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Author

Seaborg, Glenn T.

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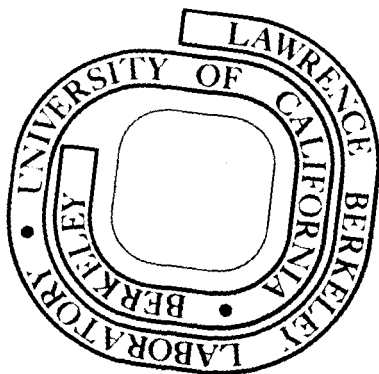
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STATUS REPORT ON THE TRANSURANIUM ELEMENTS

Glenn T. Seaborg

Lawrence Berkeley Laboratory, University of California
Berkeley, California, U.S.A.

ABSTRACT

The present status of the known transuranium elements is briefly reviewed, with emphasis on those most recently discovered. In the years following the completion of the actinide series with the discovery in 1961 of the eleventh transuranium element (lawrencium, element 103), the first two transactinide elements, elements 104 and 105, were discovered. There are competing claims by U.S. and Soviet scientists for the discovery of these two elements.

The chemical properties and the position in the periodic table of these 13 known transuranium elements are discussed.

Plans for the synthesis and identification of additional near transactinide elements and for the far transactinide elements, the superheavy elements, are discussed. Predictions are made concerning the chemical properties, electronic structures and position in the periodic table of these elements.

The possible synthesis of the transactinides, including the superheavy elements, by means of bombardments with heavy ions is described. This includes a discussion of the predicted nuclear properties of these elements and the nuclear reactions that might be used in their production.

INTRODUCTION

I shall attempt to describe in this review some of the most recent interesting results in the field of the transuranium elements, including recent experimental discoveries on the known elements and theoretical predictions and prospects for additional elements, especially the superheavy elements.

Today there are thirteen known transuranium elements with names and symbols as follows: neptunium, Np (atomic number 93); plutonium, Pu (atomic number 94); americium, Am (atomic number 95); curium, Cm (atomic number 96); berkelium, Bk (atomic number 97); californium, Cf (atomic number 98); einsteinium, Es (atomic number 99); fermium, Fm (atomic number 100); mendeleevium, Md (atomic number 101); nobelium, No (atomic number 102); lawrencium, Lr (atomic number 103); rutherfordium, Rf, or kurchatovium, Ku (atomic number 104); and hahnium, Ha, or nielsbohrium, Ns (atomic number 105).

The duplicate suggested names for elements 104 and 105 result from competing claims by U.S. and Soviet scientists for the discovery of these

two elements, and I shall have more to say about this a little later.

There is general agreement that the transuranium elements should be placed in the Periodic Table as shown in Figure 1. The actinide series, analogous to the lanthanide series of rare earth elements, is completed with the eleventh transuranium element (lawrencium, element 103), after which the elements 104 to 118 should be located in the main body of the Table as analogs of hafnium (element 72) to radon (element 86). Of these, only elements 104 and 105 have been discovered, with some indications that their chemical properties are consistent with this placement in the Periodic Table, and the others are placed in their predicted positions.

The transuranium elements are all synthetic, i.e., man-made. The studies of the chemical properties of the actinide elements have been made with macroscopic (i.e., weighable) quantities for all up to and including einsteinium (atomic number 99). Those beyond this can only be studied with amounts so small as to be unweighable, relying on their radioactive properties to work with "tracer" quantities. The chemistry of the heaviest of these has been studied using "one atom at a time" techniques. The actinide elements neptunium through einsteinium are being synthesized in increasingly large quantities through neutron irradiation processes in high neutron flux nuclear reactors in several countries. Such a production program has been underway in the United States for some 20 years, augmented by some production for special purposes through the use of fast neutrons produced in thermonuclear reactions--this production campaign is depicted in summary form in Figure 2. The total yields from the culminating stage of this program--from the High Flux Isotope Reactor-Transuranium Processing Facility (HFIR-TRU) at the Oak Ridge National Laboratory and the Savannah River Plant and Transuranium Processing Facility (SRP-TRU)--are shown in Table 1. These isotopes have been available for investigations in the numerous laboratories investigating the transuranium elements in the United States, and also to laboratories in other countries, and have been used in some of the investigations that I shall describe.

Table 1. Actinide Production (as of July 1972)

	HFIR-TRU	SRP-TRU
Cm-244	85 g	(3 kg)*
Bk-249	35 mg	80 mg
Cf-252	300 mg	600 mg
Es-253	2 mg	350 µg
Es-254		3 µg
Fm-257	1 pg	1.5 pg

* SRP-SRP

Of especial interest is the discovery, in the last few years, that as the latter part of the series is approached, the electrons become more tightly bound so that, starting with californium, the (II) state can be achieved with increasing ease toward the end of the series. It has been possible to synthesize CfBr_2 and CfI_2 , and although the short half-life of einsteinium (Es-253, 20 days) makes it more difficult to work with, there is some evidence for EsCl_2 , EsBr_2 and EsI_2 . (These new methods of synthesis have also led to the production of dipositive americium, as AmCl_2 , AmBr_2 and AmI_2 , a long predicted consequence of the stable half-filled 5f subshell of seven electrons.) With nobelium, the element just before the end (represented by lawrencium), the (II) state (with the stable filled subshell of 14 5f electrons) is more stable than the (III) state. Then lawrencium, the last element in the series, is characterized by the stable (III) state with the full 5f subshell.

Consistent with this general picture, tracer experiments at the Institute of Physical Chemistry in Moscow, using a co-crystallization method, have determined the relative stability of the (II) oxidation states of californium, einsteinium, and fermium, indicating increasing stability in proceeding from californium to fermium. Similar experiments at that Institute have indicated a (I) oxidation state for mendelevium which could be stabilized by the fourteen 5f electrons in this ion.

These considerations of the relative stabilities of the (II) oxidation states of the actinide elements, in comparison with those of the lanthanide elements, are well summarized in Figure 3, in which the standard oxidation potentials (scale $\frac{1}{2}\text{H}_2 \rightarrow \text{H}^+ + \text{e}^-$ equals 0.0 volts) for the (II)-(III) couple are plotted. The (II) oxidation state has been observed for all of the elements (except promethium) whose positions on the plots are below the upper dotted line (corresponding to an oxidation potential of +2.7 volts), with measured or indicated values for the (II)-(III) standard potential as shown on the plots; the standard potentials for the elements with points above the upper dotted line (as well as for those below) have been derived by L. J. Nugent from theoretical considerations and correlations.

For those elements with standard potentials less than +1.6 volts (i.e., those with points lying below the lower dotted line) the trihalide compound can be reduced to the salt-like dihalide compound by heating with hydrogen gas or simply by heating in a vacuum. For those elements with standard potentials between +1.6 and +2.7 volts (i.e., those with points lying between the two dotted lines) the conversion of the trihalide to the salt-like dihalide requires the heating of the trihalide with the element in metallic form or the heating of the metal with mercury(II) halide. For those elements with standard potentials more than +2.7 volts (i.e., those with points above the upper dotted line) the dihalides are unstable with respect to disproportionation, or if formed, are not salt-like but have metallic characteristics. The borderline position of californium, whose standard potential of +1.6 volts lies right on the lower dotted line, is consistent with the experimental observations that CfBr_3 can be reduced to CfBr_2 by heating with hydrogen, but there is no similar reduction of CfCl_3 .

Another interesting aspect of actinide chemistry where much progress has been made in recent years is that of organometallic complexes. Two types of compounds where much work has been done on synthesis and understanding of the resultant products are the tricyclopentadienyls (Cp) and the cyclooctatetraenyls (COT). The tricyclopentadienyl compounds, with formula MCp_3 , of uranium, plutonium, americium, curium,

berkelium, and californium, have been synthesized and characterized; although the bonding in these actinide compounds, which are relatively volatile, is more covalent than in analogous lanthanide compounds, they are still properly characterized as being highly ionic. (The tetrakis(cyclopentadienyl) compounds, with formula $M(Cp)_4$, of thorium and uranium have also been synthesized and characterized.) Cyclooctatetraenyl compounds, with formula $M(COT)_2$, of thorium, uranium, neptunium and plutonium, have been synthesized and characterized--these compounds exhibit covalent bonding and the measured sandwich π -complex nature of the uranium compound, analogous to ferrocene, has led to the proposal that it be given the name "uranocene."

Other interesting organometallic complexes that have been synthesized and characterized are the tricyclopentadienyl actinide (IV) X compounds, with formula Cp_3MX , where M represents thorium and uranium and X represents halide, alkyl, $\frac{1}{2}$ oxygen and $\frac{1}{2}$ sulfur; and the tetra-kisallyl actinide (IV) compounds, with formula $M(C_3H_5)_4$, where M represents thorium and uranium.

ELEMENTS 104 AND 105

Elements 104 and 105 are the first "transactinide" elements. They have been synthesized by heavy ion bombardments and like elements 101, 102 and 103, have been identified by using "one atom at a time" techniques.

