Meetings

Transplutonium Elements

Since the discovery of the first transplutonium elements, americium and curium, research has expanded the number of known elements to 104. It became apparent as early as 1956 that relatively large quantities of the newly discovered elements, berkelium, californium, einsteinium, and fermium (elements number 97, 98, 99, and 100) must be produced if nuclear, chemical, and solid-state research in the field of transplutonium elements was going to proceed. To facilitate production of these elements, the U.S. Atomic Energy Commission gave its approval, in 1960, to build a new reactor at Oak Ridge National Laboratory (ORNL). Known as the High Flux Isotope Reactor (HFIR), it is the most advanced research reactor for producing isotopes in the world today. A complex processing facility, the Transuranium Processing Plant (TRU), was built adjacent to the reactor for fabricating target rods and for separating the highly radioactive man-made transplutonium elements from the irradiated targets. The third unit of the complex is the Transuranium Research Laboratory (TRL), a specially constructed laboratory for conducting research on the heavy elements. The Second International Transplutonium Symposium was held 8–10 November 1966 at ORNL to introduce the recently completed transuranium center to the world.

Glenn T. Seaborg, chairman of the AEC, participated in the symposium and dedicated the new facilities. In the dedication, Seaborg, whose concept of the actinide series serves as the central guide in transuranium research, stressed the importance of future basic research which these new facilities will make possible. He mentioned particularly research on nuclear structure and spontaneous fission, chemical behavior, and biological properties of matter. The distribution of isotopes produced in Oak Ridge to laboratories throughout the United States and other countries makes this an international venture. However, since many transuranium elements decay rapidly they will be available only at Oak Ridge. Consequently, scientists from other laboratories will come to TRL for this special work. Seaborg emphasized the present and potential applications of the transuranium elements. The importance of ^{239}Pu as the explosive ingredient for nuclear weapons and as a nuclear fuel for generating electricity is well known. Less well known are the potential applications of isotopes such as ^{238}Pu; this isotope may be used to power devices like artificial hearts and space nuclear batteries. The projected requirements for ^{238}Pu in space nuclear batteries over the next decade or two run into tons of material. Another isotope, ^{244}Cm, could also be used as a fuel for nuclear batteries. If ^{244}Cm turns out to have the properties required for widespread use as an isotopic power source, one can imagine its future production in ton quantities. Californium-252, which will be produced in gram quantities by the HFIR, has potential application as a point neutron source for radiography or neutron activation where conventional neutron generators cannot be used (as in space).

The first of the new ORNL facilities, the HFIR, was described by A. L. Boch. The HFIR achieved its design power of 100 Mw on 9 September 1966. This reactor uses the flux-trap principle and a very high performance core to attain an unperturbed slow neutron flux of 5.5×10^{15} neutrons per cm^2 in the flux trap. This is believed to be the highest flux ever produced in a routinely operated research reactor. After special targets have been irradiated for an appropriate number of cycles, it may be possible to recover, each year, gram quantities of Cf, hundreds of milligrams of Bk, tens of milligrams of Es, and about a microgram of Fm.

The TRU was described by W. D. Burch. The irradiated HFIR targets, PuO_2-Al cermets contained in aluminum rods are processed in TRU. The heavy elements are separated by dissolving the target in HCl; separating the actinides from fission products and Al by solvent extraction from a concentrated LiCl solution; separating the transcurium elements from Am and Cm by phosphonate extraction from dilute HCl; separating Bk from Cf, Es, and Fm by oxidation of Bk to the +4 valence state, and subsequent extraction into a solvent; and finally separation of Cf, Es, and Fm by cation-exchange chromatography.

The TRL (described by E. H. Taylor) was constructed for research on heavy elements. The building contains gloved boxes and lead-shielded boxes which permit most of the work on the actinide elements. In addition, TRL contains a concrete shielded-box facility which allows working with up to 10 mg of ^{252}Cf. Visiting scientists are expected to compose about half the ultimate staff of 30 chemists and physicists.

