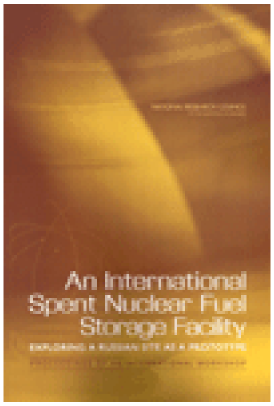


An International Spent Nuclear Fuel Storage Facility -- Exploring a Russian Site as a Prototype: Proceedings of an International Workshop

Glenn E. Schweitzer and A. Chelsea Sharber, Editors, Committee on the Scientific Aspects of an International Spent Fuel Repository in Russia, Office for Central Europe and Eurasia, National Research Council, Russian Academy of Sciences

ISBN: 0-309-55154-4, 302 pages, 8 1/2 x 11, (2005)

This free PDF was downloaded from:
http://www.nap.edu/catalog/11320.html

Visit the National Academies Press online, the authoritative source for all books from the National Academy of Sciences, the National Academy of Engineering, the Institute of Medicine, and the National Research Council:

- Download hundreds of free books in PDF
- Read thousands of books online, free
- Sign up to be notified when new books are published
- Purchase printed books
- Purchase PDFs
- Explore with our innovative research tools

Thank you for downloading this free PDF. If you have comments, questions or just want more information about the books published by the National Academies Press, you may contact our customer service department toll-free at 888-624-8373, visit us online, or send an email to comments@nap.edu.

This free book plus thousands more books are available at http://www.nap.edu.

Copyright © National Academy of Sciences. Permission is granted for this material to be shared for noncommercial, educational purposes, provided that this notice appears on the reproduced materials, the Web address of the online, full authoritative version is retained, and copies are not altered. To disseminate otherwise or to republish requires written permission from the National Academies Press.

THE NATIONAL ACADEMIES
Advisers to the Nation on Science, Engineering, and Medicine

An International Spent Nuclear Fuel Storage Facility

EXPLORING A RUSSIAN SITE AS A PROTOTYPE

PROCEEDINGS OF AN INTERNATIONAL WORKSHOP

Glenn E. Schweitzer and A. Chelsea Sharber, *Editors*

Committee on the Scientific Aspects
of an International Spent Nuclear Fuel Storage Facility in Russia

Office for Central Europe and Eurasia
Development, Security, and Cooperation
Policy and Global Affairs

NATIONAL RESEARCH COUNCIL
OF THE NATIONAL ACADEMIES

In cooperation with the Russian Academy of Sciences

THE NATIONAL ACADEMIES PRESS
Washington, D.C.
www.nap.edu

THE NATIONAL ACADEMIES PRESS • 500 Fifth Street, N.W. • Washington, DC 20001

NOTICE: The project that is the subject of this report was approved by the Governing Board of the National Research Council, whose members are drawn from the councils of the National Academy of Sciences, the National Academy of Engineering, and the Institute of Medicine. The members of the committee responsible for the report were chosen for their special competences and with regard for appropriate balance.

This study was supported by a grant from The Russell Family Foundation. Any opinions, findings, conclusions, or recommendations expressed in this publication are those of the author(s) and do not necessarily reflect the views of the organizations or agencies that provided support for the project.

International Standard Book Number 0-309-09688-X

A limited number of copies are available from the Office for Central Europe and Eurasia, National Research Council, 500 Fifth Street, N.W., Washington, DC 20001; (202) 334-2644.

Additional copies of this report are available from the National Academies Press, 500 Fifth Street, N.W., Lockbox 285, Washington, DC 20055; (800) 624-6242 or (202) 334-3313 (in the Washington metropolitan area); Internet, http://www.nap.edu

Printed in the United States of America

THE NATIONAL ACADEMIES
Advisers to the Nation on Science, Engineering, and Medicine

The **National Academy of Sciences** is a private, nonprofit, self-perpetuating society of distinguished scholars engaged in scientific and engineering research, dedicated to the furtherance of science and technology and to their use for the general welfare. Upon the authority of the charter granted to it by the Congress in 1863, the Academy has a mandate that requires it to advise the federal government on scientific and technical matters. Dr. Ralph J. Cicerone is president of the National Academy of Sciences.

The **National Academy of Engineering** was established in 1964, under the charter of the National Academy of Sciences, as a parallel organization of outstanding engineers. It is autonomous in its administration and in the selection of its members, sharing with the National Academy of Sciences the responsibility for advising the federal government. The National Academy of Engineering also sponsors engineering programs aimed at meeting national needs, encourages education and research, and recognizes the superior achievements of engineers. Dr. Wm. A. Wulf is president of the National Academy of Engineering.

The **Institute of Medicine** was established in 1970 by the National Academy of Sciences to secure the services of eminent members of appropriate professions in the examination of policy matters pertaining to the health of the public. The Institute acts under the responsibility given to the National Academy of Sciences by its congressional charter to be an adviser to the federal government and, upon its own initiative, to identify issues of medical care, research, and education. Dr. Harvey V. Fineberg is president of the Institute of Medicine.

The **National Research Council** was organized by the National Academy of Sciences in 1916 to associate the broad community of science and technology with the Academy's purposes of furthering knowledge and advising the federal government. Functioning in accordance with general policies determined by the Academy, the Council has become the principal operating agency of both the National Academy of Sciences and the National Academy of Engineering in providing services to the government, the public, and the scientific and engineering communities. The Council is administered jointly by both Academies and the Institute of Medicine. Dr. Ralph J. Cicerone and Dr. Wm. A. Wulf are chair and vice chair, respectively, of the National Research Council.

www.national-academies.org

Copyright © National Academy of Sciences. All rights reserved.

NATIONAL RESEARCH COUNCIL COMMITTEE ON THE SCIENTIFIC ASPECTS OF AN INTERNATIONAL SPENT NUCLEAR FUEL STORAGE FACILITY IN RUSSIA

Milton Levenson, *Chair*, Bechtel International (retired)
John F. Ahearne, Sigma Xi, The Scientific Research Society
John H. Kessler, Electric Power Research Institute

Study Staff

Glenn E. Schweitzer, Program Director, National Research Council
Kelly Robbins, Senior Program Officer, National Research Council
A. Chelsea Sharber, Senior Program Associate, National Research Council
Amy Moore, Program Assistant, National Research Council

RUSSIAN ACADEMY OF SCIENCES ORGANIZING COMMITTEE

Nikolay P. Laverov, Russian Academy of Sciences
Vasily I. Velichkin, Institute of Geology of Ore Deposits, Mineralogy, and Geochemistry, Russian Academy of Sciences
Yury K. Shiyan, Foreign Relations Department, Russian Academy of Sciences

Preface

After several years of contentious debate in Russia, the Russian government enacted three laws that permit the importation and storage in Russia of spent nuclear fuel from reactors. A number of foreign governments have expressed interest to the Russian government concerning the possibility of transferring their material to Russia on a temporary or permanent basis.

Much of the material that might be shipped to Russia is of U.S. origin, that is, uranium provided by the United States to other governments that has been used in fuel rods. Some might not be of U.S. origin. The transfer of U.S.-origin spent fuel to Russia requires U.S. government approval, and the U.S. government is not prepared to approve such transfers at this time.

Despite this political uncertainty, Russian officials and scientists are preparing for shipments within the next decade. The income that Russia could potentially derive during the initial few years from accepting and storing spent fuel has been estimated in billions of dollars, although the interest of governments in shipping spent fuel to Russia has yet to be determined.

The details of the plans for establishing a repository in Russia are in their early stages of formulation. The location of the site is still being debated. The exact configuration of the site is under discussion. The view of many Russian specialists is that it should be able to accommodate interim storage of 25–50 years or longer and also provide for permanent burial, even though current Russian law prohibits permanent retention in Russia. There are many technical questions concerning the design and operation of the facility and, of course, there are concerns over the ecological aspects and the security of the material both at the site and en route.

At the suggestion of the Russian Academy of Sciences to the National Academies, an interacademy workshop of international experts on the scientific issues relevant to the establishment and operation of a storage facility was held in Moscow on May 14–15, 2003. The participants discussed a range of issues drawing on related experience in the United States, Russia, Japan, South Korea, and

Switzerland. There was no attempt to be comprehensive in the discussions, and many relevant topics were left for consideration at other meetings.

Following the workshop a group of the participants traveled to Krasnokamensk, a small town near the Siberian city of Chita close to the border with China. There they viewed uranium mining activities and discussed the possibility of locating the international facility at this site.

We have not attempted to summarize in this report the papers presented at the workshop or the discussions at the workshop or in Krasnokamensk. We simply note that these two venues provided excellent opportunities for informative discussions and exchanges of ideas concerning the international facility.

ACKNOWLEDGMENTS

This publication was made possible by a grant from The Russell Family Foundation. The statements made and views expressed are solely the responsibility of the individual authors and do not represent the positions of the foundation, the National Academies, the Russian Academy of Sciences, or other organizations where the authors are employed.

This volume has been reviewed in draft form by several individuals chosen for their technical expertise, in accordance with procedures approved by the National Research Council's Report Review Committee. The purpose of this independent review is to provide candid and critical comments that will assist the institution in ensuring that the report is as sound as possible and meets institutional standards for quality. The review comments and original draft manuscript remain confidential to protect the integrity of the process.

We wish to thank the following individuals for their review of selected papers: William Arnold, Westinghouse Electric Corporation (retired); James Crook, Independent Consultant; Rodney Ewing, University of Michigan; Kathryn Higley, Oregon State University; Frank Parker, Vanderbilt University; and Alvin Trivelpiece, Oak Ridge National Laboratory (retired).

Although these reviewers have provided constructive comments and suggestions, they were not asked to endorse the content of the individual papers. Responsibility for the final content of the papers rests with the individual authors.

Special thanks are extended to Kelly Robbins for her translation of the Russian language papers into English, and to Amy Moore for her work in editing the papers.

Milton Levenson, *Chair*, National Research Council Committee on the
Scientific Aspects of an International Spent Nuclear Fuel
Storage Facility in Russia

Glenn E. Schweitzer, *Director*, Office for Central Europe and Eurasia

Contents

HANDLING SPENT NUCLEAR FUEL—INTERNATIONAL EXPERIENCE

SITE SELECTION FOR SPENT FUEL STORAGE AND DISPOSAL OF HIGH-LEVEL WASTE

PROBLEMS IN ESTABLISHING AN INTERNATIONAL STORAGE FACILITY FOR SPENT NUCLEAR FUEL IN RUSSIA

UTILIZATION OF HIGH-LEVEL WASTE

APPENDIXES

Opening Remarks

Milton Levenson
Bechtel International (retired)

I welcome all of you to this workshop. The topic we are to address in the next two days is extremely important and has significant long-range implications. At a time in history when war and terrorism fill our television programs and the headlines of our newspapers, it is not easy to focus on what are somewhat less urgent but perhaps in the long run equally important issues.

To help set the tone for our workshop I will first define the overall problem and then identify what role our workshop might play in developing a solution to the problem. The overall problem has two parts. The first part is to ensure that toxic materials do not enter the biosphere or environment in dangerous amounts. The second part is to ensure that fissionable materials are either controlled or destroyed so that irresponsible groups, including terrorists, cannot obtain critical quantities of weapons-usable material.

While science and technology may provide us with multiple options to address both parts of the problem, it is premature to select a single option for worldwide use. The development of technical options and the debate on risks versus benefits continues. The ultimate solution has not been selected. The first step, but a very important step, is to collect both fissionable material and other toxic materials and to provide secure storage until definitive solutions are decided upon. We all know that radioactive materials are not the only toxic materials; they are not even the most toxic materials, but our workshop will limit its scope to radioactive materials. Whether long-term storage becomes the method of disposal in the future, whether fissionable material is utilized or destroyed in the future, whether fission products and other radioactive materials are disposed of in geological repositories or transmuted into less dangerous materials in the future, these are all topics for future decisions. The topic of this workshop is how to start the process by collecting these materials in centralized, secure locations so that the risk they might pose to society can be minimized while ultimate solutions are developed, debated, and then implemented.

Our challenge for the next two days is to discuss the issues that must be resolved before we can be assured that the nuclear material now in temporary storage in many locations throughout the world has been placed in safe and secure storage. This is especially important for the smaller temporary sites where the cost per fuel unit of waste to ensure safety is very high because of the relatively small amounts of material involved. A centralized storage facility is certainly an attractive option for such material and that really is the focus of our workshop. We have invited speakers to cover the technical, policy, and legal issues of such a centralized facility. This is to be a workshop to illuminate the issues. No attempt will be made to resolve the issues, nor will an attempt be made to obtain a consensus. Our objective is to define what needs to be done as the next step in moving toward a solution—and identifying what might be some of the options for doing so. We will be successful if in the end we have provided elements and options for a solution that can be used by the decision makers in resolving the problem.

HANDLING SPENT NUCLEAR FUEL—INTERNATIONAL EXPERIENCE

IAEA Activities in Nuclear Spent Fuel Management

Fyodor F. Sokolov, Aleksey E. Lebedev, and *Kosaku Fukuda**

The International Atomic Energy Agency (IAEA) has implemented a subprogram on spent fuel management to assist member states. This subprogram is intended to play a catalytic role in promoting cooperation among member states through, among other things, the collection, evaluation, and dissemination of information on the current status of their national program for spent fuel management, and to assist member states in policy making, strategy planning, and implementation. The subprogram contains two projects: one is technology, strategy, and information on spent fuel management and the other is guidance on best practices for long-term storage of spent fuel. The major activities involved in these two projects are categorized as follows:

- use of the Regular Advisory Group on Spent Fuel Management
- resolution of technical and institutional issues on spent fuel storage
- implementation of burn-up credit
- support of dry storage technology for spent fuel from the water-moderated water-cooled power reactors (VVER) and high-power channel reactors (RBMK)
- other activities, such as the development of technical cooperation projects, research in spent fuel treatment, and a symposium on storage of spent fuel received from power reactors

---

*Fyodor F. Sokolov and Kosaku Fukuda, International Atomic Energy Agency; Aleksey E. Lebedev, Technabexport.

REGULAR ADISORY GROUP ON SPENT FUEL MANAGEMENT

The IAEA activities on spent fuel management have been based on the work of the Advisory Group on Spent Fuel Management (RAGSFM) that was established in 1984, upon the recommendation of an Expert Group Meeting on International Spent Fuel Management in 1982. It met biennially. In 1990 the group was renamed the Regular Advisory Group to express its recurring nature. The main objective of the RAGSFM was to review the worldwide spent fuel management situation, define the most important directions of national efforts and international cooperation, exchange state-of-the art information on spent fuel management, and elaborate recommendations for future IAEA spent fuel management programs. In 2001, following the recommendation by the Standing Advisory Group on Nuclear Energy (SAGNE) to reduce the number of advisory groups, the RAGSFM and the Technical Working Group on Nuclear Fuel Cycle Options (TWG-NFCO) were merged and a new TWG-NFCO was established. The first meeting of the new TWG-NFCO was held in July 2002, on the topic of Impact on Extended Burn-up of Spent Fuel on Back-end Nuclear Fuel Cycle Management.

TECHNICAL AND INSTITUTIONAL ISSUES ON SPENT FUEL STORAGE

Long-Term Spent Fuel Storage Facilities

Over the past three decades approximately one-third of spent fuel has been reprocessed, and the remaining is currently being held in storage. As a consequence some spent fuel has been stored for quite a long time. In addition, the recent trend of higher fuel burn-up and the use of plutonium in mixed oxide (MOX) fuel led to spent fuel characteristics requiring long-term storage due to high levels of heat emission and radioactive cooling. In several member states storage facilities have been licensed to operate for periods up to 50 years. In many cases periods of 100 years or longer are now anticipated. The IAEA implemented the coordinated research project (CRP) on long-term storage of spent nuclear fuel from 1994 to 1997. After the CRP, two Technical Committee Meetings (TCMs) under a new work program were held to address the trends in extended duration of spent fuel storage and the related technological and regulatory impacts:

1. TCM on Good Practices on Long-Term Storage of Spent Fuel Including Advanced, High Burn-up, and MOX Fuel, November 1999
2. TCM on Requirements for Extremely Long-Term Storage Facilities, October 2000

The results of these TCMs are included in the TECDOC.[1] The TCMs formed small working groups to provide information within the context of four general subjects:

1. long-term behavior of spent fuel
2. long-term behavior of dry storage systems
3. wet spent fuel storage facilities
4. regulatory concerns related to long-term spent fuel storage

The experience in spent fuel storage accumulated over the past decades is largely based on safe and effective wet storage and the effect of time on structures and materials during this limited period of time. The new challenges are to extend the life of new and existing wet and dry storage facilities, and to guarantee their safe performance for much longer periods of time.

Selection Criteria for Away-from-Reactor Storage Facilities

The survey of Away-from-Reactor (AFR) technologies has been conducted by a group of international consultants, and was published in a TECDOC on the survey of wet and dry spent fuel storage[2] in 1999.

Many utilities around the world are currently facing spent fuel storage concerns. The total spent fuel storage capacity for all commercial reactor sites worldwide is estimated to be 185,000 tons (t) heavy metal (HM). Further increases in capacity will be based on the business decisions of utilities as well as fuel cycle companies offering either reprocessing or storage services. Shipments to reprocessing facilities are expected to decrease, as new commitments by commercial utilities for spent fuel reprocessing are currently rare.

At the current rate of discharges the total inventory at reactor sites worldwide could reach about 192,000 t HM by 2010. In order to accommodate an increasing volume of spent fuel, an AFR storage facility could be constructed. AFR storage can be considered in two categories. The first is an additional interim storage capacity constructed at the reactor site (RS) but largely and entirely independent of the reactor and its at-reactor (AR) pool. The AFR/RS storage could be wet in the form of secondary or additional pools, but most often it would be a dry storage facility, which would have a capability for offsite transport. It has been suggested that some of these AFR/RS may stay operational well beyond the life of the power plants (up to 50 years or longer).

The second category of AFR storage is off the reactor site (OS) at an independent location. A large proportion of this AFR/OS capacity is in the form of pools at reprocessing plants particularly in France, the United Kingdom, and the Russian Federation. AFR/OS interim storage can also be centrally located at a selected power plant complex and receive spent fuel from other power plants.

Approximately 92 percent of AFR storage capacity is wet and the remaining 8 percent is dry.

Both wet and dry storage technologies have to address the following six requirements:

1. Fuel cladding integrity should be maintained during handling and exposure to corrosion effects of the storage environment.
2. Fuel degradation during storage should be prevented by providing adequate cooling in order not to exceed fuel temperature limits.
3. Subcriticality of the spent fuel should be maintained under both normal and emergency conditions.
4. Radiological shielding of the spent fuel should protect plant operators, the public, and the environment from exposure of radiation doses in excess of regulatory limits.
5. Environmental protection should be assured by minimizing the release of radioisotopes.
6. Fuel retrievability should always be available.

IMPLEMENTATION OF BURN-UP CREDIT

In 1997 the IAEA started to monitor the implementation of burn-up credit in spent fuel management systems, to provide a forum to exchange information and to gather and disseminate information on the status of national practices of burn-up credit implementation in the member states. In October 1997 the IAEA organized an Advisory Group Meeting (AGM) to examine and report on the status of burn-up credit for storage, transport, reprocessing, and disposal of pressurized water reactor (PWR), boiling water reactor (BWR), VVER, and MOX spent fuel. Since the proceedings of the AGM were published in 1998,[3] significant developments have served to advance the use of burn-up credit throughout the world. The IAEA held several consultancies in July and December 1998, July 1999, and July 2001 to monitor the progress in burn-up credit implementation.

In Vienna in July 2000 a TCM evaluated the implementation of burn-up credit in spent fuel management systems to determine the progress of international activities related to the use of the credit. The proceedings of this TCM were published as a TECDOC.[4]

In October 2001 a training course on the implementation of burn-up credit in spent fuel management systems was held at the U.S. Argonne National Laboratory, with 25 participants from 12 countries.

A TCM on the requirements, practices, and developments in burn-up credit application was held in Madrid, Spain, in April 2002.[5] The TCM recommended continued acquisition of data to support burn-up credit. In particular, the need for additional chemical assay to benchmark calculation methods was identified. Further studies of axial effects and verification methods for fuel burn-up values were recommended. A new TECDOC on the Implementation of Burn-up Credit in Spent Fuel Management Systems, which is the result of the CRP on Spent Fuel Performance Assessment and Research (SPAR, 1997–2002), was published in 2001 as IAEA-TECDOC-1241 (see Note 5).

SUPPORT FOR DRY STORAGE TECHNOLOGY FOR SPENT FUEL FROM THE VVER AND RBMK REACTORS

The government of Japan provided extrabudgetary funds from 1995 to 2001 to increase the safety of spent fuel storage from VVER and RBMK reactors. In the framework of this project, seven TCMs and workshops were held from 1995 to 2002, and a dry storage test was conducted. Beginning in 1997 workshops were held as follows:

- Prague, Czech Republic, October 1997
- Ignalina, Lithuania, October 1998
- Balatonfüred, Hungary, October 1999
- Prague, Czech Republic, July 2001
- St. Petersburg, Russia, June 2002

The dry storage test was performed at the Research Institute for Atomic Reactors (RIAR) in Russia, using spent fuel rods irradiated to 46 MWd/kg U burn-up on average in VVER 440.[6] The fuel rod cladding material is a Zr-1 percent Nb alloy. The dry storage tests were performed at 350°C for two months and at 390°C for another two months. Prior to the tests and after the 350°C test and the 390°C test the spent nuclear fuel was inspected by nondestructive methods. The tested fuel rods did not show any detectable changes in optical appearance, eddy current testing, profilometry, length measurement, gamma scanning, or oxide thickness measurement. In parallel, three fuel rods taken from the neighborhood of the tested spent nuclear fuel with practically identical operation performance were inspected by destructive methods to obtain the fission gas content in the fuel, the void volume, and mechanical test data from fuel rods. Detailed statistical investigation of the fuel rod diameter measurements indicated that a diameter increase of 1 to 2 μm had occurred. This value corresponds to the theoretical forecast for the diameter increase during the test. No negative effects on the fuel rods in the test were observed in the course of the test program. An extrapolation of the test data for longer-term storage shows that even under constant storage temperature the total strain at 350°C after 50 years remains at 1 percent. Assuming a pattern of falling temperature over the course of five years, for precooled fuel in a dry storage cask the total hoop strain does not exceed approximately 0.02 percent.

RELATED ACTIVITIES

Technical Cooperation Project on Spent Fuel Management

Technical Cooperation (TC) projects aim to provide technical assistance and respond to national or regional needs. In the field of spent fuel management the recent TC projects involved the following countries:

- Romania on the back end of the nuclear fuel cycle at Cernavoda nuclear power plant, ROM/4/020 (1997–1998) and ROM/4/023 (1999–2000)
- Bulgaria on reracking the existing AFR spent fuel storage facility at Kozloduy nuclear power plant for increased storage capacity, BUL/4/011 (2001–2002)
- China on the study of burn-up credit for critical safety analysis in nuclear power plant spent fuel storage and transport, CPR/4/023 (2002–2003)

Spent Fuel Treatment

Under the heading of spent fuel management, reprocessing activities have been reviewed from time to time using the terminology *spent fuel treatment*. The activities on spent fuel treatment implemented after 1990 have been an AGM in 1992, a TCM in 1995, an AGM in 1998,[7] and an AGM in 2000. After the AGM in 2000, a TECDOC will be drafted covering three major topics: national report, review of current technologies, and a review of emerging technologies. The TECDOC will be completed in 2003.

International Conference

The IAEA has held an international symposium on the storage of spent fuel from power reactors once every four years since 1987. The objectives of the symposiums are to

- review recent advances in spent fuel technology
- exchange information on state-of-the art techniques and prospects of spent fuel storage
- review and discuss the worldwide situation and the major factors influencing the national policies in this field
- exchange operating experience on wet and dry storage facilities
- identify the most important directions that national efforts and international cooperation in this area should take

The last symposium, in November 1998, was attended by 130 experts from 32 countries and 3 international organizations, including the IAEA.[8] The dominant messages to come out of the symposium were that the primary spent fuel management solution for the next decades will be interim storage, the duration time of interim storage is becoming longer than earlier anticipated, and storage facilities will have to be designated also to receive spent fuel from advanced fuel cycle practices, that is, high burn-up and MOX spent fuel. The next international conference is planned for Vienna on June 2–6, 2003.

SUMMARY AND CONCLUSIONS

The major emphasis of spent fuel management in the near term program will be placed on

- continuous review of spent fuel incidents and storage capacity worldwide
- long-term issues of spent fuel storage covering optimization of cask and container loading and fuel failure classification
- burn-up credit, including a particular depletion code for burn-up credit using chemical assay
- extension of the operation of interim storage facilities

The IAEA continues various activities associated with spent fuel management by taking into account the General Conference Resolutions, requests from member states, recommendations of major meetings, and the IAEA Medium Term Strategy.

NOTES

1. IAEA-TECDOC-1293, Long term storage of spent nuclear fuel—Survey and recommendations, Final report of a coordinated research project, 1994–1997, (2002).
2. IAEA-TECDOC-1100, Survey of wet and dry spent fuel storage, (1999).
3. IAEA-TECDOC-1013, Implementation of burnup credit in spent fuel management systems, Proceedings of an Advisory Group meeting held in Vienna, October 20–24, 1997, (1998).
4. IAEA-TECDOC-1241, Implementation of burnup credit in spent fuel management systems, Proceedings of a Technical Committee Meeting held in Vienna, July 10–14, 2000, (2001).
5. H. P. Dyck, Overview on the burnup credit activities at the IAEA, Proceedings of the Technical Committee Meeting on the Requirements, Practices, and Developments in Burnup Credit Applications, Madrid, Spain, April 22–26, (2002).
6. H. P. Dyck (IAEA), V. P. Smirnov (RIAR), and S. Pavlov (RIAR), Results of VVER-440 FR examination after testing under simulated dry storage conditions, to be published as a TECDOC.
7. IAEA-TECDOC-1103, Status and trends in spent fuel reprocessing, Proceedings of an Advisory Group Meeting held in Vienna, September 7–10, 1998, (1999).
8. IAEA-TECDOC-1089, Storage of spent fuel from power reactors, Proceedings of a symposium held in Vienna, November 9–13, 1998, (1999).

Analysis of U.S. Experience with Spent Fuel

John F. Ahearne
Sigma Xi: The Scientific Research Society

The United States has 103 reactors at 65 sites, representing a generation capacity of about 88 GWe. As of December 2001 the United States was storing approximately 45,000 metric tons of heavy metal of spent fuel from civilian nuclear power plants. As of December 31, 2001, 42,000 metric tons of heavy metal was stored in pools, with only 3000 in dry casks. Use of dry casks is growing, however, as utility pools fill up. A repository or offsite storage facility is not available.

Table 1 summarizes the U.S. fuel, along with that in Russia. A smaller amount of spent fuel from the U.S. weapons program is also being stored for eventual disposal. Most of this fuel has been reprocessed to extract the plutonium or highly enriched uranium.

There are many nuclear power plant designs used in the United States, but all except three are and have been light water reactors, either pressurized water reactors or boiling water reactors. The fuel elements are zirconium alloy tubes, containing ceramic UO_2 pellets of 3–5 percent enrichment. Current burn-up goes to at least 45,000 MWD per metric ton of heavy metal. The three nonlight water reactors were

- Fermi 1, a 60 MWe breeder, closed in 1972
- Peach Bottom 1, a 40 MWe high-temperature gas-cooled reactor, closed in 1974
- Fort St. Vrain, a 330 MWe high-temperature gas-cooled reactor, closed in 1989

TABLE 1 Amounts of Spent Nuclear Fuel in Storage and Rate at Which the Amount is Increasing

Type of Spent Nuclear Fuel	Russian Federation Spent Nuclear Fuel (metric tons of heavy metal)	United States Spent Nuclear Fuel (metric tons of heavy metal)
Power Reactor	14,000 + 850 per year	45,000 + 2000 per year
Naval	70 + fuel from 15–18 nuclear power stations per year	19.5 + 45.5 over 33 years[a]
Production Reactor	Not available[b]	2100 + 0 per year
Research Reactor	28,500 assemblies	23 + 0.07 per year

[a] Citing an annual rate for discharges from naval reactors may not be accurate so the expected total for a known period is given.

[b] Approximately 1.5 metric tons of separated plutonium are produced each year by the three dual purpose reactors. The spent nuclear fuel from these reactors is stored only briefly before going through chemical separations.

SOURCE: National Research Council. End Points for Spent Nuclear Fuel and High-Level Radioactive Waste in Russia and the United States. Washington, D.C.: The National Academies Press, 2003, Table 1.1.

GOVERNMENT-MANAGED SPENT FUEL

The U.S. Department of Energy (DOE) currently manages about 2500 metric tons of heavy metal of spent fuel, of about 250 different types. This includes fuel from plutonium production reactors, naval reactors, and research and demonstration reactors.

The United States halted reprocessing to obtain weapons plutonium in 1988, and the Hanford reprocessing canyons shut down in 1989. One Savannah River canyon continues to operate for processing unstable fuel. The Idaho Chemical Processing Plant, which had been used for naval fuel, shut down in 1992.

Table 2 lists quantities of U.S. government fuel and current disposition plans.

NAVAL REACTORS

The United States produced 191 submarines each with 1 reactor, 8 aircraft carriers with 2 reactors each and 1 carrier with 8 reactors, 9 cruisers with 2 reactors each, a deep submergence research vessel with 1 reactor, and 1 civilian cargo ship with 1 reactor. The only nuclear ships still in service are the carriers and 72 submarines.[1]

All navy spent fuel is shipped to Idaho National Laboratory for storage. After decommissioning the ship and removal of the fuel, the reactor compartment is removed and shipped to the Hanford site for storage.

TABLE 2 Quantities of U.S. Government Spent Nuclear Fuel and Unirradiated Nuclear Fuel Grouped According to Near-Term Management[a]

Near-Term Management	Quantity (metric tons of heavy metal)	Examples
Processed to HLW at ANL-W	61.3	Sodium bonded EBR-II and FFTF fuel
In foreign research reactors	14.3	HEU in Al plates in France, Pakistan, and four other nations
Storage until repository disposal (no further processing)	2465	N-reactor fuel, fuel from isotope production reactors, ANP fuel
Special treatment	0.041	Cutting fines from SNF assay, MSRE fuel
Processed to HLW at SRS	23.9	Declad EBR-II uranium metal fuel, declad uranium/thorium fuel
Treatment at ORNL Y-12	0.27	Failed fuel from Rover[b]
Unknown	996	Unirradiated fuel for the N-reactor, FFTF, EBR-II
Unknown	25.2	Various fuel forms (unclad natural uranium, polyethylene matrixes, aluminum) from test piles and research reactors, also unirradiated but damaged fuel (managed as spent fuel)

[a] All wastes are planned ultimately to be disposed of in a repository.
[b] Rover was a nuclear rocket prototype reactor with niobium-based fuel.

NOTE: **ANL-W:** Argone National Laboratory West; **ANP:** Aircraft Nuclear Propulsion; **EBR-II:** Experimental Breeder Reactor-II at Argonne National Laboratory West; **FFTF:** Fast Flux Test Facility at Hanford; **HEU:** Highly Enriched Uranium; **HLW:** High-Level Waste; **MSRE:** Molten Salt Reactor Experiment; **ORNL:** Oak Ridge National Laboratory; **SNF:** Spent Nuclear Fuel; **SRS:** Savannah River Site.

SOURCE: U.S. Department of Energy, Office of Spent Fuel Management. Spent Fuel Database (SFD), Version 4.2.0. March 25, 2002; National Research Council. End Points for Spent Nuclear Fuel and High-Level Radioactive Waste in Russia and the United States. Washington, D.C.: The National Academies Press, 2003, Table 2-4.

RESEARCH REACTORS

In the 1970s the United States had 70 research reactors at universities and several dozen research and test reactors at government and industrial facilities. Today 36 are operating, and 19 are being closed down. Most are small, with the largest being 20 MWth. The United States has provided fuel to 110 research reactors in other countries, and there is a program for the highly enriched uranium fuel to be shipped back to the United States. When the fuel is returned, it is sent to Savannah River or Idaho for storage. Approximately 2.7 metric tons of heavy metal of highly enriched uranium fuel are still in other countries.

DISPOSITION OF 34 METRIC TONS OF WEAPONS PLUTONIUM

The United States began to consider various approaches for disposition of weapons plutonium in the early 1990s and asked the National Academies to study the possible approaches. The study examined vitrification and various reactor options. Following this study and development of technical information by the national laboratories, in 1997 DOE announced a dual-track strategy:

1. fabrication of clean plutonium into MOX (mixed oxide) fuel and use in civilian reactors (26 metric tons)
2. vitrification of the plutonium thought to be unsuitable for MOX fuel (8 metric tons)

In 2001 DOE concluded that this plan would be too expensive and would take too long, and decided to proceed only with MOX. Of the 8 metric tons of unsuitable plutonium, 6.2 metric tons will be processed and then made into MOX fuel. Other plutonium will be used to make up the required total of 34 metric tons; the 1.8 metric tons of impure plutonium do not yet have a disposal path.

The U.S. program to build a MOX facility has not developed as rapidly as hoped. The plant design is currently in the licensing process.

LIQUID HIGH-LEVEL WASTE

Production of nuclear weapons has also produced large volumes of liquid high-level waste. Table 3 shows the quantities of liquid high-level waste stored at sites in the United States.

YUCCA MOUNTAIN, HIGH-LEVEL WASTE, AND THE NATIONAL ACADEMIES

For over 80 years the U.S. government has asked the National Research Council, the study arm of the National Academies, to provide advice on issues of importance to the government. In the area of radioactive material the Board on Radioactive Waste Management (BRWM) has produced over 100 reports, primarily on high-level waste and the environmental management issues relating to legacy wastes, those wastes from the nuclear weapons program. I am glad to note that Academician Laverov is a member of BRWM.

I will now address the end point for spent nuclear fuel as seen in the United States. In 1955 the National Research Council recommended isolation in stable

TABLE 3 Quantities of High-Level Waste (HLW) Stored at Sites in the United States

Site	HLW in Tanks (cubic meters)	Vitrified HLW (canisters)	Total Radioactivity ($\times 10^8$ Ci)	Percent of Total Volume	Percent of Total Radioactivity
Hanford Site	200,000	0	384	58.9	15.8
Savannah River Site	130,000	719[a]	1730	38.3	71
Idaho National Engineering and Environmental Laboratory	9360	0	300	2.8	12.3
West Valley Demonstration Project	109[b]	241[b]	23.3	<0.1	1
Total	339,469	960	2437.3	100	100.1

[a] 1337 as of October 2002.

[b] HLW from the tanks at West Valley has been vitrified in 275 canisters. Residual HLW encrusted on the tanks is being characterized and sluiced.

SOURCE: U.S. Department of Energy, Office of Environmental Management. Summary Data on the Radioactive Waste, Spent Nuclear Fuel, and Contaminated Media Managed by the U.S. Department of Energy. April 2001. National Research Council. End Points for Spent Nuclear Fuel and High-Level Radioactive Waste in Russia and the United States. Washington, D.C.: The National Academies Press, 2003, Table 3.3.

geologic formations as the approach for handling high-level waste. In 1982 the U.S. Waste Policy Act directed the DOE to develop a deep geologic repository. In 1987, after several years of screening for potential sites and then narrowing to four, the U.S. Congress selected the Yucca Mountain Site in Nevada, adjacent to the Nuclear Test Site, where the United States has conducted many above- and below-ground nuclear weapons tests.

In 1990 BRWM produced a 40-page report, *Rethinking High-Level Radioactive Waste Disposal*,[2] which had a major impact on the DOE program. The report begins as follows:

> There is a worldwide scientific consensus that deep geological disposal, the approach being followed in the United States, is the best option for disposing of high-level radioactive waste (HLW). There is no scientific or technical reason to think that a satisfactory geological repository cannot be built.

The report further stated that the U.S. program as conceived and implemented in the past is unlikely to succeed. Note that this report, as do many, seems to address only science and technology; the political and other social science aspects were not highlighted.

Since the report was written, the DOE program did change, but it still has had problems, both technical and political. The United States appears to be the only country to have taken the approach of writing detailed regulations specifying what must be shown in the license application before the requirements for the general site data are determined. "As a result, the U.S. program is bound by requirements that may be impossible to meet."[3]

At the request of Congress the National Research Council conducted a study on what should be the technical bases for Yucca Mountain requirements and produced *Technical Bases for Yucca Mountain Standards*, known as the TYMS report,[4] in 1995. In addition to recommending a risk-based approach rather than a dose-based approach, the study advised on how to treat intrusion (specifically, do not make it a licensing requirement) and integrated population dose (specifically, do not count carbon 14 in all of the world's population). Although the U.S. Environmental Protection Agency (EPA) eventually disregarded the report, the report did introduce the issues to a wide audience, including many in Congress who had been unaware of the issues. It may eventually be used in court.

In 1996 the National Research Council published a large report, *Nuclear Wastes: Technologies for Separations and Transmutations* (the STATS report),[5] which addressed whether science and technology could avert the need for a long-term geologic repository. The report recommended that DOE continue to develop a repository for spent nuclear fuel, since no science and technology concept would eliminate that need; that retrievability in the repository should be on the order of 100 years; and that research and development should continue on science and technology concepts.

The next major study was published in 2001, *Disposition of High-Level Waste and Spent Nuclear Fuel: The Continuing Societal and Technical Challenges*.[6] This was an international study and included members from many countries (Academician Laverov was a member). This study reviewed the arguments for and against geologic repositories, reviewed the status of national programs, and recommended processes for governments to follow and to develop geologic repositories. These recommendations included technical topics and also political and social science-based topics. In this second report the committee recognized that involving the local public is essential if there is to be successful site selection and development.

THE U.S. PROCESS

In 2002 Secretary of Energy Spencer Abraham recommended to President Bush that DOE should go forward with developing an application to construct the Yucca Mountain repository. The president accepted that recommendation and forwarded it to Congress. As the Waste Policy Act provides, the host state, Nevada, could refuse to accept the repository, a veto that could be overridden by the Congress. Nevada did veto the recommendations, and finally the Congress did override it. The issues, often couched in technical language, were in my opinion primarily political. (The congressional and DOE approaches are case studies of how not to work with the public. These are classic examples of the model characterized as "decide, announce, and defend" in which the interested and affected public is not asked to contribute before the decision is made—only after.)

DOE now must submit a license application to the Nuclear Regulatory Commission, which by law has three to four years to review it. A legal hearing, in our terms an adjudicatory hearing, will be held to decide whether the DOE application meets the licensing requirements. This is the first such license application, and with intense public interest, the commission will move carefully.

If the application is successful and a license to construct is issued, it will take several more years to construct the above-ground facilities and extend the existing 11 km of tunnels. DOE must then apply for a license to emplace waste.

The current schedule—which DOE has had for several years—is for first fuel going underground in 2010. A recent National Research Council report noted, "Most external commenters believe this ambitious schedule is unrealistic based on the time needed for each step. In addition, several lawsuits that attempt to block the various steps in the process have been filed."

Other issues regarding Yucca Mountain include working on the transportation of spent nuclear fuel to Yucca Mountain and the security of Yucca Mountain and related facilities against terrorist attacks.

The latest Academy study was released earlier this year, *One Step at a Time: The Staged Development of Geologic Repositories for High-Level Radioactive Waste*.[7] While addressing the general case of developing a repository, the study has direct applicability to Yucca Mountain. The study recommends an approach called adaptive staging. There is no simple definition of this concept. It provides flexibility in responding to new information, allows examination of performance and consideration of new knowledge before moving to the next step, and provides more access by the public to the repository program. To an engineer this looks like a feedback loop.

OTHER STUDIES

The National Research Council has also studied both narrower and broader issues. Examples of narrower studies are

M. Levenson and K. D. Crowley. *Research Reactor Aluminum Spent Fuel: Treatment Options for Disposal*. Washington, D.C.: National Academy Press, 1998.

National Research Council. *Research Needs in Subsurface Science.* Washington, D.C.: National Academy Press, 2000.

National Research Council. *Characterization of Remote-Handled Waste for the Waste Isolation Pilot Plant (WIPP).* Washington, D.C.: National Academy Press, 2002.

Examples of broader studies are

National Research Council. *A Strategic Vision for Department of Energy Environmental Quality Research and Development.* Washington, D.C.: National Academy Press, 2001. This report addressed all the programs that relate to the environmental quality research and development portfolio across many DOE divisions.

National Research Council. *Making the Nation Safer: The Role of Science and Technology in Countering Terrorism.* Washington, D.C.: The National Academies Press, 2002. This report recommends how science and technology could contribute to many areas and addresses the vulnerabilities to terrorist threats; one section addresses radiological threats.

National Research Council. *End Points for Spent Nuclear Fuel and High-Level Radioactive Waste in Russia and the United States.* Washington, D.C.: The National Academies Press, 2003. This study was cochaired by Academician Laverov; the Russian members of the committee included Academician Melnikov, Academician Myasoedov, and Dr. Pek. The study provides a high-level view of the entire fuel cycle in both countries and recommends near-term and long-term actions.

NOTES

1. National Research Council. End Points for Spent Nuclear Fuel and High-Level Radioactive Waste in Russia and the United States. Washington, D.C.: The National Academies Press, 2003.
2. National Research Council. Rethinking High-Level Radioactive Waste Disposal: A Position Statement of the Board on Radioactive Waste Management. Washington, D.C.: National Academy Press, 1990.
3. National Research Council. Rethinking High-Level Radioactive Waste Disposal: A Position Statement of the Board on Radioactive Waste Management. Washington, D.C.: National Academy Press, 1990, p. vii.
4. National Research Council. Technical Bases for Yucca Mountain Standards. Washington, D.C.: National Academy Press, 1995.
5. National Research Council. Nuclear Wastes: Technologies for Separations and Transmutations. Washington, D.C.: National Academy Press, 1996.
6. National Research Council. Disposition of High-Level Waste and Spent Nuclear Fuel: The Continuing Societal and Technical Challenges. Washington, D.C.: National Academy Press, 2001.
7. National Research Council. One Step at a Time: The Staged Development of Geologic Repositories for High-Level Radioactive Waste. Washington, D.C.: The National Academies Press, 2003.

Problems of Spent Nuclear Fuel Management and Storage Site Selection*

M. I. Solonin
Russian Ministry of Atomic Energy

In this report I will cover the complex fate of the products of existing nuclear technologies, including spent nuclear fuel and radioactive wastes, as well as the degree to which they affect prospects for the development of the international atomic energy sector. I will also discuss the Russian experience and our long-range plans in this area.

As you know, there are currently a number of different assessments regarding the rates at which the world economy will develop in the twenty-first century. All these projections conclude that overall demand for energy will increase. These forecasts also promise growth in the absolute amounts of electricity to be produced at nuclear power plants. It is interesting to consider the objective capacities for increasing the nuclear production of electricity around the world.

Let us recall that the share of electricity produced by the nuclear power sector now totals 16 percent, and the majority of representatives of this industry express optimistic views regarding the long-term prospects for its development. It is noted that no substantial growth should be anticipated in the next 10–20 years. The most typical view is as follows: Hydrocarbon fuel will serve as the main source of energy in the coming decades because the entire current world infrastructure for energy production and consumption is based on this type of fuel. In addition, currently exploited deposits of hydrocarbon fuel are being exhausted, while bringing new resource deposits into exploitation requires ever increasing volumes of investments. As a result, it will become unavoidable to change the supply structure in the energy market, which in conjunction with

---

*Translated from the Russian by Kelly Robbins.

severe environmental restrictions will lead in turn to sharp price competition and corresponding instability. It is nuclear power, with its greater fuel resources using existing technologies, that may be able to stabilize the energy sector in the twenty-first century on an environmentally safe basis.

The competitive advantages of this type of energy production are commonly known. Here I would like to focus on history, however: specifically those very joyous predictions that were made with regard to the rates of development of the nuclear energy sector in the 1960s. It should be acknowledged that the large-scale plans for building nuclear reactors that were made in the majority of the leading countries never materialized, and the hopes associated with these plans were not justified either. The basic reasons for this are well known. In the initial stage of development of the nuclear power industry, insufficient attention was paid to matters of comprehensive safety.

The accidents at the Three Mile Island and Chernobyl power plants had a severely negative impact on the views of the general population regarding the safety of nuclear technologies. The fact that in the mind of the average observer, the effects of radiation are intangible and therefore uncontrollable, led to widespread radiophobia among the masses. Here, naturally, there appeared certain individuals and even groups that for various reasons took an interest in ensuring that this fear of radiation became constant among people. We encountered this in Russia in this previous stage in the industry's development partly because our nuclear power industry had to operate in a closed manner because of its close ties with military programs. Now, however, we are changing: We are building information centers, conducting scientific debates, and publicizing our activities in the media, and radiophobia is lessening among the population. Unprecedented measures have been taken regarding the safety of existing nuclear reactors as well as those under construction. Their high reliability has been proven by many years of operating experience, and the public recognizes this. What then is hindering the development of the nuclear power industry, its full renaissance? I will try to provide one answer to this question later in this paper.

Russia has created its Strategy for the Development of Nuclear Power in the First Half of the Twenty-First Century, the basic elements of which include the doubling of energy output and the formation of a closed nuclear fuel cycle. As a result, we must ensure fuller utilization of natural fissionable materials as well as those created during reactor operations, minimize the volume of radioactive wastes, and promote nuclear and radiation nonproliferation. This approach was presented in the initiative of the President of the Russian Federation at the United Nations Millennium Summit in September 2000. This initiative called for broad international cooperation on nuclear energy to ensure stable development, nonproliferation of nuclear materials, and environmental, nuclear, and radiation safety. As you know, in the United States, efforts to improve reactors are being carried out under the Nuclear Energy 2010 Program, while work on innovative nuclear technologies is coordinated under the international program Generation

IV. In support of the initiative of President Vladimir Putin, work on innovative projects has recently been included in the International Project on Innovative Nuclear Reactors and Fuel Cycles (INPRO) project of the International Atomic Energy Agency (IAEA). International cooperation in this area is developing actively, for which we can only be glad. In working on current types of reactors we are eliminating old mistakes, but we must not make new ones when questions of the end stage of the nuclear fuel cycle are pushed into the background.

Here is my answer to the question raised a bit earlier: It seems to me that the development of the nuclear energy sector is held back mainly by the lack of clear and generally understandable solutions regarding the safe management of spent nuclear fuel and radioactive wastes.

What is spent nuclear fuel? It is well known that on the one hand, more than 90 percent of it is made up of materials suitable for further industrial use, and therefore it represents a valuable raw material source for obtaining recycled nuclear fuel components and important isotopes. On the other hand, it contains, albeit in small quantities, potentially dangerous radioactive substances having no suitable application, given the existing level of technology, that is, substances that are radioactive wastes.

It is because of this product duality and the corresponding contradictory nature of approaches to it (raw material or waste) that there is no end to stormy debates among nuclear, environmental, and economics specialists on the correctness of the choice of this or that means of managing spent nuclear fuel.

It should be emphasized that the problem of reprocessing spent nuclear fuel is one faced by all countries that operate nuclear power facilities. Spent nuclear fuel is accumulating in many geographic regions, in decentralized fashion and according to varying standards, a fact that represents a potential threat to global security and hardly coincides with the objectives of nuclear nonproliferation. The fact is that plutonium suitable for the creation of a nuclear explosive device can be separated from spent nuclear fuel. The spent fuel itself can be used to create a radiological bomb—or what IAEA Director General Mohamed ElBaradei calls a "dirty" bomb—which is formed by combining conventional explosives with a radiation source. This problem gives rise to a political aspect as well.

As you know, there are three concepts in the world regarding how to manage spent fuel: (1) reprocessing of the material at radiochemical plants in order to reuse the uranium and plutonium in the fuel cycle; (2) direct burial of it; and (3) long-term storage with the decision on final disposition left for a later date.

The various concepts involved in the management of spent nuclear fuel depend on the strategies adopted by each nation regarding development of the nuclear power industry, the role of this industry in the country's future energy supply, and the capacities of its natural resource base.

Russia signed the Joint Convention on the Safety of Spent Fuel Management and on the Safety of Radioactive Waste Management in 1997, but the final formal procedures for its ratification have not yet been completed. I will review the basic approaches to the management of spent fuel and radioactive wastes included in the convention.

As I see it, the joint convention signified progress in international cooperation regarding the management of spent fuel and radioactive wastes. One of the most important disputed questions was the inclusion in the convention of the very topic of spent nuclear fuel, inasmuch as the approach to assessing the essence of spent fuel (raw material or waste) fundamentally differs in countries oriented toward reprocessing the material and countries that plan for its direct isolation. As a result, only spent fuel not subject to reprocessing was included within the scope of the convention.

As for the position of each country on the matter of spent fuel and radioactive waste management, the parties to the convention recognize, first, that setting policy regarding the nuclear fuel cycle remains the prerogative of each individual state and, second, that some states consider spent fuel a valuable resource that can be reprocessed, while others prefer to bury it.

The convention also notes that radioactive wastes must be buried in the country in which they were produced, recognizing that in certain circumstances the safety of spent fuel and radioactive waste management could be enhanced by an agreement among the parties regarding the use of the facilities of one country in the interests of other countries. It is acknowledged that any state has the right to ban the importation of foreign spent fuel and radioactive wastes into its territory.

The convention creates good opportunities for international cooperation, and nothing in the convention infringes upon or affects the rights of the signatories, for instance,

- the rights of a signatory to export spent fuel for reprocessing
- the rights of a signatory receiving imported spent fuel for reprocessing to return (or ensure the return) to the producing state of the radioactive wastes and other products created during reprocessing operations

In analyzing international law on this topic, one may conclude that there is an internationally recognized right of each state to set its own policy on managing spent nuclear fuel and radioactive wastes and, while observing international norms for ensuring security, to create its own legal and regulatory base. The question of legislative regulation of the conditions for managing spent fuel and radioactive wastes has special characteristics in each country depending on the concept selected there for spent fuel management. For example, when direct burial of spent fuel is selected, spent fuel is deemed by the legislation of a number of countries to fall into the category of high-level radioactive waste. The dynamic nature of the legal base in foreign countries bears particular notice. Effective and timely changes in laws and regulatory documents, taking into account changes in the political, economic, and social spheres, are necessary in order to ensure that the laws correspond with current realities. Russia is not standing idle in this regard.

As you may already know, we in Russia have passed new legislation that expands our capabilities with regard to international cooperation on spent fuel management and makes Russia's position on this question more flexible. The legislation includes provisions that

- facilitate relevant shipments involved in the nuclear fuel cycle (fresh and spent nuclear fuel)
- enhance the regime for nonproliferation of nuclear weapons
- strengthen the future raw material base for the power industry through the use of regenerated uranium and plutonium
- help to attract financial resources for the resolution of Russian environmental problems that built up during the creation of nuclear weapons in the country
- facilitate the development of the national infrastructure for spent fuel management in accordance with current international requirements
- encourage the development of international cooperation on spent fuel management in the scientific-technical and industrial spheres

Now is an appropriate time to discuss our practical experience in the management of spent nuclear fuel. In Russia at present spent nuclear fuel from water-moderated water-cooled power reactors and fast neutron reactors (VVER-440, BN-600, and BN-350), the majority of research reactors, and nuclear reactors from atomic-powered ships and submarines is reprocessed. This reprocessing is carried out at the RT-1 plant at the Mayak Production Association. The plant has been operating since 1976 as an experimental production facility. The spent fuel from VVER-1000 reactors is currently shipped to a centralized wet repository at the Mining-Chemical Complex in the city of Krasnoyarsk, and the spent fuel from high-power channel reactors (RBMK) is stored in facilities at each relevant power station.

Adherence to the principle of demand for the output from the regeneration process means that a substantial increase in spent fuel reprocessing volumes makes sense only after the construction of a new generation of fast reactors begins on a full and regular basis. Therefore, it is necessary to store the spent fuel from thermal reactors for a long period. This material must be stored in dry conditions, which ensures that the process will be reliable and the spent fuel will be kept hermetically sealed, and it also reduces the costs of construction and operations. At the same time, dry storage also makes reprocessing easier, as during the period of storage the radioactivity level of the fuel is substantially reduced. Thus, storage becomes a process of technological restraint.

Plans for a 33,000 metric-ton dry container-type repository at the Mining-

TABLE 1 Management of Spent Nuclear Fuel at Russian Nuclear Power Plants

Type of Reactor	Current System	Planned System
VVER-440	Temporary storage of spent fuel in holding pools (3–5 years) with subsequent shipment for reprocessing	Temporary storage of spent fuel in holding pools (3–5 years) with subsequent shipment for reprocessing
VVER-1000	Temporary storage of spent fuel in holding pools (3–5 years) with subsequent shipment to centralized repository	Temporary storage of spent fuel in holding pools (3–5 years) with subsequent shipment to centralized repository or for reprocessing
RBMK-1000	Permanent storage in holding pools; filling of pools to capacity in 2005–2007	Containerized storage (3600 spent fuel rods per year at each power plant) with subsequent shipment to centralized repository
BN-600	Temporary storage of spent fuel in holding pools (3–5 years) with subsequent shipment for reprocessing	Temporary storage of spent fuel in holding pools (3–5 years) with subsequent shipment for reprocessing
EGP-6 and AMB	Permanent storage in holding pools	Containerized storage (Bilibino Atomic Power Plant: 8200 spent fuel rods; Beloyarsk Atomic Power Plant: 7200 spent fuel rods)

Chemical Complex for spent fuel from VVER-1000 and RBMK reactors are now being developed and subjected to expert review. Tables 1 and 2 present information on the current capacities of existing industrial facilities to manage spent nuclear fuel, as well as plans for their modernization. The tasks are clear and precisely formulated, so I do not believe that any detailed commentary is required.

Considering the volume of spent nuclear fuel that has accumulated in the world, it is my opinion that the situation demands the creation of international spent fuel storage facilities. The creation of such large-capacity facilities, especially in nuclear countries that have no need for weapons-grade plutonium but on the contrary are involved in reprocessing it, in my opinion increases the safety of spent fuel management and reduces the possibility of theft and unauthorized proliferation of nuclear materials.

I will now say a few words about the practices and plans in the area of radioactive waste management. Russia's Ministry of Atomic Energy (Minatom) has adopted the Convention on the Management of Radioactive Wastes (ratified by the Decree on Minatom of 2000). Also in 2000 a resolution by the government of the Russian Federation launched the Federal Targeted Program on the Nuclear and Radiation Security of Russia. One of the top-priority objectives of

TABLE 2 Management of Spent Nuclear Fuel at the Mayak Production Association, City of Ozersk, Chelyabinsk Oblast

Type of Reactor	Current System	Planned System
VVER-440	Reprocessing at current RT-1 plant	Reprocessing at modernized RT-1 plant
VVER-1000	Reprocessing has not yet begun	After modernization of RT-1 plant, storage in centralized 1700 metric-ton repository and reprocessing
BN-600	Reprocessing at current RT-1 plant	Reprocessing at modernized RT-1 plant
Research reactors and those powering naval vessels	Reprocessing at current RT-1 plant	Reprocessing at modernized RT-1 plant Construction of facilities for underground isolation of wastes from spent fuel reprocessing
VVER-1000	Storage in centralized wet repository with capacity of 6000 metric tons	Increase in capacity of wet repository to 9000 metric tons, storage in centralized "dry" facility with capacity of 9000 metric tons, and reprocessing at plant
RBMK-1000	Facilities not yet ready	Storage in centralized dry facility with capacity of 24,000 metric tons Construction of facilities for underground isolation of wastes from spent fuel reprocessing

this program is the comprehensive resolution of problems regarding the management of radioactive wastes.

Subprograms involved in this targeted effort include the management, recycling, and burial of radioactive wastes and the organization of a system for state accounting and control of these wastes. Anticipated results of the targeted program include the development and use of modern technologies for ensuring safe operations with radioactive wastes, the recycling and reliable isolation of wastes, and the creation of radioactive waste burial sites and repositories.

Planned activities include the following:

- improvement of technologies for managing radioactive wastes at enterprises involved in the nuclear fuel cycle
- improvement of technologies for managing radioactive wastes at nuclear power plants
- improvement of technologies for managing radioactive wastes accumulated during the production of nuclear materials for weapons purposes
- management of radioactive wastes in scientific organizations

- management of radioactive wastes at specialized radon complexes
- creation of burial sites and repositories for radioactive wastes
- development of a methodology for selecting appropriate geological environments for waste repositories
- development of principles for selecting and creating new types of protective barriers to ensure the security of radioactive waste storage facilities

Russia has accumulated radioactive wastes with a total activity of about 2000 MCi, with more than 95 percent of them being concentrated at Minatom organizations and enterprises.

The main problems involved in radioactive waste operations at enterprises involved in the initial stages of the nuclear fuel cycle are as follows:

- removal from service and mothballing of quarries, mines, and mine-tailing storage pits
- reprocessing of wastes from the production of enriched uranium
- reprocessing of solid radioactive wastes at enterprises manufacturing nuclear fuel

The main problems involved in radioactive waste operations at nuclear power plants are as follows:

- creation of installations for the concentration and conditioning of various types of radioactive wastes
- creation of liquid and solid waste storage facilities at the plants
- transportation of radioactive wastes to a centralized repository

Separate mention should be made of the problems of two previously discussed enterprises engaged in spent fuel management: the Mayak Production Association and the Mining-Chemical Complex. With these enterprises, the problems of managing radioactive wastes accumulated during the production of weapons-grade nuclear materials have come to the forefront, including

- mothballing and elimination of pulp storage tanks, specialized reservoirs, and test sites for the underground storage of liquid radioactive wastes
- cleanup of radiation-contaminated land sites and bodies of water
- decontamination of structures, buildings, and equipment

The saddest thing here is that these problems, which arose from the military sphere, are viewed by the public as being purely problems of the nuclear power industry and not as a national inheritance created by the well-known conflict that occurred in an ideologically bipolar world. As a result, Minatom truly has already become an environmental agency with its own environmental doctrine.

The arms race, nuclear weapons testing, major radiation accidents, and the wastes and emissions of enterprises during the early years of the nuclear industry led to the appearance in the biosphere of engineered radionuclides, the radioactive contamination of certain areas, and the presence to this day of a large number of facilities, mainly from the defense sector, that are hazardous from a nuclear or radiation standpoint and must be decommissioned or returned to environmentally safe status. Overcoming the negative consequences of past activities is one of the most important tasks assigned to and being addressed by Minatom.

A Minatom decree in 2003 set forth the Foundations for the Environmental Policy of the Russian Ministry of Atomic Energy. The main goal of Minatom's environmental policy is the creation of conditions in which nuclear industry enterprises can operate most effectively to achieve the strategic goal of the environmental policy of the Russian Federation; this policy seeks the preservation of natural systems and maintenance of their integrity and life-sustaining functions to ensure the stable development of society; improve the quality of life, public health, and the demographic situation; and promote the environmental security of the country. These conditions must promote

- the environmental security of existing production facilities as well as those under construction or being planned and those removed from operation
- the resolution of previously accumulated environmental problems
- the development and implementation of new economically efficient and environmentally safe technologies in the nuclear power industry and other sectors where nuclear energy is used

To provide an example of how the previously listed conditions are being put into practice, I will discuss the existing and potential future technologies for spent fuel reprocessing at Mayak's RT-1 plant. Besides transforming high-level wastes into aluminum-phosphate glass, the plant also carries out the fractionation of nuclides (cesium, strontium, transplutonium elements, and rare earth elements) by activity level and other nuclear properties as shown in Table 3.

The introduction of the most dangerous radionuclides into mineral-like matrices is viewed as a promising technology. While for common radionuclides one can find existing analogs for chemically stable minerals, for plutonium and minor actinides this requires a great deal of research, as such materials do not occur naturally.

Plans for modernizing the RT-1 plant calls for improving radiochemical technologies, sharply cutting radioactive waste volumes, and immobilizing medium- and low-level wastes.

TABLE 3 Fractionation of Radionuclides at the Mayak Production Association

Year of Operation	Volume of High-Level Wastes Processed (m^3)	Specific Activity of High-Level Wastes (Ci/dm^3)	Cs-Sr Concentrate Extracted $\times 10^6$ Ci	Transplutonium and Rare Earth Elements Extracted	
				α-activity $\times 10^3$ Ci	β-activity $\times 10^3$ Ci
1996	210	32.4	7.5	—	—
1998	95	20.0	4.8	—	—
1999	62	20.3	1.5	1.9	37.3
2000	276	27.3	6.8	125.8	313.8
2001	586	27.0	16.112	290.2	970
Total	1229	—	36.712	417.9	1320.8

CONCLUSION

In implementing even the temporary storage of wastes, we representatives of today's nuclear power industry can by no means sit by peacefully with our hands folded. We must discuss this problem more widely, collaborate more closely, and seek alternative new technologies—and having reached a decision, we must act. Let us act together!

I would like to note the need to expand scientific-technical and commercial cooperation among nuclear countries with regard to the management of spent nuclear fuel and radioactive wastes. In this regard, the creation of major international complexes for storing and reprocessing spent fuel and possibly manufacturing new fuel and recycling radioactive wastes should be based on existing enterprises that have the necessary technologies and, most importantly, the experience—for example, such enterprises as the radiochemical plants in France and Great Britain and the industrial enterprises of Minatom.

Feasibility of Transmutation of Radioactive Elements

Sekazi K. Mtingwa
Massachusetts Institute of Technology
North Carolina A&T State University

One of the most formidable obstacles to exploiting the full potential of nuclear energy is the long-term disposal of highly radioactive waste that is generated by the burning of fuel in nuclear reactors. There is a hope by many that much of the highly radioactive waste can be transmuted to a form that poses much less of a hazard to the environment. To develop such a transmutation technology, the U.S. Department of Energy (DOE) has instituted several additional programs.

In October 1999 DOE's Office of Civilian Radioactive Waste Management submitted a Report to Congress called "A Roadmap for Developing Accelerator Transmutation of Waste (ATW) Technology."[1] At the time it seemed feasible to use accelerator-generated spallation neutrons to transmute high-level radioactive waste. In the report DOE identified a host of technical issues for the ATW Program, proposed a program and schedule to resolve those issues, estimated the cost of such a program, proposed international collaborations, and assessed the impact of ATW technology on spent fuel from civilian nuclear reactors.

At about the same time DOE established the Accelerator Transmutation of Waste Subcommittee under its Nuclear Energy Research Advisory Committee (NERAC), the top advisory panel to the secretary of energy on matters related to nuclear energy, science, and technology. DOE charged the ATW Subcommittee with reviewing its overall ATW Program and making recommendations on future ATW research and development (R&D). Soon after the establishment of the ATW Subcommittee, DOE merged its ATW Program with its Accelerator Production of Tritium Program, calling the new program the Advanced Accelerator Applications Program. Guided by the recommendations of its ATW Subcommittee, DOE decided that fast neutrons from the fleet of next-generation nuclear reactors, called Generation IV (GEN-IV) reactors, might be preferable to accelerator-generated spallation neutrons for transmuting radioactive waste. Consistent with its recommendation, the ATW Subcommittee changed its name to the Advanced Nuclear Transformation Technology (ANTT) Subcommittee, thereby de-emphasizing the role of accelerators in the U.S. transmutation program.

Most recently DOE decided that its GEN-IV and transmutation programs needed more coordination to maintain consistency between the two sets of technologies. To promote this coordination Congress established a new program in 2003 called the Advanced Fuel Cycle Initiative (AFCI), under which DOE is charged with developing both advanced fuels for GEN-IV reactors and technologies for spent fuels reprocessing and transmutation.

Research on transmuting radioactive waste is in its infancy, and there is much to be done to make it a reality.

STATEMENT OF THE PROBLEM

The long-term storage, by which I mean the permanent burial, of highly radioactive waste from nuclear reactors is a major obstacle to exploiting fully the potential of nuclear energy. In the United States high-level waste from its roughly 100 civilian nuclear reactors is stored temporarily near the reactors at some 130 sites around the country until some long-term storage facility is commissioned. The most likely site for a permanent repository is Yucca Mountain in the state of Nevada, about 100 miles northwest of Las Vegas. The waste would be buried some 800 feet below the surface and about 1000 feet above the water table. The 5000-foot mountain is located in a desert region that receives about 6 inches of rainfall per year, most of which evaporates. Tentatively the spent fuel would be sealed inside containers made of a corrosion-resistant steel alloy containing nickel, chromium, molybdenum, and tungsten, and the spent fuel would be protected further by titanium drip shields.

The statement of the problem facing the long-term storage of high-level waste from nuclear reactors is as follows:

> To be licensed by the U.S. Nuclear Regulatory Commission as a permanent repository for high-level waste from nuclear reactors, the containers to be used to encase the reactor waste at Yucca Mountain must be corrosion-resistant and leak-proof for 10,000 years.[2]
>
> If the reactor waste is not transmuted, it will be highly radioactive for about hundreds of thousands of years.
>
> In case of waste leakage after 10,000 years can we trust the geologic integrity of the site to prevent the waste from diffusing into the water table or other parts of the environment?

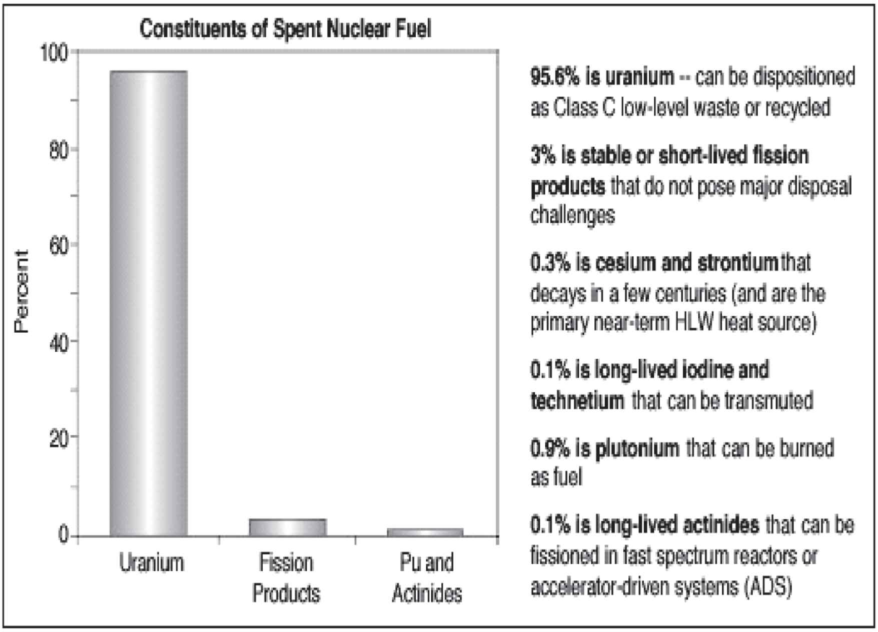
The goal of the U.S. transmutation program is to solve this problem by reducing the radioactivity of the high-level waste, in a period not to exceed 10,000 years, to a level less than that of the natural uranium ore from which the original fuel was made.[3]

To better understand the composition of waste from reactors, I show in Figure 1 a rough breakdown of the constituents of spent fuel from a typical civilian reactor in the United States.

A more detailed breakdown of the composition of 1 metric ton of pressurized water reactor (PWR) fuel (approximately 2 fuel assemblies) at 50 MWd/kg burn-up after cooling for 10 years is in Table 1.

As for the principal contributors to the radioactivity of PWR spent fuel at 50 MWd/kg burn-up, 10 years' cooling, we have the data in Table 2.

If it proves to be feasible, transmutation could have a major benefit for the size and cost of Yucca Mountain and future repositories. Transmutation means the transformation of one atom into another by changing its nuclear structure. In the present context this means bombarding a highly radioactive atom with neutrons, preferably fast neutrons, from either a fast nuclear reactor or spallation



95.6% is uranium -- can be dispositioned as Class C low-level waste or recycled

3% is stable or short-lived fission products that do not pose major disposal challenges

0.3% is cesium and strontium that decays in a few centuries (and are the primary near-term HLW heat source)

0.1% is long-lived iodine and technetium that can be transmuted

0.9% is plutonium that can be burned as fuel

0.1% is long-lived actinides that can be fissioned in fast spectrum reactors or accelerator-driven systems (ADS)

FIGURE 1 Constituents of spent fuel from a typical U.S. nuclear reactor. SOURCE: U.S. Department of Energy. Report to Congress on Advanced Fuel Cycle Initiative: The Future Path for Advanced Spent Fuel Treatment and Transmutation Research. Washington, D.C.: U.S. Department of Energy, Office of Nuclear Energy, Science, and Technology, January 2003, p. II-2.

TABLE 1 Detailed Composition of 1 Metric Ton of Spent Nuclear Fuel

	Fission Products
	10.1 kg lanthanides
955.4 kg U	1.5 kg ^{137}Cs
8.5 kg Pu (5.1 kg ^{239}Pu)	0.7 kg ^{90}Sr
0.5 kg ^{237}Np	0.2 kg ^{129}I
1.6 kg Am	0.8 kg ^{99}Tc
0.02 kg Cm	0.006 kg ^{79}Se
34.8 kg fission products	0.3 kg ^{135}Cs
	3.4 kg Mo isotopes
	2.2 kg Ru isotopes
	0.4 kg Rh isotopes
	1.4 kg Pd isotopes

SOURCE: James Laidler. Development of Separations Technologies Under the Advanced Fuel Cycle Initiative. Report to the ANTT Subcommittee. December 2002.

neutrons created by bombarding protons from a high-energy accelerator on a suitable target. Two examples of transmutation are shown in Figure 2.

In addition to transmuting the highly radioactive constituents of spent fuel, there is the possibility of separating out the uranium to sufficient purity that it could be disposed of as Class C low-level waste or reused in reactors. The potential benefits of transmutation and such uranium separation can be seen in Figure 3, which displays a graph of the accumulation over time of civilian spent fuel in the United States both with and without transmutation and uranium separation.

Currently there are about 44,000 metric tons of spent nuclear fuel residing at commercial nuclear power plants in the United States, with some 2000 metric tons being generated each year. The statutory limit for Yucca Mountain is 63,000 tons, and the United States should reach that limit by the year 2015. After 2015 either Yucca Mountain will have to be expanded greatly or a new repository will have to be constructed. If transmutation and uranium separation prove implementable on a commercial scale, the quantity of waste sent to the repository could be stabilized at a level that would eliminate the need for a significant expansion of Yucca Mountain or even a second repository.

The significant cost benefits derived from reprocessing the spent fuel going into Yucca Mountain are shown in the DOE estimates in Table 3.

With the separation of uranium and transmutation of other highly radioactive components of the spent fuel, mainly the transuranics, a second repository

TABLE 2 Principal Contributors to the Radioactivity of PWR Spent Fuel

Isotope	Sievert/Metric Ton (1 sievert = 100 rem)
U-236	6.0E + 02
U-238	5.0E + 02
Np-237	3.0E + 03
Pu-238	3.5E + 07
Pu-239	2.8E + 06
Pu-241	2.0E + 07
Pu-242	2.0E + 04
Am-241	1.9E + 07
Am-243	7.7E + 05
Cm-244	4.9E + 07
Sr-90	9.2E + 07
Cs-134	1.4E + 07
Cs-137	6.3E + 07
Y-90	8.9E + 06
Ce-144	3.7E + 04
Pr-144	3.5E + 02
Pm-147	6.6E + 04
Sm-151	5.0E + 03
Eu-154	8.7E + 05
Eu-155	1.5E + 04
Ru-106	2.0E + 05

SOURCE: James Laidler. Development of Separations Technologies Under the Advanced Fuel Cycle Initiative. Report to the ANTT Subcommittee. December 2002.

would not be needed. With no reprocessing, a second repository is estimated to cost $35 billion. The total savings for disposal from separation of uranium and transmutation is estimated at about $53 billion.

In addition to the significant cost savings for the first repository and the elimination of the need for a second repository, uranium separation and transmutation could serve the worthwhile goal of reducing the time required for the radiotoxicity of the waste in the repository to settle to the level of natural uranium, and this reduction would be from about 300,000 years to several hundred years, as shown in Figure 4.

I have discussed some of the main problems facing the long-term storage of spent nuclear fuel and have suggested that if they could be implemented on a commercial scale, transmutation and uranium separation would have major benefits for the cost and size of the first repository, and even eliminate the need for future repositories.

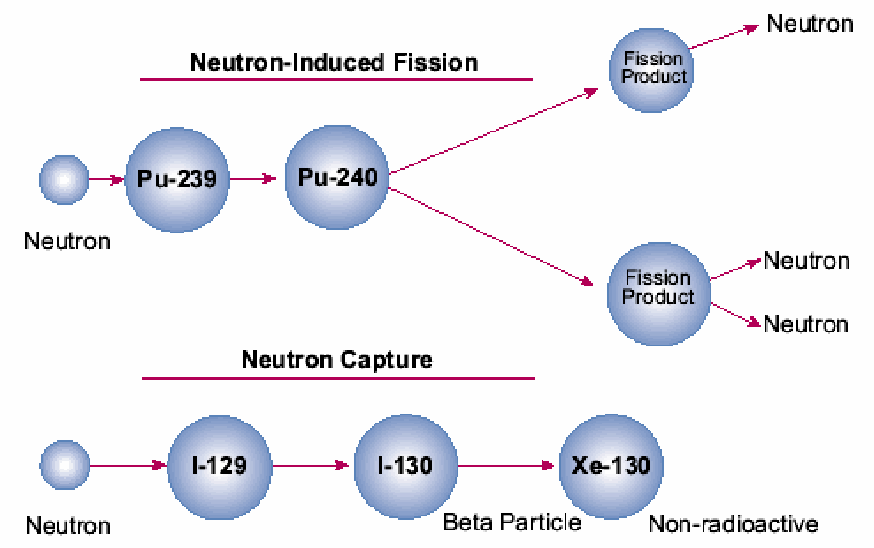


FIGURE 2 Examples of transmutation. SOURCE: U.S. Department of Energy. A Report to Congress: A Roadmap for Developing Accelerator Transmutation of Waste (ATW) Technology, DOE/RW-0519. Washington, D.C.: U.S. Department of Energy, Office of Civilian Radioactive Waste Management, October 1999, pp. 1–4.

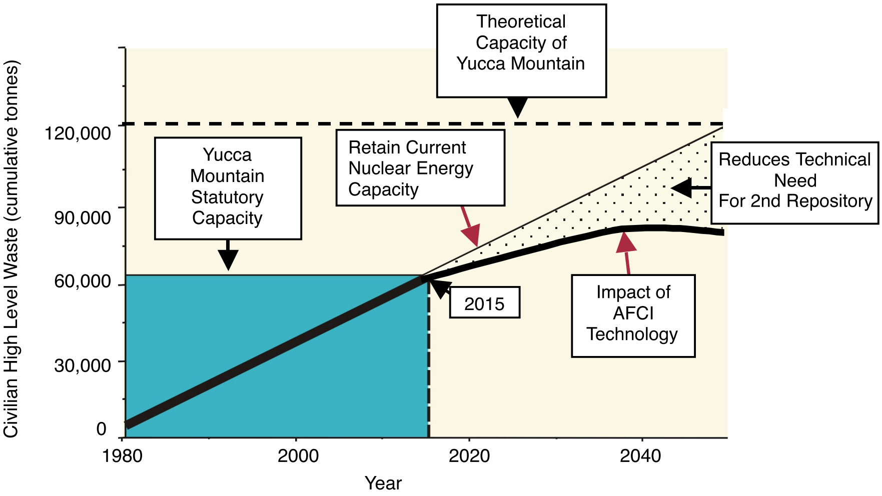


FIGURE 3 Civilian spent fuel accumulation in the United States both with and without transmutation and uranium separation under the AFCI Program. SOURCE: U.S. Department of Energy. Report to Congress on Advanced Fuel Cycle Initiative: The Future Path for Advanced Spent Fuel Treatment and Transmutation Research. Washington, D.C.: U.S. Department of Energy, 2003, pp. I-3, II-4.

TABLE 3 Disposal Cost Benefits Derived from Reprocessing Spent Fuel (in billions of U.S. dollars)

Cost Element	No Reprocessing	Reprocessing in 2010
Site characterization	6.7	6.7
Surface facilities	7.7	5.0
Subsurface facilities	9.0	5.8
Waste pkg/drip shield	13.4	5.5
Performance confirmation	2.3	2.3
Management	3.1	3.1
Waste acceptance and transport	6.0	3.0
Nevada transport	0.8	0.8
Program integration	4.3	4.3
Site characterization	4.6	3.5
TOTAL	57.9	40.0

SOURCE: James Laidler. Development of Separations Technologies Under the Advanced Fuel Cycle Initiative. Report to the ANTT Subcommittee. December 2002.

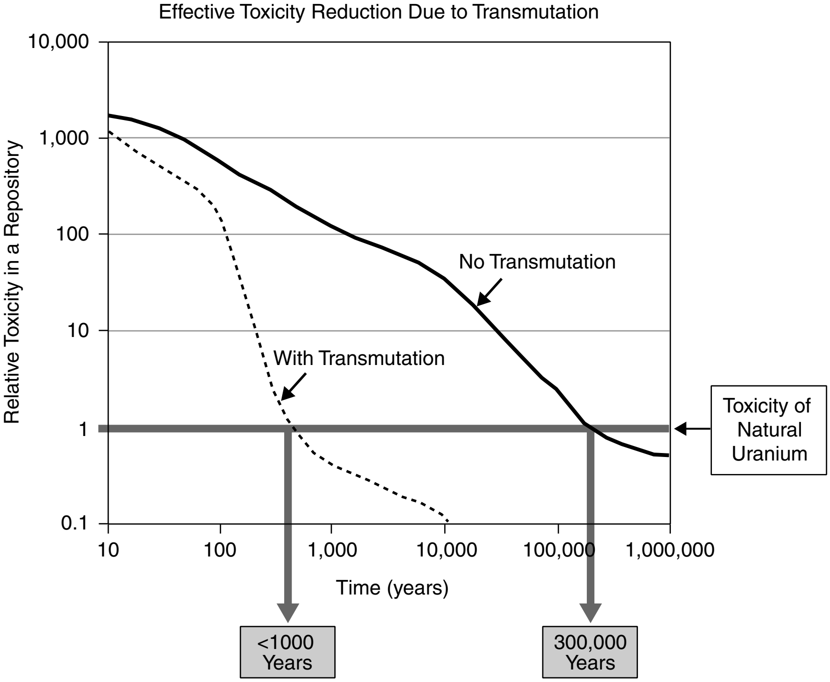


FIGURE 4 Radiotoxicity relative to natural uranium versus time, with and without uranium separation and transmutation. SOURCE: U.S. Department of Energy. Report to Congress on Advanced Fuel Cycle Initiative: The Future Path for Advanced Spent Fuel Treatment and Transmutation Research. Washington, D.C.: U.S. Department of Energy, 2003, p. III-2.