There are competing claims for the discovery of, and hence competing suggestions for the name of, the elements with the atomic numbers 104 and 105. Since I have reviewed the history of these claims many times, I will not attempt to do so again here. G. N. Flerov and co-workers at Dubna have based their claim on the observation of activities that decay by the spontaneous fission process, while A. Ghiorso and co-workers at Berkeley have based their claim on the observation of alpha particle emitters (with the advantage of exact alpha energy determination) and the observation of the previously known alpha emitting daughters of these isotopes.

The Berkeley research group has proposed that element 104 be named rutherfordium (symbol Rf) after Lord Ernest Rutherford, the British nuclear physicist, while the Dubna group has proposed kurchatovium (symbol Ku) after the Soviet nuclear physicist, Igor Kurchatov.

Ghiorso and co-workers suggest that element 105 be named hahnium (symbol Ha), after Otto Hahn, the discoverer of nuclear fission in uranium, while Flerov and co-workers suggest nielsbohrium (symbol Ns), after Niels Bohr, who played the key role in developing the early theory of electron structure of atoms.

Just within the last year C. E. Bemis and co-workers at the Oak Ridge National Laboratory have applied a daughter X-ray identification technique to the identification of element 104. The characteristic K-series X-rays from the daughter nobelium ($Z=102$) isotope, formed from the alpha-decaying parent, the 4.5-second $^{257}_{104}$, were observed in coincidence with the alpha particles, providing an unequivocal determination of the atomic number of the parent as $Z=104$. This observation confirms the work of Ghiorso and co-workers, who first reported the production and identification of 4.5-second $^{257}_{104}$.

The chemical properties of element 104, because it is the first transactinide element, are of especial interest. It is predicted that, in its role as eka-hafnium, it should have chemical properties like hafnium, which lies just above in the Periodic Table. Scientists working at the SuperHILAC (the rebuilt HILAC) at Berkeley have devised a computer-controlled, fast automatic chemistry apparatus, which has been dubbed Fast Automatic Chemistry Experiment (FAKE), for studying the chemical properties of element 104 using the 65-second $^{261}_{104}$. Previous experiments have indicated that element 104 is indeed predominantly tetra-positive, like hafnium. The FAKE apparatus should also make it possible to study the chemical properties of element 105, which should be similar to those of tantalum, by using the 40-second $^{262}_{105}$; such experiments are also planned.

BEYOND ELEMENT 105

Some detailed estimates have been made recently of the half-lives of the isotopes of element 106 that might be synthesized and identified. In line with the observation that the longest lived alpha emitting isotope of each of the elements fermium, mendelevium, nobelium, lawrencium, element 104, and element 105, is the one with 157 neutrons, it is expected that $^{263}_{106}$ should be the longest lived alpha emitting isotope of element 106. J. Randrup, C. F. Tsang, P. Möller, S. G. Nilsson, and S. E. Larsson have made estimates for the half-lives of even-even isotopes of element 106 for decay by spontaneous fission, which is expected to be the dominant mode of decay, and their results are summarized in Figure 4. They estimate that the retardation factor for the decay of the odd mass isotope $^{263}_{106}$ should be about 10^2 to 10^3 , leading to a predicted half-life of 10 to 100 milliseconds for the decay of this isotope, which is apparently shorter than the predicted half-life for alpha emission; they predict that $^{263}_{106}$ should be the longest lived isotope of element 106.

On the basis of the simplest projections it is expected that the half-lives of the elements beyond element 105 will become shorter and shorter as the atomic number is increased, and this is true even for the isotopes with the longest half-life for each element. This is illustrated by Figure 5, in which the half-lives of the longest lived isotopes (for alpha and spontaneous fission decay) of transuranium elements on a logarithmic scale are plotted against the increasing atomic number. Thus, if this rate of decrease could be extrapolated to ever-increasing atomic numbers, we would expect half-lives of the order of 10^{-10} seconds for the longest lived isotope of element 110, and 10^{-20} seconds for element 115, and so forth, with decay by spontaneous fission becoming of dominating importance beginning with element 106. This would present somewhat dismal future prospects for heavier transuranium elements, but fortunately other factors have entered the picture in recent years. These have led to an increased optimism concerning the prospects for the synthesis and identification of elements well beyond the observed upper limit of the Periodic Table--elements that have come to be referred to as "superheavy" elements.

SUPERHEAVY ELEMENTS

Theoretical calculations, based on filled shell (magic number) and other nuclear stability considerations, have led to extrapolations to the far transuranium region. These suggest the existence of closed nucleon shells at $Z=114$ and $N=184$ that exhibit great resistance to decay by spontaneous fission, the main cause of instability for the elements beyond element 105. Earlier considerations had suggested a closed shell at

Z=126, by analogy to the known shell at N=126, but due to the Coulomb field of repulsion this shell is predicted to be weak in the proton case. Although Z=164 and N=318 may represent additional hypothetical points of stability, this possibility has not been investigated in as much detail. It also corresponds to a much more extensive extrapolation and any predictions are therefore much more uncertain.

Table 3 illustrates the known closed proton (Z) and neutron (N)

Table 3. Closed Proton (Z) and Neutron (N) Shells with Closed Electron (Noble Gas) Shells for Comparison (Predicted shells shown in parentheses)

Z	N	e ⁻
2 (He)	2 (⁴ He)	2 (He)
8 (O)	8 (¹⁶ O)	10 (Ne)
20 (Ca)	20 (⁴⁰ Ca)	18 (Ar)
28 (Ni)	28 (⁵⁶ Ni)	36 (Kr)
50 (Sn)	50 (⁸⁸ Sr)	54 (Xe)
82 (Pb)	82 (¹⁴⁰ Ce)	86 (Rn)
- - - -	126 (²⁰⁸ Pb)	- - - -
(114)	- - - -	(118)
(126)	(184)	
		(168)
(164)	(318)	

shells ("magic" numbers) and the predicted closed nucleon shells (shown in parentheses) that might be important in stabilizing the superheavy elements. The indicated shells at Z=164 and N=318 are included for completeness, although there appears to be little hope of ever observing these on earth; it is, of course, conceivable that nuclei containing such extremely large numbers of protons or neutrons may someday be observed by identification of their characteristic radiation emitting from stars. Included by way of analogy are the long-known closed electron shells observed in the buildup of the electronic structure of atoms. These correspond to the noble gases and the extra stability of these closed shells is reflected in the relative chemical non-reactivity of these elements. The predicted (in parentheses) closed electronic structures at Z=118 and Z=168 will be further discussed later.

Enhancing the prospects for the actual synthesis and identification of superheavy nuclei is the fact that the calculations show the doubly magic nucleus ²⁹⁸114 not to be the single long-lived specimen but to be merely the center of a rather large "island of stability" in a "sea of spontaneous fission." In Figure 6, nuclear stability is depicted in a scheme that shows regions of known or predicted stability as land masses in a sea of instability representing forms of radioactive decay. The grid lines show "magic" numbers of protons or neutrons giving rise to exceptional stability. The doubly magic region at 82 protons and 126 neutrons is shown by a mountain; a predicted doubly magic but less stable region at 114 protons and 184 neutrons is shown by a hill at the island of stability. The ridges depict areas of enhanced stability due to a single magic number. The submerged ridges show isotopes that are unstable but more stable than nearby ones because of increased stability

afforded by closed shells of nucleons in the nucleus.

Stability against decay by spontaneous fission is highest near the center of the island, and instability increases as the edges of the region are reached. The doubly magic nucleus $^{298}114$, for example, is predicted to have a very long half-life for decay by the spontaneous fission process, a substantially shorter half-life for decay by the emission of alpha particles, and it is predicted to be stable against beta particle decay. Adjacent odd A or odd Z nuclei may be even longer lived. Half-lives as short as nanoseconds (billionths of a second) are discernible with the instruments now used in the detection of new elements, so that there is quite a margin for error in employing the predictions. These indicate that maximum stability should be found at 110 protons and 184 neutrons ($^{294}110$), taking into account decay by both spontaneous fission and alpha particle emission, and this nucleus is possibly stable against decay by beta particle emission.

If the half-life of a superheavy nucleus should be as long as a few times 100 million years, this would be long enough to allow the isotope to survive and still be present on the earth (as in the case of uranium-235, which has a half-life of 700 million years), provided that it was initially produced in the cosmic nuclear reactions that created the matter of the solar system. Although so long a half-life is unlikely, adventurous scientists have pursued this remote possibility. Consultation of the Periodic Table (Figure 1 or Figure 7) will show that element 110, for example, is a homolog of platinum and should have chemical properties similar to those of that precious metal. Therefore searches have been made for element 110, and also for its neighboring elements, including element 114, in naturally occurring platinum, lead, and other ores by workers in a number of laboratories.

Every attempt to find evidence, direct or indirect, of the superheavy elements in nature has not been conclusive enough to give a clear-cut answer. Because of the physical limitations inherent in any experimental technique, it is not possible to say that the superheavy elements do not exist on earth, but such existence appears unlikely. The results of such searches establish that the concentration, if they are present, is extremely small--for example, much less than one part in a million million parts of ore. Searches have also been made in cosmic rays, meteorites and moon rocks with no generally accepted positive results except for some indirect evidence of former presence in meteorites during the early history of the meteorite's life. The postulated current synthesis of a broad range of chemical elements, possibly including superheavy elements, in some unusual stars might enhance the prospects for finding even shorter lived (half-life much less than 100 million years) superheavy elements in cosmic rays; elements as heavy as uranium have apparently been found in cosmic rays emanating from such stars.

