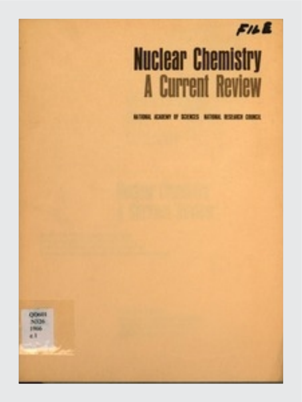

Nuclear Chemistry: A Current Review

DETAILS

56 pages | 7 x 10 | PAPERBACK
ISBN 978-0-309-36426-3 | DOI 10.17226/21526

AUTHORS

Panel on Nuclear Chemistry; Committee for the Survey of Chemistry; Division of Chemistry and Chemical Technology; National Research Council

Nuclear Chemistry
A Current Review

Report of the Panel on Nuclear Chemistry
of the Committee for the Survey of Chemistry
Division of Chemistry and Chemical Technology
National Academy of Sciences National Research Council

Publication 1292-C
NATIONAL ACADEMY OF SCIENCES
NATIONAL RESEARCH COUNCIL
WASHINGTON, D. C. 1966

PANEL ON NUCLEAR CHEMISTRY*

Gerhart Friedlander, Brookhaven National Laboratory, Co-Chairman
John O. Rasmussen, University of California, Berkeley, Co-Chairman
John R. Huizenga, Argonne National Laboratory
Thomas T. Sugihara, Clark University
Anthony Turkevich, University of Chicago

COMMITTEE FOR THE SURVEY OF CHEMISTRY

Frank H. Westheimer, Harvard University, Chairman
William O. Baker, Bell Telephone Laboratories, Inc.
Theodore L. Cairns, E. I. du Pont de Nemours and Company
Melvin Calvin, University of California, Berkeley
Bryce L. Crawford, Jr., University of Minnesota
Herbert S. Gutowsky, University of Illinois
Franklin A. Long, Cornell University
Robert W. Parry, University of Michigan
Kenneth S. Pitzer, Rice University
Charles C. Price, University of Pennsylvania
John D. Roberts, California Institute of Technology
Harrison Shull, Indiana University
Walter H. Stockmayer, Dartmouth College
Gilbert Stork, Columbia University
Henry Taube, Stanford University

Martin A. Paul, National Academy of Sciences—National Research Council, Special Consultant

*James R. Arnold, University of California, San Diego, was initially a panel member and drafted part of Section IIIC but was unable otherwise to participate after the first meeting.

Available from

Printing and Publishing Office
National Academy of Sciences
2101 Constitution Ave., N.W.
Washington, D. C. 20418
Price $2.00

The Committee on Science and Public Policy of the National Academy of Sciences has appointed committees to survey several of the basic sciences. In the course of preparing its report, Chemistry: Opportunities and Needs (National Academy of Sciences—National Research Council Publication 1292, November 1965), the Committee for the Survey of Chemistry appointed panels of distinguished scientists to prepare reports on specific areas of chemistry. These panel reports, of widely varying scope, provided much of the information and expert opinion that constitute the Survey.

Many of the panel reports necessarily contained more valuable information and more detailed discussion than we could adequately summarize in the space at our disposal. We have therefore presented here the panel report on Nuclear Chemistry in its entirety. The Committee for the Survey of Chemistry does not necessarily agree with or endorse all the opinions or conclusions presented; they are of course those of the authors. We believe, however, that this report contains important subject matter, and that it will be interesting and informative to many readers. We wish to thank the panel members, and especially the two cochairmen, Dr. Gerhart Friedlander and Dr. John O. Rasmussen, for their contribution to chemistry. We wish also to acknowledge a grant from the American Chemical Society and additional support from the National Science Foundation that enabled this study to be undertaken.

Frank H. Westheimer, Chairman
Committee for the Survey of Chemistry

CONTENTS

I

INTRODUCTION

In the recent decades of expanding scientific research, the classical boundary lines between various branches of science have often lost their clarity and usefulness. The searchlights of scientific inquiry have been directed on problems calling for a range of skills and insights transcending the bounds of a classical category of science. Astronomy, biology, chemistry, geology, and physics especially have found intimate new interdependence in many areas.

Since the beginnings of studies on radioactivity and nuclear transformations, the skills and methods of approach of both chemistry and physics have been demanded. Nuclear science and its applications have advanced under the combined efforts of scientists trained in various disciplines and in different countries. Had nuclear research remained the sole province of researchers with similar educations, its progress would have been far more restricted and unbalanced.

The intrinsic nature of the problems led to a complementary research effort in present-day nuclear science. While there is considerable overlapping of interests between nuclear physicists and chemists, a predominance of physicists exists in research areas where the experimental demands are primarily in physical instrumentation and a predominance of chemists exists where there are research demands for chemical separations and purifications. At present, there is also some complementary division of work manifest in the predominance of physicists' interests in the simpler systems (such as lighter nuclei, the few-nucleon problem, elucidation of the fundamental nature of nucleon-nucleon forces, and the weak interactions), whereas chemists' interests are in the more complex phenomena (structure of heavier nuclei, high-energy nuclear reactions, cosmological and geological applications, and nuclear fission). The dividing lines shift with time, and some research areas have passed back and forth between physicists and chemists. This has happened, for example, in atomic and molecular spectroscopy and in solid-state studies as well as in the investigations of many nuclear phenomena.

This report will deal mainly with research by scientists trained as chemists; however, future progress in interdisciplinary areas may depend in varying proportions on scientists trained in physics, geology, biology, astronomy, or other disciplines. An effort has been made to strike a balance in pointing out the actual or potential contributions of the chemists, without minimizing the credit to contributors from other fields.

It is difficult to give a universally acceptable definition of nuclear chemistry.* For the purposes of this survey, included as research in nuclear chemistry are: most of the research on nuclear structure, reactions, and radioactive transformations being carried out by persons trained as chemists; research by chemists on atomic and molecular phenomena, insofar as nuclear processes are involved more than incidentally; and research by chemists on geological, astronomical, or other problems where nuclear processes are intimately involved.

---

*For recent discussions of definitions as well as a general review of nuclear chemistry see References 1-3.

The older term radiochemistry sometimes used as a synonym for nuclear chemistry, now has a narrower meaning—the use of radioactivity as a tool in the study of chemical systems. Historically, radiochemistry and nuclear chemistry have developed together, since both rely heavily on radioactivity measurements, and the same investigators have often been involved in both. Some important aspects of radiochemistry will be mentioned although they are not considered a part of nuclear chemistry today. Many radiochemical techniques such as the use of radioactive tracers and isotopic substitution are so widespread in chemistry, and the nuclear aspects are now so incidental in these applications, that they are more properly discussed in the context of the chemical research to which they are applied. For similar reasons, the field of radiation chemistry—the study of chemical reactions induced by radiation—is not included.

II

HISTORY AND MAJOR ACCOMPLISHMENTS

Although the term nuclear chemistry, designating a separate discipline, has come into use only in the past two decades, since the days of the Curies chemists have played a vital role in exploring the phenomenon of radioactivity and studying atomic nuclei. Nuclear chemists, in partnership with physicists, have advanced our understanding of atomic nuclei and helped to open up exciting new areas of research in chemistry, geology, archaeology, astrophysics, meteorology, and other fields. One indication of their important contributions is that over 10 percent of all Nobel prizes in chemistry have been awarded for work that would today be designated nuclear chemistry.

Nuclear chemistry had its origin in the early work of the Curies, since their characterization and concentration of the new elements polonium and radium depended strongly on chemical manipulations and chemical reasoning. In 1900 the fruitful collaboration began between the physicist, Ernest Rutherford, and the chemist, Frederick Soddy, which culminated in the recognition of radioactive disintegrations as subatomic changes and thus revolutionized the then accepted concept of atoms as immutable entities. Soddy's chemical insight eventually led him to clear recognition that several radioelements, each with its own distinctive characteristics, could occupy a single position in the periodic table and thus he arrived at the concept of isotopes, a term he coined. Soddy was also the first to propose that the occurrence of isotopes was not confined to radioelements, but "that each known element may be a group of non-separable elements occupying the same place (in the periodic table), the atomic weight not being a real constant, but a mean value."

Other pioneers in those early years of nuclear chemistry included K. Fajans, who contributed much to the understanding of the connection between radioactivity and atomic structure and formulated some of the rules governing the chemical behavior of trace quantities of materials; O. Hahn, who discovered a number of naturally occurring radioactive species, cleared up the decay sequence in the ^{235}U series, and made important contributions to our knowledge of the behavior of substances at extremely low concentrations; and G. von Hevesy, who with F. A. Paneth originated the idea of using radioactive isotopes as tracers or indicators and later pioneered the application of this notion to biological problems as well as extending it to the use of man-made radioactive isotopes.

Further major accomplishments have occurred in recent times; some of these will be discussed more fully in Section III, in connection with current trends in research. Since the mid-1930's, chemists have played an important role both in the investigation of nuclear reactions and in the characterization and detailed study of new radioactive species. The complex patterns of reactions induced in heavy nuclei by bombarding particles with energies of hundreds and thousands of MeV (million electron volts) were unravelled almost exclusively by chemists. Reactions at lower energies were more nearly a joint province of physicists and chemists. Much of the data on nuclear energy levels on which the development of models for nonspherical nuclei was based resulted from studies of radioactive decay processes by nuclear chemists. The synthesis and characterization of elements that do not occur in nature, particularly those beyond uranium in the periodic table, were largely accomplished by nuclear chemists, led by G. T. Seaborg and his co-workers. This work not only produced a significant extension of the periodic table, but led to a wealth of new information on nuclear structure and nuclear systematics (4, 5). On the heels of the discoveries of the first transuranium elements, radiochemical methods were used to achieve the spectacular development of processes for large-scale production of some of these new species (especially plutonium). Chemists have played a pivotal role throughout the development of the atomic energy program. In fact, the phenomenon of nuclear fission, which made atomic energy possible in the first place, was discovered in 1938 by the nuclear chemists O. Hahn and F. Strassmann. It was only through their painstaking development of firm chemical evidence that nuclear fission, which ran counter to the then-accepted ideas of nuclear physics, was made credible to them and to the world (6).

Without doubt, nuclear fission has had more far-reaching consequences for mankind than any other single scientific discovery in recent decades. However, it is not the only example of advances in other disciplines that are directly traceable to developments in nuclear chemistry. The entire vast field of the use of isotopic (radioactive and stable) tracers, which has become so important in chemistry, biology, and medicine as well as in agriculture and industry, sprang from nuclear chemistry. Not only was the tracer method itself invented, developed, and first used by nuclear chemists, but also they discovered and characterized some of the most important radioactive tracer isotopes, such as ^{14}C and ^{131}I. By the same token, nuclear chemists were responsible for the discovery of most of the radioactive nuclides used as powerful radiation sources in industry and medicine such as ^{60}Co and ^{137}Cs and they also developed many techniques for handling and packaging such sources.

Parallel with the exploration and application of the tracer method itself, two other interesting and fruitful developments were: (1) the study of the small but significant chemical effects of isotopic substitutions; these, in turn, provide a most useful tool in the study of chemical reactions and chemical bonds and form the basis of the most successful method for separating the isotopes of hydrogen and other light elements; (2) the invention of various ingenious techniques for labeling complex molecules with radioactive atoms; among these, labeling with the energetic atoms produced in nuclear reactions—so-called hot atoms—is particularly worthy of note (7).

In analytical chemistry, the method of radioactivation has opened up previously inaccessible ranges of sensitivity. In this method, characteristic and easily detected radioactive elements are generated by bombarding the sample with neutrons or other radiation. The achievement of the extremely high purities required for transistor materials was dependent on such a method for measuring trace impurities.

Geology and geochemistry were revolutionized by the introduction of radioactivity clocks which, for the first time, established absolute and accurate measurements of geologic time. By this means, geologists have been able to determine the time elapsed since the solidification not only of various terrestrial rocks but also of meteorites (8). Another radioactivity clock, somewhat different in character and applicable to more recent dates, was provided by the ^{14}C-dating method of W. F. Libby (9). This method was also found capable of widespread applications and has provided significant impetus to various branches of science, notably archaeology and anthropology. A final example of cross-fertilization is the impact that physicists' and chemists' investigations of nuclear reactions have had on present-day thinking in astrophysics (10). As notable milestones we may cite the discovery of spectral lines of the man-made and relatively short-lived element technetium in stellar spectra, which stimulated the development of theories of element synthesis in stellar interiors; and the discovery, by chemical means, of the process of many successive neutron captures on a rapid time scale in the first man-made thermonuclear explosions; this discovery led to the realization that such processes may be important in supernovae and thus to new ideas about the mechanisms of these gigantic stellar explosions and their roles in element synthesis.

