Accomplishments and promise of transplutonium research

The 16 new elements added to the periodic table beyond uranium not only have contributed to our understanding of nuclear and atomic physics, but some have practical uses as well.

O. Lewin Keller Jr, Darlene C. Hoffman, Robert A. Penneman and Gregory R. Choppin

Fifty years ago the periodic table of chemical elements appeared to be complete. Most of the gaps left in the table by Dmitri Mendeleev in 1869 had been filled in, and it appeared that with elements 43, 61, 85, and 87, the periodic table of 92 elements—hydrogen to uranium—would be fully realized.

One of the major scientific advances in the last half century involved the addition of 16 new elements to the periodic table beyond uranium. These discoveries not only extended the periodic table into a new realm of unstable nuclides but also led to a new understanding of the position of the actinide elements in the periodic table. The discovery of the transuranium elements and their development as an area of research were built on a ferment of activity in the general areas of nuclear physics and chemistry in the 1930s—particularly on the discovery of the neutron by Sir James Chadwick and the invention of the cyclotron by Ernest O. Lawrence. Thus, Edwin M. McMillan and Philip H. Abelson in 1940 used neutrons produced at the 60-inch cyclotron at Berkeley to bombard uranium and discovered neptunium in the products; later in the same year, Glenn T. Seaborg, McMillan, Joseph W. Kennedy, and Arthur C. Wahl used the same cyclotron to discover plutonium.

Significant scientific discoveries are frequently coupled with new developments in instrumentation as well as theory. Research on transuranium elements is no exception, and new generations of accelerators are now providing the sort of breakthroughs once seen at cyclotrons. Heavy-ion accelerators at Berkeley, Darmstadt and Dubna can accelerate even uranium ions over the Coulomb barriers of the heaviest target atoms. The heavy ion beams are especially useful when coupled with transplutonium target materials such as curium-248, californium-249 and einsteinium-254 (elements 96, 98 and 99) obtained from the production facilities at Oak Ridge (see figure 1). These beam–target combinations produce isotopes of elements above 100 that provide exciting and unique opportunities for investigating new ideas about fission, the limits of nuclear stability, and for the determinations of atomic and chemical properties in this heaviest element region. The known transplutonium isotopes (see figure 2) now number about 150.

An especially intriguing search is going on for superheavy elements with atomic numbers in the range 110–120 and around 184 neutrons. These elements, if they are discovered, should have especially interesting chemical and physical properties: Their chemistry should be heavily influenced by relativistic effects, and the spontaneous fissioning of their spherical nulcei should be decidedly different from the fissioning of the known nonspherical nuclei.

Solid-state physics in the transuranium region is a little-explored field now offering an opportunity for the increased understanding of electronic phenomena of considerable interest, such as valency fluctuations. The electronic properties of the transuranium elements have long been of interest to the chemist: The periodicity of the electronic properties of atoms underlies the periodicity of their chemical properties. Our understanding of this periodicity is, of course, based on numerous insights in both physics and chemistry. In 1944, for example, Seaborg boldly moved thorium, protactinium and uranium from their long-accustomed places in the last row of the periodic table to a newly proposed "actinide" series placed under the lanthanides (figure 3). The actinide series, as proposed by Seaborg, would show properties based on the filling of a 5f shell of electrons analogous to the filling of the 4f shell in the lanthanides. There is a difference, however, because in the lanthanide series, the 4f orbitals are contracted into an inaccessible inner shell from the very beginning, while the actinide series must reach americium and curium (in the middle of the series, with a $5f^7$ electron configuration) before the 5f orbitals have contracted sufficiently for the characteristic "rare-earth" chemistry to become prominent. Using his actinide hypothesis, Seaborg correctly predicted the chemical properties of elements 95 and 96 (americium and curium); these properties provided the key to their discovery in 1944. Similarly, Seaborg predicted chemical properties of the remainder of the series (elements 97–103) to aid in their discoveries. The

High Flux Isotope Reactor at Oak Ridge is used to prepare transuranium elements. The photo shows a spent fuel element being removed from the pressure vessel; the element will be moved under water to a storage area. The water above the reactor vessel and storage area serves as both shielding and coolant. Figure 1

O. L. Keller is director of the chemistry division, Oak Ridge National Laboratory; D. C. Hoffman is the leader of the isotope and nuclear chemistry division of Los Alamos National Laboratory; R. A. Penneman is leader of the isotope and structural chemistry group in the isotope and nuclear chemistry division at Los Alamos; G. R. Choppin is R. O. Lawton Distinguished Professor in the department of chemistry, Florida State University.

0031-9228 / 84 / 0300 34- 08 / $01.00

ultimate success of these predictions led to world-wide acceptance of the new formulation of the periodic system with the incorporation of the actinide series.

Developing an understanding of the architecture of the periodic system is still an active field of research. This is especially true toward the end of the actinide series (lawrencium, $Z = 103$) and at the beginning of the transactinide series (rutherfordium and hahnium, elements 104 and 105). Electronic relativistic effects can be expected to be of particular importance in this heavy region, especially for s and $p_{1/2}$ electrons. For example, calculations indicate that lawrencium will have a $7s^2\,7p_{1/2}$ electronic configuration rather than the 6d $7s^2$ configuration expected by analogy with lutetium. Similar displacement of 6d electrons by the $7p_{1/2}$ orbitals may occur in the transactinide series somewhere between rutherfordium and eka-platinum ($Z = 110$), thus disrupting the electronic periodicity shown in the lighter analogs. The periodicity of chemical properties, though based on the periodicity of electronic properties, is a broader question because the wavefunctions of the outer electrons are rearranged when the atoms are bound in molecules. The experimental determination of the electronic and chemical properties of lawrencium and of the transactinide elements is therefore an important goal to be pursued.

