility that limits the total release of iodine-129. Groundwater transport and sorption contribute essentially nothing. Similarly, it is mainly the low solubility of uranium that limits the long-term release of radium-226.

Whereas the calculation of solubility-limited transport is on clear physical grounds, it introduces important uncertainties due to (1) uncertainties in the solubilities and (2) the assumption that only dissolved radionuclides are transported by groundwater. The solubilities for many of the important radioelements are sensitively affected by the local oxidizing conditions, the acidity, and the presence of complexing agents, and they may be affected by the other dissolved constituents. The uncertainties in these solubility data have been discussed in Chapter 7. A thorough study of solubilities is warranted, both to understand the uncertainties in solubilities better and to reduce them.

The radionuclides transported by groundwater are not necessarily limited to dissolved complexed and noncomplexed species. For such low-solubility species the presence of radionuclides in colloidal form, or in otherwise suspended forms, can increase or even decrease the rates of release to groundwater and the rate of transport of radionuclides toward the biosphere, as discussed in Sections 5.3.2, 5.6.7, and 5.7.4.

The uncertainties in the solubilities of the low-solubility radionuclides have been discussed by K. Krauskopf (Stanford University, personal communication, 1982), who has suggested probable values as well as ranges of solubilities for individual elements (cf. Table 7-1). The effect of the range of neptunium solubilities on the calculated radiation dose from neptunium-237 is shown in Figure 9-16. A solubility tenfold lower than the most probable value of 1 ppb may be more

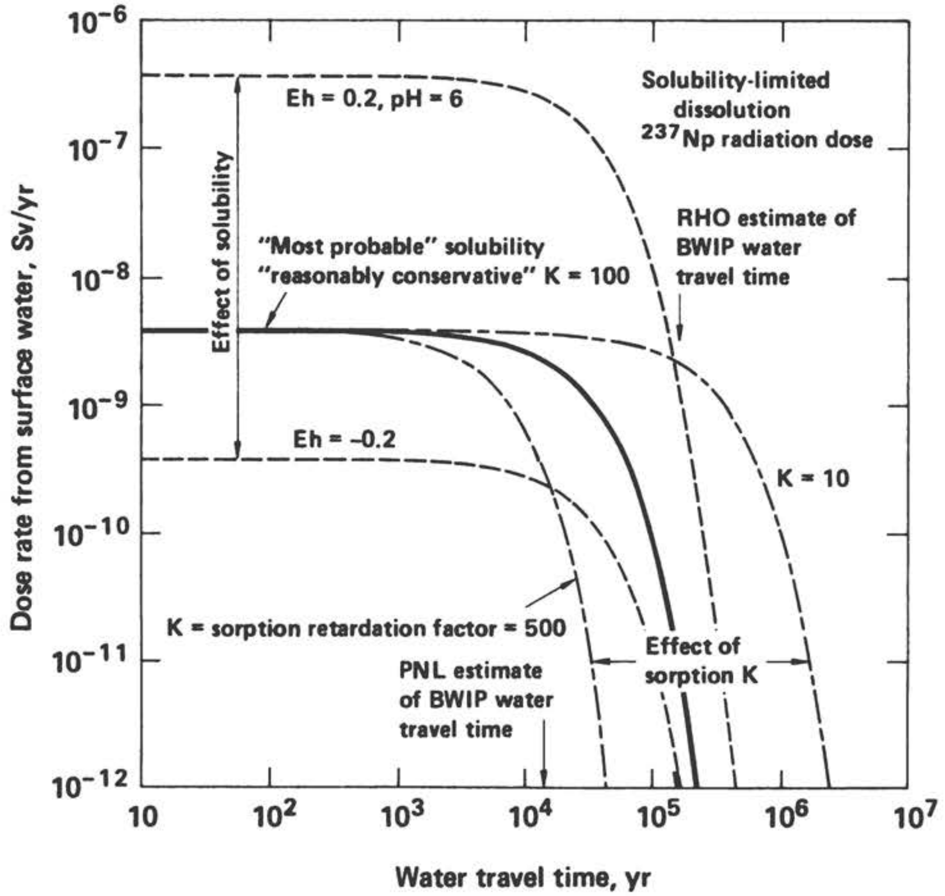

FIGURE 9-16 Individual radiation dose from neptunium-237 as a function of water travel time in basalt, showing the effect of uncertainties in solubilities and retardation: reprocessing waste from 10^5 Mg uranium fuel, solubility-limited dissolution.

representative of the ambient reducing conditions of basalt. For slightly oxidizing conditions (Eh = 0.2), as might occur during the time period of water resaturation of the repository and for some time thereafter, the neptunium solubility could increase one to two orders of magnitude above the most probable value.

If these solubility changes persist over very long periods of time, comparable to the radionuclide travel times implied in Figure 9-16, the radiation doses would increase or decrease proportionally. Such long-term effects are assumed in calculating the data in Figure 9-15. If they persist for only a few hundred years, dispersion would result in a much smaller increase in radiation dose. This is similar to the effect of thermally induced changes in dissolution rate discussed in Section 9.13.

9.21. EFFECT OF UNCERTAINTIES IN RETARDATION PROPERTIES

K. Krauskopf (Stanford University, personal communication, 1982) has discussed the uncertainties in retardation properties for the different radioelements in various rock media. The two principal areas of uncertainty are (1) the sorption equilibrium constants, i.e., the K_d values, and (2) the effective porosity of the sorbing media between the repository and the biosphere. Our performance calculations have been based on Krauskopf's "reasonably conservative" values of retardation coefficients, listed in Table 7-1. He also lists for each sorbing element the range of sorption retardation coefficients.

The effect of this range on the predicted radiation doses from neptunium-237 is shown in Figure 9-17. The effect of the possible tenfold decrease in the retardation coefficient is to extend the time period of the neptunium-237 dose beyond the mean water travel time to the biosphere, as predicted by BWIP. Consequently, it is important to resolve the uncertainties in the retardation coefficient for neptunium. A possible fivefold increase in the retardation coefficient would increase the margin between the time for neptunium-237 to decay and the mean water travel time estimated by BWIP. Among the radionuclides indicated as important in Figures 9-9 through 9-12, neptunium and cesium are the most important with respect to retardation properties, followed by selenium.

Although much attention has been given to variations in the values of the sorption equilibrium constants, a comparison between Krauskopf's predicted retardation coefficients for basalt and those recommended by BWIP indicates that the greatest difference lies in the effective porosity of the media between the repository and the biosphere. Whereas Krauskopf conservatively adopts an effective porosity of 20 percent, BWIP (D. J. Brown, Rockwell Hanford Operations, personal communication to T. H. Pigford, 1982) suggests a value of 1 percent based on site data. For the same distribution, this results in 25-fold greater values of BWIP's predicted retardation coefficients, and the calculated curves of Figure 9-15 would be shifted to water travel times 25-fold lower, as shown in Figure 9-17. Even with the lower equilibrium constants implied by Krauskopf's lower neptunium retardation coefficient of 10, neptunium would decay before it reaches the biosphere.

Although the panel adopts the data recommended by Krauskopf for the purpose of its evaluation, this comparison shows the importance of resolving the hydrological properties that enter into the calculation of retardation coefficients.

9.22. EFFECT OF FRACTURED MEDIA ON RADIONUCLIDE TRANSPORT

The performance calculations presented herein have been carried out for one-dimensional transport through equivalent porous media, assuming local equilibrium between dissolved radionuclides and those sorbed on the rock. Neretnieks (1982) has pointed out that in fractured rock the flow is predominantly through planarlike fractures, with radionuclides in the groundwater diffusing into and out of micropores penetrating the fracture surfaces. Because of the appreciable time for diffusion into and out of the micropores as the chromatographic band progress through the fractures, the width of the band of radionuclides can be appreciably increased. If most of the sorption is within the micropores rather than at the fracture surfaces, the leading edge of the band can progress more

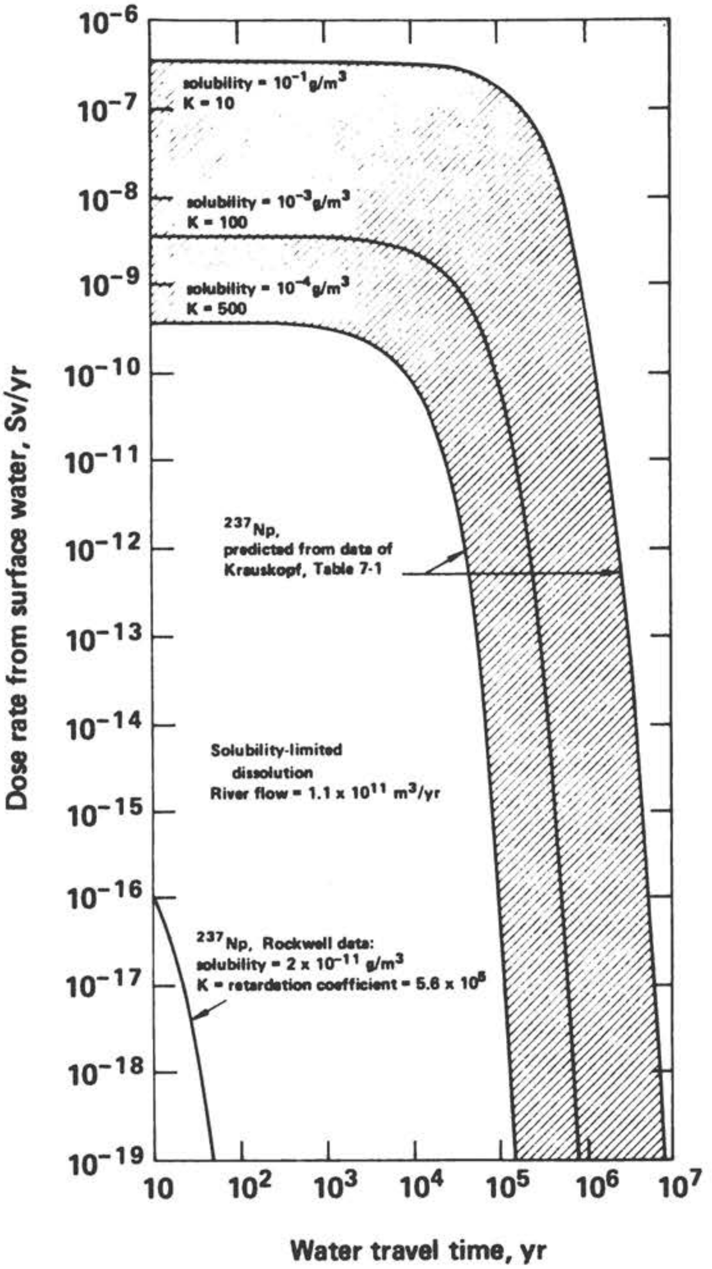

FIGURE 9-17 Individual radiation dose from neptunium-237 as a function of water travel time in basalt, showing the effect of Rockwell data on solubility and retardation: reprocessing waste from 10^5 Mg uranium fuel, solubility-limited dissolution.

rapidly than in the equivalent porous medium. Such fracture-flow transport offers a mechanism for sorbing radionuclides to reach the biosphere sooner than in porous advective transport, and it offers a mechanism for spreading the radionuclide band and attenuating the concentrations.

Neretnieks (1982) and Kanki et al. (1981) have provided mathematical analyses of fracture-flow transport. Kanki has pointed out that the most important effect is to reduce the maximum concentrations and dose rates in groundwater and surface water, qualitatively similar to the effect of a large axial dispersion constant.

Fracture-flow transport may be important in reducing the maximum doses for geologic repositories in fractured media and should be taken into account in estimating the probable radiation doses, and uncertainties therein, for the geologic repositories.

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10

NATURAL ANALOGS RELEVANT TO GEOLOGIC DISPOSAL

10.1. INTRODUCTION

The relevance of a comparison of nuclear waste deposits with natural or man-made analogs can be questioned on the basis that the differences may be far more important than the similarities. Nevertheless, to the extent that it is possible to make realistic estimates of releases from natural sources and to stipulate that nuclear repositories shall not exceed these releases, the analogy with ore bodies is useful, particularly in the absence of relevant long-term field experience with actual waste deposits. If differences in release, transport, and biological uptake mechanisms can be properly taken into account, statutory release limits that can be presented as a fraction of present contributions to human radiation exposure from ore deposits or generalized crustal abundances will probably be more widely understood than standards that refer only to peak exposures.

In addition, studies of the dissolution and leaching rates of minerals and glasses under field conditions may be uniquely relevant to the problem of long-term waste storage. They are consistently lower than rates measured in the laboratory, sometimes by several orders of magnitude. These differences are presumably due to processes not readily studied by short-term experiments in the laboratory but that are rate controlling in the long term (e.g., volume diffusion, surface film diffusion, or solubility-limited transport).

10.2. DISPERSION OF URANIUM AND RADIUM IN NATURE

Several reports have been published dealing with the dispersion of uranium and radium from ores and from trace quantities in a variety of host rocks.

In a Pacific Northwest Laboratory report (Wick and Cloninger 1980), potential hazards from a spent fuel depository are compared with those due to natural uranium deposits. These comparisons are made on the basis of intrinsic characteristics (inventory, depth, hydrology, geographic distribution) and on calculations or radiological consequences based on specific transport models. In terms of inventory and average uranium concentration, modestly large ore bodies and waste deposits may be considered comparable. Thus, the authors describe a nominal repository as roughly equivalent to an ore body 3.8 m thick with an average concentration of 0.16 percent uranium extending over 3.13 square miles. However, there are important differences in detail; e.g., most U.S. deposits occur in permeable aquifers, whereas repository locations will presumably be located in regions with much more favorable hydrology.

Their calculations of the biological hazards demonstrate that the natural ore problem is dominated by radium-226 and its daughters. By comparison, uranium is a minor hazard. The radium-226 concentration in U.S. groundwater varies from 3.7 x 10^{-3} Bq/l in the west to 3.7 x 10^{-2} Bq/l in parts of Arkansas and southern Missouri. Various radioactive springs contain several orders of magnitude more radium-226 than the general background.

In the calculations it was assumed that fractional dissolution rates in ore bodies vary from 10^{-7}/yr to 10^{-8}/yr. In calculations on reference waste repositories, these rates were varied from 10^{-2}/yr to 10^{-6}/yr. Groundwater travel times were varied from 500 to 10^{6} years in a reference granite repository, 500 to 3.7 x 10^{5} years in a reference salt repository, and 500 to as much as 1.5 x 10^{5} years for two reference ore bodies. Based on the results of these calculations, the report concludes that a reasonably constructed repository presents no greater hazard than a large ore deposit.

Williams (1980) attempts to estimate the contribution of hazardous radioactivity from unmined ore bodies. The chief result is that radium-226, the principal hazard, is calculated to be released to the biosphere at a fractional rate of 3 x 10^{-8}/yr. The corresponding uranium release is a factor of 10 less. An estimated number of 230 fatal cancers in 10^{4} years is reported for a repository containing waste from 10^{4} Mg of uranium fuel.

The same report also derives experimental uranium and radium releases from poorly documented premining data published in environmental impact statements filed for three commercially minable uranium ore bodies in Wyoming. The conclusion appears to be that these deposits lose 10^{-6} fraction of contained uranium per year and 10^{-4} fraction of radium-226 per year. If, indeed, such dispersion rates persisted for large unmined ore bodies, their lifetimes would be orders of magnitude shorter than their observed lifetimes. In addition, they would be relatively easy to find by chemical analysis of the surrounding groundwater. In fact, groundwater analyses have been notably unsuccessful in identifying buried ore bodies precisely because the uranium burden of close-in water samples is rarely, if ever, significantly higher than in the larger surrounding environment. Uranium ore that is close to the surface and exposed to oxygenated water can be readily oxidized and transported to an adjacent unoxidized layer of rock. This is the phenomenon that produces "roll fronts" in the Colorado Plateau. However, the interaction of oxidizing water with uranium in an insoluble reduced mineral form is a special case that is encountered only in nonreducing soils or very near the surface. Therefore, it should not be assumed that such high release results are generally relevant in a comparison of ores with waste forms.

The presently proposed Environmental Protection Agency (EPA) limit on the release of radium-226 is 1.1×10^{13} Bq in 10^4 years (Table 8-1). Based on the EPA health consequence model (C. B. Smith et al. 1981, J. M. Smith et al. 1981), at this limit the average yearly release consequence is 0.1 cancers per year, consistent with the stated EPA objective of holding total consequences to 10^3 deaths per 100,000 Mg of uranium fuel in the first 10^4 years.