The foregoing examples of the revolutionary impact of nuclear chemistry emphasize that none of these developments was, or could have been planned or even foreseen too far in advance. Such developments grew out of basic research that was undertaken without thought of useful applications. It was Lord Rutherford who said, very late in his life, "I am doubtful, however, whether the most imaginative scientific man, except in rare cases, is able to foresee the result of any discovery." Unknowingly proving his point, Rutherford, the discoverer of nuclear transmutations, wrote in 1936, 2 years before the discovery of fission, "The outlook for gaining useful energy from the atoms by artificial processes of transformations does not look very promising."

III

CURRENT TRENDS IN NUCLEAR CHEMISTRY

The goals, intellectual content, and status of present-day nuclear chemistry research are outlined in this part of the report. Section A discusses research aimed at understanding the properties and transformations of nuclei. Section B turns attention outward from the world of nuclei to nuclear chemical research directed at atomic and molecular problems. Section C examines the applications of nuclear chemistry to the solution of problems in the macroscopic universe. Although this report deals principally with pure research, the role of nuclear chemistry in the nation's nuclear energy programs is so basic that it could not be omitted; this role is discussed briefly in Section D. Finally, in Section E, attention is focused on the interplay between advances in instrumentation and the development of research in nuclear chemistry.

The relative amounts of space devoted to Sections A, B, and C should not be interpreted as reflecting the panel's judgment about the over-all importance of various research areas. Rather it was felt that the work on nuclear properties and reactions deserves the most detailed attention because, clearly, it constitutes the heart of nuclear chemistry. The fields of research covered in Sections B and C have developed largely out of nuclear studies but are, for the most part, being assimilated into other disciplines. The panel is fully aware that not everyone will agree with the classifications of some of these research areas as parts of nuclear chemistry, and that the appropriateness of these classifications may require revision to meet changes as time goes on. One of the strengths of nuclear chemistry has been the fact that from it have sprung many ideas and techniques that have become important in other fields of science. For a proper perspective of the field, it was felt justifiable to consider briefly the research in borderline fields that has strong interdependence with nuclear problems.

A. RESEARCH ON NUCLEAR PROPERTIES AND REACTIONS

The research in this area will be discussed under the categories of synthesis of nuclei, their structure and properties, and nuclear transformation, with some further subdivision of the last category into different classes of reactions.

1. Synthesis of New Elements and New Isotopes

A major achievement of postwar nuclear chemistry is the synthesis and identification of nuclei of new transuranium elements (4, 5). Seven new elements and over seventy new transuranium isotopes have been discovered in the United States since World War II.* Only about a dozen isotopes of transuranium elements were known at the close of World War II.

To judge the future needs of research in the synthesis of transuranium isotopes its development must be traced briefly. The heaviest isotope available in nature is ^{238}U, and it is the starting material for all synthesis. The main line of synthesis of weighable quantities of heavier atoms is the long-term irradiation by neutrons in nuclear reactors of the highest available flux. Military requirements have led to extensive production of ^{239}Pu. As by-products, small amounts of isotopes of elements 95 and 96, americium (Am) and curium (Cm) became available for basic research in the United States in the early postwar years. These materials in microgram quantities were sufficient in 1950 to serve as cyclotron target materials for helium-ion bombardment to make detectable amounts of elements 97 and 98, berkelium (Bk) and californium (Cf). Since cyclotron production could not make these elements in the quantities needed as target materials to reach higher elements, investigations toward long-term irradiations of americium and curium in reactors were pursued.

A new avenue of synthesis was also explored, that of the development of beams of ions heavier than helium. With such heavy ions there was hope of depositing more than two protons at once in a nucleus, and thus by-passing the limitation imposed by the unavailability of target material of elements beyond curium. Before either of these two approaches

---

*The discovery of element 104 was reported in 1964 by G. N. Flerov and his co-workers in the Soviet Union (by bombardment of ^{242}Pu with ^{22}Ne ions from a cyclotron).

proved successful, a surprise breakthrough came with the first giant fusion-fission bomb explosion in 1952. The exceedingly high neutron flux in this device produced a rapid successive absorption of as many as 17 neutrons by ^{238}U; the subsequent chain of beta decays gave rise to detectable amounts of new elements 99 and 100, einsteinium (Es) and fermium (Fm), as well as thirteen new isotopes of Pu, Am, Cm, Bk, and Cf. By 1955 the years of reactor-neutron irradiations at the high-flux Materials Testing Reactor in Idaho finally produced enough einsteinium to serve as a cyclotron target for helium ions, leading to the discovery of element 101. Elements 102 and 103 came initially from the alternate approach of heavy-ion bombardments.

New element synthesis via the heavy-ion approach becomes increasingly difficult with increasing atomic number because reaction yields become exceedingly small as a result of strong competition from fission reactions and also because the relatively neutron-deficient isotopes attainable by heavy-ion bombardment are very short-lived. Synthesis via the route of slow buildup by neutron capture in reactors has a serious blockage at isotopes such as ^{256}Fm, which is so unstable with regard to spontaneous fission that its half-life is only 3 hr. It may be that greater stability toward spontaneous fission is regained beyond mass 256 and that a host of longer-lived nuclei lie beyond the present barrier. A tiny amount of the isotope ^{257}Fm with the encouragingly long half-life of 80 days has been obtained from long irradiations with thermal neutrons.

A possible future approach to synthesis of super-heavy elements is to employ highly endothermic reactions between heavy nuclei in an inverse fission process, such as reactions induced by xenon ions in which the compound nuclei would be formed with relatively low excitation energy. A new type of heavy-ion accelerator would be needed to attain the energies of hundreds of MeV needed for such reactions.

For the present, synthesis via the route of fast successive neutron captures in underground nuclear explosions appears to be the most hopeful method of by-passing the short-lived fermium isotopes, and this method deserves intensive effort (4, 11). It is also important to continue, by long irradiation in reactors, the buildup of all the attainable longer-lived nuclides, that is, heavy isotopes of curium, ^{249}Cf, ^{251}Cf, ^{254}Es, and possibly fermium isotopes beyond 256. The new High Flux Isotope Reactor at Oak Ridge is specifically designed for this purpose.

The contributions of nuclear chemists to synthesis of new nuclei have not been confined to the transuranium-element region alone, but have ranged over the whole periodic table. Approximately 1400 radioactive nuclides are now known. It is of interest that nearly half of them were discovered by nuclear chemists in the United States.

The present knowledge of radioactive nuclei near the region of beta-stability is fairly extensive, since these nuclei are readily made. On the other hand, much is yet to be learned about the generally shorter-lived products further removed from stability. Bombardments with heavy ions or with high-energy projectiles give access to the neutron-deficient region, whereas fission (particularly of heavy elements such as californium) and multiple neutron capture in reactors with high neutron fluxes or in fusion explosions can produce hitherto unexplored neutron-rich isotopes.

The production of new types of nuclei is not confined to the production of new isotopes of transuranium elements or of nuclei with unusual neutron-proton constitution. New techniques for studying short-lived radioactivity have recently made possible the discovery of nuclei exhibiting a novel type of radioactivity—delayed proton emission (beta decay followed quickly by proton emission). Another unusual class of nuclei has been discovered in the Soviet Union, where heavy-ion bombardments have produced short-lived (seconds down to milliseconds) excited states of odd-odd americium nuclei that decay by spontaneous fission. Similarly the discovery of relatively long-lived highly excited states of high spin in ^{212}Po and ^{177}Lu has been a significant stimulant to theory, and indications are that there are many more such excited states of common nuclei as yet undiscovered.

2. Detailed Studies of Nuclear Structure and Properties

While a sharp line cannot be drawn between nuclear synthesis and the detailed and more intensive studies of nuclear properties, the pioneering discovery work usually involves characterizing a few gross properties such as half-life, mass, atomic-number assignments, and preliminary measurements of the energy and abundance of the various radiations accompanying the decay. Such measurements are only the beginning of the fundamental information to be gained through intensive study.

The nuclear spectroscopist employs various devices to measure the energies and intensities of the alpha, beta, or gamma rays emitted by a given radioactive species. In this way, he attempts to establish precise energies for various excited energy states of the product nuclei. By a variety of direct and indirect measurements, other properties of these energy states are determined, among them their spins, modes of decay, lifetimes, magnetic moments, and electric quadrupole moments (a measure of nuclear shapes). Every few years during the last two decades, new or radically improved instruments were developed for such nuclear measurements; each new development provided the opportunity for fruitful re-examination of many nuclei. Nuclear chemists along with physicists have always been actively involved in the exploitation of these advances in instrumentation for a wide variety of nuclear spectroscopy studies.

The detailed measurements of nuclear properties have accomplished far more than merely to supply a large collection of numbers; indeed, the past 15 years have witnessed the complementary rapid advance of nuclear theories and models giving order and meaning to the measurements. A great landmark in modern theory is the 1949 formulation, principally by the physicists Maria Goeppert-Mayer at the University of Chicago and J. H. D. Jensen in Germany, of an atom-like picture applicable to a large class of nuclei (12). Their independent-particle (or shell) model assigns protons and neutrons to separate orbits analogous to the assignment of electrons to orbits in atoms. The special stability of nuclei with certain numbers of neutrons or protons became understandable in the same way that the relative chemical stability of the noble gas atoms is understood.

A second important theoretical development was the 1953 formulation of a model for a class of distorted, cigar-shaped nuclei by physicists A. Bohr, a Dane, and B. R. Mottelson, an American, both working in Copenhagen. Their model was the first to combine with the shell model the important concept of collective motions, such as rotations and vibrations of the nucleus as a whole. The nuclei to which this model applies should have properties closely resembling those of certain molecules—energy levels bunched closely into rotational-band patterns, and protons and neutrons moving in elongated orbitals.

During the past decade there has been much progress in testing, modifying, and refining these basic models, and in this work many nuclear physicists and chemists have contributed in important ways with their complementary skills. Especially in the testing of the Bohr-Mottelson model, the work of nuclear chemists has been of great significance, for most of the cigar-shaped nuclei lie in the transuranium region and the rare-earth region, where most nuclear studies require special chemical separation techniques. Studies of the fine details of alpha-particle energies emitted by transuranium nuclides provided the richest early source of information on the rotational-band structure. The alpha-decay studies have also proved of great value in establishing the correspondence between experimentally observed bands in the heavy nuclei and the calculated orbits of neutrons and protons. Most of our modern detailed understanding of the alpha-emission process stems from the pioneering postwar work of American nuclear chemists both in experiment and in theory, with important contributions in recent years from Western European and Soviet physicists.

Heavy-ion bombardments have been used in two important ways to obtain information on rotational-band structures. One is the so-called multiple Coulomb excitation process, raising nuclei into various states of rotational excitation by electromagnetic interaction. Another method involves observation of gamma rays corresponding to the de-excitation of rotational states of nuclei formed as a result of nuclear reactions with heavy ions. These new spectroscopic tools for the investigation of states with extremely high spins have been pioneered by nuclear chemists, and the results have had an impact on theory, especially in showing the absence of a sharp transition from the superfluidity of nuclear matter in ground states to the normal state of matter in a highly excited condition. With recent advances in the field of fission research, studies of the radiations from fragments of known masses (produced in the fission of heavy elements such as californium) are giving detailed information on excited states of a large number of neutron-rich fragment nuclei in a single experiment.

In another area of detailed studies of nuclear properties, chemical know-how is of importance to the research on ultra-low-temperature nuclear orientation. When radioactive atoms are incorporated into special alloys or single crystals of magnetic salts and cooled to below 0.1°K, the nuclear spins can be oriented along an applied magnetic field or along a crystalline axis. Such orientation may manifest itself in the angular dependence of nuclear radiation accompanying the decay of the radioactive atoms. Studies of gamma-ray anisotropies are now numerous and provide much detailed information on nuclear moments, spins, and gamma multipolarities. Less common and more difficult are experiments on electron or alpha radiation from oriented nuclei. Such experiments on alpha radiation have provided the best information derived on the angular momentum mixtures involved in the alpha-decay process. Experiments have also been carried out to study the angular distribution of fission fragments from the neutron-induced fission of aligned nuclei. These experiments give information about the quantum states for the saddle-point nuclei.