Chemistry and spectroscopy

Many elements can form compounds requiring different valences. These variations in effective valence arise because the outer electrons—in nominally "filled" shells as well as in the partially filled valence shell—can form hybrid orbitals, consisting of coherent superpositions of the original wavefunctions, at a smaller cost in energy than is gained by the molecular bond. The most familiar example is carbon, whose s and p electron wavefunctions can hybridize to form tetrahedrally oriented sp^3 orbitals or planar sp^2 orbitals, for example. The energy to form atoms with such hybrid orbitals from free atoms is called the promotion energy. It is, of course, a function of the difference in energy between the two hybridizing levels.

In the first half of the actinide series, the promotion energy (5f–6d) is less than that (4f–5d) for the lanthanides, while the reverse is true for the actinide elements heavier than curium. For the light actinides, the valences (up to VII) are thus much higher than for the light lanthanides; in the middle of the series the valence abruptly drops to III, followed by an increasing general tendency towards divalency, culminating in the extraordinary stability of divalent nobelium ($Z = 102$). An important consequence of the unusual electronic hybridization possible for uranium, neptunium, plutonium, and even americium is the appearance of oxo-cations of the form MO_2^+ and MO_2^{++}. These acid-soluble, singly and doubly charged cations are monomeric and linear—a chemical behavior that is unique to the actinides.

Rapid progress is being made in understanding of the electronic structures the actinide elements and of the nature of the bonding in their molecular species. For example, the recent experimental measurement of the atomic spectrum of einsteinium represents a real *tour de force*. Our knowledge of the spectrum of einsteinium (taken with Es^{253}, which has a 20-day halflife) is now on a par with that of many lighter, nonradioactive elements. The use of laser sources to explore the photochemical consequences of selective optical excitation will be an important area of study, particularly for volatile actinide compounds. Techniques are now available to separate isotopes of the lighter actinides; these are based on the coupling of vibronic structure to electronic transitions in the visible and near infrared. Such techniques may work for the heavier members as well.

The spectra of divalent actinide ions in crystals of $CfCl_2$ and $EsCl_2$ have been measured to give the magnitude of the crystal-field interaction. This is the first experimental basis for estimating a complete set of interaction parameters in the f^N-configuration for divalent actinides or lanthanides.

Electronic structure

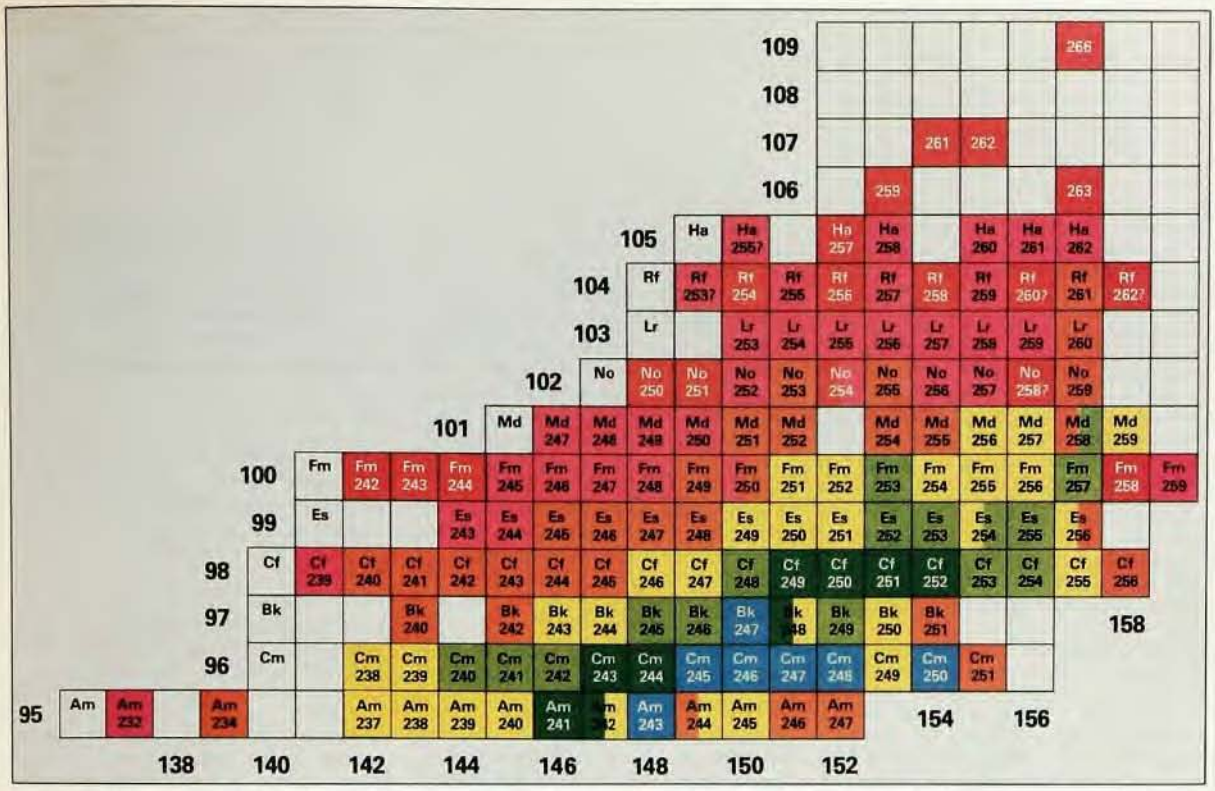
The behavior of the 5f electrons in the readily available lighter actinide metals has been investigated by many experimental techniques involving measurements of, for example, magnetization, electrical transport properties, Mössbauer effect, deHaas–van Alphen effect, x-ray and neutron scattering, heat-capacity and photoelectron spectra. Because pure metal samples of curium-248, berkelium-249 and californium-249 are now available in more than milligram amounts, there is an opportunity to extend the systematics to much higher atomic number. As a result of such systematic studies we should be able to gain a clearer understanding of the very complicated lighter members of the series by extending principles learned from the simpler heavier members. For example, studies of americium superconductivity under high pressure (causing the 5f orbitals to become more available for interaction) would provide very useful information, and studies of americium and of its alloys with plutonium are opening up new insights into the behavior of plutonium.

