RECENT WORK WITH THE TRANSURANIUM ELEMENTS

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In this discussion of the heaviest known elements, I intend to describe some recent research which appears to offer the most promise for future investigations. As a general background, I shall review briefly the history and properties of the transuranium elements.

The first transuranium element to be discovered, neptunium with atomic number 93, was identified by E. M. McMillan and P. H. Abelson at the University of California in Berkeley in 1940. Plutonium, element 94, was discovered the following year. Soon after, the impetus of wartime work resulted in the intensive investigation of plutonium and the discoveries of americium (element 95) and curium (element 96); and, in the postwar years, berkeliun (element 97), californium (element 98), einsteinium (element 99), fermium (element 100), mendeleevium (element 101), and element 102 have been prepared and identified. For all practical purposes, these elements are synthetic in origin; in fact, all of them have been discovered as the transmutation products of other elements, and our current supplies of the transuranium elements have been prepared in this way. Traces of neptunium and plutonium, however, do occur in nature in uranium ores, being produced by the action of the neutrons that are present. All of the known transuranium elements have been found to belong to a rare-earth-like series of elements which begins with actinium and presumably ends with the undiscovered element 103. This series, called the actinide series, is similar in chemical properties to the lanthanide or rare-earth series. The position of the actinide and lanthanide elements in the periodic table is shown in Fig. 1. As in the rest of the periodic table, the elements in corresponding vertical columns are
chemical homologues; thus the homologue of the newly discovered element 102 is ytterbium; that of mendelevium, terbium; etc. This chemical resemblance between corresponding members of the two series, particularly with respect to their ion-exchange behavior, has been the key to the discovery of most of the transuranium elements.

One of the recent accomplishments in the field of the transuranium elements has been the determination of the electronic structure of the ground state of curium. The electronic structures of the ground states of all the elements in the first half of the actinide series are now known and constitute some of the best presently available evidence for the actinide concept, i.e., that the elements from actinium through element 103 are members of a second rare-earth group in which the 5f electron shell is being filled. The similarity between the electronic structures of corresponding members of the actinide and lanthanide series can be seen in Table 1. Here the known electronic structures for all the lanthanide elements and for the actinide elements through curium are given. Also included are the predicted, but not yet determined, electronic configurations for the transcurium elements.

The electronic configurations of curium and most of the preceding actinide elements were determined by studies of the hyperfine structures of free atoms using the atomic beam technique. In combination with chemical, paramagnetic, optical, and crystal structure data, these investigations have made it possible to assign ground state electronic configurations. The atomic beam technique is adaptable to the use of tracer amounts and thus offers much promise for the determination of the electronic structures of elements not available in weighable or macroscopic amounts, presumably including at least some of the transcurium elements. The basic elements of the method
Table 1. Electronic Structures of Actinide and Lanthanide Elements

(Predicted structures are given for the transcurium elements.)

<table>
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<td>6d(^2)7s(^2)</td>
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<tr>
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or

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<td>71 Lutetium</td>
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are as follows: A crucible is filled with the material under investigation and heated to 1000 - 3000° C to produce a beam of neutral atoms which passes through a system of collimating and analyzing slits and three magnets, two of which produce deflecting fields. The system is arranged so that normally no atoms can reach the detector. If, however, an appropriate transition between magnetic substates in the atoms is induced in the homogeneous field produced by the third magnet, a portion of the beam can reach a detector. One can thus study the hyperfine structure of free atoms, which is a function of both electronic and nuclear structure, by this experimental method. The hyperfine structure interaction involves predominantly the interaction of the nuclear magnetic dipole moment with the magnetic field at the nucleus arising from valence or unpaired electrons. The principal electronic information derived is the electronic angular momentum and Lande g-factor of all electronic states within several thousand cm⁻¹ of the ground state.

Increasing amounts of the heavier transuranium elements are becoming available, being prepared by the neutron irradiation of plutonium or higher elements for long periods of time in a high flux reactor such as the Materials Testing Reactor in Arco, Idaho. Just this year, S. G. Thompson and B. B. Cunningham succeeded in isolating for the first time weighable amounts of berkelium and californium which had been synthesized by the approximately five-year neutron irradiation of about eight grams of Pu²³⁹ and its transmutation products. The reaction paths for this method of preparation are shown in Fig. 2. Prior to these isolations, studies of berkelium and californium were made with tracer amounts of material, and enough was known from such investigations to permit isolation of the two elements from the
plutonium and transmutation products present, the final concentration and purification being accomplished by the ion-exchange technique.

