LBL-26112

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

Presented at the Symposium on Transuranium Elements
Today and Tomorrow, European Institute for
Transuranium Elements, Karlsruhe, FRG,
October 26, 1988

Recent Research on the Heavy Transuranium Elements

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September 1988

NUCLEAR SCIENCE DIVISION

Prepared for the U.S. Department of Energy under Contract Number DE-AC03-76SF00098.

LBL--26112

DE89 002961

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Talk at the Symposium, "Transuranium Elements Today and Tomorrow," European Institute for Transuranium Elements, Karlsruhe, Federal Republic of Germany, October 26, 1988.

This work was supported by the Director, Office of Energy Research, Office of High Energy and Nuclear Physics Division of the U.S. Department of Energy under Contract DE-AC03-76SF00098.

Recent Research on the Heavy Transuranium Elements

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We are nearing the 50th anniversary (1990) of the discovery of the first transuranium element. This half century has seen a tremendous amount of research on these elements, whose number now stands at 17 (Figure 1). A recent review covering only their chemical properties [1] devoured nearly 2,000 pages and included a total of about 7,000 references.

This review is devoted to recent research (performed in the 1980s) on the heavy members of the group, defined as the transcurium elements (with a few references to related work on lighter elements). It covers the discovery of the three heaviest elements, the discovery of new isotopes, investigation of interesting decay properties of some previously known isotopes, investigations of some heavy ion reaction mechanisms in this region, and recent investigations of nuclear and chemical properties. It is not exhaustive in its coverage, is necessarily succinct, and inevitably places emphasis on those aspects with which the author is most familiar.

There have been excellent reviews recently written by G. Münzenberg [2] on recent advances in the discovery of transuranium elements, which includes descriptions of experimental techniques; Y. T. Oganessian and Y. A. Lazarev [3] and P. Armbruster [4], which include descriptions of the synthesis of the heavy transuranium elements by heavy ion reactions; D. C. Hoffman and L. P. Somerville [5] on spontaneous fission; and O. L. Keller, Jr. [6] on the experimental methods used to investigate the chemical properties of heavy actinide and transactinide elements.

Element 107, 108, 109--and more?

During the 1980s, investigators working at the Gesellschaft für Schwerionenforschung (GSI) at Darmstadt, Federal Republic of Germany, synthesized and identified (i.e., discovered) isotopes of the elements with atomic numbers 107, 108, and 109, produced by the "cold fusion" reaction bombardments of target nuclides in the region of closed nucleon shells at lead and bismuth with heavy ions furnished by the UNILAC accelerator. The potential efficacy of such a reaction was first suggested in 1974 by Oganessian and demonstrated by Oganessian and coworkers [7] working at the Joint Institute for Nuclear Research at Dubna in the Soviet Union. The expectations were to observe product nuclides with odd numbers of nucleons (odd-even, odd-odd, or even-odd) because these have substantially longer half-lives for decay by alpha-emission or spontaneous fission than do even-even nuclides.

Element 107

Münzenberg et al. [8] in 1981, working at GSI, clearly identified the nuclide $^{262}107$ produced in the "cold fusion" reaction $^{209}_{83}Bi + ^{54}_{24}Cr \longrightarrow ^{262}107 + ^{1}_{0}n$. The recoiling product nuclei from the nuclear reaction were passed through a velocity separator called SHIP (Separator for Heavy Ion Reaction Products) [9] which guaranteed that they had the characteristic velocity of the product of complete fusion of projectile and target nuclei. The products were implanted into position sensitive surface barrier detectors. The mass number of the velocity-separated product nucleus was determined approximately by measuring its time of flight and its atomic number and mass number were determined by observing the time-correlated alpha-decay to the previously known daughter, $^{258}_{105}Ha$, granddaughter, $^{254}_{103}Lr$, etc. (the method of genetic relationship). A total of about five alpha-decays of $^{262}107$ were observed with an energy of 10.4 MeV and half-life of approximately 5 milliseconds (ms). The cross section for producing these nuclei was 2 x 10^{-34} cm^2, i.e., 0.2 nanobarns (nb), approximately 1/5,000,000 of the production cross section observed in the first one-atom-at-a-time experiment in the discovery of mendelevium (atomic number 101) [10]. In other experiments Münzenberg et al. [11], with the same target-projectile combination and by the method of genetic relationship, identified a few atoms of the isotope $^{261}107$ with a 6 ms half-life and an alpha-energy of about 10.1 MeV and a few atoms of an isomer of $^{262}107$ with a 56 ms half-life and an alpha-energy of about 9.8 MeV. By 1988, a total of 38 atoms of Element 107 had been observed [12], 14 atoms of $^{262m}107$ with a half-life of 8 ms and 8.3 MeV alpha-particles, 15 atoms of 100 ms $^{262}107$ (9.9 MeV alpha-particles), and 9 atoms of 9 ms $^{261}107$ (10.1 MeV alpha-particles). Also using the same target-projectile combination Oganessian et al. [13] identified ^{246}Cf, which they claimed to be a decay descendent of $^{262}107$.

Oganessian et al. [14] in 1976, working in the Joint Institute for Nuclear Research at Dubna in the Soviet Union, had reported the production in a cyclotron of an isotope, which decayed by spontaneous fission (SF) with a half-life of approximately 2 ms, from the reaction of $^{209}_{83}$Bi with $^{54}_{24}$Cr, which they attributed to $^{261}107$. However, the evidence is not sufficient to assign an atomic number.

Element 108

Münzenberg et al. [15] in 1984, identified three atoms of Element 108, in the form of the isotope $^{265}108$, produced by the $^{208}_{82}$Pb ($^{58}_{26}$Fe, 1n) reaction, again by the method of genetic relationship. The nuclide decayed with a half-life of approximately 1.8 ms, emitting alpha-particles with an energy of 10.36 MeV. The cross section for production of these nuclei was 1/10 of that observed for the production of Element 107 (i.e., approximately 2 x 10^{-35} cm^2 or 20 picobarns). In a second experiment [16] one atom of the isotope $^{264}108$ (half-life approximately 80 microseconds) was produced by the ^{207}Pb (^{58}Fe, 1n) reaction and identified by the method of genetic relationship. It was notable that this heaviest known even-even nuclide decays by alpha-particle emission rather than by spontaneous fission and that it has so relatively long a half-life.

Also in 1984, Oganessian et al. [17] reported the very indirect observation of the possible alpha-decay of $^{263}108$, $^{264}108$, and $^{265}108$ produced in the reactions $^{209}_{83}$Bi ($^{55}_{25}$Mn, 1n), $^{207}_{82}$Pb ($^{58}_{26}$Fe, 1n), and $^{208}_{82}$Pb ($^{58}_{26}$Fe, 1n), respectively. None of the alpha-particle decays of the Element 108 isotopes were observed directly. Identification was surmised on the basis of observation of descendants from alpha decay--$^{255}104$ from $^{263}108$, $^{256}104$ from $^{264}108$, and ^{253}Es from $^{265}108$. However, such inferences cannot be accepted as proof of the discovery of an element because the observed activities could have been produced directly.