Plutonium, easily the most complicated metal known, has unusual crystallographic and electronic properties that have been a source of scientific curiosity since the days of the Manhattan Project. Band structure calculations show that in the lighter actinides (thorium through plutonium), the 5f orbitals are somewhat delocalized; they can thus form hybrids with orbitals in the 6d-7s band, resulting in complicated behavior. The electronic behavior becomes simpler in the heavier members because the 5f electrons become quite localized with increased nuclear charge. Some new phenomena appear, however, such as superconductivity (in americium metal). In americium metal, also, the application of sufficiently high pressures can induce uranium-like behavior, apparently by causing overlap of the 5f orbitals with the 6d-7s band. At a pressure of 65 kbar, the crystal structure of americium metal changes to face-centered cubic from the double hexagonal close-packed structure found at atmospheric pressure. At 152 kbar, the americium metal assumes a more complicated orthorhombic structure characteristic of α-uranium and α'-cerium, as shown in figure 4.

The 5f orbitals of the actinides are more spatially extended than the 4f orbitals of the lanthanides; comparing physical and chemical properties of homologous compounds can thus provide valuable insights. Such comparisons, of course, have often been used to advantage in both physics and chemistry. As an example, consider AmB_6 and SmB_6. The latter is an archetypical mixed valency material in which the Sm configuration shifts between $4f^5$ and $4f^6$. However, AmB_6 principally shows the $5f^6$ non-magnetic configuration with fluctuations into the $5f^7$ configuration whose magnetic moment is due only to spin, a situation that is far simpler to deal with experimentally and theoretically.

Nuclear fission and stability

Because isotopes of the transplutonium elements have become available through the US transplutonium production program, one can now study their spontaneous fission. Studies of such isotopes as Cf^{252} ($T_{1/2} = 2.7$ years) Es^{255} ($T_{1/2} = 40$ days) and Fm^{257} ($T_{1/2} = 100$ days) have contributed greatly to our understanding of spontaneous fission. Although spontaneous fission was discovered by Russian scientists in 1940, early studies were hampered by the low specific activity of the available nuclides, such as U^{238}, whose halflife for spontaneous fission is 10^{16} years. With the increasing availability of sources having a high specific activity and with the development of techniques for measuring fission fragment



Isotopes of the transplutonium elements. The chart shows all the nuclides that have been studied to date with atomic numbers greater than 94. The colors indicate halflives, with the shortest-lived (a second or less) nuclides in red, the longest-lived (thousands of years) in blue. Some nuclides have isomeric states with different decay modes and different halflives. Figure 2

kinetic energies with solid-state detectors, it became possible to obtain information concerning details of mass, charge and kinetic-energy distributions, as well as neutron and photon emission for a few spontaneously fissioning isotopes such as Cf^{252}.

Until about 1970, it was generally believed that all spontaneous and low-energy fission resulted in asymmetric mass division and that the spontaneous fission properties of heavier nuclides could readily be extrapolated from the known systematics of lighter nuclides. However, in 1971 researchers at Los Alamos discovered that the spontaneous fission of Fm^{257} produced much enhanced yields of symmetric mass division accompanied by unusually high total kinetic energies. The discovery sparked a renewed interest in spontaneous fission and low-energy fission. In 1979, 10^9 atoms of Fm^{257} became available, making it possible for a Livermore–Los Alamos collaboration to use the (t,p) reaction to produce and to measure the properties of Fm^{259} ($T_{1/2} = 1.5$ sec), the most neutron-rich isotope so far identified. Spontaneous fission of Fm^{258} became accessible for study when the exceedingly rare target Es^{255} was bombarded with α particles, producing via an (α,n) reaction Md^{258}, which decays by electron capture to Fm^{258}. A special preparation of Cf^{254} made possible the production of Cf^{256}, which contains the same number of neutrons as Fm^{258} and also decays by spontaneous fission. More recent work has provided information concerning the spontaneous fission of many additional isotopes—including Md^{259}, No^{252}, Rf^{260}, and Ha^{262}—allowing a comparison of the data for the fermium isotopes with both heavier and lighter nuclides.