CRITERIA SET FOR THE U.S. TRANSMUTATION PROGRAM

NERAC is the highest panel of experts that provides advice to the U.S. secretary of energy on issues pertaining to civilian nuclear energy, science, and technology. In 1999 NERAC established the ATW Subcommittee to provide advice on the accelerator transmutation of high-level waste from civilian nuclear reactors. Accelerator-induced transmutation has a unique set of problems, such as accelerator reliability and the uncertain problems of coupling an accelerator to the remaining transmutation complex. Given the formidable time and expense to develop this accelerator-based technology, and given the ability of fast reactors (such as those already contemplated for the next generation of nuclear reactors called GEN-IV) to do the same job, the subcommittee early in its deliberations decided to change its name to the Advanced Nuclear Transformation Technology (ANTT) Subcommittee and recommended that DOE not emphasize the use of accelerators for transmutation but place more emphasis on the use of fast reactors.

The current membership of ANTT is as follows:

- Burton Richter, Chair, Nobel Laureate, Stanford Linear Accelerator Center
- Darleane Hoffman, University of California, Berkeley
- Sekazi Mtingwa, Massachusetts Institute of Technology and North Carolina A&T State University
- Ronald Omberg, Pacific Northwest National Laboratory
- Joy Rempe, Idaho National Engineering and Environmental Laboratory

In order to maintain the focus and effectively evaluate the U.S. transmutation research and development program, the ANTT Subcommittee has established the following four criteria to be met by any transmutation and separations technology:

1. **Reduce the long-term radiological impact of spent nuclear fuel.** The minimum goal should be the reduction of the radiological impact of spent fuel to below that of the ore from which it came in a time period equal to or less than the Nuclear Regulatory Commission's licensing period, now set at 10,000 years. To accomplish this goal the maximum allowable amounts of plutonium and higher actinides in the final waste stream must be severely limited.[4]
2. **Provide substantial benefits to the repository, making it simpler and cheaper.** Given that the legislated limit is 63,000 tons for the capacity of the first repository using only the once-through fuel cycle in the United States, and given that the existing U.S. reactors will produce this amount by the year 2015, the first repository is in dire need of any help that it can derive from transmutation and separations technology. The situation is especially critical when one consid-

ers that the quantity of spent fuel will double the 63,000 ton statutory limit of Yucca Mountain in about 50 years, and even sooner if nuclear power expands. Luckily it appears that uranium separation and the transmutation of long-lived elements in the waste could reduce the mass going to the repository by a factor of about 20 and the volume by a factor of about 4.

3. **Reduce the proliferation risk.** Without spent fuel processing and transmutation, even at a constant level of nuclear power, the world's plutonium inventory will continue to increase. With the transmutation of plutonium, the inventory could be stabilized at an equilibrium level lower than what exists now, and it could be put in an isotopic form less amenable to terrorists and more difficult to use for the creation of weapons of mass destruction. A big challenge is that the decrease in quantity of plutonium to the repository is accompanied by an increased availability of plutonium in the system from material in process.
4. **Improve the long-term prospects of nuclear power.** Having the first and third criteria certainly helps in satisfying this criterion, since any long-term prospects for nuclear power hinges on proving to the public that the proliferation risk and radiological impact of spent fuel can be minimized. The economic benefits of transmutation and uranium separation are only now beginning to be analyzed, and the early numbers are encouraging. It appears that transmutation and uranium separation could save tens of billions of U.S. dollars on repository costs and even eliminate the need for additional repositories.

To date, the above ANTT criteria have served the U.S. transmutation program well in maintaining its focus.

GENERATION IV ACTIVITIES

The grand purpose of the GEN-IV Program is to identify and down-select the most promising technologies for sustaining, and even increasing, nuclear energy production for the rest of the twenty-first century. This truly has been a coordinated international effort. In 2001 the United States and eight other countries established an international working group called the Generation IV International Forum (GIF) in order to create a common, international nuclear research and development agenda for the next generation of nuclear reactors. The current membership includes 10 countries: Argentina, Brazil, Canada, France, Japan, Republic of South Africa, Republic of Korea, Switzerland, United Kingdom, and the United States. Working with GIF, another DOE NERAC Subcommittee, called the GEN-IV NERAC Subcommittee (GRNS), cochaired by Professor Neil Todreas of the Massachusetts Institute of Technology and Salomon Levy of Levy & Associates, produced the Generation IV Technology Roadmap, which has identified six nuclear reactor technologies that should receive highest priority for future consideration. The main goals of the GEN-IV Program are the following:[5]

- provide sustainable energy generation that meets clean air objectives
- minimize and efficiently manage GEN-IV nuclear waste in order to protect the public health and the environment
- ascertain the economic competitiveness of GEN-IV nuclear reactors versus other energy-producing technologies
- ensure the high level of safety and reliability for GEN-IV systems
- maximize the proliferation resistance of weapons-usable material, as well as protect such materials from theft by terrorists

The down-selected reactor technologies that GIF has assigned the highest priority are the following:

- very-high temperature reactor (VHTR)
- supercritical water-cooled reactor (SCWR)
- sodium-cooled fast reactor (SFR)
- gas-cooled fast reactor (GFR)
- lead-bismuth-cooled fast reactor (LFR)
- molten salt reactor (MSR)

While the international community will study all six concepts, DOE's GEN-IV program will place highest priority on the first four. As of May 2003 the VHTR is of highest priority because of its hydrogen-generating capability, SCWR is next, and the SFR and GFR seem to be a distant third. As for compatibility with transmutation, the VHTR and SCWR are not capable of adequately burning the minor actinides in the recycled fuel and the GFR is of limited capability. In its most recent report to NERAC and DOE the ANTT Subcommittee emphasized the importance for DOE to continue research and development on the SFR, because it is the most compatible of the four for performing a final burn of the minor actinides.[6]

THE ADVANCED FUEL CYCLE INITIATIVE

In order to improve the coordination of the GEN-IV and transmutation research and development programs so that technologies considered for next-generation reactors are compatible with the transmutation option, the U.S. Congress established a new program in 2003 called the Advanced Fuel Cycle Initiative (AFCI), under which DOE is charged with developing both advanced fuels for GEN-IV reactors and technologies for waste transmutation.

To ensure a high level of coordination DOE has appointed Ralph Bennett of the Idaho National Engineering and Environmental Laboratory to serve as the national technical director of systems analysis for the AFCI. The program has been divided into the following three subprograms, with the corresponding director and sample responsibilities:

1. fuels development—Kemal Pasamehmetoglu, Los Alamos National Laboratory; fuel forms: oxide, nitride, metal, dispersion, ceramic, coated particles; fabrication techniques
2. separations technologies—James Laidler, Argonne National Laboratory; advanced aqueous chemical fuel treatments; pyroprocessing; waste forms; group separations
3. transmutation technologies—Michael Cappiello, Los Alamos National Laboratory; materials; physics; targets; accelerator-driven systems (ADS)

Responding to the advice of the ANTT Subcommittee, DOE has set the following overall waste treatment goals for the AFCI:

- reduce the radiotoxicity of high-level nuclear waste to that of natural uranium ore within 1000 years
- reduce high-level nuclear wastes: mass by a factor of 20 and volume by a factor of 4
- reduce the civilian inventories of plutonium in forms that are conducive to weapons proliferation and terrorist activities
- reduce the cost of geologic waste disposal, with a possible net savings over $35 billion during the period 2007–2040

The AFCI is divided into two broad initiatives, called Series One and Series Two.

AFCI SERIES ONE PROGRAM

The mission of the AFCI Series One Program is as follows:

- Develop technologies applicable to current and near-term reactors.
- Address the intermediate-term goals for separations, transmutation, and GEN-IV fuels technologies. Among other things, this goal will address the reduction in cost of spent nuclear fuel disposal by decreasing the mass and volume of high-level waste to the repository. This goal will also address the reduction of the long-term proliferation threat posed by plutonium contained in spent fuel.
- Make policy recommendations to the U.S. Congress by the period 2007–2010 about the need for a second repository.
- Research the extraction of unspent energy from the reactor waste. The transmutation of plutonium and the minor actinides could provide a 25 percent increase in the energy extracted from reactor fuel compared to the current once-through cycle used in the United States.
- Continue with the development of the Uranium Extraction (UREX) and UREX+ separations technologies. In the late 1940s the United States developed an aqueous chemical treatment technology called Plutonium-Uranium Extrac-

tion (PUREX), and currently this technology is in use in France, Russia, and the United Kingdom. In the PUREX process spent fuel is dissolved in acid and fed through a solvent extraction process, separating both uranium and plutonium. Although separating out the uranium has a definite benefit for a repository, a major problem with PUREX is that the plutonium is partitioned out, which poses a proliferation risk. Recently DOE has pursued a different aqueous chemical treatment technology called UREX, which enhances the proliferation resistance of the spent fuel by separating out the uranium, while at the same time, keeping the plutonium combined with other radioactive species. In an even more advanced process called UREX+, selected actinides and fission products can be separated out in various combinations after the uranium has been removed; for example, mixtures of plutonium and certain of the minor actinides could be partitioned together to enhance the proliferation resistance of the plutonium-containing material. Also, long-lived fission products, such as iodine-129 and technetium-99, could be incorporated into targets for destruction in reactors.

In August 2002 a DOE team under the leadership of James Laidler of Argonne National Laboratory performed experiments at the Savannah River Technology Center and demonstrated that UREX could recover nearly all the uranium at 99.999 percent purity from spent light water reactor (LWR) fuel, thus yielding a level of contamination below Nuclear Regulatory Commission criteria for disposal as Class C low-level waste.[7] Flowsheets for UREX, UREX+, and related processes are shown in Figures 5–8.

The new UREX/UREX+ technology could play an important role in reducing the cost of the first repository by separating out the uranium and disposing of it as Class C low-level waste and by rendering the reprocessed spent nuclear fuel proliferation-resistant by keeping the plutonium mixed with other radioactive elements. An important goal of the AFCI Series One Program will be both laboratory-scale and engineering-scale demonstrations of UREX/UREX+.

The AFCI Series One Program will pursue laboratory and engineering-scale demonstrations of pyrochemical dry treatment (PYROX) technology using spent LWR fuel, including actinide recovery. The program will also pursue the demonstration of large-scale metal waste form technologies, treatment facility designs, and cost estimates.

AFCI SERIES TWO PROGRAM

The main focus of the AFCI Series Two Program is on the fuel cycle technology associated with the next generation of nuclear reactors, especially the fast neutron spectrum reactors. On the advice of the ANTT Subcommittee[8] DOE has divided Series Two into Phases I, II, and III, and there is some overlap with AFCI Series One.

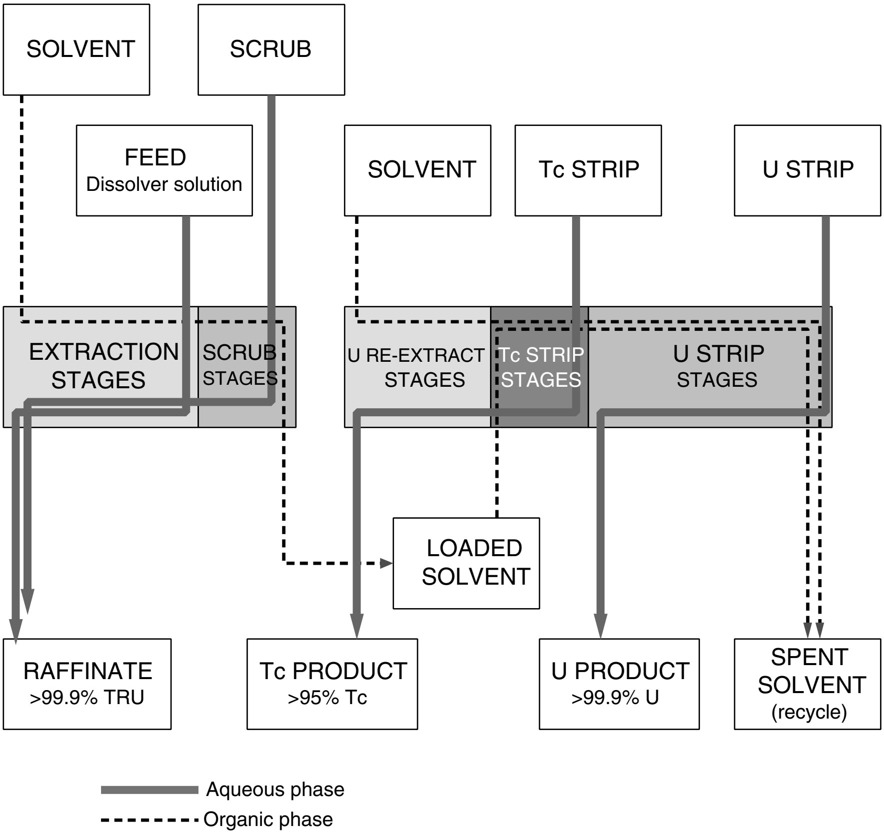


FIGURE 5 UREX process flowsheet. SOURCE: James Laidler. Development of Separations Technologies Under the Advanced Fuel Cycle Initiative. Report to the ANTT Subcommittee. December 2002.

Phase I: Basic Technology Evaluation

DOE considers Phase I to be essentially completed as of the year 2002 and mainly involves an initial evaluation of promising fuel and fuel processing technologies. Major Phase I accomplishments are as follows:

1. treatment of spent fuel to demonstrate the feasibility of extracting uranium to high purity; the initial success of the UREX process in AFCI Series One should allow UREX to be a viable option for the front-end fuels treatment for transmuting Series Two fuels

2. successful manufacture of transmutation fuels containing various combinations of plutonium and the minor actinides in preparation for irradiation testing in 2003

3. construction and commissioning of a lead-bismuth materials loop at Los Alamos National Laboratory to investigate materials behavior in a high-temperature liquid metal environment

4. completion of studies that analyzed several transmutation systems to determine which have the highest potential to reduce the radiotoxicity of spent nuclear fuel

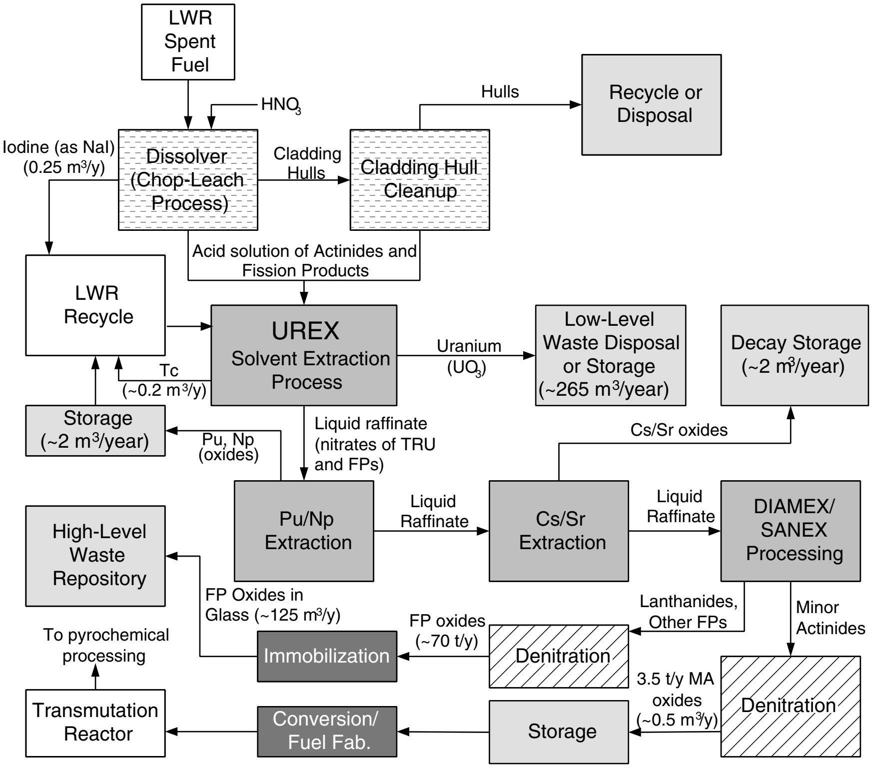


FIGURE 6 UREX+ process flowsheet. (Storage volumes based on 2000 tons spent nuclear fuel per year, 10-year cooling; 2210 m^3 per year if disposed directly in a geologic repository). SOURCE: James Laidler. Development of Separations Technologies Under the Advanced Fuel Cycle Initiative. Report to the ANTT Subcommittee. December 2002.

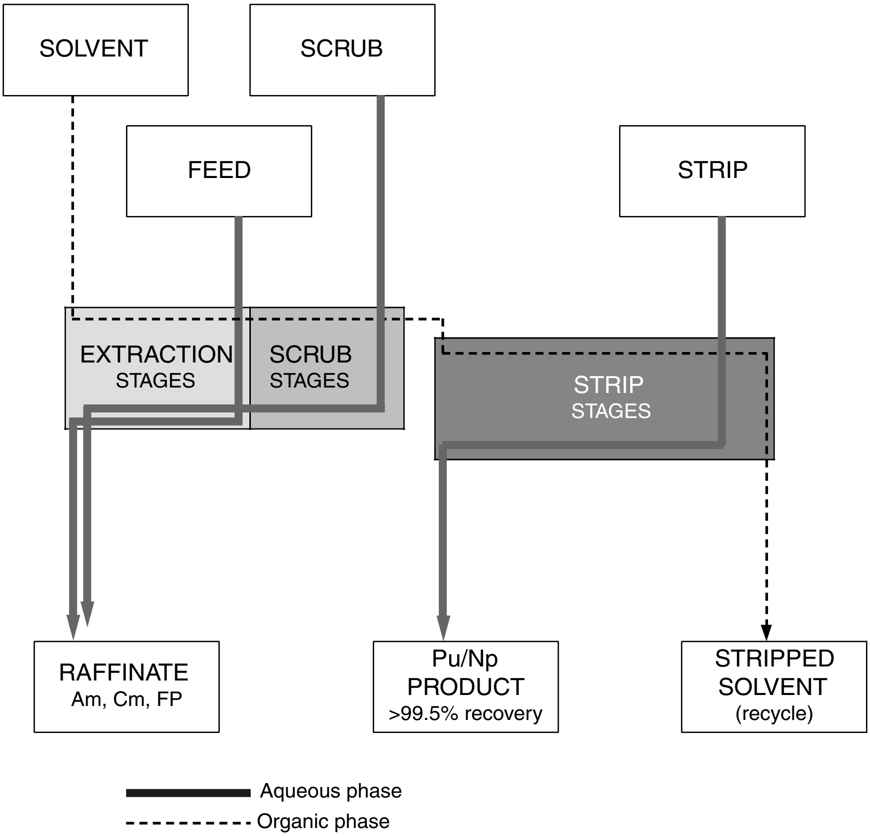


FIGURE 7 Pu/Np extraction flowsheet. SOURCE: James Laidler. Development of Separations Technologies Under the Advanced Fuel Cycle Initiative. Report to the ANTT Subcommittee. December 2002.

Phase II: Proof of Principle

Under Phase II of the AFCI Program, which is considered the proof-of-principle phase, the following activities will be pursued:

- The most promising technologies from Phase I, such as UREX/UREX+, will be identified for focused research and development. While many experts believe that UREX/UREX+ can be implemented on a commercial scale, dry processes like pyroprocessing may have advantages in handling large concentrations of transuranics. Some believe that pyroprocessing may be more efficient and proliferation-resistant. These advantages must be verified for implementation at a commercial scale and will be pursued in Phase II.

- Laboratory and larger-scale testing will be done to clarify various technology options, thereby providing important information needed to choose the best path forward for Phase III.
- More work will be pursued on the development and demonstration of advanced proliferation-resistant treatment technologies.
- The development and testing of advanced transmutation fuels will receive greater study, especially since there is little work to date on making minor actinide fuels. Irradiation tests of fuel forms, especially those containing mixtures of plutonium with the minor actinides, will be performed at the advanced test reactor, a 250 MW light water reactor located at the Idaho National Engineering and Environmental Laboratory. An international collaboration to test minor actinide fuel forms is being planned for experiments at PHENIX, a fast

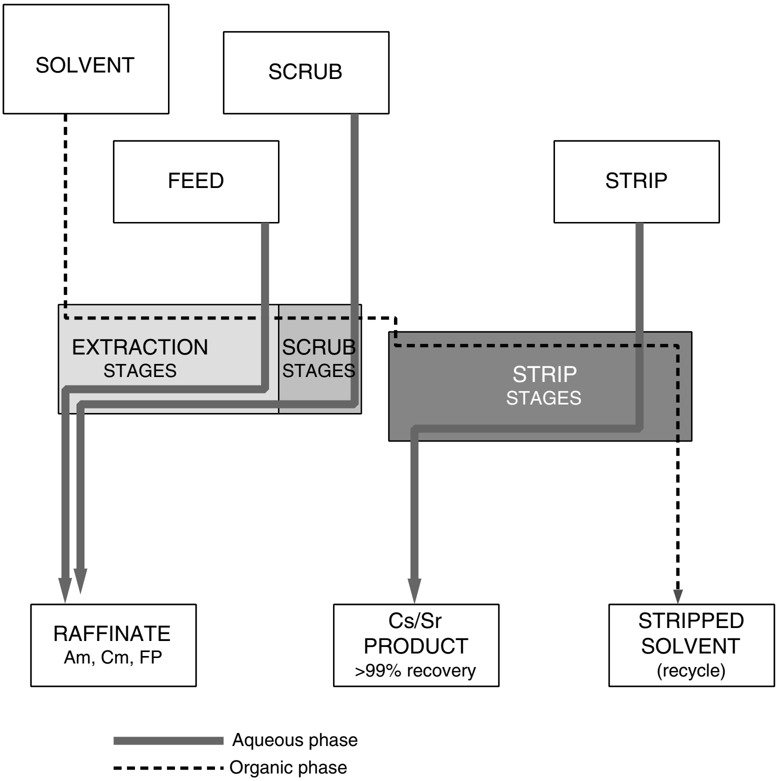


FIGURE 8 Cs/Sr extraction flowsheet. SOURCE: James Laidler. Development of Separations Technologies Under the Advanced Fuel Cycle Initiative. Report to the ANTT Subcommittee. December 2002.

spectrum reactor located in France. Another goal for Phase II is to use experimental data to compile a fuels handbook of the physical, chemical, and thermal properties of the transmutation fuel forms.

- Preliminary technology will be developed for an ADS. DOE has retreated from pursuing the large-scale deployment of ADS complexes as the primary means of transmutation and has decided to concentrate on the use of fast reactors for that purpose. It has been estimated that the cost of implementing an accelerator-only approach could be as high as $280 billion.[9] If a suitable fast reactor is constructed among the GEN-IV fleet of future reactors, the large-scale use of ADS technology should not be necessary. There is some interest in studying the smaller-scale use of a centralized ADS facility for a final burn of the transuranics before disposition in a repository. An example of an ADS concept is depicted in Figure 9.

Phase II may be the most important part of the overall AFCI Program, since the efficacy of the chosen technologies will have to be proven, at least at a level with potential scalability. Important results from Phase II should include the final selection and demonstration of optimal fuel forms and advanced fuels treatment processes, the selection of an ideal ADS target material, and a comprehensive assessment of the costs and benefits of each down-selected technology.

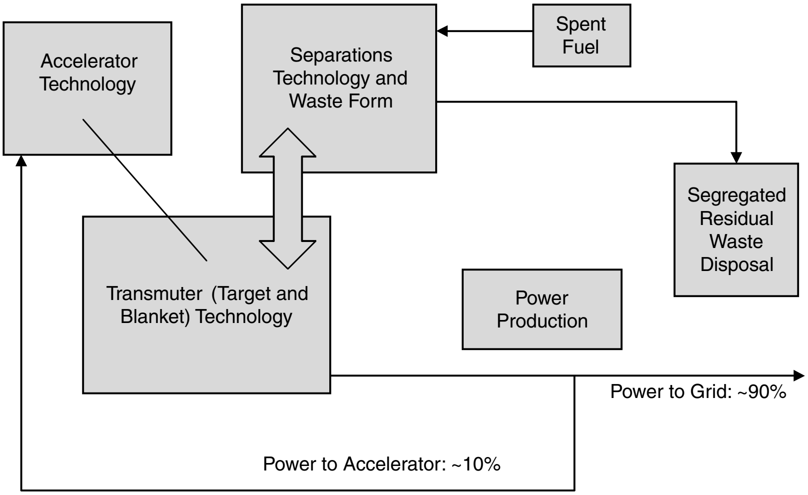


FIGURE 9 Components of an accelerator-driven system. SOURCE: U.S. Department of Energy. A Report to Congress: A Roadmap for Developing Accelerator Transmutation of Waste (ATW) Technology, DOE/RW-0519. Washington, D.C.: U.S. Department of Energy, Office of Civilian Radioactive Waste Management, October 1999, p. E-3.

This work should focus the AFCI research and development as it proceeds into Phase III.

Phase III: Proof of Performance

In this final phase of the AFCI Program the research and development will proceed to a full-scale proof-of-performance demonstration of separations and transmutation technologies down-selected from Phase II. It is during this stage of AFCI that full-scale prototypes of the technologies will be constructed and tested. If the tests prove successful, the next stage will be the commercial-scale implementation of the next-generation technologies.

To ensure the success of the AFCI Program it will be crucial to organize international collaborations to study the many unsolved issues.

INTERNATIONAL COLLABORATIONS

Focusing on reactor concepts, the Generation IV international community has proceeded much more swiftly to initiate international partnerships than has the transmutation world community. The formation of GIF is an excellent example of the kind of world cooperation that is essential in order to tackle the many difficult issues surrounding next-generation reactors and the permanent disposal of reactor wastes. It will be a big step forward when Russia joins the GIF partnership.

Notwithstanding the worldwide cooperation on GEN-IV issues and the importance of coordinating those efforts with separations and transmutation activities, there is minimal international cooperation on fuel separations and transmutation R&D. Keeping the transmutation program alive in the United States has not been easy, and the situation is not significantly better in other countries; this creates a dire need to form a Generation IV International Forum on Transmutation and Separations (GIFTS). Already there are the seeds of some limited partnerships. In the United States, DOE is fortunate to have received important transmutation research data from France and Switzerland that would have cost over $100 million to duplicate in the United States. Another seed of international collaborations is MEGAPIE, which seeks to demonstrate the safe operation of a liquid-metal lead-bismuth spallation target at a beam power of 1 MW in the SINQ target station of Switzerland's Paul Scherrer Institut. Originally launched by Switzerland, France, and Germany, the project currently involves a number of other countries, including the United States. The TRIGA Accelerator Driven Experiment (TRADE) is to be performed in the 1 MW TRIGA reactor of the ENEA Casaccia Centre in Italy. We can see that the seeds are now planted for a bolder push for more international transmutation and separations collaborations.

It is clear that budgets are strained worldwide, and there seems to be a lack of adequate funding to explore transmutation and separations technologies fully;

for example, since it has been difficult in the United States to keep the transmutation program alive, work on such topics as minor actinide fuel forms has been hampered. At the same time Russia is independently pursuing similar investigations with limited funding.[10] There is no better time than now to call for an international summit on separations and transmutation to share the research and development progress in countries actively pursuing this technology, gain a consensus on the important questions to be answered, and establish GIFTS to decide which international collaborations would be most meaningful.

FEASIBILITY OF TRANSMUTATION TECHNOLOGY

Saying that the transmutation of radioactive waste elements is feasible may be too strong a statement at the present time. A better statement would be that it seems plausible, and significantly more work is needed to further down-select competing technologies. The only way to move from plausibility to feasibility is to proceed along the lines of the U.S. AFCI Program through to the completion of Series Two/Phase II. Getting there will not be easy. There are many difficult problems that must be solved, and there is a need for a pooling of world resources through international collaborations. In the meantime there is the need for a close coordination between GIF and a GIFTS-type organization to ensure that GEN-IV and transmutation technologies are compatible by, for example, including a suitable fast reactor for burning the actinides in the fleet of future reactors. More work is needed on separations technologies, such as UREX/UREX+ and the complementary (or possibly superior) pyroprocessing techniques. On the physics front there is a critical need for such data as fast neutron cross-sections for plutonium and the minor actinides for input and checks on reactor simulation codes.

It is clear that if transmutation is to be realized, the effort will take over a decade of hard work. In the United States there is already a shortage of students pursuing nuclear engineering and radiochemistry. Although other countries may not be experiencing the same problem, as the United States produces a large fraction of the world's researchers in these fields, it becomes a world problem. It is critical that appropriate attention and resources be focused to confront this problem head-on. There are many problems to be solved, but the future still seems bright for the possibility of a full commercial-scale demonstration of nuclear transmutation.

NOTES

1. U.S. Department of Energy. A Report to Congress: A Roadmap for Developing Accelerator Transmutation of Waste (ATW) Technology, DOE/RW-0519. Washington, D.C.: U.S. Department of Energy, Office of Civilian Radioactive Waste Management, October 1999.
2. There is a dose and groundwater concentration limit for the first 10,000 years.
3. B. Richter, D. Hoffman, S. Mtingwa, R. Omberg, and J. Rempe. Report of the Advanced

Nuclear Transformation Technology Subcommittee of the Nuclear Energy Research Advisory Committee. April 2002. http://www.ne.doe.gov/nerac/ANTT2-02ReporttoNERAC1.pdf

4. B. Richter, D. Hoffman, S. Mtingwa, R. Omberg, and J. Rempe. Report of the Advanced Nuclear Transformation Technology Subcommittee of the Nuclear Energy Research Advisory Committee. April 2002. http://www.ne.doe.gov/nerac/ANTT2-02ReporttoNERAC1.pdf

5. R. Bennett. Systems Analysis Overview. Report to the ANTT Subcommittee. December 2002.

6. B. Richter, D. Hoffman, S. Mtingwa, R. Omberg, and J. Rempe. Report of the Advanced Nuclear Transformation Technology Subcommittee of the Nuclear Energy Research Advisory Committee. January 2003. http://www.ne.doe.gov/nerac/antt14Jan_03.pdf

7. J. Laidler. Development of Separations Technologies Under the Advanced Fuel Cycle Initiative. Report to the ANTT Subcommittee. December 2002.

8. B. Richter, D. Hoffman, S. Mtingwa, R. Omberg, and J. Rempe. Report of the Advanced Nuclear Transformation Technology Subcommittee of the Nuclear Energy Research Advisory Committee. April 2002. http://www.ne.doe.gov/nerac/ANTT2-02ReporttoNERAC1.pdf

9. U.S. Department of Energy. Report to Congress on Advanced Fuel Cycle Initiative: The Future Path for Advanced Spent Fuel Treatment and Transmutation Research. Washington, D.C.: U.S. Department of Energy, Office of Nuclear Energy, Science, and Technology, 2003, p. I-2.

10. For example, see the talk at this workshop by V. Matveev et al.

The High-Level Waste Disposal Technology Development Program in Korea

Jongwon Choi
Korea Atomic Energy Research Institute

Korea launched a long-term research and development program for high-level waste (HLW) disposal technology development in 1997. The main purpose of this program is to establish a reference HLW repository system by 2006. The disposal concept being conceived in this program is to encapsulate the intact spent fuel in corrosion-resistant containers and the packaged spent fuel is then to be disposed in a mined underground facility located at about 500 m below surface in a crystalline rock mass. No site for the underground repository has been specified in Korea, but a generic site with granitic rock is considered for this study. The waste packages containing spent fuel are placed in the boreholes drilled in the floor of deposition tunnels. Many different alternatives concerning the emplacement patterns of the container, waste packaging methods, as well as the distance between deposition holes and tunnels were proposed, as seen in most countries that have established their own disposal concepts for high-level waste.

From the comparison of the proposed alternatives[1] the VSA concept, or vertical emplacement of separate packaging containers in separate areas, was suggested as the reference disposal concept and horizontal emplacement of containers in separate areas (the HAS concept) as an alternative concept. The reference concept accepts the separate-package method of the respective pressurized water reactor (PWR) and Canada deuterium uranium reactor (CANDU) fuel in different containers with consideration of the significantly different geometry and radiological characteristics of spent PWR and CANDU fuels generated from domestic nuclear power plants.

This paper addresses the preliminary conceptual design of the geological repository system based on the reference disposal concept suggested. The conceptual design study includes the layout of the underground facilities, the engineered barrier system, and the dimensions of the major repository system components needed to support the reference repository system design. This study is not site specific, because there are no site characterization data requiring repository performance to a specific location. Many design parameters and criteria are necessarily general or assumed.

DESIGN BASES

These design bases provide the information that identifies the specific functions to be performed by the repository system, as well as the specific values or ranges of values chosen as controlling parameters to bound the design. As a general guideline for this study[2] the conceptual repository will be located in granitic rock, the major rock type in Korea, at a depth nominally of 500 m between two large fault zones that extend from this depth to the surface. The rock quality varies from highly weathered surface deposits to competent rock at the repository horizon. For this conceptual design the distance between faults should be about 7 to 10 km. Therefore, the repository layout must be capable of fitting within these major structural features. The most stringent requirement for the base-case underground facility is that the temperature of the bentonite buffer material remains below 100°C throughout the lifetime of the repository.

The development of the underground repository system conceptual design requires specifications of (1) design constraints and criteria, (2) disposal canister design, and (3) waste form and throughput. Key design constraints are summarized in Box 1.

BOX 1
Specifications for the Conceptual Design of an Underground Repository

- Total spent fuel inventory to be packaged for disposal: 36,000 tHM[2]
- Decay heat (40-year cooling): 385 watt/assembly for PWR, 2.28 watt/bundle for CANDU
- The container filling material for void space within the container: carbon steel
- Emplacement sequence: CANDU spent fuel first, followed by PWR spent fuel
- Backfill design: vertical boreholes with bentonite buffer
- Repository layout: single-level repository
- Throughput: dictated by a 50-year operational life of repository
- Thermal constraints: surface temperature of disposal container limited to 100°C
- Waste package configuration: 4 PWR assemblies and 297 CANDU bundles
- Borehole spacing: 3 m (CANDU) and 6 m (PWR)
- Drift spacing: 40 m

REPOSITORY SYSTEM DESIGN

The major factors that influenced the base-case conceptual study include waste package size and weight, waste package thermal output, waste package receipt and emplacement rate(s), vertical borehole emplacement, and geologic setting of the proposed repository in terms of rock type, water conditions, topography, and rock quality.

Underground Facilities

The conceptual repository layout builds on the information and key constraints that define the emplacement tunnel dimensions and borehole separation distances, borehole layouts, and waste-receipt schedules. Figure 1 shows the isometric view of the various underground openings, including the disposal area, the service shaft complex, and the ventilation exhaust shaft complex. The entire facility is assumed to be constructed using drill and blast techniques. Drill and blast construction allows the flexibility of layout with the least amount of wasted space with excavations intersecting at right angles. The layout also assumes that the CANDU waste will be emplaced separately from the PWR waste. A separate CANDU emplacement area is identified at the lower left of the layout nearest the ventilation shaft complex.

As shown in the figure the base-case repository system includes two service mains that run the length of the repository from right to left connecting each shaft complex. The entire facility is bounded by a perimeter tunnel that functions for ventilation and access/operations. The disposal area consists of 8 disposal panels. Based on a 40-m emplacement tunnel spacing, each panel consists normally of 42 emplacement tunnels (for PWR) and includes 2 panel tunnels that are used for ventilation and to access the emplacement tunnels during concurrent construction and operation. The CANDU panel, located at the lower left, consists of 38 emplacement tunnels. Each emplacement tunnel is 250 m long. This includes a 6-m end standoff and a 12-m standoff at the entrance for placement of a bulkhead. This allows for the emplacement of 11,375 PWR containers and 2926 CANDU containers within 302 PWR and 34 CANDU emplacement tunnels at 6-m and 3-m spacing (to ensure that neither the maximum container surface temperature nor the maximum buffer temperature of 100°C is exceeded). There is an expansion area on the right side of the repository, which provides a 20 percent increase in the potential emplacement area (84 emplacement tunnels). The tentatively recommended dimensions of the emplacement tunnel and the borehole are shown in Figure 2. Regarding the proper configuration of the spent fuel containers satisfying the technical and safety criteria, the entire underground facility requires an area of about 4 km^2.

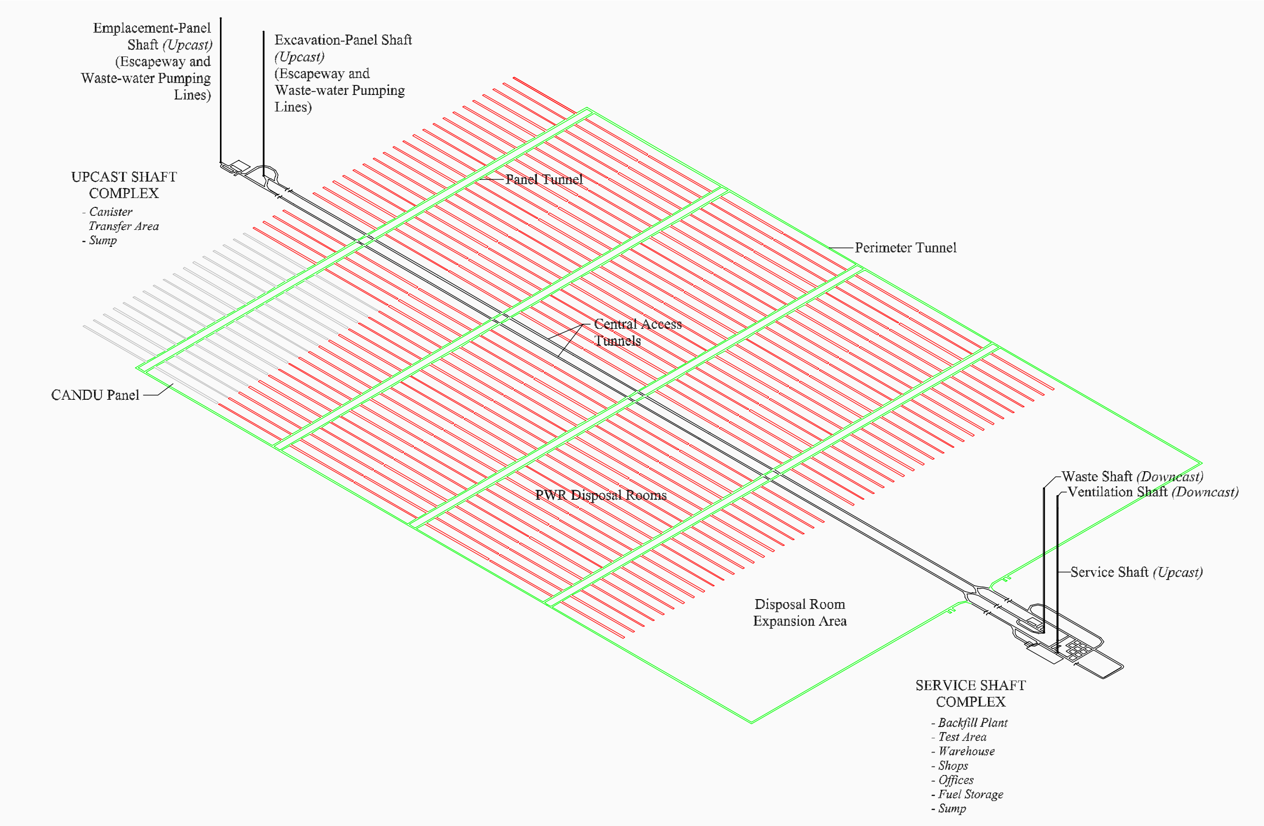


FIGURE 1 Isometric view of the preliminary conceptual geological repository.

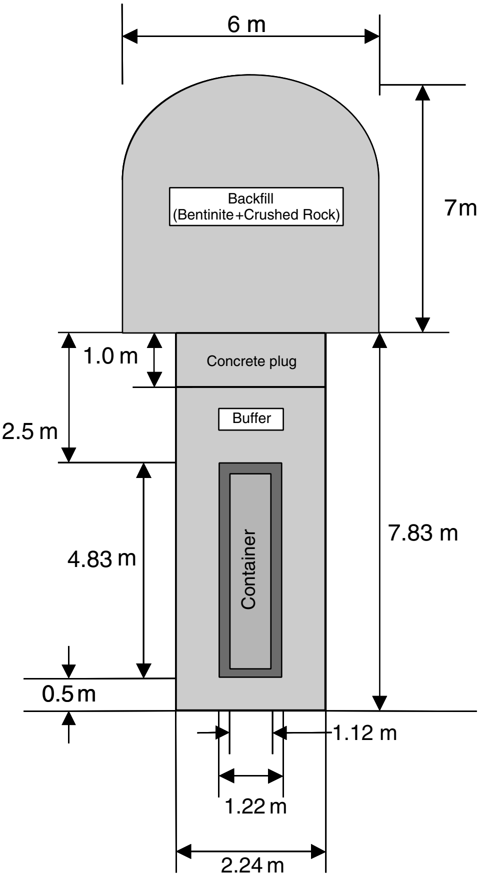


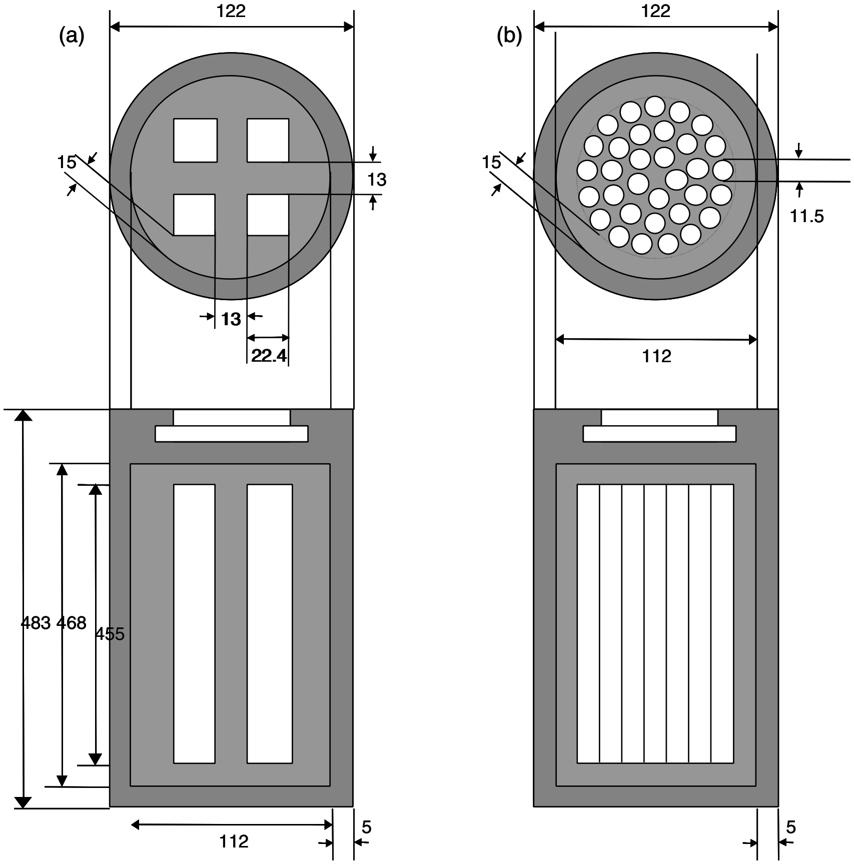
FIGURE 2 Engineered barrier design concept for the reference repository.

Engineered Barrier System

For this preliminary concept the repository containment concept of an engineered barrier system provides the primary containment and is protected by the natural barrier (that is, the geological formation) system. The engineered barrier system is composed of the waste types, the surrounding waste package, other engineered items in the underground facility, and buffer and backfill material. Design features of the engineered barrier system are the waste type, the waste canister package, and a bentonite buffer. Figure 2 shows the reference waste package and engineered barrier system concept applied for this study.

The reference spent PWR and CANDU fuels are defined in terms of initial enrichment, burn-up rates, dimension, gross weight, etc.[3] As a basis for the design, spent fuel inventories are estimated to be 36,000 tHM, based on the long-term National Nuclear Energy Plan. PWR fuel comprises approximately 20,000 tHM (55 percent of the total inventory to be disposed) of the projected spent fuel inventory. The reference PWR spent fuel has the average burn-up of 45,000 MWd/tHM (initial enrichment of 4 percent by weight) and is cooled for 40 years after irradiation before the encapsulation and disposal. PWR assembly weight and dimensions are 665 kg and 21.4 cm^2 (cross-section) × 453 cm (length), respectively. The spent CANDU fuel inventories are approximately 16,000 tHM, 45 percent of the total inventory to be disposed. The reference CANDU fuel has an average burn-up of 7500 MWd/tHM and the fuel dimensions are 10 cm (diameter) × 49.5 cm (length).

Because of the significantly different properties of both fuel types and the retrieval potential of PWR fuel for reuse, the reference container in which the spent PWR and CANDU fuels are separately encapsulated is illustrated in Figure 3. The overall sizes and component materials of the containers for both spent fuels are designed to be exactly identical to make the encapsulation and handling processes in the repository simple. The container being studied consists of two major components: a massive cast insert and a corrosion-resistant outer shell. The insert provides mechanical strength and radiation shielding, and it keeps the fuel assemblies in a fixed configuration. For the insert, carbon steel is considered for the design basis material. For the complete isolation of waste for a long time, high-nickel alloy (alloy 22), stainless steel, and copper are considered as the candidate corrosion-resistant materials for the outer shell. As shown in Figure 3, the outer shell contains fuel storage baskets (4 square tubes for spent PWR fuel and 33 circular tubes for spent CANDU fuel), and the void space between the fuel storage basket and the outer shell is filled with carbon steel called the cast insert. The loading capacity of the container was determined from the thermal analysis to confirm that the maximum thermal load on the container satisfies the thermal constraint of the bentonite buffer surrounding the container. The temperature at the buffer should be lower than 100°C to keep the physical and chemical properties of bentonite. Four spent PWR fuel assemblies and 297 CANDU fuel bundles are loaded in each container. The heat load from the PWR and CANDU containers are about 1.54 kW and 0.68 kW, respectively. The dimensions of the container were determined from the mechanical structural analysis under the expected mechanical loads in underground repository conditions.[4] The spent fuel received at the encapsulation facility, located on the same site as the underground repository, is transferred by a remote-handling system to the hot cell for the packaging process where the spent fuel is inserted into the disposal containers. In the packaging process each spent fuel assembly or bundle is identified to comply with safeguard requirements. The total number of contain-



Canister Outer shell: Ni-Alloy (or Copper or SUS)
Cast Insert (Fuel region): Carbon steel
Capacity: 4 PWR spent fuel assemblies
Total No. of the Required Containers: 11375
Residual Heat in Canister: 1540 Watt
Total Volume: 5.64 m^3
Surface Area: 19.67 m^2
Total Weight: 39,112 kg

- Fuel wt.: 2660 kg
- Cast insert: 27,345 kg
- Outer shell (Ni-Alloy): 9107 kg
  (Copper): 9211 kg

Canister Outer shell: Ni-Alloy (or Copper or SUS)
Cast Insert (Fuel region): Carbon steel
Capacity: 297 bundles (33 tubes x 9 stacks)
Total No. of the Required Containers: 2836
Residual Heat in Canister: 760 Watt
Total Volume: 5.64 m^3
Surface Area: 19.67 m^2
Total Weight: 39,099 kg

- Fuel wt.: 7425 kg
- Cast insert : 22,567 kg
- Outer-shell (Ni Alloy): 9107 kg
  (Copper): 9211 kg

FIGURE 3 Schematic diagram of the reference disposal container for spent PWR (a) and CANDU (b) fuels.

ers to dispose of the 36,000 tHM of spent fuel is 11,375 containers for PWR fuel and 2529 containers for CANDU fuel. The detailed encapsulation process will be designed further regarding the technical feasibility of the fabrication and the cost aspects.

Bentonite is under consideration as the buffer material because of its low permeability, high sorption capacity, self-sealing characteristics, and durability. The need for, extent, and required performance of seals in the underground facilities and access shafts and ramps for the repository must be developed as a result of performance assessments of the system. The disposal concept being considered at present includes borehole emplacement in the floors and subsequent backfilling of the emplacement tunnels. The base-case repository may include backfilling, particularly of the emplacement tunnels, immediately after waste emplacement or 50- to 100-year monitored retrieval operations. Because the backfilling is intended to provide additional support for the closure of underground openings, the backfill timing and method are important parameters in terms of disposal safety, cost, and political issues, for example, retrieval for reuse or change of safety constraints. The backfill composition will be also determined through more detailed performance assessments and engineering tradeoff studies.

Operation, Decommissioning, and Closure Concepts

The surface-waste-handling facilities are designed to receive spent fuel from the transportation system, unload the spent fuel into a storage facility, repackage the spent fuel into disposal containers, and transport the disposal canisters to the entrance of the repository for emplacement. The base-case design allows for two separate process lines: one for CANDU fuel and one for PWR fuel. The CANDU and PWR waste will arrive at the Spent Fuel Receiving and Packaging Building (SFRPB) in transportation casks. Although both waste types will be handled and packaged at the SFRPB, because of the great variation in size, radiation levels, and packaging processes of the two waste forms, CANDU and PWR fuel will be handled and packaged using different equipment. The CANDU and PWR fuel will share storage facilities.

For transfer to the repository a disposal container of about 40 tons will be loaded into a shielded flask in the packaging plant and transferred underground using the waste-handling shaft. At the underground facility the disposal container is handled in a shielded flask for radiation protection during the transportation and emplacement processes. Prior to the receipt of the container, the compacted bentonite buffer with a density of 1.8 g/cm^3 is placed in the deposition hole. This compacted buffer is in the form of ready-made blocks to provide the hole with a diameter of 124 cm. After container emplacement, the gap between the container and buffer is filled with bentonite powder. Once the container has been lowered from the shielded flask into the prepared borehole, the empty flask is returned to the surface. Additional buffer material is placed over the container up to the emplacement tunnel floor. Although all emplacement work in an emplacement tunnel is completed, the emplacement tunnel may be kept open without backfill during the operation period of 50 to 100 years for monitored retrieval operation. After the designed monitored-retrieval operation, all access and deposition tunnels will be backfilled with a mixture of crushed rock and bentonite and an especially designed concrete bulkhead is constructed at the tunnel entrance to complete sealing for repository closure.

The monitored retrieval operation program will monitor for unexpected radioactivity and provide warning for the underground and surface facilities. These monitoring systems should sample the air at locations throughout the repository for radionuclides and provide an alarm to allow for control measures to be initiated. Control of inadvertent radionuclide release is, under normal operating conditions, achieved by the use and activation of the high-efficiency particulate air (HEPA) filtration system located at the surface facilities of the upcast emplacement shaft. Decommissioning and closure time and processes of the base-case repository will be studied further with cost and safety analyses

ACKNOWLEDGMENTS

This project has been carried out under the Nuclear Research and Development Program of the Ministry of Science and Technology of Korea.

NOTES

1. J. W. Choi, J. E. Kuh, and C. H. Kang. Technology assessment of the repository alternatives to establish a reference HLW disposal concept. Journal of the Korean Nuclear Society 31(6) (1999).
2. C. H. Kang, et al. High-Level Radwaste Disposal Technology Development—Geological Disposal System Development, KAERI/RR-2013/99. South Korea: Korea Atomic Energy Research Institute, 2000.
3. J. W. Choi, W. I. Koh, and C. H. Kang. Reference spent fuel and its characteristics for a deep geological repository concept development. Journal of the Korean Nuclear Society 31(6) (1999).
4. Y. Kwon, S. Kang, J. W. Choi, and C. H. Kang. Structural analysis for the determination of design variables of spent nuclear fuel disposal canister. KSME International Journal 13(3)(2001): 327–338.

The Use of Sodium-Cooled Fast Reactors for Effectively Reprocessing Plutonium and Minor Actinides*

V. I. Matveev, V. A. Yeliseev, and *Ye. V. Poplavskaya*
State Scientific Center Institute of Physics and Power Engineering

The most urgent problem associated with nuclear power industry operations is that of the accumulation of high-level radioactive wastes in the form of fission products and actinides. The latter are defined as plutonium and the so-called minor actinides—neptunium (Np), americium (Am), and curium (Cm). The water-moderated water-cooled power reactor (VVER-1000), for example, produces about 20 metric tons of spent fuel annually, with this material containing approximately 1 percent plutonium (about 200 kilograms) and about 0.1 percent minor actinides (about 20 kilograms). If you consider the fact that the capacity of all the atomic power stations in Russia is on the order of 22 Gwt, then the nuclear power industry produces approximately 500 metric tons of wastes annually, including about 10 metric tons of plutonium and about 0.5 metric tons of minor actinides.

To date, studies have been made at the conceptual level regarding various means of using reactor technology to recycle wastes from the nuclear power industry. These studies primarily focus on fast reactors, although VVER-type thermal reactors, for instance, might also be used for the effective reutilization of plutonium.

The most important problem lies in recycling minor actinides. Plutonium may be used as an efficient nuclear fuel in thermal and fast reactors to produce electricity. It is impossible to use minor actinides (Am, Np, Cm) for these purposes in thermal reactors, and their constant accumulation as a result of nuclear

---

*Translated from the Russian by Kelly Robbins.

power operations creates a significant environmental threat. Minor actinides may be reprocessed as a side function in fast oxide-fueled reactors. However, this method is not sufficiently effective. A more efficient method involves burning minor actinides in specialized cores using fuel that does not contain uranium-238. Including fast reactors with such cores in the nuclear power industry's closed fuel cycle would make it possible to stabilize the accumulation of minor actinides. In such a system all actinides produced by the industry are also recycled by it, and only the portion of these materials that cannot be separated out through chemical reprocessing must be stored as wastes. Organizing such a system, however, requires major capital investments for the creation of a large number of external fuel cycle enterprises. Therefore, the burning of plutonium and minor actinides in the open fuel cycle is of interest.

This report considers various means of using sodium-cooled fast reactors for reprocessing plutonium and minor actinides produced in nuclear power industry operations.

THE CONCEPT OF FAST REACTORS AS ACTINIDE BURNERS

In analyzing the possible use of BN-type fast reactors[1] to recycle wastes from the nuclear power industry, let us highlight two problems: reprocessing of plutonium and reprocessing of minor actinides.

Efficient recycling (burning) of plutonium can be carried out in mixed-oxide-fueled fast reactors with reduced internal breeding due to increased fuel enrichment, but the use of mixed oxide (MOX) fuel for these purposes is not the best option. From the standpoint of secondary plutonium output, it would make sense to consider fuel compositions without uranium-238, as this isotope is the source that produces both plutonium and other very environmentally harmful actinides. Fuel without uranium-238, with this isotope being replaced by some inert matrix, is of great interest in this regard. A number of technological laboratories in Russia, France, and Japan have been working recently to develop such a fuel. For example, a power reactor (fast or thermal) could burn approximately 750–850 kg/GW (el) of plutonium per year without producing any secondary plutonium. Burning plutonium in reactors using fuel without uranium-238 is much more efficient in comparison with using MOX fuel.

Another important problem involves the reprocessing of minor actinides. Plutonium may be used as an efficient fuel in thermal and fast reactors for electricity production purposes. Minor actinides cannot be used in thermal reactors for these purposes, and their continual accumulation as a result of nuclear power industry operations creates a significant environmental threat. Let us note once again that plutonium from spent fuel from thermal reactors contains about 7 percent minor actinides, an amount that increases over time as a result of the decay of plutonium-241 ($T_{1/2}$ = 14.7 years) and its transformation into americium-

241. Because the activity and radioactive danger of minor actinides are substantially (about three times) greater than the analogous characteristics for plutonium, reprocessing only plutonium will not in and of itself resolve the problem of the environmental safety of nuclear wastes. To resolve this problem we might also use fast reactors with fuel not containing uranium-238. It should be noted that the reprocessing of minor actinides could be carried out only as a side function in oxide-fueled fast reactors. Here, in assessing the possible (permissible) amount of minor actinides that could be included in the fuel, it is essential to take into account the nuclear safety of the reactor, one of the main criteria of which is the sodium void reactivity effect (SVRE). For example, assessments made for the BN-800 reactor demonstrate that it would be possible to burn approximately 20 kilograms of minor actinides over the course of a year. At this rate the fast reactor could serve one VVER-1000 reactor. However, to this end it would be necessary to significantly expand the number of fast reactors in the nuclear power sector—up to a level totaling 50 percent of all operating reactors. Meanwhile, about 100 kilograms of minor actinides per year could be burned in specialized cores in BN-800-type fast reactors using fuel without uranium-238, replaced instead with an inert matrix. In the long term it may be possible to burn minor actinides more efficiently in subcritical reactors using accelerators (an accelerator-driven system). In such a system it would be possible to burn up to 250 kilograms of minor actinides per year using an 800-MWt (thermal) fast-neutron subcritical reactor.

Using fuel without uranium-238 in BN-type fast reactors and in advanced subcritical fast reactors opens up real possibilities for eliminating high-level and long-lived wastes from nuclear power production.

USING BN-800-TYPE REACTORS WITH FUEL WITHOUT URANIUM-238 TO BURN ACTINIDES

To date, a rather broad range of studies has already been conducted to analyze and select various fuel compositions without uranium-238, with this isotope being replaced by an inert matrix. Compositions based on zirconium carbide, aluminum nitride, magnesium oxide, and other compounds are being considered. Current research studies have focused on a composition based on magnesium oxide.

The core using fuel without uranium-238 has a number of special characteristics. First of all, excluding uranium-238 ensures that the SVRE will have a negative value, which is an undoubted advantage of such active zones. Meanwhile, there is a noticeable reduction in the magnitude of the Doppler effect, which in traditional cores is determined by uranium-238. The Doppler effect could be partially restored by introducing resonance absorbers into the fuel composition, with the best of these being iron, niobium, and tungsten. Excluding

TABLE 1 Basic Characteristics of a BN-800 Reactor Using Fuel Without Uranium-238

Characteristic	Value
Nominal power, MWt (electrical)	800
Number of fuel rods in active zone	565
First subzone	145
Second subzone	420
Fuel composition	(Pu, MA)O$_2$ MgO
Effective fuel density, g/cm^3	4.5
Actinide content (Pu, MA) in fresh fuel, g/cm^3	1.6
SVRE in core, %Δk/k	~0
Doppler effect in active zone, %Δk/k	–0.235
Rate of reactivity change, %Δk/k/month	3.5
Time between reloadings, in months	3.0
Maximum fuel pin linear power Wt/cm	500

TABLE 2 Quantity of Actinides Produced in Spent Fuel from Thermal Reactors, kg/year/GW (electrical)

Isotope	Uranium Fuel	Uranium-Plutonium Fuel (30%)
All	235	385
Pu^{238}	2.8	6.8
Pu^{239}	121.7	160.1
Pu^{240}	53.2	96.7
Pu^{241}	27.5	56.9
Pu^{242}	12.8	31.8
Np^{237}	7.1	11.2
Am^{241}	6.4	8
Am^{242}	0.006	0.03
Am^{243}	2.6	9.1
Cm^{242}	~0[a]	0.05
Cm^{244}	1	4
Cm^{245}	~0[b]	0.2

[a] Approximately 0, due to decay.
[b] The number is too small to value.

uranium-238 also leads to a substantial increase in burn-up reactivity rates, which in turn leads to a decrease in the interval between reloadings. However, the most painful shortcoming of such cores is the great difference in power of fresh and spent fuel assemblies, which significantly increases the power inequality. In developing a core with fuel without uranium-238 for the BN-800 reactor, the latter of the above-mentioned problems was resolved as follows. The core, consisting of two subzones having different plutonium contents (analogous to the zones of low and high enrichment in traditional reactors), is separated into six concentric subzones (three subzones in each of the subzones with different plutonium contents), and reloading is carried out through the internal subzones by means of shifting fuel subassemblies from the periphery to the center, thus forming an equalized power field. In this process the equalization is optimized with a fuel burn-up of approximately 35 percent. It is possible to increase the fuel burn-up, but this will increase the inequality of the power field and will require the core to have a greater volume. The basic characteristics of the BN-800 reactor using fuel without uranium-238 are presented in Table 1.

These characteristics could be improved through the selection of a core for the specialized reactor. The core option cited was selected on the basis of the existing limitations of the BN-800 reactor, for example, the number of control rods and scram system and the volume of the core.

A BN-800 reactor with the above-described core can effectively fulfill the function of a reactor-burner of plutonium and minor actinides. Here we will consider a simple model for the operation of a nuclear power system consisting of VVER-1000 thermal reactors and BN-800-type fast reactors. In this model the BN-800 reactors use plutonium as well as minor actinides (7 percent), which are extracted from the spent fuel from the VVER reactors and which are mixed with plutonium and minor actinides extracted from the spent fuel from the BN-800 reactor. Table 2 shows the actinide content of the spent fuel from the VVER-type thermal reactors.

The dynamics of changes in fuel cycle characteristics have been studied for

TABLE 3 Established Isotope Composition of Fresh Fuel for Fast Reactors (kg/t)

Isotope	T_{outer} = 1 Year	T_{outer} = 3 Years
Pu^{238}	46.6	53.5
Pu^{239}	249	250.1
Pu^{240}	358.7	358.3
Pu^{241}	80.2	67.9
Pu^{242}	118.6	119
Np^{237}	17.1	17.8
Am^{241}	47.2	62.1
Am^{242}	3	3.5
Am^{243}	46.6	43.7
Cm^{242}	0.6	0.06
Cm^{244}	28.3	20.1
Cm^{245}	5.3	4

two periods of cooling of spent fuel from fast reactors, T_{outer} = 1 year and T_{outer} = 3 years. Calculations have shown that during the process of recirculation, the content of minor actinides in the fuel increases steadily, from approximately 7 percent (initial loading of fuel from the VVER) to 10–13 percent in quasiequilibrium mode. This mode is established after about 25 cycles. The calculated characteristics of the fuel in quasiequilibrium mode are presented in Table 3.

In the nuclear power model under consideration one BN-800-type reactor using fuel without uranium-238 could reprocess the plutonium and minor actinides produced in 1.8 VVER-1000 reactors.

RECYCLING MINOR ACTINIDES IN A BN-800-TYPE REACTOR

The previously discussed study has shown that by repeatedly recirculating plutonium fuel containing 7 percent minor actinides through the core of a BN-800 reactor using fuel without uranium-238, it is possible to burn approximately 36 kilograms of minor actinides per year, thus processing the output of 1.8 VVER-1000 reactors. The amount of minor actinides increases in a stable manner toward a state of equilibrium at the level of about 12 percent.

How much might we increase the minor actinide content in fresh plutonium fuel to be loaded into a BN-800 reactor? The answer to this question depends on the results of studies of many factors, including the radiation characteristics and heat release characteristics of the fresh fuel, which have a substantial impact on the organization of the technological process involved in manufacturing the fuel. Here we note only the most important physical factor, namely, the magnitude of the SVRE. Calculations indicate that for this sort of core design, the permissible level of minor actinide content in fuel is approximately 15 percent if the goal is to ensure a zero SVRE value in the active zone. We shall also note that here the reserve SVRE provided by the sodium cavity is not considered.

Research on the dynamics of the fuel cycle given 15 percent minor actinide content in the fresh fuel indicates that the level of minor actinides in quasiequilibrium status will total approximately 18 percent. Setting up the fuel cycle in such a manner makes it possible to recycle about 1.5 times more minor actinides per year than if the fuel cycle included fuel with 7 percent minor actinide content, meaning that about 55 kilograms of minor actinides would be reprocessed per year.

Increasing the efficiency of minor actinide burning is possible by using fuel based on uranium-235. Calculations indicate that when using this type of fuel, a zero value for SVRE is maintained when adding 35 percent minor actinides. In this case minor actinide burning efficiency is increased by 60 percent over the preceding case and will total about 90 kilograms per year. Thus, the core of a BN-800 reactor using uranium-235-based fuel could serve six or seven VVER-1000 reactors.

It is interesting to note that in quasiequilibrium mode, the proportion of

TABLE 4 Calculated Composition of Unloaded Fuel in Reactors Using Fuel Feed Based on Plutonium and Uranium-235 (Relative Percentages)

	Fuel	
Isotopes	85% Pu, 15% MA	65% U^{235}, 35% MA
U^{234}	0.18	2.90
U^{235}	0[a]	36.96
U^{236}	0[a]	29.99
Np^{237}	7.23	10.11
Pu^{238}	4.65	8.62
Pu^{239}	41.57	1.67
Pu^{240}	24.09	2.35
Pu^{241}	7.41	0.26
Pu^{242}	5.48	0.81
Am^{241}	6.39	2.88
Am^{242}	0.19	0.15
Am^{243}	2.13	1.94
Cm^{243}	0.03	0.01
Cm^{244}	0.58	1.00
Cm^{245}	0.05	0.15
Cm^{246}	~0[b]	0.07

[a] This isotope was absent in this fuel composition.
[b] The number is too small to value.

minor actinides is reduced to approximately 18 percent. Table 4 presents the results of calculations by composition of loaded fuel for reactors using two types of fuel, one containing 85 percent plutonium and 15 percent minor actinides, the other containing 65 percent uranium-235 and 35 percent minor actinides. It may be noted that the plutonium composition that arises when the uranium-235 fuel is used includes a large amount of plutonium-238.

FAST REACTORS AS BURNERS OPERATING IN THE OPEN FUEL CYCLE

In developing cores for fast burner reactors it is essential to select fuel compositions that would ensure the high stability of the spent fuel during its subsequent burial for storage. A number of Japanese researchers have developed a proposal for using highly stable practically insoluble rock-like fuel (ROX fuel) in thermal reactors for the efficient reprocessing of plutonium. It would seem expedient to use this sort of fuel in actinide-burning fast reactors as well.

In developing ROX-fueled cores for operation in the open fuel cycle, one of the main criteria of efficiency is the achievement of maximum possible fuel burn-up, but maximum burn-up is limited by a number of factors. There is a purely physical limitation associated with the fact that increasing the fuel burn-up requires increasing the load of fissile materials in the fresh fuel. The properties of ROX fuel are maintained when the content of fissile materials in the matrix is no more than 40 percent, that is, maximum loading by fission materials should not exceed 2.4 grams per cubic centimeter (initial density of ROX-fuel is approximately 6.0 grams per cubic centimeter). Another limitation associated with the operating capacity of the engineering materials (fuel subassembly and fuel rod casings) occurs during fast neutron irradiation: a permissible neutron flux on the order of 200 displacement per atom (dpa) may be taken as a preliminary norm.

The Institute for Physics and Power Engineering has conducted studies on the efficiency of burning plutonium and minor actinides in a fast BN-800-type reactor fully loaded with ROX fuel. The following ROX-fuel composition was used in the study:

$$(PuO_2+ZrO_2+Y_2O_3)+MgAl_2O_4 \text{ and } (PuO_2+ThO_2)+MgAl_2O_4$$

Various possible ratios between the fuel and the inert matrix were considered. Special parametric studies showed that the greatest burning in the active zone could be achieved by using fuel based on pure plutonium dioxide (without a matrix).

Power in this core is equalized by repositioning the fuel subassemblies (SA) during reloading.

As a result it is possible to achieve an average along the height of SA burn-up of approximately 83 percent of unloaded power plutonium fuel in the core. The maximum burn-up in the central plane totals about 88 percent. The same assessment for the burning of weapons-grade plutonium shows an average burn-up per fuel rod of about 88 percent and a maximum of 92 percent. The burn-up rate for the main isotope, plutonium-239, is 97 percent. These are the maximum burn-up levels possible in principle due to the physics of fast reactors.

The option for the core based on pure plutonium dioxide cannot be technically feasible first due to the very high irradiation integral for the engineered materials (more than 1000 dpa) and second because its fuel must be pure plutonium dioxide, not a ROX composition. Therefore, the following aspects were taken into account during further development work on a ROX-fueled core:

1. The requirements of deep and fast fuel burn-up are mutually exclusive. Basic parameters for the core should be selected on the basis of the conditions of technical feasibility for the core.

2. The maximum permissible limit for radiation damage to engineered materials is defined as 200 dpa. This condition in fact clearly defines the permissible fuel burn-up, including the fuel-matrix ratio and the thickness of this composition.

3. The fuel composition must have ROX properties, that is, it must be suitable for direct burial in geologic formations. The maximum plutonium content with which the composition maintains its rock-like (ROX) properties is 40–45 percent (by weight). This is an additional condition that determines the selection of a possible fuel composition.

Studies on the selection of a composition that would fulfill the above-listed restrictions have shown that the composition must have a density (for plutonium dioxide) of 2.61 grams per cubic centimeter and consist of 42 percent PuO_2 and 58 percent $MgAl_2O_4$. The density of the fuel composition itself will be about 6.22 grams per cubic centimeter.

For comparison purposes Table 5 presents the basic characteristics of three different types of active zones. The first is the so-called designed option, all of the parameters of which are included in full in the original design for the BN-800 reactor.[2] The second is the maximum option, in which the maximum fuel burn-up is achieved, and the third is the optimal or technically feasible option.

One can see that the spectrum of characteristic changes is very broad. In the maximum option the neutron spectrum is very hard (since there is no uranium-238, which is a moderator of neutrons); therefore, this option features a large positive SVRE value due to fission of threshold isotopes, and the Doppler effect is very low (due to the very low proportion of intermediate neutrons, which determine this effect). In the initial option the inert matrix substantially mitigates the spectrum, and a significant proportion of intermediate neutrons appears, as a result of which the SVRE goes from a positive to a negative value and the Doppler effect increases by almost two orders of magnitude.

In order to increase fuel burn-up further the idea arose to use radial leakage neutrons. To accomplish this a post burn zone of burned SA is placed around the

TABLE 5 Calculated Range of Changes of Basic Characteristics of a Core Burner

Option	Designed	Maximum	Optimal
Fuel density (for plutonium dioxide), g/cm^3	1.05	8.6	2.61
Average (maximum) fuel burn-up, % heavy atoms (h.a.)	30 (36)	83 (88)	50 (58)
Ratio of power in fresh/spent fuel	1.97	8.4	2.8
Radial power inequality, K_r	1.78	3.24	2.21
Vertical power inequality, K_z	1.28	1.25	1.14
Volume power inequality r, K_v	2.28	3.73	2.52
Proportion of power in the core/post burn zone, %	—[a]	93/7	93/7
Rate of reactivity drop (over 30 days), %Δk/k	4.27	0.77	1.77
SVRE of core, %Δk/k	–1.6	+1.9	+0.07
Doppler effect = $\Delta k/k/\ln(T\text{-}1500/T_0\text{-}1500)$, %Δk/k	–0.0112	–0.00048	–0.0068

[a] Post burn zone is absent in this core option.

TABLE 6 Calculated Values of Average Burn-up Depth of Unloaded Fuel, Percent Total Activity (Numerator: From Core; Denominator: From Post Burn Zone)

Option	Designed	Maximum	Optimal
Post burn zone without moderator	$\frac{29.1}{38.8}$	$\frac{83.3}{89.1}$	$\frac{47.4}{59.2}$
Post burn zone with moderator (zirconium hydride)	$\frac{29.0}{45.9}$	$\frac{83.2}{84.5}$	$\frac{48.5}{61.4}$

core (the volume of the former was set as equal to the volume of the core itself, and the additional burning time of the fuel in it equaled the time of core lifetime). Inasmuch as this zone has a weak effect on reactor criticality, any radical changes in the neutron spectrum, for example, making a fast reactor into a thermal one by adding a moderator to the periphery zone, are possible without damaging the core. Table 6 provides burn-up data for the three previously selected options, taking into account the additional burning of fuel on the periphery.

The optimal option, which meets all technical limitations, produces a burn-up in unloaded fuel of about 55–60 percent h.a., with burn efficiency in the periphery zone, in this case being somewhat higher than in the maximum option, as the moderator somewhat increases this efficiency rate.

STUDY OF THE POSSIBILITY OF DEEP BURNING OF MINOR ACTINIDES IN THE CORE OF A BN-800 REACTOR NOT FULLY LOADED WITH FUEL WITHOUT URANIUM-238

This study of the burning of minor actinides used a calculated model of an expanded core in a BN-800 reactor (702 fuel assemblies in the core and 30 control and scram system rods). The fuel subassemblies in this model were loaded with common MOX fuel, but part of the fuel subassemblies were replaced with burn subassemblies containing only minor actinides (AmO_2 and NpO_2) in an inert matrix (spinel Al_2MgO_4) or in a moderator (zirconium hydride). At this stage the placement of these materials was considered homogeneous. The quantity of plutonium or minor actinides in the burn subassemblies varied across a very wide range—from 100 percent (about 25 kilograms per subassembly) to 0.1 percent (25 grams per subassembly). These subassemblies were placed in regular order throughout the entire core. The total quantity of these burn subassemblies in different core options varied from 84 to 137. This theoretical research produced the following correlations between the burn-up for plutonium and minor actinides and the composition of the burn subassemblies over time as presented in Table 7 and Figure 1.

As shown in Table 7 and Figure 1, increasing the loading of minor actinides in the fuel elements of the fuel subassemblies being burned leads to a small reduction in the fuel burn-up. This is associated with a certain reduction in minor actinide cross-sections as a result of the increase in resonance blocking.

The introduction of a strong moderator into the fuel subassemblies leads to a sharp increase in the burn-up of minor actinides (up to approximately 95 percent h.a. if minor actinide [MA] content is low). This is because of the fact that in the presence of a strong moderator the neutron spectrum becomes close to thermal, in which the cross-sections of nuclide interaction increases by hundreds of times.

This research allows us to make the following conclusions:

1. It is possible to achieve deep burn-up rates for minor actinides (more than 90 percent h.a.) in a relatively short period of time (that is, in one or two fuel lifetimes), but only with a strong moderator and comparatively small quantities of loaded minor actinides (about 2–3 kilograms of minor actinides per fuel SA).
2. The efficient burning of minor actinides in a strong moderator is accompanied by noticeable flashes of power at the borders of neighboring fuel subassemblies. In order to suppress this phenomenon one might use, for example, a thin (0.2–0.5 millimeter) layer of cadmium to cover the fuel subassembly wrapping.

TABLE 7 Calculated Values for Burn-up (Percentage) for Americium and Neptunium Depending on Content and Use of a Matrix or Moderator over 800 Days (Figures in Parentheses over 400 Days)

Quantity of MA in Fuel Composition, Percentage	Mass of MA in Fuel Rods, kg	Burn-up, Percentage, h.a.			
		Americium + Matrix	Americium + Moderator	Neptunium + Matrix	Neptunium + Moderator
0.1	0.025	25.2 (11.0)	98.6 (88.3)	24.1 (10.5)	99.4 (94.3)
0.5	0.125	24.7 (10.8)	98.4 (86.1)	23.3 (10.2)	99.4 (93.9)
1	0.25	24.3 (10.7)	98.2 (84.3)	23.1 (10.1)	99.3 (93.1)
5	1.25	23.1 (10.1)	92.8 (62.3)	22.2 (9.8)	96.9 (78.2)
10	2.5	22.1 (9.7)	79.1 (44)	21.5 (9.4)	91.1 (58.1)
20	5	20.8 (9.1)	56.8 (28)	20 (9)	65.3 (32.1)
30	7.5	19.8	42.8	19.2	43.9
50	12.5	18.7	29.3	18.3	25.6
70	17.5	17.9	22.3	17.7	20.1
80	20	17.6	20.1	16.8	18.8
90	22.5	17.4	18.1	17.4	17.9
100	25	17	16.9	17.3	17.1

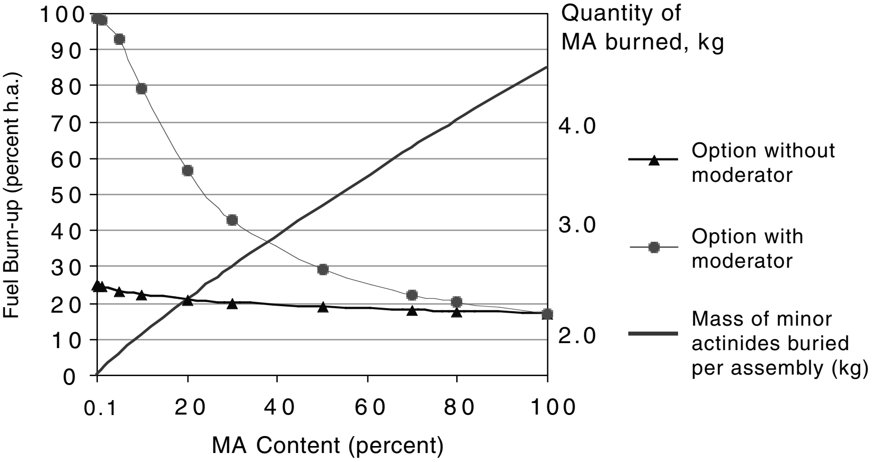


FIGURE 1 Dependence of fuel burn-up and mass of burned minor actinides (per burn subassembly for matrix option) on content of minor actinides in matrix (moderator) (time of irradiation taken as equal to two reactor lifetimes or 2 × 400 days).

3. The duration of burning the above-mentioned materials in a moderator could be either 400 or 800 days, and in the latter case the level of radiation damage to the casings of the burn subassemblies will be somewhat greater than 90 dpa. This is fully permissible if existing EP-450-type steels are used.

Preliminary research has shown that for destroying minor actinides, a core not fully loaded with fuel based on an inert matrix is highly efficient. This sort of core makes it possible in real time (no more than three years) to achieve a minor actinide burn-up rate of more than 90 percent by total activity. This burning is carried out in special burn subassemblies loaded with elements containing zirconium hydride as well as other elements containing minor actinides in an inert matrix. When 84 such burn subassemblies are installed in the expanded core of a BN-800 reactor using oxide fuel, it is possible to burn about 65 kilograms of americium or neptunium per year. This amount could be increased by approximately 50 percent (up to 100 kilograms) if the number of burn subassemblies is increased to about 137. This would make it possible to recycle the minor actinides produced in about five VVER-1000 reactors.