To the extent that justification for this limit can be derived by analogy with an ore body containing an equivalent amount of uranium, the technical rationale is as follows: The amount of natural uranium to produce 10^5 Mg of slightly enriched uranium in light-water reactor fuel is about 6.5×10^5 Mg, which contains about 8.2×10^{15} Bq of radium-226. An EPA release limit of 1.1×10^{13} Bq in 10,000 years, if applied to the natural uranium, corresponds to a fractional release rate for radium of about 10^{-7}/yr.

It is technically more sound to derive a radium-226 limit from generalized background data, which are voluminous and reasonably accurate, than from less well established leaching data attributed to single ore bodies. Cohen (1977) stated that 300 g or 1.1×10^{13} Bq of radium-226 are leached per year in the United States (with a total area of 8×10^{12} m^2). This corresponds to a leach rate from the top 600 m of soil and rock of 2.5×10^{-8}/yr. On the average, an area 10 km on a side releases 1.4×10^8 Bq of radium-226 to the biosphere per year, or 1.4×10^{12} Bq in 10^4 years. Water in rivers contains an average of 7.4×10^{-3} Bq/l of radium-226. Many regions in the United States presently exceed this average level by an order of magnitude. If a repository area encompassed by the interface with the biosphere in an average region is assumed to be 280 km on a side, the limit of 1.1×10^{13} Bq of radium-226 would add 1 percent to the runoff in the enclosed area if it were not further impeded during transport.

10.3. THE OKLO NATURAL REACTOR

Of the various uranium ore bodies that have been studied as useful analogs for waste deposits, the most thoroughly investigated is the Oklo mine in the Republic of Gabon, West Africa. This mine contains a series of adjoining natural "fossil" fission reactors that sustained neutron chain reactions 2×10^9 years ago, generated 12 Mg or more of fission products and 4 Mg of plutonium, and shut down after a few hundred thousand years of low-level operation. Searches for additional natural reactors have been conducted in rich Precambrian ore deposits in Saskatchewan, Canada, and in the Northern Territory, Australia, but no additional examples have been found.

With reference to the waste management interest, the unique features of these natural reactors are (1) they generated known amounts of fission products and depleted uranium and plutonium at a given site and within a known period in prehistoric time and (2) the mineral grains were exposed to elevated temperatures, high radiation, and heavy loading of the entire spectrum of fission products. The retention or loss of each of these products by the uraninite grains in which it was produced serves as a measure of its mobility in this particular environment and under these temperature, radiation, and loading conditions.

The products that were largely retained are zirconium, niobium, ruthenium, palladium, silver, tellurium, the rare earths, bismuth, thorium, uranium, and plutonium (Bryant et al. 1976). The fractional loss of these elements is about 10^{-9}/yr (Cowan et al. 1978). In a more recent Los Alamos analysis of Oklo data (Curtis et al., in press), evidence is reported for significant technetium, ruthenium, and neodymium losses in one of the reactor zones. These losses indicate dispersion rates of the order of 10^{-7}/yr to 10^{-8}/yr. Lead-206 is characteristically about 50 percent retained, indicating a fractional loss rate of 10^{-10}/yr. These rates are probably controlled by the rate of volume diffusion to the surface of the grain. The dissolution rate of the uraninite grains themselves is also characteristically no greater than 10^{-10}/yr, although there are regions in which the rate is considerably higher.

10.4. THE THORIUM DEPOSIT AT MORRO DO FERRO, BRAZIL

A deposit containing 20,000 Mg of thorium, largely in a monazitelike mineral, cheralite, located on Morro do Ferro, a hill on the Pocos do Caldas plateau in the state of Minas Gerais, Brazil, is presently being studied as a waste deposit analog (Eisenbud et al. 1982). Although the investigation is not yet completed, analysis of water and sediments from the stream that drains the hill indicates that the thorium is being mobilized, largely by erosion of the outcropping mineral, at a fractional rate of 5.9×10^{-7}/yr from the hillside. The mobilization by dissolution is estimated at 7.5×10^{-10}/yr.

10.5. THE IRIDIUM ANOMALY AT THE CRETACEOUS-TERTIARY BOUNDARY

The recently observed iridium anomaly at the Cretaceous-Tertiary boundary (Alvarez et al. 1980) is relevant as a natural analog because it has resisted dispersion and remains in a narrow, well-defined band of sediments observed in several locations around the world. Pillmore (1982) has reported such a band 5 cm wide peaking at an iridium concentration about 2,000-fold higher than the immediately adjacent samples. If the mobilization rate significantly exceeded 10^{-8}/yr, the band would not have remained sharply defined 6.5×10^{7} years after it was laid down.

10.6. THE NEVADA TEST SITE EXPERIMENT

A nuclear explosion, detonated May 14, 1965, in tuffaceous rock 294 m below the surface and 73 m below the water table in Frenchman's Flat, Nevada, produced a yield of 0.75 kT and a cavity 10.9 m in radius containing plutonium, uranium, the entire fission-product spectrum, tritium (as HTO), and a variety of neutron capture products.

As a part of a field study designed to measure the migration of these elements, the cavity was reentered in the spring of 1974. The distribution of the elements of interest between soil melt and water was determined by analysis of liquid and solid samples retrieved from various parts of the cavity and chimney. In general, the nuclear components and fission products were trapped in solid debris, although some of the more volatile elements were not entirely condensed or solid immediately following explosion and, consequently, were highly fractionated. Ten years after the event, the retention factors in soil ranged from 10^7 for plutonium-239, 10^6 for promethium-147, and 10^2 to 10^4 for strontium-90, ruthenium-106, antimony-125, and cesium-137. Only technetium and strontium-90 concentrations exceeded permissible drinking water levels in a few of the highest level samples of water taken directly from the explosion cavity.

Water has been pumped continuously at a rate of about 2.3 m^3/min from a satellite well 91 m from the cavity since October 1975. Tritium broke through two years later and now appears to be peaking in concentration. The water also contains krypton-85 and, possibly, some traces of ruthenium-106. Calculations that include retardation by sorption indicate that strontium-90, for example, should break through in 1,500 years. The pumping and analyses are continuing to ensure that unknown phenomena do not produce unexpectedly high transport rates for some species.

10.7. SOME CONCLUSIONS FROM NATURAL AND FIELD ANALOG DATA

All of the field data available to us demonstrate that leaching rates of solids under natural conditions tend to be very much slower than laboratory measurements made by Soxhlet extraction under standard International Atomic Energy Agency (IAEA) conditions. The differences are frequently two orders of magnitude or more. This observation has been repeated by a number of investigators. In a Chalk River paper on the comparative rates of leaching of fission-product-loaded syenite glass in the field and laboratory, the reported field rates are approximately 45-fold slower than in the laboratory after correcting for temperature differences. Mendel et al. (1981) report on Soxhlet extraction experiments with some common minerals (quartz crystals, dolomite, garnet, corundum, orthoclase, granite, quartzite, felsite, marble, calcite, and basalt). All of these minerals dissolve in the laboratory at rates comparable to or greater than high-level waste (HLW) glass. They comment that ". . . clearly [these] leach rates do not prevail in nature [or] the land masses would have dissolved away long ago. . . . There are rate-inhibiting processes in the natural surroundings that protect natural materials and that may also slow the leaching of waste forms."

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GLOSSARY

accessible environment	Defined in the proposed Environmental Protection Agency high-level waste standard as "surface waters, land surfaces, the atmosphere and underground formations which might provide ground water for human consumption including any locations greater than 10 km from the original location of the emplaced wastes (beneath the earth's surface)."
actinide	An element in the series beginning with element 90 and continuing through element 103. All the transuranic radionuclides discussed in this report are actinides.
activity	A measure of the amount of a radioactive material, expressed in terms of its rate of radioactive decay. In the SI system, the unit of activity is the becquerel (Bq), where 1 Bq = 1 disintegration per second. The unit in the alternate system is the curie (Ci), where 1 Ci = 3.7 x 10^{10} disintegrations per second.
advective transport	The convective-diffusive motion of a dissolved species that sorbs locally in a porous solid.
ALARA	The philosophy that radiation exposures be kept "as low as is reasonably achievable." As defined by the International Commission on Radiological Protection, it includes the concepts that the exposures be justified and that cost-benefit analyses be performed to determine the "optimum" level of protection.
alluvium	Deposits laid down by streams or running water.
Anadarko Basin	A small basin, or subbasin, within the major geologic region identified as the Permian Region that covers most of the central United States. It

	is located in the Texas panhandle, extending northeastward into Oklahoma.
analcime	A mineral: $NaAlSi_2O_6\cdot H_2O$. It is an isometric zeolite found in diabase and in alkali-rich basalts.
anhydrite	A mineral consisting of calcium sulfate, $CaSO_4$. It is gypsum without its water of hydration and is denser, harder, and less soluble than gypsum.
anion	A negatively charged ion in solution.
anticline	A fold of rocks whose core contains the stratigraphically older rocks; it is convex upward.
aquifer	A water-bearing layer of permeable rock that will yield water in usable quantities to wells.
aquitard	A confining bed that retards, but does not prevent, the flow of water to or from an adjacent aquifer; a leaky confining bed. It does not readily yield water to wells or springs but may serve as a storage unit for groundwater.
backfill	Material such as crushed rock, sand, or clay used to fill the space between emplaced waste canisters and the emplacement-hole rock surface.
basalt	Rocks or deposits of igneous origin, including extrusive flows and intrusions into existing rocks.
bedded salt	Deposits of salt (NaCl, or halite when in pure form) laid down in layers or beds.
bentonite	A soft, plastic, porous light-colored rock composed primarily of clay minerals of the montmorillonite (smectite) group plus colloidal silica.
biosphere	Generally includes the earth's surface, the oceans, and the atmosphere; those zones that contain or support life.
borosilicate glass	A glass in which the principal network formers are boron and silicon; a primary candidate high-level waste form for incorporating calcined fission products and actinides.
breccia	A coarse-grained rock composed of angular broken fragments held together by a mineral cement or in a fine-grained matrix.

breccia pipe	A cylindrically shaped, more or less vertical body of breccia material residing within horizontal beds of rock. Commonly found in salt and gypsum beds.
brine aquifer	An aquifer containing highly saline or mineralized water.
brine inclusion	Aqueous liquids included as droplets in natural salt deposits.
Bullfrog member	The lower welded tuff of the Crater Flat tuff; contains well-formed quartz crystals, biotite, and white pumice lenticles.
calcine	The product of the calcination process wherein the water portion of slurried waste is driven off by evaporation at high temperature in a spray chamber. The calcine is the residue of dry unmelted particulate solid.
calcrete	A conglomerate consisting of surficial sand and gravel cemented into a hard mass by calcium carbonate.
Calico Hills Formation (bedded tuffs)	A volcanic formation of Miocene age composed of beds of nonwelded vitric tuff and tuffaceous sandstone whose matrix commonly is altered to zeolite and clay minerals.
canister	A container, usually cylindrical, in which solidified reprocessing waste or spent fuel is contained for handling prior to emplacement in a repository. Additional containers or packaging materials may be added to comprise the emplaced waste package.
Capitan Reef	Massive limestone beds of Permian age in western Texas and southeastern New Mexico.
Castile Formation	A series of largely anhydrite-gypsum beds in western Texas and southeastern New Mexico. It underlies the Salado Formation, the proposed location for the Waste Isolation Pilot Plant.
cataclastic process	Brittle failure of materials involving dilation due to displacements along discontinuities and formation of new fractures with the opening of voids.
cation	A positively charged ion in solution.

cermet	A material or body consisting of ceramic particles bonded within a metal matrix.
cladding waste	Radioactive zircaloy cladding originally containing uranium dioxide reactor fuel.
clastic	A rock or sediment composed principally of broken fragments that are derived from preexisting rocks or minerals and that have been transported some distance from their places of origin.
clinoptilolite	A zeolite mineral: $(Na,K,Ca)_{2-3}Al_3(Al,Si)_2Si_{13}O_{36} \cdot 12H_2O$.
collective dose (equivalent)	See population dose.
colloid	A fine-grained material in suspension, or any such material that can be easily suspended; when mixed with a liquid, such particles will not settle but tend to remain suspended. The colloidal suspension thus formed has properties that are quite different from the simple solid-liquid mixture, or a solution.
committed dose (equivalent)	The dose equivalent that will be accumulated for a specified period of years following an initial intake of radioactive material into the body. For occupational situations, the 50-year committed dose is generally used.
competent	Referring to structurally strong rock; able to support tectonic forces without yielding.
complex	A chemical compound involving coordination bonds.
congruent dissolution	A process of dissolving wherein the ratio of the rates of dissolution of constituents is proportional to their concentration ratios.
constitutive model	Mathematical model describing the behavior of a material, e.g., yield and failure criteria, flow laws, and/or relations between stress and strain.
Cordilleran	A series of more or less parallel ranges or chains of mountains together with their associated valleys, basins, plateaus, rivers, and lakes.
Crater Flat tuff	A volcanic formation of Miocene age erupted from the Sleeping Butte Caldera and composed of the Bullfrog and Prow Pass members.

creep	Plastic flow of rock, particularly significant in salt formations, under the influence of lithostatic pressure.
crystalline rock	An inexact but convenient term designating an igneous or metamorphic rock as opposed to a sedimentary rock.
Dalhart Basin	A subbasin or region within the Permian Basin located in the Texas panhandle extending into New Mexico to the west and into Oklahoma to the northeast.
Darcy flow	Flow in a porous medium whereby the superficial velocity is proportional to the gradient of a flow potential.
decrepitation	Structural deterioration due to heating characterized by loss of cohesion between mineral grains or crystals.
deviator stress	Principal stress difference or shear stress.
devitrification	Change from a glassy state to a crystalline state.
diapir	A dome or anticlinal fold in which the overlying rocks have been ruptured by the squeezing out of plastic core material. Diapirs in sedimentary strata usually contain cores of salt or shale.
digitate margin	A boundary of a rock formation or structure that is characterized by a fingerlike or finger-shaped outline.
dilatency	Material volume increase during shear. It is expressed as the change in volume per unit volume.
discharge (area)	An area in which subsurface water is discharged to the land surface, to bodies of surface water, or to the atmosphere.
dispersion	The expansion of a moving plume or band of contaminant carried by a moving fluid in a porous medium. It is caused by nonuniform distribution of fluid velocities that are due to effects of pores and fractures in the rock. The principal effect is to reduce the average concentration of contaminant in the plume.
dissolution front	The interface or boundary between where a mineral within a rock (or an entire rock) has been dissolved and the undissolved material.

dissolution time	The time period for complete dissolution of a solid.
domal salt	A diapir or piercement structure with a central, nearly equidimensional salt plug, generally 1 or 2 km or more in diameter, that has risen through the enclosing beds from a "mother" salt bed below. The mother bed may be as much as several kilometers below the top of the raised dome or plug.
dose equivalent	The product of the absorbed dose and a modifying or weighting factor called the quality factor, Q. Values are assigned to Q according to the relative ability of the several types of radiation--alpha, beta, gamma, neutron, etc.--to induce deleterious effects. The unit of dose equivalent is the sievert (Sv) or the rem, where 1 Sv = 100 rem.
dose (equivalent) commitment	The infinite time integral of the individual dose equivalent rate (or committed dose equivalent rate for internally deposited radionuclides).
dose factor	A numerical conversion factor that gives the committed dose (or dose equivalent) from an initial intake of radioactive material by an individual. The units are sieverts per becquerel, rem per curie, or the equivalent.
effective cross section	The actual cross section of rock through which water can flow freely, consisting of the connected pores and cracks but excluding voids filled with trapped air or water trapped by adhesion.
effective dose equivalent	The sum of the "weighted" committed dose equivalents to each of the several exposed organs or tissues. The weighting factor for an organ is the ratio of the stochastic risk from irradiation of that organ to the total risk from uniform irradiation of the whole body. The effective dose equivalent provides a means of summing the doses from nonuniform exposures. The unit is the sievert (Sv) in the SI system or the rem in the alternate system of units, where 1 Sv = 100 rem.
effective porosity	A measure of the void space that actually contributes to flow through a porous medium; it is usually less than the theoretical porosity. Expressed as the ratio of void volume to total volume in percent.
Eh	Oxidation potential, measured in volts.