Element 109

Münzenberg et al. [18] in 1982, after eleven days (equivalent) of bombardment, observed one unusual time-correlated decay sequence that occurred for a reaction product that had been velocity-separated by SHIP from the $^{209}_{83}$Bi + $^{58}_{26}$Fe reaction. The product $^{266}109$, which decayed in 5 ms (half-life 3.5 ms) with the emission of an 11.1 MeV alpha-particle, was partially identified by the method of genetic relationship. Such a yield corresponds to a formation cross section of very approximately 10^{-35} cm^2, or 10 picobarns. In a second experiment [19], in 1988, two more time-correlated decay sequences similar to the first were found.

Oganessian et al. [13] used the ^{209}Bi (^{58}Fe, n) reaction to identify ^{246}Cf, which they inferred to be a decay descendent of ^{266}109.

Element 110

Y. T. Oganessian et al. [20] in 1987 reported the production in the reaction of $^{44}_{20}$Ca with ^{232}Th, with a cross section of 8 picobarns, of a 9 ms spontaneous fission activity, which they assigned to an isotope of element 110 (possibly ^{272}110). A similar activity was also produced, and so assigned, in the reaction of $^{40}_{18}$Ar with $^{235,236}_{92}$U. The evidence is not sufficient to assign an atomic number. An attempt [21] by a GSI team to observe this activity from the reaction of ^{40}Ar with ^{235}U, using SHIP, led to negative results. Additional exhaustive attempts [21] by a GSI team to produce and identify element 110 by the reaction $^{208}_{82}$Pb + $^{64}_{58}$Ni ----> ^{271}110 + 1n have also led to disappointment. A. Ghiorso [22] is attempting another approach through the reaction $^{209}_{83}$Bi + $^{59}_{27}$Co ----> ^{267}110 +·1n, using a rebuilt version of SASSY (Small Angle Separating System) [23], a gas-filled on-line recoil separator, to separate and identify the expected product. An electrostatic separator device, called "VASILISA," has been built at Dubna [24] for the separation of heavy-ion beams from reaction products of complete nuclear fission reactions.

Heavier elements?

Considerations by theoretical physicists, beginning more than 20 years ago, led to the prediction that there should be an "Island of Stability" in a region of spherical nuclei at or near atomic number 114 and neutron number 184, which hopefully might be reached by bombardment of heavy target nuclei by heavy ions. There have been more than 25 publications describing futile efforts to reach this region of "Superheavy Elements" [25, 26]. The most recent efforts, by both the Dubna group [27] and the collaborative work of the

GSI-Berkeley-Los Alamos-Mainz-Bern-Göttingen groups [28] have used the promising approach of bombarding ${}^{248}_{96}$Cm with ${}^{48}_{20}$Ca projectiles to produce a product such as ^{294}116 (N=178), but these comprehensive experiments have also yielded negative results.

However, the product of even this reaction is some half dozen neutrons short of the objective of N=184 (although P. Möller et al. [29] suggest that this spherical shell is pushed down to N=178). A closer approach to N=184 might be achieved by using ${}^{254}_{99}$Es as a target, but this is hampered because it is presently available in very limited (microgram) quantities. An interlaboratory group in the United States (Berkeley-Livermore-Los Alamos-Oak Ridge) is proposing that this nuclide be produced in larger amounts (40 micrograms) for this purpose [30]. As will be seen below, the use of ^{254}Es, in even the limited quantities, has already led to the production and identification of many new heavy transuranium nuclides through some interesting reaction mechanisms.

More recent calculations [29, 31] suggest that there should be stabilizing, deformed nuclear shells (or subshells) at lower neutron numbers, such as N=162. Some of the above described attempts to synthesize element 110 were designed to reach a neutron number near such a subshell. However, an attempt by Hulet et al. [32] to detect the alpha-decay of ^{272}108 (N=164) as the electron capture daughter of ^{272}109 (N=163), produced in the reaction ${}^{254}_{99}$Es (${}^{22}_{10}$Ne, 4n) was unsuccessful, leading to the conclusion that the stability is less than anticipated. Similarly, M. Schädel et al. [33] failed to detect ^{266}107 (N=159) in the reaction ^{254}Es (^{16}O, 4n).

The effects of a rather distinct deformed shell at N=152 were clearly seen [34] as early as 1954, in the alpha-decay energies of isotopes of californium, einsteinium, and fermium. In fact, a number of authors [35] have suggested that the entire transuranium region is stabilized by shell effects with an influence that increases markedly with atomic number. Thus, the effects of shell structure lead to an increase in spontaneous fission half-lives of up to about 15 orders of magnitude for the heavy transuranium elements, the heaviest of which would otherwise have half-lives of the order of that for a compound nucleus (10^{-14} seconds or less) and not of milliseconds or longer, as found experimentally (Figure 2). This gives hope for the synthesis and identification of several elements beyond the present heaviest (Element 109) and suggests [35] that the peninsula of nuclei with measurable half-lives may extend up to the "Island of Stability" at $Z=114$ and $N=184$ (or $N=178$).

Recent nuclides and decay modes

Recent nuclides

There are a total of about 130 known curium and transcurium nuclides (not counting so-called spontaneous fission isomers) ranging in half-lives from milliseconds to centuries (Figure 3). As an example, notable recent additions are five new nuclides identified by E. K. Hulet and coworkers as the result of the bombardment of microgram quantities of ^{254}Es with heavy ions such as ^{22}Ne in the Berkeley 88-Inch Cyclotron.

In chemically separated lawrencium (Element 103) fractions they [36] identified ^{261}Lr and ^{262}Lr, which decay by spontaneous fission (SF) with half-lives of 39±12 and 216±15 minutes, respectively, and with formation cross sections of 240 and 37 nb, respectively. These half-lives are substantially longer than those of previously known lawrencium isotopes and, thus, now allow detailed studies of the chemical properties of lawrencium. In follow-up work they [37] found that ^{262}Lr decays partially by electron capture to ^{262}No, which decays via bimodal SF with a half-life of 5 ms. Earlier, they [38] reported the identification of the new nuclide ^{260}Md, which decays via SF with a half-life of 32 days. The fifth new nuclide identified by this group [39] is the 60-minute ^{258m}Md, an isomeric state of ^{258}Md which decays by electron capture to ^{258}Fm, the 360-microsecond SF activity.

L. P. Somerville et al. [40], using actinide targets, have identified a 50 ms SF activity assigned to ^{262}Rf, have tentatively identified a 100 ms SF activity as due to ^{260}No, and have established the SF half-life of ^{260}Rf as 21 ms (thus, refuting the claim of G. N. Flerov et al. [41] to the discovery of Element 104 in 1964 based on a reported SF half-life of 0.3-second for ^{260}104) [42].