SYNTHESIZING SUPERHEAVY ELEMENTS

Unless they are observed as cosmic rays, it appears that the superheavy elements will be observed on earth only as the result of their creation by man on earth. There appear to be nuclear synthesis reactions that might make this possible, and these should be effected by bombarding target nuclei with sufficiently energetic projectiles consisting of heavy ions, possibly very heavy ions. Unfortunately, the yield of the desired product nuclei is predicted to be very small because the overwhelming proportion of the nuclear reactions lead to fission rather than to the desired synthesis of superheavy nuclei through amalgamation of the heavy

ion projectile and the target nucleus. We even have the question of whether such amalgamation or joining together of the projectile and target nuclei occurs to an observable extent.

The accelerators required to effect such nuclear reactions are complicated machines. Each projectile heavy ion must be multiply charged to a high degree by the removal of orbital electrons so that it can be accelerated in a machine of reasonable size, and it is very difficult to obtain intense beams of such ions. And each such ion must be accelerated to a sufficient energy in order that it can overcome the repulsion of its positive charge by the positive charge of the target nucleus (that is, overcome the so-called Coulomb repulsion) which is necessary in order to allow the amalgamation of the projectile with the target nucleus. So-called heavy ion accelerators that meet these criteria to varying degrees are in operation or under construction in various parts of the world. Two of these that have special capabilities are the SuperHILAC situated at the Lawrence Berkeley Laboratory, and the tandem cyclotron combination at the Dubna Laboratory. In addition, powerful machines are under construction--notably the UNILAC heavy ion accelerator of the Gesellschaft für Schwerionenforschung (GSI) at Darmstadt, Germany. Scientists now working with or planning to work with these machines will be devoting a tremendous effort toward the synthesis and identification of the super-heavy elements.

An inherent difficulty in the synthesis of the superheavy nuclei situated near the center of the island of stability is the simultaneous requirement that there be a sufficient number of neutrons as well as protons in the product nucleus. The desired product nuclei have a larger ratio of neutrons to protons than the constituent projectiles and target nuclei and hence somewhat unusual nuclear reactions are required. There appears to be no way of knowing prior to the actual experimental attempts which nuclear reactions will be most effective, although theoretical predictions may be helpful in pointing toward some preferred directions.

The identification of the presumed small yield of superheavy elements after their synthesis will require extraordinary ingenuity by the participating scientists. One method will be to steer the charged superheavy product nuclei, individually projected from the target as the result of the reaction by which they are produced, through a series of electric and magnetic fields interspersed with counters and detectors in order to determine the atomic and mass number. Another method will be to subject the recoil superheavy products to "in situ" or "on line" chemical reactions in the target area, or, after catching them on some kind of a collector, to subsequent chemical separation procedures, based on chemical properties such as those predicted in the following sections. It is, of course, possible that even if the superheavy elements are relatively stable toward radioactive decay, it will be impossible to detect them because there exist no nuclear production reactions with sufficient yields of these desired products. In that case, we would be faced with the tantalizing and philosophical dilemma of feeling certain that the island of stability is there in principle, but man lacks the ingenuity to reach it or nature has imposed insuperable roadblocks to make arrival at the island impossible.

Another approach to the synthesis of superheavy elements, the use of extremely high neutron fluxes (such as produced in thermonuclear explosions) to produce instantaneous multiple neutron capture, followed by multiple beta decay, does not seem too hopeful because of the termination of such paths by spontaneous fission decay. H. W. Meldner has suggested

the circumvention of this spontaneous fission roadblock by the use of successive thermonuclear explosions with intermediate beta decay between explosions. This approach offers great technical, financial and political difficulties but may be the only ultimate hope for reaching the island of stability should it prove to be inaccessible through the route of heavy ion bombardment.

An interesting intermediate possibility, short of the actual synthesis of superheavy nuclei, has been investigated theoretically by Walter Greiner and co-workers, based on earlier calculations with lighter atoms. Their calculations indicate that the collision of two very heavy nuclei might lead to the transient creation of a superheavy nuclear quasimolecule surrounded by a complete atomic shell structure; each individual electron would behave approximately as though the two central nuclei of the atom were one nucleus with nuclear charge equal to the sum of the two. In fact, the two nuclei are calculated to remain almost completely separated with only very little binding of the quasimolecule coming from the additional electronic binding energy of the combined system. Very short lived quasimolecules might be detected by observing the characteristic X-ray energies corresponding to a nucleus of atomic number equal to the sum of the atomic numbers of the original two colliding heavy nuclei; actually, the low energy M X-rays of such transient superheavy nuclei ($Z=132, 143$ and 145) may have already been observed by P. H. Mokler, H. J. Stein and P. Armbruster in Germany during the bombardment of gold, thorium, and uranium with iodine ions. However, to observe the higher energy and more interesting K X-rays, the system probably must hold together longer than the time involved in such a simple collision.

Even if the two nuclei do not hold together for any longer than the transit time for a collision, such systems offer some exciting possibilities. One of the effects that might be observed, even if the quasimolecule does not stick together, is the autoionization of positrons (coming from the creation of electron-positron pairs) when the combined charge of the superheavy nuclear quasimolecule is equal to or greater than about 170; the observation of such positrons might yield interesting information about the electromagnetic interaction.

CHEMICAL PROPERTIES OF SUPERHEAVY ELEMENTS

If the nucleus is sufficiently stable, we can then, and only then, begin to consider experiments concerned with the chemical properties of the superheavy elements. (The upper limit so far as stability of electronic structure is concerned seems to be near $Z=170$.) Turning first to the consideration of electronic structure, upon which the chemical properties must be based, modern high speed computers now make possible the calculation of such structures. Such calculations have been carried out by scientists at a number of places throughout the world, with perhaps the leading effort taking place at the Los Alamos Scientific Laboratory by J. B. Mann, J. Waber and co-workers, and in Germany by B. Fricke and W. Greiner. The calculations show that elements 104 to 112 are formed by the filling of the 6d subshell, which makes them, as expected, homologous in chemical properties with the elements hafnium (number 72) through mercury (number 80). Elements 113 through 118 result from the filling of the 7p subshell and are thus similar to the elements thallium (number 81) through radon (number 86). Thus these calculations are consistent with the Periodic Table shown in Figure 7 and are better illustrated in the Periodic Table shown in Figure 8, which shows the filling of all the electron subshells. It is of much interest here that element

114 proves to be homologous with that very stable element, lead; it can therefore be called "eka-lead," using the terminology of Mendeleev to denote an undiscovered element in a periodic column, or family, that should have properties similar to those of the preceding members of the family. The calculations show that element 119 should be an alkali metal, element 120 an alkali earth, and element 121 similar in properties to actinium and lanthanum, again consistent with Figure 7.

The author--and also the Russian chemist Vitalii I. Goldanskii and others--has speculated for some years that another inner transition series of elements, somewhat like the lanthanide and actinide series, should begin in the vicinity of element 120. Quantum theory indicated that this series would be formed by the addition of 18 electrons to an inner 5g subshell or 14 electrons to a 6f subshell, but the order of filling these shells could not be predicted until the computer techniques became available. The calculations now suggest that after the addition of some electrons to the 7d and 6f subshells, the filling of the inner 5g subshell takes place in an orderly manner. This is followed by the filling of the inner 6f subshell, leading to an inner transition series totalling 32 elements, ending somewhere near element 153 (Figures 7 and 8). The lower members of the series, perhaps through element 124, might be generally homologous with the lower members of the actinide series. Throughout the series, however, the correlation would be indistinct, although the tri-positive (III) oxidation state might be the distinctive one. Since the difference in energy levels of successive electrons is very small, this series of elements will exhibit multiple, barely distinguishable oxidation states, leading to quite complicated chemistry. I have termed this series of 32 inner transition elements the "superactinide" elements. This nomenclature is equivalent to suggesting for element 121 the name "superactinium," rather than "eka-actinium," in recognition of the prediction that this element should serve as an approximate prototype for the following 32 or so, rather than following 14, elements. More strictly speaking, since the 5g subshell is filled before the 6f subshell, the elements in the neighborhood of atomic numbers 140-153 could be called the "eka-actinide" elements to correctly designate the filling of the 6f subshell so as to reflect the analogy with the actinide elements in which the 5f subshell is filled.

After the completion of the superactinide series, the addition of electrons to the remaining positions in the 7d subshell would form elements 154 through 162 (eka-104 through eka-112). Filling the 8p shell of six electrons would result in elements 163 through 168, the latter expected to be a noble gas (or noble liquid). Note that element 164, a possible point of high nuclear stability, would on this basis be an eka-114 or dvi-lead, according to the nomenclature of Mendeleev. Thus the known or predicted centers of nuclear stability at tin (element 50), lead (element 82), element 114, and element 164, would be members of the same column or family in this oversimplified Periodic Table. This is a curious kinship, because the interactions that determine nuclear stability are different from those that determine electronic structure, which governs an element's position in the Periodic Table.