A summary of mass-yield data for some of the fermium isotopes is shown schematically in figure 5. Note the abrupt transition between Fm^{257} and Fm^{258} to narrow, symmetric mass distributions. Furthermore, the total kinetic energy for Fm^{258} and Fm^{259} is some 40 MeV higher than the value extrapolated from lighter isotopes. However, Cf^{256}, which has the same number of neutrons as Fm^{258}, has properties similar to the lighter actinides. The only other known nuclide with 158 neutrons is Md^{259}, produced by bombardment of Cm^{248} with O^{18}. Although Md^{259} fissions symmetrically, the total kinetic energy of the fragments is not anomalously high—unlike Fm^{258} and Fm^{259}. Thus, the addition of only one proton results in a profound change in the fission properties of Md^{259} relative to Fm^{258} and Fm^{259}. The widths of both the mass and total kinetic energy distributions for Md^{259} are unusually large, perhaps indicating that the fragment shapes at scission range from spherical to highly distorted. Experiments have ruled out the suggestion that the relatively low total kinetic energy for Md^{259} might be due to the emission of a $Z = 1$ particle at scission. Another possible explanation is that because of the odd proton, one of the fragments is distorted and the kinetic energy from Coulomb repulsion is much lower than for two spherical fragments. Recently, Rf^{260} was prepared from reactions of O^{18} ions with a Bk^{249} target; its properties are similar to those of Md^{259}. An additional intriguing feature of spontaneous fission of the fermium isotopes is that for the Fm^{257} fissions having the highest total kinetic energy, the average number of prompt neutrons is less than 1, even though the average neutron emission per fission is 3.8.

It appears that the fission properties of the heavy fermium isotopes can be explained qualitatively on the basis of fragment shell effects. The high total kinetic energies result from symmetric mass division into two spherical fragments having nearly the doubly magic Sn^{132} configuration, that is, with closed shells of $Z = 50$ and $N = 82$. Such

H 1																	He 2
Li 3	Be 4											B 5	C 6	N 7	O 8	F 9	Ne 10
Na 11	Mg 12											Al 13	Si 14	P 15	S 16	Cl 17	Ar 18
K 19	Ca 20	Sc 21	Ti 22	V 23	Cr 24	Mn 25	Fe 26	Co 27	Ni 28	Cu 29	Zn 30	Ga 31	Ge 32	As 33	Se 34	Br 35	Kr 36
Rb 37	Sr 38	Y 39	Zr 40	Nb 41	Mo 42	Tc 43	Ru 44	Rh 45	Pd 46	Ag 47	Cd 48	In 49	Sn 50	Sb 51	Te 52	I 53	Xe 54
Cs 55	Ba 56	*	Hf 72	Ta 73	W 74	Re 75	Os 76	Ir 77	Pt 78	Au 79	Hg 80	Tl 81	Pb 82	Bi 83	Po 84	At 85	Rn 86
Fr 87	Ra 88	*	Rf 104	Ha 105	106	107	(108)	109	(110)	(111)	(112)	(113)	(114)	(115)	(116)	(117)	(118)

*Lanthanide series	La 57	Ce 58	Pr 59	Nd 60	Pm 61	Sm 62	Eu 63	Gd 64	Tb 65	Dy 66	Ho 67	Er 68	Tm 69	Yb 70	Lu 71
*Actinide series	Ac 89	Th 90	Pa 91	U 92	Np 93	Pu 94	Am 95	Cm 96	Bk 97	Cf 98	Es 99	Fm 100	Md 101	No 102	Lr 103

Periodic Table. This, the modern arrangement, was proposed by Glen Seaborg in 1945. The elements shown in color were discovered after 1935, and in Seaborg's table they were still represented by blank spots. In the earlier version, the actinide elements continued below hafnium as homologues of the transition metals. Seaborg recognized that their chemical properties associate the actinide elements with the rare earths—which had been classified as a separate group of the periodic table since around 1900—and clarified the electronic basis for their chemical properties. Figure 3

spherical fragments will have the maximum kinetic energy from Coulomb repulsion and little internal excitation energy; they thus emit few neutrons or photons. While the properties of the fragment shells can explain some of the fission data, they will not explain the broadly symmetric mass distributions with "normal" total kinetic energies and large half-widths observed for Md^{259} and Rf^{260}. Nor is there any comprehensive model for fission that can explain the sudden changes in properties in going from Cf ($Z = 98$) to Fm ($Z = 100$) to Md ($Z = 101$) while keeping the neutron number fixed. These sudden changes may be due to the disappearance of the second barrier to fission in the potential-energy surface of the fissioning nuclide. Or, perhaps, fission is an "adiabatic process" in which fragment-shell effects dominate; but if so, how are they suddenly "turned off" in going from Fm^{258} and Fm^{259} to Md^{259}? These are a few of the questions remaining to be answered. Future measurements of the properties of still more neutron-rich isotopes of mendelevium and heavier elements are essential if we are to understand these phenomena and their effects on nuclear fission and the limits of nuclear stability. These discoveries concerning fission make necessary a reassessment of the theory of low-energy fission; a comprehensive, dynamic theory of fission that can explain the descent from saddle to scission point, nuclear viscosity and the time scales involved still eludes us.

The experimental results have also had a profound effect on our ideas concerning nuclear stability and the decay properties of the postulated island of superheavy elements with 110 to 120 protons and about 184 neutrons. It should be noted that although relatively stable superheavy elements with high neutron numbers have not yet been produced, elements through atomic number 107, as well as 109, have now been reported, so the very-neutron-deficient edge of the "superheavy" region is already being explored.

