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Prospects for Further Considerable Extension of the Periodic Table

In the modern periodic table, familiar in detail to every physical scientist and indeed in a more general sense to almost every educated person, the elements through 103 have been accounted for satisfactorily. Thus, with element 104 we enter the relatively unexplored region of the periodic system with the first member of what I have chosen to call the "transactinide" elements, that is, all elements beyond the inner transition series formed by the filling of the 5f electron subshell which is commonly called the actinide series and which includes elements 90 through 103. Once we have established the location of the 14 elements in which the 5f electron shell is filled, it is possible to locate the positions of elements 104 to 121 and in the style of Mendeleev to predict their chemical properties with varying degrees of detail and reliability by comparing them with their homologs in the periodic table.

Consultation of Figure 1 shows that element 104

should be a homolog of hafnium, 105 a homolog of tantalum and so forth until element 118, a noble gas homologous with radon, is reached. If I use the notation of Mendeleev, I may call element 104 "eka-hafnium," element 105 "eka-tantalum" and so on. By this time your eyes will have caught my apparent brashness in extending the periodic system all the way to element 168, resulting in a periodic table of somewhat breathtaking dimensions. The most striking feature of this arrangement is the addition of another inner transition series of elements starting with about atomic number 122 and extending through atomic number 153. I call this grouping the "superactinide" series, because of the rough analogy to the known actinide and lanthanide series, but hasten to point out that each element of this series does not correspond to an actinide (or lanthanide) element on a one-by-one basis. This could hardly be the case, because the superactinide series is postulated to contain 32 elements, whereas the lanthanide and actinide series each contains 14 elements. Following the superactinide transition series, elements

Figure 1. Conventional form of the periodic table showing predicted locations of new elements.

154 through 168 are shown as being homologous with elements 104 through 118, with element 168 again being a noble gas (or should I say a "noble liquid"). However, I must warn that calculations which are now in progress indicate a more complicated order of filling of electron shells in elements 154 through 164, which would be inconsistent with placing these elements in the periodic table in the straightforward method shown in Figure 1 and in Figure 2 (below).

If this hypothetical extension of the known periodic table appears to be reckless, consider that calculations have confirmed much of it, that serious experimental efforts are being made to synthesize elements in the region around atomic number 114 and that searches for elements 110 and 114 in nature have been under way for more than a year. I will discuss these intriguing affairs in much greater detail later, but for the moment I ask acceptance for the proposition that the thought being given to what the periodic table might look like, in regions heretofore considered beyond the point of experimental verification, is much more than idle speculation.

Prediction of Chemical Properties of Transactinide Elements

Therefore, accepting the periodic classification shown in Figure 1 as valid, I would like first to offer some 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 according to the considerations that I shall describe a little later.

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. It should be predominantly tetrapositive both in aqueous solution and in its solid compounds, although it should be possible to form solid halides of the II and III oxidation states as well.

One probably can predict some of the crystallographic properties of tetrapositive element 104 by extrapolation from those of its homologs zirconium and hafnium. The ionic radii of tetrapositive zirconium (0.74 Å) and hafnium (0.75 Å) suggest an ionic radius of about 0.78 Å for tetrapositive element 104, allowing for the smaller actinide than lanthanide contraction. Since the ionic radius of tetrapositive thorium is 0.98 Å, one would expect element 104 to have a crystal chemistry like that of hafnium and quite different from that of thorium. Further, one would expect the hydrolytic properties of element 104 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 state. Some of these and other properties of element 104 are tabulated in Table 1, which has been prepared with the assistance of O. L. Keller, Jr., and J. L. Burnett of Oak Ridge National Laboratory.

Table 1. Some Predicted Properties of Element 104 (Eka-Hafnium)

Atomic weight	272
Atomic volume, cm³/mole	14
Density, g/cm³	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 105 (eka-tantalum) should resemble tantalum and niobium, and to a lesser extent protactinium, with the pentavalent state being perhaps the most important. The chemical properties of element 106 (eka-tungsten) are predicted to be similar to those of tungsten, molybdenum, and to some extent chromium. Element 107 should be an eka-rhenium and element 108 an eka-osmium, which suggests that the latter should have a volatile tetraoxide 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 or octafluorides might be useful for chemical separation purposes. As I shall describe later, there is a special interest in element 110 and its expected eka-platinum character. Element 112 (eka-mercury), because of the distinctive chemical properties of its homolog, mercury, should have properties that can be exploited in its chemical identification.

As I will explain in substantial detail shortly, 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, I have tabulated some predicted properties of elements 113 and 114, eka-thallium and eka-lead, respectively, in Table 2. Details of the methods of extrapolation employed to arrive at the values shown will appear in the literature, but are too exhaustive to describe at this

Table 2. Some Predicted Properties of Elements 113 and 114

	Element 113 Eka-thallium	Element 114 Eka-lead
Chemical group	III	IV
Atomic weight	297	298
Atomic volume, cm³/mole	18	21
Density, g/cm³	16	14
Most stable oxidation state	+1	+2
Oxidation potential, V	$M \rightarrow M^+ + e^-$	$M \rightarrow M^{2+} + 2e^-$
	-0.6	-0.8
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

time. However, I would like to dwell for a moment on the oxidation states and oxidation potentials of these two hypothetical elements because they are of particular interest to me.