equilibrium distribution coefficient (K_d)	The ratio of the mass of a radioactive chemical species sorbed onto the solid phase per unit weight of solid to the mass remaining in solution per unit volume of liquid in a porous medium. Usually expressed in units of milliliters per gram (ml/g).
eutaxitic	Rocks with a banded structure that results in a streaked or blotched appearance.
evaporites	Nonclastic sedimentary rocks composed primarily of minerals produced from saline solution as a result of extensive or total evaporation of the solvent. Examples include gypsum, rock salt, primary dolomite, and various nitrates and borates.
far field	An imprecise term used to designate zones or locations in rocks at some distance from an underground waste repository.
fault	A surface or zone of rock fracture along which there has been displacement.
fertile	Describes a nuclide that can be transmuted into a fissile, or fissionable, nuclide by absorption of a neutron and subsequent decay.
Fick's law	An empirical law that states that the rate of diffusion of a substance is proportional to the negative concentration gradient of the diffusing substance in the direction perpendicular to the plane.
fissile	A nuclide that undergoes fission on absorption of a neutron of any energy.
fission	The splitting of a nucleus into smaller parts, usually two of approximately equal mass, each the nucleus of a lighter element. The process is accompanied by the release of about 200 million electron volts of energy and one or more neutrons.
fission product	Any radioactive or stable nuclide produced by fission, including both primary fission fragments and their radioactive decay products.
flux (water)	Flow rate, either volume or mass, per unit time.
forced convection	Movement of a fluid under an external influence such as a difference in pressure or an unstable gradient of density.

formation (geologic)	The basic unit in the local classification of rocks. It consists of a body of rock generally characterized by some degree of compositional and structural uniformity or distinctive features.
fuel cycle	Includes the sequence of steps and processes from mining the ore, manufacture of the fuel elements, and production of power in a reactor to disposition of spent fuel after removal from the reactor.
FUETAP	Acronym for "formed under elevated temperature and pressure." A special concrete-matrix waste form, hot-pressed to reduce voids and water of hydration.
fumarolic minerals	Minerals containing voids or vesicles created by gases and vapors of volcanic origin, or minerals formed by volcanic venting processes.
genetic effects	Generally used in radiation protection to designate harmful effects to progeny that could result from radiation-induced mutations in the germ cells of one or both parents. Can include a spectrum of effects from stillbirth to various diseases of genetic origin.
geomorphic	Pertaining to the form of the earth or of its surface features, e.g., a geomorphic province.
geopressured	Usually referring to underground water bodies in which the fluid pressure exceeds the nominal hydrostatic pressure at that depth.
geosyncline	A mobile downwarping of the crust of the earth, either elongate or basin like, measured in scores of kilometers in which sedimentary and volcanic rocks accumulate to thicknesses of thousands of meters.
geothermal gradient	The rate of increase of temperature of the earth with depth. The approximate average value in the earth's crust is 25°C per kilometer or 1.4°F per hundred feet.
Grande Ronde	A principal basalt formation in the Pasco Basin in the state of Washington. It is composed of a series of beds or "flows," one of which is the Umtanum, a principal candidate for a repository.
granitoid	A general term indicating grain size and mineral distribution typical of granite.
groundwater	Water that exists or flows within underground rock formations.

gypsum	A mineral consisting of hydrous calcium sulfate, $CaSO_4 \cdot 2H_2O$. It is soft and, when pure, white.
halite	The mineral rock salt, NaCl.
health effects	Term used by the U.S. Environmental Protection Agency to denote fatal cancers and first-generation genetic effects (leading to early death in individuals) predicted to occur in population groups exposed to ionizing radiation.
high-level waste (HLW)	The highly radioactive materials resulting from the fission process in nuclear reactors. Can include the separated radioactive residues from the chemical reprocessing of spent fuel or the unreprocessed spent fuel, when the latter is intended for disposal.
horizon	An underground level extending in all horizontal directions from a specific location.
host rock	The rock formation in which a high-level waste repository is located. Intended to distinguish between surrounding or nearby rock formations and the specific formation containing the repository.
human intrusion	Actions of humans in the future that result in contact with radioactive materials placed in a repository. Includes drilling of wells or sinking of shafts and withdrawal of contaminated water or rock materials.
hydraulic conductivity	The ratio of flow velocity to driving force for viscous flow under saturated conditions in a porous medium. It is measured in meters per day or equivalent units.
hydraulic gradient	The change in hydraulic head of water with distance in an aquifer at a given point and in a specified direction.
hydraulic head	The height of a column of water that can be supported by the static pressure. A measure of the potential energy at a point in a body of fluid or in a saturated porous medium.
hybrid medium	A concept wherein a repository is located in a geologic setting of different rock types, that in combination provide conditions favorable for long-term performance; for example, a repository

	constructed in a hard rock that is overlain by a sedimentary regional aquifer.
hydriding	The process of chemical combination with hydrogen.
illite	A general name for a group of three-layer micalike clay minerals that are widely distributed in argillaceous sediments (especially in marine shales and soils derived from them). They are intermediate in composition and structure between muscovite and montmorillonite.
incongruent dissolution	A process of dissolving where the ratios of dissolution rates of constituents are not proportional to their concentration ratios.
interbed	A bed, typically thin, of one kind of rock material occurring between or altering with beds of another kind.
isolation	Prevention of the movement of radionuclides placed in a repository to locations where future humans could be exposed to the radiation. Isolation is considered to be achievable if results of performance predictions satisfy the applicable performance criterion (such as a specified dose to future individuals).
isotherm	A line connecting points of equal temperature.
isovolumetric ductile behavior	Material plastic flow at constant volume.
joint	A fracture or parting in a rock along which little or no displacement of rock material has occurred.
ligand	A molecular group bonded about a central atom.
limit equilibrium analysis	Ultimate or limit load calculations at equilibrium of forces.
linear hypothesis	A set of assumptions used to predict the numbers of fatal cancers associated with exposure of populations to low levels of ionizing radiation.
lithophysical	Referring to the physical properties or structure of rocks.
lithopysae	Hollow bubblelike structures composed of concentric shells of finely crystalline alkali feldspar,

	quartz, and other minerals. Found in silicic volcanic rocks, e.g., rhyolite and obsidian.
magnetite	A black, isometric, strongly magnetic, opaque mineral of the spinel group: $(Fe,Mg)Fe_2O_4$. Constitutes an important ore of iron and often contains viable amounts of titanium oxide.
matrix	The material in which radioactive waste is incorporated as a solid. The matrix material and the contained radionuclides together comprise the waste form.
maximally exposed individual	A hypothetical person who because of the combination of assumptions used to estimate the dose is predicted to receive the largest dose among all individuals who could receive exposure from a postulated release of radioactive material.
megascopic	An object or phenomenon or its characteristics that can be observed with the unaided eye or with a hand lens.
Mesozoic	An era of geologic time from the end of the Paleozoic to the beginning of the Cenozoic, or from about 225 million to about 65 million years ago.
meteoric water	Pertaining to water of recent atmospheric origin.
MgHM	Megagrams of heavy metal, referring to the amount of fissile material, which consists of uranium, plutonium, and other actinides present.
miarolitic	Small irregular cavities in igneous rocks, especially granites, into which small crystals of the rock minerals protrude; characteristic of, pertaining to, or occurring in such cavities. Also used to refer to rock containing such cavities.
midden	Remains of nests or burrows of rodents or other animals. Generally refers to those used by animals in the distant past.
montmorillonite	A group of expanding-lattice clay minerals of general formula $R_{0.33}Al_2Si_4O_{10}(OH)_2 \cdot nH_2O$, where R includes one or more of the cations Na^+, K^+, Mg^{+2}, Ca^{+2}, and possibly others.
Nash Draw	A shallow 5-mile-wide valley open to the southwest located to the west of the proposed Waste Isolation Pilot Plant site in southeastern New Mexico.

natural background radiation	Radiation in the human environment from naturally occurring radioactive isotopes and from cosmic radiation.
near field	Referring to a zone within the repository host rock that is in the proximity of the emplaced waste packages.
nepheline glass	A glassy mineral composed of crystals or colorless grains: $(Na,K)AlSiO_4$.
Nevada Test Site	A U.S. government-owned site in Clark and Nye counties in southern Nevada dedicated to Department of Energy and Department of Defense research and test projects, including underground testing of nuclear weapons.
nuclide	A species of atom characterized by its mass number, atomic number, and nuclear energy state.
overpack	A container in which the waste form (in its canister) is placed for emplacement in a repository. Several potential functions have been identified for overpacks. These include provision of a corrosion-resistant layer to delay the beginning of the release of nuclides from the waste package.
Paintbrush tuff	A volcanic formation of Miocene age erupted from the Claim Canyon Cauldron; composed of several members of welded and nonwelded ash-flow tuff and bedded tuff.
Paleozoic	The geologic era covering the time span from about 575 million to about 225 million years ago, or from the end of the Precambrian to the beginning of the Mesozoic eras.
Palo Duro Basin	A subbasin within the Permian Basin of the United States; located within the Texas panhandle. Contains bedded salt formations that are under evaluation as potential repository sites.
Paradox Basin	An area of approximately 10,000 square miles in southeastern Utah and southwestern Colorado underlain by a series of salt-core anticlines.
performance criterion	As used in this document, it is a specified quantity (10^{-4} Sv/yr to an individual) against which results of the calculated performance of the waste-isolation system are compared.

permeability	A measure of the ability of a porous medium to transmit fluids. It is also called the intrinsic or specific permeability and is defined to be independent of the fluid flowing through it. It has units of length squared or darcies, where 1 darcy = 9.87×10^{-9} cm^2.
Permian Basin	A region in the central United States where during Permian times (280 million to 225 million years ago) there were many shallow seas that laid down vast beds of evaporites, i.e., salt and gypsum beds.
pH	The negative logarithm of the hydrogen ion concentration. It denotes the degree of acidity or basicity of a solution. At 25°C, 7 is the neutral value; higher values denote increasing basicity; lower values denote increasing acidity.
pintle	A protuberance or receptacle for the attachment of mechanical lifting or grappling devices.
pluton	A body of intrusive igneous rock of any shape or size.
pollucite	A colorless, transparent zeolite mineral: $(Cs,Na)_2Al_2Si_4O_{12}H_2O$. It occurs in massive cubic form and is often used as a gemstone.
population dose	The sum of the doses to all the individuals in a specified group. In units of person-sievert or person-rem. Also called collective dose.
pore velocity	The actual linear velocity of water moving in a porous medium.
potential flow	A flow field wherein the velocity can be derived from the gradient of a potential; from hydrodynamic theory.
potentiometric gradient	See hydraulic gradient.
Precambrian	The era in geologic time prior to about 570 million years ago. It includes all geologic time and the corresponding rocks before the beginning of the Paleozoic era; it spans about 90 percent of all geologic time.
Priest Rapids member	A series of three or four basalt lava flows in the vicinity of the Priest Rapids Dam in south-central Washington. It is the youngest subdivision of the

	Wanapum basalt that overlies the Grande Ronde basalt at the Basalt Waste Isolation Project site.
prolate spheroid	A spherically shaped body elongated at the poles. The longitudinal cross section is elliptical.
pyrite	A common pale bronze or brass yellow isometric mineral: FeS_2. It has a brilliant metallic luster, an absence of cleavage, and is often mistaken for gold.
Quaternary	The most recent geologic time period, covering the present back to about 1.5 million years ago.
radiogenic	Of radioactive origin.
radiolysis	Breaking of chemical bonds in molecules by the action of ionizing radiation. Can result in the production of new chemical species.
radionuclide inventory	The amounts of radionuclides in a particular part of the waste-isolation system; usually listed as the activities (in curies or becquerels) of each nuclide present. It is a time-dependent quantity.
recharge	The processes involved in the absorption and addition of water to rock formations; also the amount of water added.
repository	An underground mined facility, and the associated engineered structures and equipment, where high-level radioactive wastes are placed for disposal.
retardation factor (or constant)	A measure of the delay due to sorption on the rock pore surfaces of chemicals moving in groundwater through porous rock. See K under Units and Nomenclature.
retrievability	The capability to safely remove previously emplaced wastes from a repository prior to final closure and sealing.
rhyolitic tuff	Rock formed of compacted volcanic fragments composed of fine-grained materials consisting of quartzes and feldspars set in a glassy groundmass.
Rustler Formation	The evaporite beds, including mudstones, probably of Permian age that immediately overlie the Salado Formation (proposed Waste Isolation Pilot Plant host formation) in southeastern New Mexico.

Saddle Mountains Formation	A basalt formation on the Hanford Reservation. It is one of the major formations overlying the Grande Ronde Formation, the candidate formation for the Basalt Waste Isolation Project repository.
Salado Formation	A formation composed of salt and gypsum beds in southeastern New Mexico. It is the formation that is the prime candidate for the Waste Isolation Pilot Plant repository.
sericite	A white, fine-grained potassium mica occurring in small scales and flakes as an alteration product of various aluminosilicate minerals, having a silky luster and found in various metamorphic rocks.
shaft	A vertical or steeply inclined drilled or excavated hole that connects the surface with the underground working areas of a mine or repository.
shaft pillar	The volume of rock through which a shaft passes. Major openings are excluded from the pillar to ensure adequate protection of the shaft.
shotcrete	Pneumatically applied portland cement mortar or concrete. Used to line underground excavations to give support, to provide a smooth surface, and to limit the weathering of exposed rock.
shunt flow	Flow parallel to a tunnel or shaft.
silicic tuffs	Silica-rich consolidated pyroclastic, i.e., explosively formed, areally deposited tuff containing varying proportions of silicic glass, silica polymorphs, feldspars, zeolites, and clays, plus generally minor amounts of metal oxides and mafic silicates.
smectite	A name for the montmorillonite group of clay minerals. It includes montmorillonite and saponite and their chemical varieties. These minerals exhibit swelling properties when wetted and have high ion exchange properties.
sodalite	A mineral of the feld spathoid group: $Na_4Al_3Si_3O_{12}Cl$. It is usually blue or blue-violet, but it may be white, greenish gray, pink, or yellow, and it occurs in various sodium-rich igneous rocks.
sol-gel ceramic	A candidate high-level waste form based on a sol-gel production process wherein very small particles of

waste material in the form of salts as oxides or nitrates are suspended in a colloidal suspension. As the liquid is removed, the viscosity increases, and a gel is formed. The material is then extruded into small beads (up to about 100 microns), which can then be incorporated into large monoliths by sintering, or hot pressing, with a suitable matrix material such as various powdered metals.

solubility	A measure of the amount of a soluble material that can be contained dissolved, in solution, in a liquid; the equilibrium concentration of a solute in a solution saturated with respect to that solute at a given temperature and pressure. Units in grams per liter or equivalent.
solubility-limited dissolution	Where the rate of dissolution of radioactive species from the waste package is limited by solubility, diffusion, and convection in groundwater.
sorbate	Material that is sorbed onto another material.
sorption	The binding on a microscopic scale of one substance to another, such as by adsorption or ion exchange, e.g., the sorption of soluble radionuclides from the liquid phase onto the solid phase in a porous rock.
Soxhlet extractor	A laboratory leach-testing apparatus. It moves leaching solution past the test specimen. Aliquots of the solution are removed periodically, and the concentration of leachant is measured to determine the fractional leach rate of the specimen.
spent fuel	Fuel assemblies removed from a nuclear power reactor after their useful life, usually 2 to 3 years of operation at power.
stability field	The range of chemical and thermodynamic conditions for which a chemical species or a mineral tends to remain in a given state.
strike-slip faulting	Faulting on which the movement is parallel to the strike or trend of the fault.
Strombolian eruption	A type of volcanic eruption characterized by jetting of clots or fountains of fluid basaltic lava from a central crater.
subareal	Referring to conditions and processes such as erosion that exist or operate in the open air on or

	immediately adjacent to the land surface; also of features and materials, such as aeolian deposits, that are formed or situated on the land surface.
syncline	A fold of which the core contains the stratigraphically younger rocks; it is generally concave upward.
SYNROC	A candidate high-level waste form, "synthetic rock." It is a ceramic based on titanates, zirconates, and aluminates. The base material is mixed, as a finely divided powder, with finely divided calcined waste, and then formed into cylinders by hot-isostatic pressing.
TBM	Tunnel boring machine.
tectonic	Rock structure or forces resulting from deformation of the earth's crust.
Tertiary	The geologic period within the Cenozoic, or most recent, era covering the time from about 70 million years ago to the advent of early man at about 1.5 million years ago.
tertiary creep	An accelerating rate of plastic flow under constant stress, culminating in material failure.
thermal period	The time during which the rock surrounding a repository is appreciably heated by radiation from radioactive decay of emplaced waste.
thermoelastic/ viscoelastic analysis	Analysis that includes the influence of mechanical and/or heat-induced stresses on material elastic response. It is coupled with time-dependent displacement (i.e., flow or creep) phenomena.
Ticode	A group of titanium-based alloys that have high resistance to corrosion in water and salt-brine environments.
titanite	A mineral, usually yellow or brown: $CaTiSiO_5$. It occurs in wedge-shaped monoclinic crystals as an accessory mineral in granitic rocks and in calcium-rich metamorphic rocks.
Topopah Spring member	The lower member of the Paintbrush tuff composed of welded and nonwelded ash-flow tuff having normal thermal remnant magnetization in contrast to reverse polarization of younger units of the Paintbrush tuff.