Fission properties and barrier parameters for the prompt fission of a host of heavy element isotopes including einsteinium-255 and 256 and fermium-255 and 256 have been deduced from studies of reactions of light projectiles such as d, t and He^3 with exotic targets such as Es^{254}. German and American researchers have carried out Coulomb fission studies at the GSI accelerator in Darmstadt, Germany, using W^{184} and U^{238} projectiles on heavy targets—including Cm^{248}. This method constitutes a valuable probe of the fission mechanism because one can select a purely electrostatic interaction. Current results are not in complete agreement with theory, and further studies are in progress in Darmstadt. In 1962 a group in Dubna, using uranium targets, discovered short-lived fission isomers: metastable states that decay by fission whereas the ground states may decay by other processes. The availability of plutonium and americium targets made it possible to study fission isomers of Am, Cm and Bk as well. A group at Oak Ridge National Laboratory's isochronous cyclotron, for example, investigated the millisecond fission isomer Am^{240m} using laser-induced nuclear polarization to determine the deformation parameter.

Nuclear structure

Nuclear structure theories, whether fully self-consistent or based on V. Strutinsky's macroscopic–microscopic method, contain parameters whose values are adjusted to reproduce properties of known nuclei. The single-particle potential plays a central role in these models and in our understanding of nuclear structure. Comparison of experimental data at the limits of stability with predictions provides a stringent test of the various nuclear models. Examination of the properties of the transplutonium isotopes is especially valuable because, although the liquid-drop model predicts smoothly changing properties with increasing charge, small changes in proton or neutron number in this region lead to rapid changes in stability and properties, as we have seen for spontaneous fission. Studies of alpha decay of transplutonium isotopes provide us with further information on nuclear single-particle levels, complementing the information from the beta and gamma decay processes common in the lighter nuclei. Direct reaction studies of transplutonium targets at accelerators have also furnished much detailed information concerning the arrangement and energies of the proton and neutron levels. For example, single-particle transfer reactions, such as (d,p), (d,t), (^{3}He,d) and (α,t), have proved to be a valuable tool for studying single-parti-

cle energy levels, nuclear structure and deformations and nuclear masses. These studies have involved a variety of targets, including Pu^{244}, Am^{243}, Cm^{246}, Cm^{248}, Cf^{249} and Cf^{250}.

Another important aspect of selecting experimental probes in the testing of nuclear models is the question of completeness. Some approaches, though less selective in terms of providing information about specific states, offer the assurance that all nuclear states within a certain range of angular momentum and parity are populated. Such complete spectral information is provided, for example, by the gamma rays and conversion electrons from resonance-averaged thermal neutron-capture. One can now obtain extensive data on secondary gamma transitions and multipolarities for a significant fraction of the total number of transitions (see the article by Fay Ajzenberg-Selove and Ernest K. Warburton, November, page 26). Such results have been obtained with the high-resolution spectrometers available at the Institut Laue–Langevin in Grenoble for Am^{244}, Cm^{249} and Bk^{250} through neutron-capture gamma-ray measurements on Am^{243}, Cm^{248} and Bk^{249} targets.

Heavy-ion reactions

In the past, the major impetus for the study of nuclear reactions involving transplutonium isotopes has been the desire to produce and study new heavy isotopes and new elements. Recently, considerable interest in studying the various interaction mechanisms themselves has been developing, and many theoretical as well as experimental studies are emerging that link observations in this region to those for lighter elements.

The production of heavy nuclides depends on many aspects of the nuclear reactions. Early experiments, for example, showed that the cross sections for synthesizing the nuclides depend critically on the values of the ratio of the partial widths for prompt neutron emission and for fission from the product nucleus. Subsequent studies have shown that nuclear reaction mechanisms that do not result in the complete fusion of the target and projectile to form a compound nucleus are also important in heavy-element production at accelerators. More recently, studies at Lawrence Berkeley Laboratory show that heavy-ion transfer reactions, as well as compound-nucleus and deeply inelastic reactions, offer hope for producing new heavy-element isotopes with relatively low excitation energies, so that loss of the product by prompt fission or particle emission can be avoided.

American and German collaborators have been particularly active in pursuing the search for superheavy elements and measuring yields of actinides produced via deeply inelastic reactions in bombardments of Cm^{248} with very heavy projectiles such as U^{238}. The use of targets of the heaviest potentially available isotopes, Es^{254} and Es^{255}, offers considerable promise for production of new neutron-rich isotopes and possible stable superheavy elements having a closed neutron shell with N near 184, and having atomic numbers Z in the range 110–120.

The recent discoveries of element 107 and especially 109 at Darmstadt make the island of stability seem tantalizingly close. If relatively stable superheavy elements are eventually discovered at Berkeley, Darmstadt, Dubna or one of the other laboratories working in the field, their chemical properties will certainly be dominated in an unprecedented way by relativistic effects. The spontaneously fissionable spherical nuclei in this region will also offer properties that are not simple extensions from the known fissionable elements, whose nuclei are deformed.

Transplutonium production

The centerpieces of the US Transplutonium Production Program are the High Flux Isotopes Reactor and the Transplutonium Processing Plant, both located at Oak Ridge National Laboratory. These unique production facilities furnish the following key isotopes

- curium-248, 150 mg per year
- berkelium-249, 50 mg per year
- californium-252, 500 mg per year
- einsteinium-253, 2 mg per year
- einsteinium-254, 3 μg per year
- fermium-257, 1 pg per year.