All of these considerations concerning electronic structure are reflected in the Periodic Table illustrated in Figure 8. Actually, recent, more careful calculations have indicated that the picture is not this simple. These calculations indicate that other electrons (8p and 7d) in addition to those that have been identified in the above discussion enter the picture as early as element 121 (or even element 103), thus further complicating the picture. These perturbations become

especially significant in the region beyond the superactinide series, leading to predictions of chemical properties that are not consistent, element by element, with those suggested by Figures 7 and 8. However, for the present purpose, Figure 7 gives an overall view that is very helpful in understanding the whole picture in an over-simplified manner.

DETAILED PREDICTIONS OF CHEMICAL PROPERTIES

Using Figure 7, and guided by the predicted electronic configurations summarized in Figure 8, it is possible to offer some detailed predictions on the properties of the elements beyond lawrencium. Of course these elements will best be produced at first only on a "one atom at a time" basis and they offer scant hope for ultimate production in the macroscopic quantities that would be required to verify some of these predictions. However, many of the predicted specific macroscopic properties, as well as the more general properties predicted for the other elements, will be useful in designing tracer experiments for the chemical identification of any of these elements that might be synthesized. After summarizing predictions of the chemical properties of the known--but not yet much chemically studied--elements 104 and 105, some predicted chemical properties of elements beyond element 105 and especially of some superheavy elements are included.

Element 104 (eka-hafnium) is predicted to resemble its homolog, hafnium (element 72), in its chemical properties; thus it should also resemble thorium, but not so much as it does hafnium. It should be predominantly tetra-positive both in aqueous solution and in its solid compounds, although it should exhibit solid halides and perhaps aqueous ions of the (II) and (III) oxidation states as well.

One probably can predict some of the crystallographic properties of tetra-positive element 104 by extrapolation from those of its homologs zirconium and hafnium. The ionic radii of tetra-positive zirconium (0.74\AA) and hafnium (0.75\AA) suggest an ionic radius of about 0.78\AA for tetra-positive element 104, allowing for the smaller actinide than lanthanide contraction. Since the ionic radius of tetra-positive thorium is 0.98\AA , one would expect element 104 to have a crystal chemistry like that of hafnium and different from that of thorium. Further, one would expect the hydrolytic properties of element 104 and the solubilities of its compounds (such as the fluoride) to be similar to those of hafnium. The sum of the ionization potentials for the first four electrons should be less than that for hafnium, which suggests that it should be easier to oxidize element 104 to the (IV) ionic state although, of course, the formation of the (IV) state in covalent form will not require complete removal of all four electrons. Some of these and other predicted properties of element 104 are tabulated in Table 4.

Element 105 (eka-tantalum) should resemble tantalum and niobium, and to a lesser extent protactinium, with the pentavalent state being the most important. It should exhibit several oxidation states, such as (IV) and (III), in addition to the more stable (V) state. There should be an extensive range of complex ions, offering a rich chemistry.

The chemical properties of element 106 (eka-tungsten) are predicted to be similar to those of tungsten, molybdenum, and to some extent chromium, with an even richer chemistry of complex ions than these elements. The hexafluoride should be quite volatile and the hexachloride and pentachloride and oxychlorides should be moderately volatile.

Table 4. Some Predicted Properties of Element 104 (Eka-Hafnium)

Chemical group	IV
Atomic volume, cm ³ /mole	14
Density, g/cm ³	18
Most stable oxidation state	+4
Oxidation potential, V	$M \rightarrow M^{4+} + 4e^- > +1.7$
Ionic radius, Å	0.78
Crystal structure	hcp
Metallic radius, Å	1.6
Melting point, °C	2100
Boiling point, °C	5500

Element 107 should, similarly, be an eka-rhenium (with a volatile hexafluoride) and element 108 an eka-osmium, which suggests that the latter should have a volatile tetra-oxide that should be useful in designing experiments for its chemical identification. Elements 109, 110, and 111 (eka-iridium, eka-platinum, and eka-gold) should be noble metals. If the upper oxidation states are stable, volatile hexa- and octafluorides might be useful for chemical separation purposes. These elements should have a strong tendency toward the formation of complex ions.

There is a special interest in element 110 and its expected eka-platinum character. It should have an extensive range of oxidation states with perhaps the (IV) and (VI) states the most important, and should be characterized by an extensive complex ion chemistry.

The prediction of the chemical and physical properties of element 111 (eka-gold) is a rather difficult task. Since there is no discernible trend in stability as one goes to higher Z in the group IB elements, the periodic system is of no direct help in predicting the most stable oxidation state. Furthermore, the most sophisticated computer calculations find the ground state of the outer electronic configuration of 111 to be $6d^9 7s^2$ compared to the $6d^{10} s^1$ ground states of copper, silver, and gold. O. L. Keller, C. W. Nestor, Jr., and T. A. Carlson of Oak Ridge National Laboratory, and Burkhardt Fricke of Gesellschaft für Schwerionenforschung, Darmstadt, Germany, have approached the prediction of element 111 chemistry by identifying those electronic factors which appear most important in determining the chemistry of copper, silver, and gold, and then evaluating how these factors will extrapolate to eka-gold. The most important property to predict is the most stable oxidation state and the relative stabilities of other oxidation states. Using the results of the computer calculations judiciously, coupled with various well-established qualitative chemical theories, these authors have made preliminary estimates of the chemical behavior of element 111. It appears that it will be most stable in the oxidation state (III), and that it will be about as reactive as gold with a chemistry similar to Au(III), but with more extensive complex ion formation. The heat of sublimation is also expected to be similar to that of gold, although a direct extrapolation is not possible. Furthermore, the possibility exists that an ion with negative oxidation state (I) analogous to the auride ion will be stable. On the other hand, the oxidation state of plus I for element 111 should be much less important than for gold. If it exists, it will probably be in cyanide complexes.

An important question concerns the relative stability of element 111 in the oxidation state of (II). For the group IB elements to achieve this state appears to require an essentially ionic mechanism. Only copper is most stable in this state. Silver can achieve the oxidation state of (II) only with difficulty and for gold it probably does not exist. The extrapolated sum of the first two ionization potentials of element 111 is so high as to indicate that it will be even less likely than gold to show an oxidation state of (II).

Similar detailed considerations concerning the chemical properties have not yet been made for element 112 (eka-mercury). More qualitative conjectures suggest that the most stable oxidation form might be the (I) state, but higher oxidation states will surely be important in aqueous solution and in compounds. It should be a distinctly noble metal and there have even been suggestions, on the basis of its calculated high ionization potential, that the interatomic attraction in the metallic state will be small, possibly even leading to high volatility as in the noble gases. A simple extrapolation of the volatility properties of its homologs zinc, cadmium and mercury also suggests that metallic element 112 should be quite volatile. Again, it should have an extensive complex ion chemistry. A number of these considerations suggest a chemistry noticeably different from its homolog, mercury.

As explained earlier, there is special interest in the properties of elements in the vicinity of atomic number 114. With the assistance of O. L. Keller, Jr., J. L. Burnett, T. A. Carlson, and C. W. Nestor, Jr., of Oak Ridge National Laboratory, some predicted properties of elements 113 and 114, eka-thallium and eka-lead, respectively, have been tabulated in Table 5.

Table 5. Some Predicted Properties of Elements 113 and 114

	Element 113 Eka-Thallium	Element 114 Eka-Lead
Chemical group	III	IV
Atomic volume, cm ³ /mole	18	21
Density, g/cm ³	16	14
Most stable oxidation state	+1	+2
Oxidation potential, V	$M \rightarrow M^+ + e^-$ -0.6	$M \rightarrow M^{2+} + 2e^-$ -0.9
First ionization potential, eV	7.4	8.5
Second ionization potential, eV		16.8
Ionic radius, Å	1.48	1.31
Metallic radius, Å	1.75	1.85
Melting point, °C	430	70
Boiling point, °C	1100	150
Heat of vaporization, kcal/mole	31	9
Heat of sublimation, kcal/mole	34	10
Debye temperature, °K	70	46
Entropy, eu/mole (25°C)	17	20

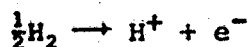
The Group IV elements show increasing stability in the (II) oxidation state relative to the (IV) state as one goes to higher atomic numbers. Carbon and silicon have very stable tetra-positive oxidation states, and germanium shows a very unstable di-positive oxidation state

In addition to a stable tetra-positive state. In tin, both the di-positive and tetra-positive oxidation states are important, and lead is most stable in the (II) oxidation state.

Thus, element 114 should be weakly, if at all, tetra-positive and the most stable oxidation state is expected to be the (II) state. The oxidation potential for the reaction



is calculated by Keller et al. to be -0.9V, on the scale where the reaction



is assigned the value of 0.0V.