Tram member	One of the deeper lying tuff formations in the Crater Flat series, varying in thickness from about 100 to 300 m and lying from 800 to 1,100 m below the surface at the Yucca Mountain, Nevada, proposed repository site.
transuranic (TRU) waste	Radioactive waste material containing elements with atomic numbers above 92 in concentrations above a specified value.
Umtanum flow	A dense basalt lava flow formation within the Grande Ronde basalt group on the Hanford Reservation. It averages about 50 m in thickness and lies about 1,000 m below the surface. It is being actively investigated as a possible host rock for a commercial high-level waste repository.
vitrification	Incorporation of radioactive waste into glassy or noncrystalline material.
vitrophyre	Any porphyritic igneous rock, i.e., large crystals set in a glassy groundmass.
volcanism	The process by which magma and the associated gases rise into the crust and are extruded onto the earth's surface and into the atmosphere.
waste form	The solid material that is comprised of the radioactive wastes, usually as a calcine, together with the incorporating matrix, e.g., borosilicate glass.
water travel time	The average time for water that has contacted wastes in a repository to move from the repository location through the surrounding rocks to a specified location where the water could be used by humans.
zeolites	A generic term for a large group of hydrous alumino-silicates that are similar in composition to the feldspars, with sodium, calcium, and potassium as their chief metals.
zirconate	A gray or brownish zircon mineral: $ZrSiO_4$. It occurs in tetragonal prisms and is a common accessory mineral in siliceous igneous rocks, crystalline limestones, schists and gneisses, and derived sedimentary and river placer deposits.

UNITS AND NOMENCLATURE

Bq	becquerel, SI unit of radioactivity, 1 Bq = 1 disintegration per second.
Bq/l	becquerels per liter, radioactivity concentration.
Bq/m^3	becquerels per cubic meter, radioactivity concentration.
Ci	curie, alternate unit of radioactivity, 1 Ci = 3.7×10^{10} disintegrations per second.
cm	centimeter.
d	day.
D_j	liquid diffusion coefficient for species j, in meters squared per second.
f	fractional release rate of species j from a waste package or test sample, in s^{-1}.
g	gram.
g/cm^3	grams per cubic centimeter, density, solubility limit concentration of a dissolved species.
GW	gigawatt, 10^9 watts.
GWe	gigawatt of electric power.
GWth	gigawatt of thermal power.
Gy	gray, SI unit of absorbed dose, 1 Gy = 1 joule per kilogram. In the alternate system of units, 1 Gy = 100 rad.
J	joule.

K	retardation constant, coefficient, or factor, dimensionless. $K = 1 + \rho K_d(1 - \varepsilon)/\varepsilon$.
K_d	equilibrium distribution coefficient, in cubic centimeters per gram, or a dimensionless concentration ratio when concentrations in the solid and liquid phases are in the same units.
kg	kilogram, 10^3 grams.
km	kilometer, 10^3 meters.
kT	kilotons, 10^3 tons (of TNT), nuclear weapon energy yield.
kW	kilowatt, 10^3 watts.
kWth	kilowatts of thermal power.
L	length of waste-form cylinder, in meters.
m	meter.
$\underline{M}$	molar.
MBq	megabecquerel, 10^6 becquerels.
Mg	megagram, 10^6 grams.
MgHM	megagrams of heavy metal (uranium and transuranic elements).
min	minute.
ml/g	milliliters per gram.
mm	millimeter, 10^{-3} meter.
MPa	megapascal, 10^6 pascals.
m/s	meters per second, velocity, hydraulic conductivity.
mV	millivolt, 10^{-3} volt.
MWthd	megawatt day, thermal power.
MWe	megawatts of electric power.
m^2/yr	meters squared per year, dispersion coefficient.
m^3/yr	cubic meters per year, water flow rate.

N_j^*	solubility of elemental species j in groundwater, in grams per cubic centimeter.
n_j	bulk density of elemental species j in a waste-form material, in grams per cubic centimeter.
PBq	petabecquerel, 10^{15} becquerels.
ppb	parts per billion.
ppm	parts per million.
R	radius of waste-form cylinder, in meters.
rad	alternate unit of absorbed dose, 1 rad = 100 ergs per gram, 1 rad = 10^{-2} gray.
rem	alternate unit of dose equivalent, 1 rem = 10^{-2} sievert, 1 rem = 1 rad x Q, where Q = the quality factor.
s	second.
Sv	sievert, SI unit of dose equivalent, 1 Sv = 100 rem.
S/V	surface-to-volume ratio, in m^{-1}.
T	temperature, in degrees Kelvin (K).
U	groundwater pore velocity, in meters per year.
W/m^2	areal thermal loading, watts per meter squared.
$W/m^{o}C$	thermal conductivity, watts per meter per degree Celsius.
yr	year.
^{o}C	degrees Celsius, temperature.
^{o}E	degrees east, compass heading or direction.
β	a geometrical factor in the equation for the diffusion-controlled fractional release rate, in m^{-2}.
ε	porosity, pore volume divided by the volume of pores plus solid, dimensionless.
μ	viscosity, in grams per meter-second.
ρ	density of solid, in grams per cubic centimeter.

APPENDIX A

CHARACTERISTICS OF WASTE MATERIALS

TABLE A-1 Summary Composition of High-Level Liquid Waste from Reprocessing 1 Mg of PWR Fuel

	Time After Reprocessing[b]												
	1 yr			10 yr		100 yr		1,000 yr		10,000 yr		100,000 yr	
Constituent[a]	g	MBq	W	MBq	W	MBq	W	MBq	W	MBq	W	MBq	W
Fission products													
Se	5.6×10	1.5×10^4	–	1.5×10^4	–	1.5×10^6	–	1.5×10^4	–	1.4×10^4		5.2×10^3	–
Rb	3.4×10^2	–	–	–	–	–	–	–	–	–	–	–	–
Sr	8.7×10^2	2.6×10^9	8.4×10	2.1×10^9	6.6×10	2.5×10^8	7.8	–	–	–	–	–	–
Y	4.6×10^2	2.7×10^9	4.0×10^2	2.1×10^9	3.2×10^2	2.5×10^8	3.7×10	–	–	–	–	–	–
Zr	3.5×10^3	2.1×10^8	2.8×10	6.7×10^4	–	6.7×10^4	–	6.7×10^4	–	6.7×10^4		6.3×10^4	–
Nb	–	4.4×10^8	5.9×10	2.8×10^4	–	6.3×10^4	–	6.3×10^4	–	6.3×10^4	–	5.9×10^4	–
Mo	3.3×10^3	–	–	–	–	–	–	–	–	–	–	–	–
Tc	7.7×10^2	4.8×10^5		4.8×10^5	–	4.8×10^5	–	4.8×10^5	–	4.8×10^5	–	3.5×10^5	–
Ru	2.2×10^3	8.1×10^9	1.3×10	1.6×10^7	2.6×10^{-2}	–	–	–	–	–	–	–	–
Rh	4.7×10^2	8.1×10^9	2.1×10^3	1.6×10^7	4.3	–	–	–	–	–	–	–	–
Pd	1.3×10^3	4.1×10^3	–	4.1×10^3		4.1×10^{-3}	–	4.1×10^3	–	4.1×10^3	–	4.1×10^3	–
Ag	7.7×10	3.7×10^7	1.7×10	–	–	–	–	–	–	–	–	–	
Cd	1.1×10^2	2.0×10^6	9.1×10^{-2}	–	–	–	–	–	–	–	–	–	–
Sn	9.0×10	1.1×10^7	7.7×10^{-1}	3.6×10^4	–	3.1×10^4	–	2.8×10^4	–	2.7×10^4	–	1.4×10^4	–
Sb	2.8×10	3.7×10^8	3.3×10	4.1×10^7	3.5	3.3×10^4	1.2×10^{-2}	3.3×10^4	1.2×10^{-2}	3.1×10^4	1.1×10^{-2}	1.6×10^4	–
Te	4.7×10^2	1.4×10^8	3.2	1.0×10^7	2.3×10^{-1}	–	–	–	–	–	–	–	–
Cs	2.7×10^3	7.4×10^9	1.1×10^3	3.2×10^9	1.1×10^2	3.7×10^8	1.1×10	1.3×10^4	–	1.3×10^4	–	1.2×10^4	–
Ba	1.4×10^3	3.5×10^9	3.7×10^2	2.9×10^9	3.0×10^2	3.7×10^8	3.8×10	–	–	–	–	–	–
La-Gd	9.9×10^3	3.0×10^{10}	2.7×10^3	5.6×10^8	4.5×10	6.3×10^6	4.8×10^{-2}	5.9×10^3	–	–	–	–	–
Total	2.8×10^4	6.3×10^{10}	7.1×10^3	1.1×10^{10}	8.8×10^2	1.3×10^9	9.4×10	7.0×10^5	2.0×10^{-2}	7.0×10^5	1.9×10^{-2}	5.2×10^5	1.2×10^{-2}
Actinides													
Th	–	–	–	–	–	–	–	–	–	–	–	5.2×10^3	4.1×10^{-3}
Ra	–	–	–	–	–	–	–	–	–	–	–	5.2×10^3	–
Pa	–	1.1×10^{-4}	–	1.1×10^4	–	1.2×10^4	–	1.3×10^4	–	1.3×10^4	–	1.3×10^4	–
U	4.8×10^3	3.3×10^2	–	4.4×10^2	–	1.1×10^3	–	1.9×10^3	–	2.4×10^3	–	5.9×10^3	–
Np	4.4×10^2	6.3×10^5	5.1×10^{-2}	6.3×10^5	5.1×10^{-2}	6.3×10^5	5.1×10^{-2}	5.9×10^5	4.8×10^{-2}	2.6×10^5	2.7×10^{-2}	1.3×10^4	1.1×10^{-2}
Pu	5.1×10	2.5×10^7	3.1	1.8×10^7	3.6	2.4×10^6	2.0	3.1×10^5	2.6×10^{-1}	2.3×10^5	1.9×10^{-1}	1.9×10^4	1.6×10^{-2}
Am	1.4×10^2	8.1×10^6	6.9	8.1×10^6	7.0	7.4×10^6	6.5	2.1×10^6	1.9	2.5×10^5	2.1×10^{-1}	–	–
Cm	2.0×10	2.1×10^8	5.6×10	3.7×10^7	3.6×10	1.3×10^6	1.2	2.8×10^3	–	5.9×10^2	–	–	–
Total	5.4×10^3	2.4×10^8	6.6×10	6.7×10^7	4.7×10	1.2×10^7	9.8	3.1×10^6	2.2	7.4×10^5	4.4×10^{-1}	9.3×10^4	6.0×10^{-2}
Total fission products plus actinides	3.4×10^4	6.3×10^{10}	7.2×10^3	1.1×10^{10}	9.3×10^2	1.3×10^9	1.0×10^2	3.7×10^6	2.2	1.4×10^6	4.6×10^{-1}	6.3×10^5	7.2×10^{-2}

NOTE: This waste is from reprocessing 3.2 percent enriched UO_2 irradiated to 33,000 MWthd/MgHM at a specific power of 37.5 MWth/MgHM. It contains all of the fuel material except 100 percent of the hydrogen and noble gases, 99.9 percent of the halogens, and 99.5 percent of the uranium and plutonium.

[a] The principal chemical constituents from reprocessing include 50 kg HNO_3, 9 kg gadolinium added as a neutron poison, 1.5 kg PO_4^{-3}, 1.4 kg iron, nickel, and chromium, and 0.05 kg sodium.

[b] The fuel is reprocessed 160 days after discharge from the reactor.

SOURCE: Adapted from the data of Croff, A. G., and C. W. Alexander, 1980. Decay Characteristics of Once-Through LWR and LMFBR Spent Fuels, High-Level Wastes, and Fuel Assembly Structural Material Wastes. ORNL/TM-7431. Oak Ridge National Laboratory, Oak Ridge, Tenn.

TABLE A-2 Summary Composition of High-Level Liquid Waste from Reprocessing 1 Mg of Blended LMFBR Fuel Core and Blankets

	Time After Reprocessing[a]												
	1 yr			10 yr		100 yr		1,000 yr		10,000 yr		100,000 yr	
Constituent	g	MBq	W	MBq	W	MBq	W	MBq	W	MBq	W	MBq	W
Fission products	3.9×10^4	1.0×10^{11}	1.1×10^4	1.2×10^{10}	8.9×10^2	1.4×10^9	9.6×10	1.1×10^6	5.1×10^{-2}	1.0×10^6	4.8×10^{-2}	7.8×10^5	2.8×10^{-2}
Actinides	6.4×10^3	1.2×10^9	4.0×10^2	4.1×10^8	3.0×10^2	1.1×10^8	8.9×10	2.8×10^7	2.2×10	5.6×10^6	3.3	3.3×10^5	2.4×10^{-1}
Total	4.5×10^4	1.0×10^{11}	1.1×10^4	1.3×10^{10}	1.2×10^3	1.5×10^9	1.9×10^2	3.0×10^7	2.2×10	6.7×10^6	3.3	1.1×10^6	2.7×10^{-1}
Chemicals[b]													
HNO_3	5×10^4												
PO_4^{-3}	2×10^3												
Gd	9×10^3												
Fe	4×10^3												
Cr	1×10^3												
Ni	8×10^2												
Mn	1×10^2												
Mo	2×10^2												
Na	4×10												

NOTE: This waste is from reprocessing a blended mixture of core and blankets having an average burnup of 45,500 MWthd/MgHM at an average specific power of 41.5 MWth/MgHM. It contains all of the fuel material except 100 percent of the hydrogen and noble gases, 99.9 percent of the halogens, and 99.9 percent of the uranium and plutonium.

[a] The fuel is reprocessed 90 days after discharge from the reactor.

[b] The principal chemical constituents include 0.7 percent of the structural materials solubilized during dissolution and gadolinium added as a neutron poison.

SOURCE: Adapted from the data of Croff, A. G., and C. W. Alexander, 1980. Decay Characteristics of Once-Through LWR and LMFBR Spent Fuels, High-Level Wastes, and Fuel Assembly Structural Material Wastes. ORNL/TM-7431. Oak Ridge National Laboratory, Oak Ridge, Tenn.