In addition, about a dozen other transcurium nuclides have been identified recently (in the 1980s) as the result of bombardments of lead and bismuth targets with heavy ions [2, 3, 4].

New decay modes

In other work the Hulet group reported that the principal mode of decay of ^{260}Md is indeed spontaneous fission [43], discovered that the spontaneous fission is bimodal [44], and that the neutron multiplicity [45] is 2.58±0.05, substantially less than for other actinides,with a peak at 1.80±0.05 neutrons/fission corresponding to fissions with high total kinetic energies (TKE) and a peak at 3.91±0.09 neutrons/fission for fissions with a low TKE. They [44] also observed bimodal fission in ^{258}Fm, ^{259}Md and ^{258}No. Bimodal fission was first observed and explained by Hulet et al. [44]; it is interpreted as due to two nearly equal minimum-energy paths (fragment-shell-directed and liquid drop fission) along the potential energy surface from saddle to scission [46, 47, 48, 49]. P. Möller et al. [46] have concluded that the fragment-shell-directed scission path, leading to products with closed shells at N=81, lowers the spontaneous fission half-life for nuclei with N=162 to the extent that such nuclei are not stabilized to the extent suggested by earlier predictions [29, 30].

Nuclear reaction mechanisms

Nuclear fission is the dominating reaction observed when transuranium target nuclei are bombarded with heavy ions to produce transuranium nuclides. The "loss" due to fission increases dramatically with increasing atomic number of the compound nucleus so that in the production of the known nuclei with the highest atomic numbers, the survival rate is less than one in 10^{10}. Rather complete studies of so-called "spallation-fission" competition in the transuranium region, with helium ions as projectiles, were made as long as 30 years ago [50]; these demonstrated the important role of other reaction mechanisms than that of compound nucleus formation.

Recent work has elucidated in detail the role of non-compound nucleus "transfer" reactions in the production of broad ranges of nuclides from reactions with heavy ions. As an example, a large collaborative effort [51, 52] (GSI-Mainz-Los Alamos-Berkeley-Bern) has used ^{48}Ca ions (and ^{40}Ca) in the UNILAC (GSI) and SuperHILAC (Berkeley) to bombard ^{248}Cm. Using radiochemical techniques they identified "transfer" products (produced by quasielastic, deep inelastic, direct interaction, etc., mechanisms) all the way from radon (Element 86) to fermium (Element 100). The peak isotopic production cross sections range from hundreds of microbarns for radon (Element 86) to uranium (Element 92), increasing to thousands of microbarns at berkelium (Element 97), then dropping steadily down to microbarns at fermium (Element 100). The differences between the positions of the maxima in the isotope yield curves for berkelium, californium, and einsteinium for ^{48}Ca and ^{40}Ca as bombarding projectiles was only about two mass numbers.

Other examples can be cited from the work of D. C. Hoffman and coworkers using the 88-Inch Cyclotron at Berkeley. In an early publication they [53] reported the production cross section for nuclides with atomic number greater than the target nucleus from interactions of ^{16}O, ^{18}O, and ^{20}Ne, ^{22}Ne with ^{248}Cm. The maxima of the yield curves are about two mass numbers heavier for reactions with the heavier ion of each projectile pair. The production cross sections were found to decrease with increasing atomic number of the products and ranged, for the peak isotopic yields, from a few millibarns for berkelium and californium isotopes, to a few microbarns for einsteinium and fermium isotopes, to the order of nanobarns for mendelevium (Element 101) and nobelium (Element 102) isotopes. The Hoffman group also investigated the yield of products with atomic numbers larger than that of the target nucleus for the interaction of ^{18}O projectiles [54] and ^{16}O projectiles [55] with ^{249}Cf. Surprisingly, the peak yields for the einsteinium and fermium isotopes are in the millibarn region, indicating that the yields of products with $\Delta Z = +1$ and $\Delta Z = +2$ are nearly the same for curium (Element 96) and californium (Element 98) target nuclei. However, unlike for ^{248}Cm, the mass number-yield peak shift for ^{249}Cf is less than two mass numbers for the ^{16}O, ^{18}O pair.

Perhaps even more spectacular are the observations of M. Schädel et al. [56] of such high yields with a target nucleus of even higher atomic number. They bombarded microgram quantities of ^{254}Es with ^{16}O, ^{18}O and ^{22}Ne ions in the Berkeley 88-Inch Cyclotron and the UNILAC at GSI and chemically separated and identified isotopes of fermium (Element 100), mendelevium (Element 101), nobelium (Element 102), and lawrencium (Element 103). (The discovery of the new isotopes, ^{261}Lr and ^{262}Lr, using such a reaction, is described above.) Surprisingly, as with curium and californium target nuclei, they find peak production cross sections for nuclides with $\Delta Z = +1$ and $\Delta Z = +2$, fermium and mendelevium, in the millibarn region. Also, as with the curium target, the peak yields of nuclides with $\Delta Z = +3$ and $\Delta Z = +4$, nobelium and lawrencium, are in the microbarn region. Clearly in these transfer-type reactions product nuclei are formed with relatively low excitation energy and, therefore, are better able to survive loss by nuclear fission. This underscores the value of obtaining a larger supply of ^{254}Es for use as target material!

As was described above, Oganessian [7] came up with the brilliant conception of the reaction mechanism of "cold fusion." This made possible the discovery of elements 107, 108, and 109 by bombardment with heavy ions of target nuclei with closed nucleon shells (^{208}Pb and ^{209}Bi). However, their report in 1974 [7] of the discovery of Element 106, through the observation of a 4-10 ms SF activity produced in the bombardment of $^{207,208}_{82}$Pb by bombardment with $^{54}_{24}$Cr, was shown in 1984 by Demin et al. [57] (Dubna) to be incorrect in that the observed SF activity was mainly due to Element 104 alpha-daughters of Element 106.

The "cold fusion" reaction may be reaching its limits for application to the synthesis and identification of still heavier elements because of the inability of still heavier projectile nuclei to fuse with these target nuclei. W. J. Swiatecki [58] has developed a schematic model that has been widely used to represent this dynamical limitation to fusion, for heavy target nuclei, which increases rapidly with increasing atomic number of the projectile for atomic number greater than 18 (argon); P. Armbruster [4] has developed a semi-empirical representation of fusion probability that is very useful, and W. Nörenberg et al. [59] have developed a Dissipative Diabatic Dynamics (DDD) theory that is applicable to the explanation of the limitation to fusion. Because of this limitation, another approach is now being explored--the use of actinide targets and lighter heavy ions. This is referred to as the "hot fusion" mechanism because of the higher degree of excitation of the compound nucleus, a result of the lack of such strong closed nucleon shells (like $Z = 82$ and $N = 126$) in the actinide region. Examples of unsuccessful attempts to synthesize element 110 using the "cold fusion" reaction mechanism are described above.