CONCLUSION

The results cited in this paper illustrate the great usefulness of continuing and developing scientific research and design efforts in the area of fast sodium reactors for use as actinide burners, including materials science and engineering research efforts focusing on fuel compositions without uranium-238, with that isotope being replaced by an inert matrix. Inasmuch as the selection of and justification for the use of this or that fuel composition depend substantially on the design of the fast burner reactor and its fuel cycle (for example, an open fuel cycle with subsequent burial of spent fuel, a closed fuel cycle with reprocessing of spent fuel, and so forth), technological research must be conducted along with conceptual research on the selection of fast burner reactors and their fuel cycles as well as on the scientific justification for this selection.

REFERENCES

Akie, H., T. Muromura, H. Takano, and S. Matsuura. 1994. A new fuel material for once-through weapons plutonium burning. Nuclear Technology 107:189–182.

Byburin, G. G., I. Yu. Krivitski, A. P. Ivanov, V. I. Matveev, and E. V. Matveeva. 1995. Plutonium burning in fast reactor cores using unconventional fuel without U-238. Unconventional options for plutonium disposition. IAEA, November 7–11, 1994, Obninsk, Russian Federation. IAEA-TECDOC-840, pp. 219–228.

Krivitski, I. Yu., M. F. Vorotyntsev, L. V. Korobeinikova, and V. K. Pyshin. 1998. Evaluation of self-protection of fast reactor core with uranium free core. Transactions of ENC ’98 Congress, Nice, France, October 25–28, 1998. 4:100–104.

Krivitski, I. Yu. 1999. Actinide and fission product burning in fast reactors with a moderator. Proceedings of the International Conference on Future Nuclear Systems, GLOBAL ’99, United States, August 29–September 3, 1999. Jackson Hole, Wyoming.

Matveev, V. I., A. P. Ivanov, and E. V. Efimenko. 1992. Use of fast reactors for actinide transmutation. A Concept of Specialized Fast Reactor for Minor Actinide Burning. IAEA, September 22–24, Obninsk, Russian Federation.

Matveev, V. I., I. Yu. Krivitski, and A. G. Tsikunov. 1996. Concept of fast reactors—plutonium burners and their fuel cycle. Proceedings of the International Conference on the Physics of Reactors, PHYSOR ’96, Mito, Japan, September 16–20, 1996. 3:p. 21.

Matveev, V. I., I. Yu. Krivitski, V. A. Elissev, and A. L. Kochetkov. 1999. The role of fast reactors in utilization of long-lived nuclear wastes. Proceedings of the International Conference on Future Nuclear Systems, GLOBAL ’99, United States, August 29–September 3, 1999. Jackson Hole, Wyoming.

Matveev, V. I., A. G. Tsikunov, and I. Yu. Krivitski. 1999. Nuclear power systems using fast reactors to reduce long-lived wastes. Pp.179–199 in Safety Issues Associated with Plutonium Involvement in the Nuclear Fuel Cycle, eds. T. A. Parish et al. Dordrecht: Kluwer.

Muromura, T., et al. 1995. Unconventional options for plutonium disposition IAEA-TECDOC-840, pp. 253–262.

Poplavsky, V. M., V. I. Matveev, V. A. Rogov, et al. 2001. Physical and engineering problems of increasing the safety and efficiency of actinide burning in a cutting-edge fast reactor. Monograph, Obninsk, Russian Federation. [in Russian]

NOTES

1. A liquid metal fast neutron reactor.

2. This option was developed in 1998, and its use led to the achievement of a burnup of about 30 percent.

SITE SELECTION FOR SPENT FUEL STORAGE AND DISPOSAL OF HIGH-LEVEL WASTE

Site Selection for Spent Fuel Storage and Disposal of High-Level Waste: Experience of European Countries

Charles McCombie
Arius

This workshop looks at methods for the long-term management of radioactive waste from the nuclear fuel cycle. In principle there are various potential approaches to long-term waste management; different options have been looked at over the past years, for example, BNWL 1974. Figure 1 illustrates the various options looked at, including the so-called exotic options such as disposal into space, ice sheets, or subduction zones. In practice, however, there are only two realistic options today. One of these is long-term storage on or under the surface. The other is deep disposal in a geologic medium. In this talk I will look briefly at

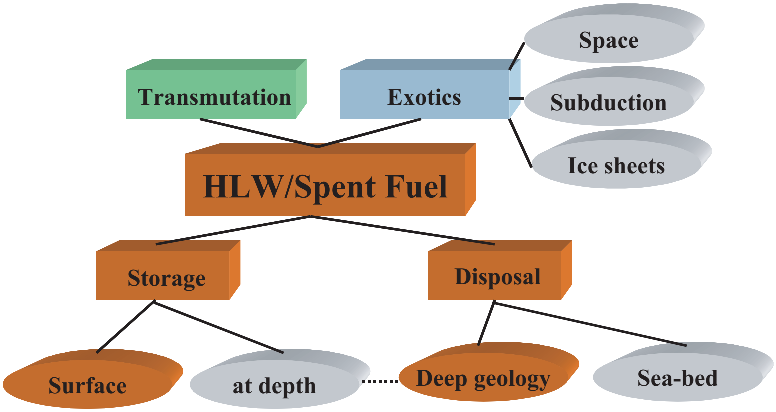


FIGURE 1 Long-term waste management.

long-term storage, addressing the concept and its status today in Europe and the world. Most of the presentation, however, is devoted to the more challenging issue of geological disposal. I will look at the concept of geological disposal in general and then at specific siting issues. I will focus on the contentious issues that have made siting a problem throughout the world, and I will give some examples in a European context—both positive and negative examples. Last of all, the paper looks at the way ahead to siting in Europe in a national context and in an international framework.

LONG-TERM STORAGE: CONCEPT AND STATUS

Long-term storage is technically straightforward. Different methods have been developed and have been tried and tested at various locations for many years and even for decades. The original method used was pool storage, that is, storage under water, normally at reactor facilities. This is still in operation today at many places around the world. Later the technique of dry storage was introduced. Dry storage can take place either in individual storage containers, as is done in various places in Europe, including the Gorleben and Ahaus facilities in Germany and the Zwilag Central Storage Facility in Switzerland. Dry storage can also take place in vaults. This is done at the reprocessing plants in Europe (in France and the United Kingdom) and also in some individual countries, such as at the Borssele storage facility in the Netherlands. Spent fuel storage is a proven methodology; it is safe as long as effort continues to be expended for the necessary maintenance and control. This is the essential difference between storage and the disposal concepts to which we will come later.

Spent fuel or high-level waste storage is needed in all national programs. It is needed because geological repositories will not be implemented in any countries in the very near future and in some cases will be implemented only in the far future. Spent fuel storage needs are in some cases urgent. When early nuclear power plants were built, they were constructed with spent fuel storage pools dimensioned under the assumption that the fuel could be moved off site, either to reprocessing facilities or to centralized storage facilities or to disposal facilities. The problems in implementing these further steps in the back-end chain have led to the spent fuel storage capacity being exhausted or nearly exhausted at various reactor plants. Implementing new centralized storage facilities has proven to be difficult because of the societal problems in establishing new nuclear sites, problems to which we will return later.

There have been, however, some successes in this area. Sweden and Finland have both implemented pool storage facilities below the surface in granitic rocks, close to operating power plants. In Belgium, the Netherlands, and Switzerland it has been possible to implement new surface storage facilities, in each case based on the dry storage technique. In other countries it has proven more difficult, or as yet impossible, to site new centralized storage facilities. This problem has been alleviated in many cases by re-racking of existing fuel storage pools in order to increase the quantities of spent nuclear fuel that can be stored in these. In Europe, Germany currently represents a special case with respect to spent fuel storage policy. Germany has available spent fuel storage facilities both at Gorleben and at Ahaus. However, massive demonstrations against transport (only to the Gorleben facility) have led the antinuclear government to seek reasons for stopping the transports. Asserting that transport is too risky, the government introduced a policy of expanding storage facilities at existing reactor sites. This is a clear example of technical reasons being given to justify political decisions. There are no safety reasons or technical reasons that prevent the safe transport and storage of spent nuclear fuel.

GEOLOGIC DISPOSAL: CONCEPT AND STATUS

Before moving into the details of this section it is worth drawing your attention to a valuable reference document produced recently by Lawrence Berkeley National Laboratory in the United States. The document, “Geological Challenges in Radioactive Waste Isolation; Third Worldwide Review,” (Witherspoon and Bodvarsson 2001) contains a very instructive summary of the status of geological disposal programs in a large number of countries around the world. Geological disposal is widely recognized as the only feasible long-term solution that does not involve continuing care and maintenance by future generations. Accordingly, geologic disposal is part of the government policy or even of the legal framework in many countries. The countries closest to implementation of deep geologic repositories are the United States, where a license application is being prepared for the Yucca Mountain site, and Finland and Sweden, which are at or very close to the choice of their final disposal site. Other countries working toward geological disposal are numerous and include Switzerland, Japan, Belgium, the Czech Republic, Hungary, Spain, South Korea, and countries of the former Soviet Union. The progress being made in these countries varies very dramatically, however. In a few countries geological disposal is being questioned once again. These include Canada and the United Kingdom; in both these countries all options, including the so-called exotic options, are once again being studied and put before the public. In France three potential options (continuing storage, transmutation, and geological disposal) are all being examined in parallel, at least until the milestone deadline of 2006.

In the countries that are progressing with geologic disposal, there are extensive scientific and technical programs that have been running for decades. In almost all cases, however, there have been major delays in implementation of the geological disposal plans, when measured against the ambitious timescales set out by the technical community in the early days. The reasons for the delays have been both technical and societal; the consensus is that nowadays the greatest challenges are indeed societal, as stated in a recent report by the U.S. National Research Council (NRC 2001). Because of this, geological disposal programs in many countries around the world have currently a very strong focus on stakeholder issues. There is much debate on when, how, and to what extent the public can and should be involved in the decision-making processes. This stakeholder issue is, of course, of greatest impact when we come to the challenge of siting geological disposal facilities, as discussed in the following section.

SITING OF GEOLOGICAL DISPOSAL FACILITIES

The siting of nuclear facilities in general has evolved over the many decades since nuclear technology was first introduced. Originally nuclear facilities were sited in remote areas. This was in part related to the military origins of nuclear technology but in part also related to the understandable wish to keep major industrial facilities far from centers of population. As societal problems with siting grew there was a tendency to try to collocate new facilities with existing facilities. Today it is still noticeable that acceptance of new nuclear facilities tends to be higher in areas where nuclear facilities have already been operating for some time. When looking for new sites, the original approach was to use expert opinion. This has been characterized as “decide, announce, defend” (DAD), an approach that has fallen increasingly into disrepute. The next phase, documented in many advisory documents of the IAEA, advocated progressively narrowing in from a nationwide search, using predominantly technical criteria to identify the “best” existing site or sites. The approach was intended to be traceable and to be defensible, that is, the finally chosen site should be easy to defend against any objections, including those of the local population. This technocratic approach was doomed to failure, primarily because the criteria for selection tend to be not purely objective but rather to have a high subjective component, which implies that controversy can never be ruled out. The tendency then has been to move to a more pragmatic approach, based on multiattribute analysis. In this approach an attempt is made to quantify as far as possible all significant characteristics of the site and of the siting approach, and then to allow different stakeholders to apply their personal weighted views on the importance of these characteristics. A final siting approach that has become increasingly important in recent years is volunteering by a potential host community. This has been tried in various countries, often with very sobering results, but occasionally has led to real success. We will return to this issue later.

Guidelines and Regulations in Siting

There are international guidelines on approaches to siting and also on the characteristics that should be sought in specific sites. The IAEA has identified four stages in the siting process.

- a conceptual and planning stage
- an area survey stage
- a site characterization stage
- a site confirmation stage

At each of these stages a set of siting guidelines must be kept in mind. These guidelines include criteria that are useful for judging the acceptability of a site in a societal and, importantly, in a safety sense. Typically the criteria fall into three categories: technical, environmental, and socioeconomic aspects. Box 1 gives examples of the technical and societal guidelines that have been used to date. As indicated above, siting is not a simple case of measuring all the characteristics

BOX 1
Examples of IAEA Siting Guidelines

Technical

- The geological setting should be amenable to characterization, should have geometrical, geomechanical, geochemical, and hydrogeological characteristics that inhibit radionuclide transport and allow safe repository construction, operation, and closure.
- The host rock and repository containment system should not be adversely affected by future dynamic processes such as climate change, neotectonics, seismicity, volcanism, and diapirism.
- The hydrogeological environment should tend to restrict groundwater flow and support waste isolation.
- The physicochemical and geochemical characteristics should limit radionuclide releases to the environment.
- Surface and underground characteristics should allow optimized infrastructure design in accordance with mining rules.
- The site should be located such that waste transport to it does not give rise to unacceptable radiation or environmental impacts.

Societal

- Potential future human activities should be considered in siting and the likelihood that such activities could adversely affect the isolation capability should be minimized.
- Site choice should mean that the local environmental quality will not be adversely affected, or such effects should be mitigated to an acceptable degree.
- Land use and ownership in the area of the site should be considered in connection with possible future development and regional planning.
- The overall societal impact of developing a repository at the chosen site should be acceptable, with beneficial effects being enhanced and negative effects minimized.

given and choosing the best site. This is neither feasible nor necessary. In practice there are a range of issues in siting that have been in the past very contentious and that are today still the subject of intensive debate. The next section looks at the more important of these contentious siting issues.

KEY CONTENTIOUS ISSUES IN SITING

Technical Selection or Volunteering?

Technical siting approaches, involving a progressive narrowing in from a large number to a smaller number of sites using quantifiable criteria, were developed 20 to 30 years ago. As indicated above, however, volunteering of sites can also be a responsible method for siting. Any community that considers that it would be prepared to host a geological waste repository should feel able to put forward this suggestion. The community should also be confident that the site would ultimately be chosen only if it could be shown to fulfill the necessary safety criteria. It is the responsibility of the repository implementer to ensure that all sites considered must fulfill the same strict criteria, irrespective of how they entered into the selection process. If this is assured, it is possible to run a technical screening process for sites and a volunteering process in parallel. In all siting approaches a high-quality scientific and societal process, which must also be transparent, is required if the necessary degree of confidence in the final choice is to be shared by all the stakeholders in the process.

Number of Sites at Each Stage

The number of sites to be considered at each stage in the siting process is a key issue, not least because of the resources that are required to evaluate each potential site. Multiple sites increase the chances of having at least one success. Multiple sites allow choices, that is, they give flexibility to the program and prevent unexpected results at any site necessarily leading to a major realignment of effort. Exploration of sites, especially if this involves investigation of the deep geology, is an expensive undertaking, and much judgment is needed in deciding the number of sites that should be included at each stage throughout the siting process.

Approaches have differed in countries around Europe. Concepts vary for the most expensive stage involving underground exploration using a deep shaft. In countries like Finland, Switzerland, Belgium, and Spain the objective has always been to sink a shaft at only one site, unless the results at the first site chosen prove that further site searching is necessary. In Sweden and France the original plans were to have a deep shaft for investigations in at least two potential sites. Sweden has since moved away from this concept, partly because there has already been extensive exploration at two underground laboratory sites. In France the policy still involves underground exploration at two sites, one in clay and one in crystalline. A crystalline site has not yet been identified, whereas work at the clay site at Bure in northern France is significantly advanced. In Germany, again an exception in the European framework, two sites have already been investigated at depth. The Gorleben salt dome in Lower Saxony has been the subject of a major site exploration program that has cost more than 2 billion Deutsche marks. The Konrad iron mine has been extensively investigated for potential disposal of non-heat-producing wastes. Mainly for societal reasons, the government in Germany charged an advisory group (AkEnd) with the development of a new siting approach, intended to reintroduce a larger number of alternative sites (AkEnd 2002). Whether the complex procedures proposed by the AkEnd group (now disbanded) will ever be put into practice is still an open question.

Predefinition of Explicit “No-Go” Criteria

“No-go” or exclusion criteria are commonly set at the initial screening phase of a program. Typically, a technical no-go criterion might be the distance from a known major geological feature. A typical societal exclusion criterion might be the avoidance of national parks. As the site investigation process proceeds there is sometimes a demand, in particular from the public or from regulators, for further quantitative exclusion criteria. The wish is often for specific criteria based on single parameters, such as the permeability of the rock or the degree of fracturing. Such criteria should not be applied in isolation, however. The safety of a site never depends on a single parameter, but on the total repository system. The repository implementer should resist calls for exclusion criteria based on some single simple parameter and should try to illustrate transparently to the public how the overall safety case for a geological repository is made.

Public Participation

Public *participation* is an important feature of geological disposal today. Earlier, public *information* was thought by many to be sufficient, but today the public in many countries wants also to be part of the decision-making process. The degree of public participation in site selection varies greatly from country to country. Sometimes, especially in the past, it has been a closed process done without any public input. Today there is increasingly an effort being made to inform the public and to seek input from the public on the siting process. A particular point that has arisen in many countries concerns the compensation of a siting community. Originally there was some reticence in many countries to offer compensation, the feeling being that this would be looked on as a form of bribery to compensate for a community accepting a dangerous facility. Increasingly it is recognized that this is not the case. Any community that accepts a facility that will serve the common good of a larger public is entitled to be compensated for providing this wider service. In various countries direct compensation of the host community is now foreseen. In some countries compensation begins during the site selection stage, that is, all potential siting areas will receive some form of compensation (Richardson 1998).

A further critical issue in dealing with the public in the context of siting of geological repositories is that subjective opinions and objective facts must both be factored into the equation. As mentioned earlier, a purely technocratic or objective approach is a myth that has not been able to be realized in any country. In all programs the importance of listening to the subjective opinions and the anxieties of the involved stakeholders has been increasingly recognized.

The last point to be made with respect to public participation is that this is a duty not only of the repository implementer but also of the regulator. In most countries the regulator tries to and is perceived as working on behalf of the public. It is, therefore, important that the regulator also have an explicit program for interaction with all stakeholders, including the public.

Responsibility for Selection of a Preferred Site

The responsibility for site selection usually lies with the repository implementer. The roles of the regulatory authorities are in specifying the selection process and in judging the acceptability of the proposed site. This is the institutional framework most commonly applied. In some cases, however, the government itself, by policy or through its regulatory authorities, has become directly involved in a choice of site. This confuses the roles of implementer and regulator to a large extent and has not proven to be the most productive approach. On the other hand it is also not an optimal approach to exclude the future regulator of the repository site from all early steps leading to the choice of that site. For example, in the United Kingdom the regulators (the Environmental Agency and the Nuclear Installations Inspectorate) were not involved in the early stages leading to the selection of the Sellafield site as a potential deep geological repository. This was counterproductive for the hearings that finally took place, and contributed to the loss of that potential site. It is prudent for the implementer to involve official bodies in the selection process even if it is not legally required. It is to be recommended generally that contacts be established and maintained between implementer and regulator throughout the process of implementing a geological repository, although this must, of course, be done without compromising the independence of the regulator.

"Best" Site or "Sufficiently Safe" Site?

It has already been noted that identifying a "safest" site is neither feasible nor necessary. The tools of safety assessment that are used to judge the safety of the site do not normally have high enough resolution to distinguish between suitable sites. The tools are also not very precise, which implies that sites will be designated as suitable only if large safety margins can be shown to exist. Although a "safest" site is not feasible, a "best" site can be chosen. This is done, using a multiparameter analysis, by choosing a site that best accords to the requirements and wishes of a wide range of stakeholders. Of course the weighting of the selection criteria will be subjective and therefore open to debate subsequently, but there is no method that is more likely to lead to sites that can achieve the necessary degree of acceptance from all participating stakeholders. In summary the brief consensus on this issue is that

- a site must be sufficiently safe to satisfy all safety criteria in a demonstrable manner
- the optimization of site selection must, however, consider criteria other than those directly related to safety
- in this way it is possible to justify the chosen site as being the best technical and societal option from a limited number of chosen potential sites

Figure 2 illustrates the stepwise answering of questions that should lead to decisions on the acceptability of a site. The process starts with deciding whether

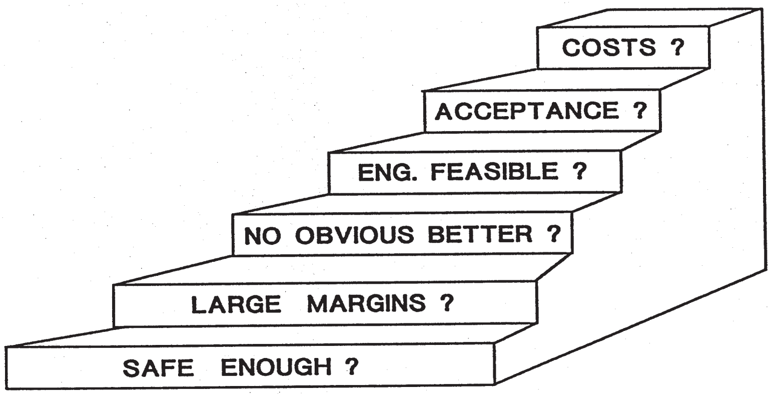


FIGURE 2 Key questions relating to disposal options.

the potential site is safe enough, that is, does it meet or is it likely to meet the safety criteria specified by the regulator? The second question asks whether there are large margins of safety. As mentioned above, the tools of safety assessment are blunt; large margins of safety give more confidence that subsequent increases in knowledge will not lead to the site's dropping out. The third question that one can ask is whether there is an obviously better alternative. Although the given site might meet all the requirements, it will be difficult to convince a large range of stakeholders that this should be the chosen site if there is an easily identifiable better alternative. The fourth question is whether it is feasible to engineer a repository at this site. It may seem late to be asking this question, but unless the safety criteria look as if they can be fulfilled, there is no sense in asking the question. In practice, engineers today are capable of implementing engineered underground facilities in even very difficult rock formations, so that only very extreme conditions will lead to failure to surmount this hurdle.

The fifth question that must be asked concerns acceptance. Even if the safety aspects are satisfied and even if the engineering looks feasible, we still must achieve a sufficient degree of acceptance. Another point of view is that one might **first of all** clarify the acceptance issue, assuming that local community support will eventually be necessary. This purely voluntary approach to siting is currently being tried by the Japanese waste management agency, NUMO. The top step, the last question, is how much will it cost. Putting this question last does not imply that costs have a low priority. It implies that unless all the other steps have been passed, it is not really relevant even to discuss the cost issue.

Stepwise Siting and the Regulatory Process

The stepwise procedure is recommended not just in the siting process but also throughout the entire repository implementation process. This has been increasingly recognized in recent years, and the U.S. National Research Council recently produced a comprehensive publication on the subject (NRC 2003). *One Step at a Time* makes clear that there are both technical and societal reasons for proceeding with development in a stepwise or staged process. This staged concept has several key characteristics.

- A reference-staged process is defined at the outset, but it is not assumed that adaptations will occur only if forced by circumstances.
- The stages are planned with the objective of gaining further knowledge that might lead to amendments subsequently.
- At the decision points between stages a broad and open participation in the decision process is designed to take place.
- To the maximum extent possible the steps are designed to be reversible, in case the chosen direction turns out to be false.

These features are not new, of course, but together they enhance the prospects of progress in controversial first-of-a-kind projects, such as the implementation of a deep geological repository.

EUROPEAN SITING DEVELOPMENTS

Current Status

The most advanced European nation in siting is Finland. In this country the government, the parliament, and the local population have agreed to the choice of a preferred site for a spent fuel repository, at Olkiluoto on the Baltic coast. In neighboring Sweden the disposal program is almost as far advanced. In Sweden two local communities have accepted that site investigations for a spent fuel repository can take place in their community. It is interesting to note that both sites are adjacent to existing nuclear facilities. France has selected one of the two potential sites to be investigated for a deep geological repository. This is in clay; an alternative site in granite has not yet been identified. In Switzerland a geological repository site for low- and intermediate-level waste was recently refused, not for technical reasons but for political reasons. For a deep disposal site for high-level waste and spent fuel, the preferred region in northern Switzerland has recently been identified. This is a region between Zurich and the German border, where the preferred host rock is an overconsolidated clay, opalinus clay. The German situation has already been described. Despite having two sites where exploration at depth has already taken place, consideration is being given to restarting the siting process. In the Czech Republic the siting process was broken off following public opposition some years ago, but this is also to be restarted in the near future. In countries such as Belgium, Spain, Holland, the United Kingdom, and several Central and Eastern European countries there are no active siting programs for deep geological repositories.

A recent development, which has caused great interest in Europe, is the issue by the European Commission (EC) of a draft directive on waste management that includes specific points concerning the implementation of repositories. With respect to repositories for spent fuel and high-level waste it was proposed that authorization for development of sites should occur no later than 2008 and that the authorization for operation of these facilities should occur no later than 2013. These dates were hopelessly optimistic and can be fulfilled by no European program. Even the most advanced program in Finland will not be able to meet these deadlines. A further aspect of the directive is of great interest to many of the smaller countries in Europe. In the memorandum accompanying the directive it is stated that "a regional approach involving two or more countries could also offer advantages, especially to countries that have no or limited nuclear programs, insofar as it would provide a safe and less costly solution for all parties." After intensive debate in the EU, the dates were removed and the directive watered down to a non-binding resolution, which does however still support the concept of shared repositories.

Following up this suggestion from Brussels, the Arius Organisation in Switzerland, together with the Decom Company in Slovakia, submitted a proposal to Framework Program 6 of the European Union. This proposal called SAPIERR (Support Action: Pilot Initiative for European Regional Repositories) has the support of a wide range of Western and Central European countries. The European Commission decided to support the project, and funding is being provided by the EC and the Swiss government.

THE WAY AHEAD

How will programs for development of geological repositories in Europe progress over the next years? It seems clear that national repositories will be implemented in the foreseeable future. The progress that has been made in the Scandinavian countries has already been pointed out, and France after its 2006 deadline may well also have a specific schedule for implementation. These national programs may show the way ahead for programs to be carried out in a wider context. The regional repository concept must be followed in Europe, given the large number of small countries that will need access to deep geological disposal. In fact, regional repositories will almost certainly be implemented at various places throughout the world. Obvious further candidates are Southeast Asia and Central and South America. It may be the case that in the future, international repositories able to dispose the wastes from anywhere in the world will be implemented in regions specifically chosen because of their extremely favorable properties. The same scientists currently involved in the Arius association for promoting regional and international disposal worked earlier in the Pangaea Project, which developed the so-called high-isolation concept for geological disposal. High-isolation sites have various factors contributing to long-term safety (Black and Chapman 2001). These are

- highly stable geology in tectonics
- extensive flat topography
- arid climate, stable for more than 100,000 years
- no fast pathways from the deep repository to the surface; horizontal sediments favorable
- absence of economic resources or major aquifers
- low-permeability rock with good construction properties
- saline, stratified deep groundwaters with chemically reducing conditions

Regions that fulfill all these characteristics have been identified in various parts of the world, in Australia, southern Africa, Argentina, China, Mongolia, and Russia. Whether any of these particularly suitable sites will be considered for the implementation of international repositories is still an open question.

We come now to the overall conclusions of this talk with respect to the siting of deep geological disposal facilities within Europe.

- Geologic disposal is the only feasible option for long-term management of spent nuclear or high-level radioactive wastes that does not depend on continuing care and maintenance.
- Europe is at the forefront of the field with respect to siting. This is illustrated by the excellent progress being made in Finland and Sweden.
- European countries are in the middle of the field with respect to siting. This is best illustrated by France, where at least one site has been chosen as a potential deep disposal site, the necessary technical work is underway, and societal consent including the local population has been achieved.
- Europe is at the back of the field with respect to siting. This is illustrated to some extent by Spain, where political opposition to the process has led the government agency simply to postpone all siting efforts for 10 years. In the United Kingdom the government has gone even further back. Following the catastrophic failure of the Sellafield program, the United Kingdom has officially decided that the geological option is only one of several alternative options, all of which must be re-examined. Worldwide, only the Canadian program, also following a major programmatic failure, has taken this radical step backwards.
- Europe needs to move ahead with repository implementation, both national and regional. This has been recognized by the European Commission, which has issued its 2002 Directive on Nuclear Waste (EC 2002).
- The whole world needs to move ahead in disposal and needs to include both national and shared international solutions in the approaches being considered (IAEA 1998).
- It is important in the scope of this workshop to discuss how Russia can contribute to meet the above objectives. Russia, with its diverse nuclear activities, certainly needs a geological disposal option to take care of its own wastes safely. Russia is one of the few countries that have expressed interest in helping other countries to take care of their waste. This initiative has also received support from the director general of the IAEA (ElBaradei 2004). In fact, Russia has a history of re-importing spent fuel delivered to various countries and of planning for further treatment of this fuel and disposal of the wastes within the Russian Federation. The numerous political, legal, and technical issues that affect the potential for Russia to become the host to a shared disposal facility should all be addressed in the coming years. This is a matter of urgency for Russia, for numerous other countries in Western, Central and Eastern Europe, and also for other countries in the world, in particular the United States.

REFERENCES

AkEnd. 2002. Site Selection Procedure for Repository Sites: Recommendations of the AkEnd Committee on a Site Selection Procedure for Repository Sites. www.akend.de

BNWL (Battelle Northwest Laboratory). 1974. High-Level Radioactive Waste Management Alternatives, 4 BNWL-1900. Richland, Wash.: Pacific Northwest Laboratories.

Black, J. H. and N. A. Chapman. 2001. Siting a High-Isolation Radioactive Waste Repository: Technical Approach to Identification of Potentially Suitable Regions Worldwide. P. 60 in Pangea Technical Report PTR-01-01. Baden, Switzerland: Pangea.

EC (European Commission). 2002. Draft proposal for a Council Directive (Euratom) on the management of spent nuclear fuel and radioactive waste. Brussels: Commission of the European Communities.

ElBaradei, M. 2004. Nuclear Non-Proliferation: Global Security in a Rapidly Changing World. Presentation at the Carnegie International Non-Proliferation Conference, June 30, 2004.

IAEA (International Atomic Energy Agency). 1998. Technical, Institutional and Economic Factors Important for Developing a Multinational Radioactive Waste Repository. IAEA-TECDOC-1021. Vienna: IAEA.

NRC (National Research Council). 2001. Disposition of High-Level Waste and Spent Nuclear Fuel: The Continuing Societal and Technical Challenges. Washington D.C.: National Academy Press.

NRC. 2003. One Step at a Time: The Staged Development of Geologic Repositories for High-Level Radioactive Waste. Washington, D.C.: The National Academies Press.

Richardson, P. J. 1998. A Review of Benefits Offered to Volunteer Communities for Siting Nuclear Waste Facilities. Stockholm: Swedish National Co-ordinator for Nuclear Waste Disposal.

Stefula, V. and C. McCombie. 2004. SAPIERR Paves The Way Towards European Regional Repository. Presentation at the 5th International Conference on Nuclear Options in Countries with Small and Medium Electricity Grids, Dubrovnik, Croatia, May 16–20, 2004.

Witherspoon, P. A. and G. S. Bodvarsson. 2001. Geological Challenges in Radioactive Waste Isolation, Third Worldwide Review. Berkeley: Ernest Orlando Lawrence Berkeley National Laboratory.

The Private Fuel Limited Liability Company National Spent Fuel Site

John D. Parkyn
Dairyland Power Cooperative

OVERVIEW

The issue of retention of unreprocessed spent nuclear fuel became a serious concern in the United States after President Jimmy Carter signed an executive order in the late 1970s that forced U.S. reprocessing companies to cease operation. The order caused utilities to stockpile 40,000 metric tons of uranium (MTU) of spent fuel with the inventory of uranium, plutonium, and thorium intact. No formal national provision was made to deal with this issue during the Carter administration (1976–1980). The subsequent president, Ronald Reagan, nullified the executive order, but U.S. industry was not interested in pursuing reprocessing due to the uncertainty of future political actions and the availability of cheap uranium feedstock. The High Level Nuclear Waste Act of 1982 committed the U.S. government to building, at utility ratepayers' expense, an underground repository for spent nuclear fuel, scheduled to open in 1998. As it became apparent that this date would not be met and the temporary site authorized for the Department of Energy would not be built, eight U.S. utilities applied for a license in 1997 to build a site on the Goshute Indian Reservation in Utah. A lease was signed with the Goshutes for such a site. A license decision is expected in 2004.

The Private Fuel Storage Company (PFS) is responsible for the central storage and transportation of spent nuclear fuel. The respective owners are responsible for the packaging of their expended fuel in conformed Nuclear Regulatory Commission certified multiple purpose canisters, useable for both storage and transportation.

POLICIES USED TO SHAPE THE PRIVATE FUEL STORAGE COMPANY

The first meetings involved almost all reactor operators and vendors in the nuclear industry in the United Sates. A full year was spent discussing options and using this field of talent to develop certain fixed practices that would guide PFS regardless of which entities finally formed the company. It was necessary to consider all options to ensure that this large number of resources was heard before ideas were dismissed. PFS was formed as a limited liability company in 1995 by eight of the nation's utilities. These utilities owned about 20 nuclear-power-producing reactors. They included both for-profit companies and a co-operative.

The initial actions were to decide major principles upon which the effort to centrally store spent fuel on an interim basis would be developed. They included site selection criteria, type of storage, cask orientation, and methods of shipment.

Site Selection Criteria

This process was built on a failed endeavor by the U.S. government called the Voluntary Host Program. The program would have rewarded communities for being willing to host a temporary surface repository. Congress cancelled it for political reasons. The concept was to do a widespread search of possible sites and consider the positives and negatives in each individual case in light of the U.S. National Environmental Policy Act, which requires a thorough review of all aspects of any major activity. This includes a consideration of alternatives for siting and for the action itself such as leaving spent fuel scattered at 70-plus locations throughout the United States. This evaluation, completed before the events of September 11, 2001, created a need for additional protection of hazardous substances. The alternate-sites review initially looked at over 40 initial potential hosts. Consideration was given to the total transportation mileage to the ultimate repository, the population density of the area, the ability of the governmental entity to represent its population, the proximity to groundwater, and the proximity to surface water. The availability of mainline rail transportation was also a major consideration.

The number of potential sites was reduced by a series of reviews, visits, and analysis until four final candidates remained. An engineering evaluation was performed to determine the best site, and a location on the reservation of the Skull Valley Band of the Goshute people in Utah was selected. The sites were all voluntary, and owners or governments were actively involved at each location. Once the site was selected, the negotiation process was started with the host location and included fixed and variable payments and employment preference for those fully qualified. The members of the host community voted on the project. The lease was signed and submitted to the Bureau of Indian Affairs of the U.S. government for ratification. This process took several months; collectively the negotiating process took a year. Educational updates to the Band members were accompanied by workshops for those wishing to get additional formal education for qualifying for site jobs.

Specific Site Principles

The two significant commitments to the final site host were that no fuel would be handled at the site (unless a canister failed) and that no radiological contamination was permissible. A decision for no bare fuel handling is a significant improvement in risk potential to a locality. A reactor's site staff is the group ultimately best qualified to handle their particular type of fuel assemblies, which are then permanently sealed in a multiple purpose canister (MPC). The handling of older fuel assemblies away from the original reactor site introduces a factor that complicates the selection of a voluntary host. The site is designed to facilitate dry transfer into a replacement canister should the process ever be required. This prevents the need to return fluke to its originating reactor site.

The canisters are to be passively air cooled, and therefore any external contamination could become airborne at the site. This is an unacceptable condition. The service agreements with customers were written to require complete decontamination of any radioactive materials from the outside of the MPC before its installation into the shipping cask. On arrival at the PFS site the entire outside of the MPC is monitored for contamination, any located is cleaned immediately, and the customer is billed for the cost. This provides both the incentive and the method to ensure a site free of contamination. Ultimate decommissioning is easy and site closure complete in a relatively short time. The total amount of contaminated material from total site removal will be little and the site will be returned to its owners, the Goshute, in excellent condition for any use they choose.

Type of Storage

It was determined that storage would be accomplished by the use of MPCs, which would allow a single design of a right circular cylinder shape to hold all fuel types in uniquely fabricated baskets. The canisters would be large enough to hold 68 boiling-water reactor fuel assemblies and 32 pressurized water reactor assemblies. The canisters would be seal welded shut at the reactor of origin and never reopened to expose the fuel. The MPC program reduces the risk of handling accidents at both interim storage sites and at the ultimate underground repository, as bare fuel is never directly handled by any staff other than fuel type experts.

The canisters would be moved around the reactor site in a shielding cask. They would then be inserted into a transportation cask and shipped by train to the Goshute site. When removed from the transportation cask, the canister would be placed in a storage cask and remain there for as long as needed. When the underground repository is ready for operation, selected canisters would be put into transportation casks for shipment by rail to the final site. The canisters would then be placed directly in the ground in storage casks specifically designed for an underground environment.

The fuel would remain in the same canisters through all operations and would share the same cask regardless of fuel type, including cladding, intactness, length, enrichment, and burn-up. Large economy of scale would be achieved with minimal shipments and risk to the public. This would ensure a higher level of local government acceptance along the shipping route.

Cask Orientation

The need for uniformity in handling was reviewed among design choices, and a decision was made to store fuel only in vertical casks of the MPC type. This reduces the risks of fuel breakage (fuel in the reactor is operated in the vertical orientation) and increases the use of space at the interim storage facility. It also reduces the need for handling equipment to the type used in vertical storage.

Methods of Shipment

The need to reduce highway shipments and decrease frequency of total shipments dictates a railroad mode only. The railroads are privately owned and can restrict access to rights of way. They can handle much heavier loads than the highway system. A multimodal system was developed to ensure loading at the reactor site into the MPC configuration and the use of heavy haul to a railhead by either truck or barge. This integrated system has the potential to reduce the shipments transversing the United States to a small of fraction of all hazardous material shipments.

Licensing Progress

After a series of meetings among the U.S. utilities in 1994 and 1995, a limited liability company was formed to serve as the operating company throughout the lifetime of the project. The company was funded by the member utilities, and they were not committed to site use as a consequence. The facility is to be open to any domestic utility and use any certified cask design. At that time there were six vendors of spent fuel storage casks in the United States, and two major business decisions were made: (1) All U.S. vendors would be allowed to bid for site acceptance, with PFS paying a significant amount toward its site-specific licensing costs and (2) all U.S. architects and engineers (AEs) would be invited to bid to become the site AE. Technical committees were set up to conduct interviews and included the best staff available in the industry.

The AE firm of Stone & Webster was selected for the site design. The Holtec and Sierra Nuclear firms were selected to provide casks. A site selection and negotiation process was completed to allow a license submittal by June 1997. The staff of the Nuclear Regulatory Commission found the submission complete in July 1997, and the process began.

The final application included two methods of reaching the site: by rail, requiring construction of a common carrier rail line (the Great Salt Lake and Southern Railroad) to bridge the 32-mile gap between existing lines and the site, or by heavy-haul truck with an intermodal transfer point on the mainline railroad.

The Nuclear Regulatory Commission reviewed the license application and gave a positive finding on safety through the final safety evaluation report in 2000. The final environmental impact statement was issued with a positive finding in 2001. The State of Utah objected to locating the facility in its proximity (though the Goshutes are a separate entity with sovereign immunity) and was allowed to intervene in the licensing process. Hearings on safety topics were held in the spring of 2000 and additional hearings on the final two safety topics (earthquakes and aircraft overflights) and environmental topics were held in spring 2002.

The licensing board made no final decisions on the hearing subjects until March through May 2003. All three financial issues were resolved in favor of PFS. Both seismic issues were resolved in favor of PFS. The F16 fighter aircraft overflights were decided against PFS, which now must review the consequences of crash and take any required protective actions. The rail line routing, an environmental contention, has not yet been ruled on.

The final license decision should be available in the summer of 2004.

Post-Licensing

The customers will be contracted and a schedule for construction, operations, and specific shipments will be set. The federal and local governments will be involved in final route selection. Construction will be on a bid basis with quality assurance oversight each step of the way. The fuel cars have been built to the national standard developed by PFS and the American Association of Railroads. Each car will be fabricated to exacting standards, including nondestructive testing of individual welds.

The entire train, including locomotives, will be operated and maintained by PFS. Features such as electropneumatic braking will provide for a rail shipment to achieve very high safety standards. Real-time transmittal to satellite will permit off-train support of security and safety needs on a continuous basis.

Site construction will include the installation of a 32-mile rail line. Once construction is complete, shipments can be coordinated at 200 casks per year from individual utilities to meet their specific needs of timing and size. Outbound shipments to the U.S. Department of Energy at Yucca Mountain or other locations as designated will be facilitated in the reverse order. This will permit selective handling of inbound shipments to the federal repository.

Site Operation

To accommodate the staff time restrictions at the originating sites the site will schedule shipments with specific customers about two years in advance. The first preference goes to owning members of PFS. The exact dates and routes of shipments will generally be held from public disclosure subject to the regulations of the Nuclear Regulatory Commission and Department of Transportation. A complete train, including locomotives, buffer cars, a security car, and one or more fuel cars, will be formed at the customer's reactor site. The train will be operated by fully trained crews in active communication with a central dispatch. Proper notifications will be given when traversing local political boundaries.

The train will arrive at the site, and each car will be unloaded in the canister transfer building. The MPC will be removed from the transportation cask, installed in a storage cask, and moved to a location on a concrete slab. Eight wireless thermocouples will be installed. The cask will be continuously monitored during its multiple-year storage period. When the canister is ready for shipment to an underground repository, the process will be reversed. Canister selection will be at the discretion of the receiving agency. The shipment order will be arranged to accommodate the movement into the underground storage repository.

Site Closure

The decommissioning is totally prefunded and will begin about 20 years before final closure. As the inventory of onsite canisters begins to decrease from its maximum level, the most exposed storage casks will be dismantled and disposed. Manufacturers anticipate no activation levels requiring radiological disposal. Once the final offsite shipments have been made the central structures will be cleared of any radiological contamination (none is anticipated), and the surveys to authorize license determination will be started. These are three site characterizations. Once all radiological materials are removed, the Nuclear Regulatory Commission will terminate the license. The Goshute people have the right to retain any structure and/or the railroad for future economic development. A fully provided nonradiological decommissioning fund will be available to dismantle and remove any structure the resident community designates. The soil and ground cover will be reshaped to the natural levels existing in the area. Even the rail embankments will be removed. The commitment is to achieve a final closure that is totally satisfactory to all parties.

ACKNOWLEDGMENTS

Thanks to John M. Parkyn for presentation assistance in Russia; Cindy Spears, Penny Schreiber, and Sarah Snodgrass for final document oversight; and the National Academies for their organizational role in addressing this critical topic on the environmental and energy future of the world.

Experience of Japan*

Koji Nagano
Central Research Institute of Electric Power Industry (CRIEPI)

After years of nuclear energy production and use, growing pressure from spent nuclear fuel accumulation is receiving serious attention in Japan in order for the nuclear power plants to keep operating without an overflow of built-in storage pools. There are other key constraints with spent nuclear fuel management, such as the schedule of Japan's first spent nuclear fuel reprocessing plant construction and operation.

A series of accidents and scandals in nuclear facilities, such as a sodium leak at the Monju fast reactor, an asphalt explosion at the PNC[1] Tokai reprocessing plant, and the latest criticality accident at the JCO uranium conversion plant, have made the public increasingly suspicious and distrustful of the nuclear power establishment as a whole. This has been further exacerbated by two recent occurrences: the scandal of maintenance information concealment revealed by an employee of the maintenance contractor for Tokyo Electric Power Company (TEPCo) in August 2002, and the invalidation judgment by the Nagoya High Court concerning a construction permit for Monju, a prototype fast breeder reactor, in January 2003. We should clearly recognize such conceptual gaps between the nuclear power establishment and the general public as a crucial constraint that influences nuclear technology development. This paper does not discuss that area, but such reorganization of nuclear governance should be based on open public participation.

These conceptual gaps may cast various uncertainties; on the one hand, plutonium use in light water reactors (LWR) has virtually ceased, which has an

---

*Excerpts from Nagano 2003a and Nagano 2003b reprinted with permission of the Atomic Energy Society of Japan.

indirect implication for spent nuclear fuel reprocessing and related management of its residues. Direct impacts increase difficulties of siting new nuclear facilities, including not only power plants but even nuclear fuel cycle facilities, namely final repositories of high-level radioactive waste (HLW). Both direct and indirect impacts give clear rise to the role and importance of storage of spent nuclear fuel.

In the next section recent efforts at managing spent nuclear fuel in Japan are reviewed. The paper also discusses the implications for the temporal and geographical aspects of the process.

CURRENT STATUS OF SPENT NUCLEAR FUEL MANAGEMENT IN JAPAN

Present Status of Spent Nuclear Fuel Management

Table 1 shows the recent status of spent nuclear fuel accumulation at all the nuclear power stations (NPSs) as of March 2001, reflecting changes 6 months

TABLE 1 Spent Nuclear Fuel Stored at NPSs in Japan (in metric tons of uranium)

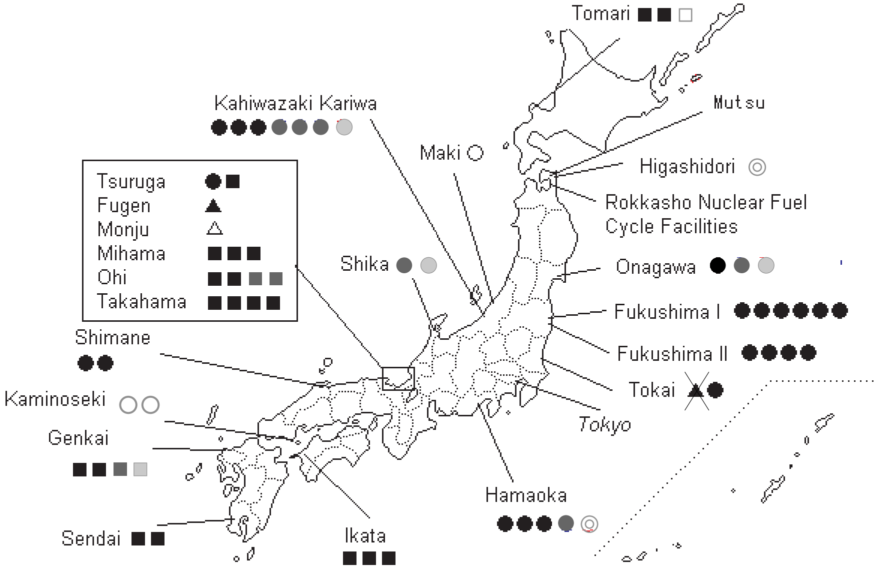
Utility Company	NPS	Loading in Core	Fuel per Batch	SF in Store	Storage Capacity
Hokkaido	TOMARI	100	30	250(+10)	420
Tohoku	Onagawa	160	40	200(+10)	370
Tokyo	Fukushima-1	580	150	1140(+40)	2100
	Fukushima-2	520	140	1280(+30)	1360
	Kashiwazaki-Kariwa	960	250	1470(+100)	1890
Chubu	Hamaoka	420	110	730(+10)	860
Hokuriku	Shika	60	20	50(+20)	100
Kansai	Mihama	160	50	280	300
	Takahama	290	100	850(+50)	1100
	Ohi	360	120	740(+70)	1370(+530)[a]
Chugoku	Shimane	170	40	340(+70)	440
Shikoku	Ikata	170	60	330	980(+450)[b]
Kyushu	Genkai	270	100	420	1060
	Sendai	140	50	580(+10)	900(+200)[c]
JAPCo	Tsuruga	140	40	440(+10)	870
	Tokai-2	130	30	220	260
Total	4630	1330	9290(+380)	14,380(+1190)	

[a] Re-racking of Units 3 and 4.
[b] Re-racking of Unit 3.
[c] Re-racking of Units 1 and 2.

SOURCE: Federation of Electric Power Companies (FEPC) (http://www.fepc.or.jp). Changes in parentheses are from September 2000.

after September 2000. Japan's current nuclear power generation, with a total capacity of 45.9 GWe with 53 reactor units (see Figure 1), discharges about 900 MTU (metric tons of uranium) of spent nuclear fuel per year. This spent nuclear fuel discharge accumulates primarily in the built-in reactor pools at those power reactor units. As spent nuclear fuel accumulation approaches the capacities of those reactor pools, some nuclear power stations are forced to supplement at-reactor (AR) storage capacity in order to avoid an overflow of the reactor pools. At TEPCo's Fukushima Daiichi NPS, a 1120 MTU water pool storage facility was implemented in 1997, as well as an auxiliary dry metal cask storage capability. At Japan Atomic Power Company's Tokai Daini NPS a dry metal cask storage device with a capacity of 260 MTU (24 casks) was constructed. Several other stations have added storage capacity by re-racking storage pools, some of which are found in Table 1.

It is clear that opportunities for enhancing existing AR storage capacity is almost exhausted, which strongly suggests urgent needs for away from reactor (AFR) storage measures. In November 2000 Mutsu City in Aomori Prefecture



Reactor Type	In Operation	Under Construction	Under Construction Preparation
PWR	■	◇	□
BWR	●	◎	○
GCR, FBR	▲	△	

Total Capacity	In Operation		Under Construction		Planned		Total	
	Units	GWe	Units	GWe	Units	GWe	Units	GWe
Sep. 1991	41	32	11	12	3	2	55	36
Dec. 1996	51	43	4	3	2	2	57	48
Jan. 2002	53	45.9	4	4.12	6	7.24	63	57.26

FIGURE 1 Nuclear power stations in Japan as of January 2002, with changes since 1991. (Tokai NPS Unit 1 was shut down on March 31, 1998.)

announced an invitation to TEPCo for site investigation for AFR storage in its territory. TEPCo, responded immediately and initiated a feasibility study. The report was submitted to the city on April 3, 2003. On request from the city a supplementary business plan was submitted on April 10, 2003. Its details will be touched upon in a subsequent section.

Institutional Developments in Spent Nuclear Fuel Management[2]

Spent nuclear fuel storage was first mentioned in the 1987 Long-Term Program for Development and Utilization of Nuclear Energy, which is regularly revised and published by the Atomic Energy Commission of Japan as the fundamental nuclear policy document in Japan. In the 1994 Long-Term Program for Research, Development, and Utilization of Nuclear Energy a special section was added for future methods of spent nuclear fuel storage, as well as ways to manage spent mixed oxide (MOX) fuel.

The Steering Committee for Nuclear Energy under the Council for the Comprehensive Energy Policy for then-Ministry of International Trade and Industry, which is the primary engine to formulate Japan's energy policy, published an interim report on spent nuclear fuel storage in January 1997, which urged preparedness for possible prolongation of spent nuclear fuel storage, and actual deployment of AFR storage in about 2010. The cabinet supported the report in February 1997.

In order to plan steps to realize conceptual views provided in the interim report the Working Group for Spent Nuclear Fuel Storage Measures was formed with representatives from the government and major electric power companies (EPCos). The working group, after a series of intensive discussion during March 1997–March 1998, submitted its final report, revealing the concept of storage of recyclable fuel resources, as well as possible regulatory schemes for storage service providers and the related legal framework. The Steering Committee for Nuclear Energy published another report in June 1998, which was primarily an endorsement of the working group's report, with an emphasis on legal procedure and site selection principles.

One should not overlook the fact that it was emphasized in both of these reports that because spent nuclear fuel storage is a safe and static process, virtually any business venture may be able to enter into the market of storage services, as long as it meets relevant regulatory principles. While this statement was intended partly for better public acceptance, it is still worth paying attention to the fact that such a competitive atmosphere was already anticipated positively by those representatives from the government and the power utility industry.

The law for regulation of nuclear power reactors and other nuclear-related operations (the Regulation Law, hereafter) was amended in June 1999, as a follow-up to the interim report. In this amendment, "operation of storage for recyclable fuel resources" was identified and introduced,[3] which opened up ways for new business ventures to be allowed this service provider operation. Related regulatory schemes, such as safety design criteria of facilities, are under preparation accordingly.

The latest Long-term Program for Research, Development, and Utilization published in December 2000 simply followed up the series of arguments and discussions described above.

Future Prospects of Spent Nuclear Fuel Management

Future prospects of spent nuclear fuel management, namely, the demand for additional storage measures, are influenced largely by the following factors:

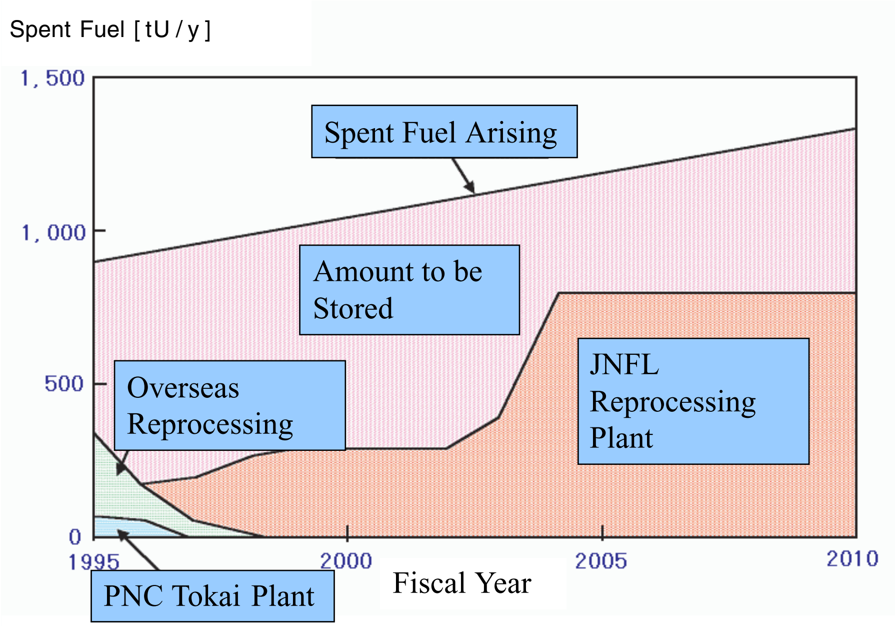
- the JNFL (Japan Nuclear Fuel Limited) reprocessing plant of 800 MTU/year design capacity, currently under construction in Rokkasho-mura of Aomori Prefecture
- one-time full-core discharge upon decommissioning of reactor units foreseen beginning in 2010, with built-in storage pools also dismantled at a certain stage of decommissioning

Even if the Rokkasho reprocessing plant is successfully operated at its design capacity, it cannot take in the whole discharge of Japan's NPSs every year, nor the past discharges. While the government's official views are shown in Figure 2 and Table 2, these underlying assumptions, especially the schedule of the reprocessing plant, are already obsolete, since the JNFL Rokkasho reprocessing has been rescheduled to start its operation in July 2005. This clearly illustrates the importance of repeating projections whenever there are any changes in the above-mentioned factors.

In the long run it is obvious that large-scale storage devices are needed. This is particularly true after 2010, when the first commercial LWR plant reaches the end of its 40-year lifetime. After that, a series of LWR plants will be shut down, which means, on the one hand, a large amount of one-time discharge of spent nuclear fuel, and on the other hand, a loss of the storage capacity of the reactor pools.

Based on the existing amount of spent nuclear fuel stocks, Nagano (2002a,c; 2003a) projected spent nuclear fuel balances in Japan up to the year 2050, using an integrated tool SFTRACE (spent fuel storage, transportation and cost evaluation system). The results are shown in Table 3. As a result the Japanese nuclear industry should prepare a storage capacity at around 10,000 to 15,000 MTU in the medium term, for example, by 2030. Then, in the long-run, up to 2050, the storage needs would differ significantly, from a decrease to none to a continuous increase up to the level of 25,000 MTU.

Special attention should be given to the plutonium utilization in LWR plants. Spent MOX fuel will have to be stored, as the Rokkasho reprocessing plant is



Assumptions:

Fiscal Year	1995	2000	2010
Nuclear Power Generation Capacity [GWe]	4150	4500	7000
Annual Discharge of Spent Fuel	900–1300 tU/y		
PNC Tokai Reprocessing Plant	Receipt of spent fuel ends in 1996.		
JNFL Reprocessing Plant	Receipt of spent fuel is started in 1997. Operation starts at 800 tU/y in 2003.		

FIGURE 2 Prospects of spent nuclear fuel projected increase and management. SOURCE: Ministry of International Trade and Industry (MITI).

TABLE 2 The Official Perspective of Spent Nuclear Fuel Management

Spent Fuel	1997–2010 (tU)	2011–2020 (tU)
Cumulative Projected Increase (a)	15,200	16,000
Shipment to JNFL/Rokkasho Reprocessing Plant (b)	5900	8000
Shipment to Overseas Reprocessors (c)	70	—
AR Storage Capacity (d)	5300	4200
Requirements for AFR Storage (a-b-c-d)	3900	3800
Cumulative AFR Requirements	3900	7700

SOURCE: Agency of Natural Resources and Energy, On the Interim Storage of Spent Nuclear Fuel, 1999.

TABLE 3 Projected Spent Nuclear Fuel Storage Needs in 2050 in Japan (in metric tons of uranium)

Simulation Cases	Spent Low Burn-Up UO_2 Fuel	Spent High Burn-Up UO_2 Fuel	Spent MOX Fuel	Total Amount to Manage (A)
O	0	39,000	10,000	49,000
N	7,000	30,000	11,000	
O+2Rep	0	21,000	13,000	34,000
N+2Rep	0	23,000	11,000	

Assumptions for Projection Cases: **Case O:** No second reprocessing plant; the first reprocessing plant receives older spent fuel as prioritized. **Case N:** No second reprocessing plant; the first reprocessing plant receives newer spent fuel (higher burn-up) as prioritized. **Case O+2Rep:** The second reprocessing plant starts in 2030; no MOX fuel is reprocessed; older spent fuel prioritized. **Case N+2Rep:** The second reprocessing plant starts in 2030; spent MOX fuel and older spent fuel prioritized.

not licensed for the type of spent nuclear fuel with higher generation of heat and radiation. Significant uncertainty has been cast on MOX fuel utilization because of local opposition, which was vividly demonstrated by the negative results of the vote by Kariwa villagers[4] on May 27, 2001. Up to now no MOX fuel has been loaded in any NPS unit in Japan.

As already mentioned, TEPCo announced its plan to install the Recyclable Fuel Reserve Center (Reserve Center) in Mutsu City. According to TEPCo's business plan submitted on April 10, 2003, the facility will have a storage capacity of 5000–6000 MTU separated to two phases, around 3000 MTU each, all using the dry metal cask storage technique. Storage duration is set at a maximum of 50 years. The facility will be constructed and operated by a new private company to be established by TEPCo and other electric utility companies as they volunteer to join, but no utility has responded positively[5] to TEPCo's invitation up to now. The Reserve Center is expected to commence by 2010 and to accept spent nuclear fuel from those utility companies that joined the operating company.

The author draws attention to three points. (1) This facility's capacity will satisfy roughly one-half of the necessity up to 2030, as discussed previously in this section, at about 10,000 MTU. Since there seem to exist initiatives undertaken by other utility companies, another facility of a similar scale will reach Japan's management capability at a flexible enough level until 2030. (2) If it is a multiutility venture, it will be a centralized facility. In this case the issue is local acceptance of the receipt of fuel from other regions or other utility companies in the country, as well as the region's utility. (3) Similar to the second point, this could become an important milestone for HLW repository siting. Except for the Rokkasho Fuel Cycle Center whose siting took place years ago,

Total AR Storage Capacity (B)	Needs for Additional Storage (C = A – B)	Second Reprocessing Plant to Commence in 2020	Range of Needs for Storage
24,000–27,000	22,000–25,000	—	0–25,000
	7,000–10,000	Δ8,000	

The total nuclear power generation capacity is 70 GWe in 2010 and 80 GWe in 2050, respectively. AR capacity is assumed at 300 MTU/GWe, slightly larger than the current average (270 MTU/GWe). The second reprocessing plant is assumed with capacity of 800 THM/year to commence in 2030. UO_2 fuel for reload is of low burn-up (3300 MWd/MTU at average) until 1992, and thereafter of higher burn-up at 45,000 MWd/MTU.

this Mutsu initiative is almost the only example of new central nuclear facility siting. Nuclear Waste Management Organization (NUMO), the entity responsible for HLW disposal is promoting a stepwise siting processes, and now it is in its first stage of inviting local townships to nominate themselves as candidates for preliminary site investigation. Its schedule is to reach a final selection by about 2035. The Mutsu initiative may have two implications to this NUMO process; first, as already mentioned, to allow time for NUMO to exercise its plan, and second, to demonstrate successful experience of facility siting, with full transparency of all relevant information and a sufficient level of public participation.

Costs and Economics of Spent Nuclear Fuel Storage in Japan

Since spent nuclear fuel storage remains in a precommercial stage, we only have a few examples of cost analysis and little is known about possible market prices. Based on IAEA (1994), CRIEPI has conducted cost analyses of different spent nuclear fuel storage techniques by calculating the levelized unit cost of storage. The Net Present Value (NPV) of a project is a measure of the value of a project, defined as a sum of all the discounted cost streams associated with the project, that is,

$$NPV = \sum \frac{C_i}{(1+d)^i} \tag{1}$$

where C_i is the cost or expenditure in the i-th year, d is the discount rate, i is the year index. The levelized unit cost (LUC) of storage is the unit price of storage service that equalizes the NPV of the cash flow of income and the NPV of the expenditure for the whole lifetime of the project, that is,

$$LUC = \sum \frac{C_i}{(1+d)^i} \Big/ \sum \frac{M_i}{(1+d)^i} \tag{2}$$

where M_i is the amount of spent nuclear fuel transported into the storage facility in the *i*-th year. Thus, the formula (2) is based on the assumption that the storage fee is paid upon receipt of spent nuclear fuel at a uniform unit price, which is *LUC*, namely,

$$\sum \frac{LUC \times M_i}{(1+d)^i} = \sum \frac{C_i}{(1+d)^i} \tag{3}$$

Figure 3 shows an example from Saegusa (1998), where a comparison among storage options is presented at 3000 MTU AFR storage under the Japanese circumstances. Although the water pool storage is a mature technology with plenty of experience from existing reactor pools, its economics may suffer from high capital investments as well as high operation and maintenance costs due to requirements for forced circulation and quality control of the cooling water. The metal cask has received the highest priority in implementing storage facilities in the short and medium terms, with its superb modularity and economics compared with the water pool. For a longer perspective, research is ongoing for other

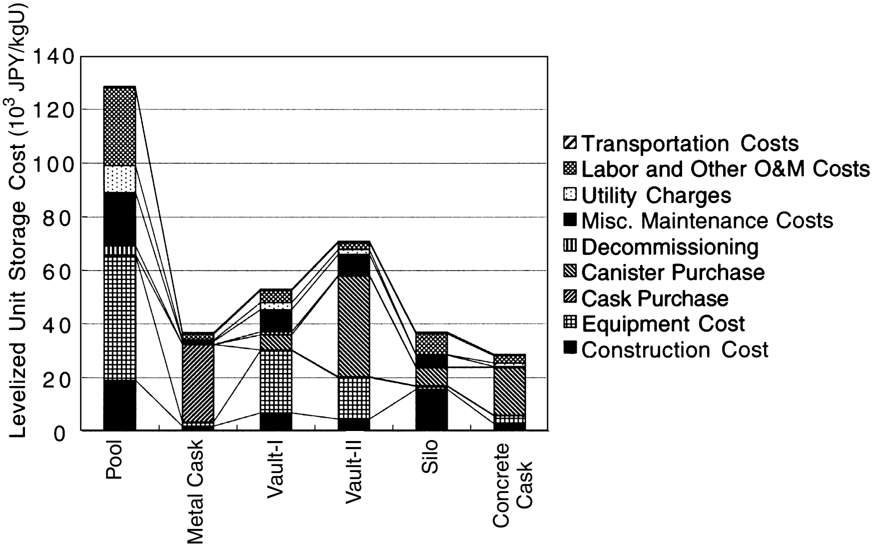


FIGURE 3 Comparison of levelized unit storage costs for AFR storage of 3000 MTU. SOURCE: Saegusa (1998).

dry storage technologies, aiming at better economic performances. Key issues of research include

- long-term integrity of massive concrete structures
- long-term integrity of thin metal canisters
- safety standards in operation and maintenance, especially unloading and loading for transportation

The applications of similar methodology include Yamaji et al. (1987) and Nagano and Yamaji (1989) for comparison of pool and metal cask techniques, and Nagano et al. (1990) for AFR storage, though cost data used in those early studies are now obsolete. The latest example is Ito et al. (2000, 2001), whose main result is shown in Figure 4. Conclusions rather similar to those in Figure 3 are found, though cost data reflect further development and improvement during the period. Figure 5 shows the cost data used for the case of 5000 tU storage capacity in Figure 4, both by lump sum and NPV in the year of the facility's commencement.

If we try to convert these results to cost value in terms of generated power, a simple formula is applied. Take 30,000 JPY/kgU as metal cask storage of 5000 MTU in Figure 4. At the average burn-up of 40 GWd/t, and a thermal efficiency of 35 percent, and neglecting the time difference between power production to storage, we will then obtain 30,000 [JPY/kgU]/336,000 [kWhe/kgU] = 0.09 [JPY/kWhe]. This can be compared to 5.9 JPY/kWhe, a widely cited number for standard nuclear power generation in Japan.

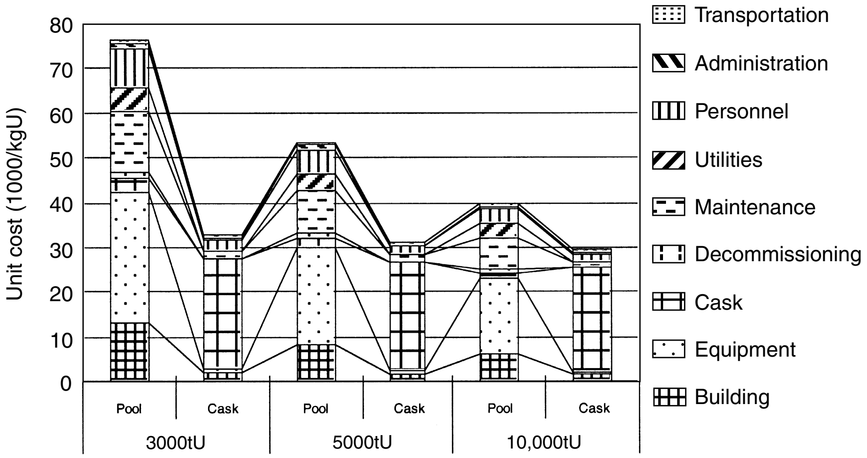


FIGURE 4 Comparison of unit AFR storage costs for a storage capacity of 3000–10,000 MTU. SOURCE: Ito et al. (2000, 2001).

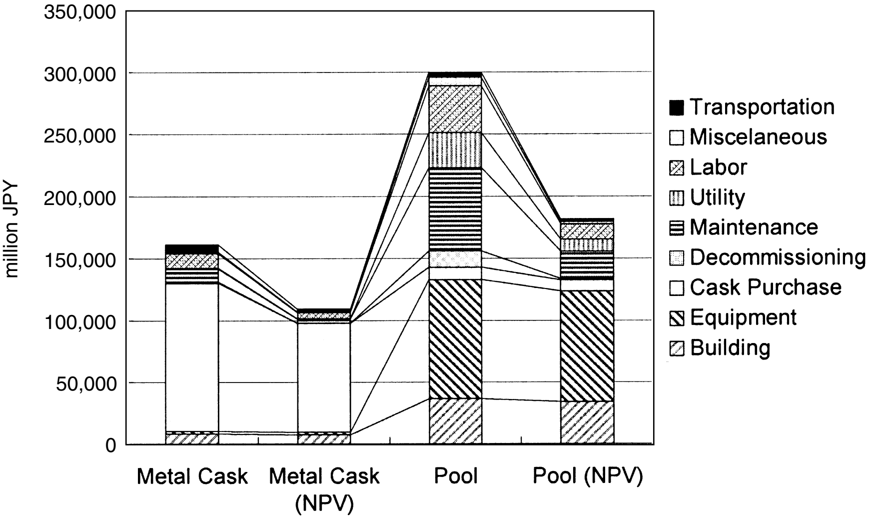


FIGURE 5 Cost data for 5000 tU storage facility. SOURCE: Ito et al. (2000).

CONCLUDING REMARKS

This paper discussed recent developments and the current status of spent nuclear fuel management in Japan. With the increasing pressure of spent nuclear fuel discharge from the power plants in operation and, by contrast, uncertainties in their processing and management services, spent nuclear fuel storage in the short and medium terms has been receiving the highest priority in nuclear policy formulation in Japan. Small-scale interim storage devices, as well as capacity expansion, for example, re-racking, and shared uses of existing devices, are being introduced at a number of power stations. Large-scale AFR storage of recyclable fuel resources is to be realized in the medium and long terms. Commercial operation of the storage of recyclable fuel resources concept may be in the offing, as the amendment to the law for regulation of nuclear power reactors and other nuclear-related activities has passed in the Diet. Most recently TEPCo submitted its Feasibility Study Report (FSR) to the candidate site, Mutsu City, on April 3, 2003, as well as its supplementary business plan on April 10, 2003. TEPCo's Recyclable Fuel Reserve Center is expected to commence operation by the year 2010, and other power utility companies are expected to follow similar directions. In the Mutsu initiative the following three points should be heeded:

1. This facility's capacity will satisfy roughly one-half the requirements up to 2030, as discussed in a previous section, at around 10,000 MTU.

2. If it is realized with a multiutility venture, it will be a centralized facility, and the issue of local acceptance must be carefully dealt with and overcome.

3. This could be an important milestone for HLW repository siting.

REFERENCES

Atomic Energy Commission, Japan. 2000. The Long-Term Program of Research, Development and Utilization of Nuclear Energy. Unofficial English translation version is available at: http://aec.jst.go.jp/jicst/NC/tyoki/siryo/tyoki_e/siryoe.htm

CRIEPI (Central Research Institute of Electric Power Industry). 2001. Demonstration Tests of Recycle Fuel Resources—Development of Concrete Modular Storage Technologies. Final Report for Fiscal 2000 submitted to the Government. [in Japanese]

CRIEPI. 2002. Demonstration Tests of Recycle Fuel Resources—Development of Concrete Modular Storage Technologies. Final Report for Fiscal 2001 submitted to the Government. [in Japanese]

IAEA (International Atomic Energy Agency). 1994. Cost Analysis Methodology of Spent Fuel Storage. Technical Reports Series No. 361. Vienna: IAEA.

Ito, C., T. Saegusa, and K. Nagano. 2000. Economical Evaluation on Spent Fuel Storage Technology away from Reactor. Research Report No. U99047. Tokyo: CRIEPI. [in Japanese]

Ito, C., T. Saegusa, and K. Nagano. 2001. Economical evaluation on spent fuel storage technology away from reactor. Journal of Nuclear Fuel Cycle and Environment 8(1). [in Japanese]

Nagano, K., and K. Yamaji. 1989. The Needs and Economics of Spent Fuel Storage in Japan. Proceedings for ASME/JSME/AESJ Joint International Waste Management Conference (JIWMC), Kyoto, Japan.

Nagano, K., T. Saegusa, C. Ito, et al. 1990. Economic Analysis of Spent Fuel Storage Technology in Japan. IAEA-SR-171/22. In Spent Nuclear Fuel Storage: Safety, Engineering and Environmental Aspects. IAEA Working Material No. IAEA-SR-171. Vienna: IAEA.

Nagano, K. 1998a. A Mathematical Analysis of Storage in the Fuel Cycle Back-End System. Denryoku-keizai Kenkyu (Economics and Public Utilities) 39:1–12. [in Japanese]

Nagano, K. 1998b. An Economic Analysis of Spent Fuel Management and Storage. Proceedings for PBNC-1998 International Conference. Banff, Canada.

Nagano, K. 1999. Interim Storage of Spent Fuel in Japan—Facts and Theoretical Prospects. Paper presented at Workshop on Interim Storage of Spent Nuclear Fuel and Management of Excess Fissile Material, Harvard University, February 18–19, 1999.

Nagano, K. 2002a. A strategic consideration on nuclear fuel cycle with spent nuclear fuel management as its core. Journal of Nuclear Fuel Cycle and Environment 8(2). [in Japanese]

Nagano, K. 2002b. A Systems Analysis of Spent Fuel Management in Japan. Paper presented at PBNC-2002 International Conference, October 21–25, 2002, Shenzhen, China. [To be published]

Nagano, K. 2002c. A Systems Analysis of Spent Nuclear Fuel Management and Storage. Doctoral thesis, Univ. of Tokyo.

Nagano, K. 2003a. Systems analysis of spent fuel management in Japan I. Projection of material balances and storage needs. Journal of Nuclear Science and Technology 40(4):173–181.

Nagano, K. 2003b. Systems analysis of spent fuel management in Japan II. Methodologies for economic analyses of spent fuel storage. Journal of Nuclear Science and Technology 40(4): 182–191.

OECD/NEA (Organisation for Economic Co-operation and Development/Nuclear Energy Agency). 1985. The Economics of Nuclear Fuel Cycle. Paris: OECD.

OECD/NEA. 1989. Plutonium Fuel: An Assessment. Paris: OECD.

OECD/NEA. 1994. The Economics of Nuclear Fuel Cycle. Paris: OECD.

Saegusa, T. 1998. Activities and future issues on research of spent nuclear fuel storage in Japan. Nuclear Eye 44(4):14–19. [in Japanese]

Saegusa, T. et al. 1999. Dry Storage Technologies for High Burnup and MOX Spent Nuclear Fuels. Summary Report No. U27. Tokyo: CRIEPI.

Steering Committee for Nuclear Energy. 1998. Towards Realization of Storage of Recycle Fuel Resources. Interim Report to the Council for Comprehensive Energy Policy, June 1998. [in Japanese]

Yamaji, K., K. Nagano, and T. Saegusa. 1987. Comparative Economic Evaluation of Spent Nuclear Fuel Storage Technologies. Research Report L87001. Tokyo: CRIEPI. [in Japanese]

NOTES

1. The Power Reactor and Nuclear Fuel Development Corporation, currently Japan Nuclear Cycle Development Institute (JNC).

2. For more detail see Nagano (2002c, 2003b).

3. In the Regulation Law those entities licensed for specific “operations” for various activities related to nuclear energy and nuclear materials are regulated, such as power reactor operation, fuel fabrication, material transportation, spent fuel reprocessing, and radioactive waste disposal. This “regulation by operational entity” principle shows clear contrast with “regulation by materials” in the United States, for example, where nuclear materials are put under regulation regardless of who owns or handles them. As there was no specification of “storage of spent fuel” before the amendment, virtually none was allowed to stay as this was a situation of possessing stocks of spent nuclear fuel for the purpose of their storage. The amendment in 1999 meant that such an operation of spent nuclear fuel storage became recognized and put in the normal regulatory formula.

4. Among the electorate, 88.2 percent voted, of which negative votes for MOX fuel loading were 1925, supportive 1553, reserved 131, and invalid 16.

5. Tohoku Electric Power Co., which has Mutsu City in its supply territory and is considered the most desirable partner of the group, has not shown any interest in joining. Japan Atomic Power Co. was said in a newspaper article to have shown interest, but the company denied it.

The Current Status of Spent Nuclear Fuel in Korea

Hyun-Soo Park and *Jongwon Choi*
Korea Atomic Energy Research Institute

Despite the slowdown of the nuclear energy industry in western countries, Korea is steadily promoting the nuclear power generation business in response to Korea's increasing electricity demand, seeking new sites for nuclear power plants, and supporting the development of commercial technology. Nuclear energy has certainly been a great contribution to the energy supply in Korea because of the shortage of natural energy resources. In 2002 nuclear power was the primary energy source, with a 29.2 percent share of the total, followed by bituminous coal (27.4 percent), gas (25.3 percent), and oil (8.7 percent). The only domestic energy resources were anthracite coal and hydropower, which had a share of 2.2 percent and 7.2 percent of the total, respectively. In gross power generation nuclear power produced a share of 38.9 percent over other energy sources, such as coal (38.5 percent), oil (8.2 percent), gas (12.7 percent), and hydro (1.7 percent).

Since the first nuclear power plant, Gori Unit 1, a pressurized water reactor (PWR), was commissioned in Korea in April 1978, the nuclear power generation capacity has grown steadily and remarkably in Korea. By the end of 2002, 18 nuclear power plants with 14 PWRs and 4 CANDU (Canada deuterium uranium) reactors were in operation with a total capacity of 15.7 GWe (see Table 1). In addition, 2 PWRs produced by the Korean Standard Nuclear Plant (KSNP) of 2 GWe are under construction. The construction of an additional 4 PWRs produced by KSNP and 4 evolutionary PWRs (Korean next-generation reactors, 1.4 GWe each) is planned by 2015. A total of 26 nuclear power plants will be in commercial operation by the year 2015, after the potential retirement of the two oldest units (Gori Unit 1 and Wolsung Unit 1). The total installed nuclear power capacity is then expected to reach 26.1 GWe.

TABLE 1 Status of Nuclear Power Plants in Korea as of December 2002

	Reactor Sites			
	Gori	Yonggwang	Uljin	Wolsung
Number in operation	4	6	4	4
Number under construction	—	—	2	—
Reactor type	PWR	PWR	PWR	CANDU

Korea's demand for uranium and nuclear fuel cycle service has continuously increased. The radioactive waste and spent fuel has also been rapidly accumulated, and radioactive waste management is one of the important concerns in the Korean nuclear community. A new radioactive waste management plan was proposed by the Ministry of Commerce, Industry, and Energy in January 1997 and was approved by the Atomic Energy Commission in September 1998. According to the new plan, a low- and intermediate-level radioactive waste repository will be constructed by 2008, and spent fuel will be stored at each nuclear power plant site until interim storage facilities are constructed in 2016.The site-securing program is currently underway.

SPENT FUEL MANAGEMENT

The effective management of spent fuel remains a challenge for the future of the nuclear industry. The current at-reactor (AR) storage capacities of PWRs and CANDUs are 4996 tU and 4807 tU, respectively. The cumulative amount of spent fuel by the year 2002 reached about 2893 tU from the existing PWRs and 3089 tU from the CANDUs. Using the above to generate a long-term projection, it can be estimated that approximately 11,000 tU and 20,000 tU of spent fuel would be accumulated by the years 2010 and 2020, respectively. The accumulated amount of spent fuel and the expected year of losing full core reserve in each power station in Korea are shown in Table 2, as of December 2002.