Ion-exchange separations are performed by a method which can be outlined briefly as follows. The solid ion-exchange resin (an organic polymer with, in this example, exchangeable cations) is stirred into an aqueous solution containing trispositive actinide or lanthanide ions which replace the exchangeable cations of the resin. (Similar separations can, of course, be made using a polymer with exchangeable anions, in which case, the actinide or lanthanide elements must be present in the initial solution as complex ions.) This solid resin is then usually placed at the top of a glass column filled with untreated resin. A solution containing anions that form complex ions with the actinide or lanthanide, the eluant, is poured through the column to effect their removal. The drops of eluant are collected and analyzed from their radioactive properties. Solutions of chloride, nitrate, lactate, citrate, or alpha-hydroxyisobutyrate, for example, can be used as the eluant. The adsorption and elution reactions can be written as follows:

\[
\begin{align*}
M^{+3}(aq) + 3 \text{NH}_4^+(s) & \rightarrow MR_3(s) + 3 \text{NH}_4^+(aq) & \text{ADSORPTION STEP} \\
M^{+3}(aq) + 4 A^-(aq) & \rightarrow MA_4^-(aq) & \text{ELUTION STEP,}
\end{align*}
\]

where \(M\) represents the actinide or lanthanide ion; \(\text{NH}_4^+\), the ion-exchange resin, ammonium ions being the exchangeable cation in this case; and \(A^-\), the eluant. The actual formula for the complex ion (given here as \(MA_4^-\)) or ions may be more complicated and may not be known. These two reactions, of course, occur many times during the passage of the eluant down the column. In certain systems the actinide and lanthanide elements are eluted in the inverse order of their atomic numbers, as is illustrated in Fig. 3.
Thompson and Cunningham isolated 0.23 microgram of berkelium (as Bk$^{249}$) and 0.06 microgram of californium (as a mixture of the isotopes Cf$^{249}$, Cf$^{250}$, Cf$^{251}$, and Cf$^{252}$) and attempted to measure two macroscopic properties of the elements, the absorption spectrum and the magnetic susceptibility of their trivalent ions. The data obtained for the absorption spectra of Cf$^{+3}$ (and Dy$^{+3}$ for purpose of comparison) in dilute hydrochloric acid are summarized in Fig. 4. Visual observation failed to reveal absorption for either ion in the region from about 4600 to 7400 Å, but a photographic method showed that broad and quite faint absorption bands occur in californium around 7800 and 8300 Å. Rough estimates indicate that the absorption intensities are much weaker than those observed in the lighter actinides and, in fact, have intensities comparable to those found typically in lanthanide ions. This indicates that as the 5f electron shell becomes more deeply embedded in the second half of the actinide series the elements begin to resemble the lanthanide elements even more. The magnetic susceptibilities of the trivalent ions of berkelium and californium were measured over the temperature range 77 to 298° K. In Fig. 5 the data obtained for californium (and gadolinium for comparison) are shown. Results obtained for both berkelium and californium show a close agreement with the predicted values as may be seen from Fig. 6. In this figure are plotted the magnetic moments, determined by experiment and by theory, for a number of actinide elements. The data are consistent only with the filling of the 5f electronic shell in the actinide elements. A plot of the magnetic moments of the lanthanide elements falls almost exactly on the theoretical curve shown.

Einsteinium is the only remaining transuranium element capable of being isolated in macroscopic amount but not yet so isolated. The isotopes with
the longest half lives are the 280-day isotope $^{254}$E and the 20-day isotope $^{253}$E.

We do not expect to find isotopes of fermium and higher elements with sufficiently long half lives to permit their isolation in macroscopic quantity, although new techniques permitting special measurements, such as measurement of spectroscopic properties of gaseous atoms, may be developed.