3. Nuclear Dynamics

An area of research that has steadily attracted many nuclear chemists is the investigation of the dynamics of nuclear reactions. It is concerned with the details of the processes that occur when nuclear species are transformed into other nuclear species on interaction with some bombarding particles. The particles used to bring about nuclear reactions may be elementary particles (such as photons, neutrons, protons, and mesons) or complex nuclei (such as deuterons, helium nuclei, ^{12}C, ^{20}Ne, and the like).

Chemists became involved originally in the study of nuclear reactions largely because the unambiguous identification of a reaction product almost always requires the determination of its atomic number, in other words, its chemical identification. However, the interest of chemists in nuclear dynamics has gone beyond this and has become much broader; it now encompasses most aspects of nuclear reaction studies. The chemist's traditional concern with many-body problems and with statistical mechanical aspects of complex systems has probably been responsible, at least partly, for this development.

The subject of nuclear dynamics has been divided into three parts in the discussion that follows. Studies of the fission process at low energies are discussed separately because this reaction is quite different from other reactions; it is at the same time extraordinarily important. Other types of reactions are divided according to the bombarding energies into low-energy (≤ 100 MeV) and high-energy (≥ 100 MeV) reactions—a somewhat arbitrary classification that has, however, some justification in the dominant reaction mechanisms that appear to prevail.

a. Low-energy nuclear fission. Among nuclear reactions, the process of nuclear fission has been one of the special concerns of chemists since its discovery, primarily because of the particular suitability of the techniques of chemistry. Both the gross features of the asymmetric mass-split in low-energy fission and the quantitative yields of the approximately one hundred fission products were established by chemists. Likewise, the dependence of these yields on the nature of the fissioning nucleus and on the amount of excitation has been determined by radiochemical techniques. Finally, studies of the charge distribution (variation of yield with atomic number for a given fission-product mass number) are still being actively pursued.

The fission process is a unique nuclear reaction in that it involves the excitation of gross collective motion of one part of a nucleus relative to another. Studies of nuclear fission thus represent the only available means of learning about the dynamical properties of nuclear matter. Several features of the process are understood in terms of a model in which the fission is considered as the breakup of a charged liquid drop. However, for many other aspects of the process, including the observed striking asymmetry in the mass division, a satisfactory explanation has not yet been found. So there is still need for both theoretical and experimental investigations of this reaction.

Such studies of fission have two different objectives. First, the investigations on the distribution of mass, charge, kinetic energy, and the angle of emission of the fragments yield information about the nature of the fission process and the quantum properties of the fissioning nucleus. Second, the study of the de-excitation mechanism of the fission fragments, by investigation of the properties of the prompt radiations that the fragments emit, deals as much with the nuclear spectroscopy of these neutron-rich fragment nuclei as with the mechanism of splitting of the nuclear matter. In particular, studies of the angular correlation of the fission fragments relative to the direction of motion of the particle initiating fission and the interpretation of these in terms of the quantum states of the transition state nucleus, have led to significant advances in our understanding of the properties of a nucleus with an unusually large deformation, which is not met in stable nuclei. The new, tandem Van de Graaff accelerators capable of producing monoenergetic particle beams of good energy resolution are expected to yield important further information of this type. In a similar way the important role of the angular momentum of the fissioning nucleus in determining the probability and the nature of fission has recently received much attention.

The production of fissionable nuclei with a wide range of angular momenta has become possible through use of heavy-ion accelerators, and studies of fission induced by heavy ions are being pursued intensively.

Our understanding of the details of the de-excitation of the fission fragments immediately after fission occurs has been significantly improved in the last few years by the availability of ^{252}Cf spontaneous-fission sources. Using such sources, the radiations associated with the de-excitation process (neutrons, nuclear gamma rays, atomic x rays, and conversion electrons) can be investigated without the background radiation present in studies of induced fission. In addition to these radiations shedding light on the events accompanying the fission process, they also provide spectroscopic information on the energy levels of very neutron-rich nuclei. There is at present no other means for studying such nuclei.

One of the least explored aspects of nuclear fission is that of division into three or more fragments of approximately equal mass. Such splittings should be energetically favored, have long been sought, but apparently are rare events in the low-energy fission of the commonly available fissile nuclei. Some considerations suggest that the probability of such events may increase markedly with increasing excitation energy and atomic number. Further search for this phenomenon is definitely warranted.

From these examples it should be clear that the challenge of attaining a more complete understanding of the process of nuclear fission is likely for some time to enlist the newest experimental and theoretical tools that nuclear chemists and physicists can devise.

b. Low-energy reactions. Attempts to understand low-energy nuclear reactions have centered around two extreme models. In the compound nucleus theory suggested in 1936 by Niels Bohr, the energy brought into the nucleus by a bombarding particle is assumed to be distributed rapidly among all the neutrons and protons, thus forming an intermediate, excited "compound nucleus" analogous to, but much more rigorously defined than, the activated complex in the theory of chemical-reaction rates. The de-excitation of this compound nucleus by the emission of particles (gamma rays, neutrons, protons, and heavier entities) is thought to take a time long compared with its formation (although still a small fraction of a second). In contrast to this two-step theory with equipartition of energy in the first step, the direct-interaction concept assumes an interaction of the incident particle with one of the neutrons or protons in the target nucleus, or with a small aggregate of particles. This interaction may lead to the ejection of the struck particle or aggregate, either alone or in combination with the projectile. In another type of direct interaction, a complex bombarding particle such as a deuteron may be stripped, that is, a part of it is retained in the target nucleus, the remainder continuing essentially in the original direction.

Much of the work by nuclear chemists and physicists has been concerned with attempts to characterize particular reactions as proceeding either via compound-nucleus or via direct-interaction mechanisms. Measurements of the formation probability (cross section) of particular radioactive nuclei as a function of bombarding energy (so-called excitation function measurements) have been of particular importance, especially in conjunction with calculations of the results expected on the basis of the compound-nucleus theory. At the high excitation energies of compound nuclei, the shell and collective nuclear models, so useful in accounting for the properties of lower-lying energy levels, are not applicable. The energy levels, in fact, are so closely spaced and so broad that they overlap. It then becomes useful to adopt the approaches of thermodynamics and statistical mechanics and to consider average nuclear properties analogous to temperature and entropy. The emission of particles from compound nuclei has thus been treated by the methods of statistical mechanics in analogy with evaporation of molecules from a liquid. Nuclear chemists have contributed substantially to refinements of these evaporation calculations, in particular by including angular-momentum factors. The inclusion of these effects can significantly alter the shapes of calculated excitation functions. Many instances of reported disagreement between measured excitation functions and expectations from evaporation theory thus need re-examination.

Another important test of compound-nucleus behavior is the measurement of momentum transfer. If the projectile is completely amalgamated with the target nucleus, the reaction products will recoil in the direction of the bombarding beam with the full projectile momentum. The distances to which reaction products penetrate in stacks of catcher foils are used to measure the imparted momenta, and such range measurements are proving very useful in the interpretation of reactions.

When the bombarding particle is an alpha particle or an even heavier nucleus, compound nuclei may be formed with especially large amounts of spin, that is, rotational angular momentum. The way in which such compound nuclei get rid of energy may be considerably altered from the case where the same amount of excitation energy is present but with much lower spin. An interesting technique has provided the essential knowledge of how much of the nuclear excitation energy gets tied up in the ordered rotational motion and how much is left over for the random thermal motion to raise the nuclear temperature. Some nuclei, which are radioactive in their ground states, also have a low-lying excited state with a long lifetime (called an isomeric state). These metastable nuclear states are the analogs of the long-lived phosphorescent states in molecules. The long lifetime of a nuclear isomer is usually a consequence of a large difference in spin from the ground state. Many studies have been carried out on the rate of formation of isomeric pairs of states in various nuclear reactions. The relative rates of formation are indicators of the amount of energy present as energy of rotation in high-spin states of the highly excited compound-nuclear intermediates of the nuclear reactions.

Heavy-ion bombardments are of particular value in the investigation of the effects of high angular-momentum transfers to the reacting system. They also offer unique opportunities for studying other phenomena, such as transfers of nucleon clusters between target and projectile in grazing collisions, and the production of neutron-deficient nuclear species.

Although most reactions induced by neutrons, protons, or alpha particles with energies up to 30 or 40 MeV appear to be interpretable as compound-nucleus processes, there is also considerable evidence for direct interactions even at these low energies, and some reactions appear to exhibit intermediate behavior. It is rather clear that the compound-nucleus and direct-interaction models represent merely limiting cases of what must be formulated eventually as a more general theory of low-energy nuclear reactions.

Reactions proceeding by a direct-interaction mechanism have an additional interest in that they provide information on nuclear energy levels complementary to that obtained by conventional nuclear-spectroscopic techniques. Such direct reactions often lead directly to ground and low-lying energy levels of product nuclei. The nuclear spectroscopist who works with radioactive decay is often limited in the number and kind of excited states of a nucleus that he can study; the nuclear-reaction spectroscopist can frequently form and measure many others. Both physicists and chemists are actively using such direct reactions as deuteron in, proton out (d, p); deuteron in, triton out (d, t); (d, α); (α, d); etc. Such studies are best carried out with accelerators of modest energy and sharp energy definition in the beam. The tandem Van de Graaff accelerators and the spiral-ridge cyclotrons developed in recent years are proving to be ideal for such studies. For the national effort, the importance of basic nuclear research in expanding this type of work has been spelled out in greater detail in the study prepared for the National Science Foundation (13).

c. High-energy reactions. The study of interactions between complex nuclei and high-energy bombarding particles has become predominantly the concern of nuclear chemists. This state of affairs came about in part because such interactions always produce a wide variety of reaction products, and chemical separations are required to sort these out. These radiochemical techniques are now frequently augmented by various techniques of physical measurement.

The study of high-energy reactions depends of course on the availability of high-energy accelerators. Thus, nuclear chemists have explored the energy range of bombarding particles provided by each successive generation of accelerators: the synchrocyclotrons (100-450 MeV) in the late 1940's, the proton synchrotrons (1000-6000 MeV) in the 1950's and the strong-focusing synchrotrons (up to 30,000 MeV) since 1960. A variety of interesting phenomena have been discovered in each of these energy regions and many of them require further detailed investigation. When still higher energies become available (perhaps up to hundreds of thousands of MeV), these explorations should definitely be extended.

Spallation reactions, characterized by the emission of from one to several dozen small fragments (neutrons, protons, alpha particles, and the like), have received the greatest amount of attention up to the present time, both experimentally and theoretically. Such reactions, even with a given target bombarded at a given energy, lead to a large variety of product nuclei (both stable and radioactive). The complex patterns of these reactions and their dependence on bombarding energy, on mass, and on charge of the target nucleus are accounted for in terms of the cascade-evaporation model, in which the reaction is envisaged as taking place in two stages. In the cascade stage the incident particle sets off a rapid ($<10^{-22}$ sec) sequence of collisions between individual particles in the nucleus, leading to the ejection of some of the particles involved in this cascade and to the formation of a more-or-less highly excited intermediate nucleus. This excited intermediate, like the compound nucleus in low-energy reactions, is then de-excited on a slower time scale ($\sim 10^{-16}$ to 10^{-21} sec) by the evaporation of additional particles, as discussed in the preceding section on low-energy reactions. Detailed computer calculations of both reaction stages have led to satisfactory agreement with experimental data on the probabilities of formation of spallation products.

Fission, pictured as the breakup of an intermediate excited nucleus into two or more fragments of comparable mass, also occurs in high-energy bombardments. This process is under detailed study with the use of a variety of chemical and physical techniques, but theoretical understanding of the process is quite limited at present.