TABLE A-3 Estimated Characteristics of Nuclear Fuel Services Neutralized High-Level Waste

			Radioactivity (MBq)					
	Mass (kg)		1978			1985	Thermal Power (W)	
Constituent	Sludge	Supernate	Sludge	Supernate	Total	Total	1978	1985
Fission product								
Sr			3.0×10^{11}	3.1×10^{9}	3.1×10^{11}	2.8×10^{11}	9.8×10^{3}	9.0×10^{3}
Y			3.0×10^{11}	3.1×10^{9}	3.1×10^{11}	2.8×10^{11}	4.6×10^{4}	4.2×10^{4}
Zr			9.3×10^{6}		9.3×10^{6}	9.3×10^{5}	2.0	2.0
Nb			4.1×10^{6}	-	4.1×10^{6}	4.1×10^{6}	-	-
Tc				7.0×10^{7}	7.0×10^{7}	7.0×10^{7}	3.0	3.0
Ru			2.0×10^{9}	2.0×10^{7}	2.0×10^{9}	2.0×10^{7}	3.0×10	-
Rh			2.0×10^{9}	2.0×10^{7}	2.0×10^{9}	2.0×10^{7}	1.2×10^{3}	1.2×10
Sb			2.2×10^{9}	2.2×10^{7}	2.3×10^{9}	4.1×10^{8}	5.1×10^{2}	1.0×10^{2}
Te			-	9.3×10^{8}	9.3×10^{8}	6.7×10^{8}	-	-
Sn			1.5×10^{6}	-	1.5×10^{6}	1.5×10^{6}	-	-
Cs			-	4.1×10^{11}	4.1×10^{11}	3.3×10^{11}	1.3×10^{4}	1.1×10^{4}
Ba			-	4.1×10^{11}	4.1×10^{11}	3.1×10^{11}	3.6×10^{4}	3.0×10^{4}
La-Gd			4.4×10^{10}	1.4×10^{8}	4.4×10^{10}	1.7×10^{10}	3.1×10^{3}	2.6×10^{3}
Total fission products	4.7×10^{3}	1.3×10^{3}	6.7×10^{11}	8.1×10^{11}	1.5×10^{12}	1.2×10^{12}	1.2×10^{5}	9.6×10^{4}
Actinides								
U	6.7×10^{3}	-	-	-	-	-	-	-
Np	3.2×10	-	8.9×10^{6}	-	8.9×10^{6}	8.9×10^{6}	4.7	4.7
Pu	3.5×10	-	2.0×10^{9}	-	2.0×10^{9}	1.4×10^{9}	1.5×10^{2}	1.5×10^{2}
Am	7.0	-	7.0×10^{8}	-	7.0×10^{8}	7.0×10^{8}	6.3×10^{2}	6.3×10^{2}
Cm	1.2×10	-	4.8×10^{8}	-	4.8×10^{8}	3.5×10^{8}	4.4×10^{2}	3.3×10^{2}
Total actinides	6.8×10^{3}	-	3.1×10^{9}	-	3.1×10^{9}	2.5×10^{9}	1.2×10^{3}	1.1×10^{3}
Total fission products plus actinides	1.2×10^{4}	1.3×10^{3}	6.7×10^{11}	8.1×10^{11}	1.5×10^{12}	1.2×10^{12}	1.2×10^{5}	9.7×10^{4}
Chemicals								
$(Na,K)_2SO_4$	-	9.0×10^{4}						
$(Na,K)\ NO_3/NO_2$	-	1.1×10^{6}						
$(Na,K)OH$	-	1.5×10^{4}						
$(Na,K)Cl$	-	5.0×10^{2}						
$Fe(OH)_3$	5.0×10^{4}	-						
$FePO_4$	2.9×10^{4}	1.3×10^{3}						
$Cr(OH)_3$	3.9×10^{3}	2.1×10^{3}						
$Ni(OH)_2$	1.7×10^{3}	7.5×10^{2}						
$Al(OH)_3$	8.3×10^{2}	-						
AlF_3	-	1.6×10^{3}						
$Na_3PO_4 \cdot 12\ MoO_3$	9.8×10^{2}	5.0×10^{2}						
Mn	2.2×10^{3}	-						
Total chemicals	8.9×10^{4}	1.2×10^{6}						
Grand total	1.0×10^{5}	1.2×10^{6}						

SOURCE: U.S. Department of Energy. 1981. Spent Fuel and Radioactive Waste Inventories and Projections as of December 31, 1980. DOE/NE-0017. Washington, D.C.

TABLE A-4 Estimated Characteristics of Nuclear Fuel Services Acid High-Level Waste

	Radioactivity Inventory (MBq)	Chemical Composition (kg)
Th	1.2×10^{5}	
U	3.7×10^{2}	
Fission products (including 1.8×10^{10} MBq ^{90}Sr and 2.8×10^{10} MBq ^{137}Cs)	9.0×10^{10}	
$Th(NO_3)_4$		32,000
$Al(NO_3)_3$		3,400
NaF		200
HNO_3		3,000
$HPO_4^{=}$		200
H_3CO_3		400
$M(NO_3)_x$ (M = Fe, Cr, Ni)		800

SOURCE: U.S. Department of Energy. 1981. Spent Fuel and Radioactive Waste Inventories and Projections as of December 31, 1980. DOE/NE-0017. Washington, D.C.

TABLE A-5 Summary Characteristics of Cladding Waste from Reprocessing 1 Mg of PWR Fuel

	Time After Reprocessing[a]										
	1 yr			10 yr		100 yr		1,000 yr		10,000 yr	
Constituent	g	MBq	W	MBq	W	MBq	W	MBq	W	MBq	W
Activation products											
C	–	3.5×10^4	–	3.5×10^4	–	3.5×10^4	–	3.1×10^4	–	1.0×10^4	–
Si	4.1×10^2	–	–	–	–	–	–	–	–	–	–
Cr	1.1×10^4	–	–	–	–	–	–	–	–	–	–
Mn	8.6×10^2	1.1×10^6	1.6×10^{-1}	7.8×10^2	–	–	–	–	–	–	–
Fe	3.1×10^4	1.4×10^8	5.3	1.3×10^7	4.8×10^{-1}	–	–	–	–	–	–
Co	–	2.4×10^8	9.9×10	7.4×10^7	3.0×10	5.2×10^2					
Ni	1.2×10^4	2.4×10^7	2.9×10^{-1}	2.3×10^7	2.7×10^{-1}	1.2×10^7	1.6×10^{-1}	2.0×10^5	3.3×10^{-2}	1.8×10^5	3.0×10^{-2}
Zr	2.3×10^5	5.9×10^6	8.3×10^{-1}	4.8×10^3	–	4.8×10^3	–	4.8×10^3	–	4.8×10^3	–
Nb	7.0×10^2	1.4×10^7	1.8	4.8×10^4	1.3×10^{-2}	5.2×10^4	1.3×10^{-2}	4.8×10^4	1.3×10^{-2}	3.7×10^4	9.3×10^{-3}
Mo	–	9.3×10^2	–	9.3×10^2	–	9.3×10^2	–	7.8×10^2	–	1.3×10^2	–
Sn	3.0×10^3	5.9×10^7	7.8×10	5.2×10^3	1.0×10^{-3}	–	–	–	–	–	–
Sb	–	4.1×10^7	3.5	4.4×10^6	3.7×10^{-1}	–	–	–	–	–	–
Te	–	1.0×10^7	2.3×10^{-1}	1.1×10^6	2.4×10^{-2}	–	–	–	–	–	–
Total activation products[b]	2.9×10^5	5.6×10^8	1.1×10^2	1.1×10^8	3.1×10	1.2×10^7	1.7×10^{-1}	2.9×10^5	4.6×10^{-2}	2.3×10^5	4.0×10^{-2}
Total fission products	1.7×10	3.2×10^7	3.6	5.6×10^6	4.4×10^{-1}	6.3×10^5	4.7×10^{-2}	3.6×10^2	1.0×10^{-5}	3.4×10^2	1.0×10^{-5}
Total actinides	4.8×10^2	2.3×10^6	8.9×10^{-2}	1.5×10^6	9.8×10^{-2}	1.3×10^5	9.4×10^{-2}	3.2×10^4	9.7×10^{-2}	8.1×10^3	6.7×10^{-3}
Grand total[b]	2.9×10^5	5.9×10^8	1.1×10^2	1.2×10^8	3.2×10	1.3×10^7	3.1×10^{-1}	3.2×10^5	1.4×10^{-1}	2.4×10^5	4.7×10^{-2}

NOTE: This waste is from reprocessing 3.2 percent enriched UO_2, irradiated to 33,000 MWthd/MgHM at a specific power of 37.5 MWth/MgHM. It contains all of the cladding and structural material plus 0.05 percent of the nonvolatile fission products and actinides, and 30 percent of the ^3H in the spent fuel.

[a] The fuel is reprocessed 160 days after discharge from the reactor.

[b] Total of all elements, listed or not.

SOURCE: Adapted from the data of Croff, A. G., and C. W. Alexander, 1980. Decay Characteristics of Once-Through LWR and LMFBR Spent Fuels, High-Level Wastes, and Fuel Assembly Structural Material Wastes. ORNL/TM-7431. Oak Ridge National Laboratory, Oak Ridge, Tenn.

TABLE A-6 Summary Characteristics of Cladding Wastes from Reprocessing 1 Mg of Blended LMFBR Fuel Core and Blankets

	Time After Reprocessing[a]										
	1 yr			10 yr		100 yr		1,000 yr		10,000 yr	
Constituent	g	MBq	W	MBq	W	MBq	W	MBq	W	MBq	W
Activation product											
C	–	4.1×10^{4}	–	4.1×10^{4}	–	4.1×10^{4}	–	3.7×10^{4}	–	1.3×10^{4}	–
Si	4.6×10^{3}	–	–	–	–	–	–	–	–	–	–
Cr	1.5×10^{5}	–	–	–	–	–	–	–	–	–	–
Mn	1.6×10^{4}	8.1×10^{7}	1.1×10^{2}	5.6×10^{5}	7.4×10^{-2}	–	–	–	–	–	–
Fe	5.6×10^{5}	3.2×10^{8}	1.2×10	2.9×10^{7}	1.1	–	–	–	–	–	–
Co	–	1.4×10^{8}	4.0×10	2.1×10^{7}	8.9	1.6×10^{2}	–	–	–	–	–
Ni	1.2×10^{4}	8.9×10^{6}	1.1×10^{-1}	8.1×10^{6}	1.1×10^{-1}	4.1×10^{6}	6.5×10^{-2}	1.3×10^{5}	2.1×10^{-2}	1.1×10^{5}	1.9×10^{-2}
Nb	–	1.0×10^{4}	2.8×10^{-3}	1.0×10^{4}	2.8×10^{-3}	1.0×10^{4}	2.8×10^{-3}	1.0×10^{4}	2.7×10^{-3}	7.4×10^{3}	2.0×10^{-3}
Mo	2.0×10^{4}	7.8×10^{3}	–	7.8×10^{3}	–	7.4×10^{3}	–	6.3×10^{3}	–	1.1×10^{3}	–
Tc	–	3.1×10^{3}	–	3.1×10^{3}	–	3.1×10^{3}	–	3.1×10^{3}	–	3.0×10^{3}	–
Total activation products[b]	8.7×10^{5}	1.3×10^{9}	1.6×10^{2}	5.9×10^{7}	1.0×10	4.4×10^{6}	6.8×10^{-2}	1.8×10^{5}	2.4×10^{-2}	1.4×10^{5}	2.0×10^{-2}
Total fission products	1.9×10	4.8×10^{7}	5.4	6.3×10^{6}	4.5×10^{-1}	7.0×10^{5}	4.8×10^{-2}	5.9×10^{2}	2.6×10^{-5}	5.2×10^{2}	2.4×10^{-5}
Total actinides	4.8×10^{2}	1.0×10^{7}	5.4×10^{-1}	6.7×10^{6}	5.7×10^{-1}	6.7×10^{5}	5.2×10^{-1}	2.3×10^{5}	2.0×10^{-1}	8.5×10^{4}	7.1×10^{-2}
Grand total[b]	8.7×10^{5}	1.3×10^{9}	1.7×10^{2}	7.4×10^{7}	1.1×10	5.9×10^{6}	6.4×10^{-1}	4.1×10^{5}	2.2×10^{-1}	2.2×10^{5}	9.1×10^{-2}

NOTE: This waste is from reprocessing a blended mixture of core and blankets having an average burnup of 45,500 MWthd/MgHM at an average specific power of 41.5 MWth/MgHM. It contains all of the cladding and structural material plus 0.05 percent of the nonvolatile fission products and actinides in the spent fuel.

[a] The fuel is reprocessed 90 days after discharge from the reactor.

[b] Total of all elements, listed or not.

SOURCE: Adapted from the data of Croff, A. G., and C. W. Alexander, 1980. Decay Characteristics of Once-Through LWR and LMFBR Spent Fuels, High-Level Wastes, and Fuel Assembly Structural Material Wastes. ORNL/TM-7431. Oak Ridge National Laboratory, Oak Ridge, Tenn.

TABLE A-7 Properties of Plutonium Having an Isotopic Composition Typical of That to be Present in TRU Waste

Isotope	Weight Fraction	Radioactivity (MBq/g Pu)
^{238}Pu	0.01	6.4×10^3
^{239}Pu	0.6	1.4×10^3
^{240}Pu	0.24	2.0×10^3
^{241}Pu	0.11	4.0×10^5
^{242}Pu	0.04	5.6
Total	1.00	4.1×10^5
Total alpha activity		9.7×10^3

APPENDIX B

CALCULATED INDIVIDUAL DOSES PER UNIT CONCENTRATION OF RADIONUCLIDES RELEASED TO THE ENVIRONMENT: COMPARISON OF RESULTS FROM MODELS OF PACIFIC NORTHWEST LABORATORY (PNL) AND ATOMIC ENERGY OF CANADA LIMITED (AECL)

Table B-1 compares results from two different environmental pathways and dosimetry modeling computer codes that have been used to assess radioactivity releases to the environment from conceptual high-level waste repositories. It is of interest to examine results of independent calculations to gain understanding of the sensitivity of calculated results to assumptions and parameters used to characterize environmental transport and human exposure. As a first step toward this objective, we have used data from results of preliminary calculations performed by the Canadian National Nuclear Fuel Waste Management Program, as reported by Wuschke et al. (1981), for several radionuclides. These are compared with the Battelle Pacific Northwest Laboratory (PNL) data used in the waste-isolation system performance calculations described in Chapter 9.

The most useful comparison, consistent with the performance assessment approach described in Chapter 9, examines dose rates to an individual that result from a unit concentration (1 Bq/m^3) of a radionuclide reaching the environment. In this discussion, "environment" refers to a location or an environmental medium where radionuclides calculated to have been released from a repository are available for causing radiation exposure to humans. However, because of the somewhat different approaches used by the PNL and AECL groups in calculating environmental transport and doses, and because only limited data were available from the AECL calculations (Wuschke et al. 1981), the bases for the comparison in Table B-1 are not completely consistent.

The Table B-1 data taken from Table 9-1 of Chapter 9 give the average yearly dose to an individual calculated from an assumed continuous 70-year intake of contaminated water and food grown from this water. The PNL computer code PABLM, from which these data are derived, calculates an individual's intake as a time-dependent function of the radionuclide concentration in environmental media (Napier et al. 1980). For the comparison in Table B-1, only those pathways to man initiated by contaminated surface water are considered. In the calculation used here, it is assumed that a constant-unit concentration (1 Bq/m^3) of a radionuclide is in the surface water during the 70-year lifetime of an individual. The PABLM code calculates the committed dose equivalent for each year's intake of radioactivity and adds it to the cumulative committed dose from previous yearly incremental intakes. That is, for the first year of the individual's life, the 70-year committed dose that

TABLE B-1 Average Annual dose to an Individual per Unit Concentration of Radioactivity in Water

Radionuclide	Average Annual Dose per Unit Concentration ($Sv\text{-}m^3/Bq\text{-}yr$) Table 9-1[a]	Wuschke et al.[b]
^{79}Se	1.6×10^{-7}	7.9×10^{-9}
^{99}Tc	7.0×10^{-10}	6.1×10^{-10}
^{129}I	2.0×10^{-8}	9.1×10^{-11}
^{135}Cs	5.3×10^{-8}	1.6×10^{-8}
^{210}Pb	7.7×10^{-6} [c]	4.9×10^{-6}
^{225}Ra	1.4×10^{-6}	1.7×10^{-7}
^{226}Ra	2.4×10^{-6} [d]	4.1×10^{-6}
^{229}Th	5.6×10^{-7}	6.4×10^{-6}
^{230}Th	8.0×10^{-8}	3.4×10^{-4}
^{233}U	3.8×10^{-8}	4.8×10^{-8}
^{234}U	3.8×10^{-8}	4.8×10^{-8}
^{238}U	2.9×10^{-8}	4.2×10^{-8}
^{237}Np	1.3×10^{-5} [e]	6.5×10^{-6}
^{239}Pu	9.8×10^{-9}	6.1×10^{-8}

NOTE: All data rounded to two significant digits.