Chemical and related properties

Curium (Element 96)

Curium occupies a special position in the transition series of actinide elements. It stands at the midpoint and is characterized by a half-filled 5f electron shell, with the electronic structure: [Rn] $5f^7 6d7s^2$. This gives it rather unique chemical properties in the series with its especially stable tripositive oxidation state.

Recently W. Kot et al. [60] (Berkeley and Oak Ridge), have made an electron paramagnetic resonance (EPR) study of Cm^{3+} in single crystals of $LuPO_4$. Cm^{3+} (electronic configuration [Rn] $5f^7$) is the actinide analog of the lanthanide Gd^{3+} which has the electronic configuration [Xe] $4f^7$. Unlike the other lanthanide ions, Gd^{3+} has a long electronic relaxation time because the ground state is primarily ${}^8S_{7/2}$ and room temperature EPR spectra can be observed. Until now EPR spectra of all actinide ions have been observable only at low temperature.

Kot et al. have observed EPR spectra of Cm^{3+} diluted in single crystals of $LuPO_4$ at room as well as at low temperatures. The total crystal-field splitting of the ground state crystal-field term (which is approximately 80% ${}^8S_{7/2}$) is about 10 cm^{-1}, which is approximately 100 times greater than that found for Gd^{3+}. This enormous difference can be accounted for by the greater spin-orbit coupling constant for Cm^{3+} which mixes higher lying $J = 7/2$ terms into the ground state term. From an analysis of the EPR spectra as a function of temperature, the temperature dependence of the zero field splitting was determined. From the angular dependence of the EPR spectra the crystal field parameters were determined. These crystal field parameters (and the Slater and spin-orbit parameters from Cm^{3+} in $LaCl_3$) were then used to calculate the energy levels, which agree reasonably well with the experimental values.

In summary, the interpretation of the $Cm^{3+}/LuPO_4$ EPR spectra represents a dramatic example of the much greater effect of spin-orbit coupling in the actinide compared to the lanthanide series. In addition it was shown that the crystal field parameters derived from the ground state EPR data do reproduce the general features of the higher energy levels.

L. R. Morss et al. [61, 62, 63] (Argonne National Laboratory and University of Liège), have made the first determination of the enthalpy of formation of Cm_2O_3 (and Am_2O_3 and Cf_2O_3) by measurement of the enthalpy of solution in dilute hydrochloric acid. The enthalpy of solution (for all three oxides) was less exothermic by 5-20 kJ per mol than those of lanthanide sesquioxides with similar unit cells and coordination geometry. Since an exothermic enthalpy of solution reflects a relatively unstable substance, it was concluded that the actinide +3 ions are more stable in oxides (with respect to +3 ions in aqueous solution) than are the corresponding lanthanide +3 ions.

D. E. Hobart et al. [64] (Oak Ridge and University of Tennessee, Knoxville) have measured the Raman spectra of crystalline curium orthophosphate, as well as americium, berkelium, californium, and einsteinium orthophosphates, and of curium, as well as americium and californium trimetaphosphates. With other collaborators Hobart [65] has studied the spectroscopic and redox properties of curium (and californium) ions in concentrated aqueous carbonate-bicarbonate media; no conclusive evidence was found to indicate the existence of any higher than III oxidation states of curium (or californium) in carbonate solution.

Turning to a practical application, the determination of the concentration of Cm^{3+} species in ground water, J. V. Beitz et al. [66] (Argonne National Laboratory), have demonstrated for the first time the optical detection of Cm^{3+} in aqueous solution at the part-per-trillion level using laser-induced fluorescence. The sensitivity achieved corresponds to the detection of as little as 10^8 Cm^{3+} ions in the laser beam, which represents over a hundred-fold improvement over other laser-based methods. The power of the laser-induced fluorescence method arises not only from its sensitivity but also from the more detailed spectroscopic and photophysical characterization of species it can provide by fully exploiting the formally parity forbidden, but characteristic, 5f-5f absorption bands of actinide elements. Thus, the method is applicable to heavier tripositive actinides with the prospect that detailed speciation studies at the level of 10^8 Md^{3+} ions in solution should be possible using picosecond laser excitation and photon counting detection.

F. David and B. Fourest [67] have estimated the hydration number for Cm^{3+} ions in aqueous solution (and all of the trivalent actinide ions).

Berkelium (Element 97), Californium (98), Einsteinium (99)

Significant developments in the chemistry of transcurium elements result from the work of J. R. Peterson and his students and coworkers (University of Tennessee, Knoxville and Oak Ridge). He has carried out much research here in the European Institute for Transuranium Elements in separate collaborations with J. C. Spirlet and U. Benedict, who will talk later in this afternoon's program.

Peterson et al. have used optical absorption spectroscopy (300-1100 nanometers [nm]) to characterize many oxide and oxyhalide compounds using samples typically weighing only a few micrograms. With correlation to crystal structure, he has essentially devised a spectral probe of structure which provides identification of atomic number, oxidation state, and metal ion coordination. He and his coworkers have used these characterization tools to determine the chemical and physical consequences of alpha- and beta-decay in the bulk-phase solid state and to monitor via spectral analysis the effects on selected halide compounds of applying pressure up to 350,000 atmospheres (atm).

In a pioneering experiment Peterson et al. [68] prepared a sample of hexagonal $BkCl_3$ using ^{249}Bk, which decays to ^{249}Cf by the emission of negative beta-particles with a half-life of 320 days. Starting with pure ^{249}Bk they monitored by means of optical absorption spectrophotometry the chemical (oxidation state) and physical (crystal structure) consequences as ^{249}Bk transformed into ^{249}Cf over a period of 976 days. Their measurements showed clearly that the hexagonal $BkCl_3$ transformed into hexagonal $CfCl_3$.

Using these techniques they [69] performed a classic experiment to produce $CfBr_3$ in the previously unknown orthorhombic form. Starting with orthorhombic $BkBr_3$, the ^{249}Bk was allowed to decay for more than three years (converting it to more than 90% ^{249}Cf), leading to the production of orthorhombic $CfBr_3$, as confirmed by observing its x-ray diffraction pattern. After heating to 360°C this was converted to the usual monoclinic structure, as shown both by the optical absorption spectrophotometry and x-ray diffraction measurements.

Then, using the experimental techniques he learned here in the Transuranium Institute at Karlsruhe during his sabbatical year, he employed a diamond anvil cell (DAC) to apply pressure (1.7-3.4 GPa) to the monoclinic form of $CfBr_3$ to transform it to the orthorhombic form [70]. Thus, he accomplished in a single day what had required three years in the ^{249}Bk beta-decay process!