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 tetrapositive oxidation states, and germanium shows a very unstable dipositive oxidation state in addition to a stable tetrapositive state. In tin, both the dipositive and tetrapositive oxidation states are important and lead is most stable in the II oxidation state. In a publication in 1958, R. S. Drago showed from thermodynamic considerations that this trend arises from a decrease in the strength of the covalent bonds formed by the metal atom as the atomic number increases in Group IV. From the point of view of valence bond theory, the tetrapositive state is made possible by sp^3 hybridization, which involves all four outer electrons in bonding. Thus the tetrapositive state requires strong enough covalent bonds to supply both the promotion energy for one s electron and the Gibbs free energy required for compound stability. In lead, weak covalent bonds, involving only the p electrons, tend to be formed.

Drago explained this tendency toward only p electron involvement with increasing Z by noting that (1) the heavier elements have more inner electrons to repel the inner electrons of the bonded anion and (2) the valence electrons in the higher Z elements are spread over a larger volume so that there is less overlap with the orbitals of the anion.

Since element 114 would have the largest atomic volume and the most inner electrons of the Group IV elements, sp^3 hybridization would probably be very unimportant and thus this element would be weakly, if at all, tetrapositive and the most stable oxidation state is expected to be the II state. The oxidation potential for the reaction

$$114 \rightarrow 114^{2+} + 2e^-$$
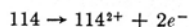
is calculated by Keller *et al.* to be -0.8 V, on the scale where the reaction

$$\tfrac{1}{2}H_2 \rightarrow H^+ + e^-$$
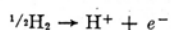
is assigned the value 0.0 V.

For reasons similar to those that lead to the expectation of a stable II oxidation state in element 114, ele-ment 113, a member of Group III of the periodic system, is expected to have a preferred oxidation state of I; an oxidation potential of -0.6 V is predicted.

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.

Elements 117 to 120 lend themselves to fairly detailed predictions of their macroscopic properties. B. B. Cunningham of the Lawrence Radiation Laboratory, University of California, Berkeley, O. L. Keller, Jr., and J. L. Burnett have made estimates of the properties of elements 117, 119, 120 shown in Table 3; the properties of element 118 are those predicted by A. V. Grosse in 1965. These properties are obtained by extrapolation of known properties of lower homologs in the periodic table.

The atomic weights given in Tables 1, 2, and 3 correspond to the mass numbers of the isotopes predicted to have the longest overall half-lives, taking into account the possibilities for decay by spontaneous fission, alpha-particle and beta-particle emission. These atomic weight estimates do not affect the prediction of chemical properties and are given to meet the formal requirement for assignment of an atomic weight. I shall describe a little later the basis for these estimates, which in some cases are of a very preliminary nature and are surely subject to change.

Electronic Structure of Transactinide Elements

The electronic configurations of the elements are, of course, what determine their position in the periodic table. For background, consultation of Figure 2, which is a form of the periodic table that shows the filling of electron shells, shows that the lanthanide series of elements, cerium through lutetium, is a transition series formed by the sequential filling of a 14-member inner electron subshell, the $4f$ subshell. This is the reason why the chemical properties of the lanthanides are similar—but not identical. In analogous fashion, the actinide series is formed by the filling of the inner $5f$ electron subshell, with each element being somewhat similar in properties to its lower lanthanide homolog. The 14-member actinide series is completed with element 103, as I noted earlier. Substantiation of the actinide concept has come from determination of the chemical properties of the actinide elements and from

Table 3. Some Predicted Properties of the Heavier Transactinides

	117 eka-astatine	118 eka-radon	119 eka-francium	120 eka-radium
Chemical group	halogen	noble gas	alkali metal	alkaline earth
Atomic weight	311	314	315	316
Atomic volume, cm³/mole	45	50	80–90	45
Density, g/cm³			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

Figure 2. Modified form of the periodic table showing known and predicted electron shells.

various kinds of spectroscopic measurements of the electronic energy levels of these elements.

Although exceedingly complex and voluminous calculations are involved, it is possible in principle to calculate the electronic structures of the actinide elements, and of the undiscovered transactinide elements well into the region of the superheavy elements. Modern high speed computers now make such calculations feasible, and they have been carried out at the Los Alamos Scientific Laboratory for elements as high as atomic number 132, by J. T. Waber, D. T. Cromer, D. Liberman, A. C. Larson, J. B. Mann, R. D. Cowan, and D. C. Griffin. Their ground state electronic con-

7p subshell and are thus similar to the elements thallium through radon. Element 119 should be an alkali metal, and element 120, an alkaline earth, formed by addition of electrons to a new 8s subshell. Element 121 should be similar in properties to actinium and lanthanum and result from the addition of the first electron to a new 7d subshell.