A third process, fragmentation, has been postulated to account for the observation of reaction products not easily ascribable to fission or spallation, particularly low-mass fragments formed in large yield from heavy-element targets. Fragmentation is clearly a high-energy phenomenon (threshold ≥ 200 MeV) and appears to be characterized by nuclear breakup on a short time-scale ($\leq 10^{-20}$ sec). Experiments designed to obtain information on the mechanism of this reaction are being pursued intensively.

Attempts to understand the reactions of high-energy bombarding particles with complex nuclei usually start from the premise that they should be accountable in terms of the interactions between elementary particles, as modified by whatever restraints are imposed by the close proximity of other particles and by the nuclear binding forces. Thus, on the one hand, there is hope of using high-energy projectiles as probes for some nuclear structure details and on the other hand, complex nuclei can aid in the study of elementary interactions including some that are not otherwise readily accessible. In this last category are the interactions of short-lived particles, for example, the excited states of neutrons and protons. These entities have such a transitory existence that, when formed in interactions between free particles, they decay before having a chance to interact. However, when formed inside complex nuclei in the cascade stage of a reaction, such short-lived particles may interact with protons or neutrons and thus play a significant part in determining the course of the reaction.

Effects related to details of nuclear structure (such as shell effects) are probably most readily observed in relatively simple reactions and have been searched for in (p, pn) and (p, 2p) reactions in particular. Some striking fluctuations in cross sections have, indeed, been observed among such reactions in neighboring nuclei and have tentatively been interpreted in terms of shell effects in deep-lying nucleon shells; but more extensive and more accurate experiments, for example, on momentum transfers in these reactions, as well as more detailed theoretical analyses, are required to establish the conclusions.

The general trend in the investigation of high-energy reactions is toward supplementing radiochemical and mass-spectrometric cross-section measurements with investigations of the momenta, angular distributions, and angular correlations of reaction products. Such studies have already given important information on the mechanism for various spallation reactions (particularly some simple ones), on the time scale of the fragmentation process, and on the nature of some high-energy fission reactions. They promise to be of much value in sorting out mechanisms of many more reactions in the next few years. The rapid development of solid-state detectors, multiparameter analyzers, and computer methods for data handling and analysis is of particular importance in this work.

The bulk of the high-energy reaction studies up to the present time has been concerned with incident protons. A small number of experiments with incident π-mesons and μ-mesons have been reported. Much work with these and other projectiles (such as antiprotons, K-mesons, and perhaps other strange particles) is needed in order to explore their interactions with nuclei. Such experiments, for the most part, must await increased beam intensities at existing accelerators or the construction of new high-intensity machines such as the meson factories that have been proposed by several laboratories. Pion-induced reactions are of particular interest because the production and subsequent interactions of pions in nuclei have been postulated as an important mechanism for the deposition of large amounts of energy in nuclei by protons with energies above approximately 500 MeV. Direct tests of this hypothesis with incident pions are needed. Also, the variation of cross sections with incident energy for some pion-induced reactions may reveal some otherwise hard-to-observe pion-pion resonances (since virtual pions presumably exist in nuclei).

The study of high-energy reactions by nuclear chemists has already borne fruit by providing new ideas, new insights, and new tools for other areas of science. In particular, the knowledge gained from reaction studies in the laboratory has led to an understanding of the interactions of cosmic rays with meteorites in space. Since there is mounting evidence of important mechanisms for the acceleration of charged particles to high energies in the universe, high-energy reactions are of interest to the astrophysicist also. Such reactions have, for example, been invoked in certain stages of the element-building processes to account for the existence and abundance distribution of some light nuclei in the solar system and in other parts of the universe.

B. RESEARCH ON ATOMIC AND MOLECULAR PROBLEMS

1. Chemistry of Radioactive Atoms

The pioneering work in the inorganic and physical chemistry of the transuranium and other artificially produced elements, as well as some of the fission-product elements, was accomplished largely by nuclear chemists. To this day, chemical research in this area is concentrated in laboratories where nuclear chemistry is strong. There have been other important interactions between nuclear and inorganic chemistry. The requirements for more specific and cleaner separation schemes in nuclear studies have fostered the development of ion-exchange and solvent-extraction methods. Such developments have stimulated work that is leading to detailed understanding of the thermodynamics and kinetics of the chemical processes involved.

2. New Analytical Techniques

One of the most important contributions of nuclear chemistry is the development of new methods of chemical analysis based on nuclear properties. The importance of these methods derives partly from the extreme sensitivities frequently present in the techniques of measurement of nuclear properties, partly from the wide applicability of the techniques, and perhaps most significantly in regard to future trends, from the instrumental character of the techniques that makes them suitable for automatic or control operations. All these methods depend on the measurement of some nuclear property to determine the amount of a given nuclear species present. The property measured may be a decay rate measured by a radiation detector, an isotopic abundance determined mass spectrometrically, or the energy of a nucleus in a magnetic or electric field.

The most widely used nuclear analytical technique, activation analysis, was developed early by nuclear chemists. The sample to be analyzed is exposed to nuclear bombardment, in a reactor or cyclotron, and the amounts of nuclear reaction products are subsequently determined by means of their characteristic radioactive properties (half-lives and radiation energies). The method is specific, since a given constituent gives rise to a particular identifiable product, and the sensitivity for most elements exceeds that of most other analytical techniques. Neutron activation in reactors is most frequently used. Other interesting variations have been developed; e.g., a powerful method for analysis of trace amounts of oxygen and nitrogen by ^{3}He bombardments was reported recently (14).

Another important technique is analysis by isotope dilution. This technique involves adding to the unknown containing the substance to be determined, a known amount of the same substance, but isotopically labeled with either a stable or radioactive isotope. Subsequently, a sample of the substance is isolated (not necessarily quantitatively) and its content of the isotope is determined. The amount of the substance in the unknown can then be computed. The method is exceedingly useful for the analysis of complex biochemical systems and geochemical and other samples.

These new methods of chemical analysis have been widely adopted; their specific applications are far too numerous to list. One of the most important applications has been to the analysis for trace elements in valuable materials. For example, the trace element constitution of meteorites (see below) has been determined by activation analysis. The same technique has been used in archaeological studies tracing the origin and history of ancient glasses and coins. Also, activation analysis has important applications in analyzing materials relevant to crime detection and prosecution. Both activation analysis and isotope dilution techniques are widely used to study complex biological systems including living organisms.

As a final comment on the role of nuclear chemists in analysis, it should be noted that measurement techniques already in use and those likely to be developed in the near future, lend themselves particularly well to simultaneous analysis for numerous elements. In the foreseeable future, chemical analysis for at least twenty elements of a complex system such as a rock will probably be possible on a completely automated basis, with neutron-activation analysis and gamma-ray detection utilizing detectors of high resolution. These new methods of analysis will make possible a better understanding of systems under investigation, more complete control over production, and the liberation of analytical chemists from the drudgery of repetitive analyses.

3. Chemical Effects of Nuclear Transformations

As a result of a nuclear process, whether it is the spontaneous decay of a nucleus or a reaction between nuclei, the atoms of the product species may have large kinetic energies and may be ionized or otherwise energetically excited. Studies of the fate and eventual chemical reactions of such species, observed through their radioactivity, have been carried out in organic and inorganic systems under a wide variety of conditions and in all phases.

The availability of atoms or ions of high kinetic energies (hot atoms) resulting from nuclear processes allows the study of chemical behavior under conditions that are difficult, if not impossible, to achieve by other means. New types of chemical reactions, unknown in the more conventional energy regions previously explored, have been observed in numerous systems, such as those involving recoil tritium, carbon, and the halogens. For example, in the interaction of methane molecules with tritium atoms, generated initially by the nuclear reaction ^{3}He(n,p)T, there occurs, in addition to the well-known reaction of hydrogen abstraction, a previously unknown process in which tritium replaces a hydrogen atom in CH_4 to give CH_3T and H.

The hot-atom work in organic systems has been concerned principally with the reaction mechanisms involved and with the problem of how these can be related to the kinetic energy, excitation energy, or charge state of the hot atom. Most of this work has been with tritium, ^{11}C, ^{14}C, and radioactive halogens, produced in a variety of nuclear reactions. More recently, the hot-atom chemistry of other elements, particularly phosphorus and sulfur, has received increasing attention. In addition to interest for mechanism studies, hot-atom techniques in organic systems have also been applied to the preparation of labeled molecules.

When an energetic atom is produced in a solid by recoil from a nuclear process, a study of the state of chemical combination of such atoms can give information about the nature of the solid state and of crystal defects. Particularly useful in this regard are data on the annealing behavior of recoil atoms. Most work of this type has concentrated on inorganic crystals.

Hot-atom chemists have used simple neutron sources, nuclear reactors, and various types of accelerators to bring about the nuclear reactions that interest them. The use of accelerators has increased in recent years; with these devices it is possible to control and minimize radiation damage to the systems under study and they generally allow great flexibility in experimental conditions.

In the radioactive decay of nuclei, catastrophic extranuclear consequences are often observed. For example, in a process where an orbital electron is captured by the nucleus, filling this vacancy is usually accompanied by ejection of other electrons. If the atom in which this process takes place is bound in a molecule, bond rupture will occur as a result of extensive electronic rearrangements. In the gas phase, ions in high-charge states have been observed as a result of electron-capture decay.

4. Mössbauer Effect and Other Nuclear Methods as Chemical Probes

Most nuclear properties and processes are not subject to ordinary chemical influences. However, a few nuclear phenomena are sensitive to the electronic environment and can thus serve as chemical probes.

The best known of these probes is provided by the process of recoilless resonance absorption, announced in 1958 by the German physicist, Rudolf Mössbauer (15). This process involves the emission of nuclear gamma rays from a radioactive source (such as ^{57}Co emitting gamma rays due to a transition in ^{57}Fe, the product of its decay) and reabsorption of these gamma rays in material containing other nuclei of the emitting species (in this case ^{57}Fe). Mössbauer discovered that under certain conditions (low-energy gamma rays, tight binding of emitting and absorbing atoms in their host materials) the recoil momentum usually imparted to the emitting and absorbing atoms can be taken up by the entire system (for example, a whole crystal lattice), and thus the energies of absorption and emission match almost perfectly. The natural linewidths under these resonance conditions are so small (of the order of 10^{-5} or 10^{-6} eV) that various small effects of the environment, either in the emitter or absorber, can destroy the resonance requirement. These effects in turn can be measured by restoration of the resonance conditions through Doppler shifting by means of modest relative velocities of source and absorber.

Among the chemical effects that can be explored in this manner are electron densities at the nucleus in different chemical compounds (which give rise to different "chemical shifts" of the resonances), as well as the magnetic and electric fields at a nucleus resulting from electron configurations (which cause hyperfine splitting of resonance lines due to the slightly different energies that nuclei with different spin orientations assume in these fields). Use of the Mössbauer effect in studies of crystal field effects, bonding in alloys, and even certain fast-kinetics problems appears promising, but exploitation has only begun.

There are other nuclear methods for probing electric and magnetic fields and thus the chemical environment of the nuclei under study. All of these depend on the small energy differences between otherwise identical nuclei that differ only in the angles between their spin directions and the directions of the magnetic or electric fields. The polarization of nuclear spins at ultra-low temperatures, mentioned in Section IIIA in connection with its use in nuclear studies, can thus be used to obtain chemical information. In 1959, the Soviet physicist Samoilov and his collaborators found that by alloying diamagnetic radioactive atoms into iron, the spins were appreciably polarized at low temperatures. In this manner they established that there are often high magnetic fields at the nuclei of impurity atoms in ferromagnetic hosts. This powerful method is now being utilized by nuclear chemists in this country.

By means of a related technique, the effects of chemical form on angular correlations between successively emitted radiations from radioactive nuclei (such as alpha-gamma or gamma-gamma correlations) are being investigated.

5. Other Interactions of Nuclear Particles in Matter

Some of the problems of atomic structure and chemical behavior can be attacked in quite novel ways through study of systems in which not all the positive charge is carried by protons and the negative charge by electrons. The systems with lifetimes greater than a billionth of a second include those resulting when μ-mesons (muons) or positrons are stopped in matter.

The negative muons usurp the role of electrons as they are captured into orbits of host atoms; their orbits have associated higher binding energies and a far lower distance scale than those of electrons, since the muon mass is more than 200 times larger than the electron mass. This field of study, opened by physicists, appears to hold much intrinsic value for future research by chemists.