In an invited lecture, B. B. Cunningham, Lawrence Radiation Laboratory, Berkeley, stressed the importance of thermodynamics in understanding the chemistry of the actinides. The oxidation states assumed by an element and its compounds reflect a balance between the ionization potential and heat of sublimation of the element on the one hand, and the heats of formation and lattice energies of its compounds on the other. Knowing the ionization potentials of the actinides, which may have to be obtained by some still undiscovered method, is therefore of the greatest chemical significance. Although heats of formation can be measured for compounds containing many different transuranium isotopes, only $^{247}Cm(T_{1/2} = 1.67 \times 10^7$ years) and $^{244}Pu(T_{1/2} = 7.6 \times 10^7$ years) and possibly $^{242}Pu(T_{1/2} = 3.79 \times 10^5$ years) have half-lives long enough for measuring low-temperature heat capacities, which are needed to obtain the entropy. Measurements on these isotopes, which can only be made available through electromagnetic separation, will then be used for extrapolating data to other actinide elements. Magnetic, spectroscopic, and x-ray crystallographic results will be important in allowing these extrapolations to be made.

In the second invited lecture I. Perlman, Lawrence Radiation Laboratory, Berkeley, outlined the essential features of the Bohr-Mottelson Unified Nuclear Model applicable to the transuranium nuclei. Evidence for the rotational structure in the even-even transuranics is available mostly from data on alpha decay. Alpha decay from even-even

nuclei populates the ground-state rotational band uniformly and predictably. Confirmatory evidence for the existence of nuclear rotational motion in even-even nuclei is available from data on gamma decay. The pattern of de-excitation in even-even nuclei is a series of cascade E2 transitions. The pattern of nuclear excited states in odd-A nuclei was illustrated by a discussion of the excited states of ^{237}Np. Levels in ^{237}Np are populated in the β-decay of ^{237}U, in the electron-capture decay of ^{237}Pu, in the α-decay of ^{241}Am, and in Coulomb excitation. The use of ultra-high resolution techniques such as conversion electron spectroscopy with iron-free $\pi\sqrt{2}$ spectrometers and magnetic alpha spectroscopy have led to a fairly complete scheme consisting of over 25 levels. Dr. Perlman pointed out the features of this scheme, such as the various rotational states built upon Nilsson single-particle levels. He also added that theoretical alpha decay rates in the transuranium element region are in excellent agreement with experimental data if pairing effects are included in the calculations.

A special report was given on the production of heavy elements in nuclear explosives (G. A. Cowan, Los Alamos Scientific Laboratory). This production route to the heavy actinide elements proceeds via multiple neutron capture on a fast time scale, ^{238}U·(mult·xn) . . . ^{260}U followed by negatron decay. The mass yield decreases exponentially with increasing mass number. The slope is determined by the time-integrated neutron flux seen by the sample. Integrated neutron fluxes of 10^{24} neutrons cm^{-2} are not uncommon in thermonuclear explosions. One of the interesting results at Los Alamos and the Lawrence Radiation Laboratory is that the nuclide ^{259}Fm may have a very long half-life.

Representatives of various laboratories throughout the world summarized the major efforts and goals of present and planned research in their institutions. O. L. Keller discussed the types of research that the transuranium facilities will make possible at Oak Ridge. The topics include: electron microscopy of hydrous oxides, electronic and molecular spectroscopy of solutions and crystals, single-crystal and powder x-ray crystallography, thermochemical studies, electron spectrometry as applied to problems in nuclear physics and chemistry, application of the Oak Ridge isochronous cyclotron to preparing nuclides of interest in decay scheme studies and nuclear reaction mechanism studies, Mössbauer studies on alloys and compounds, and magnetic structure determinations by neutron diffraction.

A. Ghiorso discussed the production and chemical identification of ^{242}Cf and ^{243}Cf on the Berkeley HILAC accelerator, theoretical calculations of neutron-fission competition in heavy-ion-induced reactions, production and identification of isotopes of element 102, plans for production of elements 104 and 105, modifications to the HILAC for acceleration of krypton ions, plans for production of element 126 in Kr-ion-induced reactions and methods for production of element 114.