[a] Average annual "total-body" dose resulting from 1 Bq/m^3 in water (constant concentration over 70 years); derived from data of B. A. Napier (Battelle Pacific Northwest Laboratory, personal communication to T. H. Pigford, 1982) for 70-year committed dose from 70-year intake of water and food.

[b] Fifty-year committed effective dose equivalent from water and food taken in during first year; derived from data of Wuschke et al. (1981).

[c] Increased by a factor of 4.4 above data of B. A. Napier (Battelle Pacific Northwest Laboratory, personal communication to T. H. Pigford, 1982) to allow for ICRP-30 corrections (Runkle and Soldat 1982).

[d] Reduced by factor of 90 below data of B. A. Napier (Battelle Pacific Northwest Laboratory, personal communication to T. H. Pigford, 1982) to allow for ICRP-30 corrections (Runkle and Soldat 1982).

[e] Increased by a factor of 200 above data of B. A. Napier (Battelle Pacific Northwest Laboratory, personal communication to T. H. Pigford, 1982) to allow for ICRP-30 corrections (Runkle and Soldat 1982).

results from the intake is calculated; for the second year's intake, the 69-year committed dose is calculated and added to the result from the first year's intake; and so on to obtain the total 70-year lifetime dose. To obtain the average yearly contribution to the committed dose per unit concentration in water (Table 9-1 data in Table B-1), the 70-year lifetime dose per unit concentration of each radionuclide in surface water is divided by 70.

As explained in Appendix C, the doses calculated by the PABLM code for internally deposited radionuclides are based on the physical dosimetry models of the International Commission on Radiological Protection (1959) Publication 2, hereafter referred to as ICRP-2. The ICRP-2 model for the whole body or total body assumes that the internally deposited radionuclide is concentrated at the center of a 70-kg sphere of tissue. The time-dependent metabolic behavior of each radionuclide is characterized by an "effective biological half-life" representative of the body in its entirety (B. A. Napier, Battelle Pacific Northwest Laboratory, personal communication to T. H. Pigford, 1982). It should be noted that in this instance the effective biological half-life is distinguished from the "effective half-life" used in the ICRP-2 dosimetry system, as the latter term includes radioactive decay. The data for lead-210, radium-226, and neptunium-237 from B. A. Napier (Battelle Pacific Northwest Laboratory, personal communication to T. H. Pigford, 1982) have been adjusted to account for ICRP-30 corrections (International Commission on Radiological Protection 1979) according to the method of Runkle and Soldat (1982). The numerical adjustment to each is shown in the footnotes of Table B-1.

The AECL models (Wuschke et al. 1981) calculate 50-year committed dose equivalents to an individual in the form of effective dose equivalent, defined by International Commission on Radiological Protection (1977), Publication 26, as opposed to the total body dose calculated by the PABLM code (Napier et al. 1980), which is used to obtain the PNL doses. The doses from Wuschke et al. in Table B-1 represent the 50-year dose commitment to an individual that results from a unit concentration (1 Bq/m^3) of a radionuclide reaching the "environment" from a repository. These values are, in effect, the maximum annual dose commitment rates to an individual from 1 Bq/m^3 (average annual concentration) in water.

The Canadian models calculate dose to man from terrestrial and aquatic food chains from radionuclides released to groundwater, soil pore water, and surface water. The distribution of radioactivity between the solid and liquid phases in the soil compartment is characterized by the equilibrium distribution coefficient K_d. The value selected for K_d affects the transfer of radioactivity from the soil compartment to the water compartments and to plants. The data from Wuschke et al. in Table B-1 are for a case where the concentrations in all the water compartments are equal. The results given here are for values of K_d selected at the low end of the range of K_d values given for each radionuclide (Wuschke et al. 1981).

The doses shown in Table B-1 from both data sources are calculated for an individual who obtains all his drinking water from the contaminated water and all his food from plants and animals whose water is obtained from the same contaminated water sources. The individual is thus, in effect, the maximally exposed individual at the site characterized by the respective environmental pathway and dosimetry models.

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APPENDIX C

INGESTION DOSE FACTORS FOR RADIONUCLIDES OF INTEREST IN ASSESSMENTS OF HIGH-LEVEL WASTE-ISOLATION SYSTEM PERFORMANCE

Table C-1 contains ingestion dose factors from four separate sources for 30 long-lived radionuclides contained in high-level radioactive waste. The factors give the committed dose equivalent to an individual resulting from the intake in food, liquids, or drinking water of a unit amount of a radionuclide. The table is arrayed to allow comparisons of dose factors from the four sources for each organ-nuclide combination of interest. Each dose factor is given in units of sieverts per becquerel and, immediately beneath, in units of rem per curie.

The first row of dose factors, identified as EPA, are from Appendix F of the report by Smith et al. (1981), which was prepared as part of the supporting analysis for the proposed Environmental Protection Agency (EPA) high-level waste standard. The eight organs included are those used by EPA for calculating the number of fatal cancers in the derivation of the 10,000-year release limits as contained in the proposed standard (U.S. Environmental Protection Agency 1982, Table 2). Not included in this comparison are the dose factors for reproductive organs used by EPA to estimate "first generation" genetic effects. The EPA dose factors are 50-year committed dose equivalents from an initial intake at time zero. The principal source for the EPA dose factors is an early compilation prepared at Oak Ridge National Laboratory of the INREM II internal dosimetry model (Killough et al. 1978; G. G. Killough, Oak Ridge National Laboratory, personal communication to B. J. Mann, 1982). The EPA report by Smith et al. (1981) contains detailed explanations of adjustments to the INREM dose factors made by EPA and identifies other sources of dose factor data used by EPA. The EPA entry in Table C-1 under the total body column heading is identified as the dose to "other organs" in the EPA report (Smith et al. 1981). It is essentially the sum of the dose contributions to a volume equivalent to the "whole body" from the radionuclide deposited in the 22 source organs of the INREM II model (Killough et al. 1978). Care should be taken to distinguish this entry in the table from the total body entries of the other data sources.

The INREM II results in Table C-1 are from a recent update (Dunning et al. 1981) that includes results of some modifications in the INREM II dosimetry and metabolic models, as well as changes in metabolic data for some elements. The total body data entries here are effective dose equivalents, based on the International Commission on Radiological

TABLE C-1 Ingestion Dose Factor Compilation

Nuclide	Bone[a]	Red Marrow[a]	Lung	Liver	GI-LLI	Thyroid	Kidney	Total Body[b]
^{14}C								
EPA[c]	3.2×10^{-10}	9.2×10^{-10}	2.3×10^{-10}	3.2×10^{-10}	4.0×10^{-10}	2.4×10^{-10}	3.0×10^{-10}	5.1×10^{-10}
	1.2×10^{3}	3.4×10^{3}	8.5×10^{2}	1.2×10^{3}	1.5×10^{3}	8.9×10^{2}	1.1×10^{3}	1.9×10^{3}
INREM II[c]	3.2×10^{-10}	9.2×10^{-10}	2.3×10^{-10}	3.2×10^{-10}	4.0×10^{-10}	2.4×10^{-10}	3.0×10^{-10}	4.2×10^{-10}
	1.2×10^{3}	3.4×10^{3}	8.5×10^{2}	1.2×10^{3}	1.5×10^{3}	8.9×10^{2}	1.1×10^{3}	1.5×10^{3}
ICRP-30[c]	5.6×10^{-10}	5.6×10^{-10}	5.6×10^{-10}	5.6×10^{-10}	5.6×10^{-10}	5.6×10^{-10}	5.6×10^{-10}	5.6×10^{-10}
	2.1×10^{3}	2.1×10^{3}	2.1×10^{3}	2.1×10^{3}	2.1×10^{3}	2.1×10^{3}	2.1×10^{3}	2.1×10^{3}
PNL[d]	7.8×10^{-10}	—	1.5×10^{-10}	1.5×10^{-10}	1.5×10^{-10}	1.5×10^{-10}	1.5×10^{-10}	1.5×10^{-10}
	2.9×10^{3}	—	5.7×10^{2}	5.7×10^{2}	5.7×10^{2}	5.7×10^{2}	5.7×10^{2}	5.7×10^{2}
^{90}Sr								
EPA	3.2×10^{-7}	1.2×10^{-7}	4.3×10^{-15}	1.5×10^{-9}	5.4×10^{-8}	1.6×10^{-9}	1.6×10^{-9}	2.6×10^{-8}
	1.2×10^{6}	4.3×10^{5}	1.6×10^{-2}	5.7×10^{3}	2.0×10^{5}	6.0×10^{3}	6.0×10^{3}	9.5×10^{4}
INREM II	3.2×10^{-7}	1.2×10^{-7}	1.6×10^{-9}	1.5×10^{-9}	2.1×10^{-8}	1.6×10^{-9}	1.6×10^{-9}	2.3×10^{-8}
	1.2×10^{6}	4.3×10^{5}	6.0×10^{3}	5.7×10^{3}	7.8×10^{4}	6.0×10^{3}	6.0×10^{3}	8.7×10^{4}
ICRP-30	4.2×10^{-7}	1.9×10^{-7}	1.5×10^{-9}	1.5×10^{-9}	2.0×10^{-8}	1.5×10^{-9}	1.5×10^{-9}	3.9×10^{-8}
	1.5×10^{6}	7.2×10^{5}	5.6×10^{3}	5.6×10^{3}	7.3×10^{4}	5.6×10^{3}	5.6×10^{3}	1.4×10^{5}
PNL	1.0×10^{-6}	—	0.0	0.0	2.1×10^{-8}	0.0	0.0	2.7×10^{-7}
	3.9×10^{6}	—	0.0	0.0	7.7×10^{4}	0.0	0.0	1.0×10^{6}
^{93}ZR								
EPA	5.4×10^{-11}	8.9×10^{-11}	1.0×10^{-11}	3.8×10^{-10}	4.6×10^{-9}	4.6×10^{-9}	5.4×10^{-11}	6.8×10^{-11}
	2.0×10^{2}	3.3×10^{2}	3.9×10	1.4×10^{3}	1.7×10^{4}	1.7×10	2.0×10^{2}	2.5×10^{2}
INREM II	—	—	—	—	—	—	—	—
	—	—	—	—	—	—	—	—
ICRP-30	9.1×10^{-9}	7.4×10^{-10}	1.2×10^{-13}	8.3×10^{-14}	1.0×10^{-9}	7.3×10^{-14}	1.1×10^{-13}	3.6×10^{-10}
	3.4×10^{4}	2.7×10^{3}	4.3×10^{-1}	3.0×10^{-1}	3.7×10^{3}	2.7×10^{-1}	4.0×10^{-1}	1.3×10^{3}
PNL	1.1×10^{-11}	—	0.0	5.9×10^{-13}	7.3×10^{-10}	0.0	2.3×10^{-12}	3.0×10^{-13}
	4.2×10	—	0.0	2.2	2.7×10^{3}	0.0	8.6	1.1
^{99}Tc								
EPA	9.7×10^{-11}	8.7×10^{-11}	0.0	1.7×10^{-10}	8.7×10^{-11}	3.8×10^{-9}	1.2×10^{-10}	5.7×10^{-11}
	3.6×10^{2}	3.2×10^{2}	0.0	6.3×10^{2}	3.2×10^{3}	1.4×10^{4}	4.6×10^{2}	2.1×10^{2}
INREM II	9.7×10^{-11}	8.7×10^{-11}	8.7×10^{-11}	1.7×10^{-10}	8.7×10^{-10}	3.8×10^{-9}	1.2×10^{-10}	2.7×10^{-10}
	3.6×10^{2}	3.2×10^{2}	3.2×10^{2}	6.3×10^{2}	3.2×10^{3}	1.4×10^{4}	4.6×10^{2}	1.0×10^{3}
ICRP-30	6.0×10^{-11}	6.0×10^{-11}	6.0×10^{-11}	8.2×10^{-11}	1.1×10^{-9}	1.6×10^{-9}	6.0×10^{-11}	3.7×10^{-10}
	2.2×10^{2}	2.2×10^{2}	2.2×10^{2}	3.0×10^{2}	4.1×10^{3}	6.0×10^{3}	2.2×10^{2}	1.5×10^{3}
PNL	3.5×10^{-11}	—	4.3×10^{-12}	4.9×10^{-11}	1.8×10^{-12}	0.0	6.2×10^{-13}	1.3×10^{-11}
	1.3×10^{2}	—	1.6×10	1.8×10^{2}	6.8×10^{3}	0.0	2.3×10^{3}	5.0×10
^{126}Sn								
EPA	2.3×10^{-8}	2.3×10^{-8}	8.4×10^{-10}	4.6×10^{-10}	3.2×10^{-8}	1.3×10^{-10}	7.6×10^{-10}	7.6×10^{-10}
	8.6×10^{4}	8.6×10^{4}	3.1×10^{3}	1.7×10^{3}	1.2×10^{5}	5.0×10^{2}	2.8×10^{3}	2.8×10^{3}
INREM II	—	—	—	—	—	—	—	—
	—	—	—	—	—	—	—	—
ICRP-30	5.1×10^{-9}	2.7×10^{-9}	6.0×10^{-10}	6.9×10^{-10}	4.3×10^{-8}	5.5×10^{-10}	8.2×10^{-10}	6.2×10^{-9}
	1.9×10^{4}	1.0×10^{4}	2.2×10^{3}	2.5×10^{3}	1.6×10^{5}	2.0×10^{3}	3.0×10^{3}	2.3×10^{4}
PNL	2.3×10^{-8}	—	0.0	4.3×10^{-10}	7.3×10^{-9}	1.3×10^{-10}	0.0	6.5×10^{-10}
	8.4×10^{4}	—	0.0	1.6×10^{3}	2.7×10^{4}	4.9×10^{2}	0.0	2.4×10^{3}
^{151}Sm								
EPA	1.3×10^{-12}	8.7×10^{-13}	3.0×10^{-14}	4.6×10^{-12}	1.6×10^{-9}	2.7×10^{-14}	1.5×10^{-12}	6.2×10^{-12}
	4.9	3.2	1.1×10^{-1}	1.7×10	5.9×10^{3}	1.0×10^{-1}	5.5	2.3×10
INREM II	—	—	—	—	—	—	—	—
	—	—	—	—	—	—	—	—
ICRP-30	3.5×10^{-10}	2.8×10^{-11}	6.5×10^{-16}	9.2×10^{-11}	1.0×10^{-9}	3.3×10^{-17}	5.2×10^{-16}	9.1×10^{-11}
	1.3×10^{3}	1.0×10^{2}	2.4×10^{3}	3.4×10^{2}	3.7×10^{3}	1.2×10^{4}	1.9×10^{3}	3.4×10^{2}
PNL	1.9×10^{-11}	—	0.0	3.0×10^{-12}	1.6×10^{-9}	0.0	3.5×10^{-12}	7.8×10^{-13}
	6.9×10	—	0.0	1.1×10	5.8×10^{3}	0.0	1.3×10	2.9