I, as well as the Soviet chemist V. I. Goldanskii and others, have speculated for some years that another inner transition series of elements, somewhat like the lanthanide and actinide series, should begin somewhere around element 120. Quantum theory indicated that this series would be formed by the addition of 18 electrons to an inner g subshell (the g notation appearing here for the first time in the periodic system) or 14 electrons to an inner 6f subshell, but the order of filling these shells could not be predicted until the Los Alamos computer techniques became available. The calculations suggest, as shown in Table 5 that, after the addition of some electrons to the 7d and 6f subshells, the filling of the 5g subshell takes place in an orderly manner. More recent calculations by Mann at Los Alamos now indicate the involvement of a new 8p subshell as well as the 7d and 6f subshells in the early part of this transition series (Table 6). It is reasonable to expect that the filling of the inner 5g subshell would be followed by the filling of the inner 6f subshell, leading to an inner transition series of 32 elements, ending with element 153. The lower members of the series might be generally homologous with the lower members of the actinide series. Throughout the series, however, the correlation would be indistinct although the tripositive oxidation state might be the distinctive one. Since the difference in energy levels of successive electrons is very small, this series, which as indicated earlier I have termed the "superactinide" elements, will exhibit multiple, barely distinguishable, oxidation states, leading to very complicated chemistry. (More strictly speaking, apparently the elements 140–153 could be called the "eka-actinide" elements.)

Table 4. Calculated Electronic Ground States for Gaseous Transactinide Elements 104–121

Atomic no.	Electronic structure[a]	Atomic no.	Electronic structure[b]
104	$6d^27s^2$	113	$7s^27p^1$
105	$6d^37s^2$	114	$7s^27p^2$
106	$6d^47s^2$	115	$7s^27p^3$
107	$6d^57s^2$	116	$7s^27p^4$
108	$6d^67s^2$	117	$7s^27p^5$
109	$6d^77s^2$	118	$7s^27p^6$
110	$6d^87s^2$	119	$7s^27p^68s^1$
111	$6d^97s^2$	120	$7s^27p^68s^2$
112	$6d^{10}7s^2$	121	$7s^27p^67d^18s^2$

[a] In addition to a "core" of radon, element 86, plus $5f^{14}$.
[b] In addition to a "core" of radon, element 86, plus $5f^{14}6d^{10}$.

figurations for elements 104 to 121 are summarized in Table 4. These electronic structures are consistent with the arrangement of transactinide elements shown in Figures 1 and 2. Consistent with my earlier remarks, elements 104 and 112 are formed by the filling of the 6d subshell, making them homologous in chemical properties with the elements hafnium through mercury. Elements 113 through 118 result from the filling of the

Table 5. Calculated Electronic Ground States for Gaseous Superactinide Elements 121–132

Atomic no.	Electronic structure[a]	Atomic no.	Electronic structure[a]
121	$7d^18s^2$	127	$5g^26f^37d^18s^2$
122	$5g^06f^07d^28s^2$	128	$5g^46f^37d^18s^2$
123	$5g^06f^17d^28s^2$	129	$5g^56f^27d^28s^2$
124	$5g^06f^37d^18s^2$	130	$5g^66f^27d^28s^2$
125	$5g^16f^37d^18s^2$	131	$5g^76f^17d^38s^2$
126	$5g^26f^37d^18s^2$	132	$5g^86f^17d^38s^2$

[a] In addition to a "core" of element 118.

Table 6. More Recent Electronic Ground States for Gaseous Superactinide Elements 121–126

Atomic no.	Electronic structure[a]
121	$8p^18s^2$
122	$5g^06f^07d^18p^18s^2$
123	$5g^06f^17d^18p^18s^2$
124	$5g^06f^37d^08p^18s^2$
125	$5g^16f^37d^08p^18s^2$
126	$5g^26f^27d^18p^18s^2$
	or $5g^26f^38p^18s^2$
	(tentative)

[a] In addition to a "core" of element 118.

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 $8p$ subshell of six electrons would result in elements 163 through 168.

Elements 104 and 105

I would now like to examine what has been done experimentally to extend the periodic system to the region immediately beyond the previously accepted upper boundary of known elements at element 103 and to outline the prognosis for making a major advance in discovering much heavier (superheavy) elements.

Albert Ghiorso and colleagues announced this spring that conclusive proof of the discovery of two isotopes of element 104 has been obtained. Soviet workers have discussed previously their less conclusive evidence for the discovery of an isotope of element 104 and two isotopes of element 105.

The significance of the work on elements 104 and 105 requires that I review briefly the nature of the Soviet and American research and the results that have been obtained. In 1964, Flerov and his coworkers published results obtained by use of the 310-cm cyclotron at the Dubna Laboratory with the postulated nuclear reaction

$$^{242}_{94}Pu + ^{22}_{10}Ne \rightarrow ^{260}104 + 4_0n^1$$
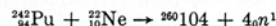
The half-life of the product isotope was measured to be only about 0.3 sec, with decay occurring only by spontaneous fission, a nonspecific decay mechanism which does not characterize an isotope with an identifying, precisely measurable energy as in alpha-particle decay. In 1966, Zvara and colleagues in the Soviet group reported experiments aimed at separating this product from the other materials present by a technique based on the premise that element 104 would be the first transactinide element and hence would have chemical properties similar to those of hafnium, below which it would fall in the periodic table (see Figs. 1 and 2). The technique consisted of attempting to convert to chlorides any product atoms formed and making use of the fact that the tetrapositive elements of Group IV (hafnium, zirconium) have very volatile chlorides when compared with the relatively nonvolatile chlorides of the actinide elements in the III oxidation state. They concluded that element 104 indeed showed the properties of a Group IV element, that is, they believed that they had observed the 0.3-sec spontaneous fission activity in the volatile tetrachloride fraction. The Soviet scientists have been using the name "kurchatovium" (symbol Ku) for the new element they believe they have observed.