An increase by perhaps an order of magnitude of the production of einsteinium-254 in the near future appears possible. Because it is the highest-Z neutron-rich target producible in microgram quantities, this isotope is particularly useful as a target at heavy-

Transplutonium workshop

Since their construction in the mid-1960s, the High Flux Isotope Reactor and the associated Transuranium Processing Facility at Oak Ridge National Laboratory have provided the western world's supply of elements beyond curium (atomic number 96), either directly or by furnishing starting materials for further nuclear synthesis reactions. Both facilities have a remarkable record of reliability and minimal downtime.

Because the operating costs for the HFIR/TRU complex are relatively high ($13.1 million in FY 1983), the US Department of Energy's Office of Basic Energy Sciences, which bears most of these costs, requested the National Research Council to convene a workshop to provide DOE with an assessment of the importance and future directions of research with transplutonium elements, viewed in the broader context of chemistry and physics.

The workshop, organized under the auspices of the NRC Board on Chemical Sciences and Technology and its Committee on Nuclear and Radiochemistry, was held at the National Academy of Sciences in Washington 28 February to 2 March 1983. Attendance was by invitation, and the approximately 60 participants were carefully selected to include not only practitioners in the various areas of research with transplutonium elements but also experts in related fields not themselves working with these materials, but well qualified to judge their importance.

The plenary meetings of the workshop were interspersed with working sessions of five panels dealing with nuclear reactions and the synthesis of new transuranium species, nuclear structure and fission phenomena, chemical properties, spectroscopy, and solid-state physics and chemistry. Each of the five panels concluded that there are important and challenging scientific problems in its respective area of interest that can be solved only if products of the HFIR/TRU remain available. The uniqueness of the facilities was a central issue in all the workshop discussions. If these facilities were to cease operation, most of the rich body of investigations discussed in the accompanying article would simply come to a halt. A particular point implicit in the workshop considerations is the fact that some of the important products, such as Bk^{249}, Cf^{252}, Es^{253}, Es^{254}, Es^{255} and Fm^{257}, are relatively short lived; they therefore cannot be stockpiled but must be produced continuously.

At the conclusion of the workshop, the participants agreed on the following recommendations:

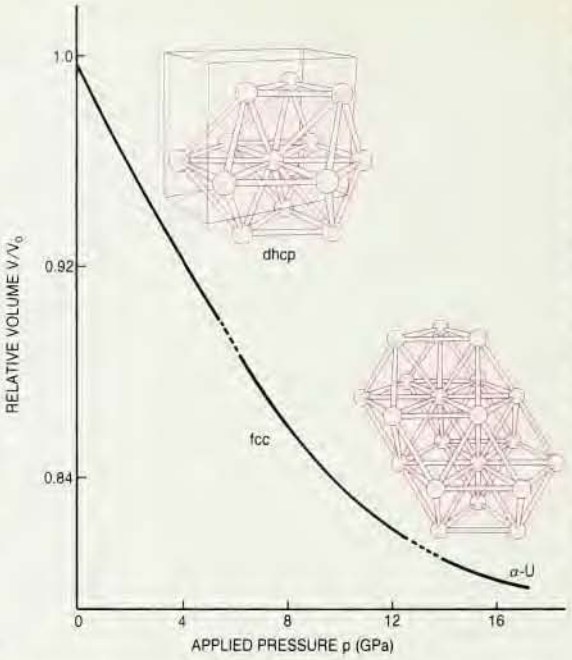
- The operation of these facilities should be continued for at least several years and their status should be reviewed again three to five years from the date of the workshop's report. Such a follow-up review should take into account, in addition to the scientific merits of transplutonium research, a broader range of issues than could be addressed in this workshop, including, for example, the importance of the facilities for training, for research on fuel reprocessing, for neutron scattering, and for medical and defense applications.
- In the meantime, the highest priority in research with transplutonium material should be given to the support of those experimental programs identified in the panel reports as particularly interesting, promising, and crucially dependent on products of the HFIR/TRU.

The workshop report, entitled "Opportunities and Challenges in Research with Transplutonium Elements," is available from the Board of Chemical Sciences and Technology, National Research Council, 2101 Constitution Avenue, Washington, DC 20418.

Gerhart Friedlander
Brookhaven National Laboratory

ion accelerators in the search for superheavy elements, in exploring chemistry of elements 100–106, and in studying the fission of mendelevium and heavier elements.

The processing plant acts as the production, storage and distribution center for these transplutonium isotopes. The plant both fabricates the americium and curium targets for irradiation in the reactor and chemically processes them after irradiation. Novel remote chemical separations processes have been developed for the purification of heavy actinides—through fermium-257—from the multitude of fission products and other elements found in these highly radioactive materials. Remote handling and inspection methods assure the satisfactory performance of the targets under the extreme conditions in which they operate: The reactor flux is in the range $2\text{–}3\times10^{15}$ neutrons/cm^2 sec at the target.