Peterson et al. [71] have also used the absorption spectra technique to characterize the dipositive oxidation state in the identification of $EsCl_2$, $EsBr_2$, and EsI_2.

Most recently they [72] have developed the use of phonon Raman spectroscopy as a spectral probe of crystal structure for transplutonium elements. As examples, they have used this technique at room temperature to characterize $AlCl_3$-type monoclinic $CfBr_3$, orthorhombic $CfCl_3$ and hexagonal (UCl_3-type) $CfCl_3$, $CmCl_3$, and $AmCl_3$. This technique offers some advantages over x-ray diffraction because the high specific activity of these elements can cause rapid and extensive damage to the long-range order in the solid, which significantly degrades the quality of the diffraction patterns that can be obtained. A Raman spectrum can be obtained in a relatively short time and immediately following the annealing treatment of the sample. In contrast, the three to six or more hours required to obtain an x-ray diffraction powder pattern is often long enough for significant self-irradiation damage to occur.

G. M. Jursich et al. [73] (Argonne National Laboratory), have observed the laser-induced fluorescence-spectrum of Bk^{4+} doped (0.1-0.01%) into CeF_4, which provides a basis for developing considerable new insight into the interpretation of f-element spectra. It also yields a clear example of the extent of the crystal-field splitting in the actinide tetrafluorides, which form an isostructural series from ThF_4 through CfF_4. In BkF_4, the trend in increased splitting and mixing of states culminates in a clear complete splitting pattern for the ground state. As it turns out, this BkF_4 spectrum can provide a very important basis for testing the understanding of the mechanisms that drive the ground state splitting in the special case of the f^7-configuration. In addition, the complete splitting pattern can be interpreted as defining one of the crystal-field parameters that measure the crystal-field splitting in the isostructural series of compounds $An(Ce)F_4$. Since their interpretation of the spectroscopic measurements for $Bk^{4+}:CeF_4$, coupled with new spectroscopic studies of the lighter actinide tetrafluorides,

UF_4, NpF_4, and CmF_4, is providing the basis for the first sytematic interpretation of An^{4+} spectra over the whole series [74], this type of investigation encourages them to predict the electronic energy level structure in heavier actinides. Their current measurements of the spectrum of $Cf^{4+}:CeF_4$ are consistent with their predictions. They are attempting to prepare and make spectroscopic measurements on $Es^{4+}:CeF_4$ and even hope to study the spectrum of FmF_4, if the compound can be prepared.

J. C. Sullivan et al. [75] (Argonne National Laboratory) have used the powerful technique of pulse radiolysis to prepare Bk(II) in aqueous solution for the first time. (This technique had been used earlier by Argonne scientists to provide evidence for the existence of divalent oxidation states of americium, curium, and californium as transient species.) They demonstrated that the hydrated electron reacts preferentially with a 0.002 M solution of Bk(III) in aqueous perchlorate solution at pH 5 to reduce the Bk(III) to a transient (half-life 4.5 microseconds) and very unstable Bk(II). They determined the absorption spectrum of the Bk(II) with the Argonne streak camera system and found this new absorption band has a maximum at 310 nm.

Hobart et al. [76] (Los Alamos, Oak Ridge, and University of Tennessee, Knoxville) have determined the spectroscopic and redox properties of berkelium in complexing aqueous carbonate and citrate solutions. The spectra were compared with those of berkelium in noncomplexing perchloric acid. They also prepared a new, solid, hydroxy-carbonato-Bk (III) complex.

D. E. Morris et al. [77] (Los Alamos, Oak Ridge, and University of Tennessee, Knoxville) made a voltametric investigation of the berkelium (IV/III) couple in concentrated aqueous carbonate solution. Direct evidence for the presence of an equilibrium between two (or more) Bk (IV) species with differing reduction potentials was found for several solution conditions.

Fermium (Element 100), Mendelevium (101), Nobelium (102), Lawrencium (103)

The enthalpy of vaporization of fermium metal, which is a measure of its cohesive energy, has been measured for the first time by R. G. Haire and J. K. Gibson [78] (Oak Ridge). From the value of the cohesive energy of fermium (enthalpy of sublimation, 32-35 kcal/mole) it can be deduced that fermium is a divalent metal, i.e., that two of a fermium atom's electrons are involved in the metal's conduction band.

The cohesive energy of fermium was determined by measuring the vapor pressure of 20.1-hour ^{255}Fm (produced in the HFIR) over fermium-samarium and fermium-ytterbium alloys (with nanogram amounts of fermium, 10^{-7}-10^{-8} atom % fermium) as a function of temperature (100-600°C). It was found that the cohesive energy for fermium was not influenced by the nature of the solvent; the same value was obtained when the solvent was divalent ytterbium metal or when it was trivalent samarium metal. Further, alloys containing both einsteinium (20.5-day ^{253}Es) and fermium at comparable concentrations were studied so that the einsteinium could serve as an internal standard. The results of these experiments have shown that fermium is the most volatile of the actinide metals that have been studied.

Demonstrating that both einsteinium and fermium are "divalent" metals, whereas all of the earlier actinide metals provide three or more electrons to a conduction band, establishes a "trend of divalence" for the heavier members of the series. This is a pivotal finding that cannot be deduced from a comparison of the expected electronic configurations of the actinide (5f series) and lanthanide (4f series) elements. The lanthanide homologs of einsteinium and fermium are both trivalent metals. The trend in the actinides must, therefore, reflect an increasing influence of the relativistic electrons in these heavier atoms.

The measurement of the cohesive energy of fermium, the derivation of other thermodynamic functions from this cohesive energy, and the deduction of the metallic nature of fermium all strengthen our basic understanding of these heavier elements and their relationship to other elements in the Periodic Table. Such information, derived experimentally, provides a framework for testing existing theories, for developing new theoretical interpretations, and for making extrapolations to heavier elements of the Periodic Table.

W. Brüchle et al. [79] (GSI, Mainz, Berkeley), using a mixture of mendelevium isotopes (27-minute ^{255}Md and 1.3-hour ^{256}Md) produced by the bombardment of ^{249}Bk with ^{18}O ions in the Berkeley 88-Inch Cyclotron, determined the elution position of mendelevium, eluted from a cation exchange column with ammonium alpha-hydroxyisobutyrate in relation to radioactive lanthanide tracers of known ionic radius. Their measurements were aided by the use of an Automated Rapid Chemistry Apparatus (ARCA) [80]. From the elution position the ionic radius was calculated for Md^{3+} (0.0896±0.0001 nm) which, with the use of semi-empirical models, allowed the calculation of its heat of hydration as -3654 kJ/mole.

G. F. Payne et al. [81] (Oak Ridge), have started an experimental program to elucidate the status of Md^{1+} (which might be stabilized by the $5f^{14}$ closed electron shell). Since Md^{1+} cannot be made in protic solutions they have investigated conditions under which it might be made in aprotic solutions. They believe that dimethyl sulfoxide (DMSO) might be a suitable solvent in which to attempt a reduction of mendelevium to the +1 oxidation state.