The policy for spent fuel management in Korea is based on the guidelines provided by the Korea Atomic Energy Commission (AEC), which is the nation's top policy-making body on nuclear energy. The government has not yet established a definite policy on whether to recycle or to permanently dispose of its spent fuel for long-term management. The AEC set a goal for spent fuel interim storage as a mid- and long-term expedient. The government effort to construct a centralized interim storage facility for spent fuel and a repository for radioactive waste packages had come to nothing due to strong dissension about the site acquisition from the local communities. After the Guleop Island Project was dealt a final blow in 1995 with the confirmation of active fault zones near and along the island during the site investigation, attempts to secure a site were started again at the beginning of the new millennium. The Nuclear Environment Technology Institute (NETEC) of Korea Hydro and Nuclear Power Co., Ltd. (KHNP) is an organization dedicated to a new site-securing program under governmental supervision. Site selection involves three basic principles: voluntary subscription of local governments, democratic and open procedures for public acceptance, and financial support programs for the local community. In this regard, continuing efforts for the improvement of safety and reliability, which are the most important objectives in the National Radioactive Waste Management Program in Korea, are needed.

According to the revised spent fuel management plan announced by the government in September 1998, a centralized interim storage facility will be completed by the year 2016. The storage system, dry or wet, will be determined by considering the circumstances of the facility site and the research and development progress at that time. The facility is to be run on the 2000 tU scale in its early stage and will be expanded gradually to a total scale of 20,000 tU, as needed. Spent fuel generated will be stored at the reactor sites until the year 2016. As shown in Table 2, the current storage capacities at reactor sites are insufficient to meet the target year of 2016 for operation of the centralized interim storage facility. Therefore, the expansion of AR storage capacity is being implemented at each site, taking into consideration an appropriate combination of technical and economic factors. For PWRs the AR expansion is currently being carried out by transshipment between neighboring units and re-racking with high-density storage racks, which increase the storage density by using boral or borated stainless steel neutron absorbers, have been installed partially or fully in spent fuel pools. Storage density was increased to about 200 percent by replacing old storage racks with the high-density storage racks. For a while this type of temporary measure will be taken into account to expand the AR storage capacities. In the case of the CANDU reactors, spent fuel bundles, after at least six years of cooling in the spent fuel bay, are put into stainless steel baskets and

TABLE 2 Status of AR Spent Fuel Storage in Korea as of July 2002

Site Location	No. of Units	Storage Capacity (tU)	Cumulative Amount (tU)	Year of Losing Full Core Reserve
Gori	4	1737	1288	2008
Yonggwang	6	1696	895	2008
Uljin	4	1563	710	2007
Wolsung	4	4807	3089	2006
Total		9803	5982	

transferred to the onsite concrete silo-type dry storage facility. A silo can hold nine fuel baskets, and each basket accommodates 60 bundles. Additional dry storage facilities for CANDU spent fuel will be constructed as needed.

RESEARCH AND DEVELOPMENT STATUS ON FUEL CYCLE BACK-END TECHNOLOGY

The Atomic Energy Act of Korea stipulates that the minister of science and technology shall formulate the National Atomic Energy Research and Development Program according to a sector-by-sector implementation plan. Originally the National Mid- and Long-Term Atomic Energy Research and Development Program was launched in June 1992 as a 10-year (1992–2001) program. It was modified into a new research and development program to be implemented for the 1997–2006 term, to take account of the major changes in national and international situations. The program is being carried out and is funded by both the government budget and the atomic energy research and development endowment fund.

The back-end fuel cycle area of the Intermediate and Long-Term Research and Development Program covers the following projects: (1) DUPIC (direct use of spent PWR fuel in CANDU reactors); (2) high-level waste disposal; (3) the advanced spent fuel conditioning process (ACP); and (4) the pyro process for partitioning and accelerator-driven systems (ADS) for transmutation.

DIRECT USE OF SPENT FUEL

Generally, approximately 0.6 percent fissile plutonium and 0.9 percent uranium-235 (U-235) are contained in the spent PWR fuel with a discharge burn-up of 35 MWd/kgU. Hence, spent PWR fuel is potentially an attractive energy resource for recycling in a CANDU reactor. From this context a broad feasibility study was performed from 1991 to 1993 in order to identify any feasibility issues of a fundamental nature for the DUPIC fuel cycle concept.

This concept is based on the idea that the spent PWR fuel material is fabricated directly into CANDU fuel without any intentional separation of fissile materials and fission products. The fuel materials always remain highly radioactive to prevent any diversion of sensitive nuclear material. Thus, by taking advantage of Korea's strategy of using both PWR and CANDU, the DUPIC fuel cycle has been developed with keen interest as an alternative of spent fuel management in a proliferation-resistant way.

In 1992 appropriate agencies of Korea and Canada and the U.S. Department of State agreed to cooperate on a study on dry recycling of spent PWR fuel into CANDU reactors in Korea. Seven dry recycling options were investigated. These options, involving only mechanical and/or thermal processes, can be referred to collectively as DUPIC fuel cycle options. Of these options the OREOX (oxidation and reduction of oxide fuel) option was chosen as the most promising method for the DUPIC fuel cycle, considering technical feasibility, safeguards, and so forth. Based on this feasibility study the experimental verification program has been implemented in tripartite cooperation. In an earlier stage of the program the fundamental works led by Atomic Energy of Canada Limited (AECL) on the DUPIC fuel fabrication process and reactor physics assessment were carried out. Three DUPIC fuel elements were fabricated, and have been irradiated in the NRU research reactor (National Research Universal reactor at AECL's Chalk River Laboratories). The post-irradiation examination for the two elements removed from NRU at 10 MWd/kgHM and 16 MWd/kgHM burn-up have already been performed.

A comprehensive research and development program has been implemented at the Korean Atomic Energy Research Institute (KAERI) to experimentally verify the DUPIC fuel cycle concept in international cooperation with Canada, the United States, and the International Atomic Energy Agency. In Phase II of the Experimental Verification Program, which was carried out in full momentum until 2001, the proliferation-resistant dry process has been developed for direct recycling of spent PWR fuel in CANDU reactors. Following small-scale hot cell experiments at the post-irradiation examination facility (PIEF) to characterize DUPIC powder/pellets using actual spent PWR fuel, a main DUPIC fuel fabrication campaign was started to fabricate the DUPIC fuel pellets and elements for the irradiation test at a research reactor. From the start of this experiment until the year 2002 a total of 10 kg of spent PWR fuel that has undergone a nominal burn-up of 35,500 MWd/tU was fabricated into DUPIC fuel pellets and elements. On the basis of the process conditions developed by the DUPIC powder/pellet characterization study, DUPIC pellets and elements were successfully fabricated in a remote manner. The fabricated DUPIC elements were loaded into the HANARO research reactor for the irradiation test. The major achievements are summarized below.

- Quality DUPIC fuel has been remotely fabricated in hot cells.
- The in-reactor performance of DUPIC fuel has been verified through the irradiation tests in research reactors and post-irradiation examinations.
- The compatibility of DUPIC fuel with a CANDU reactor system has been verified.
- Significant progress has also been accomplished in developing the DUPIC safeguards system in close collaboration with the United States.
- The DUPIC fuel cycle has been found to be economically feasible compared with the once-through cycle.

During Phase III, scheduled to take place from 2002 to 2007, more intensive works on the symbiotic fuel cycles will be carried out in international cooperation between Korea, Canada, the United States, and the IAEA.

High-Level Waste Disposal

KAERI initiated research and development activities on high-level waste disposal technology given that it will be required for long-term consideration of the nuclear energy strategy regardless of fuel cycle options. The current research and development activities are focused on the performance assessment in the long-term postclosure period and a study to set up a Korean reference disposal system.

At this moment four major projects are underway in KAERI: (1) a total system performance assessment; (2) disposal system development; (3) geo-environmental science research: and (4) a radionuclide migration study. The Phase III study (approximately 2002 to 2007) is being carried out and covers the following research items:

- Korea standard reference disposal system
  - –reference engineered barrier system (EBS) development through spent fuel characteristics
  - –cost and sensitivity analysis
  - –strategic national disposal plan and scenario
- performance assessment
  - –multidimensional probabilistic safety assessment (PSA) code development
  - –cyber research and development platform development
  - –flow visualization lab provision
- assessment of deep geological environmental condition
- validation of performance of high-level waste disposal system
  - –engineered barrier systems (EBS) performance experiments in engineer ing scale
  - –radionuclide migration experiments in large rock block
  - –construction and operation of underground research tunnel at KAERI site

Advanced Spent Fuel Conditioning Process

With the volume reduction perspective of the spent fuels to be stored and/or disposed of, the Advanced Spent Fuel Conditioning Process, a so-called lithium reduction process, has been under development in KAERI since 1997. Technical goals of advanced spent fuel conditioning are divided into three major areas: (1) active demonstration of the lab-scale process system; (2) technical and economic verification of the process concept and development of innovative technologies to simplify process systems and to reduce costs (inactive components have been developed since 1997); (3) and the installation of a lab-scale mockup, which was completed in 2000. Tests and modification of mockup facilities are being performed. The process system will be optimized by using mockup facilities by 2003. Nuclear materials control and accounting technology for this process is being developed. More detailed research and development items are being performed as follows:

- optimization of process systems by using mockup facilities
- experimental verification of oxide reduction chemistry
- automation and remotization of all unit operations and material transfers between operations
- development of nuclear materials control and accounting technology
- establishment of continuous uranium metal casting technology
- selection and optimization of dry storage systems

The planned launch date of the Phase III study of this project is 2004, and the study will cover lab-scale verification of process concepts, demonstration of nuclear material accountability and system safeguardability, verification of technical and economic feasibility of the process concepts, and the establishment of a roadmap for further research and development.

Pyroprocess for Partitioning and ADS for Transmutation

Pyrochemical partitioning technology development in Korea was initiated on the basic concepts of adoption of the proliferation process and transmutation of transuranium (TRU) and long-lived fission products (LLFP) in the hybrid power extraction reactor (HYPER) system. Key technologies in this area were defined as follows: preparation of metal halides, electrorefining and electrowinning, reductive extraction by solvent metals, and salt regeneration. On the basis of the fundamental study conducted in Phase I, KAERI's current research and development effort in the Phase II study (2001–2003) is focused on the following items:

- development electrorefining technology to recover uranium and TRU (surrogate material) in LiCl-KCl molten salt
- development of electrowinning and cadmium distillation technologies to reduce uranium content further in TRU/RE(U) metal and TRU (surrogate) from liquid cadmium cathode, respectively
- core optimization in terms of neutronic performance for ADS system
- corrosion testing of structural material (HT-9, 9Cr-2WVTa steels) against lead-bismuth coolant in static conditions
- development of fuel fabrication technology using simulated material (uranium-zirconium)

The research and development of the phase beginning in 2004 will cover the improvement of liquid metal cathodes to reduce rare earth (RE) content further, update the electrowinning process to minimize uranium content in the recovered TRU/RE(U) metal, complete a conceptual core design for ADS, specifically a Pb-Bi corrosion test in dynamic conditions (using KAERI facility), and irradiation experiments for the transmutation characteristics of FP using the HANARO research reactor.

SUMMARY OBSERVATIONS

The Republic of Korea has 14 PWRs and 4 CANDUs with a total capacity of 15.7 GWe currently in commercial operation. An ambitious plan has been established to expand the total installed nuclear capacity up to 26.1 GWe by the year 2015.

The cumulative amount of spent fuel generated by July 2002 in Korea reached about 6000 tU. Long-term projections indicate that approximately 20,000 tU of spent fuel will be accumulated by 2020. An away-from-reactor interim storage facility, which will be run on the initial capacity of 2000 tU scale, is planned to be completed by the year 2016.

The Republic of Korea continues research and development activities in search of an optimal option for spent fuel management, keeping in mind the long-term perspective of nuclear power use, which has been and should be a vital component of a growing economy and environmental protection. According to the National Mid- and Long-term Atomic Energy R&D Program launched in 1992, the back-end fuel cycle area covers DUPIC, high-level waste disposal, advanced spent fuel conditioning process (ACP), pyroprocess for partitioning, and ADS for transmutation.

The basic concept of the DUPIC fuel cycle is to directly fabricate the CANDU fuel from the spent PWR fuel by using thermal/mechanical processes at hot cells without the separation of fission products and transuranic elements. Since 1991 KAERI has successfully fabricated several DUPIC fuel elements remotely in the hot cell, and the performance evaluation through the irradiation tests at the HANARO research reactor is under way.

The main purpose of the HLW disposal technology development program started in 1997 is to establish a Korean standard reference for HLW disposal systems by 2006. The basic concept is to encapsulate the intact spent fuel in the corrosion resistant container; the packaged spent fuels are then to be disposed of in a mined underground facility located at about several hundred meters below surface in a crystalline rock mass.

The concept of the ACP is to convert spent oxide fuel into a metallic form in a high-temperature molten salt in order to reduce the heat power, volume, and radioactivity of the spent fuel. The main objective of the ACP is to treat PWR spent fuel for long-term storage and eventual disposal in a proliferation resistant and cost effective way. Moreover, the electrolytic reduction method of the ACP can contribute to the innovative nuclear energy system as a key technology for the preparation method of the metallic fuel.

The purpose of the study on the pyroprocess for partitioning and ADS for transmutation is to propose a conceptual design of the HYPER system by 2006. In this study, the HYPER system, comprising a proton accelerator and a subcritical reactor, is considered an appropriate one for the Korean situation, with priority given to the nonproliferation attributes of the treatment of nuclear fissile materials in the back-end fuel cycle.

Safe Transport of Spent Nuclear Fuel and High-Level Waste: International Experience

Michael E. Wangler and *Ronald B. Pope*
International Atomic Energy Agency

The transport of radioactive material has an outstanding safety record. This safety record has its basis in model regulations that have been established by the International Atomic Energy Agency (IAEA). Since 1963 the IAEA through its member states has constantly reviewed and revised the regulations as appropriate. The regulations were originally designated as Safety Series No. 6, “Regulations for the Safe Transport of Radioactive Material”; since 1996 they have been referred to as TS-R-1. TS-R-1, along with its companion document, “Advisory Material for the IAEA Regulations for the Safe Transport of Radioactive Material,” TS-G-1.1, provides the basis for worldwide implementation of a harmonized approach to safe transport. TS-R-1 is now incorporated into the United Nations “Model Regulations for the Safe Transport of Dangerous Goods” (often called the “Orange Book”). The modal organizations of the United Nations (UN), International Civil Aviation Organization (ICAO), and International Maritime Organization (IMO) have incorporated the UN’s regulations into their own.

Although the safety in transport of radioactive material has continually improved over the years, other issues have begun to cause concern. Since the events of September 11, 2001, security in transport has been an issue. Because of the nature of radioactive material, its availability during public transport could lead to theft of unsecured materials for use in weapons that could disrupt society. While most of the radioactive material transported would not present a radiological hazard from a “dirty bomb,” the societal response to contamination could cause significant interruption in the populations and economies that are affected by the bomb.

Additionally, the response to an emergency in which radioactive materials are released during an incident or accident creates a liability issue that has threatened to deny transport for consignments. In particular, people have expressed growing concerns that radioactive material transported by sea, if inadvertently released, could affect a significant global area due to coastline contamination or disruption in commercial fishing. Consequently, issues of liability are continuing to be discussed by members of the IMO and the IAEA and their member states.

This paper will review these issues, with a particular focus on transport of spent nuclear fuel and high-level waste. The differences in safety and security will be discussed even though the terms have not yet been succinctly defined. The safety and physical protection in transport of spent nuclear fuel and high-level waste will be presented and the international acceptance of IAEA recommendations will be noted. A summary of the international liability regime will show the conventions that have been enabled to date. Data related to the international experience in shipping materials will also be presented. All of these discussion points demonstrate that the packagings used in transport are robust; that the likelihood of a release during transport is small; and that safety in the transport of radioactive material is not being compromised.

SAFETY VERSUS SECURITY

Before any discussion, the terms *safety* and *security* must be defined. The definitions presented here are limited to this paper. Subtleties exist in the definitions of these two terms, between languages and with different applications. In some languages safety and security mean the same thing. For this paper the discussion will focus on text derived from English dictionary definitions.

Because the terms have not been used together they are often defined in an unrelated manner, and there are certain schools that define security as a subset of safety. That is, safety of radioactive sources must involve the source being unable to be obtained by an unauthorized individual; however, a source that is simply secure may not be safe because of an unaddressed radiological hazard. Other schools identify them as separate but overlapping terms because nonradiological hazardous sources, such as poisons or flammables, that are secure may indeed be safe. Some schools consider safety as a part of security. Security of radiological sources potentially involves a threat analysis to determine the nature of security. Because of wide ranging hazards the application of safety and security is usually accomplished according to a graded approach, that is, the complexity of safety and security requirements will depend on the overall hazard of the materials. From a security perspective actions may range from a locked room for low hazards to armed guards for extremely high-hazard materials. Similarly, the application of radiological safety may range from limiting access to an area to excluding an area from most activity, for example, treatment of cancer with teletherapy devices.

For the purposes of this paper the following definitions are used: Safety relates to protection of people and the environment from unintentional exposure during normal use of and accidents involving radioactive material. On the other hand security relates to protection of people and the environment from malicious, intentional actions by man. These definitions are narrow for discussions in this paper, and as these subjects are discussed more frequently in public forums, additional limitations may further refine the meaning and intent of the terms.

SAFETY IN TRANSPORT OF SPENT FUEL AND HIGH-LEVEL WASTE

Safety in the transport of radioactive material is achieved through proper classification of the materials; use of appropriate containment through performance-oriented packages; criticality safety through limitations on fissile materials; communication for emergency response through marks, labels, placards, and shipping documents; and appropriate training of transport personnel. Worldwide, safety has been achieved through the development and implementation of a set of model regulations that have been adopted by IAEA member states through legally binding instruments imposed through the UN modal organizations and through legislative and regulatory actions at the state level.

The current regulations are the result of over 40 years of effort by the IAEA, which has the statutory function in the UN system. The IAEA establishes standards of safety by issuing its Regulations for the Safe Transport of Radioactive Material. The IAEA also provides guidance and supportive documents to assure that these regulations are implemented as uniformly as possible. The regulations are based on sound radiation safety principles. To underscore the importance of the regulations, in 1998 the IAEA General Conference, in a resolution, recognized that compliance with regulations, which take account of the agency's transport regulations, is *providing a high level of safety during the transport of radioactive material.*

Additionally, the IAEA provides for the application of the regulations through technical assistance, information exchange, education and training, research and development, and appraisal services. Of particular importance is the IAEA's Transport Safety Appraisal Service (TranSAS). TranSAS evaluates a member state's transport safety regulatory program by assessing the member state's transport safety regulatory practices, by suggesting and recommending improvements in the member state's program, and identifying and documenting good practices to assist other member states in their regulatory activities. To date, the IAEA has appraised five state programs.

The UN gave the IAEA these roles in 1959. Since that time six major editions of the regulations have been developed. The major editions were published in 1961, 1964, 1967, 1973, 1985, and 1996. Minor revisions have been incorporated within those dates. The current edition of the regulations is available in English, French, Spanish, and Russian.

The IAEA regulations serve as the basis or model for international and modal regulatory documents as follows:

- United Nations (UN) Committee of Experts: Model Regulations, that is, the Orange Book
- International Civil Aviation Organization (ICAO): Safe Transport of Dangerous Goods by Air, Technical Instructions
- International Maritime Organization (IMO): International Maritime Dangerous Goods (IMDG) Code
- Universal Postal Union: Parcel Post Manual
- UN Economic Commission for Europe (ECE): Regulations Concerning the International Carriage of Dangerous Goods by Rail (RID)
- UN Economic Commission for Europe: European Agreement Concerning the International Carriage of Dangerous Goods by Road (ADR)
- UN Economic Commission for Europe: European Provisions Concerning the International Carriage of Dangerous Goods by Inland Waterway (ADN)

All these regulatory documents cover all 9 classes of dangerous goods. Radioactive material is identified as a Class 7 dangerous good. Figure 1 depicts some of the relationships.

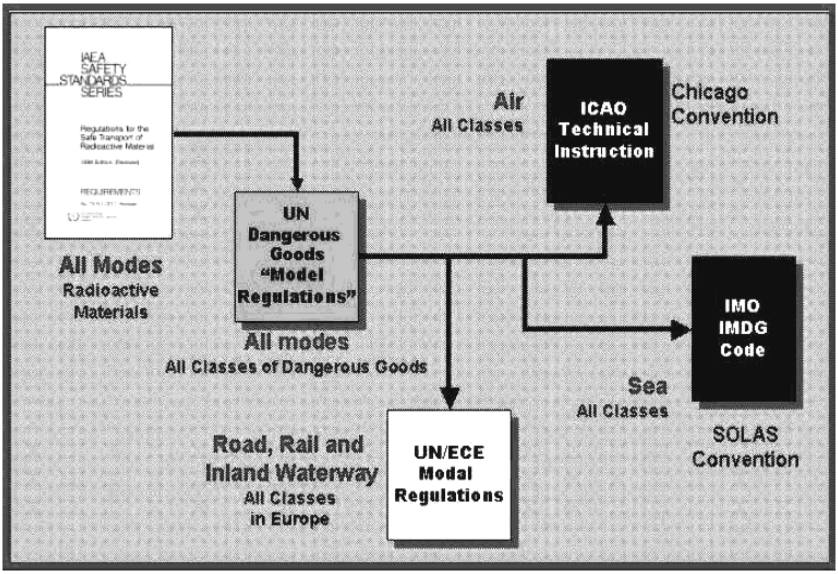


FIGURE 1 Relationships of regulations concerning transportation of dangerous goods.

APPLICATION OF IAEA REGULATIONS FOR THE TRANSPORT OF RADIOACTIVE MATERIAL

Although the IAEA regulations are not directly binding on member states, they are adopted by incorporation into the regulations of member states. Voluntary compliance by member states ensures that radioactive material is transported safely and efficiently across international boundaries. Member states expend much effort to achieve a harmonization of their regulations.

Additionally, the UN's modal bodies play an essential role in this harmonization. For air transport the ICAO's Technical Instructions are binding on all ICAO member states through the Chicago Convention. The IMO for sea mode binds all its members to comply with the IMDG Code. For travel by road, rail, and inland waterway in Europe compliance with ADR, RID, and ADN is mandatory.

For international sea transport the IMO developed the Irradiated Nuclear Fuel (INF) Code in cooperation with the IAEA, which details standards for ships that transport INF. INF in this case includes spent nuclear fuel, high-level waste (HLW), and plutonium. This code was adopted by IMO resolution in 1999 and became mandatory under the Safety of Life at Sea (SOLAS) convention on January 1, 2001.

The IMO has been particularly active in developing requirements for sea shipment. The regulatory regimes for the IMDG and INF Codes are distinct and separate. The IMDG Code addresses package design and operational requirements for transport of all radioactive material. The INF Code addresses the design of ships and the operational aspects for the limited subset of radioactive material noted above. The INF Code specifies three classes of ships with a graded approach to requirements. These classes are Class INF 1 with an aggregate cargo less than 4000 TBq, Class INF 2 with aggregate INF cargo less than 2×10^6 TBq of INF or HLW and/or 2×10^5 TBq of Pu, and Class INF 3 with no limit on INF, HLW, or plutonium. Class INF 3 Code ships have the most robust set of requirements imposed.

CONSIDERATIONS IN THE DEVELOPMENT OF AND COMPLIANCE WITH THE REGULATIONS

As mentioned earlier the regulations prescribe requirements for classification, containment, criticality safety, communication, and training. To reduce the effect of human factors during transport, the regulations provide for defense in depth to packaging requirements. The packaging is the primary defense against the release of radioactive material to the environment under all conditions. This defense is particularly important during an accident when packaging must perform in a robust manner to protect the public and the environment from the release of its radioactive contents. As statistics demonstrate, accidents involving radioactive material have occurred and will continue to occur. As the statistics further demonstrate, conformance with packaging requirements has resulted in no known deaths or injuries resulting from the radioactive nature of the cargo. The record of safety is unmatched by any other class of dangerous goods.

This emphasis on rigorous package design and test requirements by the regulations is especially important for larger quantities of radioactive material, such as spent nuclear fuel and high-level waste. The regulations prescribe rigorous design standards, demanding tests, and stringent acceptance requirements and limit the radiological exposure during and following an accident. Typical package accident test requirements include impacts onto an unyielding target, in the most damaging attitude through either a 9 m drop of the package, or a 9 m drop of a 500 kg steel mass onto the package, and a 1 m drop of the package onto a 15 cm steel bar; a full engulfing for 30 minutes with 800°C minimum average temperature; and a water immersion of 15 m, 0.9 m if fissile, and 200 m for larger quantities of radioactive material. Many tests and evaluations of the adequacy of the regulations have been undertaken over time.

In addition to these tests considering the experience with the application of the regulations, consideration has been given for a number of years to the question of whether or not the Type B performance requirements as specified in the regulations provide a sufficiently high level of safety; whether a more severe, as yet unexperienced accident could result in significant failure of a Type B package. National regulatory authorities, research institutes, and the IAEA secretariat have all attempted to address this issue. A large number of technical studies have been published addressing various aspects of the Type B performance requirements and the resulting levels of safety provided. These studies have illustrated the rigorous nature of the requirements and the resulting high level of safety. A number of these have focused on numerical risk assessments, which will not be addressed here. Others have focused on actual testing of packages in real-world environments, often at levels in excess of those required for design certification by the regulations. A few of these real-world test programs are briefly described below; they were often directed toward answering a question such as, "What happens to radioactive material packages in real-world accidents, since these may occur at speeds and with fire conditions greater than the regulatory tests?"

The technical answer to such a question is that real-world accidents, while they may appear spectacular, usually result in less damage to a package than the regulatory tests. The regulatory tests require that the impact, thermal, and immersion forces be applied to the packages in the most damaging way and in a specified sequence. This results in a very severe cumulative effect on the package structure. In real-world accidents this has never occurred, and is highly unlikely. Crushing of conveyances, rotation and displacement of the equipment involved, sliding, and so forth attenuate impact forces. Thermal forces are mitigated by the location of the package (especially an INF or HLW package since it is very heavy, and following an impact it could be expected to be lying on a horizontal surface where the fire is not as fully engulfing as the regulatory test); an intervening structure that will shield or absorb heat; the moving nature of a fire as it consumes available fuel; and the tendency to have low oxygen availability in the center of large fires, which reduces thermal input. Despite these technical and real considerations, there have always been questions about how the regulatory tests compare to real-world severe accidents.

SECURITY IN TRANSPORT OF SPENT FUEL AND HIGH-LEVEL WASTE

Since the events of September 11, 2001, added attention is being paid to security of radioactive material, including spent nuclear fuel and high-level waste. All the UN bodies, including the UN Committee of Experts, the IMO, and the ICAO have been focusing attention on security. The IAEA Board of Governors has requested that the IAEA take extraordinary actions to address security. Member states have made extrabudgetary contributions to assist the IAEA in this area. In October 2003 the IAEA will convene a Technical Meeting to address guidelines on security in transport of radioactive material.

These events could lead one to believe that transport security has never been addressed or has received only limited attention. In fact, security has been addressed for nuclear materials for many years. Guidance on security for nuclear materials is already provided for in an IAEA document, INFCIRC/225/Rev.4, "The Physical Protection of Nuclear Materials and Nuclear Facilities." This document identifies plutonium with less than an 80 percent concentration of plutonium-238, uranium-235, and uranium-233 as nuclear materials. Seventy-eight states have signed the Convention on Physical Protection of Nuclear Materials, which was a precursor to INFCIRC/225.

For security in the transport of nuclear materials INFCIRC/225 requires consideration of the following actions:

- minimizing time in transport
- minimizing number and duration of transfers during transport
- providing protection consistent with category of materials
- avoiding the use of regular movement schedules
- requiring predetermination of the trustworthiness of all individuals involved
- limiting advance knowledge to a minimum number of persons

Further, INFCIRC/225 describes the categories of materials that are to be protected. In decreasing order of security needs these categories are the following:

Category I. Material in sufficient quantity to be useful in producing a workable nuclear device (highest security level)

Category II. Material that either in total mass or need for further operation is not useful in itself for producing a nuclear device (requires significant security)

Category III. Material in quantity or quality that is insufficient in itself in producing a nuclear weapon (requires slightly more stringent security than standard transport)

Transport of all other dangerous goods faces a new paradigm since September 11. The UN has already developed security requirements for its model regulations for all dangerous goods. The IMO has issued a new code for ship and port facilities.

As already mentioned the IAEA is developing a graded approach to recommendations for security in the transport of radioactive material other than nuclear materials. IAEA actions will also address vitrified high-level waste. Meetings of experts in 2002 and 2003 in Vienna have produced draft guidance on security of radioactive material in transport, which includes a three-level graded approach to categorization of materials. This draft guidance strives for consistency with the nuclear material security requirements of INFCIRC/225/Rev. 4, with the guidance on categorization of sources for safety and security of TECDOC (IAEA technical document) 1355, and the categorization of sources for emergency response purposes.

Much work stills needs to be done. Over the next few months the IAEA will hold a series of meetings at which transport safety and security will be addressed. In July 2003 the IAEA sponsored an International Conference on the Safety of Transport of Radioactive Material, where a status report on security in transport was presented. Reports from IMO and ICAO, which cosponsored the conference, provide information on security. In October 2003 a Technical Meeting to Address Guidelines for Security in the Transport of Radioactive Material will review the draft proposed guidance and recommend changes to the text. Decisions will then be made as to which further actions are needed to issue the guidance as a TECDOC, the title of which is envisioned to be "Security in the Transport of Radioactive Material—Interim Guidance for Comment." The final document will strive for agreement on categorization activity level per conveyance and proposed security measures by category. It is anticipated that the resulting guidance document will be published within a year.

INTERNATIONAL LIABILITY

Liability is an important consideration in any normal activity. Liability is especially important for activities that can have high environmental and financial consequences. Because of its complexity, liability is only broadly and briefly presented here.

Safety and security requirements and recommendations establish the basic conditions under which transport activities are conducted. Attention is inevitably trained on transport when there is an accident that has the actual or perceived potential to cause significant physical harm to a population and financial harm to a community. Liability becomes a key issue in restoring a community to its preaccident condition.

In the international liability regime adequate protection of the public from dangers of the application of radioactive material, including those associated with nuclear energy, must be maintained without hindering the development of the nuclear industry and the other beneficial uses of radioactive material. This situation normally limits the liability of any entity, with the country of the entity indemnifying or assuming responsibility for the cost the entity is required to pay.

In the existing liability regime multiple conventions and agreements exist. A listing of these conventions and agreements is as follows:

- 1960 Paris Convention
- 1963 Vienna Convention
- 1963 Brussels Supplementary Convention
- 1971 Maritime Carriage of Nuclear Material Convention
- 1988 Joint Protocol
- 1997 Protocol to Amend the Vienna Convention
- 1997 Convention on Supplementary Compensation

In addition to these conventions, the IAEA is planning to convene a group to discuss liability in the nuclear field. To date, neither the terms of reference for a meeting nor an actual meeting date have been established.

INTERNATIONAL EXPERIENCE

In spite of the many safety, security, and liability issues transport of radioactive material continues to grow. For the Workshop on the Problems of Managing Spent Nuclear Fuel and Selection of a Site for Its Storage, the IAEA was asked to provide an overview estimate of the worldwide experience in the packaging and transport of nuclear-power-plant-related irradiated nuclear fuel and high-level waste. The current information results from an evaluation performed in 1999–2000, reported in detail in the Proceedings of the International Symposium on the Packaging and Transportation of Radioactive Materials (PATRAM) 2001. A previous attempted study to collect shipping data was reported by IAEA in the Proceedings of PATRAM ’86.This study provided a mix of data, for example, materials and modes, which were judged to be incomplete. However, from the

TABLE 1 Estimated Shipments of INF and HLW by all Modes

Shipments[a]	Estimated Quantity (MTHM)	Estimated Flask Shipments
LWR/Magnox/AGR to Sellafield and The Hague	50,000–75,0000	18,500–37,100
Other in France	10,518	>125
Canada–Japan (other than to Sellafield/The Hague)	1100	557
Slovakia	239–380	635–700
Sweden (domestic)	3300	1100
Ukraine	1300	300
United States	2274	3025
HLW from The Hague	520	8
Approximate Totals	~70,000–95,000	24,000–43,000

[a] Estimate through 2000, worldwide, excluding Russian Federation.

data in this study one could determine that an estimated 18 to 38 million packages of radioactive material are transported each year.

For this presentation a less extensive review was done. The collected data on spent nuclear fuel (SNF) and HLW shipments was obtained by personal contacts and the published literature. Contacts included consignors, carriers, consignees, and competent authorities.

Data show that multiple types of INF have been transported, including light water reactors (LWR), such as pressurized water reactors and boiling water reactors; gas cooled reactors, such as magnox, advanced gas reactors (AGR), and graphite reactors; and other types of reactors, including high-power channel reactors (RBMK), Canada deuterium uranium reactors (CANDU), high-temperature reactors (HTR), and fast breeder reactors (FBR). It is estimated that between 24,000 and 35,000 flask shipments have been made. Table 1 summarizes the data collected.

SUMMARY

The thousands of shipments of spent fuel and high-level waste have been performed safely and efficiently. Requirements in place ensure that the robustness of the package provides the first line of defense to materials being released to the environment. To date, there has been no harm to any individual as a result of the contents in the package. This record is not exceeded by the transport of any other hazard class. As the need to transport spent fuel and high-level waste increases, the transport community must be diligent in transporting the materials. All regulations, including package and communication requirements, must be followed.

Ensuring Nuclear and Radiation Safety During the Transport of Radioactive Materials in Russia*

Aleksandr M. Agapov
Russian Ministry of Atomic Energy

In accordance with the Federal Law on the Use of Atomic Energy, the management of nuclear materials and radioactive substances during transport is one of the many elements involved in the atomic energy sector as a whole. Consequently, the transport process lies within the scope of responsibilities of federal agencies dealing with atomic energy safety management and regulation in general.

One important component of the legal and regulatory base with regard to transportation safety is Resolution No. 204 of the Government of the Russian Federation, entitled "On the Competent State Authority for Nuclear and Radiation Safety During the Transport of Nuclear Materials, Radioactive Substances, and Related Products." According to this resolution, which was issued on March 19, 2001, the functions of the competent state authority are assigned to the Russian Ministry of Atomic Energy (Minatom). The provisions established by this resolution define the tasks, functions, rights, and responsibilities of the competent state authority.

Transport is one of the potentially dangerous stages in the cycle in which radioactive substances are utilized. Radiation safety is ensured during transport by means of various organizational (administrative) and technical measures.

Transport safety is promoted by the state regulatory system, the Minatom management system, and the system of safety procedures in place at the shipper level.

---

*Translated from the Russian by Kelly Robbins.

Radioactive substances are divided into two basic groups according to their uses:

- cargo from nuclear fuel cycle enterprises, including spent nuclear fuel, fresh (nonirradiated) nuclear fuel, and nuclear materials; shipments of these types of cargoes are under the complete control of Minatom
- cargo for general industrial use (so-called "isotopic" products)

Depending on the properties of the materials being transported (predominantly their degree of radioactivity), the different types of shipments may be divided into five categories characterized by various safety requirements. The bulk of the shipments related to the nuclear fuel cycle are carried out by Minatom enterprises using their own specialized means of transport (trucks and rail cars).

Ensuring the safety of shipments is the dominant activity of the Minatom organizations and enterprises involved in transporting radioactive materials. Inherent in the concept of the safety of radioactive material shipments is the condition of all design, operational, technical, organizational, and other elements that play a role in the shipment process, with these elements furthermore understood not to entail any impermissible risk of harm to the lives or health of citizens, the property of individuals, legal entities, municipalities, or the state, the environment, or the lives or health of animals or plants.

The goals of ensuring safety in the transport of radioactive materials are as follows:

- preventing emergencies or unauthorized actions
- improving the system for response to emergencies and unauthorized actions
- predicting potential emergencies
- reducing the risk of emergencies and unauthorized actions
- reducing the effects of radiation on the population and the environment
- reducing the level of irradiation received by operating personnel, law enforcement officers, and rescue workers during emergency response, rescue, and transport operations
- detecting and suppressing unauthorized actions with regard to radioactive materials
- reducing the severity of the consequences of emergencies and unauthorized actions
- reducing the time required to carry out emergency response, rescue, and transport operations and to detect and suppress unauthorized actions
- improving the efficiency of personnel and equipment involved in responding to emergencies and unauthorized actions

TECHNICAL INFRASTRUCTURE FOR TRANSPORTING RADIOACTIVE MATERIALS

The safety of shipments of radioactive materials is ensured through the practical implementation of the following basic principles:

- uniform federal norms and rules governing the procedures for transporting radioactive materials
- a unified system and rules for certifying casks for shipping radioactive materials
- uniform rules and methods for testing casks and transport vehicles

In order to ensure safety the overwhelming majority of shipments of radioactive materials are carried out with the help of complex and long-lasting pieces of equipment, namely, special transport casks, vehicles, and lifting and loading apparatuses.

Minatom currently uses the following in shipping radioactive materials:

- more than 30 types of transport casks for shipping fresh nuclear fuel
- some 14 types of casks for shipping spent nuclear fuel
- about 20 types of casks for shipping fissile materials

The late 1970s and early 1980s saw the appearance of several new types of rail cars: the TK-6 for spent fuel from water-moderated water-coated power reactors (VVER-440), the TK-10 for fuel from VVER-1000s, and the TK-11 for fuel from high-power channel reactors (RBMK). Due to the lack of capabilities for reprocessing fuel from RBMK reactors, the TK-11 containers began to be used for shipping spent fuel from BN-350 and BN-600 reactors to the RT-1 plant. The improved TK-13 rail car was created in the mid-1980s for use with spent fuel from VVER-1000s, and the TK-10 container was then taken out of production. A total of 16 TK-6 rail cars were developed and manufactured for shipping spent fuel from VVER-440 reactors.

Since 1986, spent fuel from VVER-1000 reactors has been transported using TK-10 and TK-13 rail cars and TUK-10V and TUK-13V transport casks. A total of 7 TK-10 containers and 12 TK-13 containers are currently in use. At the present time the demand for rail cars and casks for shipping VVER-1000 spent fuel is being fully met. The volume of spent fuel shipments could even be increased if necessary if new power units with VVER-1000 reactors were to come online.

Plans call for bringing spent fuel from BN reactors to the Mayak Production Association for reprocessing, with the materials to be transported in TK-11 rail freight containers.

The follow two types of casks are used to transport spent nuclear fuel from research reactors for reprocessing:

- TUK-19 (for fuel rods from VVR-K, VVR-Ts, VVR-2, VVR-S, VVR-M, IRG, IVV, MR, and SM-2 reactors)
- TUK-32 (for fuel rods from SM-2 and MIR reactors)

The TUK-19 and TUK-32 packaging units are in full compliance with the requirements of OPBZ-83 (Basic Rules for Safety and Physical Protection during Nuclear Materials Transportation) and the Regulations for the Safe Transport of Radioactive Material issued by the International Atomic Energy Agency in 1985. These units are classified as type B(U), class 1 for nuclear safety.

A reinforced concrete container intended for long-term storage and transport of spent fuel from RBMK reactors is in the final design and certification stage.

Since 1994, spent nuclear fuel from atomic submarines and icebreakers has been transported using TK-VG-18 rail freight containers and new-generation TUK-18 radiation-shielded transport casks.

Work is under way in Russia to create new transport casks to handle spent fuel from VVER-440, VVER-1000, and RBMK-1000 reactors, as well as those used in atomic submarines and icebreakers.

Fresh nuclear fuel (fuel rods and assemblies intended for irradiation in reactors) is shipped in TK-S-type transport casks. All told, more than 50 types of packaging are used, ranging in weight from tens of kilograms (the TK-S35 for fuel rods) to 6.5 metric tons (the TK-S7M for fuel assemblies for the Bilibino Nuclear Power Plant).

Practically all of the packaging units have undergone testing in accordance with the requirements of the OPBZ-83 rules, which govern type B packaging, and all of the units have received Minatom certificate permits.

An experimental line of packaging units is currently in the development and testing stages.

As per Russian and IAEA requirements, type B(U), B(M), and C packaging must be subjected to a series of tests of their performance under emergency transport conditions, specifically,

- a 9 m fall
- dynamic destruction in the event of a 500 kg object falling on the packaging unit from a height of 9 m; testing for cracking or breakage resulting from the unit falling onto a spike from the height of 3 m (or a spike falling onto the unit from the same height)
- the effect on the unit of an all-encompassing external heat field with an average temperature of 800 °C lasting at least 60 minutes
- collision of the unit with a hard target at a speed of at least 90 m/sec

Experimentally or theoretically confirming that the packaging units are safe in the first three emergency situations listed does not present any serious difficulties for designers, and this confirmation may be obtained using the designers' existing test facilities or through calculations using generally accepted methodology.

Specialists from the Russian Federal Nuclear Center—All-Russian Scientific Research Institute of Nuclear Physics have demonstrated the feasibility of conducting such tests at the center's unique test facilities. The United States is the only other country that possesses an experimental facility with analogous characteristics.

Many countries transport radioactive materials, and the total quantity of packaging units is about 4 million. The major shippers include the United States, Canada, Japan, Great Britain, and various developed European countries (see Table 1).

Over the past 30 years Russia has amassed a great deal of experience in transporting fissile materials between enterprises involved in the nuclear fuel cycle (see Figure 1). During this time, more than 1600 freight car trips have been completed, covering a total distance of more than 10 million km. Primary among the products carried were uranium and uranium compounds of various degrees of enrichment and physical form.

It must be noted that over the entire period in which all types of radioactive materials have been shipped, Russia has never had any incidents or accidents as defined under the International Nuclear Event Scale (INES). During the past 10 years, there have been four incidents with low-enriched uranium and ionizing radiation sources, and three of these incidents did not involve any breach in the integrity of the packaging units. If we look at the statistics for work-related injuries, the share of such cases involving radioactive substances comprises a very insignificant fraction of all cases related to dangerous materials.

In the United States there were 282 accidents during the period 1971 through

TABLE 1 Shippers of Radioactive Materials by Country

Country	Number of Packaging Units Shipped Annually
United States	2,402,429 (domestic shipments only)
Canada	612,632
Japan	353,054
Great Britain	242,268 (incomplete data)
Italy	205,679
Germany	155,408
France	99,617
TOTAL	4×10^6

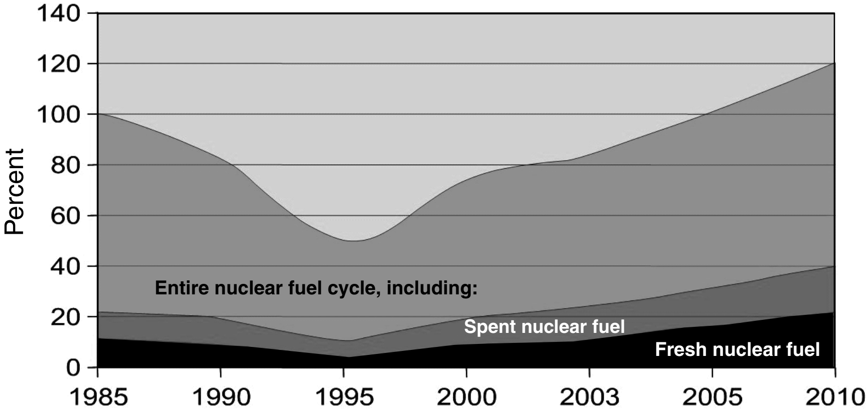


FIGURE 1 Dynamics of radioactive material shipments in Russia. (In 2001 about 1800 shipments of radioactive materials were made within the scope of the nuclear fuel cycle.)

1989, most of them involving ionizing radiation sources. None of the U.S. accidents related to the shipment of radioactive materials involved the destruction of packaging units.

LAWS AND REGULATIONS

Organizing and coordinating the creation of draft federal regulations and rules concerning nuclear and radiation safety during transport of radioactive materials is also a function of the competent state authority.

The development of a national legal and regulatory base for ensuring the safety of shipments is in complete accord with current world trends. Past practices were characterized by a closed regime for shipments of most radioactive materials in general and practically 100 percent of nuclear materials not only within the nuclear weapons complex but also with regard to those shipments pertaining to the nuclear fuel cycle. At present we believe that design-based safety, that is, safety resulting from the design of the packaging used for radioactive materials, must be augmented by organizational measures that would minimize the likelihood of transport-related violations involving radioactive materials being shipped. Using the terminology of IAEA rules, one might say that special conditions are used for shipments. This primarily pertains to shipments of those materials and substances that are potentially the most dangerous, such as spent nuclear fuel and plutonium. Special regulatory documents covering procedures for shipments of these types of radioactive materials include strict requirements of an organizational nature.

As international design requirements for radioactive materials packaging have been developed and corresponding IAEA rules have been issued, these requirements have also been introduced in Russia (and formerly in the U.S.S.R.) in the form of state standards (in 1977 for packaging for nonfissile radioactive materials and revised in 1988) and national rules (in 1983 for nuclear materials). These standards and rules were developed on the basis of IAEA rules. Russia is now preparing to introduce new national rules in full technical (design) compliance with the new IAEA rules (document ST-1/TS-R-1, 1996/2000 editions). These new national rules will cover all types of radioactive materials (fissile and nonfissile).

The transport of radioactive materials is governed by the following regulatory documents:

- Rules for the Safe Transport of Radioactive Substances (PBTRV-73), specifically in accord with the provisions of the Regulations for the Safe Transport of Radioactive Material, 1996 edition, Requirements ST-1 (hereafter referred to as “Requirements ST-1”)
- Basic Rules of Safety and Physical Protection in the Transport of Nuclear Materials (OPBZ-83), which do not contradict the provisions of Requirements ST-1
- Radiation Safety Norms (NRB-99)
- Basic Sanitary Rules for Ensuring Radiation Safety (OSPORB-99)
- Basic Industrial Rules of Nuclear Safety in the Use, Processing, Storage, and Transport of Hazardous Nuclear Fissile Materials (PBYa-06-00-96)
- Provision on Procedures for the Shipment of Fissile Nuclear Materials by Air Transport in the Russian Federation (PVPYaDM-93), which do not contradict the provisions of Requirements ST-1
- Handbook on the Shipment of Special Cargo by Rail and Truck Transport (RSP-86), which does not contradict the provisions of Requirements ST-1
- Instructions on the Shipment of Special Cargo (ISP)
- Rules for the Shipment of Dangerous Cargo by Truck Transport
- Accident reference cards
- Other regulatory documentation, for example, OST-95 10297-95 (Spent Fuel Assemblies from Nuclear Research Reactors: General Requirements for Shipment) and OST-95 745-95 (Spent Fuel Assemblies from VVER-Type Nuclear Power Reactors: General Requirements for Shipment to Reprocessing Plants)
- Recommendations from Requirements ST-1

LICENSING AND CERTIFICATION OF ACTIVITIES RELATED TO THE TRANSPORT OF RADIOACTIVE MATERIALS

The basic provisions governing the safe management of nuclear materials are set forth in Russian legislation, currently active federal norms for the transport of radioactive materials, and regulatory documents from the Russian Federal Inspectorate for Nuclear and Radiation Safety (Gosatomnadzor). One of the main requirements involves mandatory licensing for the activities of enterprises engaged in the design, manufacture, construction, installation, and operation of facilities and equipment (see Figure 2).

With regard to the handling of radioactive materials during transport, the licensing system that has been created in the Russian Federation ensures compliance with most of the requirements that have been established for this sort of activity.

The basis for the licensing requirements used by the above-listed federal executive-branch agencies generally complement one another, while the practice of independent expert reviews as instituted by Gosatomnadzor also significantly reduces the risk that licensees (enterprises) may fail to meet the requirements.

All types of work associated with the management of radioactive materials during transport are subject to licensing.

The system for certification of facilities, goods, and technologies (FGT) that has been put in place in Russia jointly by Minatom, Gosatomnadzor, and the

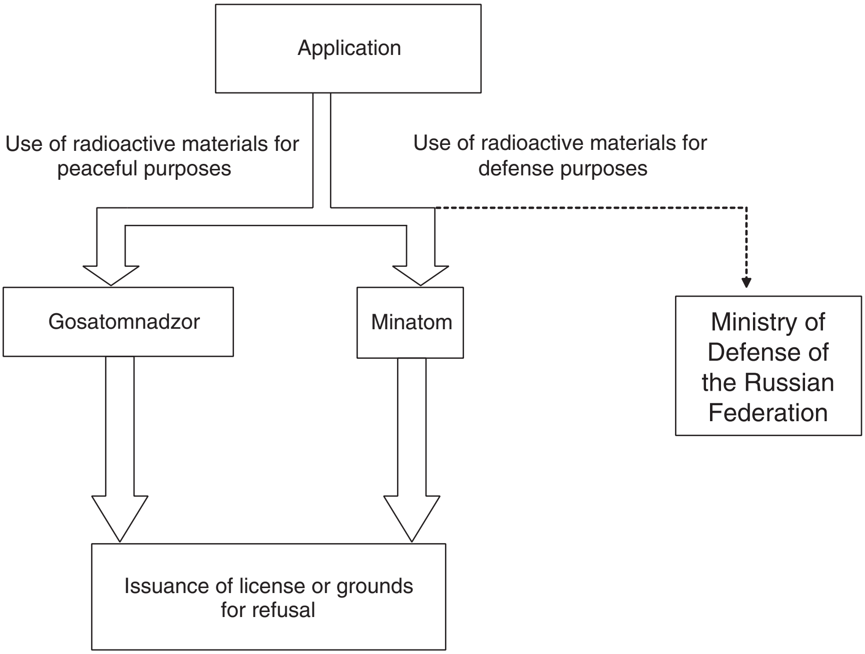


FIGURE 2 Outline of the licensing process.

Russian State Committee for Standardization and Metrology (Gosstandart) supplements the above-described licensing system with regard to compliance by Russian enterprises with requirements for equipment and technologies as generally accepted in international and domestic practice. The FGT certification system allows Minatom and Gosatomnadzor to jointly regulate the process of harmonizing international and domestic requirements and to standardize in practice the modern equipment and technologies used in managing radioactive materials.

All shipments of nuclear materials and radioactive substances within the territory of the Russian Federation must be and are made only upon the issuance of a permit certificate regarding the design and movement of the transport cask with the materials or substances.

At this time, in order to transport nuclear materials or radioactive substances in the Russian Federation, it is necessary to obtain the permit certificates from Minatom as follows:

- regarding the type of radioactive substance
- regarding the packaging design (type A, IP-2, IP-3, AF, III, B(U), B(U)F, B(M), B(M)F
- regarding the shipment
- regarding shipment under special conditions

In order to improve efforts to organize the safe transport of shipping casks with nuclear and radioactive materials, by order of the ministry (No. 192, dated July 8, 1992) the Temporary Provision on Procedures for the Issuance of Permit Certificates for Radioactive Substances of a Certain Type and for the Design and Transport of Packaging Containing Radioactive Substances (PVSR-92) was put into effect. According to this provision, functions related to the preparation of draft permit certificates are performed by the following operational agencies:

- Federal State Unitary Enterprise, Minatom Emergency Technology Center; Federal State Unitary Enterprise, All-Regional Association Isotope; and the Russian Federal Nuclear Center, All-Russian Scientific Research Institute of Technical Physics: with regard to preparation of draft permit certificates (for radioactive substances of a certain type, excluding fissile substances)
- Head Institute, All-Russian Design and Scientific Research Institute of Complex Energy Technology and the Russian Federal Nuclear Center, All-Russian Scientific Research Institute of Experimental Physics: with regard to preparation of draft permit certificates (for fissile substances of a certain type)
- Department of Nuclear Safety of the Russian State Scientific Center, Institute of Physics and Power Engineering: with regard to expert review of measures and conditions for nuclear safety and issuance of corresponding conclusions for preparation of permit certificates (for nuclear safety procedures)

On the instructions of Minatom, an industry-wide transport packaging database has been developed that will make it possible to monitor and account for the permit certificates issued by the competent state agency as well as current information on nuclear safety and expert conclusions reached by the operational agencies in preparing their permit drafts. This information system will facilitate accounting and control regarding the location and movement of packaging units as well as their technical condition. All of this will facilitate the efficient preparation of documents (certificates) regarding the technical status and availability of packaging units as well as tracking of the life cycle of the certificate from the moment of its confirmation by means of references to corresponding notices. The system also offers quick navigation between linked documents, for instance from the certificate to the nuclear safety ruling, the accident card, the notification, and other documents.

ENSURING THE SAFETY OF SHIPMENTS OF RADIOACTIVE MATERIALS

The Federal State Unitary Enterprise Atomspetstrans is responsible for operational control and organization of shipments of radioactive materials. The basic tasks of Atomspetstrans include the following:

- organizing shipments of radioactive materials
- cooperating with interested organizations on matters of physical protection of radioactive materials on a contractual basis
- monitoring the validity dates of permit certificates regarding packaging design and transport
- coordinating with the client regarding shipment schedules and conditions and entering all information obtained into the operational database
- checking that the declared cargo corresponds to the information on the accident card and is appropriate for the type of transport that has been arranged (presence and type of freight rail cars, auxiliary rail cars, and guards)
- checking permit documents regarding the transport vehicles and shipping containers against the industry-wide database by type of transport vehicle, container, operating certificate, and other control documents in order to determine the feasibility of their use in the given transport operation
- monitoring the location and movement of empty transport vehicles and shipping containers and obtaining from railway authorities and regional officials confirmation of their arrival at the intended destination

Up to now Russia has experienced no accidents or serious incidents involving the transport of nuclear materials or, in particular, of spent fuel.

In order to be able to respond quickly and efficiently in the event of a transport accident during the shipment of nuclear materials, enterprises involved in such shipments as well as state agencies have developed plans to prevent accidents and eliminate their potential consequences. These plans are submitted to the federal and regional authorities, and shippers and freight carriers have their own action plans and preparedness procedures in the event of an accident. Accident response cards are also prepared for each type of cargo.

In order to implement accident prevention and response plans Russia has created a system of regional Emergency Technology Centers (ETCs) under the auspices of Minatom. These centers constantly monitor shipments of nuclear materials within their assigned territories and respond quickly in case of any emergency. ETC personnel are trained in accident response and have the appropriate equipment and technical capabilities at their disposal. The major enterprises involved in the nuclear fuel cycle also have special emergency brigades. Forces and resources from the Ministry of Emergency Situations may also be deployed if needed to deal with the consequences of an accident.

With the aim of promoting a unified technical policy on crisis management, a Crisis Situation Center has also been created within the framework of Minatom's existing system for monitoring the security of enterprises and organizations.

Initial determination of the degree of danger presented by a radiation accident is made by the individual escorting the shipment, in accordance with the accident card that accompanies any shipment of radioactive material. This same individual is also responsible for undertaking first-response efforts regarding the accident.

The system for monitoring cargo movements throughout the territory of the Russian Federation provides the emergency technology infrastructure for the shipments. This system is responsible for maintaining forces and resources in constant readiness to prevent and eliminate the consequences of potential emergencies arising during the transport process.

The system is based on six federal, full-time, professional emergency rescue and accident response units that remain on constant alert. These units are certified by the Interagency Commission to conduct accident response, rescue, and other urgent missions in emergency situations. The system also includes Minatom-certified reserve emergency rescue and accident response units based at the various nuclear-sector organizations involved.

The Federal State Unitary Enterprise Minatom ETC is an element of the industry-wide subsystem of the Russian System for Emergency Prevention and Response. ETC is one of six emergency technology centers under the auspices of Minatom.

In accordance with Resolution No. 924 of the Government of the Russian Federation dated August 3, 1996, ETC is listed among the federal rapid response forces of the Russian System for Emergency Prevention and Response.

The Monitoring and Dispatch Service is a full-time structural unit of the professional emergency response and rescue unit of Minatom's ETC. Its basic activities are

- organizing efforts, within the scope of its competency, to ensure rapid notification of response and rescue personnel from the enterprise in the event of an emergency or change in status associated with radiation
- organizing notification of regional emergency response and rescue units regarding shipments of nuclear materials and radioactive substances in their assigned territories
- exchanging operational information with the dispatch and monitoring services of enterprises involved in the transport of nuclear materials and radioactive substances within Russian territory as well as with nuclear-hazard enterprises, the main administrations of the Ministry of Emergency Situations, and local governments in the northwestern region of Russia

As part of overall efforts to provide scientific and technical support for the response system for accidents involving the transport of radioactive materials, the Russian Academy of Sciences Institute of Problems of the Safe Development of Nuclear Power operates a Technical Crisis Center, the tasks of which include

- assessing the radiological consequences of accidents involving the transport of radioactive materials
- developing recommendations on protecting personnel, the general population, and the environment

Role-playing games and command staff training exercises are the most effective methods for addressing issues related to the training of both specialists and government officials on appropriate actions to take in emergency situations.

The primary objective of the training exercises with regard to ensuring the safe transport of radioactive materials is to improve the mechanism for cooperation among the ministries, agencies, and organizations involved in these activities. Among the major exercises that have been conducted are Urals 99, in which participants practiced actions to eliminate the consequences of an accident during the shipment of spent nuclear fuel to the Mayak Production Association; the command staff exercises held in the Southern Federal District in March 2002; and the exercises held at the Mining-Chemical Complex and the Novosibirsk Chemical Concentrates Plant. All of these exercises involved the participation of officials from the emergency services of Minatom and various regions and enterprises as well as emergency units and emergency technology centers.

The intensified threat from terrorist organizations of various types, including the threat of nuclear terrorism, has made it imperative to improve the physical protection of industrial facilities that present a nuclear or radiation risk, with special attention required for the problem of increasing the security of shipments of nuclear materials.

Given the global nature of these threats, Minatom and the U.S. Department of Energy have been working together since 1996 to create an automated system for ensuring the safety of shipments of special nuclear materials, with these efforts being a part of the international cooperation between these agencies to improve the overall physical protection, accounting, and control of nuclear materials.

Based on the results of an analysis of situational models, the system has subsystems for

- monitoring the status of the physical protection of nuclear materials during transport and the location of the transport vehicles
- detecting violations (unauthorized actions with regard to nuclear materials during transport)
- restricting access to nuclear materials
- communications and control
- emergency response in the event of an attempted attack aimed at seizing nuclear materials or conducting an act of sabotage

USING THE SCIENTIFIC POTENTIAL OF THE RUSSIAN ACADEMY OF SCIENCES

One of the most fundamental questions with respect to the management of nuclear materials in general and spent nuclear fuel in particular relates to safety. The scientific potential of the institutes of the Russian Academy of Sciences (RAS) makes it possible to carry out work in this area according to modern standards. The RAS has participated directly in the development of the Federal Targeted Program for Russia’s Nuclear and Radiation Safety; in the comprehensive analysis of radiation and chemical risks to the Russian population; in an assessment of risks to personnel, the population, and the environment during various stages in the management of spent fuel; and in other important safety-related efforts.

RAS institutes are also carrying out practical applied work in the area of spent fuel and radioactive waste management. In particular, the Institute of Geology of Ore Deposits, Petrography, Mineralogy, and Geochemistry and the Vernadsky Institute of Geochemistry and Analytical Chemistry are developing plans for burial sites in geological formations and evaluating the raw material potential of the nuclear fuel cycle. The Institute of Problems of the Safe Development of Nuclear Power is developing modern safety criteria and preparing a comprehensive analysis of the safety of atomic power and related industries.

CONCLUSIONS

The 50 years of experience in transporting nuclear materials and radioactive substances within the Russian Federation and abroad proves the effectiveness of the system for ensuring nuclear and radiation security in handling such shipments. The organizational and technical measures being taken by Minatom to improve the safety of radioactive material shipments, along with the common practice of safe shipments, provide grounds for concluding that the shipment system on the whole is effective.

During 2001, approximately 1800 shipments were made within the framework of the nuclear fuel cycle. On average there are six or seven trains carrying radioactive materials on the rails each day.

New and complex tasks with regard to transport safety have arisen in connection with manifestations of terrorism. Top priority objectives in this regard include

- completing work on developing and deploying the automated system for ensuring the safety of shipments of special nuclear materials
- completing the creation of a modern material-technical base for emergency response and rescue units and emergency technology centers and resolving issues related to the provision of social benefits for their personnel
- designing, testing, certifying, and manufacturing new standardized transport vehicles and packaging units in order to modernize the existing fleet

Regulatory activity represents another problem associated with the transport of radioactive materials. The main areas for improvement with regard to the above-mentioned Russian regulatory documents pertain to

- putting the draft Rules for Safe Transport of Radioactive Materials into effect as quickly as possible
- developing and putting into effect the draft Sanitary Rules for Ensuring Radiation Safety for the Population and Personnel during Transport of Radioactive Materials (Substances)

When the Federal Law on Technical Regulation goes into effect in 2003, it will bring with it a need for profound analysis of the entire regulatory and legal base governing matters of transport safety. Following are the main questions regarding the transport of radioactive materials that must be addressed:

- development of a technical regulation for the transport of radioactive materials
- improvement of the quality control system regarding organizations involved in transporting radioactive materials
- enhancement of the system for certification of facilities, goods, and technologies related to shipments of radioactive materials

PROBLEMS IN ESTABLISHING AN INTERNATIONAL STORAGE FACILITY FOR SPENT NUCLEAR FUEL IN RUSSIA

Creating an Infrastructure for Managing Spent Nuclear Fuel*

K. G. Kudinov
Mining-Chemical Complex, Federal State Unitary Enterprise

INTERNATIONAL SPENT NUCLEAR FUEL MANAGEMENT TODAY

At the end of 1998 there were 434 nuclear power reactors in operation worldwide. According to International Atomic Energy Agency (IAEA) estimates, about 225,000 metric tons of spent fuel had been removed from reactors by 2000, of which 75,000 metric tons are to be reprocessed and 150,000 metric tons are destined for storage. Some 10,000 metric tons of spent fuel are removed from reactors worldwide each year.

Spent fuel reprocessing plants exist in only a few countries: France (1600 metric tons per year), Great Britain (1200 tons per year), Russia (400 tons per year), and Japan (100 tons per year, with a plant capable of processing 800 tons per year currently under construction). Some of the countries involved in nuclear power production need services for the temporary or long-term storage of spent fuel due to the lack of their own storage facilities. Experts believe that the market for spent fuel storage and reprocessing services totals some $160 billion.

A total of eight nuclear power plant units with water-moderated water-cooled reactors (VVER-1000) are currently operating in Russia (at the Balakovo, Kalinin, Novovoronezh, and Rostov plants). Plans had called for spent fuel from the VVER-1000 reactors to be stored and reprocessed at the RT-2 plant (at the Mining-Chemical Complex, Federal State Unitary Enterprise in the city of Zheleznogorsk, Krasnoyarsk Krai). The first part of the plant—a wet (water-cooled) repository complex with a designed capacity of 6000 metric tons—went

---

*Translated from the Russian by Kelly Robbins.

into operation in 1985. Since 1992, construction of the plant's reprocessing complex has been on hold due to a lack of funds. The repository has been accepting spent fuel since 1985 from three Russian power plants (seven units), and four Ukrainian plants (11 units): South Ukrainian, Zaporozhye, Khmelnitsky, and Rovno. The amount of spent fuel received annually totals 250 to 350 metric tons. In 2001–2002, 82 metric tons of spent fuel was received from the Kozloduy nuclear power plant in Bulgaria (two units). The total amounts of spent fuel received annually are about 140 metric tons from Russian nuclear power plants, 220 metric tons from Ukraine, and 40 tons from Bulgaria.

The importance of issues related to spent fuel management is also confirmed by the fact that the international conference "Irradiated Nuclear Fuel Management 2002: Russian New Initiatives" was the first such conference ever held in Russia. In his speech at this conference First Deputy Minister Mikhail I. Solonin noted that "the development of the nuclear power industry is being hindered by the lack of clear-cut and broadly understandable solutions with regard to the safe management of spent nuclear fuel and radioactive wastes." Solonin proposed the creation of major international complexes for spent fuel storage and reprocessing and possibly for the manufacturing of new fuel and the recycling of radioactive wastes, with these complexes to be located at Minatom enterprises with the technologies and most importantly the experience required in this regard. I would like to emphasize this initiative and propose that the Mining-Chemical Complex be considered as a candidate for this type of operation.

According to the Strategy for the Development of the Nuclear Power Industry in Russia in the First Half of the Twenty-First Century, construction of the RT-2 plant is planned for the period 2020 through 2025, which is just prior to the wide-scale development of nuclear power production using fast neutron reactors (2030).

In accord with the closed nuclear fuel cycle concept that has been adopted in Russia, the basic stages of the nuclear waste management process are illustrated in Figure 1.

I will now discuss in greater detail the various stages involved in the transport and storage of spent nuclear fuel as carried out in practice at the Mining-Chemical Complex.

TRANSPORT OF SPENT NUCLEAR FUEL

One important element of spent fuel management is its safe shipment from the nuclear power plant to the site for its long-term storage and/or radiochemical reprocessing. In transporting spent fuel (irradiated fuel rods) care must be taken to ensure the nuclear and radiation safety and physical security of the shipment and to protect it against any damage in case of an accident. All transport casks are certified and meet Russian safety standards as well as IAEA requirements for use in normal operating conditions as well as emergency situations.

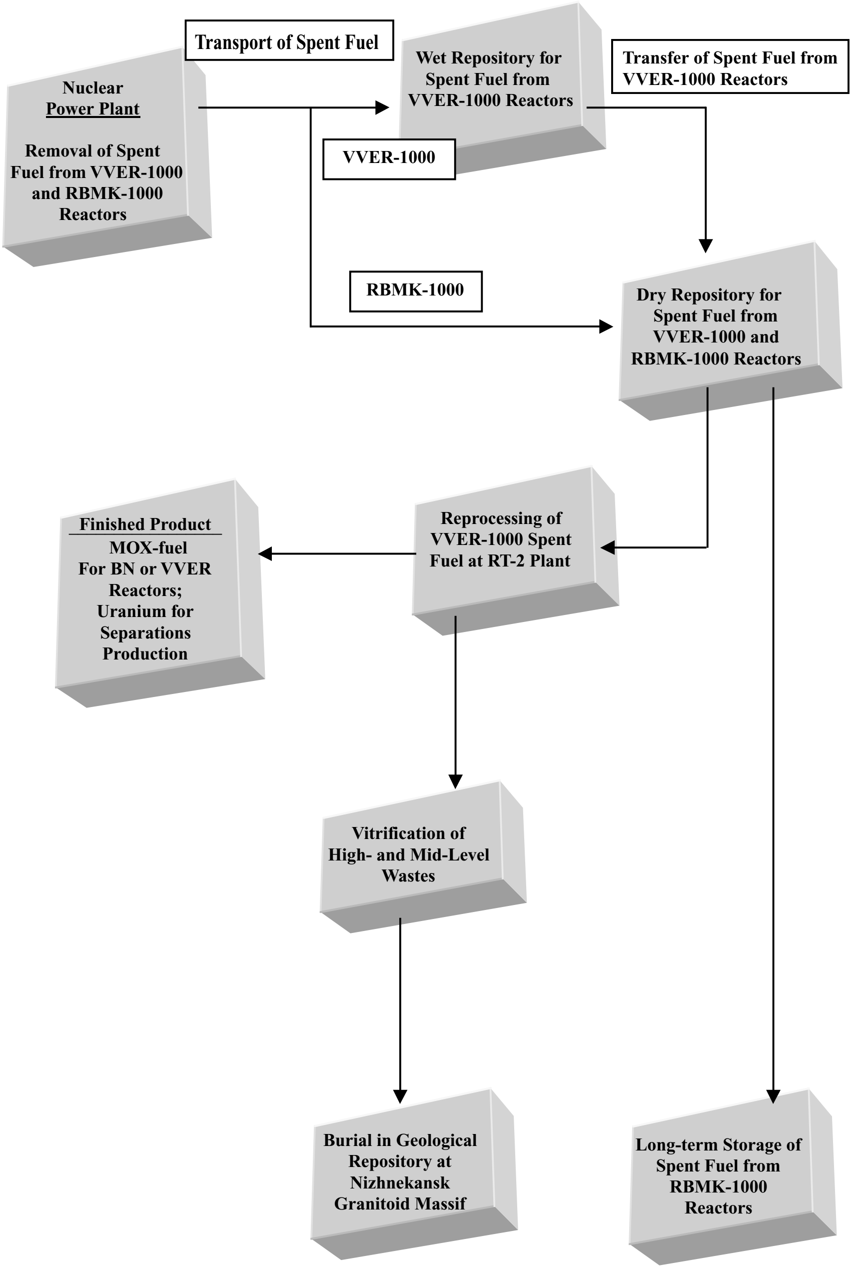


FIGURE 1 Nuclear waste management with a closed nuclear fuel cycle.

The Mining-Chemical Complex has three types of transport casks available for shipping spent fuel:

1. TUK-10V (capacity: 6 fuel rods), 7 units
2. TUK 13/V (capacity: 12 fuel rods), 5 units
3. TUK 13/1V (capacity: 12 fuel rods), 11 units

The complex also has the following transport containers for casks and auxiliary train cars: TK-10, 7 units; TK-13, 12 units; TK-13T, 2 units; TK-13M, 2 units; TK-VS, 4 units.

There have been no incidents or occurrences involving the transport of spent nuclear fuel since the system has been in operation. More than 700 train car trips have been completed to date, with the distance traveled by the freight containers totaling more than 2 million km for the TUK-10Vs and more than 4 million km each for the TUK-13/Vs and TUK-13/1Vs.

The time required to complete a shipment averages 50 days. The special trains spend 12 to 16 days in actual transit, and it takes one or two days to load and unload each container.

The transport casks have an operating lifespan of 20 years, and many of these units will soon be reaching the end of this period. It is essential to extend the operating period of existing casks or manufacture new ones with increased carrying capacity. In order to reduce costs for building new casks and freight containers, a standardized radiation-shielded transport container should be developed for shipping various types of spent fuel, as well as a standardized transport container for carrying casks in various modes of transportation. Firms should be selected to manufacture casks and transport containers on the basis of a competitive bidding system.

These objectives are included in the draft of the Program for the Handling of Irradiated Fuel Rods from Nuclear Reactors During Shipment for 2003 to 2015 (PROYAT-2003), which was developed by the All-Russian Design and Scientific Research Institute for Comprehensive Energy Technology (VNIPIET) and is currently being prepared for adoption.

CURRENTLY OPERATING WET REPOSITORY

This storage facility went into operation in December 1985. Its planned capacity is 6000 metric tons. As of January 1, 2003, it had received 3500 metric tons of spent fuel from VVER-1000 reactors, including approximately 1500 tons from Ukraine and 82 tons from Bulgaria, with the remaining 1920 tons coming from Russia.

In order to increase the capacity of the facility and improve the safety of spent fuel storage, various organizational and technical measures and other work are being carried out in accordance with the Plan of Measures for the Construction of the Unit Linking Building 1 with Building 2 at the RT-2 Plant and for the Construction of the Dry Storage Facility at the Mining-Chemical Complex as approved by the Minister. After this work is completed, the capacity of the facility will be increased to 8600 metric tons.

Plans call for spent fuel (irradiated fuel rods) to be stored at the wet facility for 30 years, after which the material is to be either sent to the regeneration plants (RT-1 and RT-2) for reprocessing or transferred for longer-term storage at the dry facility.

DRY SPENT FUEL STORAGE FACILITY

The task of building a dry spent fuel storage facility with a designed capacity of at least 6000 metric tons as the second component of the RT-2 plant was first put forth in the state environmental impact review that modified the design plans for the RT-2 plant (Ministry of Natural Resources Order No. 364, dated June 14, 1996).

At its October 1996 meeting to review the course of implementation of Russian Federation Presidential Decree No. 72, dated January 25, 1995 (On State Support for the Restructuring and Conversion of the Atomic Industry in the City of Zheleznogorsk, Krasnoyarsk Krai), the Minatom board assigned specific tasks to Minatom subunits involved in creating the dry storage facility for spent fuel from high-power channel reactors (RBMK) and VVER-1000 reactors. Based on this decision by the Minatom board, VNIPIET in 1999 prepared an investment study for a standard dry reinforced concrete-type facility with a designed capacity of 33,000 metric tons, of which 24,000 tons was to come from RBMK-type reactors and 9000 tons from VVER-1000s. Total construction costs were estimated at 10 billion rubles. The investment study passed all required expert reviews. Taking foreign experience into account, various options were subsequently worked out for the construction of other types of dry facilities, namely, those involving storage in vertical boreholes, vaults, and casks.

In 2001 VNIPIET, its Krasnoyarsk branch, and the Mining-Chemical Complex proposed that the unfinished buildings of the RT-2 plant (buildings 2, 3, 3A, and 3B) be used in the creation of a dry spent fuel storage facility. Visual and instrument studies of these buildings were conducted, and the results confirmed that using the structures for this purpose was possible. This option will make it possible to accelerate and reduce the cost of construction.

The Minatom Scientific-Technical Council and the ministry's board have approved the vault option for the dry spent fuel facility. The design for the vault-based dry facility was developed by VNIPIET in December 2002.

Taking into account the complex situation with regard to RBMK spent fuel, the Minatom board decided at its June 2002 meeting that the initial complex of the facility should be created by 2005, primarily to handle RBMK-1000 wastes, and the board also supported the proposal to use buildings 2, 3, 3A, and 3B of the RT-2 plant in the construction of the dry facility.

According to the Plan of Measures for the Construction of the Unit Linking Building 1 with Building 2 at the RT-2 Plant and for the Construction of the Dry Storage Facility at the Mining-Chemical Complex, which was approved by the minister in July 2002, construction of the initial complex of the dry repository will begin in 2003, with the facility to go into operation in 2005.

The total costs for the construction of the dry facility, including the value-added tax, will be 16.6 billion rubles at price levels of the first quarter of 2003 (432.1 million rubles in 1991 prices), of which the costs for the initial complex are estimated at 8.12 billion rubles in first-quarter-2003 prices.

The designed capacity of the facility for spent fuel from RBMK-1000 and VVER-1000 reactors totals 37,785 metric tons (26,510 tons for RMBK-1000 fuel and 11,275 tons for VVER-1000 fuel). The capacity of the initial complex of the repository is 8862 metric tons.

With the aim of getting the facility into operation more quickly, a startup unit with a calculated capacity of about 5100 metric tons of RBMK-1000 fuel has been identified for completion as part of the initial complex at the dry facility. VNIPIET has been assigned the task of completing the design for the startup unit in March 2003.

At the recommendation of the Rosenergoatom firm, the revised design must also include a cask site for the storage of reinforced concrete containers of spent fuel. According to the investment study prepared in 1999, plans call for this cask site to be big enough to hold 121 TUK-104 reinforced concrete transport packaging units. At the time when the investment study was undergoing review and revision, the cask site was eliminated, and therefore the project was never started.

DRY STORAGE FACILITY OBJECTIVES FOR 2003

The following tasks relating to the dry storage facility are to be accomplished during 2003:

- successfully pass expert review of the vault option for the dry facility
- carry out preparatory work at the site (work is under way to dismantle the existing structural elements of the buildings of the RT-2 plant)
- obtain a license from the Russian State Atomic Energy Inspectorate (Gosatomnadzor) for placement and construction of the dry facility

At the demand of the Krasnoyarsk Krai Environmental Protection Committee, phase one of the environmental impact assessment of the RT-2 plant will also be submitted for expert review along with the design for the dry facility.

PROSPECTS FOR EXPANDING THE MARKET FOR SPENT FUEL-RELATED SERVICES

As previously noted the Mining-Chemical Complex is currently accepting spent fuel from nuclear power plants in Russia, Ukraine, and Bulgaria for temporary technical storage and subsequent reprocessing. The package of "nuclear" laws passed by the State Duma of the Russian Federation in July 2001 (Federal Laws No. 92-FL, 93-FL, and 94-FL) allow the complex to accept spent fuel from other countries. Besides the countries listed above, there are others that could be potential shippers of spent fuel. However, for the above-mentioned laws to really work requires that the Government of the Russian Federation pass a number of regulatory acts or governmental resolutions, something that has not yet been fully achieved.

The dry facility is also a prerequisite for the receipt of foreign spent fuel, but in the initial phase the spent fuel could be accepted at the existing wet facility or temporarily stored in containers until the dry facility is completed. The Mining-Chemical Complex and Minatom are losing substantial amounts of hard currency due to the inability to carry out such operations. The funds required for the construction of the facility for foreign spent fuel or the cask site should be obtained as an advance payment from the prospective suppliers of spent fuel, without the need to use any Russian funds for this purpose.

It should be noted that the number of potential clients wishing to send spent fuel for temporary storage and subsequent reprocessing is substantially greater than the number of clients requiring only temporary storage, so one would hope for the spent fuel reprocessing complex to be completed more quickly.

Current Status of Government Regulation of Activities Associated with the Import of Spent Nuclear Fuel into the Russian Federation*

A. M. Dmitriev
Russian Federal Inspectorate for Nuclear and Radiation Safety

Government regulation of nuclear and radiation security activities associated with the import of spent nuclear fuel into the Russian Federation is enforced in accordance with international agreements, federal laws, decrees of the president of the Russian Federation, resolutions of the Russian Government, the Statute on the Russian Federal Inspectorate for Nuclear and Radiation Safety (Gosatomnadzor), various federal norms and rules, and Gosatomnadzor regulatory documents.

Two organizations under the Russian Ministry of Atomic Energy (Minatom) are involved in activities associated with the acceptance of spent nuclear fuel from foreign nuclear power plants—the Mayak Production Association (spent fuel from water-moderated water-cooled power reactors [VVER-440]) and the Mining-Chemical Complex (spent fuel from VVER-1000 reactors).

Located at the Mayak Production Association in the city of Ozersk, the Russian Federation's only operating spent fuel reprocessing enterprise—the RT-1 plant—can accept for reprocessing spent fuel from nuclear power plants with VVER-440, BN-600, and BN-350 reactors as well as from research reactors and atomic-powered ships and submarines. In contrast to its designed capacity of 400 metric tons per year, in recent years the RT-1 plant has processed no more than 150 metric tons per year. In 2003 Mayak plans to reprocess 132 metric tons of spent fuel of various types.