R. A. Penneman reviewed the program at the Los Alamos Scientific Laboratory. This includes x-ray and spectroscopic studies of the fluorides of the actinides, a search for alpha emission in the decay of ^{242m}Am, techniques for determining neutron cross sections from nuclear explosions, the use of the proposed high-flux meson facility for chemical studies, studies of Am borides and hydrides, and calculations on the possible electronic configurations of super actinide elements (those containing g electrons).

K. W. L. Bagnall of Atomic Energy Research Establishment (AERE), Harwell, reported that they are studying the preparative chemistry of the actinides. The aim is to accumulate magnetic and spectral data, particularly of 5f systems, from which some understanding of the energy levels and bonding involved in ions of these electronic configurations may eventually be obtained. Work of the Harwell solid-state chemistry group on actinide oxides and on PuS-Se-Te systems was mentioned, together with work on the theoretical calculation of transplutonium element buildup by multiple neutron capture.

A measurement of the fission cross section of ^{242m}Am was reported by R. W. Hoff, Lawrence Radiation Laboratory, Livermore, for neutron energies in the range 0.014 ev to 4 Mev. The measurements were made with a linear electron accelerator that served as a pulsed neutron source and with a spark chamber to detect fission fragments in the presence of intense alpha radiation. Also, studies of the optical spectroscopy of heavy actinides at Livermore were discussed. In particular, the first observations of the absorption spectra of berkelium in solution were reported. Other topics included progress reports on the study of the alpha decay of ^{255}Es and the search for ^{258}Fm.

At the European Transuranium Institute at Karlsruhe, which belongs to the Euratom Joint Research Center, shielded cells have been prepared for the processing of transplutonium targets (W. Müller). The equipment for heavy-element isolation was described. The first capsules (aluminum clad, 7 percent americium-241 oxide-aluminum cermet) have been exposed to an integrated flux of 1.7×10^{22} neutrons cm^{-2} sec^{-1} in the BR-2 reactor at Mol, Belgium. Besides anion exchange, the procedure for processing these capsules includes solvent extraction and reversed phase chromatography. Both involve the use of tertiary and quaternary ammonium salts.

S. Bjornholm, in describing the work in Denmark, particularly emphasized the study of isomeric states in odd-odd americium isotopes that decay by spontaneous fission with millisecond half-lives but not by isomeric transitions. He concludes that these isomers possess a new quality that either enhances the spontaneous fission or retards the gamma rays—or both. First of all, this unusual property does not affect the mass distribution. Upper limits for the excitation energies of the isomers of ^{240}Am and ^{242}Am have been measured by the Danish and Dubna-Bucharest groups. The excitation energy is high. Estimates of the spin for these isomers indicate that it is low. Bjornholm speculated that Strutinsky's analysis of liquid-drop energies, including the "shell" correction, could explain these results since a second minimum appears in the nuclear energy surface. A nucleus caught in such a minimum would be stable toward gamma decay to the ground state—and exhibit fast spontaneous fission too.

A. Chesne and R. Berger reported that the French transuranium program had been initiated in 1959 by irradiation of 40 grams of ^{239}Pu in the EL3 reactor (Saclay). Programs are planned which will produce 100 μg of Cf by 1972. A special laboratory at Fontenay-aux-Roses is used for reprocessing heavy-element targets, fabricating targets for sources, and process development (including a three-step Tramex-type of extraction process). Alpha-gamma coincidence counting, microgravimetry, and coulometry have been used in studies of the analytical chemistry of Am. An electromagnetic mass separator for use with transuranium elements has been in operation at Orsay since September 1965. Also, a cyclotron suitable for heavy-element

isotope production has been operating at Orsay since April 1965.

J. Kooi of Euratom discussed their production program centered on Am irradiated in the BR-2 reactor at Mol. After gross processing at Karlsruhe, the transcurium elements will be purified in laboratories placed at Euratom's disposal in the Institute for Nuclear Physics Research at Amsterdam. Studies in nuclear physics and inorganic chemistry include decay scheme studies, extraction chromatographic separation procedures, and electrochemical studies of actinides in molten salts.