As successive elements have been discovered, known techniques for their preparation and identification have been refined and new ones devised for application to the investigation of the next new element. Einsteinium and fermium were not, however, discovered as the result of such careful preliminary planning. Their discoveries were the unexpected results of routine chemical identifications of fallout materials from the first test thermonuclear explosion, the "Mike" shot which was staged in the Pacific Ocean area on November 1, 1952. It is possible that if these materials had been examined very soon after the explosion elements of atomic number greater than 100 would also have been found. But owing to the short half lives predicted for these, any such elements which were present would have disappeared before the chemical investigations were completed. Thermonuclear explosions do offer good possibilities for producing new heavy isotopes of known elements and perhaps even new elements if the products can be recovered in better yield than has yet been possible.

The occurrence of heavy isotopes such as the previously unknown $^{244}$Pu and $^{246}$Pu in the explosion debris suggested that very heavy uranium isotopes had been formed in the explosion by the irradiation of the uranium in the device by the extremely high neutron flux that existed for microseconds. Neutron capture occurred during this very small fraction of a second, and since beta decay, will not, in general, proceed with a half life shorter than about 0.1 second, there was no opportunity for beta decay to occur during the
synthesis. The nuclides found in the explosion debris were formed by the subsequent beta decay of the heavy uranium isotopes. Fig. 7 shows the reaction paths by which these heavy elements were produced. Thus, for example, the preparation of Fm\(^{255}\) was via the synthesis of \(\text{U}^{255}\) by neutron capture in \(\text{U}^{238}\), followed by a long chain of short-lived beta decays,

\[
\text{U}^{255} \rightarrow \text{Np}^{255} \rightarrow \text{Pu}^{255} \rightarrow \ldots \rightarrow \text{Fm}^{255},
\]

all of which occurred after the neutron capture reactions were completed.

The chemical investigations were made first using fall-out substances collected on filter papers attached to drone airplanes which flew through the explosion area. Preliminary chemical separations by the ion-exchange method immediately demonstrated the presence of a new element. Later experiments of the same type resulted in the positive identification of both einsteinium and fermium.\(^3\) In order to secure larger amounts of source material, it was necessary to gather up many hundreds of pounds of coral from one of the atolls adjoining the explosion area. Eventually such material was processed by the ton, the pilot plant for this going under the optimistic name of "Paydirt."

The fact that mendeleevium was successfully identified as a transmutation product of a target consisting of only about one billion atoms was due in large part to a new, efficient separation method devised for these experiments. This new method, the recoil technique, was later applied in work leading to the discovery of element 102 and presumably can be used in future work on new elements.

Plans for the preparation of mendeleevium required the bombardment of the maximum available quantity of einsteinium in the form of \(\text{E}^{253}\) with helium ions in the 60-inch cyclotron at the University of California in Berkeley.
Only about $10^{-2}$ atoms of $^{253}\text{Es}$ were available as a result of its production in the Materials Testing Reactor at Arco, Idaho as illustrated in Fig. 2. The number of atoms of mendelevium, N, expected in each bombardment was calculated by the formula $N \approx N'\sigma t$, where $N'$ is the number of einsteinium atoms used as a target; $\sigma$, the cross section or probability for the reaction; $I$, the intensity of the helium-ion beam; and $t$, the effective time of the bombardment. The cross section for the reaction was predicted to be about $10^{-27}$ square centimeters on the basis of the known values for previously observed similar reactions. As a result of some fundamental changes in the 60-inch cyclotron the intensity of the helium-ion beam was increased to $10^{14}$ particles per second per square centimeter. Predictions indicated a half life of the order of hours for the expected isotopes, so an effective value for $t$ of $10^4$ seconds was used. Substituting these numbers in the equation we obtain: $N \approx (10^9)(10^{-27})(10^{14})(10^4) \approx$ one atom per experiment! In addition, there was the formidable task of separating this one atom from the target material and effecting its chemical identification.