Construction of the RT-2 plant at the Mining-Chemical Complex in Zheleznogorsk was halted in 1989. The technology proposed for the plant, which was

---

*Translated from the Russian by Kelly Robbins.

intended for reprocessing spent fuel from VVER-1000 reactors, had become obsolete and no longer met modern requirements. The only unit of the RT-2 plant that was completed was the spent fuel storage facility, which had a planned capacity of 13,416 fuel assemblies from VVER-1000-type reactors (6000 metric tons of uranium by weight). At this time the facility has been filled to approximately 57 percent of its planned capacity.

In 2002 the above-mentioned enterprises received six special shipments of spent fuel from nuclear power plants in Ukraine and one from Bulgaria (specifically, Mayak received one shipment from the Rovno Nuclear Power Plant, while the Mining-Chemical Complex received one shipment each from the Khmelnitsky, Zaporozhye, and Rovno plants, two from the South Ukrainian Nuclear Power Plant, and one from the Kozloduy Nuclear Power Plant in Bulgaria). Last year Mayak received 27.4 metric tons. The total amount of spent nuclear fuel received by the existing storage facility at the Mining-Chemical Complex is 140 metric tons from the Ukrainian power plants and 41.5 metric tons from the one in Bulgaria. The following shipments are planned in 2003:

- Mayak: one shipment from the Rovno plant and one from the Kozloduy plant
- Mining-Chemical Complex: three shipments from the South Ukrainian plant, one from the Khmelnitsky plant, and one from Kozloduy

The way that the spent fuel is transported is important from the security standpoint. In shipping spent fuel from VVER-440 reactors, 16 TK-6 rail containers are used. Designed in 1973–1978, a period characterized by a lack of experience in such operations and of relevant regulatory documents, the TK-6 is a first-generation container. Its design does not make it completely secure. The TUK-6 transport cask used with this container is categorized as type B(M), which must be used with certain restrictions and constant monitoring by operators.

Spent fuel from VVER-1000 reactors has been transported since 1986, using TK-10 and TK-13 rail containers and TUK-10V and TUK-13V transport casks. The TUK-13/1V includes a vessel made of corrosion-resistant steel (container and documentation produced by the Izhorsk Plants Open Joint-Stock Company).

According to the rules of the International Atomic Energy Agency (IAEA), the TUK-13 transport casks are considered to be type B(U), and their design fully ensures the security of the spent fuel transported in them. In shipping spent fuel with this sort of packaging no special organizational or technical measures are required to monitor packaging and environmental parameters or to ensure that these parameters are maintained within permissible limits. The TUK-10V transport cask is analogous in design to the TUK-13; however, its capacity is 50 percent lower (6 spent fuel rods as opposed to 12 with the TUK-13), and its body is made of 20-mark carbon steel, just like the body of the TK-6 container.

At present, demand for shipping containers and casks for transporting spent fuel from VVER-1000 reactors is being met in full. There are 40 unused TUK-13V casks at nuclear power plants in Russia and Ukraine. These shipping containers and transport casks for VVER-1000 spent fuel were manufactured between 1983 and 1992, and their 20-year service life runs out in the period 2003 to 2012.

FEDERAL LAWS AND REGULATIONS OF THE GOVERNMENT OF THE RUSSIAN FEDERATION

Three federal laws (FL) were passed in 2001:

1. On Amending the Federal Law on the Use of Atomic Energy, July 10, 2001, No. 94-FL
2. On Amending Article 50 of the Law of the Russian Soviet Federal Socialist Republic (RSFSR) on Environmental Protection, July 10, 2001, No. 93-FL
3. On Special Environmental Programs for Rehabilitating Radiation-Contaminated Areas, July 10, 2001, No. 92-FL

The additions made to Article 50 of the RSFSR Law on Environmental Protection were included in full in Federal Law 7-FL on Environmental Protection dated January 10, 1992.

To effect control over the import of foreign-made irradiated fuel rods into the Russian Federation and in accordance with Decree No. 828 of the president of the Russian Federation (July 10, 2001), a special commission was to be formed, to be chaired by Russian Academy of Sciences Vice President Zhores I. Alferov, who also serves as a deputy in the State Duma of the Federal Council of the Russian Federation. A draft presidential decree has been prepared and is being processed by the president's staff.

On October 15, 2001, the Russian government issued Order No. 1371-r confirming the Plan for the Preparation of Draft Resolutions of the Government of the Russian Federation Necessary for Implementing the Federal Laws on Special Environmental Programs for Rehabilitating Radiation-Contaminated Areas and on Amending Article 50 of the RSFSR Law on Environmental Protection. This order called for the creation of five draft resolutions for the Russian government, two of which (Procedures for the Importation of Irradiated Fuel Rods into the Russian Federation and the Return to the State of Origin of Nuclear Materials or Radioactive Wastes from These Spent Fuel Rods; and Procedures and Timelines for the Development of Special Environmental Programs) were developed in cooperation with Gosatomnadzor.

In accordance with Russian Government Order IK-P7-15492 dated September 1, 2001, officials have developed the Plan for Preparation of Draft Decisions on Amending Regulatory Legal Acts of the President of the Russian Federation as Necessary to Comply with the Federal Law on Amending Article 50 of the RSFSR Law on Environmental Protection.

The changes and additions were made in the following decrees by Presidential Decree 1284 on the Making of Changes and Additions to Certain Decrees of the Russian Federation President on Matters of Spent Nuclear Fuel Management in order to bring them into accordance with the Federal Law on Environmental Protection:

- On Fulfillment by the Russian Federation of Intergovernmental Agreements on Cooperation in the Construction of Nuclear Power Plants Abroad, April 21, 1993, No. 472
- On State Support for the Restructuring and Conversion of the Atomic Industry in the City of Zheleznogorsk, Krasnoyarsk Krai, April 25, 1995, No. 72
- On Additional Measures to Strengthen Control over Compliance with Environmental Security Requirements in the Reprocessing of Spent Nuclear Fuel, April 20, 1995, No. 389

REGULATIONS

Regulations concerning activities associated with the import of spent nuclear fuel include regulations on the operation of storage facilities and nuclear plants involved in reprocessing and transporting spent fuel as well as managing the radioactive wastes created as a result of reprocessing operations.

In recent years a number of regulatory documents with a direct impact on the security of spent fuel management have been developed and put into effect.

The Rules for Safety in the Transportation of Radioactive Materials have been prepared for implementation, and their safety requirements for shipping and packaging are in full accord with analogous IAEA rules governing the safe shipment of radioactive materials by all forms of transport.

LICENSING

Mayak and the Mining-Chemical Complex operate on the basis of the following Gosatomnadzor licenses:

Mayak

- License GN-03-115-1060 dated February 14, 2002, for operation of a complex to handle nuclear materials (Plant 235) intended for the radiochemical reprocessing of spent nuclear fuel and management (reprocessing, storage) of radioactive wastes created as a result of the processing of spent fuel and of the defense-related activities of Mayak (valid until February 29, 2004)

- License 05-401-0748 dated January 31, 2002, for management of nuclear materials being transported (valid until May 1, 2005)

Mining-Chemical Complex

- License GN-03-301-0724 dated December 25, 2001, for operation of a stationary facility intended for the storage of nuclear materials (repository for irradiated VVER-1000 fuel rods containing spent nuclear fuel)
- License 05-401-0723 dated December 25, 2001, for management of nuclear materials being transported (valid until March 1, 2005)

Spent nuclear fuel from foreign nuclear power plants is imported after issuance of Gosatomnadzor permits to the reprocessing enterprise and the client organization, which must apply to the Russian Ministry of Economic Development to obtain a spent fuel import license.

The specific quantity of spent fuel to be received for reprocessing is determined by Minatom after taking into account the environmental situation in the region where the reprocessing enterprises are located. The decision is coordinated with federal environmental protection agencies and executive branch agencies of regional (oblast or krai) and local governments.

The total content of radionuclides (activity) of the solidified radioactive wastes to be returned to the country that shipped the spent fuel is determined by mutually agreed methodology, taking into account the time at which the spent fuel arrives and the time the solidified wastes are held for storage.

OVERSIGHT

State oversight over the importation of foreign spent fuel into the Russian Federation is carried out by Gosatomnadzor's Urals and Siberian branches.

The Mining-Chemical Complex accepts spent fuel at the border of the Russian Federation. At that point the Mining-Chemical Complex assumes from the spent fuel supplier the risk of accidental loss or damage as well as the responsibility for providing physical security for the nuclear materials.

Insurance policies are drawn up on each shipment to provide a contract covering civil liability to third parties for losses and damage caused by the effects of radiation during the shipment of radioactive substances, nuclear materials, and associated products. Copies of the insurance policies are submitted to Gosatomnadzor.

When a trainload of these materials is dispatched, the inspection department checks the following:

- certificates and associated regulatory documents on the technical condition of the transport casks, as well as certificates on technical and maintenance reviews of the rail containers conducted at specialized enterprises of the Russian Ministry of Railways

- certificates on the return of transport casks to service after completion of maintenance work (if such work was completed)
- certificate-permits for B(U)-category design and shipment
- agreement on the transfer of responsibility for the physical security of the spent fuel signed by the governments of all countries through which the shipment will pass

PROBLEMS INVOLVED IN THE IMPORT OF FOREIGN SPENT FUEL INTO THE RUSSIAN FEDERATION

The Russian Federation currently has very limited technical capabilities for accepting irradiated fuel rods from foreign states for reprocessing and temporary technical storage. This is due to the following circumstances:

1. The RT-1 plant (Plant 235) has been in operation for 25 years and its equipment is worn out and in need of modernization, especially taking into account the need to create sections for the conditioning of all types of radioactive wastes.
2. There is a worldwide trend toward increasing the depth of nuclear fuel burn-up, which raises the issue of the expediency of reprocessing irradiated fuel rods, that is, such fuel contains significantly less useful uranium and more fission products and has a higher total radioactivity level. Reprocessing this sort of fuel and subsequently using the regenerated materials becomes technically and economically impractical. For this reason the concept of the closed nuclear fuel cycle is being called into question throughout the world.
3. The use of the remaining capacity of the spent fuel repository at the Mining-Chemical Complex to store foreign irradiated fuel rods threatens the possibility of using this capacity to store fuel rods from Russian nuclear power plants, as this could lead to the accumulation of spent fuel rods at the power plants beyond the designed capacity and could force them to halt their operations.

As noted previously, operations involving the transport of irradiated fuel rods are currently carried out using Russian-made shipping containers and packaging units. Given the current volume of foreign shipments (shipments of Russian-made fuel rods from abroad total no more than 200 metric tons per year), the existing quantity of shipping containers and transport casks meets the demands of Mayak and the Mining-Chemical Complex. However, as soon as we start talking about shipping thousands of metric tons, the existing containers and casks will be insufficient and will have to be manufactured on a massive scale, which will require correspondingly substantial amounts of time and financial resources.

Furthermore, we must not forget that the importation of irradiated fuel rods from foreign boiling-water reactors (BWR) and pressurized water reactors (PWR) is currently problematic, as the shipping containers and other equipment at Mayak and the Mining-Chemical Complex are not appropriate for use with foreign transport casks holding fuel rods from these reactors, and the Russian casks are not certified for use abroad and could not be used to ship these sorts of fuel rods. Minatom is holding a competition for the development of a standardized transport cask for irradiated fuel rods from VVER-440, VVER-1000, high-power channel reactors (RBMK-1000), BWR, and PWR. Seven cask designs differing in form and type of materials used have been entered in the competition.

International relations involved in the management of spent fuel rods are regulated at the level of the Joint Convention on the Safety of Spent Fuel Management and on the Safety of Radioactive Waste Management as well as by intergovernmental agreements. Given the intent of the Russian Federation to expand the range of goods and services it offers on the world market, high priority should be given to implementing the provisions of this Joint Convention in Russia.

Nuclear and radiation security in the management of spent fuel rods can be achieved only through the observance of existing laws, norms, and rules regarding the use of nuclear energy. However, it must be noted that law and regulatory requirements in the Russian Federation do not fully cover the entire sphere of spent fuel rod management.

An analysis of potential accidents in normal and extreme conditions (including acts of terrorism) during the shipment of irradiated fuel rods shows that localized radiation contamination of the environment is possible. A breach in the packaging unit during an accident could lead to localized radioactive contamination up to a dose level of 20 R/hour. The fact that accidents could occur gives rise to the legal problem of who bears responsibility for the nuclear damages, a problem that could become a serious impediment to the successful activities of Russian organizations in the world market for spent fuel management services.

In 1995 the Russian Federation signed the Vienna Convention on Civil Liability for Nuclear Damage. The draft of the Federal Law on Civil Legal Responsibility for the Causing of Nuclear Damage and Provision of Financial Compensation has been under review by the State Duma since 1997. The lack of legal regulation on this matter is undoubtedly having a negative effect on insurance activities in this field.

Return to the Russian Federation of Irradiated Fuel Assemblies from the Institute of Nuclear Physics of the Republic of Uzbekistan

Aleksey E. Lebedev
Techsnabexport

FOREIGN SPENT NUCLEAR FUEL FROM RESEARCH REACTORS BUILT UNDER TECHNICAL ASSISTANCE OF THE FORMER SOVIET UNION

In the 1960s and 1970s the former Soviet Union carried out an intensive construction program of research reactors abroad. In contrast to commercial reactors, decisions on the feasibility of research reactor construction were mainly based on political grounds. As a result developing countries with unstable political and economic systems built a large number of research reactors, which did not contribute to nonproliferation goals.

The reactors were supplied with fresh fuel assemblies, containing highly enriched uranium (HEU—36–90 percent enrichment for U-235). At the beginning of the 1970s the Soviet government made the decision to begin converting foreign research reactors to lower enrichment levels; however with the disintegration of the Soviet Union this effort was discontinued.

Normally spent fuel from foreign research reactors was not returned to the country in which it had been manufactured. (Notable exceptions include both spent nuclear fuel [SNF] from the former Soviet republics and the return operation of spent fuel assemblies from the Iraq Nuclear Research Center in Baghdad, accomplished by air in 1993, in accordance with the UN Security Council Resolution.) As a result large quantities of highly enriched SNF assemblies have been accumulated at the research reactors' onsite SNF storage facilities.

In May 1997 the United States announced the start of a large-scale program of SNF return from foreign research reactors. The program involves permanent disposal of research SNF from 41 states in facilities of U.S. design in the United States.

The program's major goal is the reduction of HEU quantities contained in SNF assemblies at research reactor sites, as well as advocating that the reactors be converted to use low-enriched fuel. From the U.S. perspective the issue of nuclear proliferation risk reduction in the area of research reactor fuel cannot be resolved completely without the involvement of Russia, the second largest world supplier of corresponding facilities, technologies, and fuel.

In 1999 Techsnabexport (a company authorized by the government of the Russian Federation to conduct foreign deals related to the management of spent nuclear fuel from foreign nuclear reactors) began several rounds of negotiations with the operators of foreign research reactors. Preliminary consultations have revealed a keen interest by a number of foreign states to begin regular shipments of their spent research nuclear fuel to Russia. However, in view of the limited budgets (or in some cases, the complete lack of funds) of the research reactor operators to finance SNF operations, large-scale fuel shipments hardly seem possible without the assistance of sponsors, such as the IAEA, the United States, EUROATOM, and private funders.

The United States has expressed its readiness to provide financial assistance to countries returning their fuel to Russia, on the condition that the HEU gradually be converted to that with enrichment of less than 20 percent U-235. At the same time, during trilateral meetings among the IAEA, Russia, and the United State, possibilities were discussed for the return of the spent nuclear research fuel to the Russian Federation.

In 2001, as a result of IAEA-Russia-U.S. technical missions to research reactor sites in Uzbekistan, Ukraine, and Yugoslavia, Uzbekistan was selected to be the first state from which the return of the spent fuel assemblies would be executed. The complex political situation in Central Asia and the maximum technical and organizational preparedness for the Uzbekistan export operation were factors in this decision.

SPENT NUCLEAR FUEL FROM THE RESEARCH REACTOR AT THE INSTITUTE OF NUCLEAR PHYSICS OF THE UZBEKISTAN ACADEMY OF SCIENCES

The Uzbekistan Academy of Sciences, Institute of Nuclear Physics was founded in 1956. It is one of the largest scientific institutes in Asia, currently carrying out fundamental and applied research in nuclear and elementary particle physics, solid body physics, radiochemistry, biology, element analysis, and many other areas.

Initially the power of the VVR-SM reactor, which was put into operation in September 1959, was 2 Mwt. In the Soviet era the reactor was used for both military and civil purposes; in 1971–1979 it was upgraded and its power increased to 10 Mwt.

From 1959 to 1971 the reactor was loaded with fuel assemblies with enrichment up to 10 percent U-235. After modernizations from 1979 to August 1998, the reactor was powered by fuel assemblies with enrichment up to 90 percent U-235. In September 1998 specialists from the Kurtchatov Institute converted the reactor operation from fuel with 90 percent enrichment to fuel with 36 percent enrichment. Since October 2000, further work has been done on enrichment reduction to bring down the level to 19.7 percent U-235. Future activities on conversion to low-enriched fuel will depend on terms and volumes of financial assistance allocated by foreign sponsors.

Return of spent fuel from Uzbekistan was carried out during 1974–1991. In total, 449 fuel assemblies were transported to the Soviet Union during that period. The government of Uzbekistan is pursuing the policy of turning Central Asia into a nuclear-weapons-free zone but is perplexed by the presence of sizable quantities of highly enriched nuclear material within its borders. A particular danger is the location of the research reactor close to the combat zone in Afghanistan (the reactor is located only 137 km from the Uzbek-Afghan border).

In accordance with the agreement an IAEA mission for U.S. and Russian experts was organized at the research reactor site in July 2001. A complete check of the transport-technological scheme was performed, including loading spent fuel assemblies into the TUK-19 transportation casks, transportation of the casks to the railway station by motor vehicle (7 km), and the loading of the TUK-19 casks into special train cars. The transport-technological scheme has proven viable. Personnel who participated in past return operations are still available at the reactor, as is the equipment used during previous transports.

The overall quantity of spent nuclear fuel assemblies (mainly with high levels of enrichment) stored currently at the onsite storage facility is 259 pieces. Two hundred fifty-six assemblies are planned to be transported to the Mayak production complex in Russia (the reprocessing plant for the research and commercial spent nuclear fuel) in four trips. No difficulties of a technical nature should arise in reprocessing the given types of fuel assemblies at the Mayak plant.

LEGAL ASPECTS FOR THE RETURN OF SPENT NUCLEAR FUEL ASSEMBLIES

As has been mentioned, return of spent fuel from foreign research reactors to Russia was not possible in the 1990s, due not only to financial difficulties of reactor operators but also for legal reasons.

The order for the import of spent nuclear fuel from foreign reactors into Russia, enacted by the Russian government in 1995, is not valid for research reactor fuel. Only in the framework of new Russian federal legislation in the field of management of spent nuclear fuel from foreign reactors, adopted in 2001, has an opportunity emerged for the implementation of the research reactor spent fuel export program. Currently work is underway to elaborate and harmonize legislative acts of the Russian government that would regulate in detail the importation of spent nuclear fuel from foreign reactors into Russia, in close connection with various special environmental programs for the cleanup of radioactively contaminated areas. So far only two out of five government acts have been approved.

Taking into account the request of the government of Uzbekistan to provide assistance for the return of spent nuclear fuel assemblies from the Institute of Nuclear Physics, and in view of the present absence of the full package of legislative acts, the Russian government has issued a special directive dated April 25, 2003, No. 521-R. This dictates preparations, according to the requirements of the Russian legislation project, for the export of spent nuclear fuel assemblies from the Institute of Nuclear Physics.

After preparations for the project are completed and approval from the state ecological experts is received, Minatom will introduce corresponding proposals about the order and terms of import for given spent nuclear fuel assemblies to Russia.

Practical implementation of the given project will facilitate

- strict commitment of Russia, as a nuclear weapons state, to safeguard mechanisms for nonproliferation of nuclear weapons
- support of political stability in Central Asia
- implementation of ecological cleanup measures for rehabilitation of regions that have been radiologically contaminated as the result of the nuclear arms race in the 1950s and 1960s

Investment and International Aspects of the Problem of Spent Nuclear Fuel Management

Vitaly P. Keondjian and *Michael A. Zhdanov*
Association for Nonproliferation and Ecological Improvement

Our presentation builds upon the idea of providing services in Russia for storing and safeguarding spent nuclear fuel (SNF) coming from those countries where dealing with SNF is either impossible or not economically viable. Proceeds from the sale of this service will be managed by private organizations, the Nonproliferation Trust in the United States, and by our association in Russia. They will be used for improving fissile materials security and for ecological, restoration, and social programs in Russia.

In these areas our association has identified 49 regional programs relevant to our interests that are approved both by our U.S. partners and by respective regional authorities in Russia. The benefits for Russia will greatly outweigh the risks of dealing with additional amounts of SNF. Further development of these regional projects, specifically, preparing business plans for them, is one of the areas where funding will soon be required. Our association has developed a strategy to finance this work. How soon this occurs depends on the international aspects of the SNF management problem. The international aspects affect virtually all investment decisions concerning importing SNF to Russia because risks and returns depend greatly on the political situation in the world.

We will focus now on international aspects of the SNF management problem. A concrete model for attracting private investment to this project is being developed by our association in cooperation with our U.S. partners, and it may be discussed in person after the talk.

International SNF problems are broken down here into two parts: the problems of nuclear superstates and the problems of other countries dealing with nuclear energy. At the end of the Cold War, the United States and Russia faced additional SNF problems with the decommissioning of unneeded nuclear weapons, removal from service of excess naval vessels and their support systems, and cleaning up of defense waste sites. The latter issue is especially keen in Russia due to its totalitarian past, which resulted in the worst nuclear contamination in the world. One additional common problem is the need to consolidate SNF to protect it against terrorist attacks. To ease this problem the Yucca Mountain repository will begin to operate in 2010. This will not, however, entirely solve the problem, as much more waste has already been accumulated in the United States than can be handled at the repository.

There are substantial differences in the U.S. and Russian approaches to the problem of SNF management. While reprocessing and recycling are not banned in Russia, they are in the United States. The reconciliation of this problem is interim storage, which defers the decision. This leaves the doors open for recycling the SNF in the future, when appropriate technologies are safe and efficient.

In other countries dealing with nuclear energy the situation is even worse than in the United States. The SNF is widely dispersed, and the related risks are high. While Russia is capable of providing valuable services to the nuclear community, dealing with many countries is a difficult and lengthy process. The good news is that 80–90 percent of all SNF in the world is virtually controlled by just one country, the United States. Therefore, U.S.-Russian cooperation is extremely important to the entire world.

The SNF problem is acute now, and what are the prospects for the future? We cannot agree with an earlier presentation in which it was argued that nothing will change substantially in the next 10 years because existing energy infrastructure is based upon fossil fuels. First, nuclear energy fits well into the existing energy infrastructure. Second, the Kyoto Protocol shows that the public wants greenhouse gas reduction, and governments usually do what the public wants. Third, a hydrogen fuel cell engine will be developed in the near future; in fact, development is near completion. There are experimental buses and cars in Vancouver driven by hydrogen. The share of renewable energy is growing tremendously. We expect the growth rate of nuclear energy production to increase dramatically as soon as economic mechanisms enforcing the Kyoto Protocol come into play. Therefore, the SNF problem will be aggravated.

SNF-related problems are broken down here into political, economic, social, and technical issues. Economic problems are briefly considered here along with major political issues.

The amounts of SNF in many countries are not sufficient to make national SNF management economically viable. The costs of qualified human and natural resources are different. Exporting SNF to Russia would enforce economies of scale and scope. The costs of qualified human and natural resources are competitive in Russia.

Global political benefits of SNF management are significant contributions to nonproliferation of nuclear weapons, increased security against terrorist attacks, adherence to the Kyoto Protocol through encouraging nuclear energy production free of greenhouse gas emissions, and reduction in the risks of radioactive contamination. Cooperation between the United States and Russia is the most effective and efficient way to address these global political, economic, and social issues.

The best way to enforce this cooperation is through private-public partnership (PPP). This means private management under the U.S.-Russian agreement plus continuous consultations and problem solving in close cooperation with the governments of both countries and their respective agencies. PPP is a blend of international security, ecological improvement, and commercial interests.

The benefits of private versus public management are well understood in the United States, and the idea of private involvement is also becoming more popular in Russia. Minister Ryumiantsev of Minatom spoke recently about the necessity of private-sector involvement in and even partial privatization of the Russian nuclear industry.

CONCLUSIONS

- U.S.-Russian SNF cooperation is highly beneficial to both countries and to the entire world.
- The importance of U.S.-Russian cooperation is important beyond any other political or economic issues, such as Iraq or Iran, and it should not be used by either the United States or Russia to affect the other party in a way that may jeopardize this cooperation.
- The leadership of nuclear superstates should come to an understanding of the precedence of this problem over other national interests of the United States and Russia.
- A team of recognized experts in politics, security, and economics should be organized to prepare a report to the top leadership of the United States and Russia that will serve as a basis for the U.S.-Russian agreement to cooperate in the area of SNF management. (Our association, along with its U.S. partner, has already started working on organizing this team and has attracted to the project Harvard and Rice universities in the United States and the University of Cambridge in the United Kingdom. To finance their work on this report as well as the respective research, private investment in this project is now required.)

Creation of an Underground Storage Facility for Spent Nuclear Fuel near the City of Zheleznogorsk (Eastern Siberia)*

Ye. B. Anderson, *Ye. F. Lyubtseva*, *V. G. Savonenkov*, *S. I. Shabalev*,
and *N. L. Alekseev*
V. G. Khlopin Radium Institute Research and Production Association

At the request of the Ministry of Atomic Energy (Minatom), research has been under way for a number years to locate a site appropriate for the construction of an underground storage facility for the long-term storage of high-level radioactive wastes from the Mining-Chemical Complex (located in the city of Zheleznogorsk, Krasnoyarsk Krai), including anticipated future wastes from the RT-2 plant as well as spent nuclear fuel that will not be reprocessed. Efforts have been conducted under a program adopted by Minatom and the Russian Academy of Sciences. This program was developed taking into account criteria established by the International Atomic Energy Agency (IAEA) and the concept for the safe isolation of radioactive wastes in geologic formations.

In order to conduct this research a team of specialists was created, and included representatives from Minatom, the Russian Academy of Sciences, St. Petersburg and Krasnoyarsk universities, and various geological organizations from Krasnoyarsk Krai (see Box 1). The Radium Institute was assigned the role of providing scientific leadership and coordination for the site search and selection efforts.

SELECTION OF THE NIZHNEKANSK GRANITOID MASSIF

The territory covered in the initial search effort included portions of three global geologic structures: the Siberian platform, the west Siberian plate, and the Altai-Sayan orogenic zone. An analysis of all information gathered on the geology, tectonics, seismic activity, natural resource utilization, and other aspects,

---

*Translated from the Russian by Kelly Robbins.

BOX 1
List of Scientific Organizations and Geological Enterprises Involved in Efforts to Identify a Site for the Safe Disposal of High-Level Radioactive Wastes from the Mining-Chemical Complex and the RT-2 Plant

Minatom organizations and enterprises

- Radium Institute
- Mining-Chemical Complex
- All-Russia Scientific Research and Design Institute for Industrial Technology
- Krasnoyarsk branch of the All-Russia Scientific Research and Design Institute for Energy Technology

Universities

- St. Petersburg State University Scientific Research Institute of the Earth's Crust
- Krasnoyarsk State University Scientific Research Physical-Technical Institute

Institutes of the Russian Academy of Sciences

- Geological Institute
- Institute of the Geology of Ore Deposits, Petrography, Mineralogy, and Geochemistry

Other organizations and enterprises

- Krasnoyarsk Branch of the Nature State Research and Production Center
- Krasnoyarsk Scientific Research Institute of Geology and Mineral Resources
- Gravimetric Expedition No. 3, Krasnoyarsk
- Krasnoyarsk Geological Survey
- Northern Geological Party of the Joint-Stock Company Yeniseigeofizika
- All-Russia Institute for Prospecting and Ore Geophysics
- Ecology and Geodynamics Research and Production Center
- Center for Electromagnetic Research

including socioeconomic and demographic data, indicated that an appropriate geologic formation should be sought within the bounds of the crystalline base of the Siberian platform among the magmatic and metamorphic rock of the southern part of the Yenisei ridge, a boundary structure for the Siberian platform that reveals its crystalline base (see Figure 1).

The concept for the site search was based on the idea that the segment of Earth's crust being sought must be distinguished by a sufficient degree of homogeneity and by minimal fracturability and permeability of adjoining rock formations. Initially about 20 potentially suitable sites were identified in the southern part of the Yenisei ridge and associated geologic structures (see Figure 2). The selection process was based on the following:

- data on the geologic and tectonic structure of the region
- gravimetric and aeromagnetic mapping materials on a 1:200,000 scale
- results of the analysis of satellite and aerial survey photos on scales ranging from 1:200,000 to 1:100,000

FIGURE 1 Location of the Nizhnekansk granitoid massif, a potential site for the disposition of high-level wastes generated by the Mining-Chemical Complex in Zheleznogorsk (Krasnoyarsk-26) and the RT-2 plant.

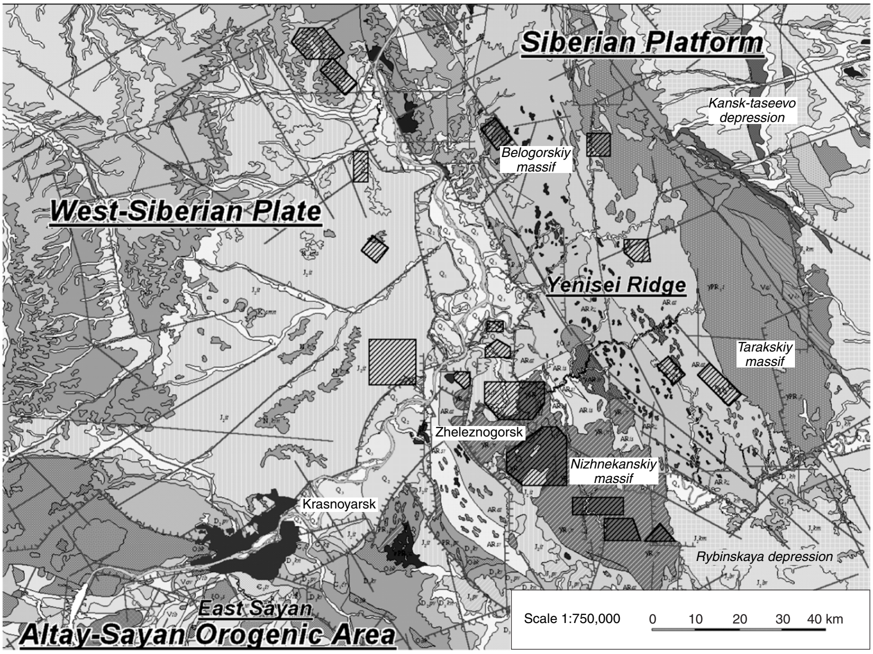


FIGURE 2 Geological structure of the territory studied and promising sites selected in the first phase (scale 1:750,000).

On the basis of specially developed search criteria—including seismic and tectonic activity of the area, dimensions of comparatively homogeneous rock formations, and distance from the Mining-Chemical Complex—the Nizhnekansk granitoid massif, one of the largest granitoid massifs in central Siberia, was selected from among all potentially suitable sites. It has an area of about 3000 km^2 and is composed of granitoids from several intrusive phases. The predominant varieties, biotite granites and granodiorites, are generally viewed as providing a favorable environment for the isolation of radioactive wastes due to various attributes, such as their high hardness, low water content, poor permeability, and low porosity. The age of the crystallization of the massif has yet to be accurately determined but lies within the boundary between the Riphean and the early Paleozoic. The central part of the massif is up to 5–6 km thick, and its northwestern boundary lies only 3 km from the Mining-Chemical Complex.

IDENTIFICATION AND STUDY OF PROMISING SITES

Along with an analysis of existing materials, a series of geological-geophysical surveys on scales of 1:100,000 to 1:25,000 were carried out to facilitate identification of promising sites within the Nizhnekansk massif. These surveys included

- a geological reconnaissance survey involving the drilling of mapping boreholes (to a depth of up to 50 m)
- hydrologic and hydrogeologic studies
- surface geophysical surveying using electric, magnetic, gravitation, and seismographic methods
- an analysis of satellite and aerial photographs
- drilling of several deep boreholes up to 700 m deep

As a result two sites in the northern part of the Nizhnekansk massif—Itatsky and Kamenny—were identified as the most promising for construction of the storage facility from among the several potentially suitable sites studied (see Figure 3). They have an area of approximately 15–20 km^2 and are located 25–30 km from the Mining-Chemical Complex. As a result of the expansion of the territory being studied using geophysical methods, two blocks of poorly permeable granitoids promising for the construction of the storage facility were identified in the northwestern and northern sections of the Itatsky and Kamenny (Itatskaya suite) sites. These blocks—Bolshoi Itatsky (12 km^2) and Maly Itatsky (14 km^2)—are separated by the mighty Bolshoi Itatsky fault zone and bounded by highly fracturable zones of geologic violations (see Figure 4).

It must be noted that the northwestern border portion of the Nizhnekansk massif, which is the closest to the Mining-Chemical Complex, is characterized by the high heterogeneity of its geologic profile, the low hardness of its granitoids, and the complex tectonic situation, all of which were confirmed by geological and geophysical investigations conducted in 2002 at the Yenisei site. Therefore, we feel that this site has little chance of being found suitable for the construction of an underground storage facility.

Special methods of geomorphological analysis based on the principle of dependence of cycles of relief formation on global climate changes have made it possible to estimate the speed of vertical movements of Earth's crust on the territory under study. Since the end of the Pliocene, about 1.5 million years ago, the southern part of the Yenisei ridge has been undergoing a slow process of elevation under calm tectonic conditions, with a maximum speed of about 1 mm per year.

From 1998 to 2001 personnel from the geological organizations of Krasnoyarsk Krai drilled a number of shallow (from 10 to 50 m) mapping boreholes as well as four deep ones (300 m, 500 m, and two 700 m boreholes) in the territory being studied. Analysis of the drilled cores and the various coring data collected revealed a complicated picture. In some cases the drilling confirmed the presence of deeply buried monolithic granitoids with a low level of fractur-

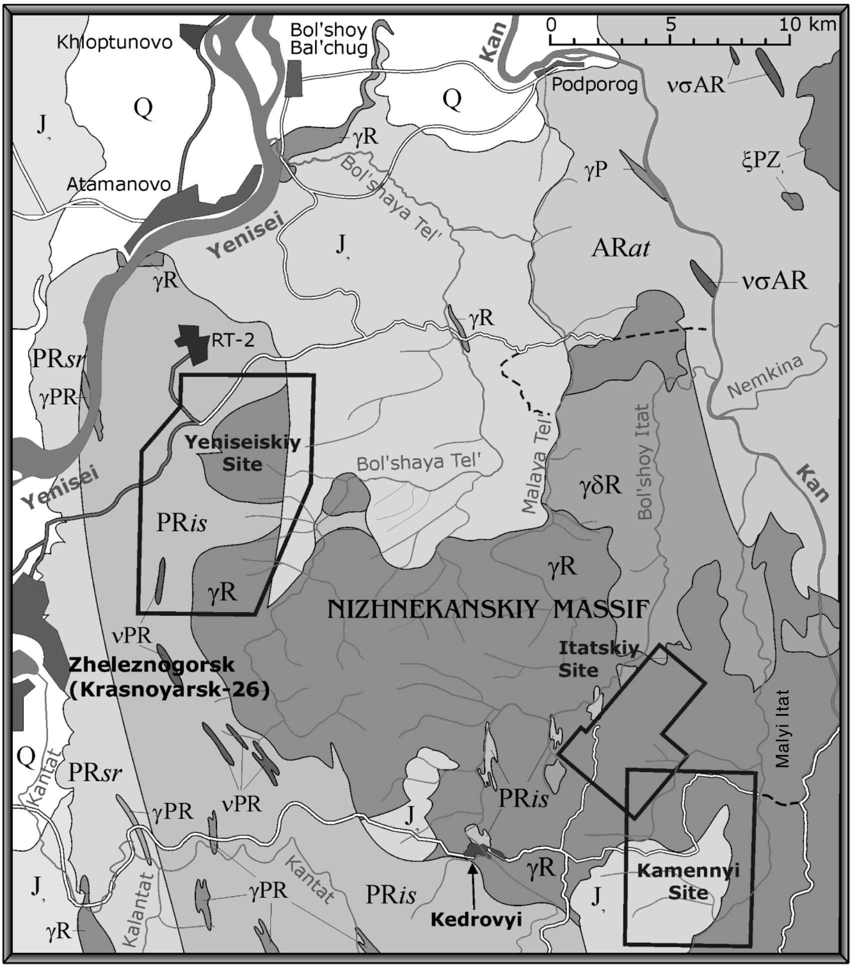


FIGURE 3 Schematic geological map of the northern part of the Nizhnekansk massif and location of the most promising sites relative to the RT-2 plant (scale 1:200,000). The most promising Itatsky and Kamenny sites, selected on the basis of reconnaissance work and tectonic structure analysis in 1:50,000 scale, along with the alternative Yenisei site.

ability water saturation, but zones of high fracturability and secondary changes were also encountered.

The Russian Academy of Sciences Institute of the Geology of Ore Deposits, Petrography, Mineralogy, and Geochemistry has conducted detailed petrographic and petrophysical studies of samples taken from drill cores 1-I and 1-K at the Nizhnekansk massif. It was established that the granitoids include zones of

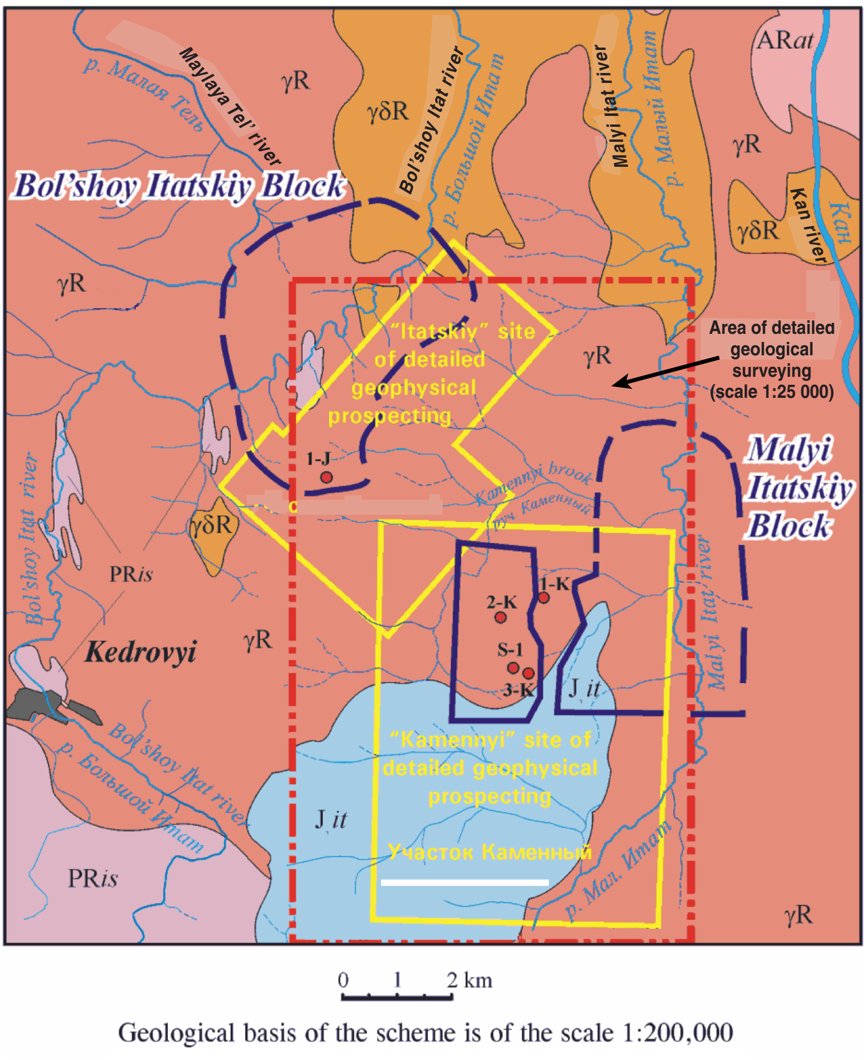


FIGURE 4 Location of the Bolshoi Itatsky and Maly Itatsky blocks in the Verkhneitatsy site (scale 1:100,000).

blastomylonite formation, cataclasm, and hydrothermal-metasomatic processing, accompanied by the crystallization of such secondary minerals as carbonates, muscovite, chlorite, and epidote. Such phenomena undoubtedly violate the homogeneity of the rock formations at the selected sites. The spatial distribution of these phenomena at the Nizhnekansk massif must be evaluated along with their connection to the fault zones, for example, and their role in the characterization of the geological environment in which the storage facility would be located. This role could be variable in nature; for example, from the standpoint of barrier properties, the presence of secondary low-temperature mineralization could, conversely, increase the sorption capacity of the granitoids. This is illustrated by several experiments that have been conducted at the Radium Institute.

The results of plutonium sorption experiments on granitoid samples with various levels of secondary changes show that the maximum sorption capacity was found in a sample of quartz diorite containing such secondary minerals as chlorite, biotite, seritsite, and carbonate. The minimum sorption capacity of all the samples studied was found in monolithic leucocratic granite, while catalyzed leucogranites occupy a middle position on the sorption spectrum (see Figure 5).

At the present stage in the process of studying potential sites, data on the deep structure of the massif have been obtained by surface geophysical methods and confirmed by direct drilling but only to a depth of 700 m. Therefore, the task

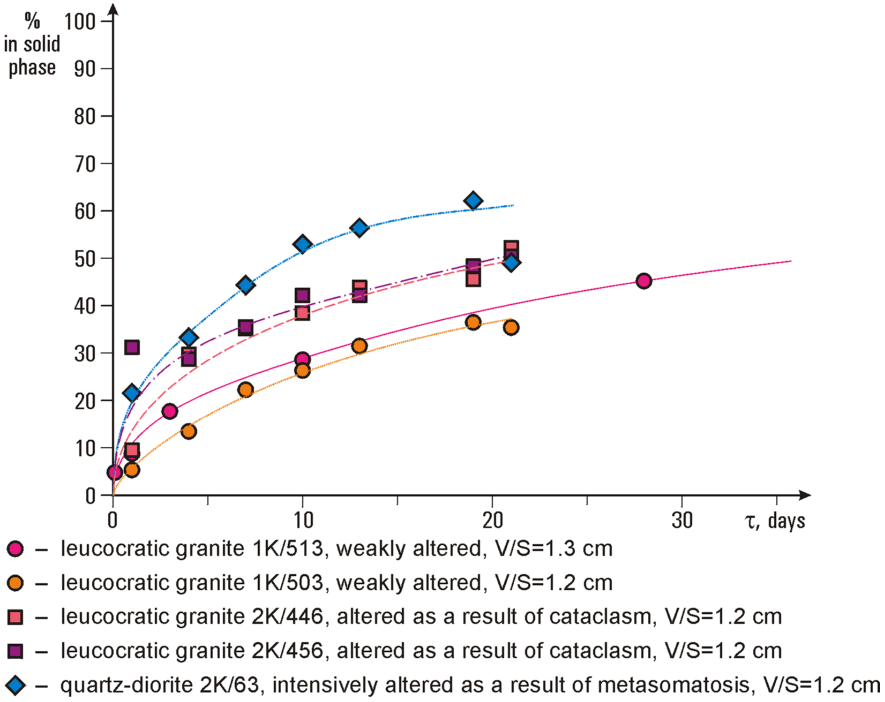


FIGURE 5 Sorption (in percentage figures) of plutonium from simulated groundwater on monolithic specimens made of granitoids of the Nizhnekansk massif depending on both time of phase contact and degree of secondary alteration.

of drilling boreholes to a depth of 2–3 km is one of the most important objectives for further study of the massif.

EVALUATION OF BARRIER PROPERTIES OF GRANITOIDS

Rocks represent the fundamental barrier to the movement of stored radionuclides into the biosphere. Therefore, great significance lies in experimental data on the permeability and radiation and heat resistance of various types of rock, as well as the restraining properties of the rock with regard to radionuclides. Special radiochemical experiments conducted at the Radium Institute demonstrate that the sorption restraint of plutonium and americium by granitoids from the

TABLE 1 Distribution Coefficients (K_d) for Long-Lived Radionuclides Between Solution and Solid Phase (Granite and Quartz Diorite from the Nizhnekansk Massif)

Radionuclide	Distribution Coefficient for Sorption (cm^3/g)	Distribution Coefficient for Desorption (cm^3/g)
Cesium	$n \cdot 10^2 - n \cdot 10^3$	$n \cdot 10^3$
Plutonium (IV)	$n \cdot 10^2 - n \cdot 10^3$	$n \cdot 10^4$
Americium (III)	$n \cdot 10^2 - n \cdot 10^3$	$n \cdot 10^4 - n \cdot 10^5$
Neptunium (V)	0.n – 2	1–2

TABLE 2 Calculated Values of Migration Rate for Long-Lived Radionuclides Through Porous Space of Granitoids of the Nizhnekansk Massif

		Rate of Migration, m/year	
Radionuclide	$K_d$eff,[a] cm^3/g	Monolithic Rock[b]	Jointy, Fractured Rock[c]
Cesium	$(5–9) \cdot 10^2$	$(4–8) \cdot 10^{-10}$	$(0.8–1.3) \cdot 10^{-7}$
Plutonium	$(7–9) \cdot 10^2$	$(6–8) \cdot 10^{-10}$	$(0.8–1) \cdot 10^{-7}$
Americium	$(1.3–3.4) \cdot 10^3$	$(1–3) \cdot 10^{-10}$	$(2–5) \cdot 10^{-8}$
Neptunium[d]	4–28	$(0.1–1) \cdot 10^{-8}$	$(0.2–2) \cdot 10^{-5}$

[a] Distribution coefficient describing the summarized process of interphase distribution for different radionuclides.

[b] Conditions: porosity = $5 \cdot 10^{-3}$, hydraulic conductivity = $3 \cdot 10^{-7}$ m/d, hydraulic gradient = $1 \cdot 10^{-2}$, actual linear rate of water migration ~$2 \cdot 10^{-4}$ m/year.

[c] Conditions: porosity $5 \cdot 10^{-2}$, hydraulic conductivity = $3 \cdot 10^{-5}$ m/d, hydraulic gradient = $1 \cdot 10^{-2}$, actual linear rate of water migration ~$2 \cdot 10^{-2}$ m/year.

[d] The rate of neptunium migration ($2 \cdot 10^{-5}$ m/year) corresponds to a migration distance of 1 m in 50,000 years.

Nizhnekansk massif is sufficiently high. This is illustrated by the high levels of the distribution coefficients for these radionuclides (see Table 1). Researchers have also detected signs of the diffusion introduction of radionuclides (Pu, Am) into several minerals, for example, monazite and zircon.

All of this substantially limits the distance that long-lived radionuclides can migrate along with underground waters in rock. Calculations have shown that the speed of radionuclide migration is extremely low, on the order of 10^{-8} m per year for monolithic rock up to 10^{-5} m per year for weakened zones (see Table 2).

Work is also under way to gather experimental data to characterize the physical-mechanical properties of granitoids of the Nizhnekansk massif.

LONG-TERM PROGNOSIS FOR THE DEVELOPMENT OF THE AREA

A number of forecast assessments have been made with regard to the geological-tectonic development of the area. These forecasts have shown the following:

- Looking at the next 10,000 to 130,000 years, there will be no change in the composition and geomorphological characteristics, orientation of tectonic pressures, average rate of tectonic elevations, and average rate of convergence along fault lines in the region under study.
- In the future the overall tectonic regime in the northern part of the massif and its separate blocks will not affect the integrity of underground repository facilities, which will be elevated to an insignificant degree, along with the block in which they are located without violating the monolithic nature of the block itself.

An analysis of paleoclimatic reconstructions give grounds to postulate that possible climatic changes over the next 50,000 years would preclude any crises that would intensify erosion processes or affect the security of an underground storage facility in the Nizhnekansk granitoid massif.

FUNDAMENTAL RESULTS AND STATUS OF EFFORTS

At present the fundamental results of research on the selection for a site for an underground storage facility in the Krasnoyarsk area are as follows:

- The Nizhnekansk granitoid massif has been selected as a geological formation suitable for the creation of an underground facility, and scientific grounds for this selection have been documented.
- Two promising sites for the construction of an underground facility in the Nizhnekansk granitoid massif have been selected, and scientific grounds for this

selection have been documented on a step-by-step basis with the help of a range of geological and geophysical methods.

- Two relatively monolithic blocks of rock with an area of 12–15 km^2 have been identified.
- Researchers have accumulated a significant volume of data necessary for subsequent engineering and geological work on the project with regard to construction of an underground research laboratory and later construction of the storage facility itself.

Based on data that has been gathered, an area encompassing the Kamenny and Itatsky sites has been announced as the site of more detailed preliminary prospecting work for the construction of an underground laboratory and the future waste burial site, and relevant materials have been submitted to the All-Russia Scientific Research and Design Institute for Industrial Technology in Moscow.

Because of a lack of financing, work on the study of the Kamenny and Itatsky sites is currently halted. However, in our opinion these sites, like the Nizhnekansk massif as a whole, remain insufficiently studied. Following is a list of the fundamental unresolved problems:

- insufficient information on the geological structure of the massif and individual sites; no direct and unambiguous data on such fundamental characteristics as the strength of the granitoids, for example
- no identification and study of fault, fracturability, and cataclasm zones and radiogeochemical and other characteristics of the main migration channels
- insufficient assessment in barrier property studies of the role of secondary changes as a factor that could play both a positive and a negative role with regard to facility security
- very little data on deep geological conditions and on the regime for transit and unloading of underground waters and gases
- practically complete lack of isotopic geochemical research that would provide unique information on the condition of the lithospheric environment
- lack of long-term geological-tectonic and geoecological forecasts regarding the area

Given the above points, it seems necessary (if financing is available) to continue comprehensive geological, hydrogeological, geophysical, and geochemical research aimed at providing a scientific basis for the selection of a specific site for the construction of an underground laboratory and subsequently the storage facility itself.

Conditions for the Creation of an International Spent Nuclear Fuel Storage Facility near the Priargunsk Mining-Chemical Production Association (City of Krasnokamensk, Chita Oblast)*

Vasily I. Velichkin,[†] *V. A. Petrov*,[†] *V. F. Golovin*,[‡] and *V. A. Ovseichuk*[‡]

Ensuring the safe isolation of the previously accumulated and regularly created stores of spent nuclear fuel is one of the serious radioecological problems facing Russia. Unresolved questions regarding the problem of spent fuel isolation also exist in other countries that use electricity produced by atomic power plants.

At present, spent fuel storage facilities at many Russian atomic power plants and nuclear fleet bases in the country's northwest and east are filled to planned capacity or nearly so. The need to construct new spent fuel storage facilities, specifically those that would facilitate the storage of spent fuel using the most environmentally acceptable methods, is one of the most urgent tasks facing Russia's Ministry of Atomic Energy (Minatom). An additional stimulus for the acceleration of practical efforts to address this problem also lies in the passage last year of legislative acts allowing the import and storage in Russia of spent nuclear fuel from foreign countries, as well as the significant economic benefits that Russia might gain from accepting and storing foreign spent fuel on its territory.

Located in the eastern Baikal region (southeastern Chita Oblast, Krasnokamensk Region), the Priargunsk Mining-Chemical Production Association (PM-

---

*Translated from the Russian by Kelly Robbins.

[†] Institute of the Geology of Ore Deposits, Petrography, Mineralogy, and Geochemistry of the Russian Academy of Sciences.

[‡] Priargunsk Mining-Chemical Production Association of the Russian Federation Ministry of Atomic Energy (Minatom).

CPA) is Russia's only enterprise for the extraction and processing of uranium ores. By virtue of its geographic location, natural and geologic characteristics, and economic and technical capabilities, PMCPA should be considered a promising site for the creation of a spent fuel storage facility that might be given international status. The type of facility and its capacity would be determined when and if the site is selected.

PMCPA processes molybdenum-uranium ores from deposits in the Streltsovskoe ore field. These deposits are concentrated in the Tulukuev volcano-tectonic caldera, which dates from the Mesozoic era (approximately 140 million years ago) and has an area of about 150 km^2. A total of 19 molybdenum-uranium deposits have been found in close proximity in the caldera, and these deposits contain unique sources of uranium totaling more than 250,000 metric tons.[1] About 50 percent of the uranium resources of the Streltsovskoe ore field have been extracted to date.

GEOGRAPHIC SITUATION, NATURAL CONDITIONS, AND GEOLOGIC CHARACTERISTICS OF THE PMCPA REGION

The region where the enterprise is located is in a lightly populated area at a significant distance from major cities, villages, and industrial centers but at the same time is linked to other economically developed regions of Russia by the rail and highway networks.

The climate of the region is arid, with precipitation levels totaling slightly more than 400 mm/yr. Lying in the steppe zone, the landscape surface is very open, with low hills and absolute elevations from 600 to between 900 and 1100 m (see Figure 1). The underground water table lies at 500 to 700 m, and the territory is categorized as slightly seismic.

Another characteristic of the PMCPA region is the fact that it has been the subject of detailed geological studies carried out in the course of many years of large-scale, comprehensive prospecting efforts, which have included geological and geophysical studies, enormous volumes of test drilling and underground mining operations, and other extensive scientific research work. The many years of mining operations that continue to this day represent a significant source of geological information.

Two separate rock formations differing in age and structural and compositional characteristics are found on the territory of the PMCPA complex.[2] The upper formation is composed of volcanogenic and terrigenous-carbonate sedimentary rock dating from the Mesozoic. The volcanogenic rocks (rhyolites, dacites, basalts, tuffs, and tuffaceous sandstones) make up the Tulukuev volcanic caldera and surround all of the extractable molybdenum-uranium and uranium deposits. These rocks are intensively tectonically violated, have been affected at various times by hydrothermal processes, and as a result are mechani-

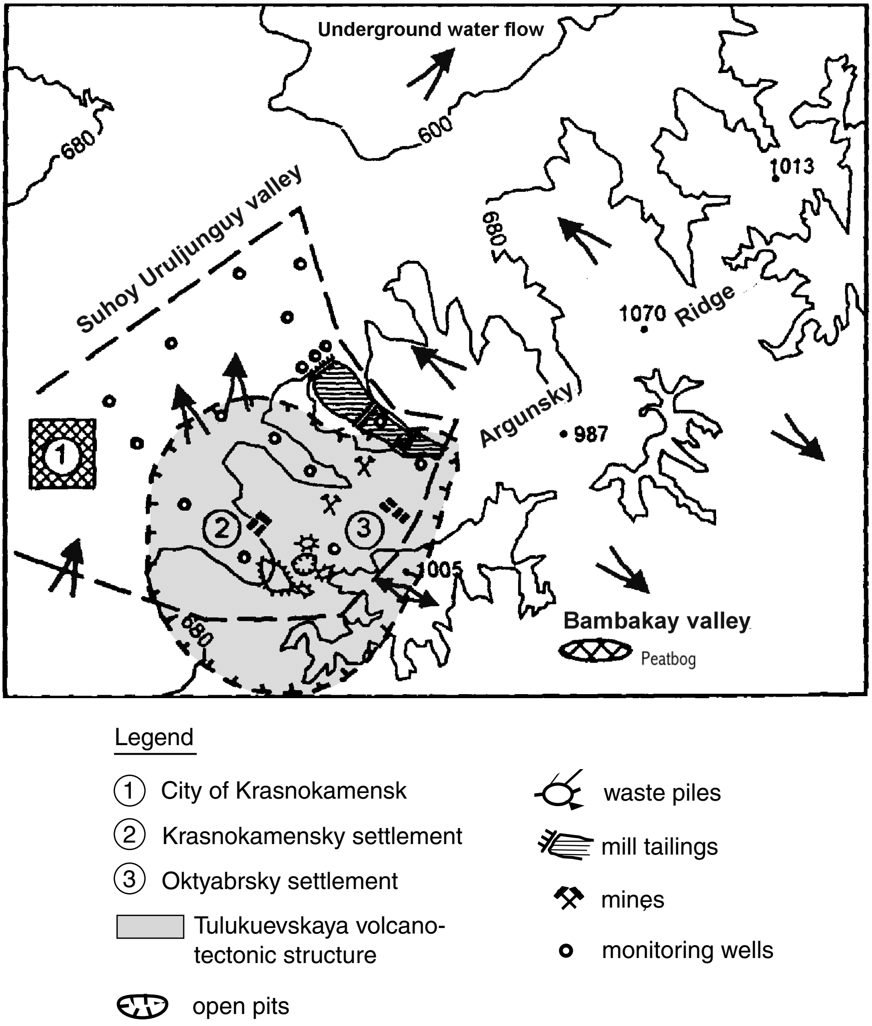


FIGURE 1 Topographic map of the region.

cally weak. For these reasons they are not suitable for underground storage of radioactive materials. The same holds true for Mesozoic sedimentary rocks. Also having insufficient mechanical toughness, they underlie the lower relief elevations and are water saturated.

The lower rock formation is made up of pre-Paleozoic gneisses, crystalline shales and marbles, and Paleozoic gneissose and normal granites. In unviolated condition these rocks are thick, massive, and poorly permeable with high hardness properties. They generally underlie elevated portions of the landscape. In the overall massif the task is to locate and study weakly violated geological blocks of the required size and then install in the most suitable block all the necessary facilities and support structures for an underground spent fuel storage facility.

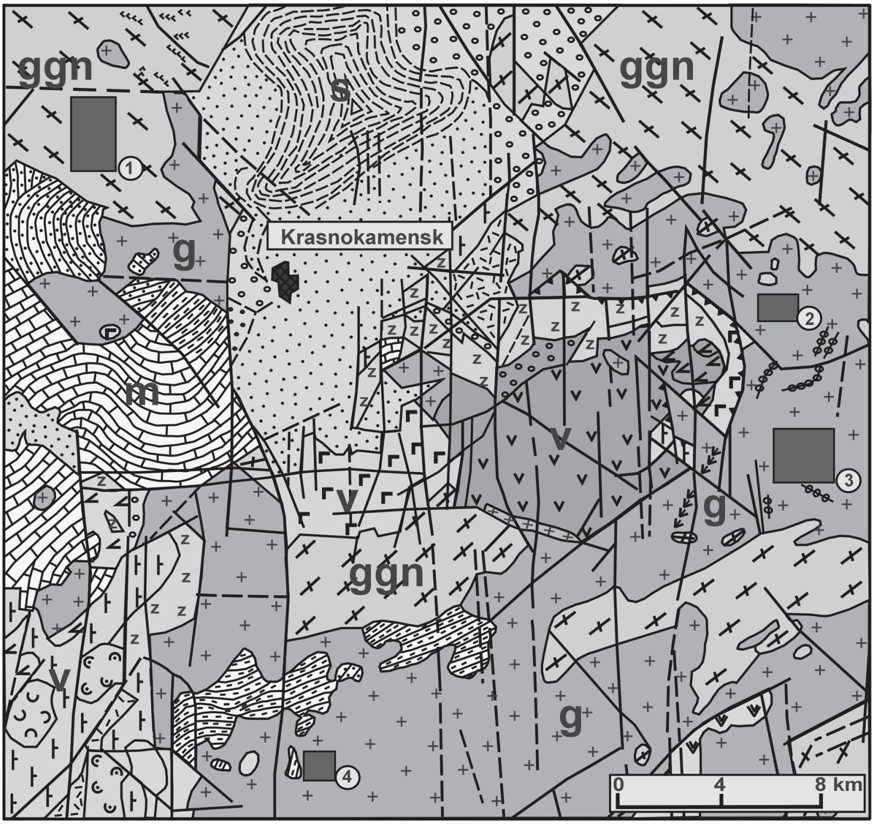
DESIGN FEATURES OF THE FUTURE REPOSITORY AND PROMISING SITES FOR ITS CONSTRUCTION

Given the characteristics of the surface landscape and the environmental, technical, and economic conditions as well as security considerations, a gallery-type underground repository appears to be the most suitable for the reliable isolation of spent nuclear fuel under the conditions prevalent at PMCPA. The following conclusions support the construction of this sort of facility in the PMCPA area:

- The location of the future facility in a rock formation having about 300 m of solid rock covering the site where the nuclear materials will be stored drastically cuts the risk of any uncontrolled outside effects on the stored materials.
- The high hardness, low permeability, and low degree of violation of the crystalline rocks that will contain the repository sharply minimize the danger of destruction or penetration into underground waters.
- In comparison with the mineshaft method of storage, the gallery-type repository eliminates the need for building underground installations and water drainage mechanisms while also significantly simplifying ventilation.

Preliminary work done by specialists from PMCPA and the Institute of the Geology of Ore Deposits, Petrography, Mineralogy, and Geochemistry of the Russian Academy of Sciences with regard to possible options for the location of the future repository have made it possible to identify four sites in the lower rock formation (see Figure 2) that occupy a position in the relief that would be suitable for the construction of a gallery-type storage facility. The most acceptable option appears to be Site 1 (see Figure 3), which is composed of weakly violated granitoid gneisses and granites and is located very close to the rail line. Some samples of the stone material have been taken in order to characterize the properties of the geological environment at this site, and these studies are already under way.

The above option for the future construction of a gallery-type spent fuel facility is fairly economical and meets the strictest requirements with regard to security against outside effects. The placement of isolated spent fuel above the underground water level—that is, in the aeration zone, which is characterized by oxidative conditions—is not a favorable factor for long-term (in this case, longer

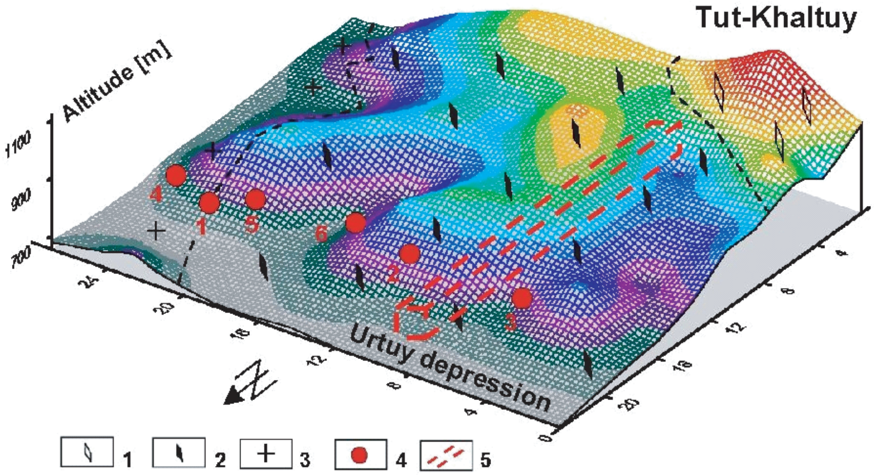


Legend	
ggn	Caledonian gneissic granites
g	Variscian biotitic granites
m	Paleozoic metamorphic rocks
v	Mesozoic volcanic rocks
s	Mesozoic sedimentary rocks

FIGURE 2 Location of the granitic potential sites denoted by numbers.

than 200 years) spent fuel storage. Consequently, the gallery-type facility option could be used only for short-term (in this case, 30–100 years) or at most medium-term (in this case, 100–200 years) storage of spent nuclear fuel.

It is commonly known that the safe long-term storage or burial of nuclear materials is ensured only by means of their complete isolation from the hydrosphere or if the repository is located in a water-saturated zone, by the restorative properties of the underground waters circulating in that zone.



Legend

1	Caledonian fine-grained gneissic granite
2	Caledonian medium-grained gneissic granite
3	Variscian medium-grained biotitic granite
4	Sampling points
5	Gallery outline

FIGURE 3 Three-dimensional image of Site 1.

Given the specific geological situation and landscape characteristics of the PMCPA region, there is also another option for spent fuel isolation that would make it possible to ensure the environmental security of this material both in the short and medium term and for a very long period, comparable to the duration of the radiobiological toxicity of the spent fuel to be isolated. This option also involves the use of gallery-type facilities, which could be not only outfitted with structural features for the short- and medium-term storage of spent nuclear fuel but also provided with special mechanical equipment for the drilling of a "blind" shaft or a spiral descending tunnel. These structures could be used to create a facility for the long-term storage of spent fuel or the burial of high-level radioactive wastes at a depth necessary to meet safety requirements. The option for a repository with a spiral tunnel has been discussed with U.S. specialists from the National Research Council's Board on Radioactive Waste Management (Milton Levenson and Charles McCombie), but this option is no longer under consideration.

STAFF, TECHNICAL, AND ECONOMIC CAPABILITIES OF PMCPA AND THE SOCIAL SITUATION IN THE REGION

In its more than 30 years of production activities PMCPA has amassed a great deal of experience in managing radioactive wastes and conducting various types of drilling, tunneling, mine construction, and extraction operations. The enterprise successfully operates special services staffed with highly professional specialists working at the modern standard to handle all matters associated with mining, drilling, and extraction operations. Production activities in these areas as well as efforts to study the geology and uranium content of the Streltsovskoe ore field and specific deposits have always been accompanied by intensive scientific research efforts. Specialists from the leading institutes of the Russian Academy of Sciences and applied research institutes of Minatom and the Russian Ministry for Natural Resources have participated in these efforts, along with enterprise personnel.

The multifaceted research and production activities carried out at PMCPA have given rise to a unified team made up of the leading institute-based scientists and professional production specialists capable of carrying out in a highly qualified and timely fashion any tasks that might arise with regard to the selection of a site for an underground spent fuel repository and its subsequent construction. PMCPA also possesses the equipment necessary for the tunneling and drilling work.

Along with the mines, the quarry, and the uranium processing and sulfuric acid plants, the production infrastructure of the enterprise includes an open-pit coal mine, a coal-fired heating plant used to generate steam and electricity, a mechanical repair and production facility, and a concrete plant (see Figure 4). The energy capacities of the existing power plant fully meet the enterprise's current electricity needs and are capable of covering additional power requirements that might arise if efforts to build an underground spent fuel repository are launched.

The population in the area around PMCPA—made up of employees of the enterprise, service workers, and their families—lives in the city of Krasnokamensk (with a total of about 60,000 residents) and the villages of Oktyabrsky and Krasnokamensky. According to a survey[3] most of the region's residents are prepared to support a decision to create an international spent fuel repository on the territory belonging to the enterprise.

CONCLUSIONS

1. The problem of safely isolating spent nuclear fuel is an urgent one both for Russia and for other countries that use electricity produced by nuclear power plants.

2. The territory surrounding PMCPA, Russia's only uranium-processing

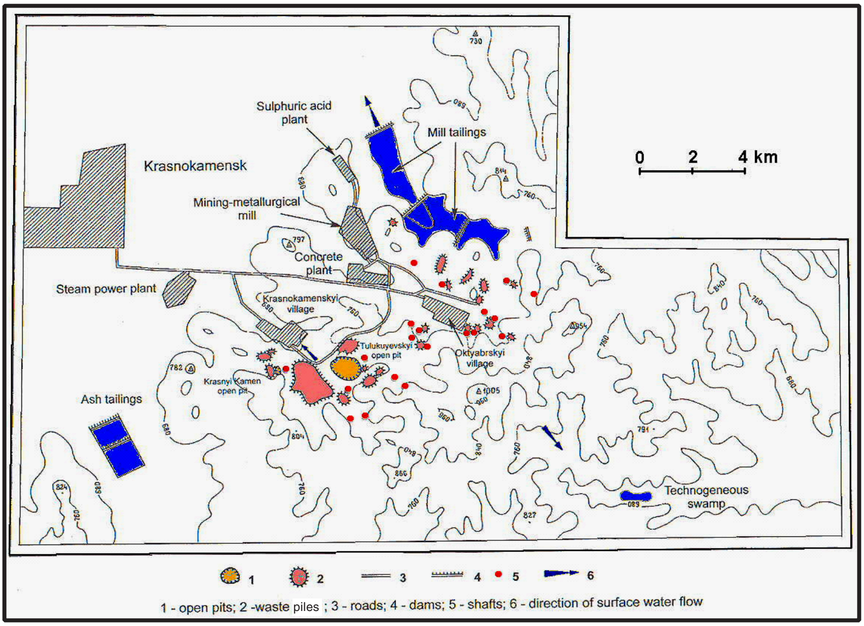


FIGURE 4 Mining and production activity area at PMCPA and main sources of environmental contamination.

enterprise, appears to be a promising site for a future international spent fuel storage site. PMCPA is located in a lightly populated area in the eastern Baikal region but is linked by rail to the country’s major industrial regions. The enterprise possesses a comprehensive and diversified industrial and energy infrastructure that allows it to resolve the most complex mine engineering and energy problems on its own.

3. PMCPA has assembled a team of highly professional specialists with enormous experience in managing radioactive substances and conducting various drilling, tunneling, and mine construction operations.

4. All parameters characterizing the natural and geological conditions in the PMCPA region are assessed as favorable for the creation of an underground repository. Four sites viewed as promising for the placement of an underground high-level waste storage facility have already been identified in the thick, hard, and poorly permeable rock formations (granitoid gneisses, granites, shales) adjoining most of the enterprise’s grounds.

5. Taking into account the special characteristics of the environment; ecological, technical, and economic considerations; and the demands of security, an underground storage facility that would be located in gallery-type horizontal

underground structures would be the most acceptable option for the reliable isolation of spent nuclear fuel at the PMCPA site.

6. Most of the workforce in the region does not suffer from radiophobia and is prepared to support the decision to create an international spent fuel storage facility on the grounds of the enterprise.

NOTES

1. S. S. Naumov. Reassessment of the status of Russia's uranium resources after the breakup of the USSR and of the direction of geological prospecting efforts in the near term. Prospecting and Protection of the Earth's Underground Resources 3, 1996.

2. L. P. Ishchukova, et al. Geology of the Urulyunguev Ore Region and the Molybdenum-Uranium Deposits of the Streltsovskoe Ore Field. Moscow: Geoinformmark, 1998.

3. Within the framework of a public opinion poll in the Krasnokamensk region, 7063 bulletins were distributed among people who have had permanent work, of which 4021 people (56.9 percent) voted in favor of international spent nuclear fuel storage facility construction. Bulletins were also distributed among 405 people who have had no permanent work, of which 170 people (41.2 percent) voted in favor of storage facility construction. Vasily Velichkin, July 12, 2004.

UTILIZATION OF HIGH-LEVEL WASTE

Types of High-Level Radioactive Wastes Formed as a Result of Dry Methods of Spent Fuel Regeneration and Technologies for Their Management

Valentin B. Ivanov
Institute of Geology of Ore Deposits, Petrography, Mineralogy, and Geochemistry
Russian Academy of Sciences

At present for the nuclear industry to adopt new nuclear fuel cycle technologies and for public opinion to be prepared to accept their implementation, it is absolutely essential that all fuel cycle-related problems without exception be resolved, especially those problems involving the management of radioactive wastes. In proposing new approaches to spent fuel regeneration, suppliers of such technologies must ensure that all aspects of the separations process are comprehensively addressed. This sort of approach was taken in the development and testing of dry technologies for spent fuel regeneration. The pyroelectrochemical technology, which is based on the use of salt melts, is the most ready for industrial application, and has been developed and tested on a semi-industrial scale at the Scientific Research Institute of Atomic Reactors (NIIAR).

A top priority in any technology used in the nuclear fuel cycle is its safety, and this means not only nuclear and radiation safety but also general industry safety as well. A safety analysis has shown the fundamental advantages of the pyroelectrochemical regeneration technology in comparison with currently used water-based methods for reprocessing spent nuclear fuel. The basic safety aspects and physical-chemical properties of the process on which it is based are presented in Box 1. This technological process is also notable for the fact that it is self-protected against the unauthorized removal of fissile materials, as it is characterized by the presence of powerful gamma radiation in all its stages. Therefore, an additional requirement for this type of regeneration process is that it should facilitate the separation of the greatest possible quantity of fissile materials with the least possible removal of fission products. Such an approach also minimizes the amount of radioactive materials, and especially radioactive wastes, that are removed from the fuel cycle.

BOX 1
Analysis of the Safety of Nonwater-Based Processes for Spent Fuel Regeneration

- **Key safety aspects:** does not use water or organic solvents; all chemical operations conducted in one closed apparatus; reprocessed product is practically ready for use after removal from apparatus; high-level wastes are in solid compact form
- **Radiation safety:** crystalline product base; operational environment is a molten salt mixture
- **Nuclear safety:** no moderators or neutron reflectors; processes take place discretely; no possibility of self-sustaining chain reaction in the event of serious accidents
- **Technical and chemical safety:** no radiolysis of the chemical environment; no pieces of equipment vulnerable to fire; explosive gases and substances not used
- **Protection against dispersion of radiation:** placement in protective chambers; automated and remote controls; chemical stability of regenerated product

BOX 2
Composition of Fuel Consisting of Plutonium Extracted During Reprocessing of VVER-440 Spent Fuel and Combined Uranium

Plutonium oxide with the following isotopic composition:

Pu-238	1.0%
Pu-239	67.4%
Pu-240	21.5%
Pu-241	6.9%
Pu-242	3.1%

Am-241 in an amount equaling 2.8 kg per metric ton of plutonium

Remainder of fuel load: combined U-238 in dioxide form and 5–10 percent in metal form, U-235 less than 0.2 percent of the total mass of uranium

To illustrate, I present the following data on the composition of plutonium-based fuel separated from spent fuel from a water-moderated water-cooled power reactor (VVER-440), as well as experimental data obtained as a result of the regeneration of spent fuel removed from a BOR-60 fast reactor, with this material being analyzed for its potential fuel content to be reloaded into a reactor (see Boxes 2, 3, and 4). It is obvious that after regeneration, the fuel contains not only fission products but also minor actinides. These elements do not hinder the phys-

BOX 3
Composition of Fuel Obtained Through Pyroelectrochemical Regeneration of BOR-60 Spent Fuel (Approximately 28 Percent Burn-Up Rate)

Plutonium oxide and uranium oxide with the following isotopic composition:

Pu-238	1.06%	U-234	0.65%
Pu-239	63.97%	U-235	48.71%
Pu-240	26.04%	U-236	3.84%
Pu-241	4.72%	U-238	46.80%
Pu-242	4.21%	(For BN-600/800 reactors only U-238)	

Am-241 in an amount equaling 5.4 kg per metric ton of plutonium and 0.376 kg per metric ton of uranium; the regenerated uranium output also includes Cm-242 (1.2 g per metric ton) and Cm-244 (14 g per metric ton)

Fission products (Curies per gram of PuO_2):

Ru(Rh)-106	$1.3 \cdot 10^{-1}$	Sb-125	$2.1 \cdot 10^{-3}$
Cs-134	$1.9 \cdot 10^{-5}$	Cs-137	$2.0 \cdot 10^{-4}$
Ce(Pr)-144	$5.3 \cdot 10^{-2}$	Eu-152	$1.4 \cdot 10^{-3}$
Eu-154	$5.8 \cdot 10^{-4}$	Eu-155	$2.0 \cdot 10^{-3}$

Corrosion elements: Mn-54 at $5.9 \cdot 10^{-5}$ and Co-60 at $3.2 \cdot 10^{-6}$ Curies per gram of PuO_2

Cation additives 8.86 mass percent of the mass of PuO_2 and by nomenclature Si, Fe, Mg, Cr, Zr, Mo, Gd, Ni, Pb, Ag, Y, La, Tb, Dy, B, Ga, Be, Ca, Ce, Pr, Eu, Ti, Cu, Na, Pd, Nd, Sm, Sc

BOX 4
Composition of Fuel Obtained Through Pyroelectrochemical Regeneration of Spent Fuel from VVER-1000 or RBMK Reactors

Left after regeneration:

U	99.9%	Pu	99.96%

Am and Cm in approximately the same quantity per kg as in fuel prepared on the basis of spent fuel from BOR-60 reactors

Standard purification coefficients for fission products:

Ru-106	13	Ce-144	19
Sb-125	120	Eu-154 and -155	33
Cs-137	30,000		

Obviously a substantial part of the fission products and minor actinides remains in the fuel to be reloaded into the active zone of the reactor.

ics of the reactor's operation, that is, they may pass through the cycle repeatedly without increasing the volumes and radioactivity of the materials removed. This is a great advantage of this technology, inasmuch as with the use of remote-controlled transport procedures this fulfills the conditions for self-protection of the fuel and reduces the quantity and activity of the radioactive substances removed from the cycle.

Dry regeneration also results in the formation of radioactive wastes. The sources and forms of these wastes are presented in Box 5. These types of wastes are subjected to special processing so that they may be placed either immediately or after a certain holding period into deep geological formations for permanent storage. Thus, enterprises manufacturing regenerated fuel for the new generation of fast neutron reactors will not accumulate radioactive wastes. The technologies and procedures for primary processing of radioactive wastes formed during the pyrochemical regeneration of spent fuel are shown in Box 6.

BOX 5
Sources of Radioactive Wastes Created During Pyroelectrochemical Regeneration of Spent Nuclear Fuel

- **Liquid wastes:** soda solution (gamma and alpha nuclides suitable for underground burial in liquid form); water solutions after flushing of precipitates (after steaming, the salts are ready to be returned to the start of the process)
- **Gaseous wastes:** technical gases from the chlorinator-electrolyzer (radioactivity 95 percent due to Sb-125 aerosols; gases are purified with a filter, absorption column, and two stages of treatment); air from protective chambers
- **Solid radioactive wastes and products:** nontechnical (equipment, pyrographite items, filters, fuel rod casings); technical (spent electrolyte, steamed salts, phosphate precipitate)

BOX 6
Primary Processing of High-Level Radioactive Wastes

1. Concentration of fission products and additives in a phosphate precipitate and a spent salt electrolyte
2. Vitrification of the phosphate precipitate in aluminofluorophosphate glass
3. Vitrification of the phosphate precipitate in aluminofluorophosphate glass along with the spent electrolyte
4. Conversion of the phosphate precipitate into a monazite-like structure and conversion of the spent electrolyte into a ceramic

The specific characteristics of high-level radioactive wastes formed during the regeneration of spent fuel with a burn-up rate of 21 percent of the heavy atoms are shown in Table 1. It should be noted that the output volume of contaminated electrolyte, 2.5 kg/kg, is valid only for this particular experiment. In real-life application of the technology the electrolyte undergoes regeneration and the output volume that must be disposed of is on the order of 10 times less.