Novel chemical considerations arise in the study of the hydrogen-like entities formed when positrons stop in matter. The *ortho*- and *para*-forms (parallel spins and antiparallel spins, respectively) of these positronium atoms have greatly differing properties; the former live much longer than the latter before undergoing annihilation into gamma rays. It has been known for some time that paramagnetic substances catalyzing the *ortho*-*para* conversion in molecular hydrogen do likewise for *ortho* and *para* positronium. Current studies, mostly by nuclear chemists in the Soviet Union, center on the effects of chemical oxidizing agents, free radicals, and so forth, on the rate of conversion of *ortho*- to *para*-positronium.

Still another area in which nuclear chemists are active is in the study of the processes by which energetic atoms and ions interact with matter. The mechanisms for the later stages of the slowing-down process of an energetic ion are not clearly understood. Experimental work frequently involves nuclear techniques for the determination of the range and the range dispersion of initially monoenergetic (radioactive) ions. The energetic ions can be generated in a nuclear reaction or in an accelerator, depending on the energy region under investigation. Some of the interest in the subject stems from the attempt to understand how the enormous kinetic energy of fission fragments is lost in traveling through matter.

C. RESEARCH ON THE MACROSCOPIC WORLD

Ever since the discovery of radioactivity, nuclear chemists have appreciated the fact that their techniques have special characteristics that are particularly suitable for the study of geochemical and cosmochemical problems. Favorable applications of these techniques derive partly from the extreme sensitivity of radioactivity measurement, and partly from the availability, in the decay constants of the different radioactive species, of methods of time measurement on scales extending back many billions of years. Some of the active areas of geochemistry and space research are summarized in the following sections.

1. The Relative Abundance of Nuclear Species

One of the most fundamental experimental pieces of information in chemistry is the relative abundance of the various nuclear species that are the raw material for all chemical studies. It has long been recognized that the crust of the earth represents a distorted version of these relative abundances, and that meteorites and stars are perhaps better sources for information of this type. The chemical analysis of meteorites has relied heavily on nuclear chemical techniques. The results of such analyses have made it possible to establish, in considerable detail, the relative abundances of all known nuclear species in our portion of the universe. In addition, nuclear chemists have provided much of the information on decay constants and reaction probabilities of nuclei under various conditions. The existence of these data has stimulated the development of theories of nucleosynthesis based on plausible nuclear reactions in stars, and of theories of stellar evolution making element synthesis compatible with other astronomical observations.

Although results of analyses of all samples of matter examined up to the present time, both terrestrial and meteoritic, are compatible with the hypothesis that all these samples come from the same process of nucleosynthesis, the exciting possibility exists that a meteorite may be found whose elements were formed at a different time from that at which the elements of the earth were formed. Evidence for this might be in a distorted isotopic composition of an element containing a long-lived radioactive element such as potassium or uranium.

The imminent availability of lunar material brought back in the course of the Apollo program will provide a new type of sample to be analyzed, to a great extent, by nuclear techniques. The information obtained may shed new light on the validity of the presently accepted theory of relative abundances of nuclear species, as well as reveal the origin and history of the moon.

2. Radioactivity Clocks

The disintegration probability of a radioactive nucleus is a characteristic property that is unaffected by external conditions except in very rare special situations. Hence, determination of the amount of product of the disintegration or of the amount of radioactive species remaining can be used to estimate the length of time a system has existed in an undisturbed state. The availability of radioactive nuclei with half-lives in the range from a thousand to many billions of years has made possible the measurements of such times for situations of archaeological and geological interest.

The first application of these dating techniques to terrestrial samples depended on the production of helium in the radioactive decay of uranium and thorium. This method has since been largely supplanted by the study of the isotopic composition of the lead formed from the same radioactive elements. Determinations of the amounts of strontium formed in the decay of rubidium, and of argon formed in the decay of potassium are even more generally useful. They provide unique information on the temporal history not only of the earth's crust as a whole, but of individual ore bodies and rocks. Most of the research in this area is now being carried out by investigators classified as geologists or geochemists, but the measurements at the limit of sensitivity and the establishment of new clocks of this type depend on the intimate knowledge of nuclear structure, as well as involving the newest measurement techniques of radioactivity, which are the domain of nuclear chemistry.

The application of these techniques to the study of meteorites establishes a value of about 4.6 billion years for the time since these rocks crystallized. This figure forms the basis for the accepted age of the solar system. The generally identical isotopic composition of the elements in meteorites and on the earth also supports the hypothesis that the chemical elements composing the various parts of the solar system were formed at the same time and place. However, the recent spectacular discovery in certain meteorites of an excessive amount of ^{129}Xe, presumably formed in the decay of ^{129}I, the half-life of which is only 17 million years, constitutes the first evidence that certain processes of nuclear synthesis occurred during a period less than 100 million years before the solidification of the meteorites.

3. Cosmic-Ray Effects

In all matter exposed to cosmic-ray bombardment, nuclear transmutations lead to the production of induced radioactivity and, in some cases, to the distortion of the usual isotopic composition of an element. In the case of the earth's atmosphere, cosmic rays produce several radioactive nuclear species ranging in half-life from fractions of a second to millions of years. The most famous of these, ^{14}C, having a half-life of 5,700 years, was shown by W. F. Libby to be a useful clock for recent human and geological events (9). It has already become widely used in archaeology and is now being applied to the study of natural phenomena such as the circulation of the atmosphere and the oceans and the times of glaciation in various parts of the world.

The use of other radioactive species produced in the earth's atmosphere is just beginning. The distribution of tritium in various waters of the earth has been demonstrated to serve as a useful medium for studying the transport of water both in the atmosphere and in the ground. Two other radioactive nuclei, ^{32}Si and ^{10}Be, have similar possibilities for the study of sedimentation and circulation rates in the oceans on thousand- and million-year time scales, respectively.

Most of these cosmic-ray-produced species, as well as many others, have also been produced by nuclear-weapons testing in the atmosphere. This has complicated the use of the naturally occurring cosmic-ray-produced radioactive species but has made possible other large-scale experiments, which are described in Section 5 below.

Since meteorites do not have a protective magnetic field or atmosphere in space, they have been subjected to far more intensive cosmic-ray bombardment than the rocks of the earth. The effects of this bombardment can be studied using the radioactive and stable products of cosmic-ray exposure. Study of the tracks of cosmic rays in certain minerals as well as the thermoluminescence of cosmic-ray-exposed material has become a useful method for determining the cosmic-ray-exposure history of meteorites. The use of these techniques has shown that the intensity of cosmic rays, in the regions of space from which the meteorites came, was essentially the same formerly, as it is at present near the earth and hence has not varied widely during the last billion years. It has also been possible to conclude that many meteorites have not always been subjected to cosmic rays; they were inside larger bodies, then exposed by disruption or by abrasion during their travels through interplanetary space.

Recent research has been directed toward detecting the effects of irregular irradiation of meteorites caused by solar protons. The quantitative interpretation of cosmic-ray-induced nuclear reactions in meteorites is dependent upon the extensive use of accelerators producing particles in the energy range of hundreds to many thousands of MeV to simulate the effects of cosmic-ray bombardment on small and large bodies.

4. Space Exploration

With the exploration of space by rocket-launched vehicles now underway, methods are in demand for the analysis of extraterrestrial bodies. Nuclear techniques are particularly adaptable for such applications because of the compact instrumentation, the high sensitivity arising from the detection of individual events, and the digital nature of the information obtained, which is especially suitable for radio transmission. The most direct methods involve measuring the radioactivity of the body being examined. These could give direct information on the amounts of the important geochemical element, potassium, as well as indirect indications of the general composition of the body, based on radiations emitted by cosmic-ray-induced radioactivities.

A practical alternative appears to be actual transport of a small fast-neutron generator to the extraterrestrial body. The gamma rays and induced radioactivity produced by the fast neutrons would be characteristic of the chemical composition, and information on these could be radioed back to earth. An even more compact system that has been proposed uses a transuranium nucleus, ^{242}Cm, as a source of energetic alpha particles. The energy spectra of the alpha particles reflected from a surface as well as those of the protons produced by (α, p) reactions can be measured on the site. From these energy spectra radioed to the earth, the chemical composition of the body's surface can be deduced.

A completely different approach to the study of extraterrestrial bodies uses nuclear chemical experiments designed to measure the neutrino flux on the earth. These experiments use a large quantity of chlorine (100,000 gallons of perchloroethylene) placed in a deep mine to protect it from unwanted, but easily absorbed, radiations. The production of radioactive ^{37}Ar from the chlorine provides a means of measuring the number of neutrinos present. The expected rates of transmutation are only a few atoms a day, an amount detectable through modern techniques. Since the neutrinos effective in producing ^{37}Ar from chlorine are expected to come from the center of the sun, their intensity would provide direct information on conditions in the center of this nearest star.

5. Meteorology and Oceanography

Some of the earliest applications of radioactive tracers to meteorology and oceanography made use of the naturally occurring radioactive species. The circulation of the lower atmosphere was studied by means of the distribution of radon, originally emanating from the earth, as a function of altitude and location. Likewise, attempts were made to study ocean processes by measurements of the degree of disequilibrium between ionium (thorium-230) and its longer-lived radioactive parents.

These classical methods are now supplemented by advanced techniques and knowledge that provide other indicators of processes in the atmosphere and oceans. One line of research has been based on the radioactive products of cosmic-ray bombardment of the atmosphere (^{14}C and tritium). Another tool is the determination of minute variations in isotopic composition of the stable isotopes of the common elements, hydrogen, carbon, and oxygen.

In the case of carbon, all available techniques combine to give a relatively complete, quantitative picture of the rates at which carbon exchanges between the major reservoirs on the earth—carbon dioxide in the atmosphere, organic carbon in living and dead plant life, and ocean carbonate. In modern times the injection of massive amounts of carbon dioxide into the atmosphere from the combustion of fossil fuels has complicated these studies; it has also presented an opportunity for observations associated with the perturbation of a system at equilibrium.

In a similar way, the production of radioactive debris in the atmosphere by nuclear test explosions has complicated the use of naturally occurring radioactive species for such studies. However, it has also made possible some unique studies of the circulation of the atmosphere. For example, the average rate and some details of the transport of air from the stratosphere to the troposphere were established, based on such radioactive debris (16). Similarly, estimates were made possible of the rate of exchange of air between the Northern and Southern Hemispheres.

Applications of these nuclear chemical techniques to oceanography have also been made. They have been hindered by the extreme sensitivities required because the oceans form such a huge reservoir. To cite an example of this type of study, H. C. Urey's measurements on the variation of the oxygen-isotope ratio in certain fossils is a method of estimating the temperatures of prehistoric oceans. Variations in the stable carbon isotope ratio and the ^{14}C content of surface waters relative to those in the atmosphere have revealed the very slow rate of equilibrium between atmospheric and oceanic carbon dioxide. Results obtained thus far on the lateral and vertical distribution of natural radioactivity and radioactive fallout debris from atmospheric-weapons testing indicate that such methods are a promising approach to the study of circulation and sedimentation rates in the oceans.

These studies on circulation on a global scale in the atmosphere and oceans present challenging opportunities for nuclear chemists. Such studies depend on the application of the most modern measurement techniques and require detailed information on nuclear processes in nature. The reward of pursuing these opportunities is information on the large-scale physical and chemical processes that affect all life on this planet.

D. ROLE OF NUCLEAR CHEMISTRY IN THE NATIONAL NUCLEAR ENERGY PROGRAM

Since their discoveries of nuclear fission and of the first transuranium elements, nuclear chemists have played indispensable roles in the nuclear energy program in this country. This has been equally true in both the military and peaceful applications of nuclear energy. The technological requirements of these programs are often so closely related to fundamental research that it becomes difficult to separate the two.

A prime example of close relationship between applied and basic research is manifest in the chemistry of the transuranium elements. The chemical properties of these elements were first established by following their radiations when the elements were available only in unweighably small quantities; on the basis of this information the original large-scale separation processes for plutonium were devised. Similarly, the development of processes for isolating weighable amounts of neptunium, americium, and curium from neutron-irradiated material has depended strongly on nuclear chemical investigations on a tracer scale. Even when availability is not a limiting factor, tracer techniques are often resorted to in the development of new and improved processes for the separation of transuranium elements; radiation hazards are encountered in work with larger quantities of most of these materials.