For reasons similar to those that lead to the expectation of a stable (II) oxidation state in element 114, element 113--a member of Group III of the periodic system--is expected to have a preferred oxidation state of (I); an oxidation potential of -0.6V is predicted. This means that eka-thallium is expected to be more noble than thallium, being more like silver in this respect. In fact, element 113 should have a chemistry somewhat between the chemistries of Tl^{+} and Ag^{+} . 113^{+} is expected to be much more easily complexed in solution than is the case for Tl^{+} . For example, the solubility of $TlCl$ in water is not increased much by adding excess HCl or NH_3 , whereas $AgCl$ dissolves. 113 is expected to be more like silver in this respect.

Keller, Nestor and Fricke have recently made some detailed predictions of the chemical properties of element 115 (eka-bismuth) based on extrapolations of the properties of elements of group VA and relativistic calculations of electronic structure. Their preliminary results indicate that the chemical properties of element 115 should be analogous to those of thallium (group IIIA) as well as to those of bismuth. Relativistic effects on $p_{1/2}$ electrons impart to them (but not to $p_{3/2}$ electrons) some properties analogous to s electrons. On this basis the $7p_{1/2}^2 p_{3/2}$ valence shell of element 115 can be expected to behave somewhat like the $6s^2 6p$ valence shell of the Group IIIA element, thallium. Thus 115^{+} is expected to be stable in analogy with Tl^{+} rather than with the unusual and barely stable Bi^{+} and is expected to have chemical properties analogous to those of Tl^{+} . Their considerations also suggest that 115(III) should have a relative stability and chemical properties like $Tl(III)$; the relation of its chemical properties to those of $Bi(III)$ is still under investigation. Element 115 is not expected to exhibit a stable (V) oxidation state.

Certain predicted volatility characteristics of elements 115, 116, 117, and 118 (eka-bismuth, eka-polonium, eka-astatine, and eka-radon) or their compounds may offer advantages for chemical identification; this, of course, is especially true for element 118. The chemical properties of element 116 should be determined by extrapolation from polonium and thus it should be stable in the (II) state with a less stable (IV) state.

Elements 117 to 120 lend themselves to fairly detailed predictions of their chemical properties. B. B. Cunningham of the Lawrence Radiation Laboratory at Berkeley, O. L. Keller, Jr., and J. L. Burnett have made estimates of the properties of elements 117, 119, and 120, shown in Table 6; the properties of element 118 are those predicted by A. V. Grosse

Table 6. Some Predicted Properties of Elements 117, 118, 119 and 120

	117 eka-astatine	118 eka-radon	119 eka-francium	120 eka-radium
Chemical group	halogen	noble gas	alkali metal	alkaline earth
Atomic volume, cm ³ /mole	45	50	80-90	45
Density, g/cm ³			³	⁷
Oxidation potential, V	$2M^- \rightarrow M_2 + 2e^-$ +0.25-0.5		$M \rightarrow M^+ + e^-$ +2.9-3	$M \rightarrow M^{2+} + 2e^-$ +2.9
First ionization potential, eV	9.3	9.8	3.4-3.8	5.4
Second ionization potential, eV	16	15	23	10
Electron affinity, eV	2.4-2.6			
Ionic radius, Å	2.3		1.8-1.9	1.5-1.7
Crystal structure			bcc	bcc
Metallic radius, Å			2.9	2.3
Covalent radius, Å	1.8	2.3		
Melting point, °C	350-550	-15	0-30	680
Boiling point, °C	610	-10	630	1700

in 1965. These properties are obtained chiefly by extrapolation of known properties of lower homologs in the Periodic Table. Although the oxidation state -1 is listed for element 117 in Table 6, extrapolation of the chemical properties of the lighter halogen homologs, and consideration of the calculated electron structure, suggests that the (III) state should be at least as, or possibly more, important. As an example, element 117 might be similar to gold(III) in its ion exchange behavior with halide media. Element 118 should certainly exhibit positive oxidation states, such as (IV), and should form fluorides and chlorides, and probably bromides and iodides, as well. The complexity of its calculated electronic structure suggests that element 119 should exhibit an oxidation state or states higher than (I), in addition to the stable (I) state.

APPLICATION OF CHEMICAL PREDICTIONS

The foregoing predictions of the chemical properties of the super-heavy elements (SHE) make possible the design of experiments for their chemical identification following their possible production in heavy ion bombardments. A few simple preliminary experiments have been performed utilizing the tandem cyclotron combination at Dubna and the SuperHILAC at Berkeley.

Flerov and co-workers at Dubna have bombarded uranium with xenon (^{136}Xe) ions and chemically isolated fractions containing the acid-insoluble sulfides of carrier elements which behave like osmium (atomic number 76) through bismuth (atomic number 83), which are the chemical homologs of the SHE of atomic numbers 108 through 115. Since simple extrapolations of the Periodic Table suggest that the sulfides of elements 108 through 115 should be insoluble in acid solution, it is expected that these SHE would be present in these carrier fractions. A few spontaneous fission events were observed in the sulfide fraction. In further experiments without the benefit of chemical separation a few additional spontaneous fission events were shown to be accompanied by the emission of 1.5 to 3.5 neutrons per fission, a much lower neutron emission than is expected for SHE. They have made a special study of a postulated "fusion-fission" reaction, in which the projectile and target nucleus are thought to fuse into some kind of an intermediate nucleus which undergoes fission, with the hope that this might provide a mechanism for the synthesis of SHE.

At Berkeley, J. V. Kratz, J. O. Liljenzin and I have made chemical separations designed to isolate the SHE following bombardments of uranium with argon and with krypton ions. The chemical separations are based on the expected marked tendency of the SHE to form strong complex ions. A survey of the chemistry of the lighter homologs of the SHE leads to the prediction that the latter should form stable oxyacids or complex ions. Especially the SHE in the range of atomic numbers 109 through 116 should be "soft" acceptor ions, in the formalism of Ahrlund, Chatt and Davis, and are expected to form strong complex ions with heavy halide ions such as iodide; in contrast to the "hard" acceptors such as the lanthanide and actinide ions which form strong complex ions (with essentially electrostatic bonding) with light halide ions such as fluoride; this should provide the means for the separation of such SHE from the lanthanides and actinides. From Figure 9, in which the logarithm of the stability constant for the addition of the first halide ion is plotted for subgroup IIB elements, we conclude that Cd^{+2} , Hg^{+2} and 112^{+2} ions are acceptors with markedly increasing softness. Our extrapolation to 112^{+2} leads to the prediction that 112^{+2} should form very stable iodide and bromide

complex ions. This plot is made simply for illustrative purposes to suggest that similar tendencies should exist for neighboring SHE ions. Because some SHE might be reduced to lower oxidation states or the metallic state by iodide ion we expect bromide ions to be best for the conversion of SHE into strong anionic complexes while not forming complexes with hard ions such as the tripositive transplutonium elements.

The separation problem thus reduces itself to a separation of anions from cations, which can be achieved by (1) cation exchange, (2) solvent extraction with aliphatic amines, and (3) by anion exchange. Keeping in mind that one will have to deal with one atom at a time, it is obvious to think of chromatographic techniques where the separation step is repeated many times and where one can expect that even a single atom will behave statistically just as a macroamount of the same substance would do. The three above mentioned techniques have been tested in chromatographic columns using radioactive tracers of a variety of elements. Best results were obtained with the cation exchange procedure. In addition, separation steps for groups of elements have been added using different volatilities from HBr/Br₂ solutions and the different degree of complexing with bromide and chloride ions.

Based on these precepts the chemical separation scheme shown in Figure 10 has been utilized on uranium targets bombarded with argon (⁴⁰Ar) ions and, in a preliminary way, on uranium targets bombarded with krypton (⁸⁴Kr) ions. With a few exceptions (bromine, iodine, mercury) no carrier material was added, i.e., carrier-free chemical separations were made leading to the desired thin fractions for measuring the radioactivity. Actually the details of this separation scheme are such that the mercury and lead, presumably acting as prototypes for elements 112 and 114, appear in fractions separated from the main SHE group and its prototype elements.

When the chemically isolated fractions which correspond to the SHE were examined to detect possible decay by spontaneous fission and alpha emission, no evidence for such decay was found, indicating the absence of SHE. This is not surprising because the indicated nuclear reactions are not those considered to be suited to the synthesis of SHE and the intensity of the bombarding projectiles was very small.

The separated chemical fractions were well suited to the determination of the yields of a broad range of isotopes of the known elements and such yields were determined by measuring the known gamma-rays of such isotopes. In the case of the argon bombardments the yields of 136 isotopes distributed among 60 elements were determined and these are summarized in Figure 11. The broad distribution of products seems to be composed of three components: (1) nucleon transfer products forming the "rabbit ears" at each end (with a production cross section of about 600 mbarn), (2) a broad distribution of fission products presumably from a fusion-fission mechanism (production cross section about 400 mbarn), and (3) a narrower distribution of more neutron-rich fission products presumably from fissioning nuclei near ²³⁸U (production cross section about 350 mbarn).

The preliminary results from the krypton bombardments are summarized in Figure 12. Although similar to the distribution measured in the argon bombardments (Figure 11), there is the striking difference that component (2), the yield of the presumed fusion-fission reactions, is relatively much less in the krypton bombardments.