D. C. Hoffman et al. [82] (Berkeley, Beijing, Livermore), were the first to make an extensive study of the chemical properties of lawrencium, employing manual batch experiments in which they used the 3-minute ^{260}Lr, produced in the bombardment of ^{249}Bk with ^{18}O ions in the Berkeley 88-Inch Cyclotron. Their experiments involved extractions with thenoyl trifluoroacetate (TTA) (to confirm the trivalency of ^{260}Lr), ammonium alpha-hydroxyisobutyrate elution from a cation resin relative to the tripositive rare earths and other tripositive actinides for which the ionic radii are known (in order to determine the ionic radius of Lr^{3+}), and reduction experiments with reverse-phase chromatography using hydrogen di(2-ethylhexyl)orthphosphoric acid (HDEHP) (to look for a lower oxidation state). In these preliminary experiments they found that the ionic radius of Lr^{3+} is very close to that of Er^{3+}, with the value 0.0886±0.0003 nm, which is surprisingly close to that of Md^{3+} (0.0894 nm by their determination). This difference of only 0.0008 nm between the ionic radius of Lr^{3+} and Md^{3+}, which differ in atomic number by two units, is comparable with the usual difference between the ionic radii of adjacent lanthanide or actinide elements. So far they have found no evidence for an oxidation state lower than +3. (L. Brewer [83] had predicted that the electronic structure of lawrencium might be [Rn] $5f^{14}7s^27p^1$, with the possibility that the $7p_{1/2}$ electron could be removed selectively, resulting in a +1 oxidation state.) U. W. Scherer et al. [84] (Mainz, GSI, Berkeley), in similar but more extensive experiments at Berkeley using the ARCA system, also find no evidence for the reduction of Lr^{3+}. They used reverse-phase chromatography with HDEHP and vanadous (V^{2+}) and chromous (Cr^{2+}) ions as reductants in dilute hydrochloric acid. In the same experiments they succeeded in reducing Md^{3+} to Md^{2+} ($E° = -0.2$ V). Thus, the reduction potential for reduction of Lr^{3+} to the divalent or monovalent state is more negative than -0.44 V. Hulet [85] has proposed a Stern-Gerlach experiment to directly determine the ground state configuration of lawrencium.

In very careful, extensive experiments, W. Brüchle et al. [79] (GSI, Mainz, Berkeley) used the 3-minute ^{260}Lr at Berkeley with the ARCA system to determine more precisely the ionic radius and hydration energy of Lr^{3+}, with the result of 0.0881±0.0001 nm for the ionic radius and -3685 kJ/mol for the heat of hydration. Thus, they find a larger value, 0.0015 nm, for the difference between the ionic radii of Lr^{3+} and Md^{3+}, which is still unusually small of a change in atomic number of two units. It would be very worthwhile to try to determine the ionic radius of the intermediate No^{3+}.

D. T. Jost et al. [86] (Würenlingen, GSI, Rossendorf, Berkeley) have investigated further the proposition that lawrencium might have the electronic structure [Rn] $5f^{14}7s^{2}7p_{1/2}$, rather than the traditionally-expected [Rn] $5f^{14}6d7s^{2}$ configuration. They also used the 3-minute ^{260}Lr produced in the ^{249}Bk (^{18}O, α3n) reaction in the Berkeley 88-Inch Cyclotron. They thought that with a $7p_{1/2}$ electron in the ground state, this would indicate a high volatility in the elemental state, similar to other p-elements such as thallium, as suggested by B. Eichler et al. [87]. They, therefore, performed on-line gas chromatography experiments in quartz and metal columns but found no evidence for lawrencium as a volatile element under reducing conditions and at a temperature of about 1000°C. S. Hübener and I. Zvara [88] (Dubna) have performed similar experiments on californium, einsteinium, fermium, and mendelevium, and Hübener et al. [89] on nobelium.

Rutherfordium (Element 104), Hahnium (105)

With the completion of the actinide (5f) elements at lawrencium (103), the transactinide elements, with the filling of the 6d electron shell, exhibit a significant change in chemical properties. Thus, these elements find their places back in the main body of the Periodic Table (Figure 1), with Element 104 having the characteristics of eka-hafnium, Element 105 of eka-tantalum, etc. These chemical characteristics have been confirmed in the experiments conducted to date.

In experiments performed in 1980 Hulet et al. [90] extended earlier investigations of the solution chemistry of rutherfordium using the 1-minute ^{261}Rf produced in the Berkeley 88-Inch Cyclotron. A hydrochloric acid solution containing the ^{261}Rf was passed through an extraction chromatography column loaded with trioctylmethylammonium chloride which strongly extracts anionic chloride complexes. Rutherfordium was shown to behave like hafnium, due to the formation of strong complexes, and differently than the actinides, which form weaker complexes.

B. L. Zhujkov et al. [91] (Dubna) used the 3.2-second ^{259}Rf (SF branching decay) to perform on-line gas chromatography experiments to test whether rutherfordium in its elemental state under reducing conditions exhibits the volatility expected of an element with $7p_{1/3}$ electrons in the ground state; as reported above for lawrencium no evidence for such volatility was found.

In 1987, K. E. Gregorich et al. [92] (Berkeley, Mainz), performed remarkable first experiments on the aqueous phase chemistry of hahnium. They were able to use the short-lived 34-second ^{262}Ha, produced by the ^{249}Bk (^{18}O, 5n) reaction in the Berkeley 88-Inch Cyclotron with the tiny yield of one atom per 30 bombardments, by conducting nearly 1000 batch experiments. The identification of 26 atoms of ^{262}Ha used in the chemical experiments was made on the basis of observation of decay by spontaneous fission and alpha-emission, including, in a number of instances, the time correlation of alpha-decays from ^{262}Ha and its 4-second daughter ^{258}Lr. Similar experiments were performed with radioactive isotopes of niobium, tantalum, zirconium, and hafnium in order to compare the chemical behavior of hahnium with that of these potentially homologous or analogous elements under identical conditions.

Hahnium, like niobium and tantalum, was found to adhere to glass surfaces upon fuming with concentrated nitric acid. Thus, by analogy with niobium and tantalum, this indicates that the most stable oxidation state for hahnium is +5 (i.e., Ha^{5+}). There is a type of reasoning whereby a relatively stable Ha^{3+} might exist; because the expected ground state electronic configuration of hahnium is [Rn] $5f^{14}6d^{3}7s^{2}$, the relativistic stabilization of the 7s electrons might be strong enough to hinder their removal, leading to Ha^{3+} with the [Rn] $5f^{14}7s^{2}$ configuration. The experimental results argue against this.