With the proliferation of reactors built for power and propulsion purposes, the industry of reprocessing reactor fuel is being born. The economics of nuclear power as well as the long-term efficiency with which available fissionable materials can be used depend largely on the cost and efficiency of chemical processes for the purification of irradiated fuel elements. For the development of these processes and for monitoring their operation, nuclear chemical research will be needed on an increasing scale.

Although the design and operation of nuclear reactors are considered primarily within the province of reactor physicists and engineers, certain problems encountered in reactor construction and operation always require the skills of the nuclear chemist. Monitoring the purity of materials needed for construction has, from the start, depended heavily on the new analytical techniques discussed in Section IIIB. In addition, detailed understanding of the neutron economy in a reactor has always been a task for nuclear chemists. Their techniques are used to establish rates of plutonium production and destruction, efficiency of utilization of the fissionable material, and breeding capabilities. And in the design of new types of reactors, extensive studies of the nuclear reaction and disintegration probabilities of individual isotopes of the heavy elements are often crucial. These studies are carried out ordinarily to obtain fundamental data on the stability and reactivity of nuclei.

The production of tritium shows a similar pattern of strong participation by chemists. They were involved in the first large-scale production of this important isotope and were among the first to investigate the special properties of ^{3}He—the product of the radioactive decay of tritium.

Chemists, particularly active in identifying, characterizing, and determining the relative production of the many products of nuclear fission, sought this information originally in order to gain a better understanding of the fission process itself. The data turned out to be essential for the design of proper chemical processes for purifying plutonium, since the fission products are the source of the main radiation hazard in the purification plants. Studies of the fission products, covering the middle third of the chemical periodic table, focused attention on the chemical behavior of many unfamiliar elements. The chemical properties of zirconium and niobium, selenium and tellurium, yttrium and the rare earths, and the new element technetium, became subjects of intensive investigations. The interest in the chemical and analytical properties of these elements contributed strongly to the development of ion-exchange and solvent-extraction procedures which have been widely used since World War II.

The large-scale utilization of the fission products has not materialized as yet. These highly radioactive products are being produced in increasingly large amounts through the expanding nuclear power programs. At present they are considered exceedingly dangerous waste products that require costly provision for long-term storage. To develop beneficial uses of their special nuclear and chemical properties poses a challenge to nuclear as well as other chemists.

Transuranium nuclides such as ^{238}Pu and ^{242}Cm as well as fission products such as ^{90}Sr have already been proved useful as compact power sources, especially in satellites (Systems for Nuclear Auxiliary Power program, known as SNAP). Such applications are destined for increasing importance; nuclear chemistry has a responsibility to help design and develop new devices for the direct conversion of heat from radioactivity into useful energy.

Finally, there is the important role of the nuclear chemists in developing and testing nuclear weapons. They not only provide much fundamental data on the properties of heavy nuclei of importance to this program, but they have also obtained diagnostic information on the behavior of nuclear weapons and have monitored the radioactive fallout following test explosions. These practical activities have been carried out hand in hand with fundamental studies on the fission process, on the production of new elements by multiple-neutron capture, and on the mixing rates in the atmosphere on a global scale.

The separation between technology and fundamental research has increased, of course, with the maturing of the nuclear-energy program. The significance of this area to the national security and economy makes it important to continue the fundamental work in nuclear chemistry in order to provide the basis for further development and the training of personnel in this field.

E. REMARKS ON INSTRUMENTATION

Effective nuclear chemical research depends so greatly on proper instrumentation that the fact must be emphasized. The availability of specialized (and costly) equipment often determines the direction of research in this area; the periodic technological breakthroughs, making possible instruments of higher quality, present opportunities for new types of research and for re-examination of old problems.

Research on induced nuclear transformations is possible only with access to a nuclear reactor or accelerator. These expensive devices have special capabilities for the types of nuclear reactions they produce. Accelerators have usually been designed primarily with the purposes of physicists in mind; nuclear chemists have had to adapt their work to the type of accelerator available. As high-energy particles of first hundreds, then thousands, of MeV were produced, the interactions of these with complex nuclei were investigated; as beams of the new particles became sufficiently intense, the reactions of π- and μ-mesons were studied; with the development of machines to accelerate heavy ions, the new types of reactions produced by such particles were characterized.

This dependence on the characteristics of available accelerators will, no doubt, continue to influence the course of much nuclear chemical research. As the beams of K-mesons and antiprotons become more intense, the reactions of these with complex nuclei will be studied. Yet, the technological developments that made reactor and accelerator design more standard and efficient have made possible the construction of machines which have, as their design goal, the requirements of nuclear chemists. For example, the High Flux Isotope Reactor at Oak Ridge has as its prime aim the production of high neutron fluxes for the manufacture of transuranium elements; the heavy-ion linear accelerator at Berkeley was designed to be effective in synthesizing new transuranium isotopes; cyclotrons at Argonne and at Berkeley were built and are operated primarily with nuclear chemical research in mind.

The dependence on instrumentation is even more pronounced in the realm of nuclear-particle-detection equipment. The bulk of nuclear chemical research has, in the past, involved the measurement of radiations emitted during spontaneous and induced nuclear transformations. In both the detectors for nuclear radiations and in the associated electronic equipment, significant advances in the last 25 years have made possible the re-examination of abandoned problems and a more searching test of developing theories.

Advances in detectors have provided increased sensitivity, improved resolving power, and decreased resolving time. For the detection of electrons, the Geiger counters of the 1940's have been superseded by proportional counters and scintillation counters with a much shorter response time. Unlike Geiger counters, they deliver an output electrical impulse that is proportional to the energy loss in the detector. In the determination of the energies of gamma rays, an essential part of the study of nuclear energy levels, the development of thallium-activated sodium iodide scintillation crystals made possible the enormous amount of research in nuclear spectroscopy in the 1950's. The resolution of these detectors, although a remarkable improvement over what was available previously, is still poor compared with that obtainable in other regions of the electromagnetic spectrum and compared with what is needed to check theoretical predictions of nuclear models.

The newest development in detectors has been that of solid-state semiconductor devices made of silicon or germanium. These detectors provide an improvement in resolution by an order of magnitude over scintillation counters in determining the energy of charged particles (such as electrons, protons, alpha particles, and fission fragments) or gamma radiation. The gamma-ray spectroscopic possibilities are particularly exciting and call for restudy of nearly all known radioactive isotopes with solid-state detectors.

Corresponding to these developments in detectors are significant advances in the electronic components of nuclear chemical instrumentation. These have brought about the increased stability, lower noise levels, higher resolving power, and faster response times required to match the capabilities of the new detectors. In addition, miniaturization and lower power dissipation have resulted from conversion to transistorized circuits.

Employing radiation detectors of higher resolving power would have meant an intolerable increase in time required for measurements, had there not been parallel development of electronic multichannel analyzers. These instruments process electrical impulses of various sizes and store them in computer-type memory systems. Multichannel analyzers make possible simultaneous measurements over an entire energy spectrum instead of at isolated points. Recent developments in this field resulted in instruments that can provide energy-correlated distributions for the outputs of detectors that measure radiations emitted in coincidence by a radioactive substance, or as a result of a nuclear transformation.

This increased efficiency of producing data by means of modern instruments has made much nuclear chemical research dependent on access to high-speed computational facilities. Whether the information generated relates to the variation in response of a detector under various conditions, or to the angular distribution or energy spectra of radiations or fragments produced in spontaneous or induced nuclear transformations, the data are naturally produced in digital form ready for computational processing. In fact, the raw data from many experiments of this type are never examined by the experimentalist in the traditional visual manner, but are fed immediately, or after temporary storage, into a computer. This dependence on real-time or almost-real-time access to computer facilities will inevitably increase in the near future.

Apart from the special relationship of nuclear research to computation that has been underscored here, this area of research, like most others, has benefited by the new computational resources that aid both the sophisticated theorist and the pure experimentalist in relating observations to theoretical considerations.

While nuclear chemists are the beneficiaries of advances in instrumentation, they have also made contributions in this field. In particular, they led in the development of techniques for the measurement of extremely low levels of radioactivity. Such developments were inspired by some of the most challenging problems of nuclear research: the establishment of the radiocarbon-dating method; the detection of a few atoms of a new element; the measurement of cosmic-ray-induced radioactivity in meteorites; and the search for neutrino-induced nuclear transformations. Such problems require the special talents resulting from combined nuclear and chemical training. In spite of the apparently inevitable contamination of the world by man-made radioactivity, the development of new techniques for the measurement of low-level radioactivity will undoubtedly make possible many new investigations of physical and chemical processes taking place on a large scale.

IV

PRESENT ORGANIZATION OF RESEARCH AND TRAINING IN NUCLEAR CHEMISTRY

A. DISTRIBUTION OF RESEARCH EFFORT

A substantial research effort in nuclear chemistry sprang up at the time of World War II. At this time, most of the small number of nuclear chemists in the United States left their research in the universities to work with the secret Manhattan Project that developed the first fission bombs. The demands of that unprecedented project brought many other chemists into direct involvement with a new and developing set of nuclear chemical techniques. At the close of the war, numerous nuclear chemists directed their interests and specialized skills toward problems in basic research; some returned to university laboratories, and some established basic research programs in the newly formed Atomic Energy Commission (AEC) national laboratories. At laboratories where nuclear physicists were using and developing accelerators or reactors, many nuclear chemists directed their efforts at discovery and characterization of new radioactivities, at nuclear reaction studies, and later at more detailed experiments on radioactive nuclei. (Their work centered predominantly in the areas described in Section IIIA.) Other nuclear chemists returned to universities and applied their knowledge of nuclear processes to investigations of extranuclear problems (discussed in Sections IIIB and IIIC).

Basic research on the nucleus itself (branch A), is greatly dependent on ready access to reactors, cyclotrons, or other accelerators. It is strongly concentrated in the large AEC laboratories, principally Argonne National Laboratory, Brookhaven National Laboratory, Lawrence

Radiation Laboratory in Berkeley, and Oak Ridge National Laboratory. These centers have a wider selection of the larger and more expensive kinds of equipment important for many nuclear research problems: reactors of the most advanced design, large particle accelerators, computational facilities, mass spectrometers, magnetic beta and alpha spectrometers, heavily shielded cave facilities, and sophisticated electronic measuring equipment. Several other AEC laboratories with primary applied research responsibilities also have such excellent facilities and carry on some active basic research in nuclear chemistry. Smaller research groups are found in several university laboratories; some of these also have accelerator facilities, but surprisingly, there is a total absence of research in nuclear chemistry at some universities where excellent accelerator facilities exist. Little basic research in this area of nuclear chemistry goes on in industrial laboratories.

Research on atomic and molecular problems (branch B), where nuclear processes are sufficiently involved to designate the work as nuclear chemistry, is found widely dispersed in university and AEC laboratories.

Research on the macroscopic world (branch C) is mainly carried on by small- or medium-sized groups in universities or in various research institutes. In this branch of nuclear chemistry, investigators require special instruments such as low-level counters and mass spectrometers, but not necessarily the larger-scale nuclear accelerators and reactors. Some of this work is carried out in departments other than chemistry.

To gain a more quantitative idea of the distribution of research effort, information has been gathered on the distribution of federal support, the number of research publications in a recent period, and the number of PhD-level nuclear chemists engaged in basic research. At the outset difficulty was encountered in covering the entire field of nuclear chemistry, since research studies in branches B and C often merge into the many related fields of research. The boundaries of branch A, research into nuclear properties, are somewhat easier to delineate, and the most complete of the following information applies only to this branch of nuclear chemistry. Less complete information on the other branches is given where available.

In branch A of nuclear chemistry the predominant support comes from the AEC; in branches B and C there is considerable financial support from the National Aeronautics and Space Administration (NASA), the National Science Foundation (NSF), and other agencies. An examination of the distribution of operating funds for the AEC physical research program in fiscal year 1964 should serve to bring out the essential features of the national support of nuclear chemistry in branch A. These funds from AEC's Research Division provide support for the nuclear chemistry effort at the large national laboratories as well as the major support for contract research at universities.