These bombardments and chemical identification procedures are continuing with the hope that the use of other heavy ions, including heavier ions than those of krypton, and greater beam intensities will lead to the synthesis and identification of superheavy elements.

PERIODIC TABLE OF THE ELEMENTS

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	105	(106)	(107)	(108)	(109)	(110)	(111)	(112)	(113)	(114)	(115)	(116)	(117)	(118)

LANTHANIDE SERIES

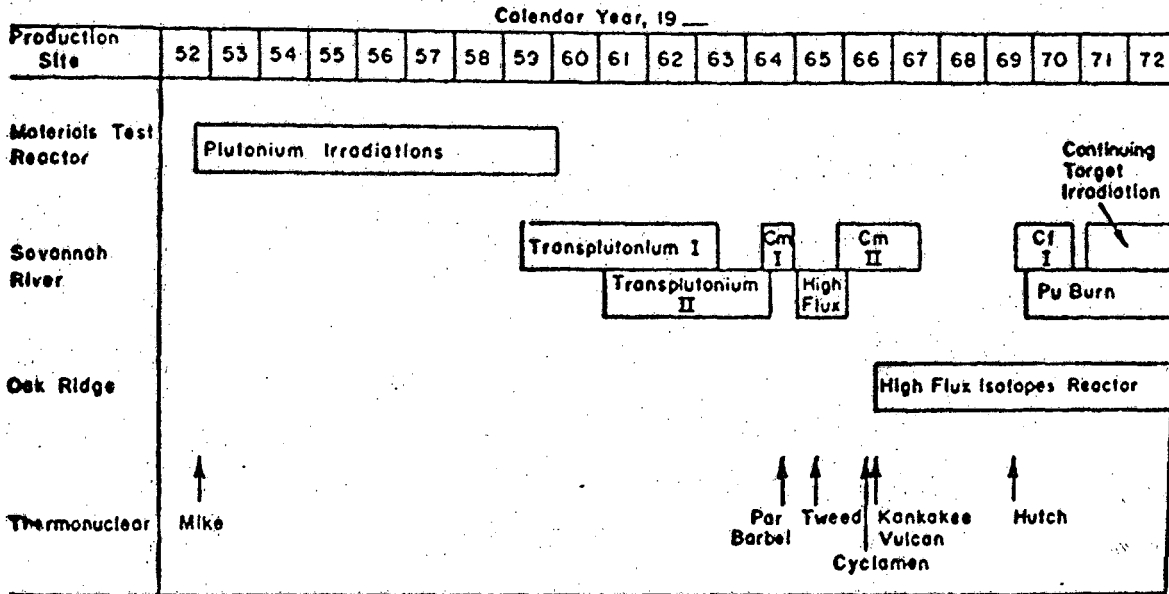
58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
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ACTINIDE SERIES

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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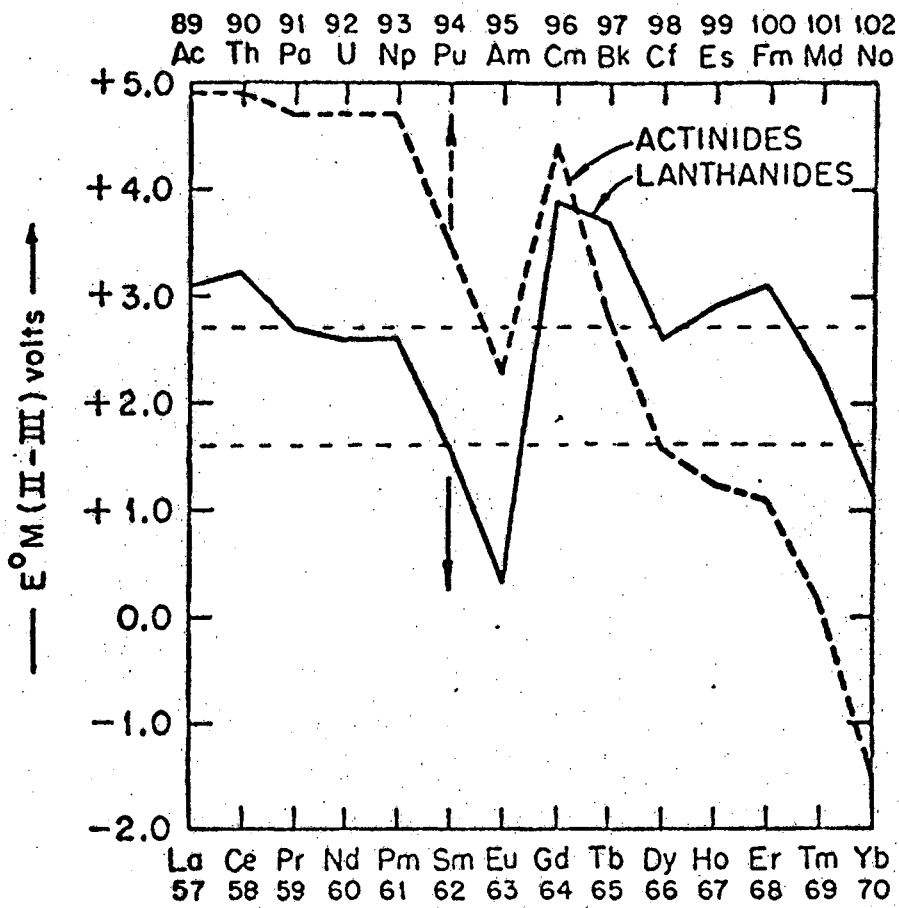
Figure 1. Periodic Table of the Elements



Principal ^{252}Cf Production Programs

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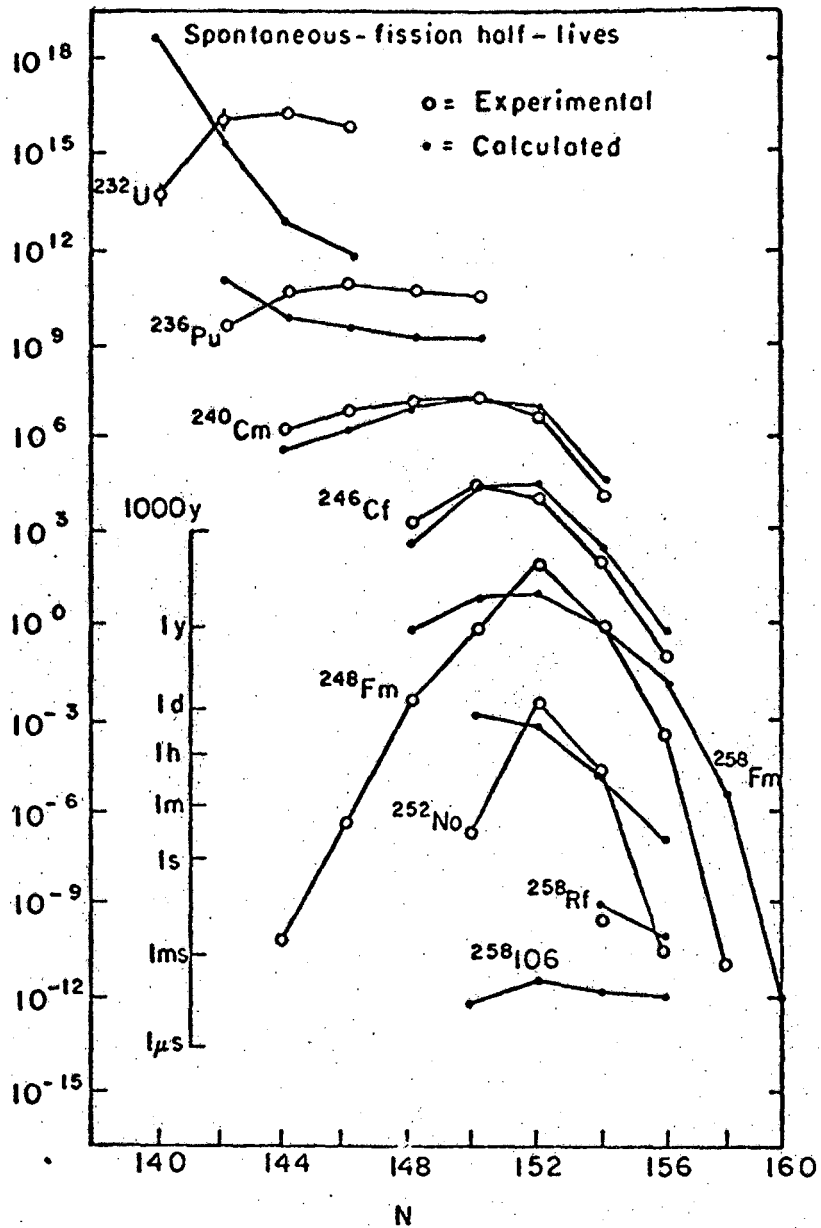
Figure 2. Principal Cf-252 Production Programs