TABLE C-1 *(continued)*

Nuclide	Bone[a]	Red Marrow[a]	Lung	Liver	GI-LLI	Thyroid	Kidney	Total Body[b]
^{225}Ra								
EPA	—	—	—	—	—	—	—	—
	—	—	—	—	—	—	—	—
INREM II	3.0×10^{-7}	3.0×10^{-7}	7.0×10^{-8}	7.0×10^{-8}	2.6×10^{-8}	7.0×10^{-8}	7.0×10^{-8}	1.7×10^{-7}
	1.1×10^{6}	1.1×10^{6}	2.6×10^{5}	2.6×10^{5}	9.7×10^{4}	2.6×10^{5}	2.6×10^{5}	6.2×10^{5}
ICRP-30	1.8×10^{-6}	1.7×10^{-7}	3.4×10^{-8}	3.4×10^{-8}	6.4×10^{-8}	3.4×10^{-8}	3.4×10^{-8}	1.0×10^{-7}
	6.6×10^{6}	6.2×10^{5}	1.3×10^{5}	1.3×10^{5}	2.4×10^{5}	1.3×10^{5}	1.3×10^{3}	3.7×10^{5}
PNL	1.8×10^{-6}	—	0.0	2.0×10^{-9}	9.2×10^{-8}	0.0	5.7×10^{-8}	3.5×10^{-7}
	6.6×10^{6}	—	0.0	7.4×10^{3}	3.4×10^{5}	0.0	2.1×10^{5}	1.3×10^{6}
^{226}Ra								
EPA	1.7×10^{-5}	5.7×10^{-7}	7.3×10^{-11}	5.1×10^{-7}	2.2×10^{-7}	2.2×10^{-7}	1.6×10^{-6}	2.1×10^{-6}
	6.3×10^{7}	2.1×10^{6}	2.7×10^{2}	1.9×10^{6}	8.2×10^{5}	8.0×10^{5}	5.8×10^{6}	7.8×10^{6}
INREM II	1.2×10^{-5}	5.9×10^{-7}	1.6×10^{-7}	1.6×10^{-7}	8.9×10^{-8}	1.6×10^{-7}	1.6×10^{-7}	3.8×10^{-7}
	4.3×10^{7}	2.2×10^{6}	5.9×10^{5}	5.9×10^{5}	3.3×10^{5}	5.9×10^{5}	5.9×10^{5}	1.4×10^{6}
ICRP-30	6.8×10^{-6}	6.0×10^{-7}	9.2×10^{-8}	9.1×10^{-8}	1.3×10^{-7}	9.1×10^{-8}	9.2×10^{-8}	3.6×10^{-7}
	3.4×10^{5}	2.2×10^{6}	3.4×10^{5}	4.9×10^{5}	3.4×10^{5}	3.4×10^{5}	3.4×10^{5}	1.3×10^{6}
PNL	1.0×10^{-4}	—	0.0	0.0	1.0×10^{-7}	0.0	0.0	6.8×10^{-5}
	3.7×10^{8}	—	0.0	0.0	3.7×10^{5}	0.0	0.0	2.5×10^{8}
^{229}Th								
EPA	—	—	—	—	—	—	—	—
	—	—	—	—	—	—	—	—
INREM II	8.7×10^{-7}	7.0×10^{-7}	2.7×10^{-9}	9.7×10^{-9}	5.7×10^{-8}	2.5×10^{-9}	2.5×10^{-9}	3.5×10^{-7}
	3.2×10^{6}	2.6×10^{6}	1.0×10^{4}	3.6×10^{4}	2.1×10^{5}	9.4×10^{3}	9.4×10^{3}	1.3×10^{6}
ICRP-30	2.4×10^{-5}	1.9×10^{-6}	4.6×10^{-9}	4.0×10^{-8}	6.3×10^{-8}	4.5×10^{-8}	4.6×10^{-9}	9.5×10^{-7}
	8.8×10^{7}	7.1×10^{6}	1.7×10^{4}	1.5×10^{5}	2.3×10^{5}	1.7×10^{4}	1.7×10^{4}	3.5×10^{6}
ICRP-2	3.0×10^{-6}	—	0.0	4.1×10^{-8}	1.5×10^{-7}	0.0	1.9×10^{-7}	1.4×10^{-7}
	1.1×10^{7}	—	0.0	1.5×10^{3}	5.7×10^{5}	0.0	7.1×10^{5}	5.3×10^{5}
^{230}Th								
EPA	—	—	—	—	—	—	—	—
	—	—	—	—	—	—	—	—
INREM II	3.2×10^{-7}	2.7×10^{-7}	1.2×10^{-8}	5.9×10^{-9}	4.9×10^{-8}	1.2×10^{-9}	1.2×10^{-9}	1.7×10^{-7}
	1.2×10^{6}	1.0×10^{6}	4.6×10^{3}	2.2×10^{4}	1.8×10^{5}	4.6×10^{3}	4.3×10^{3}	6.3×10^{5}
ICRP-30	3.6×10^{-6}	2.9×10^{-7}	6.8×10^{-10}	5.9×10^{-9}	4.9×10^{-8}	6.8×10^{-10}	6.8×10^{-10}	1.5×10^{-7}
	1.3×10^{7}	1.1×10^{6}	2.5×10^{3}	2.2×10^{4}	1.8×10^{5}	2.5×10^{3}	2.5×10^{3}	5.5×10^{5}
PNL	6.8×10^{-7}	—	0.0	4.1×10^{-8}	1.8×10^{-8}	0.0	1.9×10^{-7}	2.1×10^{-8}
	2.5×10^{6}	—	0.0	1.5×10^{5}	6.7×10^{4}	0.0	6.9×10^{5}	7.7×10^{4}
^{231}Pa								
EPA	—	—	—	—	—	—	—	—
	—	—	—	—	—	—	—	—
INREM II	4.9×10^{-6}	4.1×10^{-6}	5.9×10^{-8}	7.6×10^{-6}	5.1×10^{-8}	5.9×10^{-8}	9.7×10^{-7}	2.6×10^{-6}
	1.8×10^{7}	1.5×10^{7}	2.2×10^{5}	2.8×10^{7}	1.9×10^{5}	2.2×10^{5}	3.6×10^{6}	9.5×10^{6}
ICRP-30	7.2×10^{-5}	5.8×10^{-6}	6.8×10^{-11}	4.4×10^{-9}	5.3×10^{-8}	6.3×10^{-11}	6.8×10^{-9}	2.9×10^{-6}
	2.7×10^{8}	2.1×10^{7}	2.5×10^{2}	1.6×10^{4}	2.0×10^{5}	2.3×10^{2}	2.5×10^{4}	1.1×10^{7}
ICRP-2	1.3×10^{-6}	—	0.0	5.4×10^{-8}	2.2×10^{-8}	0.0	3.0×10^{-7}	5.7×10^{-8}
	5.0×10^{6}	—	0.0	2.0×10^{5}	8.0×10^{4}	0.0	1.1×10^{6}	2.1×10^{5}
^{223}U								
EPA	—	—	—	—	—	—	—	—
	—	—	—	—	—	—	—	—
INREM II	2.1×10^{-6}	6.5×10^{-8}	4.6×10^{-9}	4.1×10^{-9}	4.6×10^{-8}	4.3×10^{-9}	4.6×10^{-7}	7.0×10^{-8}
	7.9×10^{6}	2.4×10^{5}	1.7×10^{4}	1.5×10^{4}	1.7×10^{5}	1.6×10^{4}	1.7×10^{6}	2.6×10^{3}
ICRP-30	1.2×10^{-6}	7.4×10^{-8}	2.6×10^{-9}	2.6×10^{-9}	5.0×10^{-8}	2.6×10^{-9}	4.7×10^{-7}	7.8×10^{-8}
	4.3×10^{6}	2.7×10^{5}	9.7×10^{3}	9.7×10^{3}	1.8×10^{5}	9.7×10^{3}	1.7×10^{6}	2.9×10^{5}
PNL	2.3×10^{-7}	—	0.0	0.0	1.9×10^{-8}	0.0	5.4×10^{-8}	1.4×10^{-8}
	8.7×10^{5}	—	0.0	0.0	6.9×10^{4}	0.0	2.0×10^{5}	5.3×10^{4}

TABLE C-1 *(continued)*

Nuclide	Bone[a]	Red Marrow[a]	Lung	Liver	GI-LLI	Thyroid	Kidney	Total Body[b]
^{234}U								
EPA	5.4×10^{-6}	2.2×10^{-7}	2.2×10^{-10}	1.6×10^{-7}	2.4×10^{-8}	1.6×10^{-7}	1.6×10^{-7}	4.6×10^{-7}
	2.0×10^{7}	8.0×10^{3}	8.2×10^{2}	5.8×10^{5}	8.9×10^{4}	5.8×10^{5}	8.5×10^{5}	1.7×10^{6}
INREM II	2.1×10^{-6}	6.2×10^{-8}	4.6×10^{-9}	4.3×10^{-9}	4.6×10^{-8}	4.3×10^{-9}	4.6×10^{-7}	7.0×10^{-9}
	7.8×10^{6}	2.3×10^{5}	1.7×10^{4}	1.6×10^{4}	1.7×10^{5}	1.6×10^{4}	1.7×10^{6}	2.6×10^{5}
ICRP-30	1.1×10^{-6}	7.2×10^{-8}	2.6×10^{-9}	2.6×10^{-9}	4.9×10^{-8}	2.6×10^{-9}	4.7×10^{-7}	7.7×10^{-8}
	4.2×10^{6}	2.7×10^{5}	9.5×10^{3}	9.5×10^{3}	1.8×10^{5}	9.5×10^{3}	1.7×10^{6}	2.9×10^{5}
PNL	2.3×10^{-7}	—	0.0	0.0	1.8×10^{-8}	0.0	5.1×10^{-8}	1.4×10^{-8}
	8.4×10^{5}	—	0.0	0.0	6.8×10^{4}	0.0	1.9×10^{5}	5.2×10^{4}
^{235}U								
EPA	—	—	—	—	—	—	—	—
	—	—	—	—	—	—	—	—
INREM II	1.9×10^{-6}	4.9×10^{-8}	4.3×10^{-9}	3.2×10^{-10}	4.9×10^{-8}	4.1×10^{-9}	4.1×10^{-7}	5.9×10^{-8}
	7.1×10^{6}	1.8×10^{3}	1.6×10^{4}	1.4×10^{4}	1.8×10^{5}	1.5×10^{4}	1.5×10^{6}	2.2×10^{5}
ICRP-30	1.1×10^{-6}	5.8×10^{-8}	2.5×10^{-9}	2.5×10^{-9}	5.3×10^{-8}	2.5×10^{-9}	4.3×10^{-7}	7.2×10^{-8}
	4.1×10^{6}	2.5×10^{5}	9.1×10^{3}	9.1×10^{3}	2.0×10^{5}	9.1×10^{3}	1.6×10^{6}	2.7×10^{5}
PNL	2.2×10^{-7}	—	0.0	0.0	2.3×10^{-8}	0.0	4.9×10^{-8}	1.3×10^{-8}
	8.0×10^{5}	—	0.0	0.0	8.6×10^{4}	0.0	1.8×10^{5}	4.9×10^{4}
^{238}U								
EPA	—	—	—	—	—	—	—	—
	—	—	—	—	—	—	—	—
INREM II	1.9×10^{-6}	5.1×10^{-8}	4.1×10^{-9}	3.2×10^{-10}	4.0×10^{-10}	3.8×10^{-9}	4.1×10^{-7}	5.9×10^{-8}
	7.0×10^{6}	1.9×10^{3}	1.5×10^{4}	1.3×10^{4}	1.6×10^{5}	1.4×10^{4}	1.5×10^{6}	2.2×10^{5}
ICRP-30	1.0×10^{-6}	6.8×10^{-8}	2.3×10^{-9}	2.3×10^{-9}	4.6×10^{-8}	2.3×10^{-9}	4.1×10^{-7}	6.9×10^{-8}
	3.7×10^{6}	2.5×10^{5}	8.5×10^{3}	8.5×10^{3}	1.7×10^{5}	8.5×10^{3}	1.5×10^{6}	2.5×10^{5}
PNL	2.1×10^{-7}	—	0.0	0.0	1.7×10^{-8}	0.0	4.6×10^{-8}	1.2×10^{-8}
	7.7×10^{5}	—	0.0	0.0	6.1×10^{4}	0.0	1.7×10^{5}	4.5×10^{4}
^{237}Np								
EPA	5.1×10^{-6}	1.7×10^{-6}	2.4×10^{-10}	2.2×10^{-6}	4.1×10^{-8}	1.7×10^{-8}	3.0×10^{-7}	4.3×10^{-7}
	1.9×10^{7}	6.2×10^{6}	8.9×10^{2}	8.2×10^{6}	1.5×10^{3}	6.1×10^{4}	1.1×10^{6}	1.6×10^{6}
INREM II	2.0×10^{-6}	1.7×10^{-6}	3.2×10^{-8}	4.3×10^{-6}	6.2×10^{-8}	3.2×10^{-8}	3.7×10^{-7}	1.3×10^{-6}
	7.3×10^{6}	6.1×10^{6}	1.2×10^{5}	1.6×10^{7}	2.0×10^{5}	1.2×10^{5}	2.1×10^{6}	4.7×10^{6}
ICRP-30	1.9×10^{-4}	1.5×10^{-5}	2.4×10^{-9}	4.1×10^{-5}	5.4×10^{-8}	8.2×10^{-10}	3.1×10^{-9}	8.5×10^{-6}
	7.1×10^{8}	5.6×10^{7}	8.8×10^{3}	1.5×10^{8}	2.0×10^{5}	3.0×10^{3}	1.1×10^{4}	3.1×10^{7}
PNL	4.6×10^{-7}	—	0.0	2.5×10^{-7}	2.4×10^{-8}	0.0	1.5×10^{-7}	2.0×10^{-8}
	1.7×10^{6}	—	0.0	9.3×10^{5}	8.8×10^{4}	0.0	5.4×10^{5}	7.3×10^{4}
^{238}Pu								
EPA	1.3×10^{-7}	4.6×10^{-8}	2.1×10^{-14}	5.9×10^{-8}	3.0×10^{-8}	4.3×10^{-10}	7.8×10^{-9}	1.2×10^{-12}
	5.0×10^{5}	1.7×10^{5}	7.9×10^{2}	2.2×10^{5}	1.1×10^{5}	1.6×10^{3}	2.9×10^{4}	4.3×10^{4}
INREM II	5.4×10^{-8}	4.6×10^{-8}	8.7×10^{-10}	1.2×10^{-7}	5.7×10^{-8}	8.7×10^{-10}	1.5×10^{-8}	3.8×10^{-8}
	2.0×10^{5}	1.7×10^{5}	3.2×10^{3}	4.4×10^{5}	2.1×10^{5}	3.2×10^{3}	5.7×10^{4}	1.4×10^{5}
ICRP-30	1.8×10^{-6}	1.5×10^{-8}	8.7×10^{-15}	4.0×10^{-8}	5.7×10^{-8}	3.9×10^{-16}	1.2×10^{-14}	1.1×10^{-7}
	6.8×10^{5}	5.4×10^{4}	3.2×10^{2}	1.5×10^{5}	2.1×10^{5}	1.5×10^{-3}	4.4×10^{-2}	4.1×10^{5}
PNL	1.2×10^{-7}	—	0.0	7.0×10^{-8}	2.2×10^{-8}	0.0	2.3×10^{-8}	5.9×10^{-9}
	4.4×10^{5}	—	0.0	2.6×10^{5}	8.1×10^{4}	0.0	8.7×10^{4}	2.2×10^{4}
^{239}Pu								
EPA	1.5×10^{-7}	5.1×10^{-8}	1.7×10^{-14}	6.8×10^{-8}	2.7×10^{-8}	5.1×10^{-10}	8.7×10^{-9}	1.3×10^{-8}
	5.7×10^{5}	1.9×10^{5}	6.1×10^{-2}	2.5×10^{5}	9.9×10^{4}	1.9×10^{3}	3.2×10^{4}	4.8×10^{4}
INREM II	5.9×10^{-8}	5.1×10^{-8}	9.7×10^{-10}	1.3×10^{-7}	5.4×10^{-8}	9.7×10^{-10}	1.7×10^{-8}	4.3×10^{-8}
	2.2×10^{5}	1.9×10^{5}	3.6×10^{3}	4.9×10^{5}	2.0×10^{5}	3.6×10^{3}	6.3×10^{4}	1.6×10^{5}
ICRP-30	2.1×10^{-6}	1.7×10^{-8}	5.3×10^{-15}	4.4×10^{-8}	5.3×10^{-8}	4.4×10^{-16}	1.4×10^{-14}	1.2×10^{-7}
	6.5×10^{5}	6.1×10^{4}	2.1×10^{-2}	1.6×10^{5}	2.0×10^{5}	1.6×10^{-3}	5.1×10^{-2}	4.4×10^{5}
PNL	1.5×10^{-7}	—	0.0	8.1×10^{-8}	2.0×10^{-8}	0.0	2.7×10^{-8}	7.0×10^{-9}
	5.4×10^{5}	—	0.0	3.0×10^{5}	7.4×10^{4}	0.0	1.0×10^{5}	2.6×10^{4}