C. H. Ice of the Savannah River Laboratory reported on research aimed toward large-scale production of transplutonium elements. Using chemical technology developed in the transplutonium program and a newly-developed high flux mode of production reactor operation, the production of 4.5 kg of ^{244}Cm was undertaken in 1963. This program should also yield 9.2 kg of ^{242}Pu and 3.0 kg of ^{243}Am. If additional ^{244}Cm is produced by irradiating the ^{242}Pu and ^{243}Am intermediates, 5 to 10 mg of ^{252}Cf per kilogram of ^{244}Cm would be produced as a by-product. In the high-flux mode, one of the large reactors at SRL was operated at fluxes up to 6×10^{15} neutrons/cm^2. During the program, 520 grams of ^{242}Pu was irradiated to produce about 2 mg of ^{252}Cf and the associated transplutonium elements. SRL experiments are designed to measure cross sections of ^{252}Cf precursors. Calculations indicate that the production of ^{252}Cf in a resonance reactor may be increased 100-fold over that in a highly thermalized flux. From this, one can project the possibility of producing hundreds of grams of ^{252}Cf per year at a cost that would make a variety of applications attractive. The pioneering work in Mössbauer spectroscopy of the actinides has been carried out at SRL with the first observations of the effect in ^{237}Np and ^{231}Pa.

D. Cohen discussed the transplutonium chemistry program at Argonne National Laboratory, mentioning reduction of actinide oxides in fused chlorides to the metals; Mössbauer studies on neptunium compounds; absorption spectroscopy; extraction chromatography with a quaternary amine of Am-Cm and Es-Cf; electrical resistivity and Hall effect studies on neptunium and americium metal; and the curium-oxygen system.

A. M. Friedman reported the following topics under study in nuclear physics at Argonne National Laboratory: fission kinetics in the ^{240}Pu (α,α' fission) and ^{239}Pu (d,p fission) reactions, alpha-particle spectra of ^{254}Es and ^{255}Es, production of ^{253}Md and ^{243}Cf in (^{3}He,xn) reactions, extensive studies of states in odd-A nuclei between ^{229}Th and ^{249}Cm as observed in (d,p) and (d,t) reactions.

M. Givon of the Israeli Transuranium group reported a comparison of the spectrophotometric and thermodynamic complex constants for the systems Np, Pu, Am-halide, and nitrate.

The Symposium was sponsored by the Oak Ridge National Laboratory. D. E. Ferguson served as program chairman.

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Note

* Operated by Union Carbide Corporation for the U.S. Atomic Energy Commission.

Calendar of Events

Courses

Actinomycetes and Mycology. Indiana State Univ., 12–16 June. Oriented towards industrial problems. Limited enrollment. Fee, $125. (F. M. Rothwell, Dept. of Life Sciences, Indiana State Univ., Terre Haute 47809)

Bacteriology and Virology. Indiana State Univ., 5–9 June. Oriented towards industrial problems. Limited enrollment. Fee, $125. (F. M. Rothwell, Dept. of Life Sciences, Indiana State Univ., Terre Haute 47809)

Basic Infrared Spectroscopy. Fisk Univ., 14–19 Aug. Designed to introduce beginners to theory and applications of subject. Enrollment limited to 50 participants. Fee, $150. Partial tuition fellowships available for academic personnel. (Director, Fisk Inst., Box 8, Fisk Univ., Nashville, Tenn. 37203)

Behavior of **Liquid Propellants in Space Vehicles 805.9**. Univ. of California, Los Angeles, 19–30 June. Designed for control system and propulsion system design engineers concerned with design of advanced launch vehicles and missiles. Prerequisite: Bachelor's degree or equivalent in science or engineering. *Deadline: 12 June*. Fee, $300. (Engineering Extension, Room 6266, Boelter Hall, Univ. of California, Los Angeles 90024)

Gas Chromatography. Fisk Univ., 14–18 Aug. Enrollment limited to 50 participants. Fee, $150. Partial tuition fellowships available for academic personnel. (Director, Fisk Inst., Box 8, Fisk Univ., Nashville, Tenn. 37203)

Immuno-serological Methods. Mississippi State Univ., 1–7 June. Orientation towards industrial problems. Enrollment limited to 20 participants. Fee, $125. (J.