Figure 1 shows the distribution of research funds for fiscal year 1964 over the whole AEC physical research program. Basic research

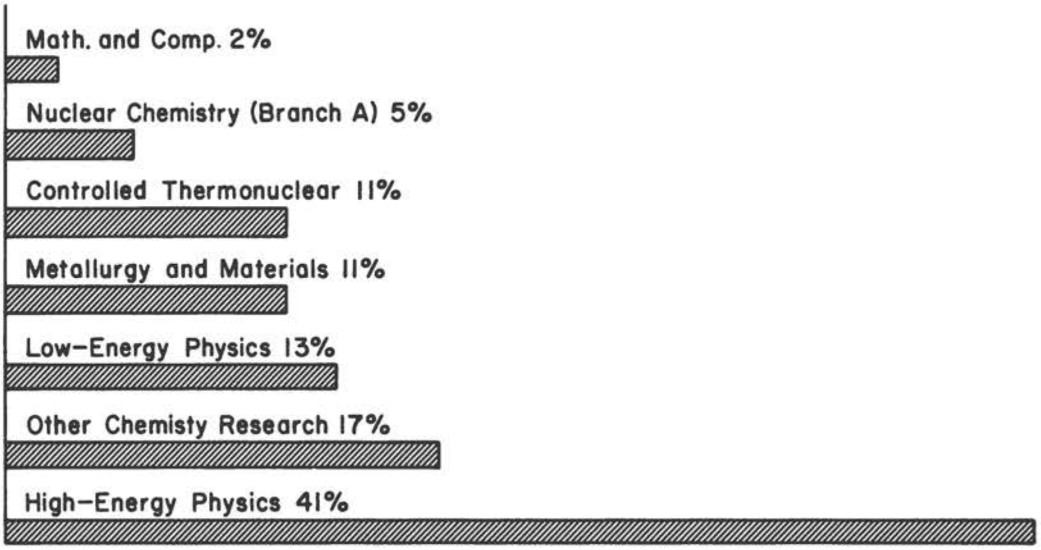

Figure 1. Amount and distribution of AEC funds supporting operations in the physical research program, fiscal year 1964. Total amount, $197,337,000.

in nuclear chemistry branch A received just under 5 percent of the total support of $197 million. Most of this support goes to the four main AEC laboratories engaged in basic research in nuclear chemistry: Argonne National Laboratory, Brookhaven National Laboratory, Lawrence Radiation Laboratory at Berkeley, and Oak Ridge National Laboratory. Of the $9.257 million total support to branch A, $7.657 million was allocated to the above four AEC laboratories,* $1.410 million to research contracts at various universities, and $0.190 million to other AEC laboratories. Research in nuclear chemistry at the Los Alamos and Livermore laboratories is supported by other AEC funds, not from the physical research division.

One of the great problems in measuring value or productivity of fields of basic research is a lack of absolute yardsticks. Alvin M. Weinberg,

---

*The nuclear chemistry research funds for these laboratories include full support of two major accelerator facilities at Berkeley, the Heavy Ion Linear Accelerator and 88-inch variable-energy cyclotron, one cyclotron at Argonne, and partial support of other accelerator and reactor operations at all four sites. Sustaining an up-to-date competitive research effort on a major accelerator calls for considerable electronics and other engineering support, all of which makes for a relatively high support cost per research investigator in nuclear chemistry, compared with other branches of chemistry. For example, the fiscal year 1964 operating expenses exclusive of research costs, for the Heavy Ion Linear Accelerator at Berkeley were $918,000, and those of the 88-inch variable-energy cyclotron were $907,000.

Director of Oak Ridge National Laboratory, has attempted to formulate meaningful criteria in a recent article (17). Journal publications constitute one such measure; publication of results in scientific journals permits criticism and evaluation, provides the principal means by which a given basic research field exercises selectivity, and rates the value of research programs and investigators within the field. It is meaningful, therefore, to count numbers of research papers, despite obvious shortcomings in the use of such figures to gauge scientific value.

The general category, Nuclear Properties and Reactions, was searched in Nuclear Science Abstracts. This category includes most of the published work of branch A of nuclear chemistry. In the first 9 months of the 1955 issues of Nuclear Science Abstracts, there were 689 papers in the above category and in a similar period in 1963, a total of 1,765 papers. Figure 2 illustrates the origin of these papers; in 1955 about half came from outside the United States, and of the papers from U.S. laboratories, about one sixth included at least one chemist as author (in most cases all authors were nuclear chemists). From 1955 to 1963 the U.S. papers had increased by some 50 percent, with chemists' contributions constituting one fifth of the 1963 list. The increase of nuclear research papers from abroad in this 8-year period is tremendous. A nearly threefold increase in the number of papers from abroad reflects in part some variation in coverage by Nuclear Science Abstracts between these two periods, and an increase in Soviet publications after information was declassified at the time of the 1955 Atoms for Peace Conference. The proportion of all published papers in this field that originates outside the United States has grown from one half to nearly three quarters in a period of 8 years.

The NSF report on nuclear-structure physics (13) counted articles in a different way. This report corroborates the growth of published articles in nuclear-structure physics from foreign laboratories and a constant level from the United States. Table 2-1 in the above-mentioned report shows 450 articles from U.S. institutions in 1956 compared with

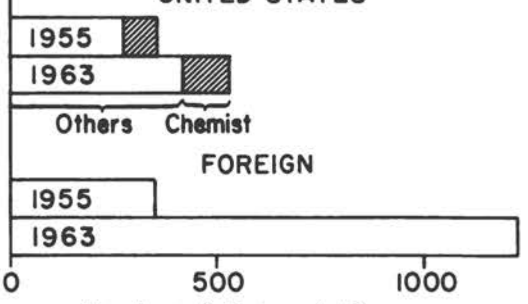

Figure 2. Research papers abstracted over 9-month periods by Nuclear Science Abstracts under the classification "Nuclear Properties and Reactions."

454 in 1960. The number of U.S.S.R. publications jumped from 20 to 136 in the same period, and other non-U.S. publications from 86 to 186. Table 2-2 of the same report shows the predominance of universities concerned with nuclear-structure physics, the ratio of university to nonuniversity publication being 2 to 1 in 1956 and 3 to 1 in 1960. These ratios stand in marked contrast to the distribution in nuclear chemistry, where the four larger AEC laboratories in basic research (including the Lawrence Radiation Laboratory at Berkeley) contributed just under two thirds of the 62 research papers of chemists in 1955, and about the same fraction of the 105 papers in the 1963 period. In this latter period one fourth of the chemists' 105 papers came from universities (other than the University of California, Berkeley) with about one tenth from other AEC laboratories and from industry.

Publications were checked for national origin in areas of nuclear-structure research that are particularly close to the atomic energy field. Research publications on the basic nature of the fission process were counted in Nuclear Science Abstracts for an 18-month period beginning in January 1963. The total of 41 papers was surprisingly small, considering the challenge and importance of the subject and the new instrumentation available. Only one seventh of the papers came from countries other than the United States and the Soviet Union and the remainder were equally divided between these two countries. A check of all publications on nuclear properties of isotopes of actinide elements (thorium and above) shows 59 papers in the years 1962 and 1963. One fourth of the papers came from the United States, one fourth from the Soviet Union, and one half from other countries—principally France and Denmark.

It seemed worth a try to estimate numbers of U.S. nuclear chemists in basic research today. A postcard survey of universities (described in more detail in the next section) and supplementary letters to AEC laboratories supplied names of researchers at the PhD or equivalent level. Complete information was not obtained on nonfaculty postdoctoral personnel in universities, and some laboratories were undoubtedly missed, but the following may be taken as rough estimates of total research personnel at the postdoctoral level.

For branch A nuclear chemistry there are 70 research persons (46 faculty, 24 nonfaculty) in chemistry departments at 31 different universities. The chemistry divisions of the four major AEC laboratories have altogether about 100 nuclear chemists at the PhD level and, in addition, about 20 persons with PhD degrees in physics. Those who hold a joint appointment at a university and an AEC laboratory (7 persons) are included in the AEC total. For the other AEC laboratories as well as all of the other kinds of government laboratories and research institutes, the figures are less complete. It is estimated that another 100 professional-level nuclear chemists devote varying portions of their time to basic research.

For branch C, figures are more fragmentary. The postcard survey of nuclear chemists involved in research in chemistry departments yielded the names of 20 professors in 15 different universities. (Six people were included in both this total of 20 and the branch A total of 46, since their work is in both branches.) The number 20 is probably only a lower limit for two reasons: some scientists classified as nuclear chemists may be identified by department chairmen as geochemists or under some other designation; some nuclear chemists are on faculties of departments other than chemistry, but the number is uncertain. There are a few branch C nuclear chemists in the large AEC laboratories.

B. TRAINING AND EDUCATION

A continuing vital research effort depends on a constant infusion of young scientists from the university graduate schools. The American Chemical Society Directory of Graduate Research (18) provides information on the numbers of PhD degrees awarded in nuclear chemistry over various 2-year periods.

For the 2-year period beginning October 1, 1953, there were 29 PhD degrees in branch A nuclear chemistry awarded from 11 universities, and 8 years later (in the 2-year period beginning July 1, 1961) 44 degrees were awarded from 14 universities. There were 1,981 chemistry PhD degrees awarded (19) in the United States by 89 universities reporting in the 1953-1955 period and 2,345 awarded in the 1961-1963 period by 119 universities. In comparison, 143 doctoral degrees in physics were awarded in the field of nuclear structure (13) during the academic year 1955, a figure nearly the same as in 1951 and 1959. It appears that the nuclear chemistry PhD degrees constitute about 2 percent of all the degrees in chemistry, and slightly more than 10 percent of all the degrees in the field of nuclear-structure research.

In branch C 10 and 14 PhD degrees were awarded in the 1953-1955 and 1961-1963 periods, respectively. The Committee for the Survey of Chemistry reports that 45 PhD degrees were awarded in nulcear chemistry in 1964 (20). Branches A and C averaged 29 degrees yearly for 1961-1963. The parent committee derived its count from a questionnaire to chemistry department chairmen, while our panel counted theses reported in the American Chemical Society Directory of Graduate Research. Part of the difference in these numbers may arise from branch B nuclear chemistry or from radiochemistry and tracer work, not included in nuclear chemistry by the definitions adopted by the panel (cf., Section II). There might be incomplete reporting in the ACS directories, and perhaps the number of degrees in nuclear chemistry was larger in 1964 than in 1962.

In order to determine the extent to which nuclear and radiochemistry are covered in chemical curricula, a postcard questionnaire was sent to chairmen of 325 ACS-accredited chemistry departments asking if separate graduate or undergraduate courses in nuclear chemistry or radiochemistry were given and if other standard laboratory courses included experiments with radioactivity. The names were requested of those teaching a separate course in recent years.

The results of 287 replies are summarized in Table 1. The American Chemical Society Directory of Graduate Research and other ACS reports were used to divide the departments according to the highest degree awarded in chemistry.

As might be expected, a majority of departments granting PhD degrees offer a separate course, a large majority of undergraduate departments do not, and those departments limited to MS degrees are about equally divided. Summed over all departments, about half offer a separate course. In about two thirds, or 181 of the departments, radioactivity experiments are performed in standard laboratory courses. Such experiments range from one half-life determination in one course to several exercises each in physical chemistry and instrumental analysis. A few departments report the incorporation of radioactivity experiments into all of their undergraduate chemistry laboratory courses.

The coverage of nuclear chemistry and radiochemistry in undergraduate and graduate curricula appears to be fairly broad, but there are some disturbing gaps. Of the 141 PhD-granting departments in the United States, replies were received from 134. In 14 of these there is neither a separate course nor the use of radioactivity in standard laboratory courses. This is also the case in 20 of 99 undergraduate departments and 6 of 54 MS departments. No doubt an appreciable number of chemistry graduates at all levels have had no opportunity to study the nucleus in a chemistry course.

The manner in which nuclear chemistry or radiochemistry is taught in the 99 PhD departments offering a separate course can be judged in part by the names of those who have taught such courses. The research interests of the instructors were appraised by considering their recent publications as listed in the 1963 edition of the American Chemical Society Directory of Graduate Research. In 32 departments there was at least one instructor active in branch A or C nuclear chemistry research. Another 18 departments listed at least one hot-atom or radiation chemist. In the remaining 49, the course was given by instructors whose chief interests lay in tracer work or in quite different areas, such as colloids or electrochemistry.