One method of disposing of the wastes that are created is vitrification. This procedure is carried out directly with the separated product without any intermediate stages. The characteristics of the glass matrices that are produced are presented in Table 2. The high radiation resistance of the matrix and the low rate of leaching are particularly noteworthy. When wastes are placed in a ceramic form, the leaching rate is slightly accelerated, but this process provides a substantially greater degree of thermal stability, which is especially important in the regeneration of spent fuel that has been stored for a relatively short period of time (see Table 3).

The technology for radioactive waste reprocessing is illustrated in Figure 1. As the schematic clearly shows, the products separated out by the process undergo only two or three operations, and the wastes are then ready for burial.

A full listing of the various types of wastes and their isotopic composition is provided in Table 4. One can see that the bulk of the radioactivity is concentrated in the phosphate precipitate (Ce(Pr)-144), the spent electrolyte, and the steamed salts (Cs-134 and -137). All of these substances are contained in the same chlorinator-electrolyzer device both during the regeneration process and after its completion. They are not transported and consequently do not contaminate any other equipment or hot chambers. All of this is very significant for reducing the volume of radioactive wastes created during the decontamination process.

The fluoride gas technology for regenerating spent nuclear fuel is less ready for industrial application. Nevertheless, a certain amount of research has been completed with regard to the management of radioactive wastes created as a result of this process. In regenerating only uranium irradiated fuel, an average of 86

TABLE 1 Characteristics of Solid High-Level Wastes (Spent Fuel from BOR-60 Reactors with a Burn-up Rate of 21 Percent of Heavy Atoms, Stored for Two Years)

Type	Fuel Output, kg/kg	Specific Heat, Wt/kg	Temperature of Spontaneous Combustion, °C
Phosphate precipitate	0.14	14.8	40 (180 g)
Electrolyte	2.5	0.95	30 (2 kg)

TABLE 2 Vitrification of High-Level Wastes (HLW) Using the Pyroelectrochemical Process

Type of HLW	Type of Glass Matrix	Means of Incorporation
Phosphate precipitate	$Pb(PO_3)_2$, $NaPO_3$	Vitrification, T-950°C
Spent salt electrolyte	$NaPO_3$, AlF_3, Al_2O_3	Vitrification without conversion of chlorides, T-950°C
Phosphate precipitate plus spent salt electrolyte	$NaPO_3$, AlF_3, Al_2O_3	Joint vitrification without conversion of chlorides, T-950°C

TABLE 3 Ceramization of High-Level Wastes (HLW) Using the Pyroelectrochemical Process

Type of HLW	Type of Ceramic	Means of Incorporation
Phosphate precipitate	Monazite	Pressing, kilning, T-850°C
Spent salt electrolyte	Kosnarite (sodium zirconium phosphate —NZP)	Conversion into NZP from melt or water solution, pressing, kilning, T-950°C

TABLE 4 Solid Technical Wastes and Products

Waste Product	Mass, kg	Mn-54	Co-60
UO_2-1 product after electrolysis of restored melt	0.489	< 4.4	< 1.6
UO_2-2 product after electrolysis of acidified melt	1.510	0.3	0.5
PuO_2 product after precipitation crystallization	0.504	2.2	0.1
Phosphate precipitate	0.442	24.8	1.3
Spent electrolyte	8.114	—	0.1
Steamed salts	0.968	2.1	0.4
Sublimates	0.495	traces	traces
Pyrographite materials, filters, fuel rod casings	20	0.8	—

Amount of Wastes Incorporated (percent)	Cs-137 Leaching Rate Over 7 Days, g/cm^2/day	Thermal Stability, °C	Radiation Resistance
28	$7 \cdot 10^{-6}$	400	
20	$7 \cdot 10^{-6}$	400	$1 \cdot 10^{7}$ Gr (γ, β) $1 \cdot 10^{18}$ α emitters/g
36	$4 \cdot 10^{-6}$	400	

Amount of Wastes Incorporated (percent)	Cs-137 Leaching Rate Over 7 Days, g/cm^2/day	Thermal Stability, °C	Radiation Stability
100	$1 \cdot 10^{-6}$	850	$5 \cdot 10^{8}$ grays
30–40	$3 \cdot 10^{-6}$	1,000	(γ, β) $1 \cdot 10^{19}$ α emitters/g

Specific Activity of Nuclides, GBq/kg on June 1, 1995						
Ru(Rh)-106	Sb-125	Cs-134	Cs-137	Ce(Pr)-144	Eu-154	Eu-155
40,700	362.6	0.4	4.1	196.1	2.9	4.8
814	35.2	0.3	3	70.3	51.8	136.9
4,810	77.7	0.7	7.4	1,961	21.5	74
151.7	229.4	16.7	170.2	96,200	888	4,440
2	1.1	207.2	2,072	244.2	6.3	—
777	74	170	1,369	2,664	21.8	144
48.1	484.7	26.3	263	29.6	traces	—
21	10.5	4.5	44.5	15.4	0.2	—

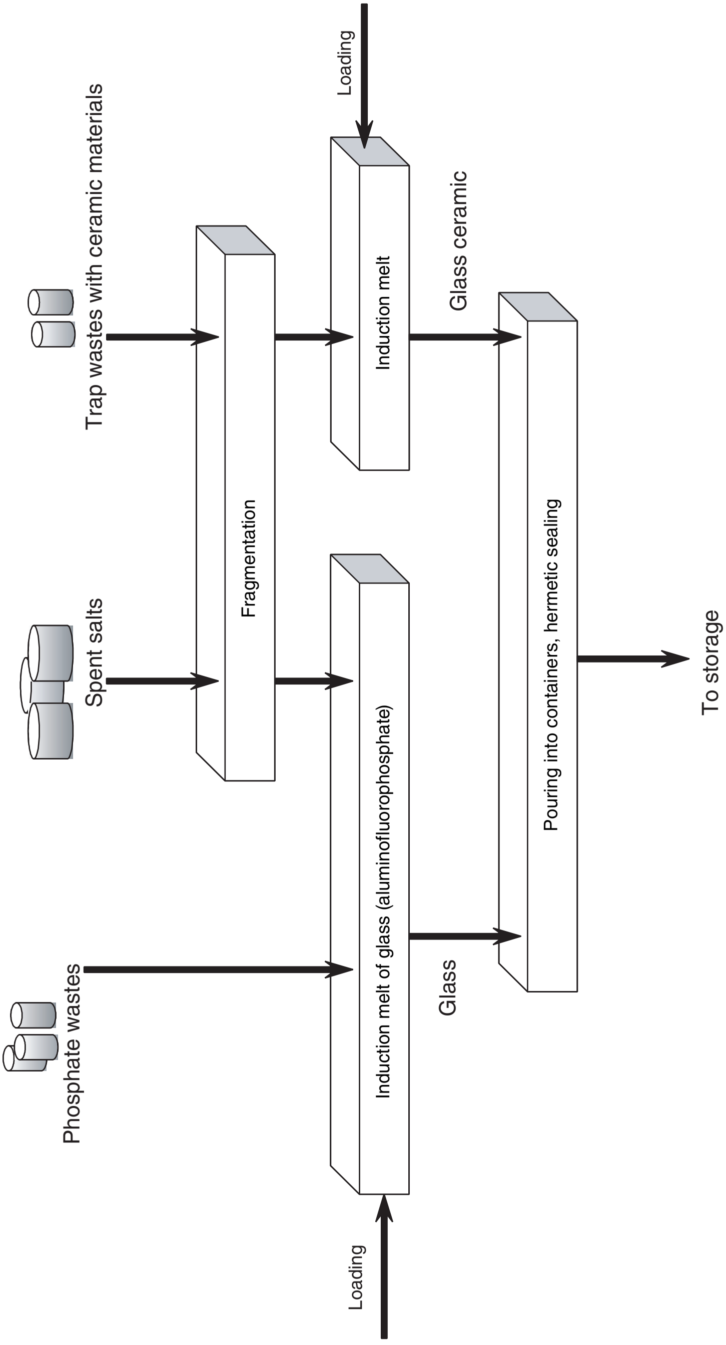


FIGURE 1 Vitrification of wastes and preparation of glass ceramics.

BOX 7
Preparation of Solid Wastes with High Specific Activity

Spent fuel and sorbent materials are first subjected to a dosing procedure and then smelted in a crucible at a temperature of up to 800°C. Then they are poured into stainless steel containers, which are hermetically sealed and inspected. The outside surfaces of the containers are decontaminated. After this process, the wastes are characterized by

- adequate radiation stability (no gas separation observed in a container of wastes up to a dose of $4.4 \cdot 10^5$ grays)
- thermal stability (depending on the proportions of components loaded, the melt temperature varies from 760 to 800°C)
- chemical stability (leaching rate for Cs-137 from melts of various compositions with high specific activity equals $(1.6 – 8.8) \cdot 10^{-2}$ g/cm^2 per day)

percent of the activity of the fission products is concentrated in the fluorination residues, whereas in regenerating mixed fuel this figure is only 53 percent, which is explained by the characteristics of the technical process of fluorination (increased temperature, large surplus of fluorine). The remainder of the activity is distributed among the chemical and other sorbents and the various tubes and pipes in the processing equipment. The output of fluorination residues totals about 0.2 kg/kg of spent fuel processed, with these residues showing concentrations of mainly long-lived radionuclides (cerium-144, praseodymium-144, cesium-134 and -137, and strontium-90) that form relatively nonvolatile fluorides.

The stability of the fluorination residues depends on the activity of the fuel being regenerated and increases over time. All of the solid wastes are highly active powders (fluorination residues) or granules (sorbents), and they require special storage technology. All of these materials can be safely stored without any special preparation in stainless steel containers; however, for long-term controlled storage a technology has been developed for the preliminary smelting of solid wastes. Box 7 describes the process for preparing solid wastes with high specific activity and presents information on their fundamental properties.

The release of gas-phase Cs-137 from the wastes increases along with increases in temperature; however, the rate of its volatilization is lower than that for the sorbents and spent fuel wastes separately (that is, the figures go from $1.3 \cdot 10^{-4}$ to $1.8 \cdot 10^{-10}$ g/cm^2 per day). The heat conductivity of the smelted wastes in the temperature range from 100 to 400°C totaled 0.51 to 0.79 Wt/m °C. The temperature of the walls of the stainless steel waste containers must not exceed 300°C in order to avoid notable corrosion of the material of the container casing.

CONCLUSION

- Research studies and the technologies that have been developed for managing high-level radioactive wastes produced in the regeneration of spent nuclear fuel using dry methods (pyroelectrochemical and gas fluoride technologies) indicate the feasibility of controlled storage of such wastes in simple hermetically sealed vessels for a prolonged period (decades).
- Radioactive wastes created as a result of pyroelectrochemical regeneration may be placed fairly easily in glass or ceramic forms suitable for permanent burial.
- A program of research work must be completed to determine the optimal form (for example, chemical composition, type of matrix) for solid high-level radioactive wastes from the standpoint of ensuring the safety of their burial in geological formations. This research is a vital element of the regeneration technologies being developed to facilitate the new closed fuel cycles.

Chemical Treatment of High-Level Waste for Utilization

Boris F. Myasoedov
Vernadsky Institute of Geochemistry and Analytical Chemistry
Russian Academy of Science

Russia is managing its irradiated nuclear fuel by reprocessing it and extracting useful isotopes. These activities will benefit the national economy as well as minimize the disposal of radioactive waste.

The RT-1 Plant at the Mayak Production Association was put into operation in 1976 within the framework of the Russian concept of preparation and transition to a closed fuel cycle. Over its 25 years of operation the RT-1 Plant has progressed from the trial reprocessing of a few kinds of nuclear fuel to the powerful and steady processing of a wide range of irradiated nuclear fuel from water-moderated water-cooled power (VVER-440), fast neutron (BN-600), transport, and research reactors. The RT-1 is the only pilot production plant in Russia that is able to rationally combine production and research activities to demonstrate the possibility of a closed nuclear fuel cycle. As a result of the development of an atomic industry, nuclear power engineering, and a nuclear powered fleet, a considerable amount of radioactive waste with total activity of about 1.5×10^9 Ci, and spent nuclear fuel (SNF) with total activity of about 4.65×10^9 Ci are accumulated in Russia at various enterprises belonging to different departments of the federal executive authority.

In order to achieve a closed fuel cycle, proper management of long-lived radionuclides contained in spent nuclear fuel, optimization of existing high-level waste reprocessing methods, and separation technologies for new and existing reprocessing plants have been studied over the last 20 years. These studies have been carried out at several Russian institutes (for example, the Khlopin Radium Institute, Institute of Chemical Technology, Institute of Physical Chemistry, and Institute of Geochemistry and Analytical Chemistry) in collaboration with the radiochemical plants of Mayak Production Association and the Mining-Chemical Association. Basic operation of the technology of separation of long-lived radionuclides involves the selective recovery from high-level waste of cesium, strontium, technetium, rare earth elements, and transplutonium elements, as well as the residues of uranium, neptunium, and plutonium remaining after the PUREX process. The objective of this presentation is to consider those developments that are now most feasible for application, are already in service at Mayak Production Association, or are soon to be introduced there.

FERROCYANIDE SORBENTS FOR CESIUM RECOVERY

An example of the successful use of sorption processes for recovery of radionuclides from high-level waste is the method of cesium recovery by inorganic ferrocyanide-containing sorbents developed by the Institute of Physical Chemistry in collaboration with Mayak Production Association.[1] In practical application, preference was given to copper-nickel ferrocyanide, which has the highest stability in the cycle of sorption-desorption-regeneration. Systematic studies on the use of this sorbent enabled the development of technology for cesium recovery from high-level waste during SNF reprocessing. When testing the technology at Mayak Production Association, a 120 liter sorption column was used. The recovery degree of cesium at sorption stage was more than 98 percent; the cesium yield into desorbate was 98–99 percent; and the concentration ratio attained was 100 percent. After 15 cycles, the sorption properties of the sorbent remained unaffected. In the course of pilot industrial tests, about 7 millicuries of Cs were separated from high-level waste.

CROWN ETHERS FOR RECOVERY OF STRONTIUM AND CESIUM

A method using macrocyclic polyethers (crown ethers) has successfully been developed for strontium recovery from high-level waste. Specifically, an elaboration of the Institute of Chemistry's technology using dicyclohexyl-18-crown-6 (DCH-6) was brought to the level of pilot industrial tests at Mayak Production Association. As a result of these tests, about 90 m^3 of high-level waste were reprocessed and more than 0.5 mCi of radiostrontium were recovered with a sixfold degree of concentration.[2] The degree of strontium recovery was 96 percent. A special advantage of the DCH-6 method is the simplicity and efficiency of conducting the strontium stripping operation using water.

RECOVERY OF CESIUM AND STRONTIUM USING CHLORINATED COBALT DICARBOLLIDE

Fundamental studies on the extraction of Cs and Sr by cobalt(III) dicarbollide have been carried out primarily in the Czech Republic. In Russia dicarbollide technology has been applied to high-level waste processing. Figure 1 shows the

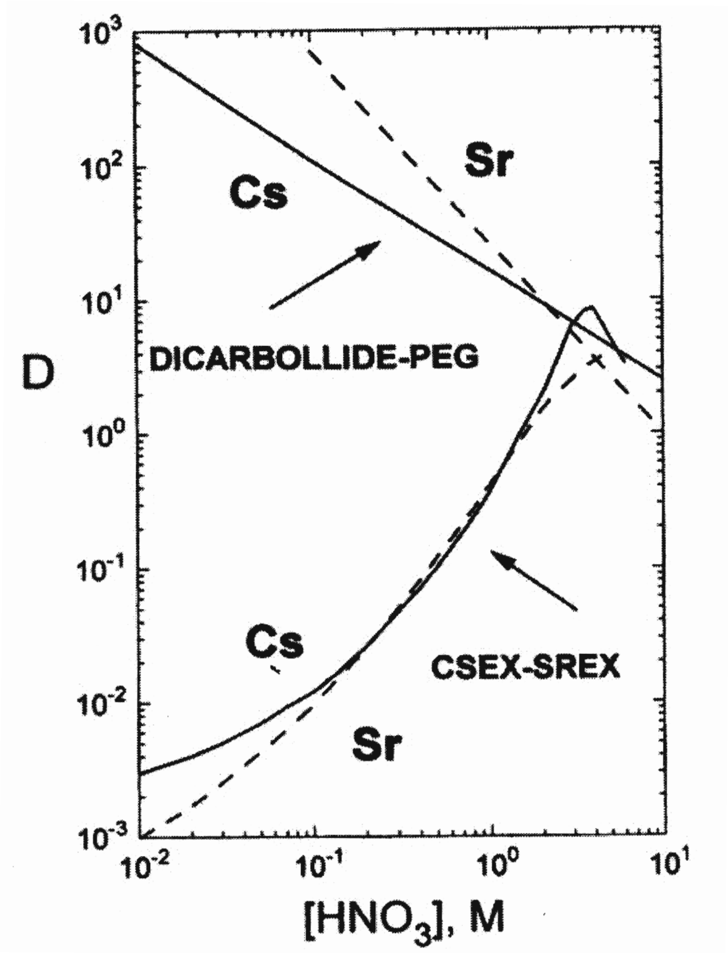


FIGURE 1 Nitric acid dependences of Cs and Sr extraction (D is the distribution coefficient) using dicarbollide and crown ether extractants (see Horwitz and Schulz, 1998). Dicarbollide (0.01M cobalt (III) dicarbollide and 0.01M PEG – 400 in nitrobenzene), CSEX-SREX (0.1 M Cs extractant – 0.005M Sr extractant – 1.2M TBP -Isopar-L – 5 vol percent lauronitrile).

nitric acid dependencies for the extraction of Sr and Cs using a 0.01 M solution of cobalt(III) dicarbollide and 0.01 M polyethylene glycol (PEG-400) in nitrobenzene. The flowsheet shows the combined recovery of cesium and strontium, as well as the extraction of barium and lead from high-level waste with HNO_3. The resulting content has no more than 3.0 M and an overall content of nitrate-ion of no more than 5.0 M. The combined stripping of cesium and strontium is carried out by means of 5.0–6.0 M HNO_3; the extractant is regenerated

TABLE 1 Industrial Operation Results of the UE-35 Separation Facility at Mayak Production Association

		Characteristics of Reprocessed High-Level Waste	
Operation Year	Duration Operation, Months	Volume, m^3	Total Activity, kCi
1996	3	210	11,886
1998	2.5	95	6,539
1999	~1	62	1,498
2000	~3	254	6,156
2001	11	558	23,436
Total	20.5	1,179	49,515

See Dzekun et al. (1991).

by a solution of 8 M HNO_3, and 20 g/L hydrazine, which is then used for the stripping operation. This extraction mixture recovers more than 99.5 percent of the cesium and strontium. The most notable achievement in the use of the ChCoDiC process in Russia relates to the reprocessing of high-level waste of varying composition at Mayak Production Association.[3] Using this technology, the first commercial facility in the world (UE-35) for the recovery of radionuclides began operation in August 1996. The first line of this facility was planned for the selective recovery of cesium and strontium from high-level waste. By the time UE-35 was put into operation in 13 storage tanks at Mayak Production Association there were 4315 m^3 of highly salted aged wastes that cannot be subject to direct vitrification. The operating experience of UE-35 shows that three tanks with high-level waste can be reprocessed yearly. This will allow for the reprocessing of the most hazardous high-level waste of Mayak Production Association within several years (see Table 1).

RECOVERY OF ACTINIDES USING NEUTRAL ORGANOPHOSPHOROUS COMPOUNDS

Neutral bifunctional extractants have been studied in Russia for three to four years. Parallel investigations have been carried out in the United States.[4] To extract TRU (transuranium) from high-level waste Russian chemists adopted a different carbamoylmethylphosphine oxide (CMPO) derivative from the one used in the United States. The TRUEX (transuranium extraction) process, namely, diphenyl-N, N-di-n-butyl CMPO, is abbreviated DPhDBCMPO. The diphenyl CMPO derivative is insufficiently soluble in paraffinic hydrocarbon diluents, even in the presence of excess TBP (tributyl phosphate), to be of practical use.

Diphenyl CMPO derivatives have a strong propensity toward third phase formation. However, Russian chemists have found that use of a fluoroether called Fluoropol-732 as a dilutent for the diphenyl CMPO eliminated the unfavorable solubility and third phase formation properties of this derivative. The D_{Am} versus aqueous HNO_3 concentration curve using a 0.05 M DPhDBCMPO solution in fluoropol-732 is shown in Figure 2. These data show that the values of D_{Am} obtained with the DPhDBCMPO-fluoropol system are significantly higher, using only one-fourth the concentration of CMPO of those obtained with the TRUEX process solvent over the entire nitric acid concentration range. The Russian TRU extraction process uses a 0.1 M solution of DPhDBCMPO in

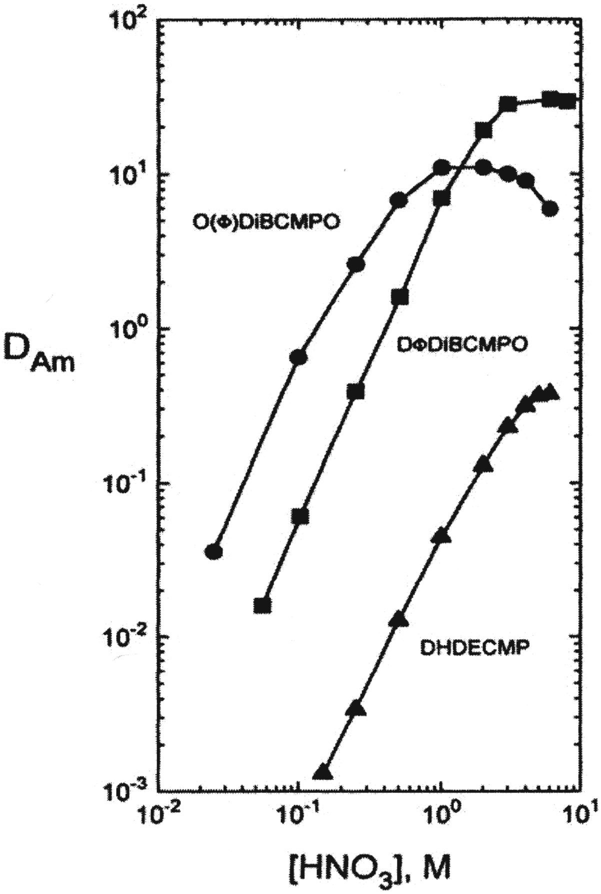


FIGURE 2 Comparison of octyl(phenyl)- and diphenyl-DiBuCMPO and DHDECMP in the presence of TBP at 25°C as extractants for Am(III) in nitric acid (see Horwitz and Schulz, 1998). 0.25M CMP or CMPO – 0.75M TBP – CCI_4.

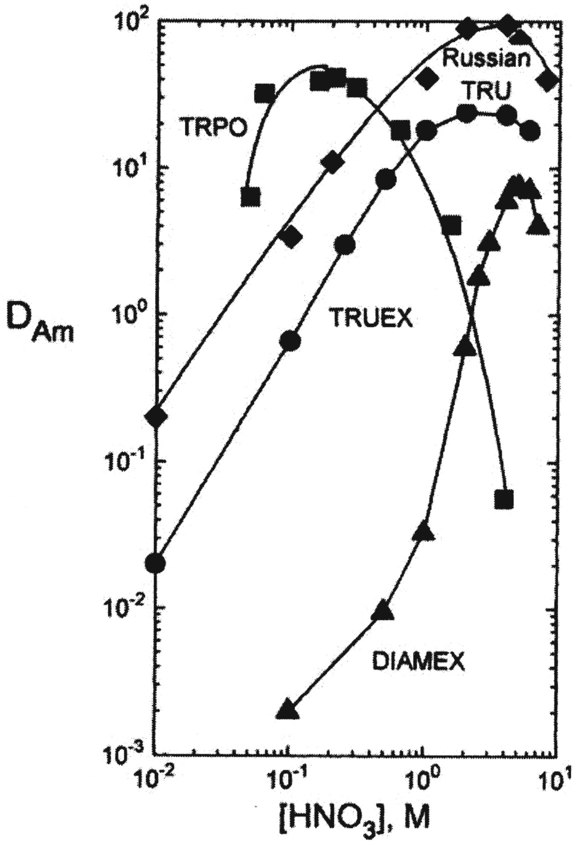


FIGURE 3 Comparison of TRUEX, Russian TRU, DIAMEX, and TRPO process solvents as extractants for Am(III) in nitric acid (see Horwitz and Schulz, 1998). TRUEX process solvent (0.02M O(0)DiBCMPO – 1.4M TBP – Conoco (C12–C14)), t=30°C. Russian TRU process solvent (0.05M D (Ø) DBCMPO – – 1.4M TBP – Conoco (C12–C14)), t=30°C. Russian TRU process solvent (0.05M D (Ø) DBCMPO -Fluoropol-732), t=23°C. DIAMEX process solvent (0.5M DMDBTDMA-TPH), t=25°C. TRPO process solvent (30 vol percent TRPO – kerosene), t=25°C.

fluoropol-732 as the process solvent (see Figure 3). An 18-stage bank of centrifugal contactors was used to test the TRU extraction. The feed solution consisted of a high-level waste simulation, 5 M in HNO_3, containing more than 13 g/L of lanthanides and actinides. An interesting feature of the flowsheet is the use of acetohydroxamic acid (AHA) to strip Fe(III), Zr(IV), and Mo(VI), which also extracts the transplutonium elements (TPE). A solution of 2 M HNO_3 10 g/L AHA was employed for this purpose. The AHA strip solution is contacted with fresh process solvent to remove any traces of TPE and possibly Pu. TPEs and lanthanides were stripped from the process solvent using 0.01 M HNO_3. More than 99.5 percent of the actinides and lanthanides were recovered and concentrated by a factor of four to six. The reduction of Fe, Zr, and Mo from the TPE fraction was >50 percent. Efforts are currently underway to apply the process to a plant-scale operation. A number of the favorable features outlined for TRUEX also apply to the Russian TRU process, namely, efficient extraction of Am(III) over a wide range of HNO_3 concentrations, and the ability to strip, using low concentrations of acid. The Russian TRU process has the added advantage of using a lower concentration of a less expensive extractant. Because of the absence of TBP in the DPhDBCMPO-fluoropol system, radiolytic and hydrolytic degradation is probably less than with the TRUEX process solvent. The technology of the modified TRUEX process was confirmed by recent tests under static conditions and for industrial high-level waste from the Mayak Production Association facility.

SEPARATION OF ACTINIDES AND LANTHANIDES BY COUNTERCURRENT CHROMATOGRAPHY

The most effective method for TPE separation from highly radioactive wastes is extraction based on the use of bidentate neutral organophosphorus compounds (BNOC).[5] A technological scheme of TPE removal with diphenyl (dibutyl-carbamoylmethyl) phosphine oxide (Ph_2Bu_2) in fluorine-containing polar solvent using an 18-step set of extractors has been proposed. The possibility of isolating >99.5 percent TPE as well as purifying the TPE from accompanying elements other than lanthanides has been demonstrated. The TPE + lanthanides fraction contains a low concentration of nitric acid (about 0.02–0.03 M) that makes it possible to choose other water systems for group separation of these elements. It is possible to increase the separation factors for the elements in the BNOC-based systems by (1) improvement of reagent selectivity by changing the reagent structure mainly by introduction of hard bridge fragments (arresting the arrangement of the donor atoms) into the extractant molecule and (2) addition of complex-forming agents to the aqueous phase that are able to react selectively with TPE or lanthanides in acid solutions. Neither the first nor the second method allows single-stage separation of TPE from lanthanides, as they have very similar properties. A multistage liquid-liquid extraction separation is needed to separate the TPE and lanthanides, in order to increase the separation factors.

Separation of actinides and lanthanides can be carried out by partition countercurrent chromatography (CCC), which is also called liquid chromatography with a free stationary phase. CCC is a relatively new method of separation. This method, suggested by the U.S. scientist Yoichiro Ito is based on the retention of the stationary organic phase in a rotating column under the action of centrifugal force while the mobile aqueous phase is pumped through continuously. Sub-

TABLE 2 Precipitation of Am (III) and Eu (III) Taken Separately from Nitric Acid Solutions Containing Potassium Ferro- and Ferricyanide

Conditions of Precipitation					
Precipitant	Element	M	Mg/mL	Percent Found in the Sediment	Percent Found in the Solution
$[K_4Fe(CN)_6]$	Am(III)	4.1×10^{-3}	1.00	99	1
	Eu(III)	8.6×10^{-3}	1.31	98	2
$[K_3Fe(CN)_6]$	Am(III)	6.4×10^{-3}	1.55	92	8
	Eu(III)	8.6×10^{-3}	1.31	0	100

See Kulyako et al. (1996).

stances are separated due to differences in their distribution coefficients when the mixture to be separated moves with the flow of the aqueous phase through the column. It was shown that TPE and lanthanides can be separated in various BNOC-based systems using CCC.[6]

SEPARATION OF ACTINIDES AND LANTHANIDES BY COUNTERCURRENT CHROMATOGRAPHY

The search for new complex-forming and oxidizing systems for TPE and rare earth elements (REE) has opened up new possibilities in the chemistry and technology of these elements. Ferrocyanide ions are known to form a multitude of salts with, in particular, cations of various elements and rare earths. The formation of salts with the same cations occurs also with ferricyanide ions but to a lesser degree. Use of the ferri/ferrocyanide system has been described in various redox reactions. Study of the behavior of some TPE and REE in weakly acid and alkaline solutions containing ions of tri- and bivalent iron is of immediate interest. When potassium ferrocyanide is added to an americium solution in 0.1 M nitric acid, a white precipitate forms, just as in the case of REE. When potassium ferricianide is added, an americium species precipitates from the acidic solution, contrary to REE (see Table 2). TPEs and REEs are separated at the stage of TPE ferricyanide precipitation. In this case REEs remain in acidic solution and can be easily separated from precipitated TPEs. Americium is isolated from curium at the stage of its alkaline-ferricyanide dissolution.

CONCLUSION

Development of efficient technologies for recovery of long-lived radionuclides from high-level wastes is urgent for implementation of promising new management methods (transmutation and disposal), as well as for the existing practice of high-level waste management. In Russia at the Mayak radiochemical plant, the UE-35 industrial facility, which recovers cesium and strontium from high-level waste, has been in operation since 1996. The next stage is aimed at development and implementation of actinide separation technology from high-level waste. For this purpose the following four processes are being studied and tested: (1) processes based on chlorinated cobalt dicarbollide (ChCoDiC-process); (2) isoamyldialkyl-phosphine oxide (POR-process); (3) diphenyl-dibutyl-carbamoylphosphine oxide (modified TRUEX-process); and (4) a combination of ChCoDiC, carbamoylphosphine oxide (CMPO), and polyethylene glycol (PEG) (UNEX-process).

ADDITIONAL REFERENCES

Dzekun, E. G., A. S. Scobtsov, et al. Proceedings of Conference in Ozersk (Zav. Lab.) pp. 29–36. [in Russian]

Horowitz, E. P., and W. W. Schulz. 1998. Metal Ion Separation and Preconcentration: Progress and Opportunities, Chapter XX, eds. A. H. Bond, M. L. Dietz, and R. D. Rogers. Cary, N.C.: American Nuclear Society.

Kulyako, Yu. M., D. A. Malikov, T. I. Trofimov, and B. F. Myasoedov. 1996. Behavior of trans-plutonium and rare earth elements in acidic and alkaline solutions of potassium ferricyanide. Mendeleev Communications 5:173–174.

Myasoedov, B. F., M. K. Chmutova, A. Yu. Shadrin, V. N. Romanovskiy, I. V. Smirnov, V. A. Babain. 1993. Proceedings of the International Conference and Technology Exhibition on Future Nuclear Systems: Emerging Fuel Cycles and Waste Disposal Options, GLOBAL '93, September 12–17, 1993, Seattle, Washington 1:581–587.

NOTES

1. E. A. Filippov, E. G. Dzekun, A. K. Nardova, I. V. Mamakin, V. M. Gelis, and V. V. Milyutin. Application of crown-ethers and ferrocyanide-based inorganic material for caesium and strontium recovery. Proceedings of the Symposium on Waste Management, Tucson, Arizona, March 1–5, 1992, 2(1992):1021–1025.

2. Ibid.

3. E. G. Dzekun, A. S. Scobtsov, et al. Proceedings of Conference in Ozersk (Zav. Lab.). 1991. p. 29–36. [in Russian]

4. E. P. Horwitz and W. W. Schulz. Metal Ion Separation and Preconcentration: Progress and Opportunities, Chapter XX, eds. A. H. Bond, M. L. Dietz, and R. D. Rogers. Cary, N.C.: American Nuclear Society, 1998.

5. V. M. Esimantovskiy, B. Ya. Galkin, E. G. Dzekun, L. N. Lazarev, R. I. Lyubtsev, V. N. Romanovskiy, D. N. Sichikin. Proceedings of the Symposium on Waste Management, Tucson, Arizona, March 1–5, 1992, 1(1992):805–808. See also Horwitz and Schulz, 1998, ibid.

6. B. F. Myasoedov and M. K. Chmutova. Proceedings of the Third Japan-Russian Joint Symposium on Analytical Chemistry, Nagoya, Japan. November 5–9, 1986.

Immobilization of High-Level Waste: Analysis of Appropriate Synthetic Waste Forms*

S. V. Yudintsev
Institute of the Geology of Ore Deposits, Petrography, Mineralogy, and Geochemistry
Russian Academy of Sciences

The immobilization matrix is the key element in the system of engineering barriers. It must ensure the isolation of radioisotopes after they are placed in geological repositories under conditions affected by underground water. The glasses that are presently being used have a low capacity to incorporate plutonium and a low chemical stability. This article presents the results of a selection of crystalline matrices for the immobilization of actinide wastes. Pyrochlore-type phases could be used for the fixation of the actinide-zirconium-rare earth fraction of high-level wastes, while ferrites with a garnet structure could be used for the immobilization of complex wastes, including corrosion products, for example, Fe, Al, and Ga.

The reprocessing of irradiated fuel from nuclear power plants produces a large quantity of radioactive wastes, including high-level wastes. These high-level wastes represent a small portion of the total by volume but are the source of the bulk of the total activity level. As shown in Table 1, they contain fission products, activated corrosion products, and actinides, including those formed as a result of nuclear transformations in the reactor, as well as numerous nonradioactive elements, such as isotopes of fission products and technological additives.[1] The most dangerous radionuclides in high-level wastes are the actinides and certain long-lived products of their fission (for example, ^{93}Zr, ^{99}Tc, ^{126}Sn). Plans call for introducing them into stable immobilization matrices that would subsequently be deposited underground.[2] Selecting optimal materials is a key point in ensuring the safe management of high-level wastes from the nuclear fuel cycle. The quest for high-level waste immobilization matrices began in the 1950s

---

*Translated from the Russian by Kelly Robbins.

TABLE 1 Basic Radioisotopes in High-Level Wastes from the Reprocessing of Spent Nuclear Fuel and Their Half-Lives

Basic Fission Products and Corrosion Elements (β- and γ-emitters)		Transuranic Actinides (α-emitters)	
Radionuclide	$T_{1/2}$, in years	Radionuclide	$T_{1/2}$, in years
^{90}Sr	29	^{237}Np	2.1×10^6
^{93}Zr	1.5×10^6	^{238}Pu	89.9
^{99}Tc	2.1×10^5	^{239}Pu	2.4×10^4
^{126}Sn	10^5	^{240}Pu	6.5×10^3
^{129}I	1.7×10^7	^{241}Pu	14
^{137}Cs	30	^{242}Pu	3.78×10^5
^{147}Pm	2.6	^{241}Am	433
^{151}Sm	93	^{242}Am	152
^{154}Eu	16	^{243}Am	7.3×10^3
Activated corrosion products		^{243}Cm	28
^{59}Ni	7.5×10^4	^{244}Cm	17.9
^{60}Co	5.3	^{245}Cm	8.5×10^3
^{63}Ni	96	^{246}Cm	4.76×10^3

with the study of various vitreous and crystalline materials based on silicates, phosphates, and titanates.[3] For industrial-scale operations for this purpose, glasses are currently used—borosilicate glasses abroad[4] and aluminophosphate glasses in Russia.[5] The shortcoming of glasses is their limited capacity to incorporate actinides (especially plutonium) and their low chemical stability.[6] The interaction of glass-like matrices with underground water is accompanied by the formation of colloidal particles[7] in which radionuclides could migrate over great distances. In addition, glasses crystallize over time, further reducing the stability of radionuclide fixation due to the appearance of soluble phases—silicates or phosphates of the alkali and alkaline earth metals, molybdates, and so forth.

To manage them more efficiently wastes may be separated into radionuclide fractions.[8] In one of these fractions the actinide content totals tens of percent by mass, and there is a significant amount of zirconium and lanthanides. The ratio of quantities of these elements in liquid high-level wastes from the reprocessing of spent fuel from various reactors is characterized as follows: actinides 10–15 percent by mass; lanthanides 60–65 percent by mass; and zirconium 20–25 percent by mass.[9] Predominant among the actinides are uranium, neptunium, plutonium, and americium, while the lanthanide group is represented by neodymium, cerium, lanthanum, and praseodymium. Another group of wastes with high actinide content is created during the conversion of metallic weapons-grade plutonium into nuclear fuel.[10]

Besides glass, crystalline matrices represent an alternative waste form with high actinide content.[11] More than 20 phases with various capacities for the given elements and different levels of chemical and radiation stability have already been proposed.[12] A matrix based on pyrochlore [$(Ca,Gd,U,Pu,Hf)_2Ti_2O_7$] has been developed in the United States to immobilize excess plutonium.[13] Zircon, zirconolite, cubic zirconium dioxide, perovskite, yttrium-aluminum garnet, britholite, monazite, and others have also been suggested for use in immobilizing plutonium-containing wastes.[14] The final selection of an actinide matrix has not yet been made and remains an urgent question.

Since 1994, researchers at the Institute of the Geology of Ore Deposits, Petrography, Mineralogy, and Geochemistry (IGEM) of the Russian Academy of Sciences have been studying artificial materials intended for the immobilization of various types of radioactive wastes. These studies are carried out in cooperation with specialists from the Radon Research and Production Association in Moscow, the A. A. Bochvar All-Russian Scientific Research Institute of Inorganic Materials, the Institute of Physics and Power Engineering, the V. I. Vernadsky Institute of Geochemistry and Analytical Chemistry of the Russian Academy of Sciences, Moscow State University, and a number of others, including foreign organizations (the University of Michigan, the Australian Nuclear Science and Technology Organization). These activities are part of a larger overall research effort aimed at creating a basis for the safe burial of high-level radioactive wastes that has been under way since the early 1990s on the initiative and under the leadership of Academician Nikolai P. Laverov of the Russian Academy of Sciences.

From a structural standpoint the advantage lies with those phases having lattices that maintain stability even with wide variations in waste content. They may be used for the immobilization of both excess plutonium and more complex wastes, for example, actinide-zirconium-rare earth element fractions of high-level wastes, actinides and fission products, such as ^{93}Zr, ^{99}Tc, and ^{126}Sn, and wastes from the conversion of plutonium into nuclear fuel. The process of seeking high-level waste matrices may be optimized by doing a preliminary analysis of the characteristics of the crystal structure of promising phases. Using the results of this analysis it is possible to reduce the number of potential compounds, and this significantly simplifies the further experimental testing of the selected material. The efficiency of this sort of approach has been demonstrated using the example of the titanates and aluminates and the structures of zirconolite, perovskite, hibonite, and hollandite.[15] I will now review the possible applications of this analytical method using the example of a matrix with a pyrochlore and garnet lattice.

GENERAL DATA ON THE STRUCTURE OF PYROCHLORE

The structure of pyrochlore (Fd3m, Z = 8) may be considered as having twice the lattice parameter of fluorite with half the polyhedrons lacking two diagonal anions.[16] As a result, instead of a cube, it forms a distorted octahedron, while the formula is transformed from A_4X_8 (the fourfold formula for fluorite) to $^{VIII}A_2{}^{VI}B_2{}^{IV}Y_6{}^{IV}X$. A and B represent the cations in two structural positions while Y and X are the anions, one of which (Y, represented by O^{2-} ions) is included in the octahedral polyhedron, while the others ($X = O^{2-}$, F^-, Cl^-, OH^-) are located in the interstices and are not involved in the structural framework.

The structural-chemical characteristics of pyrochlore-type phases are interrelated.[17] To ensure the stability of the pyrochlore lattice, another requirement besides the general condition of electroneutrality is the correlation of the sizes of the cations in the two structural positions. Based on the results of a geometric analysis it was concluded that the phases $(A^{3+})_2(B^{4+})_2O_7$ crystallize in the pyrochlore structural type if the ratio of $(R_A + R_O)$ to $(R_B + R_O)$ is in the 1.08–1.22 range.[18] Here R_A and R_B signify the sizes of the ions in positions A and B, while R_O is the ratio of the oxygen anion. In the case of strongly polarized ions with a coordination number equaling 8—Bi^{3+}, for example—the maximum value of the ratio could rise to 1.33. Similar values are cited by Wang et al., who state their view that the $(R_A + R_O)$ to $(R_B + R_O)$ ratio must lie between 1.10 and 1.24 if the pyrochlore lattice is to be maintained.[19] Therefore, the limits of variations in the correlations between the sizes of the cations in the two structural positions in pyrochlore can be assessed as 1.3–1.8. A narrower range (1.46–1.80) is given in other works.[20] Either increasing or decreasing the value of this ratio leads to the destabilization of the lattice and the appearance of a phase with a different crystalline structure instead of pyrochlore. Based on these data for various ions (B) in sixfold coordination, we may determine those elements in position (A) with a coordination number of 8 (including actinides) with which they form $(A^{3+})_2(B^{4+})_2O_7$ phases with a pyrochlore lattice (see Table 2). Thus, for titanate pyrochlores ($[B]^{VI} = Ti^{4+}$, $R_{VI} = 0.605$ Å), the radii of the ions must be between 0.8 and 1.0 Å, while stabilization of the zirconate phases ($R_{VI} = 0.72$ Å) requires larger ions with a radius of 0.9–1.2 Å.

Pyrochlore Matrices for Actinides

The pyrochlores $CaUTi_2O_7$ and $CaCeTi_2O_7$ (Ce serves as an imitator of Pu) were synthesized previously.[21] Vance et al. have also obtained the phases $Ca(Pu,U,Zr)\ Ti_2O_7$ and $Ca(Np,Zr)\ Ti_2O_7$ containing up to 45 percent by mass of PuO_2 and NpO_2 (or 0.8–1 atom of Pu or Np in the formula).[22] Titanate pyrochlore is the fundamental phase in matrices for wastes with high actinide content, for example, spent nuclear fuel.[23] By analogy with other materials, Synroc-F was named as another promising ceramic. Matrices with 80–90 percent pyrochlore content are considered in the United States to be promising forms for the fixation of excess plutonium. They are manufactured using cold pressing and then caking. More than 1000 samples of ceramics with cerium, thorium, uranium, and

TABLE 2 Pyrochlore-Type Phases $[(REE)^{VIII}{}_2B^{VI}{}_2O_7]$ and $[^{VIII}(CaAn^{4+})^{VI}(B^{4+})_2O_7]$

$A_{VIII} = REE^{3+}$ (r_{VIII}, Å)	Ti^{4+} ($r_{VI} = 0.605$ Å)	Sn^{4+} (0.69 Å)	Zr^{4+} (0.72 Å)	$A_{VIII} = An^{3+}$ or $Ca^{2+}+An^{4+}$ (r_{VIII}, Å)
La^{3+} (1.16)		+	+	
Ce^{3+} (1.14)	–	+	+	
Pr^{3+} (1.13)	–	+	+	Ca^{2+} (1.12)
Nd^{3+} (1.11)	–	+	+	Pu^{3+} (1.10)
Pm^{3+} (1.09)	–	+	+	Am^{3+} (1.09)
	–	+	+	Ca + Th (1.085)
Sm^{3+} (1.08)	+	+	±	Cm^{3+} (1.08)
Eu^{3+} (1.07)	+	+	±	Ca + U (1.06)
Gd^{3+} (1.05)	+	+	±	Ca + Np (1.05)
Tb^{3+} (1.04)	+	+	–	Th^{4+} (1.05)
Dy^{3+} (1.03)	+	+	–	Ca + Pu (1.04)
Ho^{3+} (1.02)	+	+	–	
Er^{3+} (1.00)	+	+	–	U^{4+} (1.00)
Tm^{3+} (0.99)	+	+	–	Np^{4+} (0.98)
Yb^{3+} (0.99)	+	+	–	
Lu^{3+} (0.98)	+	+	–	Pu^{4+} (0.96)

NOTE: The symbol ± indicates that the pyrochlore structure is stable, and – indicates that it is unstable. The symbol ± indicates Sm, Eu, and Gd zirconates, which have a structure that is both fluorite and pyrochlore in nature. REE indicates rare earth elements.

SOURCE: R. A. McCauley. Structural characteristics of pyrochlore formation. Journal of Applied Physics 51(1)(1980):290–294. Joint Committee on Powder Diffraction—International Center for Diffraction Data. X-ray Database PCPDFWIN2.02, 1999.

even plutonium have been produced by this method. The composition of the material with the latter element is a solid solution of $CaUTi_2O_7$, $CaPuTi_2O_7$, and $Gd2Hf_2O_7$ in a molar ratio of 2:1:1.[24]

SYNTHESIS AND STUDY OF NEW PYROCHLORE MATRICES FOR ACTINIDES

We have obtained new varieties of actinide pyrochlores. The basic composition $[(A^{2+}{}_1A_2{}^{4+})_2(B^{4+})_2O_7]$, where $A^{2+}{}_1$ = Ca or Sr, $A_2{}^{4+}$ = U or Th, and B^{4+} = Ti, Sn, or Zr, was prepared from oxides of elements ground to 20–30 µm. The powders were pressed at 200–400 MPa into tables 12–20 mm in diameter and 4–5 mm in height. These tablets were placed in aluminum oxide crucibles baked at 1300–1550°C for 0.5 to 50 hours. Achievement of equilibrium was established based on the stability of the phase content of the end product as the duration of the experiments was increased. The samples were studied using X-ray phase analysis methods as well as scanning and transmission electron microscopy.

The rate of transformation was highest for pyrochlores containing titanium and tin, while synthesis proceeded most slowly in zirconate systems.[25] In the majority of samples studied, pyrochlore is the main or only phase (see Table 3). In titanate matrices, brannerite, perovskite, and fluorite-type oxides are also present. When thorium is replaced by uranium in the content of the zirconates, a fluorite-type oxide is formed instead of pyrochlore (see Table 3).

Pyrochlores were obtained for the first time ever except for those phases in the systems Ca-U-Ti-O and Ca-Ce-Ti-O. There are no reports about them in X-ray databases. The common point in their formulas is the deficit of cations in positions with coordination numbers equal to 8. It is possible that part of the Zr^{4+} could also be in positions with coordination numbers equal to 8. It is preferable that the positions with this coordination number be filled with bivalent calcium ions instead of quadrivalent actinides. The replacement of thorium by uranium in the matrices $CaThZr_2O_7$ or $(Ca_{0.5}GdTh_{0.5})\ Zr_2O_7$ makes the pyrochlore structure unstable (see Table 3). This is probably associated with the fact that the radius of U^{4+} (1.0 Å) is less than that of Th^{4+} (1.05 Å). As a result the ratio between the sizes of ions in positions ${}^{VIII}[A]$ and ${}^{VI}[B]$ is reduced and goes beyond the bounds of the range characteristic for pyrochlore-type phases. The replacement of zirconium with smaller titanium ions leads to an increase in this ratio, which stabilizes the pyrochlore structure for the phases $(CaU)(ZrTi)O_7$ and $(Ca_{0.5}GdU_{0.5})(ZrTi)O_7$. Sr-Th pyrochlores were not obtained even with large Sn^{4+} and Zr^{4+} ions in positions with the coordination number = 6, despite the

TABLE 3 Pyrochlore Phases and Formulas Discovered in Samples

Calculated Composition	Phases Discovered	Pyrochlore Formula
$CaUTi_2O_7$	Pyr > Br	$Ca_{1.06}U_{0.72}Ti_{2.22}O_{6.94}$
$CaUZr_2O_7$	KO	Pyrochlore not found
$CaCeTi_2O_7$	Pyr > Per > KO	$Ca_{1.03}Ce_{0.99}Ti_{1.98}O_{6.98}$
$CaThSn_2O_7$	Pyr >> KO	$Ca_{0.92}Th_{0.92}Sn_{2.0}(Fe_{0.08})^{*}O_{6.96}$
$SrThSn_2O_7$	Cas > KO > Per	Pyrochlore not found
$CaThZr_2O_7$	Pyr > Per ~ KO	$Ca_{0.91}Th_{0.84}Zr_{2.25}O_{7.09}$
$SrThZr_2O_7$	Per > KO	Pyrochlore not found
$(Ca_{0.5}GdTh_{0.5})Zr_2O_7$	Pyr >> KO	$(Ca_{0.44}GdTh_{0.42})Zr_{2.13}O_{7.05}$
$(Ca_{0.5}GdU_{0.5})Zr_2O_7$	KO	Pyrochlore not found
$(Ca_{0.5}GdU_{0.5})(ZrTi)O_7$	Pyr > KO	$(Ca_{0.62}Gd_{0.97}U_{0.23})(Zr_{0.84}Ti_{1.34})O_{6.90}$
$(Ca_{0.5}GdTh_{0.5})(ZrTi)O_7$	Pyr >> KO	$(Ca_{0.47}Gd_{0.95}Th_{0.4})(Zr_{1.29}Ti_{0.89})O_{7.05}$

*Contamination during formula attrition.

NOTE: Pyr—target phase with pyrochlore structure; Br—uranium titanate (brannerite); KO—oxide with a fluorite lattice; Per—phase with perovskite structure; Cas—tin oxide (cassiterite); >—primary and secondary phases; >>—primary and secondary phases with the secondary phase present in a much smaller amount.

fact that the ratio of the ion radii is in accordance with conditions of geometric stability. With a base composition of $SrThSn_2O_7$ and $SrThZr_2O_7$, oxides with a fluorite or perovskite structure as well as tin oxide were formed instead of pyrochlore.

The data obtained on the correlation of composition and structure must be taken into account in selecting matrices for complex actinide wastes, for example, actinide-zirconium-rare earth element fractions of high-level wastes. The octahedral ions of the pyrochlore phases of rare earth elements and trivalent actinides could vary in size from 0.55 to 0.75 Å. Ti^{4+}, Sn^{4+}, Hf^{4+}, and Zr^{4+} meet this condition. As for other elements that could be located in octahedral positions in the lattice, we may note Tc^{4+} with a radius of 0.65 Å. Its long-lived isotope ^{99}Tc, which is formed during the fission of ^{235}U, could also it seems be included in a pyrochlore matrix. The capacity of the pyrochlore lattice with regard to quadrivalent actinides increases as a result of the conjugate inclusion of Ca^{2+} cations, which serve as compensators for the excess charge.

While pyrochlore-type phases have a high solubility with regard to actinides, they have a limited capacity for other high-level waste components such as iron, aluminum, and others. This could give rise to the appearance of additional phases, thus reducing the isolation capabilities of the matrix. Therefore, it is important to select crystalline phases for the immobilization of complex wastes resulting from the processing of spent fuel. Compounds with a garnet lattice are promising phases for the immobilization of these materials.

GENERAL DATA ON THE STRUCTURE OF GARNET

The structure of garnet, $A^{VIII}{}_3B^{VI}{}_2X_3O_{12}$ (la3d, Z = 8), is a three-dimensional shell made up of alternating XO_4 tetrahedrons and BO_6 octahedrons linked by common vertices.[26] The spaces in between, which have the form of distorted cubes, hold large A cations. The presence of three structural positions—A, B, and X, with coordination numbers 8, 6, and 4—makes it possible to include various elements in the lattice. Position A^{VIII} holds bivalent (Ca, Mn, Mg, Fe, Co, Cd) and trivalent (Y, rare earth elements) cations. Trivalent (Fe, Al, Ga, Cr, Mn, In, Sc, V) and quadrivalent (Zr, Ti, Sn) ions fill the B^{VI} positions. In the X^{VI} position we find trivalent (Al, Ga, Fe), quadrivalent (Ge, Si), or pentavalent (V, As) cations.

Relation Between Structural and Chemical Properties of Garnet Phases

The formation of the garnet structure is possible given a certain combination of charges and sizes of ions in various positions.[27] The cation-anion composition must satisfy the general rule of electroneutrality, while another factor in lattice stability is the relation between the sizes of the ions. Based on the example of 200 combinations of cation pairs, the field of stability for silicate garnets has been defined according to the coordinates of the radii of the ions in positions A and B,[28] and it has been shown that if the size of the cation in either of the two positions is increased, the radius of the ion in the other position must also increase. Violation of these relations will lead to the formation of compounds with a different structure. However, by no means can all garnets that may be formed based on the results of this structural analysis be synthesized at atmospheric pressure. This is especially true of compounds at the extreme ranges of the identified area of stability. Obtaining many of these compounds requires increased pressure, with more pressure being required as the variety of sizes of ions in positions with coordination numbers 8 and 6 increases.[29]

The structure of garnet may be stabilized by adding components that change the effective ionic radius in this or that position. Thus, grossular ($Ca_3Al_3Si_3O_{12}$) cannot be obtained at atmospheric pressure and at temperatures greater than 800°C; however, it was possible to synthesize garnet [$Ca_3(Al_{1.33}Cr_{0.67})Si_3O_{12}$] by replacing one-third of the aluminum atoms with chromium.[30] The same effect occurs when yttrium oxide and iron oxide are added.[31] At one atmosphere and 1400°C in an interval from 90 molar percent $Ca_3Al_2(SiO_4)_3$ plus 10 molar percent $Y_3Fe_2(FeO_4)_3$ to 100 molar percent $Y_3Fe_2(FeO_4)_3$, there is an uninterrupted solid solution with a garnet structure. Replacing the yttrium with gadolinium reduces the solubility of the grossular minal to 10 molar percent. This is probably associated with the larger size of the Gd^{3+} ion (1.05 Å) in comparison with Y^{3+} (1.02 Å), which makes it less efficient as a lattice stabilizer.

On Possible Garnet-Type Actinide Phases

Due to their sizes the ions of trivalent and quadrivalent actinides and rare earths predominantly occupy large dodecahedral spaces in the lattice. The way the polyhedrons are joined in the crystalline lattice determines the relation between the sizes of the ions in the various structural positions. For rare earth garnets the effect of the structural factor on the chemical composition of the compounds is obvious.[32] These compounds include phases with aluminate, gallate, and ferrite composition ($REE_3B_2X_3O_{12}$), B = X = Al^{3+}, Ga^{3+}, or Fe^{3+} (see Table 4). In this group there is an increase in the ion radius in positions with coordination numbers 4 and 6: Al^{3+}— 0.39 Å (IV) and 0.54 Å (VI); Ga^{3+}—0.47 Å (IV) and 0.62 Å (VI); or Fe^{3+}—0.49 Å (IV) and 0.65 Å (VI). In order to ensure the stability of the lattice the increase in ion size in positions B^{VI} and X^{IV} must be accompanied by an increase in the radii of ions in the A^{VIII} structural positions. Therefore, in this group of compounds one should expect an increase in the maximum size of the ion in the dodecahedral position. In aluminate systems one observes the formation of garnets of rare earths from Lu^{3+} to Dy^{3+} with a radius from 0.98 to 1.03 Å. For gallates the garnet-type structure is stable up to Nd^{3+} (1.09Å). If the radius of the ion falls outside these limits, a phase with a perovskite structure and an oxide will form instead of a garnet: $REE_3B_5O_{12}$ = 3

TABLE 4 Stability of Garnet Structure for Phases $[(REE^{VIII})_3(B^{VI})_2(B^{IV})_3O_{12}]$, Where B = Al^{3+}, Ga^{3+}, or Fe^{3+}

Lanthanides (r_{VIII}, Å)	Al^{3+} (r_{IV} = 0.39 Å, r_{VI} = 0.545 Å)	Ga^{3+} (r_{IV} = 0.47 Å, r_{VI} = 0.62 Å)	Fe^{3+} (r_{IV} = 0.49 Å, r_{VI} = 0.65 Å)	Actinides (r_{VIII}, Å)
La (1.16)	–	–	?	
Pr (1.13)	–	?	?	
Nd (1.11)	–	+	?	Am^{3+} (1.09)
Sm (1.08)	–	+	+	Cm^{3+} (1.08)
Eu (1.07)	–	+	+ (1460** °C)	
Gd (1.05)	–	+ (1740* °C)	+ (1460** °C)	
Dy (1.03)	+ (1920* °C)	+	+	
Y (1.02)	+ (1940*–1760** °C)	+	+ (1570*–1470** °C)	
Ho (1.02)	+ (1950* °C)	+	+	
Er (1.00)	+ (1960* °C)	+	+	
Yb (0.99)	+ (2000* °C)	+	+	
Lu (0.98)	+ (2060* °C)	+	+	

*Garnet melt temperature
**Eutectic temperature of garnet-oxide mixture (Al_2O_3, Ga_2O_3, or Fe_3O_4)

NOTE: + means garnet structure stable; – means garnet structure unstable; ? means no data available.

SOURCE: N. A. Toropov, V. L, Barzakovsky, V. V. Lapin, and N. N. Kurtseva. Diagrams of the status of silicate systems. In Dual Systems (1st ed.). Leningrad: Nauka Publishers, 1969.

$REEBO_3 + B_2O_3$, where B = Al^{3+}, Ga^{3+}, or Fe^{3+}. The melt temperature of ferrites is 400°C lower than that of aluminates and 200°C lower than that of gallates (see Table 4). The eutectic temperature of ferrite garnet and spinel, at which the amount of garnet is about 35 molar percent (70 percent by mass), is even lower (approximately 1460°C). This makes it easier to develop a technology for obtaining high-level waste matrices by melting and crystallization. Spinel accumulates many activated corrosion products (cobalt, nickel, manganese, chromium, iron). All of this makes it possible to regard a biphase garnet-spinel association as a promising matrix for high-level actinide wastes with increased content of the above elements.

Based on the similarity of the ion radii of Am^{3+} and Cm^{3+} on the one hand and Gd^{3+} and Nd^{3+} on the other, we might expect these elements to behave in a similar manner during the synthesis of a garnet matrix. The formation of phases with a garnet structure is most likely for ferrites and gallates of trivalent actinides. For the quadrivalent actinides inclusion in garnet has been experimentally confirmed only for thorium.[33] Garnet was synthesized from the mixture of $(Ca_{2.5}Th_{0.5})Zr_2Fe_3O_{12}$ and $(Ca_2Th)(ZrFe)Fe_3O_{12}$ at temperatures of 1050 and 1200°C and pressure of 1 atmosphere. The composition of the products of the experiments were studied using X-ray methods. The basic formula $(Ca_{2.5}Th_{0.5})Zr_2Fe_3O_{12}$ led to the creation of a monophase garnet ceramic. The mixture with $(Ca_2Th)(ZrFe)Fe_3O_{12}$ at 1050°C also formed a monophase garnet ceramic, but at 1200°C, thorium dioxide or thorianite also appeared along with the garnet. Researchers also synthesized a garnet in which the thorium was replaced by cerium: $(Ca_{2.5}Ce^{4+}{}_{0.5})Zr_2Fe_3O_{12}$.

We must emphasize that the published data on the composition of thorium and cerium garnets are not based on direct determinations but rather are established indirectly based on suppositions about the correspondence of the stoichiometry of the synthesized garnet with the composition of the base formula. This approach is based on the absence of other crystalline phases besides garnet among the synthetic products. From this the conclusion is drawn that the correlation of components in the initial mixture characterizes the composition of the end product. This product requires testing, using local analytical methods. We also note that the above experiments were not aimed at studying the properties of garnet as a possible matrix material for the immobilization of actinide-containing wastes. Only in the past 15 years have special studies been conducted to address this problem.

EXPERIMENTAL STUDY OF GARNET MATRICES FOR HIGH-LEVEL WASTES

Specialists from the Institute of Inorganic Materials were the first in Russia to propose garnet for the immobilization of high-level radioactive wastes.[34] Using the induction melt method in a cold crucible, they synthesized samples similar in composition to andradite ($Ca_3Fe_2Si_3O_{12}$). However, the data on the phase and chemical compositions of the samples and consequently the proof of the formation of garnet phases were not cited. These studies were later continued, which led to the creation of a matrix with a garnet content of more than 80 percent.[35] The content of oxides of rare earth elements and zirconium in the garnet exceeds 20 percent by mass, but the possibility of including actinides has not yet been studied.

Specialists from the Radium Institute are studying garnet ceramics as potential actinide matrices.[36] They have focused on a rare earth (Y, Gd) aluminum-gallate garnet as a base. Taking the place of actual high-level wastes in these studies are uranium and cerium oxides or a complex mixture representing wastes from plutonium production. The products synthesized in these experiments are polyphase ceramics consisting of garnet-, perovskite-, and hibonite-type phases as well as rarer oxides of zirconium and aluminum. The researchers have discovered an effect of the chemical composition on the phase composition of the synthesized samples: in the system Gd_2O_3—Al_2O_3, perovskite is predominant, while garnet becomes the leading phase in the systetm Gd_2O_3—Ga_2O_3. Cerium content in the garnet reaches 6 percent by mass, while for uranium the figure is less than 0.1 percent by mass. Adding 4 percent by mass of metallic tin to the initial mixture leads to an increase in the uranium content of the garnet to 5.5 percent by mass. But even in the latter case, the garnet contains less than 10 percent of the total quantity of uranium in the sample. A similar solubility value (about 6 percent by mass) in garnet has also been established for plutonium.[37] Thus, rare earth Al-Ga garnets have a low capacity for incorporating uranium, plutonium, and cerium, and evidently cannot be recommended for immobilizing high-level wastes with high quadrivalent actinide content. These results do not mean that garnet ceramics should be ruled out as actinide matrices. They only indicate that a detailed study of their structure is required in order to identify compounds that are suitable for this purpose.

It has been shown that the highest quadrivalent actinide capacity should be expected in ferrite garnets.[38] The size of the Fe^{3+} ion is greater than that of Al^{3+}, which ensures the incorporation of larger actinide ions in the garnet lattice, especially with the conjugate inclusion of Ca^{2+} as a charge compensator. The radii of Th^{4+} and Ce^{4+} equal 1.05 and 0.97 Å. The radius of Pu^{4+} (0.96 Å) is close to that of Ce^{4+}, while the sizes of the ions of Np^{4+} (0.98 Å) and U^{4+} (1.0 Å) lie between the values for Th^{4+} and Ce^{4+}. Taking previously collected data into account, we may expect that garnets of the type $(Ca_{2.5}An^{4+}{}_{0.5})Zr_2Fe_3O_{12}$ and $(Ca_2An^{4+})(ZrFe)Fe_3O_{12}$, where An = Pu, Np, or U, may be obtained experimentally.[39]

In order to test this hypothesis, phases formed in the system $CaO—Al_2O_3—SiO_2—Fe_2O_3—ZrO_2—Gd_2O_3—CeO_2—UO_2—ThO_2—PuO_2$ were studied. The initial mixtures corresponding in content to the stoichiometry of garnet (see Table 4) were pressed at 200–400 MPa into tablets 10 mm in diameter and 2–3 mm in height. The synthesis was carried out in an aluminum oxide crucible in a regular air environment (for cerium compounds, in an oxygen environment) as follows. The mixture was heated to the necessary temperature (1300, 1400, or 1500°C) at a rate of 10 degrees per minute, held for a period of from 1 to 20 hours, and cooled to room temperature. The time necessary for achieving equilibrium totaled one or two hours even at the lowest temperature of 1300°C. Those samples that were heated to 1500°C melted to form a layer 3–5 mm thick on the bottom of the crucible. The other samples maintained their initial form, indicating their higher melting points.

The content of the target garnet phase in the samples ranged from 70 to 100 percent (see Table 5). In one of the experiments (13-Th) two different types of garnet formed. The main spikes on the diffractograms were located in the ranges 3.05–3.15 Å (d_{400}), 2.72–2.82 Å (d_{420}), and 2.49–2.57 Å (d_{422}), which illustrates the significant variations in the parameters of the elemental cell depending on the content. Along with garnet the samples contained other phases, such as fluorite-type actinide oxides, a Ca-Zr oxide with a perovskite structure, and oxides of Ca, Fe, and Al (hibonite). The baked and melted garnet ceramics with a similar composition also share a similar phase structure. The basic difference lies in the

TABLE 5 Garnet Formulas Tested and Composition of Products of Experiments

Sample No.	Theoretical (calculated) Formula	Phases in Sample
3-U	$(Ca_{1.5}GdU_{0.5})(ZrFe)Fe_3O_{12}$	Gar > KO > Hib
3-Th	$(Ca_{1.5}GdTh_{0.5})(ZrFe)Fe_3O_{12}$	Gar
45-Ce	$(Ca_{1.5}GdCe_{0.5})(ZrFe)Fe_3O_{12}$	Gar >> Per
1-Pu	$(Ca_{1.5}GdPu_{0.5})(FeZr)Fe_3O_{12}$	Gar >> Hib
4-Th	$(Ca_{2.5}Th_{0.5})Zr_2Fe_3O_{12}$	Gar >> Per > KO
21-Ce	$(Ca_{2.5}Ce_{0.5})Zr_2Fe_3O_{12}$	Gar >> Per
3-Th_a	$(CaGdTh_{0.5}Zr_{0.5})Fe_2(Fe_{0.5}Al_{2.5})O_{12}$	Gar > Hib ~ KO
13-Th	$(CaGd_2)(Th_{0.25}Zr_{0.25}Fe_{1.5})(Fe_{2.5}Si_{0.5})O_{12}$	$Gar^1 > Gar^2$

NOTE: Gar—garnet; KO—cubic oxide with fluorite lattice; Hib—hibonite [$Ca(Fe,Al)_{12}O_{19}$]; Per—perovskite ($CaZrO_3$); >—primary and secondary phases; >>—primary and secondary phases with the secondary phase present in a much smaller amount.

larger size (10–100 μm as opposed to 1–20 μm) and the regular form of the grains in the samples obtained by melting and subsequent crystallization.

The results of the study of the composition of the phases (see Table 6) attest to the significant capacity of the ferrite garnets to incorporate the rare earth elements (Gd, Ce), Zr, and the actinides. The content of the latter reaches 16–18 percent by mass in Ca-Zr-Fe garnet and is reduced with the addition of aluminum and especially silicon. The concentration of actinides is reduced by a decrease in the size of the structural positions when Fe^{3+} ions are replaced with smaller Al^{3+} and Si^{4+} cations. Compression of the structural polyhedrons prevents the incorporation of larger actinide ions in the lattice. This is clearly illustrated in the composition of the product of experiment 13-Th, which included two garnets with different amounts of actinides. In the phase with the higher silicon content the thorium content was 80 percent lower. The excess quantity of thorium is included in the composition of thorianite.

In the plutonium sample only garnet, calcium oxide, and iron oxide with a hibonite structure were found. All of the plutonium incorporated into this sample (16 percent by mass) was found in the garnet phase (see Figure 1). Considering that the relative amount of garnet in the sample equaled approximately 80 percent, the concentration of plutonium in it may be assessed to be on the order of 20 percent by mass.

CONCLUSION

Using the examples of pyrochlore- and garnet-type phases, the possibilities offered by structural analysis were evaluated as part of efforts to find optimal matrices for the immobilization of high-level actinide wastes. The presence of several positions (two for pyrochlore and three for garnet) filled with cations of

TABLE 6 Compositions (percent by mass) and Calculated Formulas of Garnet in Samples

Sample No.	CaO	Al_2O_3	SiO_2	Fe_2O_3	ZrO_2	Gd_2O_3	UO_2	ThO_2	CeO_2
3-U	10.5	7.3	—	24.2	22.8	32.6	2.6	—	—
	$(Ca_{1.48}\ Gd_{1.42}\ U_{0.08})^{VIII}\ (Zr_{1.47}\ Fe_{0.53})^{VI}\ (Fe_{1.87}\ Al_{1.13})^{IV}\ O_{12}$								
3-Th	10.5	—	—	37.3	16.1	21.2	—	14.9	—
	$(Ca_{1.56}\ Gd_{0.98}\ Th_{0.47})^{VIII}\ (Zr_{1.09}\ Fe_{0.91})^{VI}\ (Fe_{2.99})^{IV}\ O_{12}$								
45-Ce	11.1	—	—	38.1	17.6	24.0	—	—	9.2
	$(Ca_{1.58}\ Gd_{1.06}\ Ce_{0.36})^{VIII}\ (Ce_{0.07}\ Zr_{1.14}\ Fe_{0.79})^{VI}\ (Fe_{3.01})^{IV}\ O_{12}$								
4-Th	19.0	—	—	31.0	32.8	—	—	17.2	—
	$(Ca_{2.57}\ Th_{0.49})^{VIII}\ (Zr_{2.02})^{VI}\ (Fe_{2.94})^{IV}\ O_{12}$								
21-Ce	19.9	—	—	33.7	33.9	—	—	—	12.5
	$(Ca_{2.53}\ Ce_{0.47})^{VIII}\ (Ce_{0.05}\ Zr_{1.96})^{VI}\ (Fe_{3.01})^{IV}\ O_{12}$								
3-Th_a	10.3	11.1	—	21.7	18.5	31.2	—	7.2	—
	$(Ca_{1.44}\ Gd_{1.35}\ Th_{0.21})^{VIII}\ (Zr_{1.18}\ Fe_{0.82})^{VI}\ (Fe_{1.31}\ Al_{1.71})^{IV}\ O_{12}$								
13-Th	4.3	—	0.6	30.9	2.9	45.3	—	16.0	—
	$Gar^1 : (Ca_{0.75}\ Gd_{2.25})^{VIII}\ (Gd_{0.21}\ Th_{0.60}\ Zr_{0.23}\ Fe_{0.96})^{VI}\ (Fe_{2.85}\ Si_{0.10})^{IV}{}_{12}$								
	7.6	—	4.2	37.4	7.7	39.9	—	3.2	—
	$Gar^2 : (Ca_{1.11}\ Gd_{1.81}\ Th_{0.10})^{VIII}\ (Zr_{0.52}\ Fe_{1.48})^{VI}\ (Fe_{2.38}\ Si_{0.58})^{IV}\ O_{12}$								

NOTE: The appearance of alumina is associated with dissolution during the melting process in the aluminum oxide crucible. In calculating the formulas it is conditionally accepted that all iron in the sample is in trivalent form and all uranium and cerium are in quadrivalent form.

various charges and sizes creates conditions for the inclusion in them of various waste components, including actinides.

Only a small part of the actinides can be incorporated into the pyrochlore $A_2B_2O_7$ in its quadrivalent form. In order to increase their solubility it is necessary to introduce Ca^{2+} ions as charge compensators. An example would be compounds in which the trivalent ions are replaced by a pair of bi- and quadrivalent cations according to the following formula: $2REE^{3+} = Ca^{2+} + Ce^{4+}$ (U^{4+}, Th^{4+}, Np^{4+}, Pu^{4+}). By comparing the radii of these pairs and the REE^{3+} ions one may suppose the existence of several phases of quadrivalent actinides with a pyrochlore lattice. The average radii of the pairs ($Ca^{2+} + U^{4+}$) and ($Ca^{2+} + Np^{4+}$) is close to the size of Gd^{3+}. For the pair ($Ca^{2+} + Th^{4+}$), the analog would be Pm^{3+} or Sm^{3+}, while for pairs involving Ce^{4+} or Pu^{4+} it would be Tb^{3+}.

As confirmation of this, actinide phases with a pyrochlore structure were synthesized as follows: $CaThSn_2O_7$, $CaThZr_2O_7$, $(Ca_{0.5}GdTh_{0.5})Zr_2O_7$, $(Ca_{0.5}$

GdU$_{0.5}$)(ZrTi)O$_7$, and (Ca$_{0.5}$GdTh$_{0.5}$)(ZrTi)O$_7$. Attempts to synthesize the phases CaUZr$_2$O$_7$, SrThSn$_2$O$_7$, SrThZr$_2$O$_7$, and (Ca$_{0.5}$GdU$_{0.5}$)Zr$_2$O$_7$ were unsuccessful, evidently due to the lack of correspondence between the sizes of the ions with the lattice stability conditions for pyrochlore. Instead of pyrochlore, phases with fluorite- and perovskite-type structures were formed along with tin dioxide.

Conditions required for the formation of high-level waste matrices with a garnet-type lattice were also studied. The incorporation of actinide ions (U^{4+}, Th^{4+}, Np^{4+}, Pu^{4+}) into the garnet was promoted by the filling of positions X^{IV} and B^{VI} with large low-valence Fe^{3+} cations (r_{IV} = 0.49 Å, r_{VI} = 0.65 Å). It has been experimentally established that the actinide content in garnet varies from 0.6–0.8 percent by mass to 16–18 percent by mass depending on its composition.

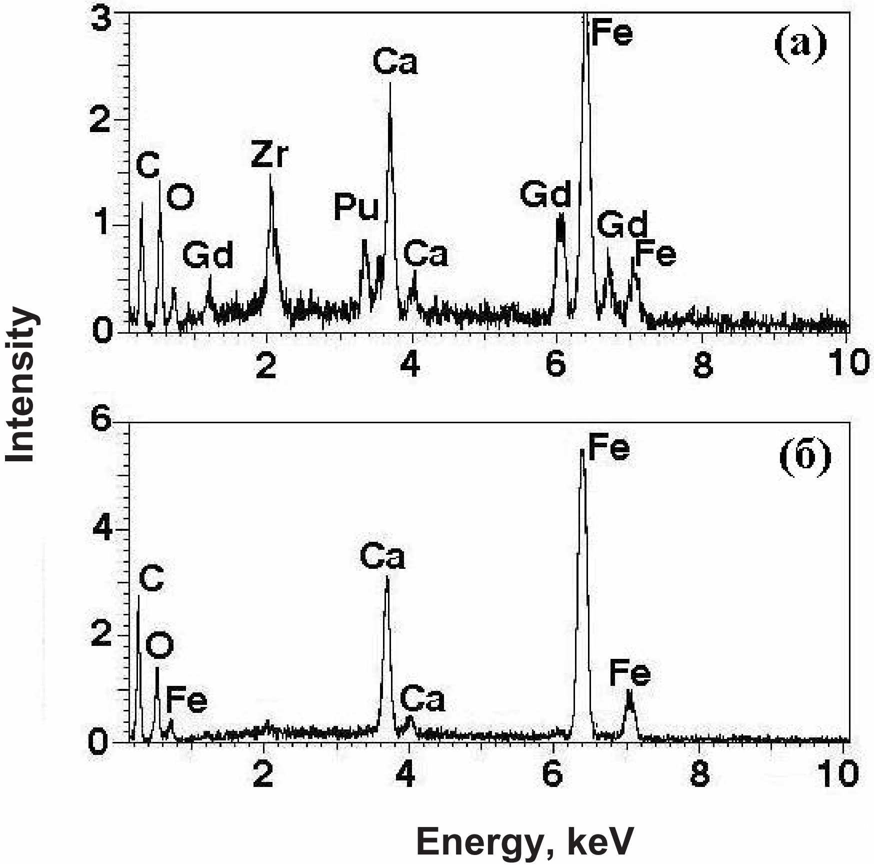


FIGURE 1 Energy dispersion spectra for garnet (a) and Ca-Fe oxide (hibonite), (b) in Sample 1-Pu. The carbon peak (C) is associated with the graphite film on the surface.

The highest actinide concentrations are characteristic of ferrites, while the lowest are observed in silicon- and aluminum-containing garnets.

Based on data that has been collected it has been concluded that it is possible to use pyrochlore matrices to incorporate the actinide and actinide-zirconium-rare earth fractions of high-level radioactive wastes. A phase with a garnet structure represents a more universal matrix. This material may be used for immobilizing both actinide and actinide-zirconium-rare earth fractions and actinide wastes with a more complex composition containing high levels of corrosion products (Al, Ga, Fe, Cr, Ni, Mn, Co). A biphase garnet-spinel association is a promising matrix for such wastes. Research on the radiation resistance and chemical stability of samples of garnet matrices under the influence of heated waters is a top priority for further study.

ACKNOWLEDGMENTS

The author is grateful to A. G. Ptashkin (Radon Research and Production Association, Moscow) and M. I. Lapina, A. V. Mokhov, and A. V. Sivtsov (IGEM) for their help in synthesizing and studying samples. This research was carried out with the partial financial support of the U.S. Department of Energy (project RC0-20002-SC14).

NOTES

1. R. H. Flowers, L. E. J. Roberts, and B. J. Tymons. Characteristics and quantities of radioactive wastes. Philosophical Transactions of the Royal Society A319(1986):5–16.

2. A. E. Ringwood. Disposal of high-level nuclear wastes: a geological perspective. (Hallimond Lecture 1983). Mineralogical Magazine 49(1985):159–176.

3. W. Lutze and R. Ewing, eds. Radioactive Waste Forms for the Future. New York: Elsevier Science Publishers, 1988.

4. L. L. Hench, D. E. Clark, and J. Campbell. High-level waste immobilization forms. Nuclear and Chemical Waste Management 5(1984):149–173.

5. A. A. Vashman, A. S. Polyakov, eds. Phosphate glasses with radioactive wastes. Moscow: Central Scientific Research Institute of Atomic Information, 1997.

6. H. Matzke and J. van Geel. Incorporation of Pu and other actinides in borosilicate glass and in waste ceramics. In Disposal of Weapon Plutonium, eds. E. R. Merz and C. E. Walter. Dordrecht: Kluwer, 1996 pp. 93–105. See also N. P. Laverov, B. I. Omelyanenko, S. V. Yudintsev, B. I. Nikonov, I. A. Sobolev, and S. V. Stefanovsky. Mineralogy and geochemistry of immobilization matrices for high-level radioactive wastes. Geology of Ore Deposits 39(3)(1997):211–228.