LANA CT (II)-(III) ELECTRODE POTENTIALS
 ACCORDING TO L.J. NUGENT

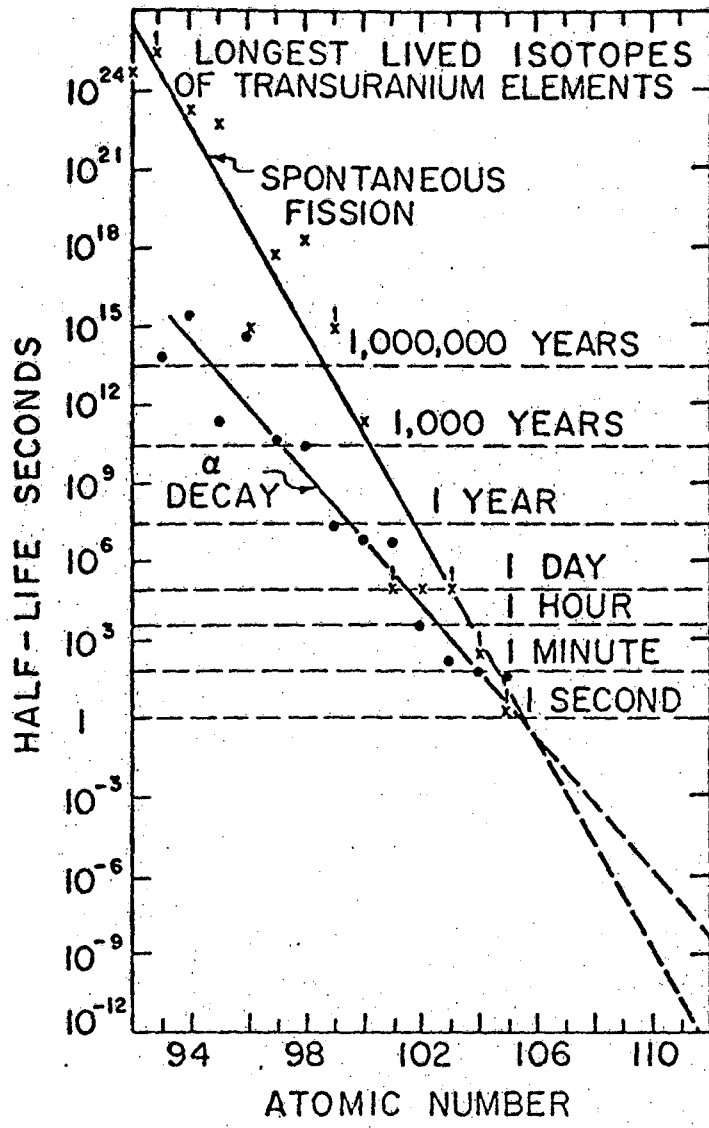
XBL 738-1117

Figure 3. LANA CT (II)-(III) Electrode Potentials
 According to L. J. Nugent



XBL733-2429

Figure 4. Spontaneous Fission Half-lives according to J. Randrup and Co-workers



XBL738-3812

Figure 5. Longest Lived Isotopes of Transuranium Elements

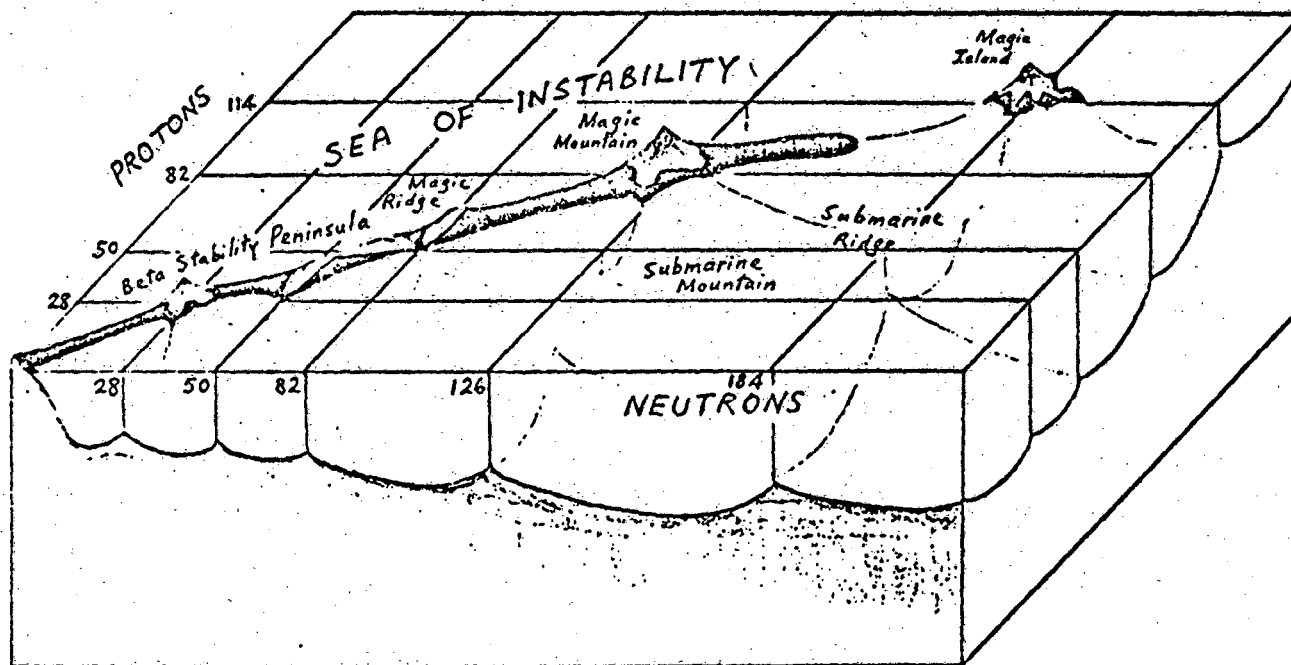


Figure 6. Allegorical Representation of Island of Stability (Magic Island)

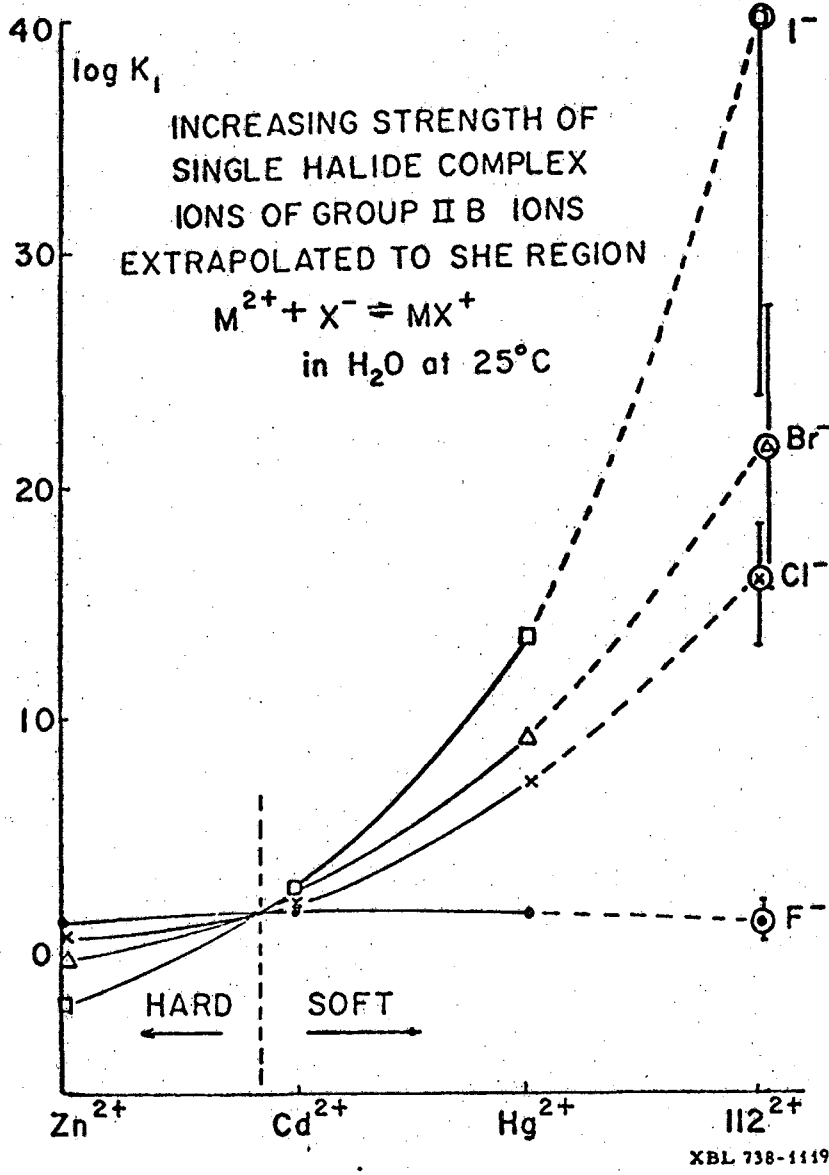


Figure 9. Increasing Strength of Single Halide Complex Ions of Group IIB Ions Extrapolated to SHE Region