The questionnaire also asked for the names of those sponsoring graduate research in nuclear chemistry. The distribution of 67 such nuclear chemists among 37 departments is given in Table 2. In 18 additional departments there is at least one radiation chemist. Another 6 departments report names of radiochemists with principal interests in analytical uses of radioactivity.

TABLE 1. SURVEY OF INSTRUCTION OFFERED IN NUCLEAR CHEMISTRY*

	Number of Chemistry Departments According to Highest Degree Granted			
	BS	MS	PhD	Total
Separate nuclear course				
With laboratory work and experiments with radioactivity also in other courses	11	12	38	61
With laboratory work on radioactivity only in the nuclear course	4	10	34	48
With lecture only but experiments with radioactivity in other courses	4	3	21	28
With lecture only and no experiments with radioactivity	2	3	6	11
Total separate course	21	28	99	148
No nuclear course				
Some experiments with radioactivity in other courses	55	17	20	92
No experiments with radioactivity	20	6	14	40
Total with no separate course	75	23	34	132
Total all categories	96	51	133	280

* Summary of questionnaires returned by 287 of 325 ACS-accredited chemistry departments. (Seven returns were not sufficiently complete to include in the table.) The postcard questionnaire was as follows:

Does your Department offer courses in Nuclear Chemistry or Radiochemistry?
(please check appropriate boxes)

☐ Graduate ☐ Undergraduate
☐ Lectures only ☐ Lectures and Labs

Name faculty member(s) teaching these courses in past 5 years

Are experiments with radioactivity performed in other standard Lab. courses?
Yes_____ No_____

Name faculty members supervising graduate research in Nuclear Chemistry

Comments:

TABLE 2. DISTRIBUTION OF 67 FACULTY MEMBERS CONDUCTING BASIC RESEARCH IN NUCLEAR CHEMISTRY (BRANCHES A AND C) IN 134 PhD DEPARTMENTS*

Number of nuclear chemists in department	5	4	3	2	1	0
Number of departments	2	2	4	8	21	97

*The Committee for The Survey of Chemistry estimates 90 faculty members engaged in nuclear chemical research, on the basis of their questionnaire (20). The disagreement (90 versus 67) represents partly the total of branch B nuclear chemists and radiochemists, who were not included in the above table.

In summary, the survey indicates that the use of radioactivity in laboratory courses is widespread and that half of the departments offer a separate course in nuclear chemistry or radiochemistry. Some departments that do not now use radioactivity or give a separate course expressed hope or mentioned plans to do so in the foreseeable future. The distribution of nuclear chemists among PhD departments is far from uniform. Eleven percent of the 134 PhD departments collectively have 70 percent of the total, and 70 percent of the departments have no nuclear chemists.

V

FUTURE REQUIREMENTS

The continuous record of exciting research accomplishments over decades, the recent advances in theory, and remarkable developments in research instruments are factors that underscore the great future promise of nuclear chemistry.

Nuclear chemists today play a key role in advancing the knowledge of atomic nuclei and their transformations. They also continue to use imaginatively their knowledge of nuclear processes to attack extranuclear research problems, ranging from the atomic-electron cloud to cosmic space. It is believed that this effort will continue, although it is impossible to predict which specific fields may benefit in the future from developments in nuclear chemistry. Trained nuclear chemists are vitally needed in many phases of the expanding atomic energy industry. For all these reasons, continued strong support for nuclear chemistry is essential. The requirements for continuing vitality in nuclear chemistry research are outlined here.

A. FINANCIAL SUPPORT

The AEC has given strong and enlightened support to nuclear chemistry and it is to be hoped that this policy will continue. A broadened base of support not limited to one agency would be desirable for nuclear chemistry, in view of its demonstrated importance in many areas of science.

The funding of fundamental research in nuclear chemistry, as in many other disciplines, must be planned with allowance for the rising costs of research. Advances in instrumentation will call for continuing increased expenditures. New advanced types of accelerators and reactors of special importance to chemists will be developed, and such devices will probably need to be financed from chemistry funds.

The information in Section IV indicates considerable variation in cost of various nuclear chemistry programs. Much excellent research is being carried out on modest budgets, particularly studies not especially dependent on accelerators, but there is also much that is quite costly when compared with standards in other fields of chemistry. There are reasons for these facts:

(a) Sustaining a major accelerator or reactor is expensive in terms of technical personnel, maintenance, and depreciation on capital investment. Regardless of the research uses, this basic overhead of operation is costly.

(b) When expensive accelerator facilities are involved, frequently the case in branch A nuclear chemistry, it is false economy to carry out experiments inefficiently with inadequate equipment. The optimization of research costs thus dictates use of modern methods, such as, multichannel pulse-height analyzers, magnetic spectrometers, and computers for data reduction.

(c) In branch C nuclear chemistry, there may be need for rare samples of meteoritic material, or samples of particles from the stratosphere, or from ocean sediments, and this sampling may involve considerable direct or indirect costs. For experiments on such material and experiments in space vehicles, thoroughly reliable equipment must be used.

(d) At installations where large amounts of radiation or radioactivity are involved, considerable costs are associated with health protection. Heavily shielded caves and special personnel charged with responsibility for necessary safeguards are indispensable for such kinds of research.

(e) Production of special research materials may be expensive, particularly the separated isotopes and transuranium nuclides, yet these are indispensable for much research.

A country with meager scientific resources might find it advisable to avoid the more expensive kinds of nuclear chemical research, but we feel the investment of money and scientific talent in nuclear chemistry by the United States has been most appropriate and amply rewarding.

B. RESEARCH MANPOWER AND DISTRIBUTION

It is vital to the future excellence of research in nuclear chemistry that able young persons continue to enter the field. In the short term, an increase in numbers of nuclear chemists in research and faculty positions is strongly called for to meet educational and research needs. A natural leveling-off of research manpower can be anticipated in the longer term, as the retirement rate rises for those who entered basic research in nuclear chemistry immediately after World War II and at the end of the Manhattan Project.

Much of the research in nuclear chemistry, branch A, is most effectively carried out in large laboratories with the most advanced facilities. The strength of these centers must be maintained with provision for continued development.

Sound development in both nuclear chemistry and physics requires broadening the research base in the universities. Research in nuclear chemistry in the universities is of excellent quality, but too few persons are involved in this work. A major requirement is for a larger number of universities to appoint nuclear chemists to their staffs. Those few university departments with more than one staff member doing research in nuclear chemistry are particularly effective centers in this field. Adding a few more such groups with a "critical mass" of nuclear chemists would contribute greatly to the strength of the field. Branch A of nuclear chemistry may, of course, remain concentrated in centers near irradiation facilities. Branches B and C are less dependent on such special facilities and can therefore be more widely dispersed.

The development and training of additional PhD's in this area should be facilitated by the availability of both postdoctoral fellowships and junior staff positions at universities and national laboratories.

C. EDUCATION AND INTERDISCIPLINARY COOPERATION

Broadening the research base of nuclear chemistry in the universities can greatly facilitate improvements in education to transmit modern concepts of nuclear behavior to new students. To nonscience students, some cognizance of the behavior of nuclei should be conveyed before and during college, since nuclear energy and use of radioisotopes have attained great importance in our lives. Science students should acquire more quantitative understanding including laboratory experience with nuclear radiation. If all chemistry undergraduates performed a few experiments with nuclear radiations and learned a few of the underlying principles, it would help to dispel the notion that nuclear phenomena are alien to chemistry. Nuclear studies should be dispersed throughout the undergraduate curriculum and should be used with molecular and atomic illustrations to teach general principles. The effectiveness of such dispersal in the chemistry curriculum depends on the interest and knowledge of nuclear subject matter by faculty members involved in teaching the various courses. Some university chemistry faculties follow this approach now; others could do so in terms of their teaching talent. For most schools with one or at most two faculty members having actual research experience with nuclear radiations, separate nuclear courses will be the more realistic and effective approach. Nuclear science has progressed far from its early stage and now can afford great enrichment and stimulation in chemistry education.

Most schools now require graduate students in physical chemistry to study graduate-level organic chemistry and graduate students in organic chemistry to study quantum mechanics. If a similar working knowledge of nuclear chemistry were to be made a basic requirement, at least for all physical chemists receiving the PhD degree, improved teaching about nuclear phenomena to undergraduate chemists could be assured for the future.

For the education of the student specializing in nuclear chemistry, required courses should not be too numerous, so that the curriculum is sufficiently flexible to permit extra courses in mathematics, physics, geology, astronomy, electrical engineering, or other disciplines. Such flexibility is needed not only in nuclear chemistry but also in other modern interdisciplinary fields.

Communication and cooperative work should be fostered among nuclear chemists and specialists in physics, geology, astronomy, and oceanography. Cooperative projects, cutting across various traditional disciplines, should be encouraged and handicaps posed by organizational or jurisdictional boundaries should be eliminated. Nuclear chemists should have close ties with the rapidly developing space program and be given opportunities to carry out experiments concerned with satellites and space vehicles. The inherent global nature of branch C nuclear chemistry puts high premiums on international cooperation for sampling and gathering data.

Many kinds of basic research thrive on the widest possible international communication and cooperation; it is to the advantage of each research group in the world to utilize all previous results and experience. Nuclear chemistry has drawn great strength in the past from the international exchange of scientists and the cooperative use of facilities both at home and abroad. The creativity and efficiency of research in the years ahead will be enhanced by expanding cooperation that spans both disciplinary and national boundaries.

VI

REFERENCES

1. V. I. Gol'danskii, "Nuclear Chemistry and Prospects of Its Development," *Vestn. Akad. Nauk SSSR*, 31:25 (1961) (in Russian).
2. M. Haissinsky, *Nuclear Chemistry and Its Applications*. Addison-Wesley, Reading, Mass., (1964). Translated by D. G. Tuck from the 1957 French edition.
3. G. Friedlander, J. W. Kennedy, and J. M. Miller, *Nuclear and Radiochemistry,* John Wiley & Sons, Inc., New York (1964).
4. G. T. Seaborg and A. R. Fritsch, "The Synthetic Elements: III," *Sci. Am.*, 208 (April):68 (1963).
5. G. T. Seaborg, *Man-Made Transuranium Elements*. Prentice-Hall, Englewood Cliffs, N. J. (1963). Cf. also, E. K. Hyde *et al*. *Nuclear Properties of the Heavy Elements, Vol. 1, Systematics of Nuclear Structure and Radioactivity*. Prentice-Hall, Englewood Cliffs, N. J. (1964), Chap. 5.
6. O. Hahn, "The Discovery of Fission," *Sci. Am.*, 198 (Feb.):76 (1958).
7. W. F. Libby, "Hot Atom Chemistry," *Sci. Am.*, 182 (Mar.):44 (1950).
8. H. Brown, "Age of the Solar System," *Sci. Am.*, 196 (Apr.):80 (1957).
9. W. F. Libby, *Radiocarbon Dating*. University of Chicago Press, Chicago (1955).
10. W. A. Fowler, "The Origin of the Elements," *Sci. Am.*, 195 (Sept.):82 (1956).
11. *Chem. Eng. News*, June 3, 1963: 58.
12. M. Goeppert-Mayer, "The Structure of the Nucleus," *Sci. Am.*, 184 (Mar.):22 (1951).
13. *Nuclear Structure Physics, Research Trends: 1962-67*, National Science Foundation Report, NSF 62-45 (1962).
14. S. S. Markowitz and J. D. Mahony, *Anal. Chem.*, 34:329 (1962); see also *Wall Street Journal* (March 18, 1963) and *The New York Times* (March 15, 1963).
15. S. DeBenedetti, "The Mössbauer Effect," *Sci. Am.*, 206 (Apr.):72 (1962).
16. J. R. Arnold and E. A. Martell, "Circulation of Radioactive Isotopes," *Sci. Am.*, 201 (Sept.):84 (1959).
17. A. M. Weinberg, *Phys. Today,* 17 (3):42 (1964).
18. *American Chemical Society Directory of Graduate Research*, 1955 and 1963 editions, American Chemical Society, Washington, D.C.
19. *Chem. Eng. News,* 33:1351 (1955); 34:1900 (1956).
20. *Chemistry: Opportunities and Needs,* National Academy of Sciences—National Research Council Publication 1292 (1965).