TABLE C-1 *(continued)*

Nuclide	Bone[a]	Red Marrow[a]	Lung	Liver	GI-LLI	Thyroid	Kidney	Total Body[b]
^{240}Pu								
EPA	1.5×10^{-7} 5.7×10^{5}	5.1×10^{-8} 1.9×10^{5}	2.2×10^{-14} 8.3×10^{-2}	6.8×10^{-8} 2.5×10^{5}	2.7×10^{-8} 9.9×10^{4}	4.9×10^{-10} 1.8×10^{3}	8.7×10^{-9} 3.2×10^{4}	1.3×10^{-8} 4.8×10^{4}
INREM II	5.9×10^{-8} 2.2×10^{5}	5.1×10^{-8} 1.9×10^{5}	9.7×10^{-10} 3.6×10^{3}	1.3×10^{-7} 4.9×10^{5}	5.4×10^{-8} 2.0×10^{5}	9.7×10^{-10} 3.6×10^{3}	1.7×10^{-8} 6.3×10^{4}	4.3×10^{-8} 1.6×10^{5}
ICRP-30	2.1×10^{-7} 7.6×10^{5}	1.7×10^{-8} 6.1×10^{4}	9.7×10^{-15} 3.6×10^{3}	4.4×10^{-8} 1.6×10^{5}	5.3×10^{-8} 2.0×10^{5}	4.8×10^{-16} 1.8×10^{-3}	1.3×10^{-14} 5.0×10^{2}	1.2×10^{-7} 4.4×10^{5}
PNL	1.5×10^{-7} 5.4×10^{5}	— —	0.0 0.0	8.1×10^{-8} 3.0×10^{5}	2.0×10^{-8} 7.5×10^{4}	0.0 0.0	2.7×10^{-8} 1.0×10^{5}	7.0×10^{-9} 2.6×10^{4}
^{241}Pu								
EPA	— —	— —	— —	— —	— —	— —	— —	— —
INREM II	1.2×10^{-9} 4.6×10^{3}	1.0×10^{-9} 3.9×10^{3}	2.0×10^{-11} 7.5×10	2.6×10^{-9} 9.5×10^{3}	2.7×10^{-10} 9.9×10^{2}	2.0×10^{-11} 7.5×10	3.2×10^{-10} 1.3×10^{3}	7.6×10^{-10} 2.8×10^{3}
ICRP-30	4.2×10^{-9} 1.6×10^{4}	3.4×10^{-10} 1.3×10^{3}	6.5×10^{-15} 2.4×10^{-2}	8.6×10^{-10} 3.2×10^{3}	2.7×10^{-10} 1.0×10^{3}	1.2×10^{-15} 4.6×10^{3}	7.6×10^{-15} 2.8×10^{-2}	2.4×10^{-9} 8.9×10^{3}
PNL	2.4×10^{-9} 9.0×10^{3}	— —	0.0 0.0	5.7×10^{-10} 2.1×10^{3}	4.3×10^{-10} 1.6×10^{3}	0.0 0.0	4.1×10^{-10} 1.5×10^{3}	9.2×10^{-11} 3.4×10^{2}
^{242}Pu								
EPA	1.5×10^{-7} 5.4×10^{5}	4.9×10^{-8} 1.8×10^{5}	4.3×10^{-14} 1.6×10^{-1}	6.5×10^{-8} 2.4×10^{5}	2.5×10^{-8} 9.4×10^{4}	4.9×10^{-10} 1.8×10^{3}	8.4×10^{-9} 3.1×10^{4}	1.2×10^{-8} 4.6×10^{4}
INREM II	5.7×10^{-8} 2.1×10^{5}	4.9×10^{-8} 1.8×10^{5}	9.5×10^{-10} 3.5×10^{3}	1.3×10^{-7} 4.7×10^{5}	5.1×10^{-8} 1.9×10^{5}	9.5×10^{-10} 3.5×10^{3}	1.6×10^{-8} 6.0×10^{4}	4.1×10^{-8} 1.5×10^{5}
ICRP-30	2.0×10^{-7} 7.3×10^{5}	1.6×10^{-8} 5.8×10^{4}	2.4×10^{-14} 8.7×10^{2}	4.2×10^{-8} 1.6×10^{5}	5.1×10^{-8} 1.9×10^{5}	3.3×10^{-15} 1.2×10^{-2}	1.1×10^{-13} 4.0×10^{-1}	1.1×10^{-7} 4.1×10^{5}
PNL	1.4×10^{-7} 5.0×10^{5}	— —	0.0 0.0	7.8×10^{-8} 2.9×10^{5}	2.0×10^{-8} 7.3×10^{4}	0.0 0.0	2.7×10^{-8} 9.9×10^{4}	6.8×10^{-9} 2.5×10^{4}
^{241}Am								
EPA	5.1×10^{-6} 1.9×10^{7}	1.7×10^{-6} 6.4×10^{6}	3.5×10^{-11} 1.3×10^{2}	2.3×10^{-6} 8.5×10^{6}	3.0×10^{-8} 1.1×10^{5}	1.7×10^{-8} 6.3×10^{4}	3.0×10^{-7} 1.1×10^{6}	4.3×10^{-7} 1.6×10^{6}
INREM II	2.1×10^{-6} 7.6×10^{6}	1.7×10^{-6} 6.4×10^{6}	3.2×10^{-8} 1.2×10^{5}	4.6×10^{-6} 1.7×10^{7}	5.7×10^{-8} 2.1×10^{5}	3.2×10^{-8} 1.2×10^{5}	5.9×10^{-7} 2.2×10^{6}	1.2×10^{-6} 4.6×10^{6}
ICRP-30	1.0×10^{-5} 3.9×10^{7}	8.4×10^{-7} 3.1×10^{6}	1.7×10^{-11} 6.4×10	2.3×10^{-6} 8.4×10^{6}	5.8×10^{-8} 2.1×10^{5}	3.1×10^{-12} 1.1×10	2.3×10^{-11} 8.4×10	5.9×10^{-7} 2.2×10^{6}
PNL	4.9×10^{-7} 1.8×10^{6}	— —	0.0 0.0	2.7×10^{-7} 1.0×10^{6}	2.2×10^{-8} 8.2×10^{4}	0.0 0.0	1.4×10^{-7} 5.0×10^{5}	1.8×10^{-8} 6.7×10^{4}
^{242}Am								
EPA	— —	— —	— —	— —	— —	— —	— —	— —
INREM II	2.0×10^{-6} 7.5×10^{6}	1.7×10^{-12} 6.3×10^{6}	3.2×10^{-8} 1.2×10^{5}	4.3×10^{-6} 1.6×10^{7}	9.2×10^{-9} 3.4×10^{4}	3.2×10^{-8} 1.2×10^{5}	5.7×10^{-7} 2.1×10^{6}	1.1×10^{-6} 4.1×10^{6}
ICRP-30	1.0×10^{-5} 3.8×10^{7}	8.3×10^{-7} 3.1×10^{6}	1.1×10^{-11} 4.0×10	2.2×10^{-6} 8.1×10^{6}	9.7×10^{-9} 3.6×10^{4}	2.2×10^{-12} 8.1	1.3×10^{-11} 4.9×10	5.7×10^{-7} 2.1×10^{6}
PNL	4.6×10^{-7} 1.7×10^{6}	— —	0.0 0.0	2.6×10^{-7} 9.5×10^{5}	2.7×10^{-8} 1.0×10^{5}	0.0 0.0	1.3×10^{-7} 4.9×10^{5}	1.8×10^{-8} 6.6×10^{4}
^{243}Am								
EPA	5.1×10^{-6} 1.9×10^{7}	8.6×10^{-6} 3.2×10^{7}	2.6×10^{-10} 9.6×10^{2}	2.3×10^{-6} 8.5×10^{6}	4.1×10^{-8} 1.5×10^{5}	1.7×10^{-8} 6.3×10^{4}	3.0×10^{-8} 1.1×10^{6}	4.3×10^{-7} 1.6×10^{6}
INREM II	2.1×10^{-6} 7.6×10^{6}	1.7×10^{-6} 6.4×10^{6}	3.5×10^{-8} 1.3×10^{5}	4.6×10^{-6} 1.7×10^{7}	5.9×10^{-8} 2.2×10^{5}	3.5×10^{-8} 1.3×10^{5}	5.9×10^{-7} 2.2×10^{6}	1.3×10^{-6} 4.7×10^{6}
ICRP-30	1.0×10^{-5} 3.9×10^{7}	8.4×10^{-7} 3.1×10^{6}	1.2×10^{-10} 4.6×10^{2}	2.3×10^{-6} 8.4×10^{6}	6.0×10^{-8} 2.2×10^{5}	3.6×10^{-11} 1.3×10^{2}	1.7×10^{-10} 6.4×10^{2}	5.9×10^{-7} 2.2×10^{6}
PNL	4.9×10^{-7} 1.8×10^{6}	— —	0.0 0.0	2.7×10^{-7} 1.0×10^{6}	2.6×10^{-8} 9.7×10^{4}	0.0 0.0	1.3×10^{-7} 5.0×10^{5}	1.8×10^{-8} 6.7×10^{4}

TABLE C-1 *(continued)*

Nuclide	Bone[a]	Red Marrow[a]	Lung	Liver	GI-LLI	Thyroid	Kidney	Total Body[b]
^{242}Cm								
EPA	—	—	—	—	—	—	—	—
	—	—	—	—	—	—	—	—
INREM II	4.6×10^{-8}	3.8×10^{-8}	7.3×10^{-10}	1.2×10^{-7}	6.2×10^{-8}	7.3×10^{-10}	1.5×10^{-8}	1.9×10^{-7}
	1.7×10^{5}	1.4×10^{5}	2.7×10^{3}	4.4×10^{5}	2.3×10^{5}	2.7×10^{3}	5.7×10^{4}	7.2×10^{5}
ICRP-30	2.3×10^{-7}	1.8×10^{-8}	1.3×10^{-14}	6.0×10^{-8}	6.3×10^{-8}	5.3×10^{-16}	1.7×10^{-14}	1.9×10^{-8}
	8.5×10^{5}	6.8×10^{4}	4.8×10^{-2}	2.2×10^{3}	2.3×10^{5}	2.0×10^{-3}	6.4×10^{-2}	7.0×10^{4}
PNL	8.4×10^{-9}	—	0.0	6.2×10^{-9}	2.4×10^{-8}	0.0	1.6×10^{-9}	3.8×10^{-10}
	3.1×10^{4}	—	0.0	2.3×10^{4}	8.8×10^{4}	0.0	6.0×10^{3}	1.4×10^{3}
^{243}Cm								
EPA	—	—	—	—	—	—	—	—
	—	—	—	—	—	—	—	—
INREM II	1.4×10^{-6}	1.1×10^{-6}	2.2×10^{-8}	3.2×10^{-6}	6.8×10^{-8}	2.2×10^{-8}	4.1×10^{-7}	7.8×10^{-7}
	5.0×10^{6}	4.2×10^{6}	8.3×10^{4}	1.2×10^{7}	2.5×10^{5}	8.2×10^{4}	1.5×10^{6}	2.9×10^{6}
ICRP-30	7.0×10^{-6}	5.6×10^{-7}	4.4×10^{-11}	1.6×10^{-6}	6.7×10^{-8}	1.3×10^{-11}	7.5×10^{-11}	4.0×10^{-7}
	3.0×10^{7}	2.1×10^{6}	1.6×10^{2}	5.8×10^{6}	2.5×10^{5}	5.0×10	2.8×10^{2}	1.5×10^{6}
PNL	2.7×10^{-7}	—	0.0	1.7×10^{-7}	2.4×10^{-8}	0.0	5.1×10^{-8}	1.1×10^{-8}
	1.0×10^{6}	—	0.0	6.4×10^{5}	8.7×10^{4}	0.0	1.9×10^{5}	4.2×10^{4}
^{244}Cm								
EPA	—	—	—	—	—	—	—	—
	—	—	—	—	—	—	—	—
INREM II	1.1×10^{-6}	8.9×10^{-7}	1.7×10^{-8}	2.5×10^{-6}	5.9×10^{-8}	1.7×10^{-8}	3.2×10^{-8}	6.2×10^{-7}
	3.9×10^{6}	3.3×10^{6}	6.4×10^{4}	9.3×10^{6}	2.2×10^{5}	6.4×10^{4}	1.2×10^{6}	2.3×10^{6}
ICRP-30	5.5×10^{-6}	4.4×10^{-7}	2.6×10^{-13}	1.3×10^{-6}	6.0×10^{-8}	2.6×10^{-14}	2.3×10^{-13}	3.1×10^{-7}
	2.0×10^{7}	1.6×10^{6}	9.5×10^{-1}	4.7×10^{6}	2.2×10^{5}	9.5×10^{-2}	8.4×10^{-1}	1.1×10^{6}
PNL	2.0×10^{-7}	—	0.0	1.3×10^{-7}	2.3×10^{-8}	0.0	3.8×10^{-8}	8.4×10^{-9}
	7.4×10^{5}	—	0.0	4.8×10^{5}	8.4×10^{4}	0.0	1.4×10^{5}	3.1×10^{4}

NOTE: First row of entries for each data source is in units of Sv/Bq; the second row is in units of rem/Ci. Dashes indicate no data available. A value of 0.0 indicates dose factor not calculated for particular organ. PNL's PABLM dose factors are based upon the ICRP-2 critical organ model, which does not yield doses from "cross irradiation" due to radionuclides deposited in other organs.

[a] EPA, INREM II, and ICRP-30 entries for bone are for bone surface. Portion of bone considered in PNL model (ICRP-2 bone dosimetry model) for bone dose is nuclide dependent. Nuclides are classified as surface seekers or volume seekers.

[b] EPA and PNL entries in "Total Body" column are for dose equivalents to a tissue volume equivalent to the whole body. The EPA dose factors (based on early INREM results) are the sums of dose contributions from a radionuclide deposited in 22 "source" organs. The PNL dose factors are the doses from a radionuclide deposited at the center of an equivalent sphere. The INREM II and ICRP-30 entries are effective dose equivalents.

[c] Values for EPA, INREM II, and ICRP-30 throughout table are for 50-year committed dose equivalents from initial intake.

[d] Values for PNL throughout table are for 70-year committed dose equivalents from intake at constant rate during first year.

Protection (1977) Publication 26 dose limitation system and described by Dunning and Killough (1981). The effective dose equivalents are from Dunning and Killough (1981). The recent INREM II data have been included in the table to enable a comparison with the EPA data.

The ICRP-30 dose factors in Table C-1 are from the recently revised recommendations of the International Commission on Radiological Protection on limits for intake of radionuclides by radiation workers. These were provided by K. Eckerman (Oak Ridge National Laboratory, personal communication to B. J. Mann, 1982) from the data developed in support of the International Commission on Radiological Protection

(1979) Publication 30 and supplements. As with the EPA and INREM II dose factors, the ICRP-30 factors are 50-year committed dose equivalents from an initial intake. Data in the total body column are effective dose equivalents.

The PNL entries in Table C-1 consist of dose factors calculated by the PABLM computer code at Battelle Pacific Northwest Laboratory (Napier et al. 1980). The data were provided by B. A. Napier (Battelle Pacific Northwest Laboratory, personal communication to B. J. Mann, 1982). The PABLM code is largely based on the physical dosimetry and metabolic models of the International Commission on Radiological Protection (1959) Publication 2. The PNL dose factors are defined as 70-year committed dose equivalents per unit (becquerels) intake, taken in at a constant rate during the first year. The total body entry is the dose equivalent to an equivalent sphere from an amount of radionuclide assumed to be concentrated at the center (Napier et al. 1980). PABLM is a fairly general environmental pathway transport and dosimetry modeling code. For high-level waste performance assessments, PABLM is usually used to calculate cumulative 70-year dose commitments to individuals from a 70-year lifetime of intake (Cloninger and Cole 1981). Dose factors calculated on this latter basis are presented in Table 9-1.

Computer codes developed at Sandia National Laboratories for assessment of high-level waste isolation use environmental pathway and dosimetry models that are derived from the PNL methods (Runkle et al. 1981). Seventy-year dose factors provided by G. E. Runkle (Sandia National Laboratories, personal communication to B. J. Mann, 1982) are not included here because they are essentially identical to the PNL dose factors.

For strontium-90 and the isotopes of uranium, neptunium, and plutonium, dose factors have been calculated for chemical compounds of differing solubility and, hence, differing metabolism and resultant dose per unit amount ingested. The effects of solubility in the ingestion dose models are described by the fractional uptake from the gut to the blood stream, designated by the parameter f_1. Dose factors shown in Table C-1 from INREM II, ICRP-30, and PNL were selected to correspond to the values of f_1 used by EPA (Smith et al. 1981). All data have been rounded to two significant digits.

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