Crystal structure of americium metal changes under compression. At low pressures the structure is double hexagonal close packed. At intermediate pressures the structure shifts to face-centered cubic and monoclinic. At very high pressures (hundreds of atmospheres) the metal assumes an alpha-uranium structure. The α-U structure is characteristic of substances such as U and Pu in which the f orbital and sd orbitals hybridize. High pressure can apparently induce such hybridization in americium. Figure 4

Atomic physics

Investigations into unique aspects of quantum electrodynamics concerned with the effects of the very strong electric fields found near high-Z nuclei are underway in Darmstadt. Targets of Cm^{248} from the US transplutonium production program are being irradiated with U^{238} beams from the UNILAC heavy-ion accelerator of the Gesellschaft für Schwerionenforschung to produce, briefly, assemblages with an effective charge of $Z = 188$. According to quantum field theory, real particles can be spontaneously created from the vacuum if the polarizable aggregate of virtual particles making up the vacuum is destabilized in a strong electric field. (See the article by Jack S. Greenberg and Walter Greiner, PHYSICS TODAY, August 1982, page 24.) Calculations indicate that such a critically strong field can be achieved in an element of atomic number greater than 173. At this critical atomic number, Z_{cr}, the binding energy of the ground electronic state exceeds $2m_0c^2$, twice the rest mass of the electron. A hole in this inner electronic state can spontaneously decay into a positron–electron pair without violating any conservation law, as the created electron goes in the hole, and the positron escapes. Although a true nucleus of an element with $Z > 173$ will probably never be made, a novel approach to forming quasiatoms of such high Z appears possible in heavy-ion collisions: If the energy of the projectile is just below the Coulomb barrier of nuclear reactions—to reduce the chances of nuclear interferences—the two nuclei can be made to approach each other so closely that the electrons dwell momentarily in a potential corresponding to $Z = 188$. Because the electrons move much faster than the nuclei, they adjust their orbits to form briefly a set of molecular orbitals. An example of spontaneous emission may have been observed recently in such collision experiments: A narrow peak appeared in the positron spectrum at about 320 keV. The narrowness of the peak implies a prolonged contact time between the two nuclei—about 40 times the Rutherford scattering collision time. Such a long contact time may be due to the formation of a metastable nuclear complex as the uranium and curium nuclei barely touch. Although it is encouraging that the 320 keV energy of the experimentally observed peak is consistent with the calculated $1s\sigma$ resonance energy of the U–Cm quasiatom, further experiments, involving other quasiatoms such as U–Cf, will be required to determine if the spontaneous emission mechanism is the source of the positrons. Other competing mechanisms, such as interference effects in the dynamical emission of positrons or processes related to nuclear dynamics, could be the explanation as well.

Californium-252

When Cf^{252} was discovered in 1953, its high rate of spontaneous fission immediately suggested the possibility of developing miniature neutron sources of high intensity that might have several applications in research, medicine, industry and education. When the promise of large-scale production of Cf^{252} began to be realized at Oak Ridge in the mid-1960s, the applications followed soon after.

A particularly important use of neutrons from the fission of Cf^{252} is in the treatment of cancerous tumors. A five-year cure rate of 37% has been achieved in, for example, advanced female pelvic tumors, and therapy that uses both neutron and photon irradiation promises further advances. A particularly useful aspect of Cf^{252} in therapy is that the tiny neutron source can be placed in the body adjacent to the tumor; the normal-tissue dose can thus be much less than if the neutrons were projected in a beam from outside.

Neutron radiography using Cf^{252} is also growing in importance. Because of its mobility, a small source containing Cf^{252} can be used, for example, to inspect bonding in aluminum aircraft structures and corrosion effects in operating aircraft.

Within the nuclear industry itself, Cf^{252} is an excellent source for use in reactor start-up. Other actual or potential uses include uranium-borehole analyzers, fuel-rod scanners, fissile waste monitors, and instruments to measure the neutron absorber content of reactor control rods and the hafnium concentration in zirconium. As a neutron source Cf^{252} could also be useful in other industries—for example, in copper and nickel ore analyzers, sulfur meters for coal or oil, analyzers for vanadium in crude oil, moisture monitors and cement analyzers.

Californium-252 sources have also been lent to educational institutions for teaching students how to monitor neutron fluxes and to perform neutron-activation analyses, and also to produce short-lived radioisotopes to be used in teaching techniques of radiochemistry and radiotracing.

The demand for Cf^{252} for applications

in medicine, industry and research has averaged 150 mg annually for many years; the demand has been met entirely by the Department of Energy production program.

Biology and the environment

The environmental and biological behavior of certain relatively long-lived isotopes of neptunium, plutonium, americium and curium are of interest in part because nuclear power reactors produce appreciable quantities of these nuclides. By the end of 1978, the nuclear power industry produced, it has been estimated, about 4500 kg of neptunium, 77 000 kg of plutonium, 1000 kg of americium, and 260 kg of curium. While these amounts may appear to be large, the total quanty of material released to local environments from the industry has been negligible compared to the global fallout of plutonium from nuclear-weapons testing. The current significance of the fallout is small, however: Plutonium currently adds only 0.4% to the natural activity produced by alpha emitters that are present in the upper two centimeters of soil in the northern hemisphere. Since the US, USSR and UK signed the Limited Test Ban Treaty on 5 August 1963, the additions of plutonium from fallout have been significantly diminished.

Alpha emission is usually the important decay process for the environmentally important transplutonium nuclides; in some cases spontaneous fission is also important. The x rays and gamma rays accompanying these decays can also have biological significance—especially in the case of decay products of the shorter-lived isotopes.