In other experiments, hahnium did not extract from mixed nitric acid/hydrofluoric acid solutions into methyl isobutyl ketone as does tantalum. Perhaps this non-tantalum behavior may be explained by extrapolation of group 5 chemical properties. The tendency to hydrolyze or to form high coordination number fluoride complexes in hahnium may be much stronger than in tantalum, leading to *non*-extractable species.

In September 1988, an international collaboration to study the chemical properties of hahnium was underway at Berkeley, again using the 34-second ^{262}Ha produced in the 88-Inch Cyclotron. Radiochemistry groups from the Institut für Kernchemie of the University of Mainz and GSI in West Germany, and the Paul Scherrer Institute and the University of Bern in Switzerland were collaborating with the LBL heavy element nuclear and radiochemistry groups. In order to explore further the unexpected differences between hahnium and tantalum in their halide complexing behavior, anion exchange studies were being carried out in mixed HCl or mixed HCl-HF and HNO_3-HF systems, using a miniaturized computer-controlled chromatographic column apparatus (mini-ARCA) which has been designed by the German groups specifically for performing repeated chemical separations on a one-minute time scale. The behavior of hahnium was being compared to that of the group 4 and group 5 elements, zirconium, niobium, hafnium, and tantalum, and to that of the pseudo-group 5 element, protactinium. Gas phase chemical separations were also being performed using an on-line isothermal gas chromatography apparatus developed by the Swiss groups. In these experiments measurements were being made of the retention times of volatile bromide complexes in quartz columns which should be formed when hahnium atoms come into contact with HBr gas at different temperatures. The behavior of hahnium was being compared with that of the group 4 and 5 elements, zirconium, niobium, hafnium, and tantalum; the heat of adsorption on quartz will be deduced from the retention times.

Reflections

As this review shows, serious research on the transcurium elements is, with some exceptions, performed by scientists working at, or with connections to, large laboratories with extensive facilities--in the United States, the national laboratories (Argonne National Laboratory, Lawrence Berkeley Laboratory, Lawrence Livermore National Laboratory, Los Alamos National Laboratory, Oak Ridge National Laboratory); in Europe, the Gesellschaft für Schwerionenforschung (GSI) in the Federal Republic of Germany, and the international laboratories, the Joint Institute for Nuclear Research, Dubna, USSR, and the European Institute for Transuranium Elements, here in Karlsruhe. (There are, of course, other laboratories that are making important contributions--this review, perhaps already too long, is necessarily selective in its survey of the recent research.) However, the potential of the transuranium field is so large that there is a need for even more specialized facilities. It is the author's dream that in the future--perhaps the distant future--additional laboratories or institutes will be created for exclusive research on the transuranium elements.

There are almost unlimited possibilities for research on these elements, which already constitute almost 20% of the total of all known chemical elements. When thinking in terms of the distant future the tendency is to underestimate potential accomplishments.

Estimates indicate that 400 transcurium nuclides would have half-lives sufficiently long to be detectable experimentally (longer than a microsecond). The synthesis and identification of another half dozen or so elements seems likely; this would include the discovery of Superheavy Elements and the extension of the present peninsula of elements to connect with the Island of Stability. Longer-lived isotopes than those now known will probably be found in the transactinide region, especially among the early transactinide elements. (As an example, the recently discovered longer-lived isotopes of lawrencium [^{261}Lr and ^{262}Lr] will make possible the detailed study of the chemical properties of this element.) As a result, it should be possible to study the chemical properties of elements beyond hahnium (Element 105), and certainly of Element 106 (already possible using the 0.9-second ^{263}106).

Much more research on the macroscopic properties of einsteinium will be possible with the availability of ^{254}Es. It will surely be possible to study the macroscopic properties of fermium and not out of the question that this will be done for mendelevium. The art of one-atom-at-a-time chemistry will advance far beyond what can be imagined today to make it possible to study the chemistry of heavier and heavier elements. All of this will result in the delineation of relativistic effects on the chemical properties of these very heavy elements, which might thus be substantially different than those expected by simple extrapolation from their lighter homologs in the Periodic Table.

And in the course of the preparation of this broad range of nuclides by heavy ion reactions and the study of their decay properties much will be learned about the dynamics of nuclear matter, the exact location of shell structure, and the energy levels and spectroscopic states of heavy nuclei. This will give the theorists information to further increase the understanding of nuclear forces and structure.

Such a research program will require, for success, the availability of apparatus and equipment of increasing complexity, versatility, and power. Central will be the need for higher neutron flux reactors, for sustained operation as a research tool and to produce large quantities of transplutonium nuclides for use in the research and as target materials as a source of the presently known and expected nuclides. (Higher neutron fluxes will be especially valuable for the production of the heaviest nuclides, ^{254}Es and ^{257}Fm, springboards to the region beyond.) Higher intensity heavy ion accelerators must be built and means of coping with their beams at the target must be developed in order to overcome limitations due to small nuclear reaction cross sections. Increase in orders of magnitude in heavy ion intensity should make possible nuclear synthesis reactions with secondary (radioactive) beams of neutron-excessive projectiles, which might greatly increase the yields of sought-after new nuclides. Improved methods for handling safely and efficiently and making chemical measurements on increasing quantities of the highly radioactive transcurium nuclides must be developed.

Improved apparatus of all kinds for the determination of chemical structure, energy levels and their electronic structure, thermodynamic data, etc., improved laser beams, and the use of new apparatus, such as the Advanced Light Source (ALS) being built at the Lawrence Berkeley Laboratory, will place new power in the hands of the chemist. The ultimate achievement will be the perfection of means of performing single atom chemistry.

Acknowledgement

I wish to express my appreciation to many friends and colleagues for furnishing information and for helpful discussions--W. T. Carnall, N. M. Edelstein, H. Gäggeler, A. Ghiorso, K. E. Gregorich, R. G. Haire, D. E. Hobart, D. C. Hoffman, E. K. Hulet, O. L. Keller, Jr., J. V. Kratz, W. Loveland, P. Möller, L. R. Morss, J. M. Nitschke, W. Nörenberg, J. R. Peterson, L. P. Somerville, and W. J. Swiatecki.

This work was supported by the Director, Office of Energy Research, Office of High Energy and Nuclear Physics, Nuclear Physics Division of the U.S. Department of Energy under Contract DE-AC03-76SF00098.

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Figure Captions
for
Recent Research on the Heavy Transuranium Elements

Fig. 1: Modern Periodic Table of the Elements (atomic numbers of undiscovered elements are shown in parentheses).

Fig. 2: Plot of SF half-life vs. atomic number. Solid line shows predictions from liquid drop model; ● designates experimental data.

Fig. 3: Chart of curium and transcurium nuclides (furnished by E. K. Hulet).