A preliminary separation of the atoms of mendelevium from the einsteinium in the target was accomplished with the new recoil technique, which is illustrated in Fig. 8. The target was prepared by plating the einsteinium as a very thin layer onto a gold foil. A beam of helium ions was sent through the back of the foil so that the atoms of element 101, recoiling due to the momentum of the projectiles, could be caught on a second gold foil, the "catcher" foil. This second foil was dissolved and the chemical separations performed. Chemical identification was accomplished in an ion-exchange experiment involving the combined products from three
successive bombardments in which a total of five spontaneous fission counts, and hence five atoms, was observed in the element 101 elution position. The first mendelevium isotope discovered has the mass number 256 and decays by electron capture with a half life of about an hour to Fm$^{256}$ which is responsible for the spontaneous fission decay. The fact that the radioactive decay involved the spontaneous fission process was a fortunate circumstance, since this mode of decay has a high, almost 100 per cent, detection efficiency and zero background. In later confirmatory experiments using larger amounts of Fm$^{253}$ in the target, hundreds of atoms of mendelevium per experiment have been prepared.

A second isotope of mendelevium with mass number 255 has recently been prepared and identified by a group at the Lawrence Radiation Laboratory. It was prepared by the helium-ion bombardment of Fm$^{253}$ and was separated in much the same way as the first isotope of mendelevium to be discovered. The new isotope has a half life of about a half hour and decays by electron capture.

The most recent research on the transuranium elements includes the synthesis and identification of an isotope of element 102.

In 1957, a team of scientists from Argonne National Laboratory in the United States, the Atomic Energy Research Establishment in England, and the Nobel Institute for Physics in Sweden announced the discovery of an isotope of element 102 as a result of research performed at the Nobel Institute. The isotope was reportedly produced by bombarding Cm$^{244}$ with cyclotron-produced C$^{13}(+4)$ ions and reported as decaying by the emission of 8.5 Mev alpha particles with a half life of about ten minutes. The name nobelium was suggested by this group for element 102. Unfortunately
neither experiments using the facilities at the University of California in Berkeley nor related experiments performed in the U.S.S.R. have confirmed this Stockholm work.\textsuperscript{7,8}

In 1958, a group at the University of California reported the positive identification of the isotope $^{102}\text{Cm}^{254}$ as a product of the bombardment of $^{246}$Cm with $^{12}$C ions accelerated in the new heavy ion linear accelerator (Hilac) at Berkeley.\textsuperscript{9} The new element was detected by the chemical identification of its known daughter $^{250}$Pu which decays by the emission of 7.43 Mev alpha particles with a half life of 30 minutes. The removal of the element 102 isotope from the target material and the separation of the daughter element from the parent element 102 were performed by the use of a new method in which two physical separations were performed. Fig. 9 shows a schematic drawing of the experimental arrangement. The target consisted of curium deposited on a thin nickel foil and was enclosed in a container filled with helium gas. The curium was bombarded with $^{12}$C ions and the transmuted atoms were knocked into the helium gas to absorb their recoil energy. With a sufficient electric field strength it was found that practically all of these positively charged atoms could be attracted to a moving, negatively charged metallic belt placed directly beneath the target. The belt was then passed under a foil (the catcher foil) which was charged negatively relative to the belt. Approximately half of the element 102 atoms undergoing radioactive decay by alpha-particle emission would cause their daughter atoms to recoil from the surface of the belt to the catcher foil. The catcher foil was cut transversely to the direction of the belt motion into five equal length sections after a time of bombardment suited to the half life of the daughter atom to
be examined, and each section was analyzed simultaneously in counters. All of the desired measurements could be made for identifying the daughter atoms caught on the catcher foils and thus the half life of the parent of the recoiling atoms could be determined. It was found that Fm$^{250}$ could be collected on the catcher foil in accordance with a parent half life (i.e., half life for 102$^{254}$) of 3 seconds. Changing the belt speed was found to change the distribution of the Fm$^{250}$ on the catcher foil in a manner conforming to a three-second parent.

The isotope Fm$^{250}$ was identified chemically by dissolving the radioactive material from the catcher foil and separating it from the other actinide elements by the ion-exchange method. Fig. 10 shows the elution curve obtained in an experiment where nine atoms of Fm$^{250}$ were observed in the element 100 position.