However, the difficulty of performing accurate chemical experiments when the half-life is so short, and the existence in the actinide region of numerous short-lived isomers that decay by spontaneous fission, lead to the possibility of confusion and incorrect assignment of atomic number and emphasize the need for independent confirmatory experiments. In fact, attempts beginning in 1967 by Ghiorso and associates at Berkeley to produce an isotope of element 104 with the same properties reported by the Soviet workers—decay by spontaneous fission with a half-life of 0.3 sec—have been unsuccessful so far. On the other hand, the Berkeley group (A. Ghiorso, M. J. Nurmia, J. A. Harris, and P. L. and K. A. Y. Eskola) in experiments performed in 1968 and 1969 has bombarded the californium isotope ^{249}Cf with ^{12}C and ^{13}C ions in the Heavy Ion Linear Accelerator (HILAC), expecting that the following nuclear reactions might occur

$$^{249}Cf + ^{12}C \rightarrow ^{257}104 + 4n$$
$$^{249}Cf + ^{12}C \rightarrow ^{258}104 + 3n$$
$$^{249}Cf + ^{13}C \rightarrow ^{259}104 + 3n$$
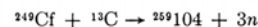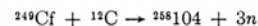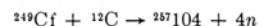
The first and third reactions resulted in isotopes which decay by alpha-particle emission with half-lives of about 4 sec and alpha energies in the range of 8.7–9.0 MeV, and the second reaction results in an isotope which decays by spontaneous fission with a half-life of about 0.01 sec. As confirmatory evidence for $^{257}104$ and $^{259}104$, they observed as their decay products the previously known isotopes $^{253}102$ and $^{255}102$; this constitutes definite proof for the identification of the two alpha-particle emitting isotopes $^{257}104$ and $^{259}104$.

More recently, Ghiorso and his group have bombarded ^{248}Cm with ^{18}O and produced $^{261}104$, which decays by the emission of alpha particles with a half-life of about a minute; this was proved by observing as its decay product the previously known $^{257}102$. Use of the isotope $^{261}104$ should make it possible to study the chemistry of element 104.

There is some very preliminary information on isotopes which could conceivably be assigned to the element with atomic number 105. Flerov and his coworkers have described the observations they have made in 1967 as the result of the bombardment of ^{243}Am with ^{22}Ne ions in the 310-cm cyclotron at Dubna. They have observed alpha-particle emitters with energies 9.7 ± 0.1 and 9.4 ± 0.1 MeV and half-life greater than 0.01 sec and between 0.1 and 3 sec which they attribute to $^{260}105$ and $^{261}105$. The observed yield is 0.05 atom/hr. The identification depends on the correlation of the alpha-particle decay with daughter alpha-particle emitters of energy 8.6 MeV which they postulate as $^{256}103$ and $^{257}103$. Again the efforts of Ghiorso and coworkers to confirm these experiments have been so far unsuccessful.

Predicted Nuclear Stability of Superheavy Elements

With regard to the development of theories of nuclear stability of the very heavy, undiscovered (superheavy) elements, we must go back twenty years, when Maria Goeppert Mayer, O. P. L. Haxel, J. H. D. Jensen, and H. E. Suess began to develop a theoretical "shell" model of the nucleus which consisted of a collection of particles moving in a nuclear force field. The collection of particles, neutrons and protons, was shown to be especially stable when the nucleus contained a "magic" number of neutrons or protons, or, more simply, nuclear shells similar to the extra-nuclear electron shells of the Bohr atom. Magic numbers of neutrons (N) or protons (Z) are generally recognized as being 2, 8, 20, 28, 50, and 82 in the elements below uranium in the periodic table. The magic number $N = 126$ is significant in this region also, as can be seen from the special stability of ^{208}Pb ($Z = 82$, $N = 126$), a doubly magic spherically symmetrical nucleus. This shell theory has progressed through many improvements, to the point where the potentials of single nucleons in a deformed or nonspherical nuclear field can be calculated, using the now widely accepted method of the Swedish physicist, Sven

Gösta Nilsson, the so-called "Nilsson orbitals."

The well-known charged liquid-drop model of the nucleus, which was so effective in developing the theory of neutron induced nuclear fission, also has been developed further to help in predicting stabilities of heavy nuclei toward decay by spontaneous fission and other modes. In the extension of the liquid-drop model formulated by W. D. Myers and W. J. Swiatecki, the potential energy of the nucleus is a function of neutron number, atomic number, and nuclear shape, and a shell correction is applied when the shape approaches sphericity. The semi-empirical mathematical expressions which describe or are related to this model can be used to calculate ground state masses and equilibrium deformations of hypothetical nuclei as functions of N and Z, as well as the barrier energies and related half-lives for spontaneous fission. Most isotopes, including those of the lanthanide and actinide elements, have more or less deformed (such as ellipsoidal) shapes, but those in the immediate vicinity of closed nucleon shells have spherical shapes. In the absence of closed shells, all superheavy elements would decay by spontaneous fission with half lives so short as to preclude their production and observation.