7. National Research Council. Glass as a Waste Form and Vitrification Tool: Summary of an International Workshop. Washington, D.C.: National Academy Press, 1996.

8. Nuclear Energy Agency. Actinide and fission product partitioning and transmutation: status and assessment report. Proceedings of the V International Information Exchange Meeting. Paris: Nuclear Energy Agency (EUR 18898 EN), 1999.

9. G. S. Sombret. Waste forms for conditioning high-level radioactive solutions. In The Geological Disposal of High Level Radioactive Wastes, ed. D. G. Brookins. Athens: Theophrastus Publishers, 1987, pp. 69–160.

10. A. F. Grachev, O. V. Skiba, A. V. Bychkov, et al. Experience in the conversion of Russian weapons-source plutonium into nuclear fuel for fast reactors. In Review of Excess Weapons Plutonium Disposition LLNL Contract Work in Russia, eds. L. J. Jardine and G. B. Borisov, UCRL-ID-149341. Lawrence Livermore National Laboratory, 2002, pp. 168–190.

11. Ringwood, op. cit. See also E. R. Vance, B. D. Begg, R. A. Day, and C. J. Ball. Zirconolite-rich ceramics for actinide wastes. Proceedings of the Symposium on the Scientific Basis of Nuclear Waste Management XVIII 353(1995):767–774. See also W. Lutz and R. C. Ewing. Glass and ceramic waste forms: applications and material properties. Ceramic Transactions 61(1995):357–364.

12. P. E. Fielding and T. J. White. Crystal-chemical incorporation of high-level waste species in aluminotitanate-based ceramics: valence, location, radiation damage, and hydrothermal durability. In Journal of Materials Research, ed. W. E. Brown 2(3)(1987):387–414. See also W. Lutze and R. C. Ewing, eds. Radioactive Waste Forms for the Future. Amsterdam, Netherlands: North Holland Physics Publishing, 1988. See also S. V. Yudintsev. Matrices for radioactive waste immobilization: status and prospects. In Excess Weapons Plutonium Immobilization in Russia, eds. L. J. Jardine, G. B. Borisov, UCRL-ID-138361. Lawrence Livermore National Laboratory, 2000, pp. 275–282.

13. V. V. Ebbinghaus, R. A. Van Konynenburg, F. J. Ryerson, E. R. Vance, M. W. A. Stewart, A. Jostons, J. S. Allender, T. Rankin, and J. Congdon. Ceramic formulation for the immobilization of plutonium. In Proceedings of Waste Management 98, Tucson, Arizona, March 1–5, 1998. Published on CD ROM. Tucson, Arizona: Waste Management Symosia, Inc., 1998.

14. Vance et al., op. cit. See also B. Ye. Burakov, Ye. V. Anderson, M. V. Zamoryanskaya, Ye. Ye. Skrykanova, and D. A. Knecht. Synthesis of garnet/perovskite-based ceramic for the immobilization of Pu-residue wastes. Proceedings of the Symposium on the Scientific Basis of Nuclear Waste Management XXII 556(1999):55–62. See also R. C. Ewing. Nuclear waste forms for actinides. Proceedings of the National Academy of Sciences 96(1999):3432–3439.

15. Fielding and White, op. cit.

16. N. V. Belov. Essays on structural mineralogy. Mineralogical Collection of the Lvov Geological Society 4(1950):21–34.

17. Belov, op. cit. See also R. A. McCauley. Structural characteristics of pyrochlore formation. Journal of Applied Physics 51(1)(1980):290–294. See also S. X. Wang, B. D. Begg, L. M. Wang R. C. Ewing, W. J. Weber, and K. V. Godivan Kutty. Radiation stability of gadolinium zirconate: a waste form for plutonium disposition. Journal of Materials Research 14(12)(1999):4470–4473.

18. V. A. Isupov. Geometric criteria for the pyrochlore-type structure. Crystallography 3(1)(1958):99–100.

19. Wang et al., op. cit.

20. P. E. Raison, R. G. Haire, T. Sato, and T. Ogawa. Fundamental and technological aspects of actinide oxide pyrochlores: relevance for immobilization matrices. Proceedings of the Symposium on the Scientific Basis of Nuclear Waste Management XXII 556(1999):3–10. See also B. D. Begg, N. J. Hess, D. E. McCready, S. Thevuthasan, and W. J. Weber. Heavy ion irradiation effects in $Gd_2(_{Ti2-x}Zr_x)O_7$ pyrochlores. Journal of Nuclear Materials 289(1–2)(2001):188–193.

21. Ringwood, op. cit. See also H. Xu, Y. Wang, R. L. Putnam, J. Gutirriez, and A. Navrotsky. Microstructure and composition of Synroc samples crystallized from a $CaCeTi_2O_7$ chemical system: HRTEM/EELS investigation. Proceedings of the Symposium on the Scientific Basis of Nuclear Waste Management XXIII 608(2000):461–466.

22. Vance et al., op. cit.

23. Ringwood, op. cit. See also A. G. Solomah, T. S. Sridhar, and S. C. Jones. Immobilization of uranium-rich high-level radioactive liquid waste in Synroc-FA. Advances in Ceramic Nuclear Waste Management 20(1986):259–265.

24. Ebbinghaus et al., op. cit.

25. N. P. Laverov, S. V. Yudintsev, S. V. Stefanovsky, and Y. N. Dzhang. Characteristics of phase formation in the synthesis of actinide matrices. Reports of the Russian Academy of Sciences 383(1)(2002):95–98.

26. G. Menzer. Die kristallstruktur der granate. Zeitschrift für Kristallographie 69(1928): 300–396. See also S. Geller. Crystal chemistry of the garnets. Zeitschrift für Kristallographie. 125(1967):1–47.

27. Geller, op. cit. See also B. V. Mill and G. Ronniger. Unusual coordination numbers for ions in the structure of garnet. Crystallography 18(1)(1973):126–132.

28. G. A. Novak and G. V. Gibbs. The crystal chemistry of the silicate garnets. American Mineralogist 56(5–6)(1971):791–825.

29. H. Nishizawa and M. Koizumi. Synthesis and infrared spectra of $Ca_3Mn_2Si_3O_{12}$ and $Cd_3B_2Si_3O_{12}$ (B: Al, Ga, Cr, V, Fe, Mn) garnets. American Mineralogist 60(1–2)(1975):84–87. See also B. A. Fursenko. Synthesis of new chromium-containing garnets under high temperatures. Reports of the USSR Academy of Sciences 250(4)(1980):945–949.

30. A. L. Gentile and R. Roy. Isomorphism and crystalline solubility in the garnet family. American Mineralogist 45(5–6)(1960):701–711.

31. S. Geller and C. E. Miller. Silicate garnet—yttrium-iron garnet solid solution. American Mineralogist 44(11–12)(1959):1115–1120.

32. N. A. Toropov, V. L. Barzakovsky, V. V. Lapin, and N. N. Kurtseva. Diagrams of the status of silicate systems. In Dual Systems (1st ed.). Leningrad: Nauka Publishers, 1969.

33. J. Ito and C. Frondel.Synthesis of zirconium and titanium garnets. American Mineralogist 52(5–6)(1967):773–781.

34. V. I. Vlasov, O. L. Kedrovsky, A. S. Nikiforov, et al. Management of liquid radioactive wastes under the closed nuclear fuel cycle concept. Vienna: IAEA, 1987, M-294/3, pp. 109–116.

35. T. V. Smelova, N. V. Krylova, S. V. Yudintsev, and B. S. Nikonov. A silicate matrix for actinides. Reports of the Russian Academy of Sciences 374(2)(2000):242–246.

36. B. E. Burakov, Ye. B. Anderson, M. V. Zamoryanskaya, and M. A. Petrova. Synthesis and study of ^{239}Pu-doped gadolinium-aluminum garnet. Proceedings of the Symposium on the Scientific Basis of Nuclear Waste Management XXIII 608(2000):419–422.

37. Burakov et al., op. cit.

38. S. V. Yudintsev, M. I. Lapina, A. G. Ptashkin, T. S. Iodintseva, S. Utsunomiya, L. M. Wang, and R. C. Ewing. Uranium accommodation into garnet host (Isomprphic capacity and radiation stability of the garnet structured actinide host). Proceedings of the Material Research Society Symposium 713(2002):JJ11.28.1–4.

39. Ito and Frondel, op. cit.

Management of High-Level Radioactive Wastes from the Mayak Production Association and Plans for the Creation of an Underground Laboratory*

Yu. V. Glagolenko
Institute of the Geology of Ore Deposits, Petrography, Mineralogy, and Geochemistry
Russian Academy of Sciences

VITRIFICATION TECHNOLOGY

In accordance with the Russian policy on solidification of high-level radioactive wastes, our enterprise has conducted a program for the creation of a complex for the vitrification of high-level liquid wastes. Implementation of this program has involved the following stages:

- 1969–1971: creation of laboratory facilities for the solidification of solutions in a glass furnace using direct electrical heating
- 1973–1977: testing of pilot facilities using model radionuclide-labeled solutions
- 1986–1988: start up of the complex and regular operation of the first electric furnace
- 1991–1997: regular operation of the second electric furnace
- 2001: regular operation of the third electric furnace

The first unit of the vitrification shop included two EP-500 electric furnaces, each capable of processing 500 liters of solution per hour. One furnace, which was put into operation in 1986, was used for one year and then shut down due to contamination of the water-cooled current feeds of its molybdenum electrodes. During its period of operation, the unit processed 997 cubic meters of high-level wastes with an activity of about 4 million curies.

---

*Translated from the Russia by Kelly Robbins.

The operation of the electric furnace revealed a number of shortcomings in its design, primary among which were the less-than-optimal location of the electrode racks and their inefficient cooling. These factors led to the unplanned shutdown of the first electric furnace and the reconstruction of the backup unit.

Trial run operations in the vitrification section began in September 1990, and in January 1991 the rebuilt EP-500/1-r furnace was put back in operation. Tests on model solutions were conducted over the next six months, and in July 1991 the unit began processing high-level wastes, with these operations continuing for five and a half years. The second unit in the vitrification section, which included two electric furnaces—EP-500/3 and EP-500/4—was put into operation in 2001.

The design for the vitrification section is based on the process of electric arc melting of phosphate glass from waste solutions and flux additives in an electric glass furnace using direct electric heating. As illustrated in Figure 1, the basic technical outline of the process for the vitrification of high-level radioactive wastes includes the following elements:

- solution preparation section, including a chain of various containment vessels
- vitrification section, including an electric furnace with systems for solution correction, glass pouring, power supply, and cooling
- gas treatment section, consisting of a number of gas cleaning devices and filters
- transport section, including a moving conveyer, a chamber for the assembly of cases, and a protective shield
- storage facility for the vitrified wastes

The technical process is carried out as follows. Liquid radioactive wastes from the repository go into a vessel where the solutions are prepared for processing. In this vessel the solution mixture is brought to a specific standard through the addition of calculated amounts of phosphoric acid and sodium nitrate. After being prepared in this manner, the solution is transferred through a number of vessels into an automated displacement elevator, from which the solution flows into the electric furnace directly onto the surface of the molten glass. At the same time that the radioactive waste solution is released into the furnace, the solution is dosed with a reducing agent (ethylene glycol) in order to create a loose foamy layer on the molten surface and reduce unwanted movement of radionuclides.

When the solution contacts the molten surface, there follows a sequence of processes by which the solution is turned to steam, the salts are dried and denitrated, and the oxides are melted. The resulting steam gas mixture moves through a gas pipe into the gas treatment system, which consists of the following devices: a sparger-refrigerator, crude and fine purification filters, ruthenium tetroxide extraction columns, and a fine purification filter and an absorption

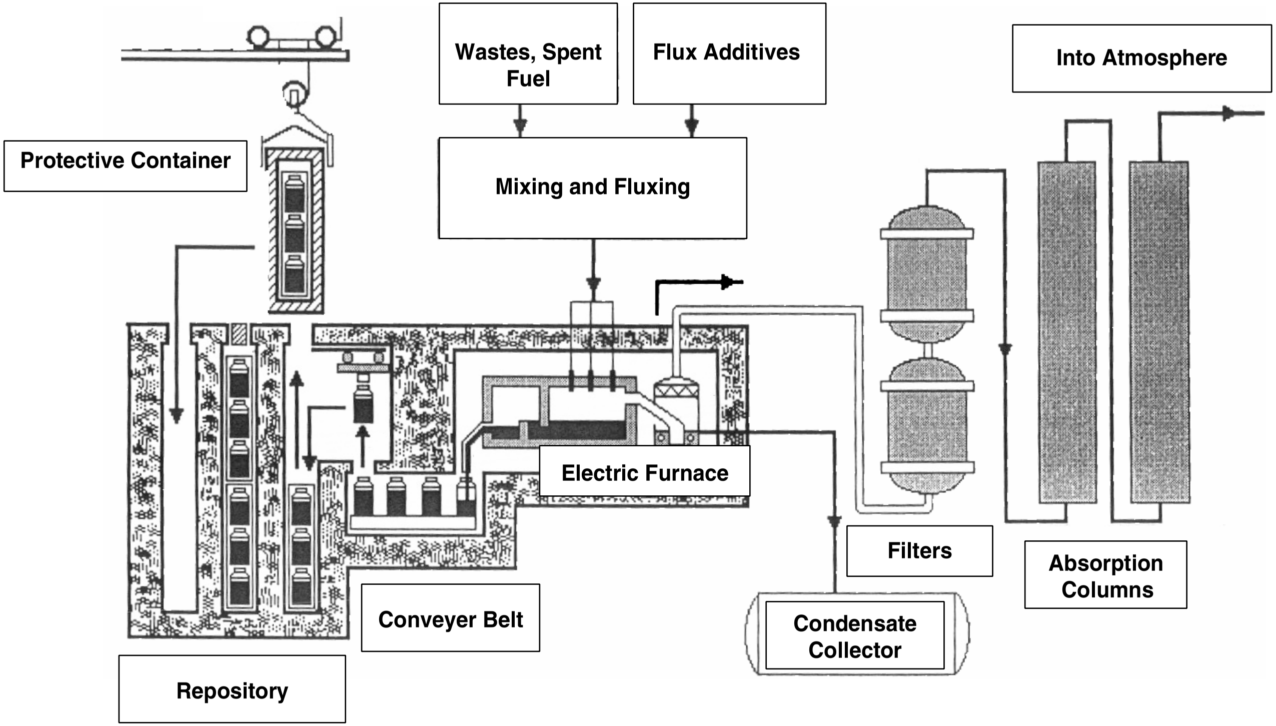


FIGURE 1 Schematic diagram of vitrification of radioactive wastes in the EP-500/1-r electric furnace.

column for extracting nitrates. The treated gases are then discharged through a pipe into the atmosphere, while the condensate that has accumulated in the sparger-refrigerator is sent for further processing to regenerate the nitric acid.

The molten glass is then transferred through the forehearth into an accumulation zone. As it builds up, the glass is periodically poured into 200-liter containers. The containers are filled with a pouring device and then the filled vessels are moved to a packaging chamber with the help of a conveyer belt. In the packaging chamber the glass-filled containers are packed in steel cases, three to a case. The case lids are hermetically welded shut, after which the cases of glass-filled containers are put into another protective housing and then placed in the repository. The repository is made up of a network of concrete emplacement holes. Each hole holds two cases, around which cooling air is circulated.

The design of the basic electric glass furnace device is shown in Figure 2. The furnace is a rectangular basin constructed of baddeleyite-corundum beams. The interior of the baddeleyite-corundum walls is lined with blocks and bricks made from clay-fired refractory material. The vault is composed of fire-resistant concrete. The furnace is housed in a hermetically sealed stainless steel structure. The space between the fire-resistant casing and the furnace structure is filled with quartz sand and refractory chips. Below the unit and up to the level of the molten glass, the side walls of the furnace structure are equipped with water-cooled refrigeration pipes.

The interior of the basin is divided into three zones for melting, transfer, and accumulation. The lower part of the partition has two transfer channels, through which the molten glass moves from the melting zone into the transfer and accumulation zones. The transfer zone is separated from the accumulation zone by a skimmer block that serves to keep the glass in the melt zone at a constant level. The lower part of the accumulation zone features two pouring devices, one for regular use and one backup unit. In the lower part of the melting zone there is a water-cooled gas pipeline that links the furnace with the sparger-refrigerator. The input solution in the electric furnace is dosed out by means of three water-cooled feeders installed in the vault.

Electricity is supplied to the furnace through water-cooled tubular stainless steel cables with attached molybdenum electrodes in the form of cylindrical bolts. In the melt zone there are two pairs of current feeder cables, each with 10 electrodes, while the transfer zone has one pair with two electrodes per cable and the accumulation zone has one pair with four electrodes per cable. The technical characteristics of the electric furnace are presented in Table 1.

From the start of operation until May 2003 the complex processed 15,331 cubic meters of fluxed high-level radioactive solutions with a total activity of 364 million curies and produced 2946 metric tons of aluminophosphate glass. The volume and activity of the vitrified high-level wastes are shown in Table 2.

The current technological set-up for the processing of high-level wastes is illustrated in Figure 3. The refined products formed as a result of extraction

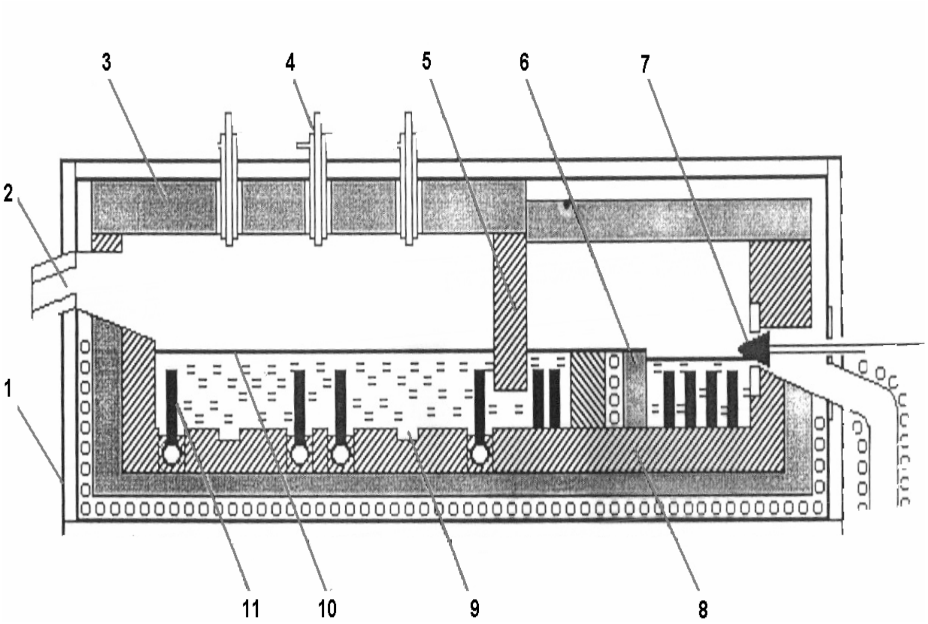


FIGURE 2 Electric furnace. 1: furnace structure; 2: gas line; 3: cover plate; 4: feeder; 5: barrier; 6: skimmer block; 7: pouring device; 8: baddeleyite-corundum blocks; 9: receptor; 10: molten glass; 11: molybdenum electrodes.

TABLE 1 Technical Characteristics of the EP-500/1-r Electric Furnace

Parameter	Value
Solution processing capacity	400–450 liters per hour
Solution input	Continual
Glass output	Periodic, 500 kilograms per pour
Melt temperature	Up to 1200°C
Required power, in kilowatts	
Melt zone	Up to 500
Forehearth zone	Up to 20
Accumulation zone	Up to 120
Dimensions, in meters	
Length	9.55
Width	4.20
Height	3.20
Area of melt surface, in square meters	
Melt zone	10.7
Forehearth zone	0.8
Accumulation zone	2
Level of melt, in millimeters	
Melt zone	350
Forehearth zone	350
Accumulation zone	240–350
Water used for cooling, in cubic meters/hour	180–200

TABLE 2 Volume and Activity Data for Vitrified High-Level Radioactive Wastes

Period of Operation	2/10/1987–2/20/1988	1/9/1991–1/4/1997	6/20/2001–5/4/2003	Totals
Equipment model	EP-500/2	EP-500/1-r	EP-500/3	
Solution processed (in cubic meters)	998	11,460	2873	15,331
Total activity (in millions of curies)	4	278	82	2200
Glass produced (in metric tons)	162	2200	584	2946

processing of spent fuel are steamed to reduce their volume, with the remaining substances being fluxed with sodium nitrate and phosphoric acid and then sent to the EP-500/3 electric glass furnace. As a result, up to 500 metric tons of alumino-phosphate glass with a total activity of 250–300 curies per liter are produced over the course of a year, with this glass being stored in a specially equipped aboveground repository. Condensates from the steaming of the refined products and from the process of vitrifying the fluxed solutions are transferred for regeneration of the nitric acid, which is reused in the technical process, while the tritium condensate is discharged into a storage reservoir.

The primary shortcoming of the existing system is the large volume of active glass that must be stored and the relatively low activity of the vitrified product. Therefore, prospects for the development of the high-level waste processing facility are tied to the use of recently developed partitioning technology that makes it possible to separate out the most ecologically dangerous radionuclides into discrete fractions. As a result of this separation the majority of the wastes may be categorized as mid-level, while the separated fractions of strontium-cesium and transuranic elements may be solidified using small-volume remote-controlled equipment. Modernization of the existing facility to incorporate the partitioning concept would make it possible to optimize the technological

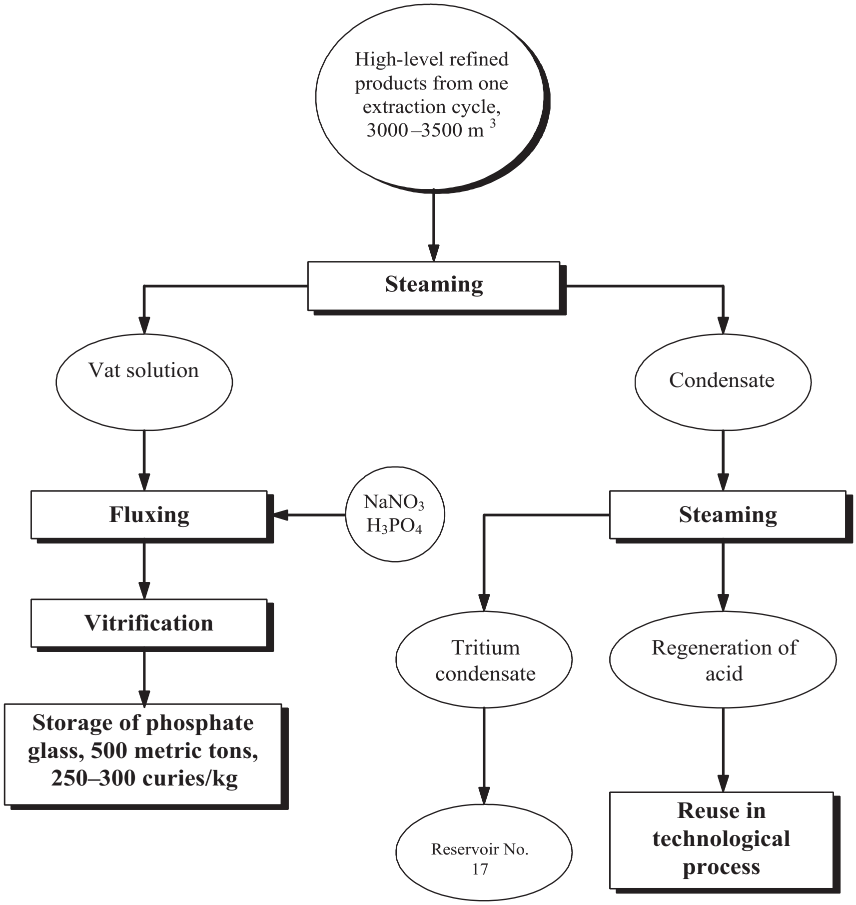


FIGURE 3 Existing system for high-level radioactive waste processing.

system from an economic standpoint by minimizing high-level waste volumes and consequently reducing costs first for temporary storage and then for permanent burial of solidified partitioned wastes.

A promising design for this sort of system for high-level radioactive waste processing is presented in Figure 4. In accordance with this system the high-level wastes are first steamed and then subjected to preliminary processing using fractionation technology. The separated fractions of strontium, cesium, and transuranic elements are solidified into borosilicate glass in the amount of 7.0–7.5 metric tons per year with a total activity of up to 15,000 curies per liter and into mineral-like ceramic (monazite) in the amount of 300 kilograms per year with a total activity of 2 curies per year for alpha radionuclides. One special feature of the system is its capacity for simultaneously processing mid-level refined prod-

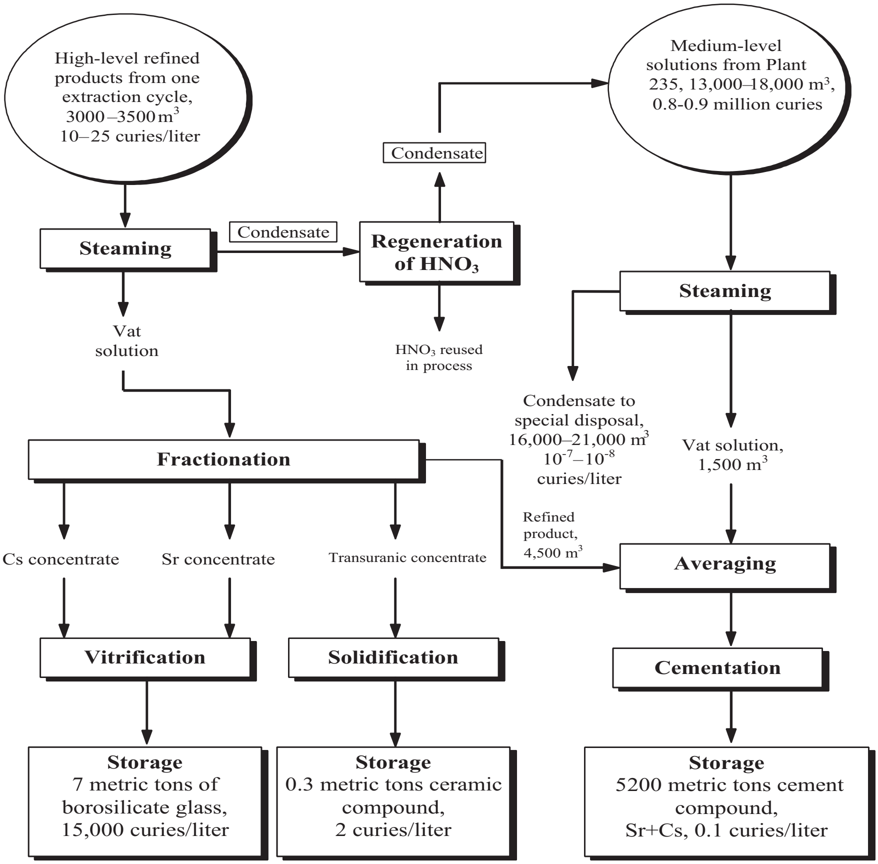


FIGURE 4 Promising system for processing high-level and medium-level wastes using preliminary fractionation.

ucts from the fractionation process and mid-level wastes from the fuel element processing plant by vitrification in the EP-500 electric furnace, with the output being blocks of aluminophosphate glass with an activity of no more than 1.2 curies per liter for strontium and cesium. Plans call for blocks of highly thermal-resistant borosilicate glass produced by this process, as well as ceramic compound containing transuranic elements, to be stored in geological formations, while the aluminophosphate glass is to be first held for temporary storage and then sent for burial in a near-surface sarcophagus.

The aluminophosphate glass, which contains 23–26 percent sodium oxide by mass, 14–19 percent aluminum oxide by mass, and 52–54 percent phosphorus oxide by mass, is characterized by a sodium-leaching rate of 10^{-5}–10^{-6} g/cm^2/hour. Comparative tests of samples of borosilicate and aluminophosphate glasses conducted in 1994 by specialists from the Savannah River Technology Center showed practically identical results for the leaching of the basic matrix components.

In addition to monazite, researchers are currently studying titanates, zircono-titanates, and aluminosilicates, all of which include radioisotopes in their crystal structures through the isomorphic replacement mechanism, as possible highly resistant mineral matrixes for actinide and rare earth elements. Also effective for the immobilization of actinides are mineral matrixes composed of zirconolite and murataite, which are three orders of magnitude more mechanically stable than aluminophosphate glasses.

PLAN FOR AN UNDERGROUND LABORATORY AT THE MAYAK PRODUCTION ASSOCIATION

From the 1960s through the 1980s specialized geological survey organizations carried out comprehensive prospecting studies in areas adjoining the Mayak Production Association in northern Chelyabinsk Oblast to search for geological formations that would be suitable for the burial of liquid radioactive wastes. As a result of experimental studies it was concluded that this area lacked the necessary geological conditions for ensuring the reliable isolation of such wastes. The question of storing liquid wastes at Mayak is not currently under consideration.

In order to implement the end stage of the technology for managing radioactive wastes, a concept for the deep burial of solid radioactive wastes in geologic formations is being developed at Mayak in accordance with the Unified Federal Targeted Program for Russia’s Nuclear and Radiation Security (2000–2006).

Underground burial is deemed the most environmentally, technically, and economically rational means of reliably isolating radioactive wastes from the ecosphere for the entire period during which they might present a potential radiobiological danger. This method makes it possible to guarantee a socially acceptable risk level in all instances with regard to possible releases of radionuclides into the sphere of human habitation regardless of cause.

Using real operating data provided by Mayak, the All-Russia Scientific Research and Design Institute for Industrial Technology (VNIPIPromtekhnologia) and the Russian Academy of Sciences Institute of the Geology of Ore Deposits, Petrography, Mineralogy, and Geochemistry are currently analyzing various options for managing wastes from spent fuel reprocessing. They are also making a comparative assessment of the ecological safety of permanently isolating radioactive wastes underground, taking into account their content of the most dangerous long-lived radionuclides, the formation of daughter radionuclides, and the dynamics of changes in the total heat exchange of various waste fractions.

To date, researchers have completed initial geological, geophysical, and hydrogeological studies with regard to the selection and evaluation of promising sites for the construction of deep underground storage facilities in accordance with existing regulatory requirements for the safe burial of radioactive wastes. The basic conclusions drawn from the results of this research are as follows:

- The geological environment of the region surrounding Mayak is well studied to a depth not exceeding 200 meters.
- The volcanic andesite-basalt rocks that make up the central block of the sanitary-protective zone at the Mayak site are suitable for the reliable isolation of radioactive wastes. They have low effective porosity (~0.2–0.4 percent) and high mechanical and thermal resistance.
- The Mayak area is characterized by intensive but irregular tectonic violation. In the least violated geologic blocks, which are located between major faults, two areas have been identified as promising potential locations for an underground laboratory and subsequently as a radioactive waste isolation site.

The following areas for future work have been defined for the next stage in the process of creating a site for the deep burial of radioactive wastes:

- detailed study of promising sites, to be conducted with the help of test wells and a range of intrawell, site, and deep-level geophysical studies
- evaluation of the tension-deformation status of the sites at depths where the future underground laboratory would be located and reconstruction of the tectonic-dynamic situation in the geologic past
- creation of a geodynamic model of the Mayak region and evaluation on this basis of the probable development of tectonic processes during the timeframes required for the long-term isolation of high-level radioactive wastes
- determination of a site for the placement and construction of an underground laboratory

The main objectives for constructing an underground laboratory lie in establishing the correctness of design solutions being developed with regard to the burial of radioactive wastes in promising sites within rock formations and confirming in practice that the input data being used are reliable and well founded. The comprehensive research being carried out in the laboratory must provide adequate information and quantitative parameters on the isolation capacities of the rock environment and the engineering barriers to be constructed, make recommendations on the optimization of technological processes, and resolve fundamental problems regarding the management of solidified radioactive wastes, methods of their isolation, and the interaction of wastes with the environment.

Regardless of the design for the facility, scientific research work in the underground laboratory is to include the following, as well as other topics:

- provision of a repository for vitrified wastes
- detailed studies aimed at proving that the site selected and the types of rocks surrounding it meet the demands necessary for ensuring the reliable isolation of wastes
- study of the thermal, mechanical, and sorption properties of the surrounding rock
- determination of the physicomechanical parameters of the structural-tectonic blocks, evaluation of lithostatic and tectonic violations, and study of the condition of the rock massif and underground structures under the influence of natural and man-made factors
- study of the effect of technological burdens (thermal, radiation, and chemical impacts) on the condition of the rock massif and on heat and mass transfer conditions
- study of the isolation capacities of various materials and engineering barrier designs under the influence of natural and engineered factors
- experiments on the distribution of nuclides within the selected site in the rock massif after the burial of real wastes
- study of the behavior and stability in real conditions of forms of solidified wastes and their packaging
- study and evaluation of the qualities, means, and methods of controlling processes within the rock massif
- development of equipment and technology for managing wastes and isolating them in real underground conditions

During the period of construction and operation of the underground laboratory, personnel are to make and further develop technical decisions on the design of the underground solidified waste sarcophagus and the entire cycle of operations from the delivery of wastes until their final isolation.

VNIPIPromtekhnologia has recommended to Mayak a well-type underground laboratory that would serve as a center for the development of technologies for the underground isolation of solidified radioactive wastes from radiochemical plants and of spent nuclear fuel. Considering the complexity of the geological and hydrogeological conditions in the volcanogenic sedimentary com-

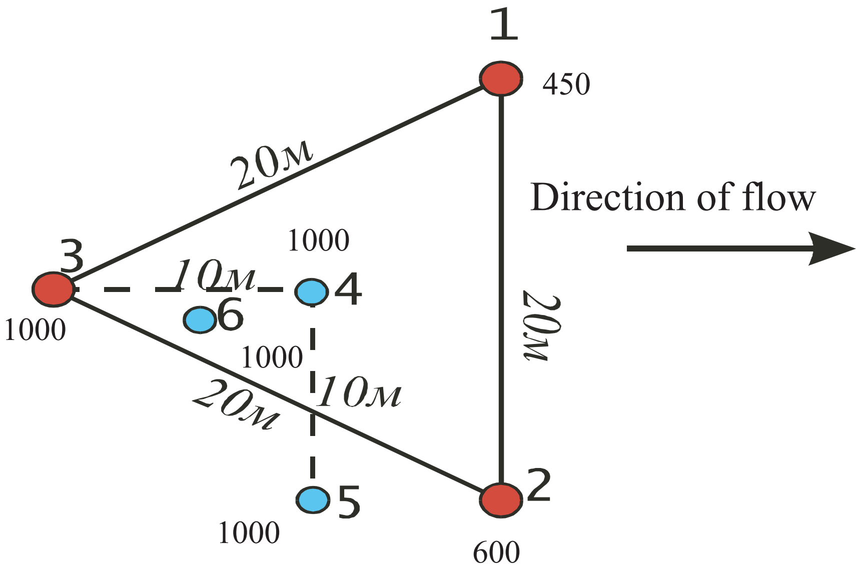


FIGURE 5 Plan for the location of test wells for the experimental laboratory.

plex of rocks that has developed in the Mayak region, a group of wells should be drilled within the promising site. One option for their possible placement is shown in Figure 5.

One important task for the laboratory is to study the process of the migration of products due to leaching from the repository. Therefore, a technical well (well 1) and observation wells (wells 2–6) must be placed, taking into account the most likely direction of underground water movement.

In the first stage, wells 1, 2, and 3 are to be drilled at the corners of an equilateral triangle in order to determine the direction of filtration of water in underground crevices at depths of more than 250 meters. All wells are therefore to be drilled from 0 to 300 meters at an initial diameter of 214 millimeters and cased with fully cemented 194-millimeter steel columns.

From 300 to 450 meters (well 1) and 300 to 600 meters (wells 2 and 3) the wells are to be drilled at a diameter of 151 millimeters without casing.

In the second stage, after the direction of natural flow filtration is determined, wells 4 and 5 are to be drilled, one in the direction of underground water flow movement and the other perpendicular to it. From the depth of 0 to 300 meters they are initially to be drilled at a diameter of 132 millimeters. This interval is to be cased with a fully cemented 127-millimeter steel column. From 300 to 450 meters the well diameter is to be 112 millimeters with the bore remaining open.

After geomigration studies are completed, wells 3, 4, and 5 are to be drilled to a depth of 1000 meters. Detailed experimental filtration and geomigration studies are conducted on the open part of the cut. Based on the results of hydrochemical and radiochemical research, studies are to be made on the effective rate of flow of underground water through fractured zones, the sorption properties of rock with regard to particular isotopes, and the migration of radionuclides in underground water under specific geological conditions. For this purpose, well geophysics methods (thermometry, gamma well logging, and resistivity measurement) will be used, and water samples will be taken for chemical and radiochemical analysis.

On the basis of the data obtained, mathematical models will be prepared along with forecasts of the migration of specific isotopes in particular zones of increased permeability within the rock massif.

Another aspect of no small importance is the study of thermal physical processes associated with the underground isolation of high-level radioactive wastes. The rock will be heated at specified depths, with the temperature regime and condition of the rock to be monitored. This will also facilitate a study of mass transfer and of the destruction of the rock under the influence of increased temperatures under natural conditions. After completion of a study of hydrodynamic characteristics and migration parameters of the filtration zones in the massif, well 3 will be turned into a technical well, which will serve as the location for the heat source, and wells 4 and 5 will be turned into observation wells. Well 6 will be drilled at a distance of 5 meters from the technical well with the same design features as wells 4 and 5.

To facilitate the study of the process of leaching of radioactive products by underground water, the technical well could be equipped with matrices containing either imitation high-level wastes or real vitrified wastes. This process would be monitored using observation wells 4, 5, and 6. Wells 1 and 2 will become research wells for application with a number of technological issues.

During the drilling efforts, detailed descriptions of the cores will be made, and samples will be taken to determine the content and physical, mechanical, and geochemical properties of the rock. A series of hydrogeological studies, for example, evacuation, discharge, and water sampling for chemical analysis, and geophysical studies, for example, stationary types of well logging, alpha well logging, gamma-gamma well logging, expenditure measurement, and resistivity measurement, are to be carried out in each well.

Completing the necessary temperature and radiation measurements and evaluations of violations in the rock massif requires creating a multichannel monitoring system that is adequate to handle both the quantity of simultaneously measured parameters and the types of measurements needed. Information collected from the measuring instruments must go to a central control point, with the results to be recorded in a form that is convenient for further processing.

The following two options are under consideration to facilitate obtaining the necessary information from the observation wells:

1. installation of a stationary system of sensors that could be replaced by an analog system if necessary
2. collection of measurements using well logging methods

The equipment at ground level must be designed to ensure the ability to raise and lower instruments into the wells and to provide for uninterrupted observation capabilities over a long period.

Research and development efforts regarding the lowering of vitrified waste cases are to be carried out in well 2, which is to be equipped with mechanisms for raising and lowering objects. The design and technical characteristics of these mechanisms must ensure not only the accident-free lowering and placement of the cases in the well hole but also their extraction for the necessary studies after a given period of time has passed.

After the raising and lowering operations are fully studied, research can then be conducted on the technology for applying buffer materials for isolating containers of radioactive waste.

Studies are planned regarding the physical-chemical processes occurring during the storage of wastes in a well with the aim of discovering possible changes in the physical-chemical and physical-mechanical properties of the environment in the “case-well-rock-liquid phase” system. Research is being carried out under natural and laboratory conditions. Objectives for the laboratory research include the following:

- verification of predictive data on gas separations during the underground waste storage process
- detection of changes in rock properties due to the effects of radiation

The following work is required to develop predictive assessments and ensure that the necessary data are available for the project:

- determination of the threshold dose at the start of surface destruction of minerals and evaluation of the permissible time of exposure at various doses
- study of the effect of temperature and irradiation on the physical-mechanical properties of various types of rock
- study of the transformation of water content during irradiation and at temperatures from 20 to 200°C
- study of a group of minerals and their compositions in the aim of using them as barriers to isolate the cases of wastes from the surrounding geological environment

Plans call for collecting initial data under natural conditions regarding the stability of construction materials under the influence of various natural and engineered burdens. For this purpose samples of various materials, for example, steel and concrete, are to be lowered into well 1 and kept there for two to three years.

The following determinations must be made based on the results of the research to be carried out in the underground laboratory:

- suitability of the selected site for the burial of solidified high-level radioactive wastes
- basic design and technical elements to be used in the construction of a sarcophagus for solidified high-level wastes

If the results of scientific research conducted in it are positive, the underground laboratory could be turned into an experimental operations storage facility and later developed as the first component of a larger storage facility for solidified high-level wastes.

The underground laboratory is expected to be built in stages. The initial laboratory complex will include one group of wells (four 1000-meter wells, one 600-meter well, and one 450-meter well). Once the lab is in full operation, the scientific research program calls for three groups of wells as described above to be located in various points in the promising site.

Costs for the initial complex total about $2 million, with the entire underground laboratory complex to cost around $5 million.

Creation of the country’s first such laboratory will allow the facility to become a research and demonstration center that will promote the comprehensive study of problems associated with the underground storage of solidified radioactive wastes and the demonstration of technical solutions and underground waste isolation methods.

Creation of Underground Laboratories at the Mining-Chemical Complex and at Mayak to Study the Suitability of Sites for Underground Isolation of Radioactive Wastes

Tatyana A. Gupalo
Federal State Unitary Enterprise, All-Russian Scientific Research and
Design Institute of Industrial Technology, Moscow

In selecting spent fuel storage sites, one must take into consideration the two possible means of further disposition that would be used, namely direct burial or reprocessing of spent fuel with subsequent burial of the output wastes.

Since the early 1970s, the Russian Ministry of Atomic Energy (Minatom) has been studying various sites and geologic formations to determine their suitability for use in the construction of underground radioactive waste isolation facilities. Underground facilities for the isolation of radioactive wastes and spent nuclear fuel differ substantially from other general industrial and power industry structures in the following ways:

- Rock formations provide reliable isolation over prolonged periods of time (during which long-lived radionuclides require isolation).
- Safety may be demonstrated on the basis of modern sanitary requirements, state standards, and construction norms and rules governing site selection, construction, operation, and maintenance of the underground facility.

All scientific research and prospecting efforts should be directed toward obtaining reliable data for design documents that will pass expert reviews at the regional, national, and international levels.

Figure 1 depicts a model of the life cycle of an underground isolation facility. The model also illustrates the application of the concept by which requirements for matrix materials, engineered barriers, and the geological formation itself must be based on the quantity and radionuclide content of the material to be stored. The region, district, and specific site for the facility are selected in accordance with established geological and environmental criteria.

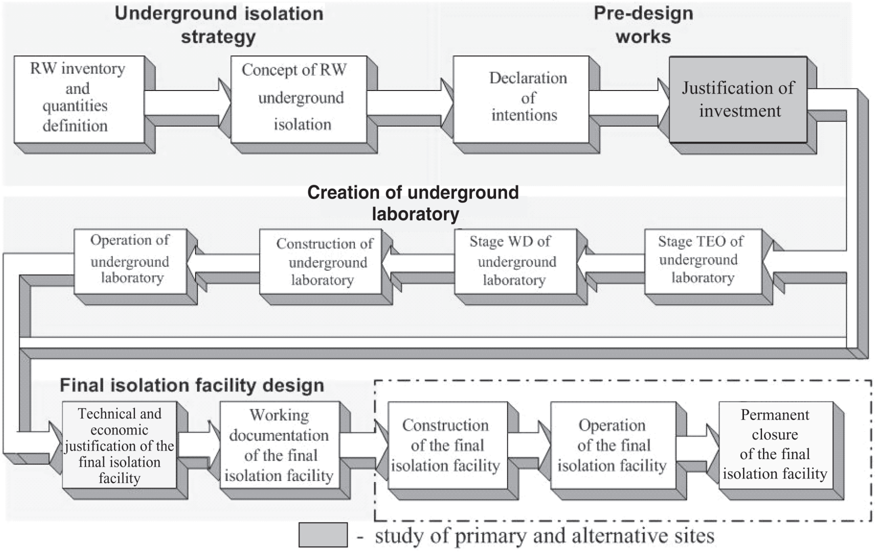


FIGURE 1 Model of the life cycle of a radioactive waste (RW) storage facility.

In geological engineering research it is necessary to take into account the entire range of legislation concerning the problem of managing radioactive wastes and spent nuclear fuel, as well as existing regulatory documents used in obtaining initial data for the design and construction of special underground structures. As shown in Table 1 the legislative and regulatory base for the creation of underground radioactive waste storage facilities includes the following related groups of documents:

- documents regulating activities at the federal level
- regulatory documents defining activities at all stages of prospecting, design, construction, and operation of the storage facility

The most important point in the process is the selection of the site where the underground facility will be constructed. A model for the selection of a promising site on the basis of programs was developed in accordance with the requirements of regulatory documents and with the correlation of the scope of research with the various design stages.

According to the regional approach that has been developed in Russia with regard to the selection of geologic sites for permanent isolation, it is most expedient to have the burial sites near the waste sources. Purposeful research for the high-level waste geological isolation has been done in the two areas where the Mayak Production Association (Chelyabinsk Oblast) and the Mining-Chemical

TABLE 1 List of the Basic Relevant Laws and Regulatory Documents Used in Creating Underground Radioactive Waste Storage Facilities at Russian Industrial Sites

Name of Document	Number, Date of Passage
Federal Laws	
1. Law of the Russian Socialist Federal Soviet Republic on Environmental Protection	No. 2060-1, February 19, 1991
2. Law of the Russian Federation on Environmental Impact Review	No. 174-FZ, November 23, 1995
3. Law of the Russian Federation on the Use of Atomic Energy, including amendments and additions of February 10, 1997	No. 170-FZ, November 21, 1995
4. Law of the Russian Federation on Radiation Safety for the Population	No. 3-FZ, January 9, 1996
5. Law of the Russian Federation on Amending Article 13 of the Federal Law on Environmental Impact Review	No. 65-FZ, April 15, 1998
6. Law of the Russian Federation on Making Additions to Article 50 of the Law of the Russian Socialist Federal Soviet Republic on Environmental Protection	No. 93-FZ, June 6, 2001
7. Law of the Russian Federation on Making Additions to the Federal Law on the Use of Atomic Energy	No. 94-FZ, July 10, 2001
8. Law of the Russian Federation on the Management of Radioactive Wastes	Bill submitted for consideration by the State Duma
Presidential Decrees	
1. On the State Strategy of the Russian Federation for Protecting the Environment and Ensuring its Sustainable Development	No. 236, February 4, 1994
Resolutions of the Government of the Russian Federation	
1. On Rules for Making Decisions on the Location and Construction of Nuclear Facilities, Radiation Sources, and Storage Points	No. 306, March 14, 1997
2. On Ratification of the Regulations on the Licensing of Activities Related to the Use of Atomic Energy	No. 865, July 14, 1997
3. On Ratification of the Regulations on Procedures for Conducting State Environmental Impact Reviews	No. 689, July 11, 1998
4. On Ratification of the Regulations for the Licensing of Activities Related to Environmental Protection	No. 168, February 26, 1996
5. On Ratification of the Rules for Making Decisions on the Location and Construction of Nuclear Facilities, Radiation Sources, and Storage Points	No. 306, March 14, 1997

Complex (Krasnoyarsk Krai) are located. There is a borehole polygon for liquid radioactive waste disposal in the reservoir beds at the Siberian Chemical Combine.

In the late 1970s in connection with the start of operations at the RT-1 Plant at Mayak in Chelyabinsk and the startup of the facility for steam reforming and vitrification, comprehensive research efforts were initiated for the first time in Russia to assess the suitability of promising sites for the burial of radioactive wastes nearby Mayak. As a result of this work, analyses were conducted on complexes of metamorphic and volcanic rocks and a number of intrusive formations both within the boundaries of the industrial zone at the complex and in adjoining areas. Further investigation at the site was justified.

Thanks to the joint research efforts of Minatom enterprises, Russian Academy of Sciences institutes, and geological organizations of the region, the structure, mineral and chemical composition, and tectonic disturbance of the geologic environment near Mayak were studied in detail. Volcanogenic rocks—tuffs and lava breccia porphyrites with negligible water permeability and high mechanical resistance and thermal conductivity—were found to be the most suitable for burial site purposes. Comprehensives studies of the geologic and hydrogeologic characteristics of these rocks at the Mayak industrial zone have been under way for more than 20 years. As a result of this work, which involved the drilling of 12 deep boreholes and more than 100 near-surface boreholes, scientists have carried out a wide range of experimental filtration work and field geophysics and thermophysics research. The lithological-petrographic composition and physical-mechanical, thermophysical, filtration, and other properties of various types of rock have also been determined under laboratory conditions. In the rock formation being studied several zones have been defined by depth with regard to their fracture toughness. The upper zone of high fracture toughness[1] is found down to a depth of 35 to 40 meters. The zone of weak fracture toughness, which lies from 40 to 100 meters below the surface, is characterized by unequal distribution of the network of open fractures. Below the depth of 200 meters, one finds monolithic rock, marked in some places by solitary cracks as well as sections having increased fracture toughness of up to 2.5 meters and a hydraulic conductivity of 10^{-3} to 10^{-4} meters per day.

After the results of all research studies were analyzed, two sites were selected as top priorities. On the basis of their hydrogeochemical characteristics, hydrodynamic subzones are being defined in these sites, with these subzones being characterized by the intensity of their water exchange processes and the depth of circulation of natural underground water.

Many years of research aimed at developing a preliminary basis for assessing the suitability of the sites under study for the underground isolation of radioactive wastes have confirmed that the sites are in fact promising.

A second Russian geological isolation site is the Nizhnekansk granitoid massif, one of the largest massifs in central Siberia with an area of more than

1500 square kilometers. It is composed of various types of magmatic and metamorphic rock, among which biotite granites, granodiorites, and gneisses are most common.

A rating system taking into account a set of geologic, technical-economic, engineering, and other criteria was applied to five sites selected by researchers from several organizations (Minatom, the Russian Academy of Sciences, and local geological organizations),[2] and as a result the two most promising sites were selected: the first,Verkhneitatsky and the second,Yeniseisky.

Engineering-geological and hydrogeological models of the sites were constructed, and forecast calculations were made of the velocity and filtration time for the various fracture toughness zones from the proposed depths for underground isolation to the surface zones for the dispersion of underground water.

In 2001 the Declaration of Intentions to Construct an Underground Laboratory was prepared and adopted in Krasnoyarsk Krai, with the above sites proposed as options.

In 2002 research at the Yeniseisky site was focused on evaluating the geological structure, degree of tectonic violation, and hydrogeologic conditions for identifying homogeneous and lightly violated blocks of native rock suitable for the construction of an underground laboratory and later a facility for the underground isolation of radioactive wastes.

Geological engineering studies were carried out on a site-specific basis and included the neotectonic mapping of 1008 square kilometers, regularly scheduled hydrological and meteorological observations, route mapping studies of 370 linear kilometers, helium surveys of 259 points, field analytical chemical studies of 85 samples, and emanation surveys of 1067 points.

In the course of geophysical work at the site, studies were conducted on five survey lines (each one 10 kilometers long) and three profiles (each one 7 kilometers long) using the following methods: magnetic surveying, electrical surveying using the audiomagnetotelluric sounding, multiparameter probe, and volatile enriched zone methods (130, 967, and 130 kilometers, respectively), seismic research (70 square kilometers), topographic-geodesic studies (70 kilometers), profile gravitation prospecting (70 kilometers), lab-based determinations of density and magnetic properties (118 samples), and preparation and description of polished core samples. Three boreholes have been drilled, each one 100 meters in depth.

Of all the promising sites in the Nizhnekansk granitoid massif, the Yeniseisky site, which has an area of 70 square kilometers, is the closest to the Mining-Chemical Complex (about 10 km away), the source of the wastes proposed for isolation. Its geologic structure is similar to the comprehensively studied massif of metamorphic rock in which the underground industrial facilities of the Mining-Chemical Complex are located (see Figure 2).

In the many years that the facility has been in operation, research has been conducted on the changes in the condition of the rock massif as a result of

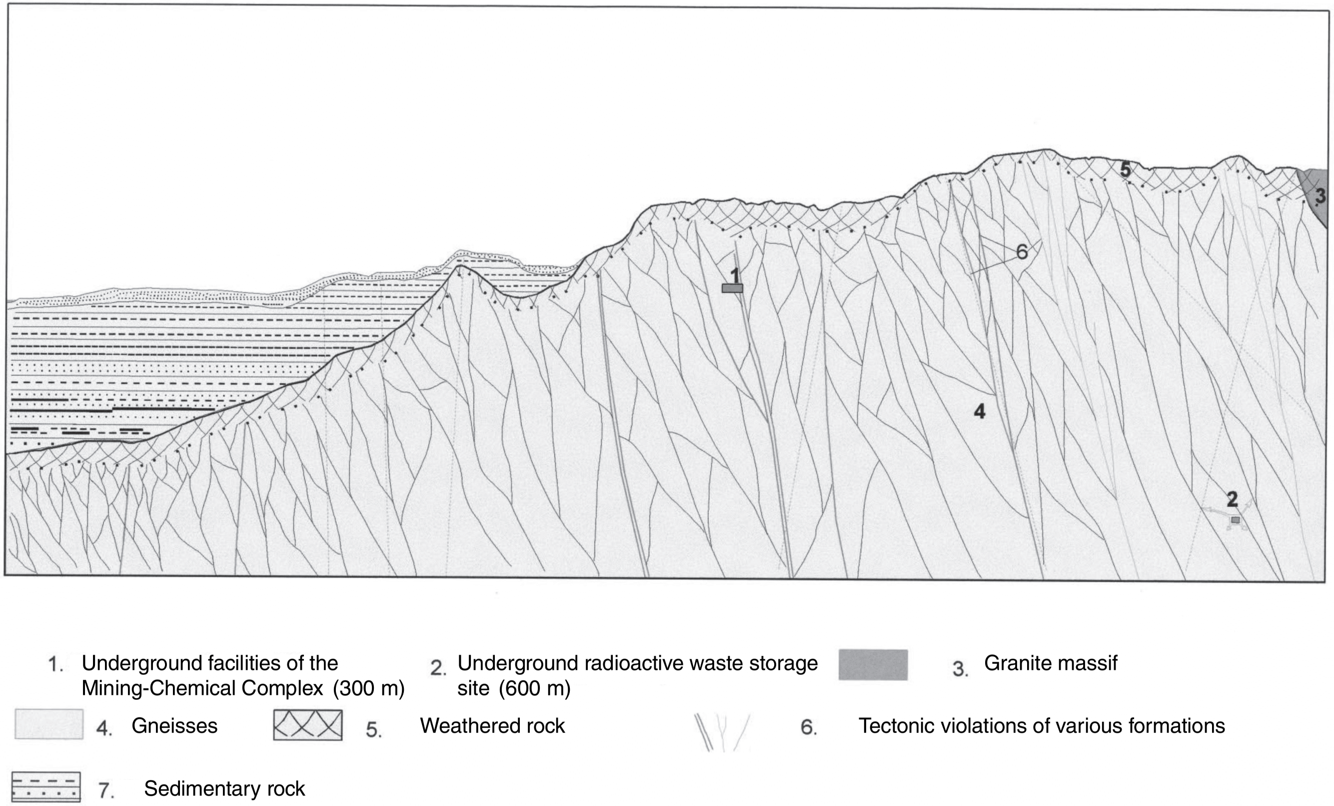


FIGURE 2 Location of underground laboratory and underground isolation facility.

natural (underground pressure, moisture, geodynamics) or industrial (for example, temperature fields) factors. Full results have been obtained through large-scale measurements of the parameters of various physical processes (geomechanical, hydrogeological, geochemical) underway in the rock massif as a result of 40 years of heat field effects. The correlations between rock condition parameters that have been found may be used as initial data in designing underground isolation facilities at the Nizhnekansk granitoid massif as well as similar facilities in any rock massif. (Prediction of the rock mass properties changes due to the natural and engineered load actions over an extended period of time.)

In order to facilitate the creation of long-range predictions a system of models for heat and mass transfer in a heterogeneous, weakly penetrable rock massif has been developed and tested. This system makes it possible to take into account the structural-tectonic characteristics of the site, the heterogeneity of the environment with regard to its heat properties, processes of underground water filtration by fatigue toughness rate, and changes of rock properties over time and space as a result of industrial and natural impacts.

The models also make it possible to predict the distance that the maximum permissible concentrations of any radionuclides will migrate as well as the heat field front, taking into account the rates of leaching from the matrices and of dissolution of radionuclides in underground water for redox conditions in the engineered barrier system and fracturing zones of the rock massif, the constructive features of underground objects, the sizes and properties of engineering barriers, the location of geologic violations,[3] and many other factors.

The unique underground facilities of the Mining-Chemical Complex currently provide an opportunity for the practical study of geophysical and geochemical processes that will occur in a rock massif during the construction, operation, and decommissioning of an underground isolation facility.

The experimental work currently being conducted is aimed at increasing our knowledge of hydrodynamic and geomigration parameters. Studies have been initiated in tectonic violation zones (crush and shear zones). Separate experiments have been set up in water-saturated and dry (but water-permeable) zones prone to crack formation. Periodic studies are also conducted regarding the mineralization of crack-vein waters in comparison with the mineralization of rain and melted snow.

Using a lab setup for studying the filtration of radioactive solutions through core models located in the underground conditions of the central factory laboratory, experiments are being conducted on various types of rocks, vitrified wastes, and real radioactive solutions at temperatures of up to 300°C[4] and pressures of up to 30 MPa. This makes it possible to simulate the behavior of a multibarrier isolation system under deep burial conditions.

In conjunction with experimental filtration work continuing at 20 boreholes, hydrodynamic and geomigration investigations have been conducted under natural conditions typical of a fracture-prone rock massif. As a result of the experiments under natural conditions, coefficients for diffusion and sorption and desorption rates have been obtained for complex radioactive solutions formed in the rock environment.

An analysis of the many years of field research regarding physical processes that determine the safety of underground isolation has made it possible to propose a new approach for determining the suitability of geologic sites for the long-term underground storage and burial of radioactive wastes and spent nuclear fuel, an approach based on the use of criteria for determining the risk that radionuclides exceeding maximum permissible concentration levels will migrate into the active water exchange zone. The new approach enables comparison of geological sites based on the prediction of their isolation characteristics dynamics during the required period of the radioactive waste isolation, according to the radioactive waste quantity and composition and the enclosing rock massif behavior. The experience of the organization that engaged in rock massif status investigations for more than 40 years for the ecologically dangerous Minatom facility placement is at the heart of this approach.

NOTES

1. The zone is most disturbed from surface to depth (40–100 meters). The depth is 4 meters at some points and 100 meters at others, and fracturing decreases with depth.
2. The basis for the ranking system is an ecological safety comparison for the environment taking into account the radioactive waste placement at the sites and different behavior of the rock massifs.
3. Fracturing zones or migration paths for the groundwater; radionuclides can only exit the rock through geologic violations.
4. This is the maximum investigation temperature; underground storage is not planned at a temperature high enough to essentially change a hydro-geological situation.

Concluding Observations

Milton Levenson
Bechtel International (retired)

It was not the objective of our workshop to achieve consensus on what is the best or optimum permanent solution to the spent nuclear fuel and high-level radioactive waste disposal problem. There are currently several alternatives—each with its own group of advocates. In some countries one option has been selected by policy officials while in other countries the decision has been deferred. It is not my intent to comment on the appropriateness of either selection by policy or deferral by policy but rather to give my personal observations on the content of the discussions at the workshop.

1. There were differing opinions as to what was the best or most appropriate final end point (for example, spent fuel geological disposal, reactor burn-out of actinide, accelerator burn-out of low-level fission products), but there was complete agreement that long-range storage and preferably centralized storage was required in all cases.
2. No one is against either reactor or accelerator burn-up or transmutation, but many have not decided on the practicality of it. This is probably due to the early stage of the science and lack of demonstration feasibility, which means estimates of cost are not credible.
3. There seemed to be concurrence that no matter how safe geological disposal is, it is prudent to minimize the amount of disposal to the extent that is practical.[1]
4. Over five decades of experience confirms that shipment of either spent fuel or high-level waste can be conducted safely although the advent of terrorism now causes a rethinking of the most appropriate methods of transportation.
5. Spent fuel and high-level waste are both currently being stored safely

under conditions that appear to be stable for as long as 50 years. The acceptable time may be much longer, but longer periods have not yet been validated.

6. Permanent solutions other than geological disposal of spent nuclear fuel, such as closed fuel cycles or transmutation, require significant research and development before they can be applied. It is not clear that funding at the levels required will be available to develop the technology in a timely manner. This supports observation 1 above.

7. Among the many considerations in the selection of a site the relationship of the chemistry of the host matrix to the chemistry of the waste form should be considered. In those cases where basic chemistry favors retention of fission products, plutonium, and actinides, natural barriers are important.

8. Since concerns over proliferation and terrorism, which were not discussed at the workshop, are current and since permanent solutions are in the future—an indeterminate time in the future—centralized, secure, long-term storage would appear to be of the highest priority.

NOTE

1. *Practical* includes consideration of costs and labor expense.

APPENDIXES

A

Workshop Agenda

Problems of Managing Spent Nuclear Fuel and Selection of a Site for Its Storage

Moscow, Russia, May 14–15, 2003

Agenda

Wednesday, May 14

9:30–10:00 Opening Session
- RAS Vice President Nikolay P. Laverov
- Representative of the U.S. National Academies, Milton Levenson
- Representative of The Russell Family Foundation, David McNelis

Session 1: Handling Spent Nuclear Fuel—International Experience

Position of IAEA on the Problem of Handling Spent Nuclear Fuel
Fyodor F. Sokolov, IAEA, presented by V. A. Lebedev, Minatom

Analysis of the U.S. Experience
John F. Ahearne, Sigma Xi: The Scientific Research Center, presented by M. Levenson

Spent Nuclear Fuel in Russia: Results of Research at the Bochvar Institute and Experience at Mayak
M. I. Solonin, Minatom

Approaches to Organization of Closed Nuclear Fuel Cycles in Russia
V. I. Rachkov, Minatom

Geological-Geochemical Basis of Creating a Long-Term Storage Site for Spent Nuclear Fuel
B. I. Omelyanenko, IGEM RAS

Feasibility of Transmutation of Radioactive Elements
Sekazi K. Mtingwa, MIT

High-Level Waste Disposal Technology Development Program
Jongwon Choi, Korean Atomic Energy Research Institute

Perspectives and Limitations of Transmutation in Reactors and With the Help of Accelerators
V. I. Matveev, GNT RF-FEI
A. V. Bychkov, NIIAR, Dimitrovgrad

Discussion

Session 2: Site Selection for Spent Fuel Storage and Disposal of High-Level Waste

Experience of European Countries
Charles McCombie, ARIUS

Site Selection for Above Ground Storage and Transportation
John D. Parkyn, Dairyland Power Cooperative

Experience of Japan
Koji Nagano, Central Research Institute of Electric Power Industry

Spent Fuel Management in Korea
Hyun-Soo Park, Korean Atomic Energy Research Institute

Problems of Safe Transportation of Spent Nuclear Fuel and High-Level Waste: International Experience
Michael E. Wangler, Transportation Safety Unit, IAEA

Experience in Transporting Spent Nuclear Fuel in Russia
Aleksandr M. Agapov, Minatom

Discussion

Thursday, May 15

Session 3: Problems in Establishing an International Storage Facility for Spent Nuclear Fuel in Russia

General Conception—Legal and Technical Aspects
V. A. Lebedev, Minatom

The Status of the Infrastructure for the Management of Spent Nuclear Fuel at the Mining-Chemical Complex
K. G. Kudninov, Mining-Chemical Complex, Minatom

Ecological Aspects
V. V. Kutsenko, Ministry for Natural Resources

Normative Requirements
A. M. Dmitriev, Gosatomnadzor

The Return to the Russian Federation of Fuel Assemblies from the Institute of Nuclear Physics of the Republic of Uzbekistan
Aleksey E. Lebedev, Techsnabexport

Possible Internal Budget Sources
Michael A. Zhdanov, Association for Nonproliferation and Ecological Improvement

Optimal Mining and Engineering-Geological Conditions: Analysis of Experience
N. N. Melnikov, Mining Institute, RAS

Conditions for Establishing a Storage Facility in Zhelznogorsk (Siberia)
Ye. B. Anderson, V. G. Khlopin Radium Institute, Minatom

Conditions for Establishing a Repository in Krasnokamensk (Baikal region)
Vasily I. Velichkin, IGEM RAS

Session 4: Utilization of High-Level Waste

Overview of High-Level Wastes and Their Characteristics
Valentin B. Ivanov, IGEM RAS

Chemical Preparation of High-Level Waste for Use
Boris F. Masoyedov, RAS Presidium

Immobilization of High-Level Waste: Analysis of the Feasibility of Synthetic Matrices
S. V. Yudintsev, IGEM RAS

Reprocessing High-Level Waste at Mayak: Plan for Establishing an Underground Laboratory
Yu. V. Glagolenko, IGEM RAS

A Model of the Life Cycle of the Repository RAO
Tatyana A. Gupalo, Scientific Research and Design Institute of Industrial Technology

Concluding Session

Discussion: Consideration of the Feasibility of Broad Cooperation in the Establishment of an International Repository for High-Level Radioactive Waste

Concluding Remarks
Milton Levenson
Nikolay P. Laverov

B

Environmental Effects of Radiation in the Russian Federation*

V. V. Kutsenko
Environmental Safety Administration
of the Russian Ministry of Natural Resources

Across the territory of Russia in 2002 the average concentration of radioactive substances in the layer of the atmosphere closest to Earth was practically unchanged from the levels in previous years; however, the potential danger from radioactive contamination and radiation accidents continues to exist.

Outside the regions contaminated as a result of the Chernobyl and Kyshtym disasters, average airborne concentrations of such radionuclides as cesium-137 and strontium-90 have practically returned to pre-accident levels. Practically everywhere except in those regions affected by these accidents, the gamma radiation exposure dose figures for 2002 were equal to background radiation levels.

The major radiation accidents of the past (Kyshtym in 1957 and Chernobyl in 1986) represent the clearest examples of violations of the stable condition of nature and sharp deviations in socioeconomic development. On the whole the period of the Cold War and the nuclear arms race left Russia with the most serious consequences in the form of radionuclide contamination of the natural environment, potentially dangerous atomic industry enterprises, and the lamentable results of the operating history of the nuclear submarine fleet of more than 30 years.

Rehabilitating areas contaminated by radionuclides requires decades of intensive work and capital investments comparable to those required for the creation of the modern nuclear weapons arsenal. At the same time we must realize that nuclear weapons cannot be eliminated in an instant, and neither can nuclear fuel cycle enterprises be shut down and dismantled or nuclear power be replaced by solar or other new power sources. Therefore, at present we must ensure the

---

*Translated from the Russian by Kelly Robbins.

environmentally safe operation of all industries involving the application of nuclear energy and radioactive isotopes.

As for the problems of ensuring environmental safety in the management of spent nuclear fuel and radioactive waste, the opinion of the Russian Ministry of Natural Resources is in support of projects and scientific innovations that facilitate the transformation of highly active nuclear materials containing long-lived radionuclides into short-lived materials. One temporary option is the creation of sites for the dry storage of spent nuclear fuel in special containers.

Taking into account that such projects are very expensive, each country must take steps on these questions to the extent allowed by its financial resources. However, in doing so countries must observe the recommendations of the International Atomic Energy Agency (IAEA) regarding radiation safety as well as national radioecological safety requirements.

ENVIRONMENTAL PROBLEMS OF STORING AND RECYCLING EQUIPMENT USED IN THE NUCLEAR POWER INDUSTRY

Potential sources of industrial radioactive contamination of the environment in Russia mainly include facilities entailing danger from radiation, such as mining, chemical, and radiochemical complexes, enterprises that enrich nuclear materials or produce or dismantle nuclear weapons, special complexes for the collection and reprocessing of radioactive waste, temporary storage points, nuclear power plants, research reactors, shipbuilding enterprises involved in repairing and decommissioning nuclear-powered ships, and facilities of the Russian Navy and civilian icebreaker fleets.

The most serious problem involved in ensuring radiation safety lies in the presence of enormous volumes of liquid and solid radioactive wastes at radiochemical enterprises, such as the Mayak Production Association, the Siberian Chemical Complex, and the Mining-Chemical Complex. More than 90 percent of all the radioactive waste in Russia is concentrated at these three enterprises. The conditions under which the waste is stored create a real threat to adjoining areas, especially if extreme natural or industrial situations were to occur. The efforts of scientific and technical development efforts must be focused on resolving problems related to the safe storage of radioactive waste, and the financing of these efforts must be given top priority.

The decommissioning of Russian nuclear submarines that have been and are being taken out of service is another serious current problem of global scope. The radioecological situation that has been created at naval facilities and in areas where nuclear submarines are based, operated, repaired, and decommissioned is critical and fraught with the possibility that extreme situations may arise at any moment. Eliminating the consequences of such situations could require financial expenditures comparable to those needed to deal with the effects of the accident at the Chernobyl nuclear power station.

In a time when massive numbers of nuclear submarines are being taken out of service, but the necessary financial and material resources for fully decommissioning and dismantling them are not being provided, it becomes unavoidable that these vessels will have to stand at anchor for long periods without their radioactive components being removed. This situation entails real nuclear and radioecological risks.

In the matter of reducing the level of danger presented by these sites, great significance must be attached to questions of ensuring environmental safety in the management of spent fuel and radioactive wastes, in the prolonged maintenance of reactor blocs in vessels at anchor, and in the long periods in which Russian nuclear submarines that have been removed from service are left standing without having their spent nuclear fuel removed. Meeting this challenge successfully depends not only on the amount of financing available but also on the technical capabilities of industrial enterprises to carry out an entire range of work involving radiation hazards.

The problem of managing liquid radioactive wastes has become somewhat less acute recently, and additional facilities for processing such wastes are being created in the Northern and Pacific fleets, including those created with the help of foreign investors (from the United States, Norway, and Japan).

A long path lies ahead if we are to resolve all the problems connected with radiation safety once and for all. The economic resources of the Russian state budget do not make it possible to resolve these problems in a compressed timeframe. Consequently, we must focus our efforts on seeking and putting into practice optimal technical innovations and low-waste technologies, making wider use of insurance mechanisms, and attracting foreign investment.

C

Geochemistry of Actinides During the Long-Term Storage and Disposal of Spent Nuclear Fuel[*]

Nikolay P. Laverov, Vasily I. Velichkin,
B. I. Omelyanenko, and *S. V. Yudintsev*
Institute of Geology of Ore Deposits, Petrography, Mineralogy,
and Geochemistry
Russian Academy of Sciences

The main source of high-level radioactive wastes (HLW) is the spent nuclear fuel (SNF) as a product of atomic power stations, as well as naval-propulsion and research reactors. Greater than 95 percent of total radioactivity of the materials involved in the human activity is concentrated in the SNF. The SNF contains 95–98 percent UO_2 and several percent of various radioisotopes formed in nuclear reactions. The SNF removed from reactors preserves greater than 90 percent of its energy resource. It contains greater than 96 percent U and up to 1 percent Pu, which are suitable for reuse. They are separated by radiochemical treatment of SNF with the formation of a large volume of liquid radioactive wastes. A part of the liquid wastes, small by volume but high in concentration of highly radioactive isotopes, belongs to high-level wastes. In accordance with current technologies, the HLW would be transformed to solidified form, stored for some time, and disposed in underground repositories.

The storage, recycling, and disposal of liquid radioactive wastes pose significant economic and environmental problems, and the expediency of SNF recycling is variously assessed. SNF is considered disposable waste in the United States, Canada, Sweden, and Finland. It is treated as recyclable nuclear material in Great Britain, France, and Japan. A decision about the SNF treatment has not been reached yet in Argentina, Brazil, and Slovenia. Switzerland plans to recycle one-third of its SNF in Great Britain and France and dispose of two-thirds of SNF (Schneider et al., 1998). It is probable that all countries will use SNF for

---

[*] Originally published in 2003, Geology of Ore Deposits 45(1):1–18; translated from Geologiya Rudanykh Mestorozhedeny 45(1):3–23. Excerpt reprinted with permission.

separation of valuable components after its controlled storage, because the ^{239}Pu, ^{235}U, and ^{238}U contents do not change in SNF while its radioactivity decreases. The fuel removed from reactors should be called irradiated and considered a potential energy resource, rather than waste.

In Russia some SNF types (from VVER-440, BN-600, and BN-350 reactors, some naval-propulsion reactors, and research reactors) are recycled at the RT-1 plant (Mayak in the Chelyabinsk region). Some other SNF types (from VVER-1000 reactors) are planned to be recycled at the RT-2 plant under construction (Zheleznogorsk near Krasnoyarsk). The SNF from RBMK, AMB, EGP-6, some transport facilities and research reactors, and faulty fuel is in storage, and its further treatment has not yet been considered. The primary portion of radioactivity (greater than 3 billion Ci) of SNF is accumulated in the RBMK reactors. This SNF has relatively low contents of fissionable species, and it will probably be recycled only in the far future. The problem of long-term safe storage and disposal of SNF in underground repositories is considered in this paper.

General geological recommendations for the long-term safe storage of SNF are deposition in the low-permeable rocks in seismically stable blocks with low velocities of vertical displacements and free of active volcanism and mineral deposits (Krauskopf, 1988a; Laverov et al., 2001). If these requirements have been met, the probability of the repository destruction, its exposure to erosion, transportation of radionuclides by magmas, and penetration of the repository by mining workings is minimized. The environmental pollution in this case can be related to radionuclide removal from the repository by groundwater in dissolved or colloidal forms. This mechanism is always taken into account in the assessment of the security level of HLW repositories.

SNF radioactivity decreases progressively during its storage (see Figure 1). For example, the radioactivity of SNF of a pressurized water reactor (PWR) (counted upon one ton of uranium) 10, 100, and 1000 years after its removal from the reactor will be approximately 400,000, 40,000, and 1700 Ci, respectively (Roddy et al., 1985). The strongest environmental impact would be expected if radionuclides escaped to groundwater at the earlier stages of SNF disposal, when the SNF contains short- and medium-lived radioisotopes with very high radioactivity. These isotopes decay in 500–1000 years. Current technology of long-term storage and disposal of HLW envisages HLW isolation from groundwater by engineered barriers for this or longer periods. The barriers comprise concrete tanks, corrosion-resistant containers, envelopes for nuclear fuel, and bentonite backfill. The SNF can interact with groundwater only after the engineered barriers lose their insulative properties. From this point on, actinides will present the main hazard for the biosphere. The geological medium will become the only barrier retaining the actinide migration. Below we analyze the conditions of safe SNF disposal provided by the insulative properties of the geological medium.

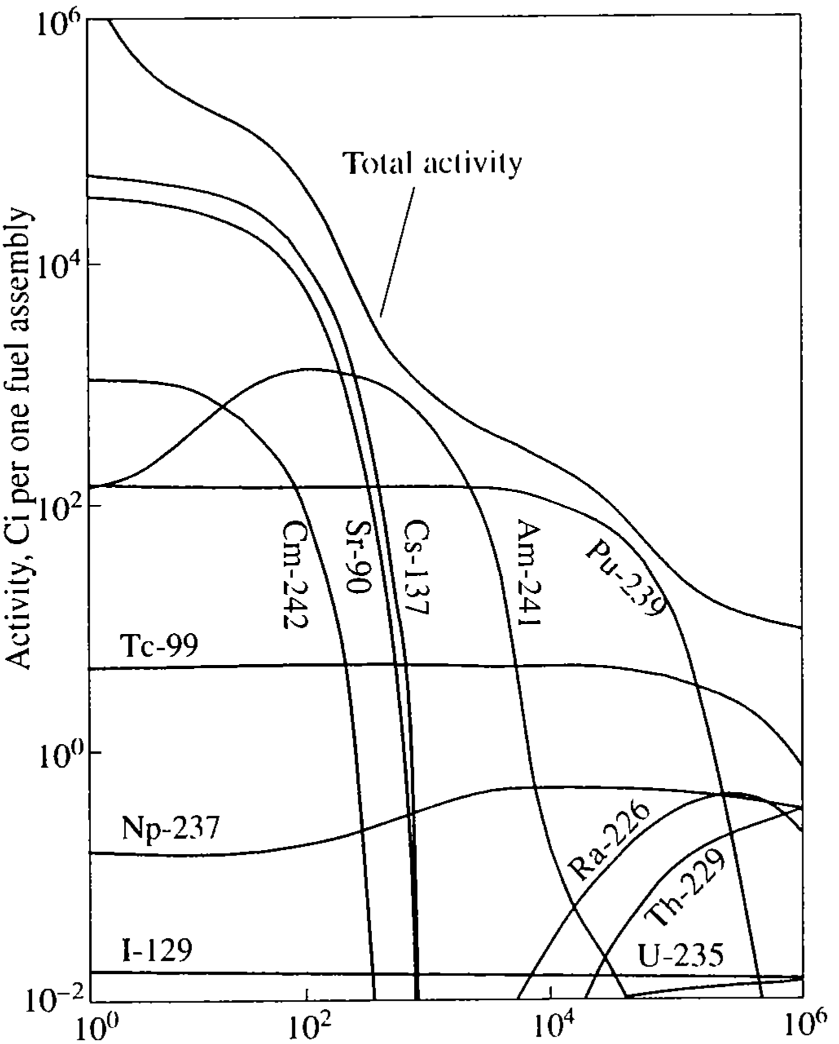


FIGURE 1 Variation of radioactivity of the SNF assembly of a PWR reactor with time (Brookins, 1984).