Inhalation is currently the most important pathway to humans. Some radionuclides, however, are readily transported through soil and water, and other pathways can take on significance, depending on the environment to which the actinides are released. For example, although ingestion (as food) is usually less important than inhalation, it can become important under certain conditions of environmental chemistry, or because of the long halflives of the actinide nuclides involved.

Several laboratories throughout the world are carrying out animal experiments to determine the biological effects of transuranium elements. Such experiments have shown that most compounds of transuranium elements are not absorbed readily into the bloodstream from the gastrointestinal tract. On the other hand, because material deposited on the lungs stays there for a long time, inhaled compounds are much more likely to be transferred to the bloodstream. Once in the bloodstream, plutonium, americium and curium are chiefly deposited in the bones and in the liver. The resulting possible health risk is cancer from alpha emissions. Because the dose is very low in a vast majority of cases, and because the cancers only show up late in life, the quantitative estimates of the long-term cancer risks are very uncertain. One approach to such estimates is based on animal experiments and the other on epidemiological studies of groups of people who have been exposed accidentally to plutonium. Both kinds of studies have detected no statistically significant cancer incidence from the sort of exposures to plutonium and the other actinides one is likely to encounter in the environment.

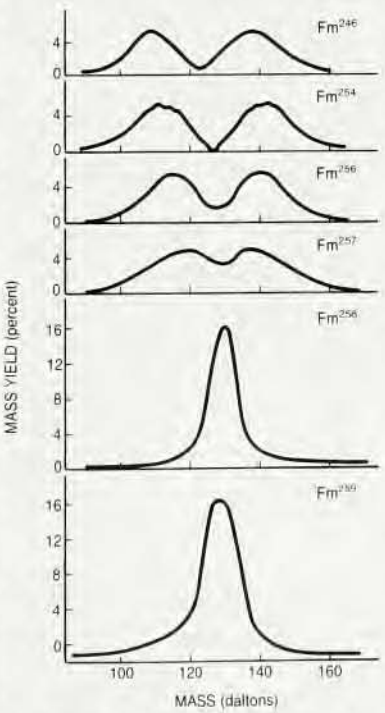
There are concerns about the problems associated with the processing and disposal of plutonium and other transuranium elements that result from energy production by nuclear fission. The concerns increase when fast breeder reactors using plutonium as a fuel are included in the discussion. Several countries are further along than the US with demonstration units of breeder reactors, and these countries are expected to have significant power production with breeder reactors in the near future.

Breeder reactors produce significant amounts of americium and curium. Much more is known about the environmental and biological behavior of plutonium than of neptunium, americium and curium. As the nuclear energy program proceeds toward greater commercialization worldwide, and as breeder reactors take on a significant part, we must maintain a strong program of research into the environmental and biological behavior of the transplutonium elements.

* * *

This paper is based on a report, A Review of the Accomplishments and Promise of US Transplutonium Research—1940–1981, *O. Lewin Keller and Raymond G. Wymer, eds., prepared under the auspices of the Subcommittee on Nuclear and Radiochemistry (Gregory R. Choppin, Chairman), Committee on Chemical Sciences, Assembly of Mathematical and Physical Sciences, National Research Council. The complete report is available from the Office of Chemistry and Chemical Technology, National Research Council, 2101 Constitution Avenue, N. W., Washington, DC 20418. Support for this project was provided by the Department of Energy under Grant No. DE-FG02-81ER 10984. See also "Opportunities and Challenges in Research with Transplutonium Elements: Report of a Workshop," Board on Chemical Sciences and Technology, Committee on Nuclear and Radiochemistry, Commission on Physical Sciences, Mathematics, and Resources, National Research Council, National Academy Press, Washington, DC (1983).*

We wish to thank a number of our colleagues for valuable suggestions in preparing this article, including Raymond G. Wymer, Glenn T. Seaborg, John E. Bigelow, Chester R. Richmond, William T. Carnall, James L. Smith, Donald E. Ferguson, Michael T. Ryan, Richard L. Hahn, C. Davis O'Kelley, Jack S. Greenberg, Jerry B. Wilhelmy, Richard W. Hoff, E. Kenneth Hulet and J. Samuel Faulkner.



Mass yields for spontaneous fission of fermium isotopes. The lower-mass isotopes split in the usual asymmetric way; however, increasing the number of neutrons above 157 makes the most probable mass split symmetric. The most probable mass split for Md^{259} is also symmetric, but the width of the mass distribution is much larger. These results raise questions for the standard theories of nuclear fission. Figure 5

References

1. *Transuranium Elements: Products of Modern Alchemy*, G. T. Seaborg, ed., Dowden, Hutchinson and Ross, Stroudsburg, Pa. (1978).
2. *Actinides in Perspective*, N. M. Edelstein, ed., Pergamon, New York (1982).
3. *Proc. IAEA Symp. Phys. Chem. of Fission, Julich*, IAEA, Vienna (1980), volumes I and II.
4. *Superheavy Elements*, M. A. K. Lodhi, ed., Pergamon., New York (1978).
5. *Transplutonium Elements—Production and Recovery*, J. D. Navratil, W. W. Schulz, eds., A. C. S. Symposium Series 161, American Chemical Society, Washington, D.C. (1981).
7. *The Environmental and Biological Behavior of Plutonium and Some Other Transuranium Elements*, L. Fitoussi, A. Aarkrog, G. F. Clemente, M. Frussel, R. Masse, C. R. Richmond, F. de los Santos Salinas, H. Smyth, F. Gera, Nuclear Energy Agency, OECD, Paris (1981). □