Recently, exciting theoretical calculations, made by scientists from many countries, including S. G. Nilsson, V. M. Strutinskii, C. Gustafson, S. A. E. Johansson, H. Medner, W. J. Swiatecki, F. A. Gareev, and P. A. Seegar, have used the liquid-drop model with shell corrections taken from Nilsson orbitals—using extrapolations of known parameters from the lanthanide and actinide regions—to establish the existence of possible closed nucleon shells in the region of superheavy elements. The survival of such nuclei from decay by spontaneous fission means that their rate of decay by alpha- and beta-particle emission becomes important; and therefore estimates of stability toward these modes of decay have also been made by Nilsson and his co-worker, Chin-Fu Tsang, at Berkeley.

Such theoretical considerations suggest the existence of closed nucleon shells at $Z = 114$ and $N = 184$, which exhibit great stability against decay by spontaneous fission. Fortunately, the nucleus with both $Z = 114$ and $N = 184$ is situated near the bottom of the potential energy valley of stability; that is, it is stable against decay by beta-particle emission and is doubly magic just like beta stable ^{208}Pb. This nucleus, $^{298}114$, is, of course, unstable toward decay by alpha-particle emission. A less stable closed shell has been computed in the past to exist at $Z = 126$ (or $Z = 124$) but this is no longer considered likely. The calculations indicate the existence of a closed shell at $N = 196$ and another possibility for a closed nucleon shell may occur at $Z = 164$. These calculations are based on the Nilsson model and must still be confirmed by calculations with other models such as the Woods-Saxon potential.

As I indicated earlier, by use of Mendeleev's nomenclature, element 114 might be termed "eka-lead" because it is a homolog of lead in the periodic table; likewise element 164 would be "eka-114" or "eka-eka-lead." It is especially interesting to note that the known and predicted centers of nuclear stability—atomic numbers 50, 82, 114, and 164—fall in the same column (Group IV) of the periodic table. This is curious because the
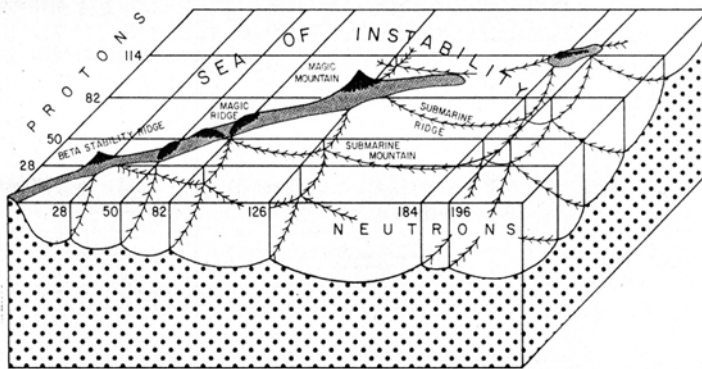
Figure 3. Known and predicted regions of nuclear stability, surrounded by a sea of instability.

principles that determine nuclear stability are apparently not related to those that determine electronic structure (which determines an element's position in the periodic table).

Enhancing the prospects for the actual synthesis and identification of superheavy nuclei is the fact that the doubly magic nucleus $^{298}114$ is actually the center of a rather large region of stability, with maximum stability against overall decay by all processes (spontaneous fission, alpha- and beta-particle emission) occurring at $Z = 110$, $N = 184$ (that is, $^{294}110$), and with decreasing stability being found as the edges of the region are reached. This is represented in an allegorical fashion in Figure 3, which I have prepared with the assistance of W. J. Swiatecki. (For the purposes of dramatization, this allegory reverses the usual method of display of centers of stability which are conventionally depicted as valleys in the potential energy surface.) The long mainland peninsula corresponds to the whole region of stable nuclei and the "island of stability" corresponds to the region of predicted relative stability among the superheavy elements. Surrounding these is a sea of instability. The grid lines (at sea level) represent magic numbers of protons (Z) or neutrons (N). The doubly magic region on the mainland peninsula at $Z = 82$, $N = 126$ is represented by a mountain. Other areas of enhanced stability on the peninsula due to single magic numbers are shown as ridges. The region between $Z = 83$ and $Z = 90$ is shown as being at a somewhat lower (but above sea level) elevation than the other parts of the peninsula of stability because of the appearance of alpha-particle emission as a predominant mode of decay here. Submerged ridges indicate that the isotopes are unstable, but are relatively more stable than the neighboring ones because of the increased stability afforded by closed nucleon shells. The "island of stability" has an irregular shape reflecting the doubly magic region (centered at $Z = 114$, $N = 184$) and the closed shell at $N = 196$, and the rate of decay is determined by competition between spontaneous fission and alpha- and beta-particle emission. This island also reflects the maximum in overall half-life predicted for the vicinity of $Z = 110$

Now that I have demonstrated where the region of enhanced nuclear stability lies by use of allegorical geography, I would like to present the results of the scientific computations which define the region in param-
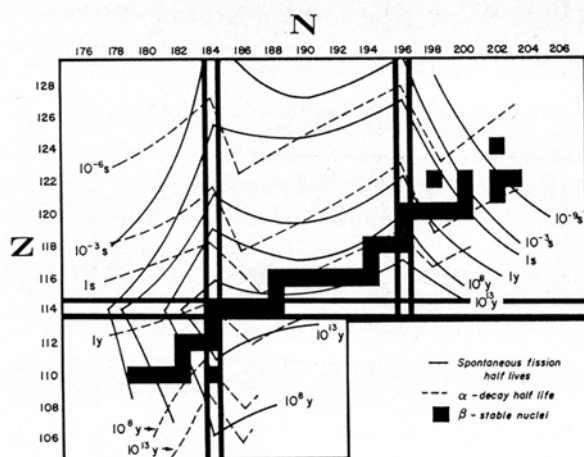
Figure 4. Half-life contours for region of exceptional nuclear stability.

eters of half-life values, atomic number, and neutron number.