1 H																	2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	57 La	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
87 Fr	88 Ra	89 Ac	104 Rf	105 Ha	106	107	108	109	(110)	(111)	(112)	(113)	(114)	(115)	(116)	(117)	(118)

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

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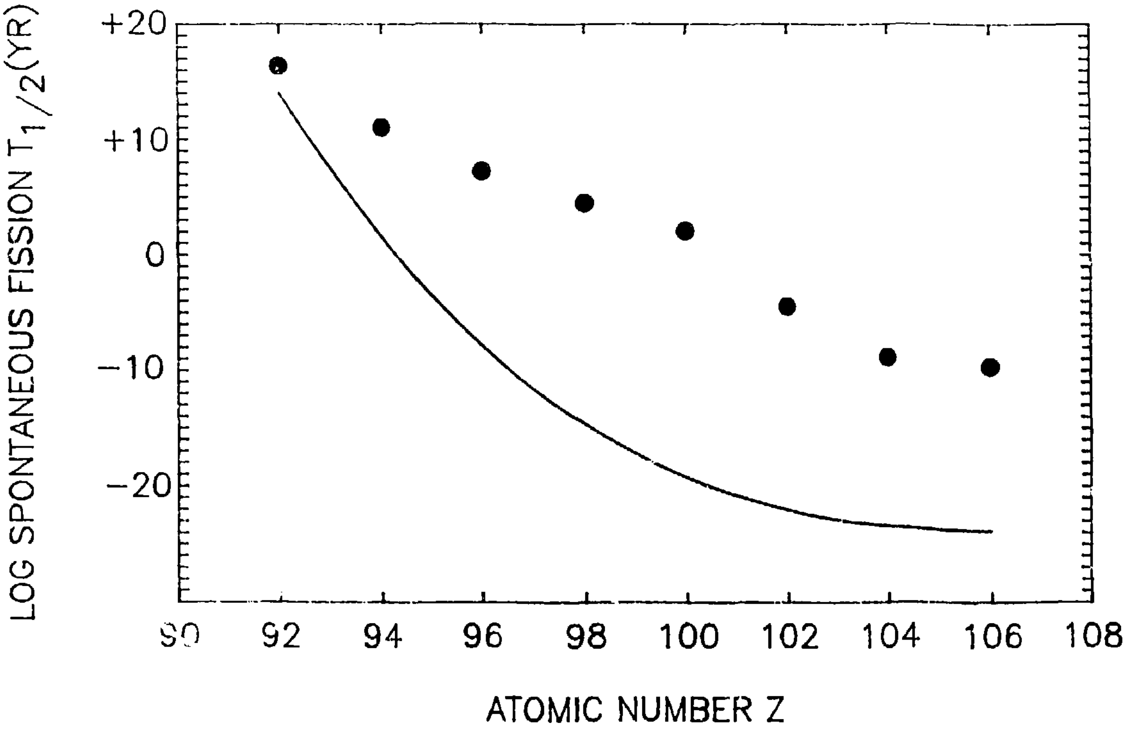
Figure 1: Modern Periodic Table of the Elements (atomic numbers of undiscovered elements are shown in parentheses).



XBL 889-3265

Figure 2: Plot of SF half-life vs. atomic number. Solid line shows predictions from liquid drop model; ● designates experimental data.

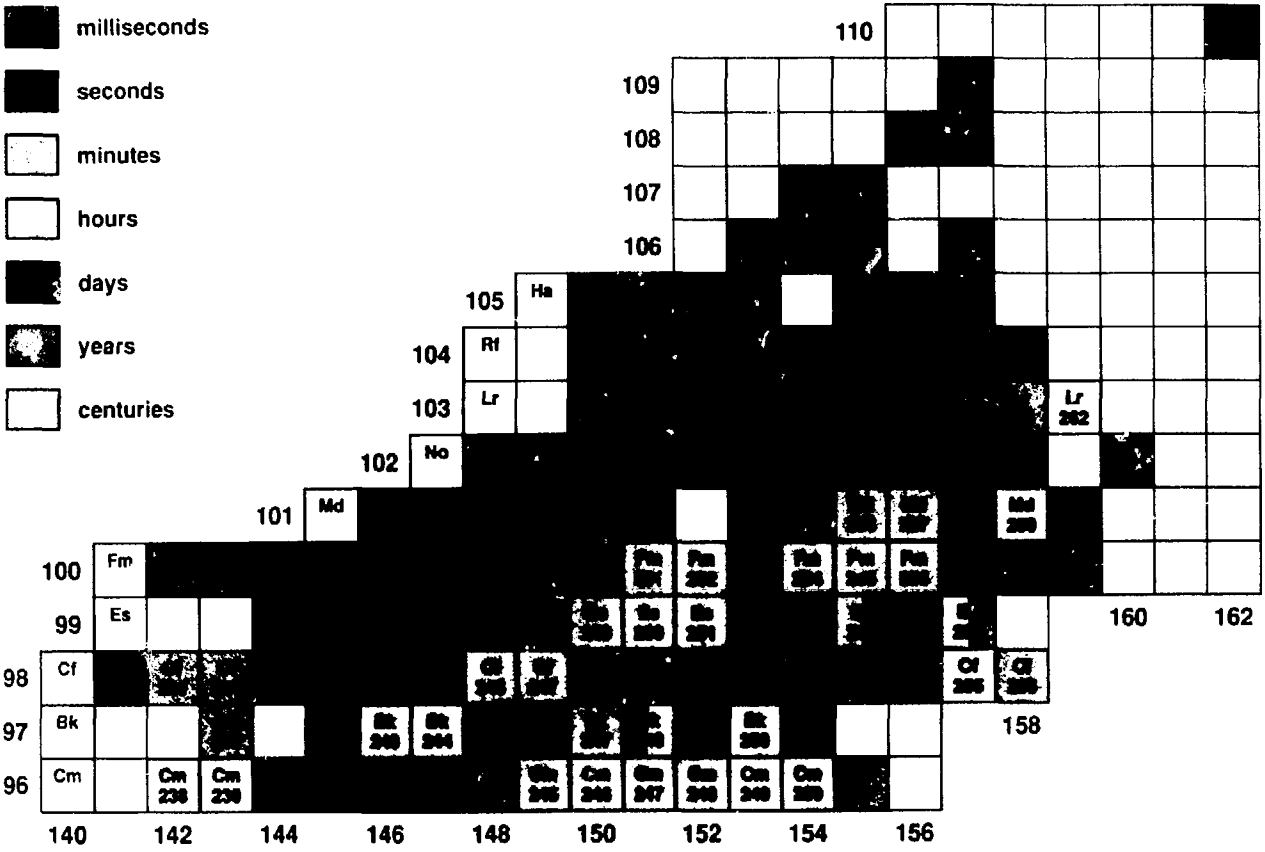


Chart of the Transcurium Nuclides

Figure 3: Chart of curium and transcurium nuclides
(Chart furnished by E.K. Hulet)