SPENT NUCLEAR FUEL PROPERTIES DEFINING THE CONDITIONS OF LONG-TERM STORAGE AND DISPOSAL

The principal constituent of the nuclear fuel is a heat-generating element (fuel rod) that is a long thin tube of corrosion-resistant zirconium alloy (or some other materials). The tube is filled with UO_2 pellets, with the proportion of ^{235}U isotopes several times higher than in natural minerals. Several fuel rods compose the fuel assemblies; for example, the fuel assembly of the RBMK reactor consists of 18 fuel rods, and the assemblies of the VVER-1000 reactor consists of 317 fuel rods. The pellets are produced by pressure and have a density of 94–95 percent of the theoretical density of uraninite. The UO_2 grains in pellets are smaller than a few microns. During irradiation the UO_2 pellets acquire numerous fractures, and the intergranular spaces expand. This results in an increase of surface area of the pellets and consequently UO_2 interaction with groundwater if the containers with SNF have failed.

The UO_2 is a conserving matrix for all elements produced in a nuclear reaction. Some elements (Pu, Am, Cm, Np, Th, rare earth elements [REE], Nb, and Zr) are incorporated into the UO_2 crystal structure. Its stability precludes release of these elements into groundwater. Some other elements (Tc, Se, I, Cs, Sn, and Sr) and their fission products are included as nonstructural admixtures. They are accumulated in the intergranular boundaries and microfractures in the UO_2 matrix. These elements can be partly leached by groundwater even in the conditions of the UO_2 stability. The leaching intensity will decrease with depletion of these elements in the areas in contact with groundwater.

In accordance with existing technologies for long-term storage and disposal, the SNF must be placed in metallic canisters and stored at depths of several hundred meters from the surface. Various schemes of SNF storage in underground workings and wells have been proposed (see Figures 2 and 3). According to these schemes the engineered barriers comprise bentonite backfill, canister, capsules, and fuel-rod coatings. These barriers must isolate the UO_2 pellets from groundwater for 500–1000 years. The short- and medium-lived radioisotopes will decay during this period, and 98 percent of the residual radioactivity of SNF will be related to Pu and Am (see Figure 1). When the geological medium becomes the only barrier retaining the radionuclide release, the repository safety in most cases will be determined by the intensity of the Pu and Am escape from the SNF and the specific conditions of their migration in groundwater. The UO_2 stability is very important for repository security, because Pu and Am occur as isomorphous impurities in this mineral. The analysis of this problem considers the data on U behavior in the geological medium and experimental results on dissolution of SNF and natural uraninite in groundwater.

URANIUM DEPOSITS AS NATURAL ANALOGUES OF SNF REPOSITORIES

The analysis of natural observations is crucial in the study of problems related to the long-term safe underground disposal of actinide-bearing wastes. It allows one to characterize the actinide behavior at various chemical conditions and obtain information about very slow processes with results notable only after thousands and millions of years.

Many researchers consider the U and Th deposits as natural analogues of the

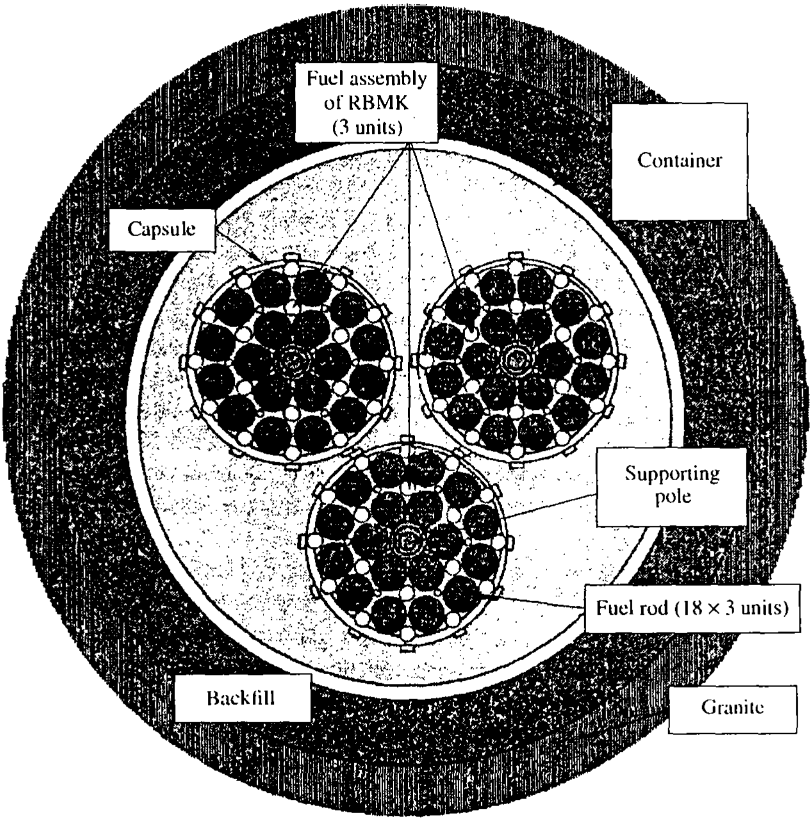


FIGURE 2 A scheme of the RBMK reactor SNF location in a well (Ivashkin et al., 2000).

HLW repositories (Brookins, 1984; Krauskopf, 1988b; Laverov et al., 1991, 1994). This is particularly appropriate for the SNF containing greater than 95 percent UO_2, which presents the conserving matrix for the other radionuclides. The study of U deposits shows that uranium oxides can be highly stable at certain conditions in the geological medium. Ores of U deposits are generally composed of uranium oxides. The ore bodies are usually located in highly permeable zones composed of cataclasites, densely fractured rocks, or water-saturated sedimentary rocks. Therefore, a high portion of U minerals of the ore bodies may have contacted with groundwater, but many U ore bodies stored for hundreds of millions of years are almost unaltered, even in the zones with high water permeability.

It was found that the occurrence of reducing or nearly neutral groundwater is the principal factor facilitating the high stability of U minerals. The groundwater attains such properties during its continuous interaction with country rocks, when its salinity decreases and oxygen is spent for oxidation of ferrous iron (Kiryukhin et al., 1993; Krainov and Shvets, 1992; Ryzhenko et al., 1996). The oxygen-rich water normally penetrates to depths from several tens to a few hundreds of meters and reaches 1.0–1.5 km only in the Alpine regions. We suggest that the conditions of high stability of UO_2 occur at depths below 500 m in most regions (except lofty mountains) (Laverov et al., 2001).

It is important that the reduced groundwater is always saturated in uranium derived from country rocks. Uranium in the rocks occurs as

1. disseminated U atoms and their clusters
2. accessory U minerals
3. isomorphous impurity in the U-bearing accessory minerals
4. U-rich segregations along grain boundaries
5. atoms sorbed on specific minerals (Omelyanenko et al., 1983)

The equilibrium uranium concentration in groundwater at the reducing hydrogeochemical conditions is very low, less than or equal to 10^{-8} mol/L (see Fig-

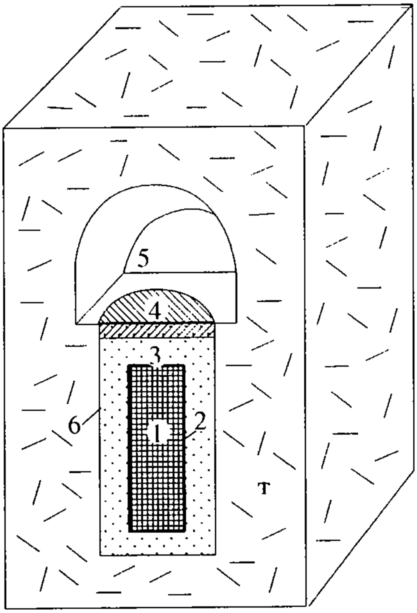


FIGURE 3 A scheme of the SNF location in an underground mining working. (1) SNF, (2) metallic canister, (3) bentonite backfill, (4) concrete seal, (5) horizontal working, (T) cell.

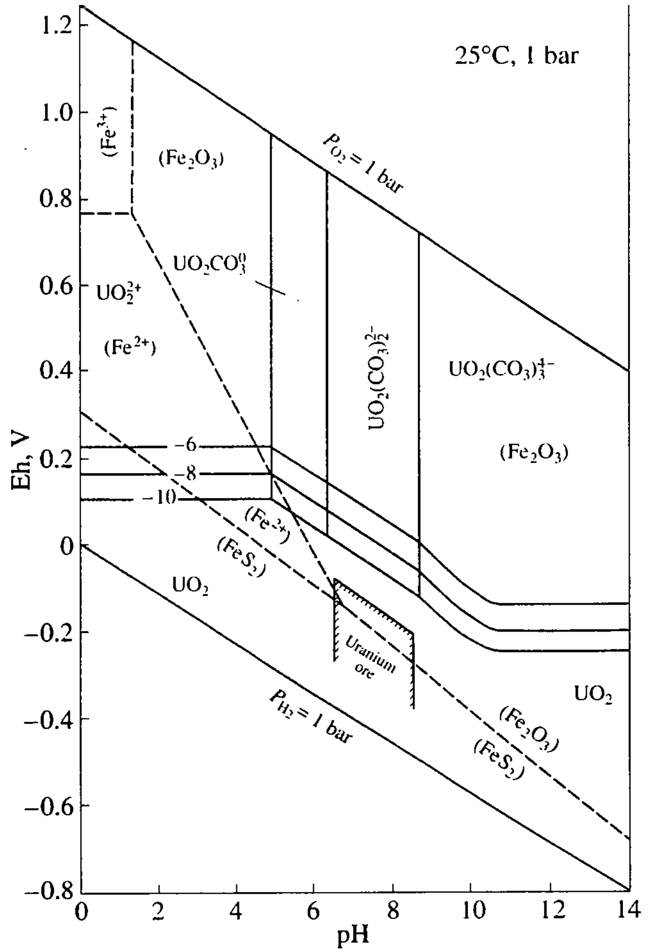


FIGURE 4 Eh-pH diagram for the U-C-O-H system and a part of the Fe-S-O-H system (Brookins, 1988). Activities of the dissolved particles: U = 10^{-6}, $^{-8}$, $^{-10}$, C = 10^{-3}, Fe = 10^{-6}, and S = 10^{-3}. The contour boundary of the field of Eh-pH values of groundwater is in equilibrium with unoxidized uranium ores.

ure 4). At such conditions uranium is redistributed among various minerals and within single grains. The proportion of sorbed U increases at the expense of the other forms. These processes are most efficient in the cataclastic zones, where the interaction area and proportion of secondary minerals (the best sorbents for U) increase. The Fe(Ti)-bearing minerals and particularly their alteration products serve as sorbents for U in crystalline rocks. Similar processes resulting in the formation of the secondary dispersion halos occur around the uranium ore bodies. These processes are very slow (but continuous) and their results become notable only over geologically long time intervals. The diffusion-driven redistribution cannot significantly affect the radionuclide release from SNF.

The Cigar Lake deposit in Northern Saskatchewan, Canada, is frequently referred to as a natural analogue of an SNF repository (Johnson and Shoesmith, 1988). The average U content in its ores is 7.9 wt percent (up to a few tens of percent in the richest blocks; Pagel et al., 1993). The deposit is dated at 1.1 Ga. Rich ores lie at a depth of 400 m, but no signs of their occurrence are seen on the surface. The ores are cut by water-permeable fractures, which may indicate that the ore contacted groundwater for a long time. The uraninite is only slightly altered by the reducing properties of groundwater. The U concentration in water from an ore body with 40 percent U is 10^{-8} mol/L and is almost equal to the background concentration (Cramer, 1986).

It is pertinent to note that the data on higher U concentrations in groundwater either correspond to the oxidizing conditions or characterize total contribution of dissolved and colloidal forms. This is also verified by experiments on uraninite dissolution under the reducing conditions, when U concentrations were determined both in the unfiltered liquids and in the solutions that passed through the filters with 2 nm pores. The results of natural observations convincingly indicate a high stability of uraninite at reducing, almost neutral hydrogeochemical conditions.

The question arises: If uraninite is irradiated in the fuel rod, does it behave in the geological medium like natural uraninite, because the intensity of an irradiation on the SNF surface is two to three orders of magnitude higher than in the uranium ore? Let us consider the data on the deposits of the Franceville uranium ore district in Gabon in West Africa to solve this problem. Oklo is the most famous deposit of the district. Some indications of natural nuclear reactions were discovered in ores of this deposit, that is, depletion of some ore bodies in ^{235}U and occurrence of radioisotopes or their fission products resulting from nuclear reaction. The deposits of the Franceville ore district formed at about 2 Ga in sandstones with organic matter at depths of $(3–3.5) \times 10^{-3}$ km. It was calculated that the ^{235}U content in ore at that time was 3.25 wt percent, which is close to that in fuel of modern power reactors. Processes similar to those in nuclear reactors occurred intermittently for about 500 Ma in some Oklo ore bodies in the presence of water serving as a neutron moderator (IAEA, 1978).

The study of these ore bodies located at different depths from the surface described the behavior of uraninite and associated products of nuclear reactions both in the reducing and oxidizing hydrogeochemical conditions; for example, the uraninite in an ore body at a depth of 250 m under reducing conditions is not affected by secondary processes (Pourcelot and Gauthier-Lafaye, 1998). Detailed studies of trace-element concentrations and isotopic compositions in such ores demonstrated that the elements incorporated in the uraninite crystal structure were preserved there until their complete decay.

Such behavior of Am, Pu, and Np is justified by the distribution of ^{209}Bi and ^{207}Pb, the final products of their fission, which were generally preserved in the uraninite, regardless of their dissimilar geochemical properties to uranium, and were only partially redeposited near the ore bodies (Brookins, 1984). The nonstructural elements, such as Rb, Cs, Sr, Mo, Cd, Xe, and I were almost completely eliminated from the ore bodies, while Ru and Sn were partially removed. It was suggested that diffusion was the main mechanism of migration of the nonstructural elements (Cowan, 1978).

Geochemical studies of natural reactor zones indicate that uraninite is highly stable under reducing conditions. It not only retains actinides but also strongly restrains migration of the nonstructural elements. Thus, the study of the Oklo ore bodies shows that the geological medium can provide the conditions for the long-term safe storage of SNF.

The interaction with oxygen-rich water facilitates the U removal and replacement of U oxides by the secondary uranyl-ion minerals. The upper parts of some steep ore bodies are strongly altered, while their lower levels are well preserved. The removal of uranium together with all mixtures from uraninite was detected in the Oklo ore body, which is located at a depth of 100 m and shows evidence of nuclear reactions (Pourcelot and Gauthier-Lafaye, 1998). The strongest decomposition of uraninite was found in the Bangombi zone located at a depth of 10 m from the surface directly beneath the lateritic weathering mantle. In this zone the uraninite was strongly modified, the organic matter in the host rocks was oxidized, and the reducing properties of these rocks declined. A part of the uranium removed from uraninite was redeposited as phosphates. The formation of secondary ore accumulations with hexavalent U is typical of the deposits located near the surface. These data, together with numerous examples of partial and complete transformation of uranium ores of many other deposits, indicate that the oxidizing conditions are inappropriate for the long-term underground storage of SNF.

EXPERIMENTAL STUDY OF SPENT NUCLEAR FUEL DISSOLUTION IN GROUNDWATER

Many experiments on interaction of natural and synthesized UO_2, as well as SNF and its imitators, and groundwater were performed under oxidized conditions and are related to the U.S. project on long-term HLW storage in the Yucca Mountain repository in Nevada (LLNL, 1998). Seventy thousand tons of HLW (generally SNF) are supposed to be stored in this repository. The experimental results show high SNF solubility in aerobic conditions (U content reached 10^{-3} mol/L) and a high rate of release of all radionuclides from the spent fuel. Scientists attempted to follow all stages of uraninite transformation under the effect of oxygen-rich water.

During its interaction with water uraninite was oxidized from the surface and along the microfractures and grain boundaries. The intergranular spaces, and consequently the interaction area, increased. The oxidized uraninite microparticles (UO_{2+x}) were separated from the main mineral mass. Secondary phases of the hexavalent U, for example, haiweeite, schoepite, and soddyite, formed as suspension on the SNF surface during the longest runs. If the Yucca Mountain repository were located in the zone with conditions of continuous interaction with groundwater, uraninite would not serve as an immobilizing matrix; however the repository is located in a tuff sequence in the aeration zone above the groundwater level. The assessment of the repository safety is based on the suggestion that without approaching the SNF, water will vaporize for several hundred years because of elevated temperature. The conditions of probable interaction of SNF with water will be reached only after the temperature decreases below 90°C. It is suggested that water will approach the SNF intermittently and only in small amounts because of the dry climate and scarce atmospheric precipitation in the repository area. Thus, the safety of the Yucca Mountain repository is controlled by the amount of atmospheric precipitation instead of by the low solubility of UO_2.

It was calculated that one SNF assembly (3140 kg) UO_2 will interact with less than 20 liters of water per year. Even in such conditions about 0.1 percent of total ^{129}I, ^{99}Tc, and ^{137}Cs amounts will escape in a year (LLNL, 1998; Chen et al., 1999). A larger ^{99}Tc release (up to 0.8 percent per year) is also reported (Finn et al., 1998). The geochemical isolation of Np in such conditions is also problematic (Buck et al., 1998; Chen et al., 1999). Generally, because of the oxidizing conditions, some experts believe that the long-term safe storage of SNF in the Yucca Mountain repository is not possible (Ewing, 2002). It is emphasized that the water volume interacting with SNF can significantly increase as a result of climatic changes or some other reasons. It is pertinent to note that Yucca Mountain is the only repository with aerobic conditions projected for long-term storage and disposal of HLW.

Numerous experiments have demonstrated that uraninite solubility is very low (U concentration is less than or equal to 10^{-8} mol/L) under the reducing almost neutral conditions and independent of temperature or composition of groundwater (Johnson and Shoesmith, 1988; Redkin et al., 1989). The extent to which the data on solubility of the natural uraninite are applicable to SNF should be especially considered. The natural uraninite and UO_2 of SNF are analogous in their crystal structure but differ in impurity elements, O/U ratio, and radioactivity. The natural uraninite grains are highly dense, and their boundaries are indistinguishable even under an electron microscope. SNF uraninite is broken by numerous microfractures; the boundaries among its grains in the SNF are wider and better seen than in natural uraninite aggregates.

Let us compare experimental results on solubility of natural uraninite and UO_2 in SNF of various types. The deionized water and the water in equilibrium with granites, clays, salts, and other rocks were used as dissolvents. The experiments showed that SNF is stable under the reducing conditions (Johnson and Shoesmith, 1988). Under similar conditions the solubility of natural uraninite, nuclear fuel imitators, and SNF are of the same order of magnitude. Insignificant differences in the solubility of individual uraninite samples are generally related to their oxidation degree: the higher the O/U ratio, the higher the solubility. The SNF solubility in water in equilibrium with atmospheric Ar and N strongly depends on the SNF-oxidation degree and is 10 times higher for the oxidized SNF than for unoxidized (Loida et al., 2001). The authors believe that SNF can be oxidized during its storage. The material of steel canisters and their corrosion products serve as inhibiting agents for SNF dissolution; for example, the solubility of the oxidized SNF decreased 20 times in equilibrium with iron powder. The radionuclide concentration in the solution in this experiment was comparable with that in runs performed under the reducing conditions. Thus, the experiments show that the reducing conditions maintain high SNF stability in the geological media.

THE EFFECT OF RADIOLYSIS ON SPENT NUCLEAR FUEL SOLUBILITY

The SNF radiation does not necessarily result in reducing conditions. The radiation can cause water dissociation on the SNF surface. Although the oxidizing (O_2, H_2O_2) and reducing (H,) components are produced in equal molar quantities, the higher diffusion rate of hydrogen can lead to the formation of the local oxidizing conditions near the SNF surface, which facilitate the UO_2 dissolution by the reaction

$$UO_2 + H_2O_2 \leftrightarrow \left(UO_2^{2+X}\right) + 2OH^-$$

The possible effect of radiolysis on radionuclide release was emphasized by many researchers (see, for example, Brookins, 1984; Chapman and Savage, 1984; Chapman and McKinley, 1988; Krauskopf, 1988b). They noted, however, that radiolysis is not capable of significantly modifying the scale of radionuclide escape because of the large reducing capacity of the geological medium.

Rocks and backfill materials continuously supply ferrous iron to groundwater. The interaction of groundwater with a metallic canister and its corrosion products with hydrogen separation by the reaction

$$3Fe + 4H_2O \leftrightarrow Fe_3O_4 + 4H_2$$

also counteract the formation of the oxidizing conditions.

The question of which processes will prevail on the SNF surface, oxidation due to radiolysis or reduction due to interaction with the ambient medium, is very important for the SNF disposal. This problem cannot be solved solely on a theoretical basis.

The experiments take into account that the SNF interaction with water will begin only after the engineered barriers have failed, that is, in ≥500 years), the main mass of β sources has decayed, and the main process capable of changing the redox conditions is radiolysis caused by α radiation. The α-particle tracks in water are about 30–50 μm long, and radiolysis can occur only in a thin water film on the SNF surface. It is induced by no more than 1 percent of all α-particles (Neretnieks, 1997). It was found that the radiolysis does not affect the uraninite solubility in the reducing conditions; for example, the runs performed over a year at 70°C in equilibrium with hydrogen verified a high stability of SNF under the reducing conditions (Spahiu and Sellin, 2001). The uranium concentration in groundwater from granite was less than10^{-9} mol/L, and the other radionuclides had not been leached from SNF. The effect of radiolysis is best pronounced in experiments in an oxygen-free atmosphere of nitrogen and argon. The role of a radiolysis of groundwater in the formation of the oxidizing conditions on the SNF surface in an underground repository was proved to be insignificant (Forsyth and Werme, 1986); however the experiments with the intensity of α radiation being one to two orders of magnitude higher than that on the SNF surface showed that radiolysis-induced oxidizing conditions can exist locally (Johnson and Shoesmith, 1988).

New experimental data on the effect of radiolysis on the SNF solubility were obtained recently. Samples of pure uraninite and uraninite with 0.1 and 10 percent ^{238}Pu isotope (a source of α particles) were used in these experiments (Rondinella et al., 1999, 2001). Specific activities at the surface of the samples doped with ^{238}Pu were 2.71×10^6 and 2.71×10^8 Bq/cm^2, which are one and three orders of magnitude higher than on the SNF surface 500 years after its removal from the reactor. The runs were performed with deionized water in equilibrium with a nitrogen atmosphere. The Eh values in the 60-day runs with uraninite with 0, 0.1, and 10 percent ^{238}Pu were 180, 380, and 570 mV, respectively. Equilibrium concentrations of uranium were less than 10^{-8} mol/L in runs with pure UO_2 and about 10^{-6} in runs with uraninite with ^{238}Pu. It was found that the dissolved uranium was mostly redeposited on the vessel walls. The amount of the redeposited uranium increased proportionally with the increasing run duration and ^{238}Pu concentration in uraninite. The experiments showed that the uranium release from uraninite doped with ^{238}Pu is comparable to that from pure uraninite in the oxidizing conditions. Plutonium concentration in the solution was only 4×10^{-12} mol in runs with uraninite containing 0.1 percent ^{238}Pu and four orders of magnitude higher in runs with uraninite containing 10 percent ^{238}Pu.

As it follows from the experiments, active radiolysis can produce highly oxidative conditions when plutonium passes into the penta- and hexavalent states; however the canister material and products of its corrosion preclude the formation of such highly oxidative conditions even at very high SNF radioactivity. A very high rate of radionuclide leaching independent of the groundwater composition was observed in experiments on the groundwater interaction with SNF powder in equilibrium with argon atmosphere (Loida et al., 2001). Radiolysis was the main process that induced the oxidizing conditions and the correspondingly high rate of SNF dissolution. Equilibrium element concentrations in water were 5.4×10^{-5} mol/L for U and 1.5×10^{-6} mol/L for Pu. After addition of iron powder to the system, the rate of SNF dissolution decreased by 20 times, equilibrium concentration of U in the solution decreased by two orders of magnitude, and that of Pu, Am, Eu, Np, and Sb by more than three orders of magnitude, which corresponds to the values observed under the reducing conditions. Note that in the 805-day runs iron was not oxidized completely, and the iron particles contacting the SNF particles were replaced by hematite (FeOOH). Thus, the reducing conditions were maintained by reactions of iron oxidation. The experimental results demonstrated that at the radiation level typical of SNF after 500 years of storage, radiolysis does not cause the transition of the tetravalent Pu to a more oxidized state.

Smith and Johnson (2000) estimated the effect of radiolysis on SNF stability from the experimental data on UO_2 solubility, theoretical calculation of the amount of the oxidizing agents produced by SNF interaction with water, and the study of natural uraninite. Admitting that precise estimation was impossible, the authors concluded that the presence of reducing agents in host rocks and canister corrosion products strongly limits the radiolysis effect on the SNF solubility. The oxidizing species resulting from radiolysis will be spent primarily for oxidation of bivalent iron.

This conclusion is also justified by the absence of uraninite oxidation in many deposits. Uranium in the Oklo ores always occurred in the tetravalent state in spite of radiolysis. The organic matter and bivalent iron in host rocks and ores maintained the reducing conditions (Oversby, 2000). Local oxidizing conditions related to radiolysis were found near the uranium ore in some deposits (Cigar Lake, Canada). The low alteration degree of uraninite in these deposits shows that the oxidizing species were spent generally for oxidation of wall rock minerals with reducing components (Liu and Neretnieks, 1995).

Available data indicate that radiolysis cannot significantly affect the radionuclide migration from SNF if the host rocks and engineered barriers contain reducing species. The radiolysis effect needs further study, however. It is necessary to analyze the situation of high solubility of the uraninite matrix due to the local oxidizing conditions in the zone of SNF interaction with groundwater. In this case, the matrix cannot isolate actinides. Let us consider the capability of the geological medium to isolate actinides stored in repository from the biosphere.

MIGRATION AND ACCUMULATION OF ACTINIDES IN GEOLOGICAL MEDIUM

Crystalline rocks are the most probable medium for long-term SNF storage in Russia and most other countries. Safe physical isolation of SNF from interaction with groundwater in crystalline rocks is impossible, because the rocks are intersected by fractures, and their amount can increase because of many factors. The most probable mechanism of biosphere pollution in this case is actinide migration from the repository with groundwater flows. The water-exchange intensity controls the rate and scale of the radionuclide migration from SNF. According to the general recommendations the repositories should be constructed in conditions of low water exchange. The interaction of groundwater with SNF in such conditions can be estimated in long-term experiments reaching steady concentrations of dissolved radionuclides (Wilson, 1990 a,b) and by results of the SNF study after its interaction with water (Finn et al., 1998b). It was found that the SNF transformation begins with uraninite oxidation accompanied by the widening of grain boundaries and detaching of tiny UO_{2+x} particles from the grain surfaces. This process results in the formation of the secondary uranyl-ion phases on the SNF surface and areas loosened by oxidation, which inhibit further corrosion. Haiweeite, uranophane, and soddyite were identified there. The rate of uranium migration from SNF is 0.1–0.3 mg/m^2 per day. The major part of the leached uranium is redeposited in the secondary uranyl-ion phases. The steady concentration of uranium in water is $(4–8) \times 10^{-6}$ mol/L, 98 percent of which occurs in colloidal form. Pu, Am, and Cm pass into water proportionally to U, while the mechanism of Np release is not yet understood. It was also found that the SNF type and degree of the fuel decay do not principally affect the rate of UO_2 dissolution. On the contrary, the UO_2 oxidation degree strongly influences the rate of its dissolution. For example, U_3O_8 dissolves two to three times faster than UO_2. This is generally caused by the fact that the uraninite oxidation is accompanied by the formation of microfractures, thereby increasing the water penetration and interaction area.

During the dissolution of the uraninite matrix, actinides can pass into water in dissolved and colloidal forms, precipitate near SNF as isomorphous impurities in the secondary uranyl-ion minerals or as individual mineral phases, and accumulate in sorbed forms in host rocks. The actinide migration in dissolved form is inhibited by sorption of these elements by rocks. Thus, a more real mechanism of the actinide migration to the biosphere is due to their removal from the repository in colloidal form. Some brief information on radioactive colloids is given below.

Three mechanisms of formation of actinide-bearing colloids are distinguished. The first mechanism is related to actinide sorption by colloidal particles abundant in groundwater. Such particles form by groundwater interaction with minerals, mechanical and chemical weathering of rocks, detaching of the particles from the minerals covering the fracture walls, or due to the changes in the hydrogeochemical conditions that result in oversaturation of solutions with respect to some minerals. The colloidal particles are from 1 nm to 1 µm in size. Actinides can migrate only with smaller particles (less than 400 nm), because the larger particles rapidly precipitate. The colloidal particles normally consist of clay minerals, chlorite, silica, iron hydroxides, zeolites, and organic matter. The amorphous phases of Si, Al, and Fe play a significant role among the colloidal particles. The content of the colloidal phases in groundwater varies significantly. The groundwater typical of granites has a constant temperature, flow velocity, ionic strength, pH, and Eh and normally contains less than 100 ng/ml colloidal particles. The disturbances in the hydrodynamic regime caused by drilling, mining, or water pumping can increase the amount of colloidal particles originated by detachment of mineral particles (including the coagulated colloids) from fracture walls. The concentration and composition of the colloidal particles near the repository can differ from the background values, which is a phenomenon related to the temperature gradient and introduction of extraneous materials into the geological medium.

The second mechanism is the formation of true radiocolloids. The radioisotopes released from the uraninite structure during its dissolution pass into saturated aqueous solution as suspended and colloidal particles, normally of amorphous hydroxides. The formation of true colloids can be favored by a decrease of actinide solubility in groundwater because of buffer reactions reducing the effect of radiolysis.

The third mechanism of the colloidal particle formation generally works in the disposed vitrified HLW. The products of their interaction with groundwater are represented by various crystalline and amorphous phases with radionuclides in isomorphous and sorbed forms. Separation of such phases from the glass surface and their migration as colloidal particles is the most probable mechanism of actinide release from the vitrified HLW. The colloidal particles from the uraninite matrix corrosion products form in similar manners during the long-term storage of SNF. The colloids can be composed of UO_{2+x} particles separated from the SNF surface or of secondary uranyl-ion minerals containing actinides in isomorphous and sorbed forms.

It was found that the migration velocity of actinides dissolved in water is two to three orders of magnitude lower due to sorption than the velocity of the water flow. On the contrary, the velocity of the colloidal particle migration is close to the velocity of the water flow. In the steady-state hydrodynamic conditions the relation between the amount of colloidal particles that entered the groundwater and were removed from it is close to equilibrium. The amount of colloidal particles can strongly increase because of tectonic activity, water sampling, mining, or some other processes disturbing the hydrodynamic regime. The main mechanism restraining the migration of colloidal particles in geological medium is their adhesion and precipitation on the fracture walls as loose aggregates. Aging of the colloids favors the particle consolidation on the fracture walls. Note that the rocks contain only natural analogues of actinides (U, Th, REE) and do not contain actinides themselves, which can be derived only from SNF. As a result, the actinide concentration in groundwater decreases with increasing distance from SNF. This favors the transition of actinides from colloidal particles to aqueous solution followed by their further sorption by rocks and other colloidal particles.

The low water permeability and water exchange, the absence of wide fractures, and the sealing of fractures by secondary minerals also counteract the migration of the actinide-bearing colloids. The bentonite backfill is an insuperable obstacle in the way of colloid migration. It was shown experimentally that the bentonite layer 2 mm thick stems the colloidal particles, and only dissolved actinides can migrate to some longer distances (Tsukamoto et al., 1995). That is because most of the modern projects of the HLW repositories envisage the use of a bentonite backfill.

When modeling the radionuclide migration in a colloidal form, it is difficult to constrain the time of occurrence of colloidal particles in groundwater and the distance of their transportation. Some suggestions should be made for calculations. For example, it was implied that for the Yucca Mountain repository, half of the colloidal particles precipitate from water to the fracture walls in 50 days (Ahn, 1997). The calculation showed that the actinide-bearing colloidal particles can migrate a distance of 50 m from the site of their formation in 30,000 years, but the reliability of this estimate and its acceptability for the other geological conditions are still uncertain.

Some reliable data indicate that actinides can migrate in colloidal form for significant distances and with high velocities (Kersting et al., 1999). The water samples collected from depth intervals of 701–784 and 1046–1183 m within the area of underground nuclear tests in Nevada contained actinide-bearing colloidal particles. The ^{240}Pu/^{239}Pu ratio testifies to the origin of these isotopes from radioactive glass produced by the nuclear explosion in 1968 at a depth of 1402 m and at a distance of 1.3 km from the sampling site. The major portion of actinides is confined to colloidal particles from 7 nm to 1 μm in size composed of illite, smectite, and zeolite. These minerals are typical products of secondary alteration of host rhyolites and tuffs. The dissolved actinides compose only 1 percent of the total actinide amount. Thus, the predominant actinide migration in colloidal form suggested from experiments was verified by natural observations.

PLUTONIUM MIGRATION IN GEOLOGICAL MEDIUM

Plutonium contents in uranium ores are negligible (Pu/U is approximately 10^{-12}). It forms there when ^{238}U atoms capture neutrons. This element is most dangerous among the SNF actinides (Korenkov, 1992). The ^{239}Pu capability of maintaining a chain reaction requires conditions excluding critical-mass formation in the HLW repositories. It was demonstrated that the HLW forms used for storage and disposal preclude the formation of critical masses in individual packages and their assemblies (Allen, 1978). Data analysis shows that the probability of critical-mass formation due to selective leaching and redeposition of ^{239}Pu in or near an SNF repository is very low (Kastenberg et al., 1996; Oversby, 1998).

This property of plutonium, together with its high toxicity, should be taken into account in projects of long-term storage of SNF.

If uraninite is dissolved and actinides pass from SNF to groundwater, the repository can be considered secure only if any one of the following conditions is met: (1) the concentration of Pu (and other actinides) in groundwater contacting with SNF is below the maximum concentration limit for drinking water or (2) the SNF actinides passed into groundwater are retained within the restricted volume of the geological medium, and only refined water suitable for domestic use reaches the zone of active water exchange. To estimate the possibility of realization of these conditions, we used the available information on the properties of synthesized actinides and their natural geochemical analogues, experimental results on SNF interaction with groundwater, data on actinide solubility in aqueous solutions and interaction of actinide-bearing solutions with rocks, experimental results obtained in underground laboratories on velocities of migration of elements that can and cannot be sorbed in fractured rocks, and information on the colloidal form of radioisotope migration with groundwater.

To predict the behavior of plutonium in the SNF under reducing conditions, we used data from the Oklo deposit. Plutonium, until its complete fission, was retained in this deposit in the uraninite matrix, regardless of its interaction with groundwater. The conservation of Pu and other actinides at the site of their formation was maintained by the reducing conditions and the high stability of uraninite under such conditions. This conclusion is also consistent with experimental data. The Pu concentration in water from granite or clay in equilibrium with SNF at 90°C under the reducing conditions is only 10^{-11} mol/L (Lelous et al., 1998). This value is close to the maximum concentration limit for water of domestic use. The highest Pu concentrations possible in groundwater under the oxidizing conditions can be estimated by experimental data on SNF dissolution in groundwater, which were obtained within the framework of the project for the HLW disposal in the Yucca Mountain repository (LLNL, 1998). The groundwater from tuffs with $pH = 7.6$ and $Eh = 0.77$ V was used as a dissolvent. It was found that, during the SNF dissolution, plutonium passed into water in dissolved and colloidal forms, precipitated on the vessel walls as solid phases, and was incorporated into the secondary uranium minerals. A high proportion of plutonium occurred in colloidal form.

The highest Pu concentration in dissolved form is approximately 2×10^{-8} mol/L at 25°C, which is close to the solubility of the amorphous $Pu(OH)_4$ in groundwater and seawater (approximately 6×10^{-8} mol/L at 25°C) (Kulyako et al., 2001). The dissolved Pu concentration is significantly lower at 85°C (approximately 2×10^{-11} mol/L) due to extensive precipitation of solid phases. In experiments on the SNF interaction with groundwater in the noble gas atmosphere (any external oxidizing or reducing agents were absent), the redox conditions were generally controlled by radiolysis. The 545-day runs in sealed ampoules filled with Ar or mixtures of Ar and CO_2 indicated a high SNF solubility in groundwater of various compositions (Loida et al., 2001). For example, the element concentrations in groundwater from granites (pH = 7–8) were 5.4×10^{-5} mol/L for U and 1.5×10^{-6} mol/L for Pu. It was found using a 1.8 nm filter that 96 percent U and 99 percent Pu occurred in colloidal form. The element concentration in water in equilibrium with iron powder decreases by two orders of magnitude for U and by more than three orders of magnitude for Pu, Am, Np, and Eu. The authors emphasize that these concentrations are comparable with those obtained under the reducing conditions.

The above data indicate that the Pu concentration in groundwater interacting with SNF under oxidizing conditions can be as high as $n \times 10^{-8}$ mol/L for the dissolved form and up to 10^{-6} mol/L for totally dissolved and colloidal forms. These values are two to four orders of magnitude higher than the maximum concentration limit for drinking water. Therefore, the Pu solubility in groundwater is not low enough for SNF repository safety with respect to Pu in oxidizing conditions.

Geochemical analogues of plutonium under reducing conditions are thorium and uranium. All three elements occur in the tetravalent state under such conditions, and their dioxides (PuO_2, ThO_2, and UO_2) are stable solid phases with very low solubility. Under the weakly and moderately oxidizing conditions, plutonium occurs in the tetravalent state and behaves like thorium, whereas uranium passes into the highly mobile hexavalent state. Plutonium can pass into the penta- and hexavalent states and behave like uranium only in highly oxidizing conditions (see Figure 5[a]), which are not possible in the SNF repositories. Unlike uranium and thorium, plutonium responds to the increase of the solution acidity by transition into the relatively mobile trivalent state in a moderately acidic medium. Acid water can form only in zones of sulfide ore oxidation or in active volcanic areas. In both cases the high acidity of the medium is caused by the formation of sulfuric acid solutions by oxidation of sulfide sulfur. The geological conditions in which the formation of such solutions is possible are unsuitable for SNF storage and disposal. The pH values of groundwater of crystalline rock massifs typically are 8 ± 1.5.

Thermodynamic modeling shows that plutonium under strongly reducing conditions (Eh is approximately –500 mV) at pH = 8 occurs generally in the trivalent state; tetravalent plutonium dominates at Eh from –100 to +150 mV; while penta- and hexavalent plutonium prevails under the strongly oxidizing conditions close to the boundary of water stability (Berry et al., 2002). Under the Eh and pH conditions typical of groundwater in crystalline rock massifs, plutonium occurs generally in the tetravalent state and behaves like thorium. The information on the thorium geochemistry in the geological medium is summarized below. These data are important for prediction of the behavior of plutonium.

Thorium belongs to hydrolyzates, shows a constant valence of four, and does not participate in redox reactions (see Figure 5[b]). Thorium can reach significant concentrations in acid solutions. The formation of the $Th(OH)_4$ col-

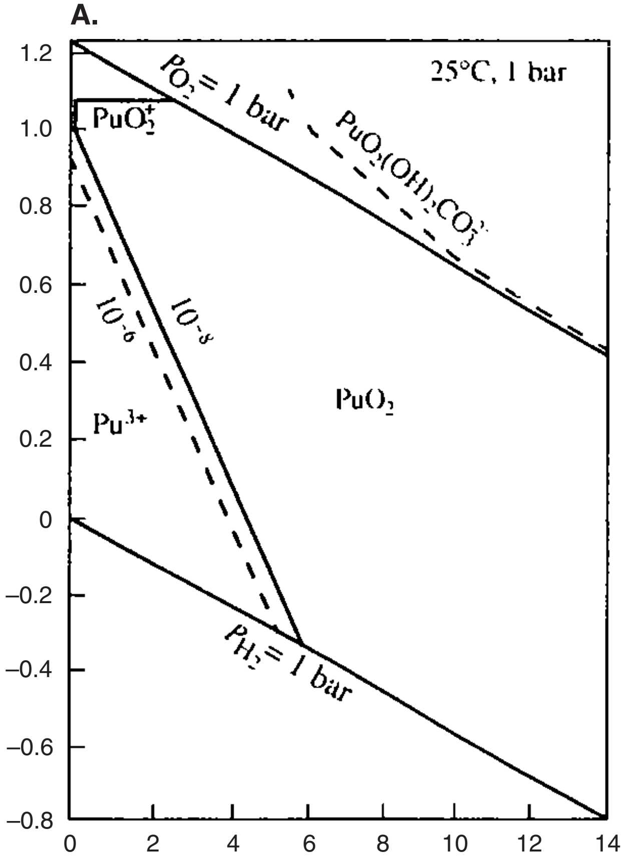
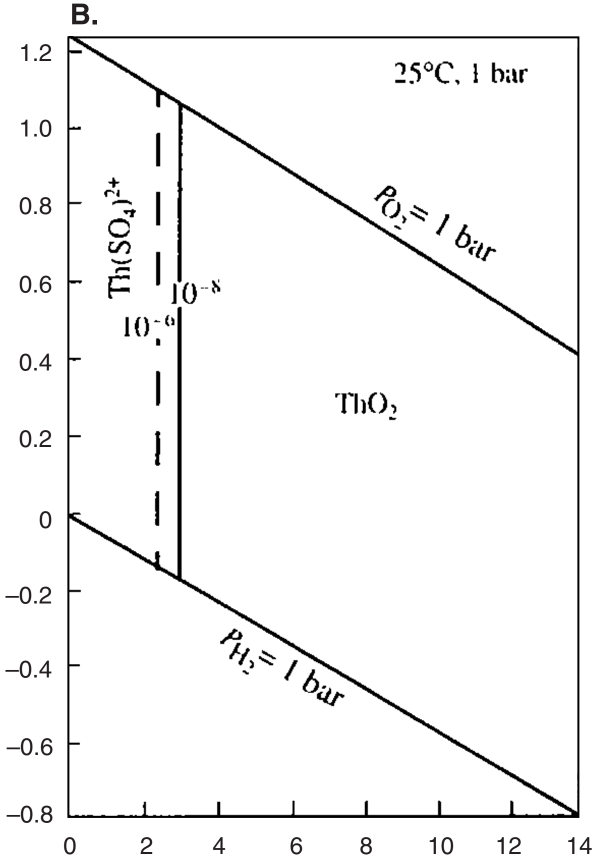


FIGURE 5 Eh-pH diagrams for the Pu-C-O-H (a) and Th-S-O-H (b) systems. Activities of the dissolved particles: Pu = 10^{-8}, C = 10^{-3}, Th = $10^{-6,\ -8}$, and S = 10^{-3} (Brookins, 1988).

loids followed by their precipitation begins at pH greater than 3.5. Aging of the sediment is accompanied by the irreversible reaction

$$Th(OH)_4 \rightarrow ThO_2 + 2H_2O$$

Thorium can form complex ions (hydroxocomplexes, carbonate complexes, as well as complex compounds with organic acids). Thorium is readily sorbed by Fe-Ti minerals and products of their alteration. Thorium is two to four times more abundant in rocks than uranium. Its concentration in groundwater is one to



three orders of magnitude lower, which determines significantly lower natural mobility of Th compared with U. The Th concentrations in groundwater vary from $n \times 10^{-11}$ to $n \times 10^{-7}$ mol/L. Its highest concentrations are typical of acid sulfate water from the oxidation zone of REE deposits rich in Th. The Th concentration in groundwater below the zone of extensive hypergenesis never exceeds 10^{-9} mol/L, even in blocks with Th ore mineralization. Thorium in ground, river, and sea water generally occurs in suspended and colloidal forms. Minerals with high Th concentrations, for example, thorite, thorianite, monazite, and zirconolite, form by crystallization from magmas or alkaline and acidic high-temperature hydrothermal solutions. These minerals are stable in natural conditions. Thorium-bearing minerals decompose only in chemical weathering in acidic conditions. Thorium can pass into aqueous solutions by incongruent leaching or complete dissolution. The study of weathered rocks indicates a very slow decomposition of these minerals (Eliseyeva and Omelyanenko, 1987). Thorium removed by acid leaching can be partly redeposited in a sorbed form below the kaolinite layer; this is related to the increase in pH values in groundwater with increasing depth.

The analysis of natural observations helped us to conclude that the interaction of the Th minerals with fracture water in the crystalline rock massifs below the hypergene zone cannot result in significant migration of thorium in a dissolved state from the disposal site. By analogy with thorium showing geochemical similarity to plutonium, we suggest that groundwater can have very low contents of dissolved plutonium whose migration will be retained by sorption and that plutonium can be transported for significant distances only as colloidal particles.

Like thorium, plutonium can be extensively sorbed by minerals. Many experiments simulated Pu sorption from aqueous solutions with rock powders. The partition coefficient values (K_d) significantly vary depending on the compositions of rocks and aqueous solutions, pH and Eh values, temperature, and some other factors. K_d values almost always exceed 100 cm^3/g and in many cases are a few thousand or even a few hundred thousand cubic centimeters per gram. K_d values are 550 cm^3/g for sand, 1200 cm^3/g for loam, and 1800 cm^3/g for organic soil (Ways of Migration, 1999). K_d values are higher under reducing conditions than under oxidizing conditions (Brookins, 1984). The effect of rock composition and CO_2 concentration (Baston et al., 2000), as well as redox conditions (Berry et al., 2002), on the Pu sorption was also studied. The basalt, sandstone, and argillite powders (less than 0.25 mm fractions) mixed with water in the proportion 1:5 were used in experiments. K_d, values ranged from 1000 to a few hundred thousand cubic centimeters per gram. The experimental results cannot be accounted for only by the influence of the studied parameters. It is more probable that the samples contained various amounts of minerals with high sorptive capacity, for example, Fe, Ti, Mn oxides and hydroxides, and organic matter; for example, the oxides concentrate up to 60 percent of the total plutonium sorbed in tuff, although the oxide content in this rock is only 1 percent (Vaniman et al., 1996).

It is pertinent to note that when characterizing the potential sorptive capacities of various rocks and minerals, the experiments with powders do not reflect real conditions of the water-rock interaction and the actual insulative properties of geological media. Unlike the experimental conditions, only a part of crystalline rocks adjacent to the water-conducting fractures participates in sorption. Experiments in underground laboratories demonstrate that radionuclides can move away from a fracture for less than 2 mm (up to several millimeters along microfractures and brecciation zones) (Neretnieks, 1993). The microfractures increase the water-rock interaction area and stimulate sorption.

Experiments on interaction of Pu-bearing solutions with plate-, cube-, or disk-shaped rock samples are also important in this context. It was found that plutonium was mostly concentrated on the sample surfaces and only partly penetrated inside along microfractures. The more prolonged the interaction, the more plutonium penetrated inside the sample along microfractures and pores (Zakharova et al., 1998). Desorption experiments indicate a stable immobilization of plutonium in microfractures.

Many experiments on Pu sorption and desorption on core samples from igneous rocks (mainly quartz monzonites) of the Canadian Shield were also performed (Vandergraaf and Abry, 1982). The groundwater from granites used in the experiments contained approximately 5×10^{-14} mol/L Pu with 19.5 percent ^{237}Pu. In this case the partition coefficient was expressed in centimeters, because K_d values in runs with samples characterized the ratio of the element content per 1 cm^2 of the sample area to its concentration in 1 cm^3 of solution ($g/cm^2:g/cm^3$). The surface area of the samples in these experiments was 9.5 cm^2; the solution volume 10 cm^3; and the run duration up to 28 days. K_d of Pu ranged for various samples from 1.4 to 70.0 cm with an average value of 14 cm.

It was found that the mafic minerals sorb plutonium better than quartz and feldspars. Consequently, the mafic rocks can sorb more plutonium than the felsic rocks. Thus, the crystalline rocks present reliable barriers for migration of dissolved Pu. The velocity of the Pu plume propagation is hundreds and thousands of times lower than the groundwater flow velocity (Krauskopf, 1998a). The distance that the dissolved plutonium can migrate in groundwater strongly depends on the water flow velocity, which is highly dissimilar in crystalline rock massifs.

The experimental results obtained in underground laboratories show that the weakly sorbed elements migrate along some fractures with a velocity of a few tens of meters per year, whereas the migration of the easily sorbed elements is much slower than the water flow (Neretnieks, 1993). The easily sorbed elements, such as Cs, Sr, Eu, Nd, Th, and U, did not pass the distance of 5–10 m after six months and were immobilized near the injection site. The increasing duration of the experiments resulted in deeper penetration of the easily sorbed elements into the rock matrix along the microfractures and did not considerably change the migration distance. These results can be extended to plutonium, which also belongs to the most easily sorbed elements. The above data indicate that the geological medium can prevent the Pu transition to the biosphere in dissolved form.

The Pu migration in colloidal form seems to be more hazardous for the environment. $PuO_2 \cdot H_2O$ and $Pu(OH)_4$ prevail among the true radiocolloids. Their role is insignificant, because Pu is extensively sorbed by minerals in rocks and colloids in groundwater. The groundwater colloids are most important for Pu transportation. Based on the suggestions that half of colloidal particles in groundwater precipitate on the fracture walls in 50 days and that the Pu-bearing colloidal particles can migrate distances of less than 50 m from the site of their formation, Ahn (1997) calculated that the Pu concentration on the fracture walls near the source in 30,000 years will be 0.025 kg/m^2. This is much lower than the critical concentration of Pu; however, the distance and scale of the Pu transportation in colloidal form are not known definitely.

Most researchers believe that Pu cannot be transported for significant distances either in dissolved or in colloidal forms; these suggestions are not consistent with the above-mentioned fact of Pu migration for 1.3 km (Kersting et al., 1999). Plutonium concentration in water in this case is 10^{-14} mol/L, and 99 percent of the total Pu amount is confined to colloidal particles from 7 nm to 7 μm in size, which are composed of illite, smectite, and zeolites. The repository safety with respect to Pu can be maintained generally by the factors preventing colloid migration, that is, low water permeability and water exchange, the absence of wide fractures, sealing of the fractures by secondary minerals, and the presence of bentonite backfill. Additional factors increasing the insulative properties of geological medium are the mafic composition of crystalline rocks and the distant location of the repository from the site of groundwater discharge (Laverov et al., 2001).

AMERICIUM MIGRATION IN GEOLOGICAL MEDIUM

Americium is the second element (after plutonium) among the actinides with risk to the environment (Korenkov, 1992). By the time of the container failure in 500–1000 years, Am and Pu will account for 98 percent of the total radioactivity of SNF. The ^{241}Am isotope will play the major part among the actinides (see Figure 1). The general knowledge of the Am behavior in geological media can be deduced from the data on its geochemical analogues.

Am has similar geochemical properties to Nd and Eu, whose average abundances in the crustal rocks are $n \times 10^{-3}$ and $n \times 10^{-4}$ percent. The Nd and Eu in rocks and minerals compose from 10–25 to 0.5–2 percent of the total rare earth element amount. The highest contents of these elements are typical of the alkaline massifs. Nd and Eu occur in many minerals as isomorphous impurities. They are mobile in high-temperature alkaline solutions, which is evidenced by the close relation of most rare earth element deposits with high-temperature alkaline metasomatites. Rare earth elements in these deposits are accompanied by Nb, Ta, Th, Zr, and U. The rare earth elements are also mobile in acid media, where they migrate as sulfate and fluoride complexes. Rare earth element contents in uraninite are variable. The uraninite from the Oklo deposit contains 0.39 wt percent Nd_2O_3. The accessory minerals, such as monazite, orthite, and xenotime are the main concentrators of Nd and Eu. These minerals are stable to interaction with groundwater, are preserved during mechanical weathering, and are accumulated in sands. These accessory minerals undergo dissolution with removal of poorly soluble elements (rare earth elements, Th, Nb, Ta, and Zr) only during chemical weathering by organic acids and accompanied by the formation of kaolinite layers. The solubility of these elements decreases with neutralization of the unconfined water, and they accumulate in the zone of rock disintegration (Burkov and Podporina, 1967). Sorption on secondary minerals is the principal mechanism of this accumulation.

The data on Nd and Eu concentrations in groundwater are scarce. The element concentrations in the CO_2-rich water passed through a 100 nm filter ranged in $(0.42–7.55) \times 10^{-10}$ mol/L for Nd and $(0.026–0.82) \times 10^{-10}$ mol/L for Eu (Michard et al., 1987). The Nd and Eu concentrations in water from a uranium mine are 6.5×10^{-10} and 0.3×10^{-10} mol/L, respectively (Ivanov, 1997). The water samples from granite massifs of the French Pyrenees collected in deep wells and thermal springs (pH = 9.0, $T = 75°C$) and passed through a 450 nm filter contain 7.1×10^{-9} mol/L Nd and 3.5×10^{-11} mol/L Eu and decrease by one order of magnitude after passing the samples through a 10 nm filter (Michard et al., 1991). The authors concluded that the colloidal forms of these elements were dominant in the studied samples. Among the natural waters in the active volcanic areas worldwide, the highest Nd (7×10^{-7} mol/L) and Eu (2×10^{-8} mol/L) concentrations were found in the acid sulfate water (pH = 1.33, $T = 43°C$) passed through a 100 nm filter (Michard, 1989). The concentrations of these elements are four orders of magnitude lower in nearly neutral water. It was demonstrated that Nd and Eu concentrations in water are independent of the rock composition but strongly depend on the solution acidity and increase with decreasing pH. The hydrothermal rock alteration typically does not result in REE leaching (Sturchio et al., 1986); for example, a rhyolite alteration with strong redistribution of major elements does not change REE concentrations. Rare earth elements were leached from the rock-forming minerals during the rhyolite alteration and were sorbed in the secondary clay minerals and zeolites. Based on the above data, we can suggest that the Nd and Eu concentrations in groundwater do not exceed $n \times 10^{-9}$ mol/L and $n \times 10^{-10}$ mol/L, respectively, and are much lower in most cases. The Nd and Eu concentrations can exceed these values only in the specific conditions of intensive weathering of the REE ores or due to an ultra acidic composition of the solution. In most cases the main part of these elements in groundwater occurs in colloidal form. REE sorption on minerals is the leading factor retaining their migration in dissolved form. The elevated activity of the complex ions in groundwater stimulates the REE transition from the isomorphous to the easily sorbed forms and facilitates REE accumulation in the suspended particles with high sorptive properties. This phenomenon can be responsible for the correlation between the carbonate-ion and REE concentrations in groundwater, as well as the domination of the colloidal form. Similar behavior in geological medium can be suggested for Am. Some additional facts supporting this conclusion are given below.

Americium can occur only in the trivalent state within the whole interval of hydrogeochemical conditions typical of the HLW repositories. The trivalent form is most stable even in the near-surface conditions (for example, in soils) (Ways of Migration, 1999). The tetravalent state is possible only in the strongly oxidizing alkaline conditions, which are unreal for the HLW repositories. Americium is rapidly hydrolyzed in water, and its equilibrium concentration in aqueous solution is determined by $Am(OH)_3$ solubility. The latter can be represented by crystalline and more soluble amorphous phases. The $Am(OH)_3$ solubility strongly depends on the composition and properties of groundwater. The CO_3^{2-} and SO_4^{2-} ions, as well as humic and fulvic acids, increase the $Am(OH)_3$ solubility because of the formation of specific complexes. The highest mobility of americium is typical of the circulation zones of acid sulfate water, which usually forms during the oxidation of sulfide deposits or in active volcanic areas. Such conditions are unsuitable for the construction of HLW repositories. $Am(OH)_3$ solubility is very low in the nearly neutral reducing conditions typical of the deep zones with low water exchange.

The hydroxocomplexes and carbonate complexes probably are the most abundant soluble forms under such conditions. According to Pokrovsky (2003), the percentage of the dissolved Am forms in seawater with pH = 8 are as follows: 43.2 for $AmCO_3^+$, 22.4 for $Am(OH)^{2+}$, 15.0 for $Am\left(OH\right)_2^+$,12.0 for $Am\left(CO_3\right)_2^-$, 4.1 for Am^{3+}, 1.5 for $AmCl^{2+}$, 1.1 for $Am\left(CO_3\right)_3^{3-}$, and 0.8 for $AmSO_4^+$. Taking into account a great abundance of carbonates in rocks and CO_2 dissolved in groundwater and the typically nearly neutral alkaline properties of groundwater, it is possible that the carbonate complexes will be the dominant dissolved species and stable solid $Am(OH)(CO_3)$ will occur together or instead of $Am(OH)_3$ under natural conditions. The solubility of these compounds in groundwater is rather high. For example, the Am concentrations in dissolved and colloidal forms in groundwater from rhyolite tuffs (pH = 8.6) in equilibrium with $Am(OH)(CO_3)$, $Am(OH)_3$(cryst), and $Am(OH)_3$(amorph) are 1.7×10^{-8}, 3.7×10^{-5}, and 6.5×10^{-4} mol/L, respectively (LLNL, 1998). It is very difficult to determine the concentration of the dissolved Am alone because of its easy sorption on the tiny colloidal particles. Passing the solution through the filters decreased the Am concentrations in all experiments. The finer the filters, the more Am was retained on them. The experiments on $Am(OH)_3$ solubility in groundwater from the Gorleben area in Germany showed a stable Am concentration of $10^{-6.3}$ mol/L (IAEA, 1992). The Am concentration in the aqueous solution decreased after it passed through filters with decreasing pore sizes. After a 1 nm filter, it decreased by almost four orders of magnitude and reached 10^{-10} mol/L. The specified values characterize the Am concentrations in water in equilibrium with highly soluble phases and are the highest possible concentrations. They considerably exceed the maximum Am concentration limit for drinking water (2×10^{-12} mol/L).

The data on Am concentrations in groundwater interacting with SNF is of great interest for current purposes. Let us emphasize that Am is incorporated in the UO_2 crystal structure and its transition into the aqueous solution strongly depends on the uraninite solubility: The experimental data on SNF interaction with aqueous solutions indicate that Am is not leached if uraninite is stable and passes into solution congruently with U and other actinides during uraninite dissolution. In the Oklo deposit uraninite was stable because of the reducing conditions, and Am, like Pu, was retained at the site of the mineral formation until its complete fission.

The experimental data help us to suggest possible Am concentrations in groundwater under the oxidizing conditions induced by radiolysis. The Am concentration in the solution was $(2.5–3.6) \times 10^{-8}$ mol/L after the interaction of SNF with a high degree of fuel decay with groundwater from granites (pH = 7–8) after 545 days in closed ampoules filled with Ar or mixtures of Ar and CO_2 (Loida et al., 2001). After ultrafine filtration, 99 percent of the total Am amount was retained on a 1.8 nm filter. The authors believe that microcolloidal particles smaller than 1.8 nm can also occur in the solution.

Many experiments on SNF interaction with groundwater in conditions open with respect to atmospheric oxygen were performed within the frames of the project for the HLW disposal in the Yucca Mountain repository (LLNL, 1998). SNF fragments 2–3 mm in size from reactors of various types and groundwater from rhyolite tuffs with pH = 7.6 were used in experiments. The runs were performed in closed and open ampoules at temperatures of 25°C and 85°C over several months. In these cases UO_2 composing 98 percent of the SNF and acting as an immobilizing matrix for the other radionuclides could not maintain their secure isolation, because the groundwater had oxidizing properties and was significantly undersaturated with respect to U. The experiments showed that the Am concentration in water rapidly increased in the beginning of the experiment, reached a maximum, and then decreased to become constant at a certain level. The authors explain the concentration decrease by the formation of secondary uranyl-ion phases on the grain surfaces. These phases have lower solubility under oxidizing conditions as compared to uraninite.

The equilibrium Am concentration in the solutions passed through a 400 nm filter was approximately 1.5×10^{-10} mol/L. After ultrafine filtration through a 1.8 nm filter, the Am concentration decreased by five times, which indicates the prevalence of the colloidal form. The measured Am concentrations were notably lower than the concentrations expected in equilibrium with $Am(OH)CO_3$ or $Am(OH)_3$. The solid phases controlling the Am solubility were not found in the material retained by filters. The authors believe that the smaller-than-expected Am concentrations could be related to high concentrations of lanthanides in SNF, whose secondary phases can confine Am as an isomorphous impurity. The equilibrium Am concentration was two orders of magnitude lower at 85°C than at 25°C. This fact indicates a more rapid formation of solid phases controlling the Am solubility at higher temperature. The highest possible concentration of

Am dissolved in groundwater at the contact with SNF was estimated to be 10^{-9} mol/L. Note that a similar value was obtained in experiments on groundwater interaction with vitrified HLW.

Thus, the repository safety with respect to Am in the oxidizing conditions cannot be maintained only by low Am solubility in groundwater. The insulative capacity of the geological medium is also very important in this case. An easy sorption of Am on solid phases is a favorable factor in the natural environment. Many experimental results on Am sorption by various rocks and minerals, as well as mixtures of bentonite with sand, are now available. Depending on the run conditions, for example, composition, pH, Eh, solution temperature, Am concentration, and pounding degree of rocks and minerals, the partition coefficients (K_d) of Am vary from several tens to several thousands of cm^3/g and reach several tens of thousands of cm^3/g during water interaction with clay minerals and zeolitized tuffs (Baston et al., 1995). The K_d value for Am in interaction with sand is 2000 ml/g and is still higher in interaction with loam and soil (IAEA, 1992). In runs at 70°C, the K_d of Am was 7.5×10^4 for the mixture of bentonite (15 percent) and sand (85 percent) and from 0.46×10^4 to 7.5×10^4 cm^3/g for tuff (Baston et al., 1995). The K_d of Am strongly increases if the rocks contain iron oxides and hydroxides, alteration products of titanium-bearing minerals, and chlorite. Depending on the rock composition and geochemical conditions, the retention factors (R_f) of Am range from a few hundred to a thousand and are comparable to those of Pu.

Summarizing the above data, we can conclude that the Am migration from the SNF repository to the biosphere in dissolved form in concentrations hazardous for the environment is impossible; its migration in colloidal form is much more probable. This conclusion is also verified by natural observations; for example, the study of Lake Trawsfynydd in northern Wales, which receives the liquid wastes from the Magnox reactor plant, demonstrated that almost all Am is confined to colloidal particles. Because of this property of Am, it is almost immobile in soils, regardless of the occurrence of organic matter (Ways of Migration, 1999). The scale of the Am release from SNF in colloidal form can be estimated only for a certain geological medium by experiments in an underground laboratory using tracers (for example, the tracers containing Nd as a geochemical analogue of Am). The secure isolation of Am, like Pu, within a repository can be maintained by the media retaining the colloid migration. The host rocks should not contain extended open fractures, while SNF should be isolated with bentonite backfill. All previous comments regarding Pu pertain equally to Am.

CONCLUSIONS

1. The spent nuclear fuel in an underground repository can interact with groundwater only after the engineered barriers fail, that is, in 500–1000 years. The main radioactivity in this case will be related to Pu and Am.

2. The SNF contains 95–98 percent UO_2 with isomorphous mixtures of Pu and Am. Thus, the scale of the Pu and Am release depends on the uraninite stability. The latter compound is insoluble in hydrogeochemical conditions of reducing, nearly neutral to slightly alkaline groundwater saturated with respect to uraninite.

3. The groundwater attains such properties at certain depths due to its long-term interaction with the host rock masses under conditions of low water exchange. The repository safety of SNF is provided due to its placement in massifs of crystalline rocks below 500 m from a surface.

4. The formation of local oxidizing conditions in the SNF repositories induced by radiolysis or some other processes is not probable, because the host medium contains reducing agents. If the oxidizing conditions do appear, the Pu and Am migration in dissolved form in concentrations hazardous for the environment will be precluded by their rapid sorption on minerals.

5. The sorption of Pu and Am on colloidal particles followed by their migration to the discharge areas is the main real mechanism of environmental pollution. The distance and scale of Pu and Am migration in colloidal form can be estimated only for a certain geological medium by experiments in underground laboratories. The absence of wide fractures, low water permeability, and isolation with bentonite backfill are efficient factors retaining the actinide migration in colloidal form.

ACKNOWLEDGMENTS

The study was supported by the Russian Foundation for Basic Research, project numbers 02-OS-64007 and 02-05-64008.

REFERENCES

Ahn, J. 1997. Transport of weapons-grade plutonium and boron through fractured geologic media. Nuclear Technology 117:316–328.

Allen, E. J. 1978. Criticality Analysis of Thermal Underground Nuclear Waste Disposal. Report no. ORNL/TM-7405. Springfield, VA: Union Carbide Corp.

Baston, G. M., J. A. Berry, M. Brownsword, et al. 1995. Sorption of plutonium and americium on repository, backfill and geological materials relevant to the JNFL low-level radioactive waste repository at Rokkasho-Mura. Proceedings of the Symposium on Scientific Basis for Nuclear Waste Management XVIII 353(part 2):957–964.

Baston, G. M., J. A. Berry, M. Brownsword, et al. 2000. Effect of carbonate concentration on the sorption of plutonium onto geological materials. Proceedings of the Symposium on Scientific Basis for Nuclear Waste Management XXIII 608:293–298.

Berry, J. A., M. Brownsword, D. J. Ilett, et al. 2002. Effect of redox conditions on the sorption of plutonium on to geological materials. Proceedings of the Symposium on Scientific Basis for Nuclear Waste Management XXV 713:693–697.

Brookins, D. G. 1984. Geochemical Aspects of Radioactive Waste Disposal. New York: Springer.

Brookins, D. G. 1988. Eh-pH Diagrams for Geochemistry. Berlin: Springer.

Buck, E. C., R. J. Finch, P. A. Finch, and J. K. Bates. 1998. Retention of neptunium in uranyl alteration phases formed during spent fuel corrosion. Proceedings of the Symposium on Scientific Basis for Nuclear Waste Management XXI 506:87–94.

Burkov, V. V., and E. K. Podporina. 1967. Rare earths in weathering crust of granitoids. Doklady Akademy Nauk SSSR 177(3):691–694.

Chapman, N. A., and L. G. McKinley. 1988. The Geological Disposal of Nuclear Waste. Chichester: Wiley and Son.

Chapman, N. A., and D. Savage. 1984. Mineralogical aspects of the safe disposal of high-level radioactive waste. Fortschr. Miner. Bd. 62(H1):17–32.

Chen, Y., E. Siegmann, P. Mattie, et al. 1999. A mechanistic model of spent fuel dissolution, secondary mineral precipitation, and Np release. Symposium on Scientific Basis for Nuclear Waste Management XXII 556:471–478.

Cowan, G. A. 1978. Migration Paths for Oklo Reactor Products and Applications to the Problem of Geological Storage of Nuclear Wastes, Natural Fission Reactors. Pp. 693–698 in Proceedings of the Technical Committee Meeting. Paris: IAEA.

Cramer, J. J. 1986. A natural analog for a fuel waste disposal vault. Pp. 697–699 in Proceedings of the Canadian Nuclear Society International Conference on Radioactive Waste Management. Toronto: Canadian Nuclear Society.

Eliseyeva, O. P., and B. I. Omelyanenko. 1987. Uranium behavior in the process of formation of weathering profiles of the Alexeyevskoe kaolinite deposit, northern Kazakhstan. Geokhimiya 6:847–854.

Ewing, R. C. 2002. Yucca Mountain. Science 296:659–660.

Finn, P. A., R. Finch, E. Buck, and J. Bates. 1998a. Corrosion mechanisms of spent fuel under oxidizing conditions. Proceedings of the Symposium on Scientific Basis for Nuclear Waste Management XXI 506:123–131.

Finn, P.A., D. J. Wronkiewicz, R. J. Finch, J. C. Hoh, C. J. Mertz, J. W. Emery, E. C. Buck, J. A. Fortner, S. F. Wolf, L. A. Neimark, and J. K. Bates. 1998b. Yucca Mountain Project-Argonne National Laboratory, Annual Progress Report, F.Y. 1997. No. ANL-98/12. Argonne, Ill.: Argonne National Laboratory.

Forsyth, R., and L. O. Werme. 1986. The corrosion of spent UO_2 fuel in synthetic groundwater. Proceedings of the Symposium on the Scientific Basis for Nuclear Waste Management IX 327–332.

IAEA (International Atomic Energy Agency). 1978. Natural Fission Reactors. Vienna: IAEA.

IAEA. 1992. Geochemistry of Long-Lived Transuranic Actinides and Fission Products. Final Report of a Coordinated Research Programme 1987–1991. Vienna: IAEA.

Ivanov, V. V. 1997. Ekologicheskaya geokhimiya elementov (Ecological Geochemistry of Elements). Book 6. Moscow: Ekologiya.

Ivashkin, N. V., G. V. Kovalenko, V. V. Leaonkov, et al. 2000. Numerical Modeling of Change in the State of Geological Environment and Migration of Radionuclides in the Period up to 10,000 Years in the Deep Disposal. Ohshchie Voprosy Radiatsionnoy Bezopasnosty 1:3–18.

Johnson, L. H., and D. W. Shoesmith. 1988. Spent fuel. Pp. 635–698 in Radioactive Waste Forms for the Future, W. Lutze and R. C. Ewing, eds. Amsterdam: North-Holland.

Kastenberg, W. E., R. E. Peterson, J. Ahn J. Burch, G. Casher, P. Chambré, E. Greenspan, D. R. Olander, J. Vujic, B. Bessinger, N. G. W. Cook, F. M. Doyle, B. Hilbert. 1996. Mechanisms for Autocatalytic Criticality of Fissile Materials in Geologic Repositories, Report Submitted to Los Alamos National Laboratory. UCB-NE-4214. Berkeley: University of California.

Kersting, A. B., D. W. Efurd, D. L. Finnegan, et al. 1999. Migration of plutonium in ground water at the Nevada test site. Nature (Lond.) 397:56–59.

Kiryukhin, V. A., A. I. Korotkov, and S. L. Shvartsev. 1993. Gidrogeokhimiya (Hydrogeochemistry). Moscow: Nedra.

Korenkov, A. P. 1992. To the problem of classification of solidified radioactive wastes. Atomic Energy 73(2):129–131.

Krainov, S. Z., and V. M. Shvets. 1992. Gidrogeokhimiya (Hydrogeochemistry). Moscow: Nedra.

Krauskopf, K. B. 1988a. Geology high-level nuclear waste disposal. Annual Review of Earth and Planetary Science 16:173–200.

Krauskopf, K. B. 1988b. Radioactive Waste Disposal and Geology. London: Chapman and Hall.

Kulyako, Yu. M., S. Perevalov, T. Trofimov, et al. 2001. Experimental studies of solubility of U(VI), Np(V), Pu(IV), and Am(III) hydroxides in simulated solutions of ground and sea water. P. 43 in Eighth International Conference on Chemistry and Migration Behavior of Actinides and Fission Products in the Geosphere. Migration '01. Abstracts. Bregenz, Austria.

Laverov, N. P., A. V. Kantsel, B. I. Omelyanenko, et al. 1991. Principal problems of radioecology in connection with radioactive waste disposal. Atomic Energy 71(6):523–534.

Laverov, N. P., B. I. Omelyanenko, and V. I. Velichkin. 1994. Geological aspects of the problem of radioactive waste disposal. Geoekologiya 6:3–20.

Laverov, N. P., V. I. Velichkin, and B. I. Omelyanenko. 2001. Insulative properties of crystalline rock: on the problem of high level radioactive waste disposal. Geologia Rudn. Mestorozhdeny 43(1):6–23.

Lelous, K., S. Constantin, J. L. Paul, D. Sambuaaro, and E. Vernaz. 1998. Leaching of spent fuel and simulated fuel in the presence of environmental materials: integral experiments. Proceedings of the Symposium on Scientific Basis for Nuclear Waste Management XXI 506:175–188.

Liu, J., and L. Neretnieks. 1995. Some evidence of radiolysis in a uranium ore body: quantification and interpretation. Proceedings of the Symposium on Scientific Basis for Nuclear Waste Management XVIII 353(part 2):1179–1186.

LLNL (Lawrence Livermore National Laboratory). 1998. Waste Form Characteristics Report. UCRL-ID-132375. CD-ROM Version. Livermore, CA: LLNL.

Loida, A., B. Grambow, and H. Geckeis. 2001. Congruent and incongruent radionuclide release during matrix dissolution of partly oxidized high burnup spent fuel. Proceedings of the Symposium on Scientific Basis for Nuclear Waste Management XXIV 663:417–426.

Michard, A. 1989. Rare earth element systematics in hydrothermal fluids. Geochimica et Cosmochimica Acta 53(3):745–770.

Michard, A., C. Beaucaire, and G. Michard. 1987. Uranium and rare earth elements in CO_2-rich waters from Vals-les-Bains (France). Geochimica et Cosmochimica Acta 51(4):901–909.

Michard, G., G. Negrel, G. Ouzounian, P. Toulhoat, et al. 1991. Major and trace elements regulation in natural granitic waters: application to deep radioactive waste disposal, high-level radioactive waste management. Proceedings, Second Annual International Conference, Las Vegas, Nevada 1:169–175.

Neretnieks, L. 1993. Solute transport in fractured rock—applications to radionuclide waste repositories. Pp. 39–123 in Flow and Contaminant Transport in Fractured Rock. San Diego: Academic Press.

Neretnieks, L. 1997. Modeling oxidative dissolution of spent fuel. Proceedings of the Symposium on Scientific Basis for Nuclear Waste Management XX 465:573–580.

Omelyanenko, B. I., P. S. Kozlova, O. P. Eliseyeva, and L. L. Simonova. 1983. Local distribution of uranium in rocks and minerals as an indicator of its geochemical history. Pp. 140–163 in Problemy Radiogeology (Problems of Radiogeology), pp. 140–163. Moscow: Nauka.

Oversby, V. M. 1998. Criticality in repository for spent fuel: lessons from Oklo. Proceedings of the Symposium on Scientific Basis for Nuclear Waste Management XXI 506:781–788.

Oversby, V. M. 2000. Using information from natural analogues in repository performance analysis: examples from Oklo. Proceedings of the Symposium on Scientific Basis for Nuclear Waste Management XXIII 608:545–550.

Pagel, M., F. Ruhlmann, and P. Bruneton. 1993. The Cigar Lake uranium deposit, Saskatchewan, Canada. Canadian Journal of Earth Sciences 30:651–652.

Pokrovsky, O. S. 2003. Forms of finding and coefficients of activity of Am(III) and Np(V) in marine water. Geokhimiya 3:337–341.

Pourcelot, L., and F. Gauthier-Lafaye. 1998. Weathering conditions and behavior of fission products in Oklo reactors. Proceedings of the Symposium on Scientific Basis for Nuclear Waste Management XXI 506:1071–1072.

Redkin, A. F., N. I. Savelyeva, E. I. Sergeyeva, B. L. Omelyanenko, I. P. Ivanov, I. L. Khodakovsky. 1989. Investigation of uraninite UO_2 solubility under hydrothermal conditions. Sciences Géologiques Bulletin Strasbourg 42(4):329–334.

Roddy, J. W., H. C. Claiborne, R. C. Ashline, P. J. Johnson, and B. T. Rhyne. 1985. Physical and Decay Characteristics of Commercial LWR Spent Fuel. Report no. ORNL/TM-9591/V. 1. Oak Ridge, Tenn.: Oak Ridge National Laboratory.

Rondinella, V. V., J. Cobos, H. J. Matzke, T. Wiss, P. Carbol, and D. Solite. 2001. Leaching behavior and α-decay damage accumulation of UO_2 containing short-lived actinides. Proceedings of the Symposium on Scientific Basis for Nuclear Waste Management XXIV 663:391–398.

Rondinella, V. V., H. J. Matzke, J. Cobos, and T. Wiss. 1999. α-radiolysis and α-radiation damage effects on UO_2 dissolution under spent fuel storage conditions. Proceedings of the Symposium on Scientific Basis for Nuclear Waste Management XXII 556:447–454.

Ryzhenko, B. N., V. L. Barsukov, and S. N. Knyazeva. 1996. Chemical characteristics (composition, pH, Eh) of the rock-water system: I. Granitoids-water system. Geokhimiya 5:436–454.

Schneider, J. W., P. Zuidema, P. A. Smith, et al. 1998. Preliminary safety studies fuel, MOX and vitrified high-level waste. Proceedings of the Symposium on Scientific Basis for Nuclear Waste Management XXI 506:1077–1078.

Smith, P. A., and L. H. Johnson. 2000. Spent fuel dissolution: an examination of the impacts of alpha-radiolysis. Proceedings of the Symposium on Scientific Basis for Nuclear Waste Management XXII 608:29–39.

Spahiu, K., and P. Sellin. 2001. Spent fuel alteration/dissolution and the influence of near field hydrogen. Proceedings of the Symposium on Scientific Basis for Nuclear Waste Management XXVI 663: 765–772.

Sturchio, N. C., K. Muehlenbachst, and M. G. Seitz. 1986. Element redistribution during hydrothermal system: Yellowstone drill cores Y-7 and Y-8. Geochemica et Cosmochimica Acta 50(5): 1619–1631.

Tsukamoto, M., T. Ohe, T. Fujita, R. Hesbol, and Y. P. Yermansson. 1995. Diffusion of radionuclides in compacted bentonite: results from combined glass dissolution and migration tests. Proceedings of the Symposium on Scientific Basis for Nuclear Waste Management XVII 353(part 1):291–298.

Vandergraaf, T. T., and D. R. M. Abry. 1982. Radionuclide sorption on drill core material from the Canadian Shield. Nuclear Technology 57:399–412.

Vaniman, D., A. Furlando, S. Chipera, J. Thompson, and L. Triay. 1996. Microautoradiography in studies of Pu(V) sorption by trace and fracture minerals in tuff. Proceedings of the Symposium on Scientific Basis for Nuclear Waste Management XIX 412:639–646

1999. Ways of Migration of Man-Made Radionuclides in the Environment: Radioecology after Chernobyl (Puti migratsii iskusstvermykh radionuklidov v okruzhayushchei srede. Radioekologiya posle Chernobylya). Moscow: Mir.

Wilson, C. N. 1990a. Results from NNWSI Series 2 Spent Fuel Dissolution Tests. PNL-7170. Richland, Wa.: Pacific Northwest National Laboratory.

Wilson, C. N. 1990b. Results from NNWSI Series 3 Spent Fuel Dissolution Tests. PNL-7170. Richland, Wa.: Pacific Northwest National Laboratory.

Zakharova, E.V., A. A. Menyailo, K. A. Menyailo, E. P. Kaimin, et al. 1998. Assessment of process of transforming rocks enclosing radioactive waste storages on exposure to chemical and thermal factors. Pp. 149–158 in Issledovanie granitoidov Nizhnekanskoyo massiva dlya zakhororreniya RAO (The Study of Granitoids from Nizhnekansky Massif for Radioactive Waste Disposal). Zheleznogorsk: KNTS.