If the nuclei in this island of stability are to be observed, they must have favorably slow rates of decay by alpha- and beta-particle emission, which are independent of and determined by different considerations than those that determine decay by spontaneous fission. Some nuclei are predicted to be stable against beta decay, some might have half-lives as long as 10^{13} yr for decay by alpha-particle emission and some perhaps 10^{16} yr for decay by spontaneous fission, although no specific nucleus would exhibit all of these favorable properties simultaneously! The actual trends predicted by Tsang for spontaneous fission, alpha- and beta-half-life values and for beta stability in the region of interest are shown in Figure 4. Quantitatively, the numbers may be in error by orders of magnitude, but even if the actual values are lower than indicated by many factors of ten, any produced nuclei that have the appropriate atomic number and mass number should still be observable. The doubly magic nucleus $Z = 114$, $N = 184$, for example, is predicted to have a spontaneous fission half-life of 10^{16} yr and an alpha-
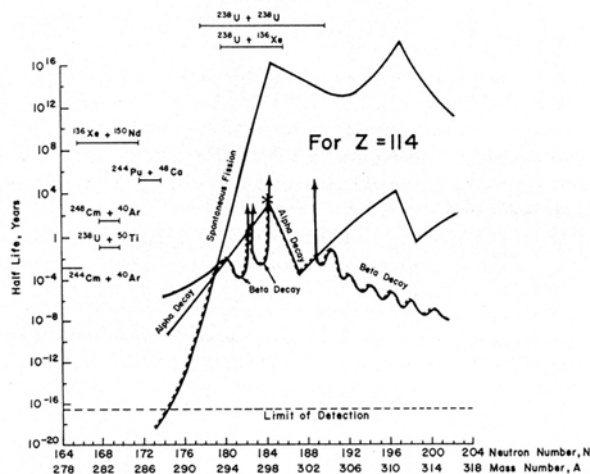
Figure 5. Cross section plot from Figure 4 for constant $Z = 114$, showing the variation of half-life with increasing N for spontaneous fission and alpha and beta decay. The dotted line represents the net or effective half-life. Bars labelled with nuclear reactions represent types of nuclear reactions available for production of superheavy isotopes.

decay half-life of greater than 1 yr. My own estimate is that the alpha-decay half-life may approach 10^3 yr. This nucleus should be stable against beta-particle decay. Half-lives of the order of seconds, even down to nanoseconds, are readily discernible with modern instrumental methods used in detection of new elements, so there is quite a margin of allowable error in the use of the predictions to select a region for experimental study. Also, the spread of predicted half-lives means that, if the island of stability can be entered experimentally, the investigator should be rewarded by the production of an isotope (or isotopes) with half-life suitable for detection.

Since the data that have been accumulated by Tsang and presented in Figure 4 are somewhat difficult to assess in a concise manner, I have chosen to narrow the discussion of the region of stability by taking two cross sections of the region, one for $Z = 114$ constant (Fig. 5) and the other for $N = 184$ constant (Fig. 6), taking into account data from Nilsson, Tsang, Swiatecki, and
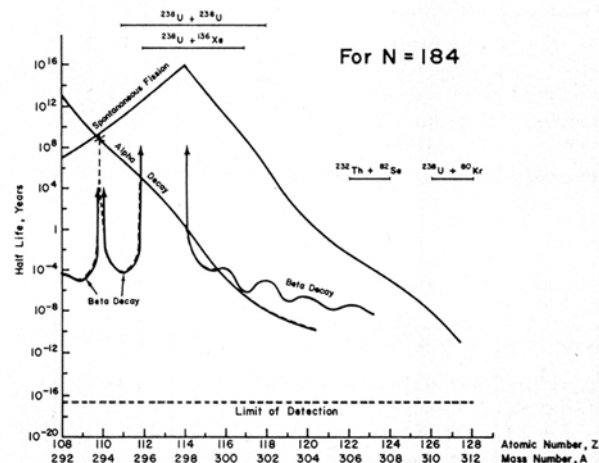

Figure 6. Cross section plot from Figure 4 for constant $N = 184$ showing variation of half-life with increasing Z.

others, and using some of my own estimates for alpha and beta stability.

In Figure 5, half-life values for spontaneous fission, alpha- and beta-particle decay are plotted as functions of neutron number. The dotted line depicts the net or effective half-life for isotopes of element 114, since it follows the controlling mode of decay (the lowest of the three lines representing the three modes of decay). It can be seen that maximum stability, i.e., a half-life of about 10^3 yr, occurs at $N = 184$, which corresponds to the isotope $^{298}114$. However, a large region of neutron numbers (or mass numbers) lies above the limit of detection line, which is set at one nanosecond (3×10^{-17} yr). Note that $^{298}114$ should be beta stable, should have a negligible spontaneous fission decay rate, and its rate of decay should be determined by alpha-particle emission with the half-life shown.