In experiments starting in 1957 and continuing in 1958, a group at the Atomic Energy Institute in Moscow has produced an isotope, emitting 8.8 ± 0.5 Mev alpha particles, in the bombardment of Pu$^{241}$ (and Pu$^{242}$) with high energy O$^{16}$ ions accelerated in their cyclotron. This product was separated from the target by the recoil technique and the energy of the alpha particles was measured by use of a photographic emulsion technique. No chemical identifications were made. It is quite possible that this alpha activity is due, at least in part, to the isotope 102$^{254}$ identified by the Berkeley group.

Although the name nobelium for element 102 will undoubtedly have to be changed, no suggestion for a new name has been made as yet.

On the basis of knowledge of the ten known transuranium elements, it has become possible to predict the nuclear and chemical properties of
undiscovered elements with atomic numbers greater than 102. The undiscovered elements up to element 118 can be given positions in the periodic table as a consequence of having determined the point of filling of the 5f electron shell which occurs at element 103 and the chemical properties of all these elements can be estimated to the extent that membership of any element in a certain group or column in the periodic table foretells its chemical properties. It is expected that the actinide series will be completed at element 103 and that elements 104, 105, 106, etc. will be fitted in under hafnium, tantalum, tungsten, etc. The filling of the 6d electron shell should be followed by the addition of elements to the 7p shell, with the rare gas structure being attained at element 118.

Owing to the short half lives expected for the elements beyond element 101, it is possible that the present criteria for the discovery of a new element, chemical identification by traditional methods and separation from all previously known elements, will have to be changed at some point. Careful investigations of decay properties and reaction yields and mechanisms, together with the use of the recoil technique combined in some cases with the chemical identification of daughter isotopes, should provide means for the satisfactory identification of isotopes with very short half lives. Decay properties may have to be measured at the target area, on recoil-product nuclei, during the bombardment. In some cases, it may be possible to make a satisfactory identification of a new element isotope by using simple and fast methods involving the migration of gaseous atoms or ions, volatility properties, reactions with surfaces, or gas-flow reactions. It is likely that the identification of the first isotopes of all the new elements that will be discovered in the future will be accomplished by such methods, and the
production of isotopes of these new elements with sufficiently long half lives to allow chemical identification by traditional methods will follow later if this is possible at all. Regardless of the method used, evidence for the discovery of a new element should include a reasonable establishment of the atomic number and this requires more than the observation of predicted decay properties and yields. The establishment of a unique genetic relationship with a known lower element, such as was done for element 102, may be taken as the required additional information. It may not, however, be possible to satisfy these requirements when the preparation and detection of elements substantially further up the atomic number scale is under investigation some years from now.

About seven dozen isotopes in the transuranium region are known, and it has been possible to determine very precisely many of their nuclear properties. This has led to a systematization of radioactive decay data so that the decay properties of undiscovered nuclides can generally be successfully predicted. The predominant mode of decay of the transuranium elements is by alpha-particle emission, but decay by the spontaneous fission process becomes more and more important as the elements of highest atomic number are approached. In fact, this latter mode of decay begins to proceed at a rate comparable to decay by alpha-particle emission for some isotopes by the time element 160 is reached. Data for spontaneous fission have also been correlated in a manner which makes predictions for undiscovered isotopes feasible. In both the alpha and the spontaneous fission modes of decay, the regularities have been found to be greatest for nuclei which contain an even number of neutrons and an even number of protons, thus making predictions of the properties for undiscovered isotopes of this type the most certain. For isotopes with an odd number of protons or an odd number of neutrons
or an odd number of both protons and neutrons, the rate of decay by both alpha
decay and spontaneous fission is slower than for the regular even type. Fig.
11 shows the predicted half lives (indicated by a broken line) of the longest
lived isotopes for elements up to element 110. Unless unexpected islands of
stability due to closed neutron or proton shells are found, the predictions
based on the regularities in the decay properties suggest that it should not
be possible to produce and detect additional elements beyond another half dozen
or so.

So far we have given no consideration to the methods by which these
undiscovered elements might be prepared. The process of multiple slow
neutron capture as a result of intense neutron irradiation over a long period
of time offers little promise for the first synthesis of transuranium elements.
Some of the necessary intermediate isotopes have half lives too short to be
present in appreciable concentrations as required. The production of new
transuranium isotopes and possibly even new elements in thermonuclear
explosions is an interesting possibility. The products must, however, be
recovered in better yield than has yet been possible.