Chemical processing of heavy ion bombarded uranium targets

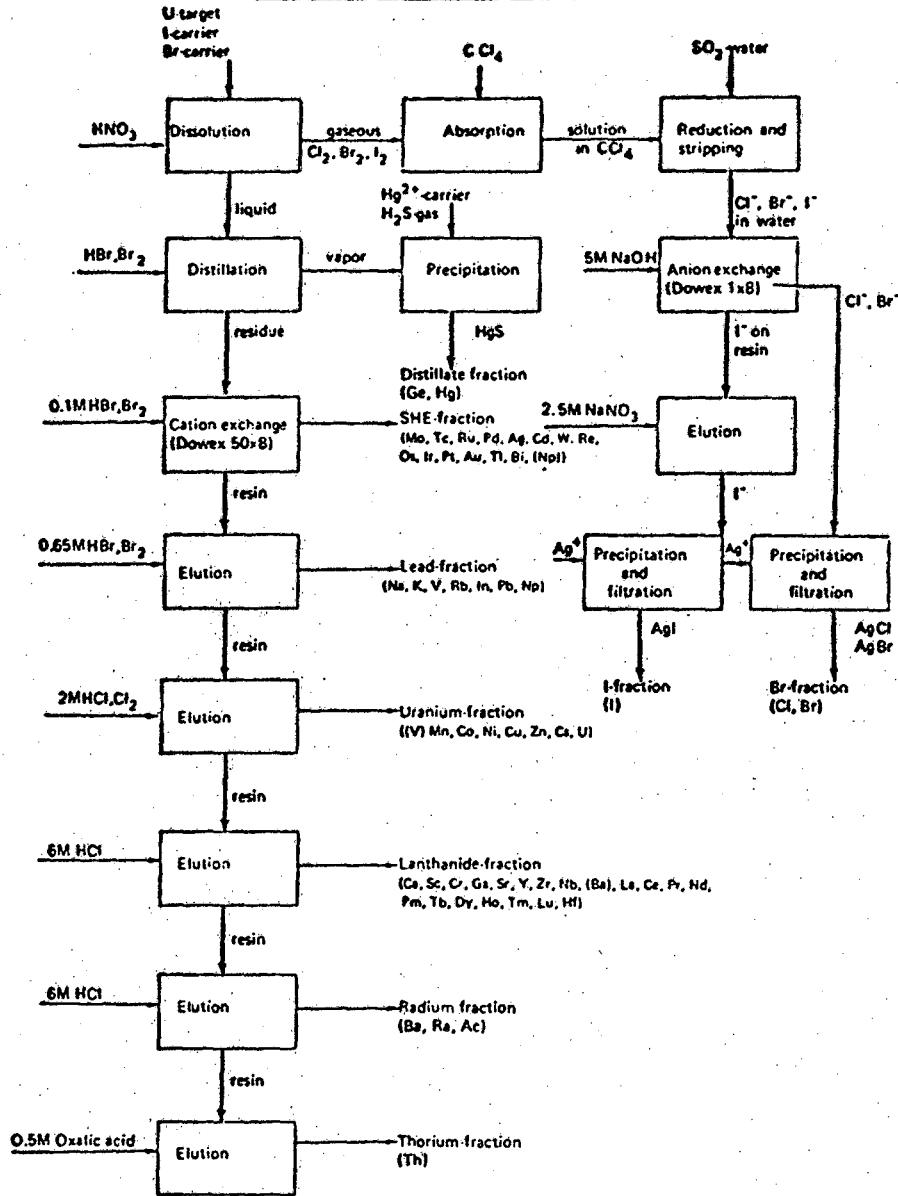


Figure 10. Chemical Processing of Heavy-ion Bombarded Uranium Targets

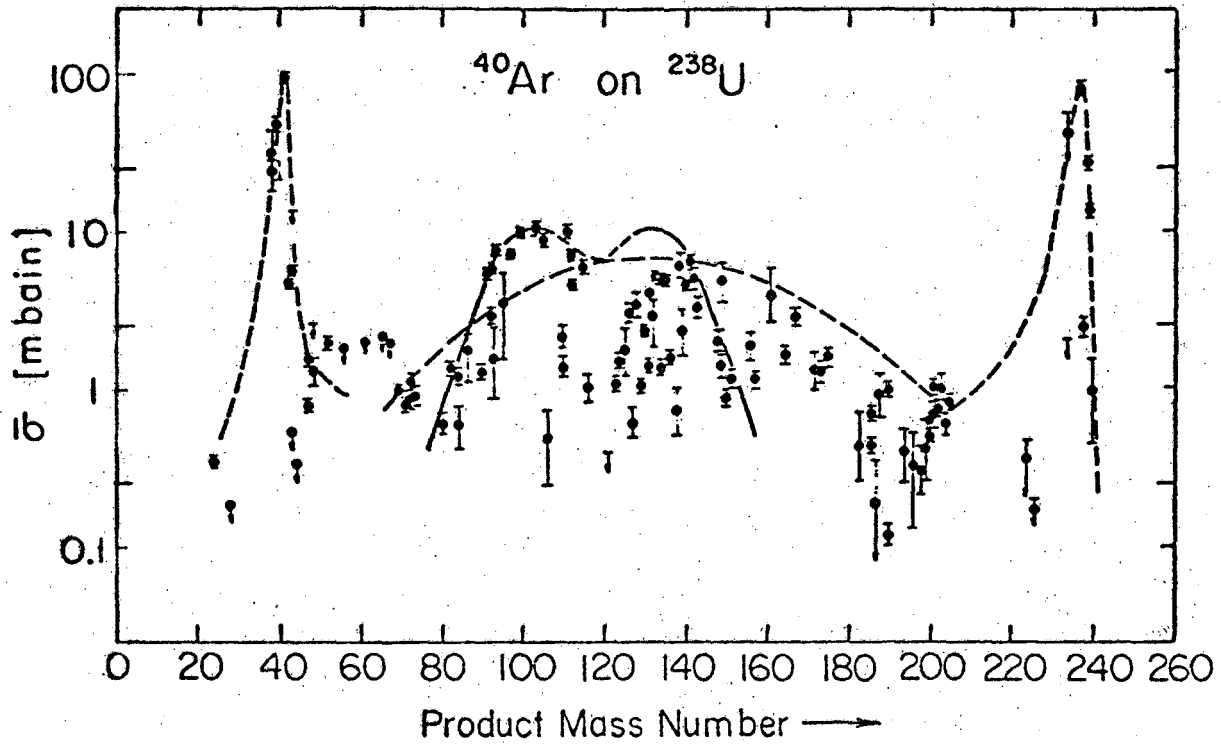
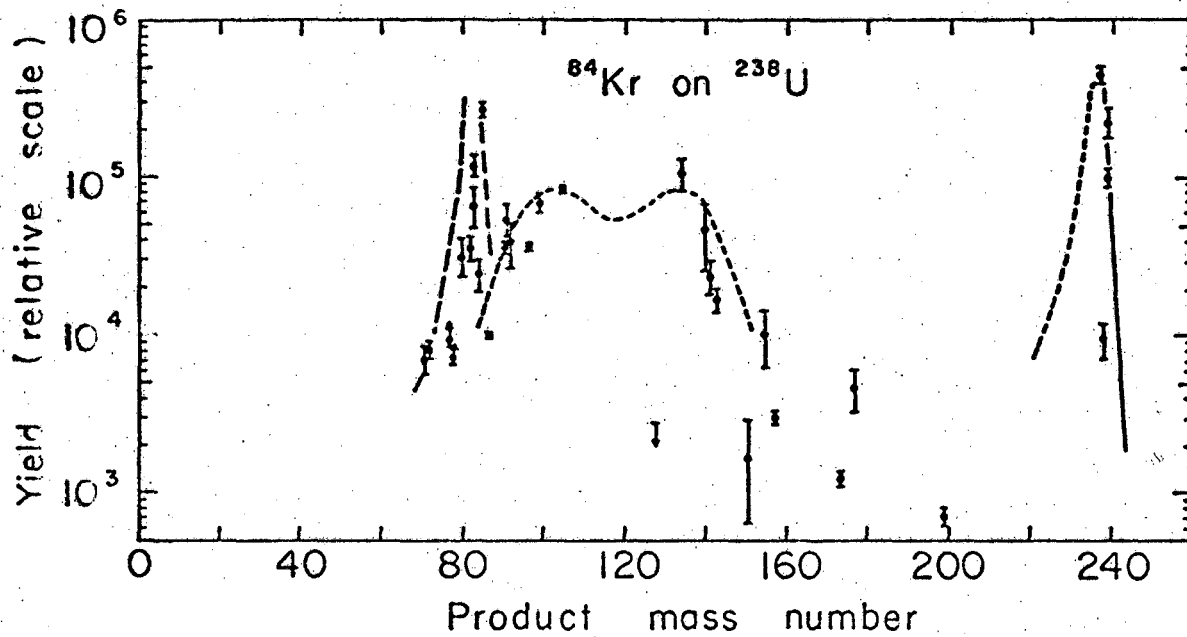


Figure 11. Yield of Products from ^{40}Ar on ^{238}U Bombardments



XBL738-3841

Figure 12. Yield of Products from ^{84}Kr on ^{238}U Bombardments

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TECHNICAL INFORMATION DIVISION
LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720