By holding the neutron number constant at 184 and varying the atomic number, Figure 6 results. This figure demonstrates that theoretically it should be possible to observe many new elements with half-lives above the limit of detection.

The bars labelled with nuclear reactions in Figures 5 and 6 represent the types of nuclear reactions available

for the production of superheavy isotopes and I shall have more to say about this later.

Search for Superheavy Elements in Nature

It appears that maximum stability should be found at $Z = 110$ and $N = 184$ ($^{294}110$), an isotope for which the half-life might be as long as 10^8 yr for decay by both spontaneous fission and alpha-particle emission and which is possibly stable toward decay by beta-particle emission. A half-life of a few times 10^8 yr is sufficiently long to allow an isotope to survive and be still present on earth (for example, ^{235}U has a half-life of 7×10^8 yr), provided it was initially present as the result of the cosmic nuclear reactions that led to the creation of the solar system. Consultation of Figures 1 and 2 will remind us that element 110 is a homolog of platinum and should have chemical properties similar to that precious metal.

Therefore, searches have been made for element 110 (and neighboring elements) in naturally occurring platinum and other metals and ores by workers at the Lawrence Radiation Laboratory in Berkeley and Livermore, at Argonne National Laboratory, at Oak Ridge National Laboratory, and at the Joint Institute for Nuclear Research in Dubna, USSR. The efforts of S. G. Thompson and associates at Berkeley are perhaps typical in this regard and a brief description of their work will serve as an example of the complexities involved. They have looked for all types of radioactive decay and, in addition, on the unlikely assumption that such elements might be so stable as to exhibit no detectable radioactivity, they also employed very sensitive methods of nuclear analysis, such as X-ray fluorescence and mass spectrometry, and all types of activation analysis in their search. So far, they have found no indications of the existence of such elements in nature and tentatively conclude that if present, the concentration must be less than one part per ten billion in platinum found in placer ores.

Flerov and coworkers, at the Dubna Laboratory in the Soviet Union, feel that they have detected spontaneous fission in lead samples that they suggest might possibly be due to the presence of eka-lead (element 114); this would be inconsistent with the predictions summarized in Figure 5 which indicate that the half-lives of the isotopes of element 114 are too short to allow their continued existence in nature today. Of course, the results of Flerov et al., if confirmed, could prove that the half-lives for alpha-particle decay have been underestimated. On the basis that the alpha half-lives may be substantially longer than indicated, consideration is being given to searches in nature for elements 111 (eka-gold), 112 (eka-mercury), and especially 113 (eka-thallium). Also, because elements with half-lives as short as 10^5 yr could be present in cosmic rays, a search for superheavy elements is being made in this possible source.

Methods for Synthesis of Superheavy Elements

From the foregoing account it is apparent that we now see a promised land of superheavy elements, a far out island of nuclear stability, and we must devise means to reach it. The indications are that it can be reached by bombarding target nuclei with sufficiently energetic heavy ion projectiles, and only by this method.

But there are many experimental difficulties to be overcome. The yield of the desired product nuclei is predicted to be very small because the overwhelming proportion of the nuclear reactions leads to the fission reaction rather than synthesis through amalgamation of the projectile and target nuclei. The presently available target nuclei and projectiles lead to neutron deficient nuclei that lie outside the perimeters of the island of stability. Projectile ions that will soon be available with the required energy should lead to nuclei just barely within the island and therefore nuclei with half-lives so short as to make them difficult to detect. To reach the center of the island of stability may require the construction of new accelerators or the modification of present accelerators in order to furnish the unusual heavy ions that may be required at the necessary intensity and energy. The leading accelerators being devoted to this research are the HILAC in Berkeley and the cyclotrons in the Dubna Laboratory in the Soviet Union. These are already being modified to accommodate experiments of the type that will be needed to reach the island of stability.

The following is an example of a nuclear reaction that is relatively easy to produce

$$^{248}_{96}\mathrm{Cm} + ^{40}_{18}\mathrm{Ar} \rightarrow \, ^{284}114 + 4_0n^1$$
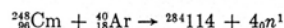
The mass number 284 shown is only illustrative, since a range of mass numbers can be expected as the number of neutrons emitted varies. The curium target and argon projectile at the required intensity and energy are available. The desired atomic number (Z) of 114 is achieved, but the neutron number (N) is only $284 - 114 = 170$, apparently placing the nucleus in the sea of spontaneous fission and making it unobservable. This is demonstrated graphically in Figure 5. This reaction has been attempted at Berkeley, but no identifiable products have been formed.

Therefore, a nucleus with additional neutrons is required to place it even at the edge of the island of stability. About the best that might be done in the near future is the reaction

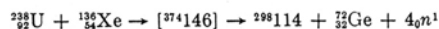$$^{244}_{94}\mathrm{Pu} + ^{48}_{20}\mathrm{Ca} \rightarrow \, ^{288}114 + 4_0n^1$$
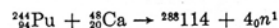
producing a nucleus with 174 neutrons which is at the edge of the predicted island of stability (see Fig. 5). This reaction uses ingredients which are relatively rich in neutrons. The isotope ^{244}Pu is a relatively rare material, but it will soon be available in sufficient quantity as the result of intensive neutron irradiation of ^{239}Pu and its transmutation products. The projectile isotope, ^{48}Ca, presents other problems; it occurs in nature only to the extent of less than 0.2 percent of the calcium isotopes. Therefore, it must be enriched by mass spectrographic techniques to a much higher proportion if it is to be an effective reactant. And it is difficult to vaporize and ionize so that it can be accelerated, although new devices for accomplishing this are being designed and tested. Other neutron-rich isotopes of interest as projectiles include ^{64}Ni and ^{50}Ti (again see Fig. 5), but they present some of the same problems as ^{48}Ca.