Synthesis by heavy-ion bombardments is the most feasible method
previously known for the preparation of elements of high atomic number. Isotopes
of californium, einsteinium, and fermium have been produced by the bombard-
ment of uranium with carbon, nitrogen, and oxygen ions, respectively, and
element 102 was discovered as a product of the bombardment of curium with
carbon ions. These heavy ions can be accelerated in cyclotrons of the
conventional type, but specifically designed cyclotrons and linear accelerators
which will be devoted to the acceleration of heavy ions to energies sufficient
laboratories throughout the world. An example is the new heavy ion linear accelerator (Hilac) at the University of California in Berkeley. In the U.S.S.R. the heavy ion cyclotron at the Atomic Energy Institute in Moscow is soon to be supplemented with a new, superior and more versatile one designed to produce large beams of heavy ions, including ions as heavy as those of magnesium and aluminum; this should greatly extend the range of experimental work that can be done in this laboratory.

The lack of target materials of high atomic number is a most serious problem. We have a program in progress in the United States for the production of milligram amounts of berkelium and californium and microgram amounts of einsteinium. According to present plans, about one milligram of Cf$^{252}$ should be obtained in about 1968. In order to prepare these elements on a more reasonable time scale, new, expensive, high-flux reactors producing $10^{15}$ to $10^{16}$ neutrons per square centimeter per second are needed.

In conclusion, it appears that the addition of new transuranium elements to the periodic table can be expected and that their study will add much to our knowledge of atomic and nuclear structure.
References


Fig. 1. Periodic table of the elements showing the heavy elements as members of an actinide series.
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**Lanthanide Series**

| 57 | La | 138.925 |
| 58 | Ce | 140.112 |
| 59 | Pr | 140.905 |
| 60 | Nd | 144.242 |
| 61 | Pm | 150.357 |
| 62 | Sm | 152.001 |
| 63 | Eu | 157.258 |
| 64 | Gd | 158.925 |
| 65 | Tb | 162.500 |
| 66 | Dy | 165.940 |
| 67 | Ho | 167.254 |
| 68 | Er | 168.934 |
| 69 | Tm | 173.040 |
| 70 | Yb | 174.966 |

**Actinide Series**

| 93 | Ac | 227.040 |
| 94 | Th | 232.050 |
| 95 | Pa | 231.050 |
| 96 | U  | 238.040 |
| 97 | Pu | 237.060 |
| 98 | Am | 243.050 |
| 99 | Cm | 247.060 |
| 100 | Bk | 251.170 |
| 101 | Cf | 259.100 |
| 102 | E  | 277.160 |
| 103 | Fm | 285.070 |
Fig. 2. Nuclear reactions for the production of heavy isotopes by intense slow neutron irradiation. Neutron capture reactions are interspersed with beta decays.
Fig. 3. Elution of tripositive lanthanide and actinide ions from Dowex-50 ion-exchange resin with ammonium alpha-hydroxyisobutyrate. The predicted positions for elements 102 and 103 are indicated by broken lines.
Fig. 4. The solution absorption spectrum of Cf$^{3+}$. The wavelength range between the dotted lines was explored.
Fig. 5. Magnetic susceptibilities of the tripositive ions of berkelium and californium in the temperature range 77 to 298°C.
Fig. 6. The magnetic moments of actinide and lanthanide elements.
Fig. 7. Nuclear reactions for the production of elements 99 and 100 in the first test thermonuclear explosion, November 1, 1952.
Fig. 8. Schematic drawing showing the recoil technique.
Fig. 9. Schematic drawing showing the experimental arrangement used in the discovery of element 102.
Fig. 10. Elution data obtained in the discovery experiments of element 102 showing the Fm$^{250}$ daughter of 102$^{254}$. The Tm, Y, E$^{253}$, and Cr$^{252}$ activities were added as tracers for calibration purposes. Dowex-50 ion-exchange resin was used, and the eluant was ammonium alpha-hydroxyisobutyrate.
Fig. 11. Predicted half-lives (indicated by broken line) of the longest-lived isotopes up to element 110.