Even reactions with heavier ions, of a type where no neutrons are emitted, such as

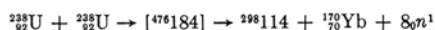$$^{136}_{54}\text{Xe} + ^{150}_{60}\text{Nd} \rightarrow {}^{286}114$$
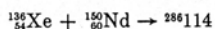
produce nuclei that are deficient in neutrons and therefore do not provide an entree into the island of stability (see Fig. 5). More likely, of course, is the production of isotopes of element 114 with even lower mass numbers, corresponding to the emission of neutrons in the synthesis reaction. However, acceleration in quantity of ions as heavy as xenon is not possible in existing accelerators. It is possible to write reactions with such intermediate heavy ions that suggest the production of isotopes such as the desired $^{298}114$, but they are of a type not expected to occur to any appreciable extent.

Perhaps the only way to provide the required ratio of neutrons to protons will be to use as ingredients nuclei like the very neutron-rich ^{238}U nuclei in reactions such as

$$^{238}_{92}\text{U} + ^{136}_{54}\text{Xe} \rightarrow [^{374}146] \rightarrow {}^{298}114 + ^{72}_{32}\text{Ge} + 4_0n^1$$

or

$$^{238}_{92}\text{U} + ^{238}_{92}\text{U} \rightarrow [^{476}184] \rightarrow {}^{298}114 + ^{170}_{70}\text{Yb} + 8_0n^1$$
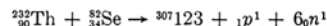
Probably the best prospect is the second of these reactions because this makes the most neutrons available. The theory here is that if an unstable, excited nucleus—and maybe "nucleus" isn't even a good descriptive term in this case—could be produced by the interaction or "fusion" of a uranium ion with uranium (or by a xenon ion with uranium), it would promptly decay by "fission" to products which were relatively stable. Actually, what is a more probable course of events would be the direct transfer of nucleons between the two reacting nuclei, with the product nuclei being formed directly and with no intermediate heavy nucleus being formed at all. Of course, a whole spectrum of products would be formed, but among them might be the desired superheavy elements. To accomplish the uranium-uranium reaction, uranium ions accelerated to about 1.5×10^9 eV would be required. Each ion must be multiply charged to a high degree by the removal of orbital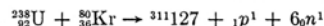 electrons in order to enable it to be accelerated in a reasonably sized machine. No machines are presently available, either in the United States or the Soviet Union, which are capable of accelerating such heavy ions to energies sufficient to overcome the Coulomb repulsion barrier. These machines can be built, or present machines can be modified, however, with essentially no limit on the atomic number of the accelerated ions.

To summarize where we are and where we hope to be, consultation of Figure 5 will show that bars are superimposed which depict the product isotopes which might be obtained from the various nuclear reactions that I have just discussed. The bars apply only to the horizontal scale; their location on the vertical axis is arbitrary and has no significance. It can be seen in Figure 5 that nuclear reactions which are now (or soon will be) achievable fail to reach the region of interest because insufficient numbers of neutrons reside in the product nuclei. Only the ^{238}U-^{238}U reaction, or possibly the ^{238}U-^{136}Xe reaction, provides enough neutrons to reach the center of the island of stability.

Likewise, in Figure 6, nuclear reactions such as

$$^{232}_{90}\text{Th} + ^{82}_{34}\text{Se} \rightarrow {}^{307}123 + {}_1p^1 + 6_0n^1$$

and

$$^{238}_{92}\text{U} + ^{80}_{36}\text{Kr} \rightarrow {}^{311}127 + {}_1p^1 + 6_0n^1$$

can be postulated which have the required neutron number of 184 in the product isotopes, but the number of protons, or the atomic number, is too large for the region of maximum stability. Again, the U-U or U-Xe types of reactions appear the most suitable.

If the superheavy elements can be produced and are as stable as predictions indicate, proof of their existence will require the use of chemical as well as nuclear detection techniques. The chemical separations must be designed on the basis of some guidance as to the nature of the element sought, and thus predictions of the electronic structure and chemistry of the new elements may well be imperative.

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It does not take much digging into history to see that throughout the ages there have always been a good number of people determined to doubt man's potential for progress. Often when it came to specific attempts at motivation or looking beyond the then known boundaries of man, there were those who suffered from what Arthur C. Clarke has called "a failure of nerve" or "a failure of imagination." Many others have sought to hold back innovation because of special economic interest. And a large number have fought change because of fear related to ignorance and misunderstanding.

...Commencement Address by Dr. Glenn T. Seaborg at Georgia Institute of Technology, Atlanta, Georgia, June 14, 1969.

Reference:

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for classroom use in Pima Community College chemistry courses.

Additional References

Additional references on the transuranium elements can be found on the Journal of Chemical Education website.

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