

Superheavy Elements

Attempts to discover elements with about 114 protons have gained momentum with the use of new accelerators.

S. G. Thompson and C. F. Tsang

Since 1965 there has been considerable interest among nuclear physicists and chemists in the possibility of discovering superheavy elements (elements with about 114 protons). Extensive experimental efforts have been made in the past 4 years to detect them in nature, but up to this time the results have been negative. Now various groups are attempting to make these superheavy elements in heavy-ion nuclear reactions, but the results so far are inconclusive. With the new Superhilac (super heavy-ion linear accelerator) being completed at Berkeley, a further major effort will soon be made to synthesize these superheavy elements.

In this article we summarize recent efforts and indicate some future possibilities. We shall begin with a general discussion of the developments leading to the expectations concerning superheavy elements. The theoretically predicted nuclear and chemical properties of these elements will be presented. Then we will summarize the efforts made in the search for these elements in nature by many groups in the United States and Europe. The possibility of producing these elements by neutron-capture and heavy-ion reactions will then be discussed, and some as-yet-unanswered problems associated with these methods will be pointed out. In

the concluding section, some current thoughts will be given on various aspects of a new field of research in which "superheavies" are only a part. We will attempt to impart some feelings regarding the significance of the push into the previously inaccessible domains. Highly technical details will not be discussed, and complete and unbiased referencing has not been attempted. For those who wish to make a study in greater depth, a number of excellent review articles (1, 2) are currently available.

Background

Superheavy elements are those elements that lie somewhat beyond the end of the present periodic table (Fig. 1) (3). Interest at present is focused on a region of isotopes—centered at proton number $Z = 114$ and neutron number $N = 184$ —expected to have special stability. The nucleus ${}_{114}^{298}\text{184}$ (signifying $Z = 114$, nuclear mass number $A = 298$, and $N = 184$) is expected to be extremely stable because the closing of both a proton shell and a neutron shell occurs at this location. Such shell closures, sometimes referred to as "magic numbers," are somewhat analogous to the closing of electronic shells in atoms that give extra chemical stability to certain elements such as the rare gases.

About 90 elements are found in na-

ture, and 15 more have been made artificially in the past 30 years. The element $Z = 105$ and $A = 262$ with the largest proton number is so unstable that it can only be produced in extremely small amounts (4), and it disappears in a few minutes by radioactive decay. These known elements form a peninsula in a plane of proton and neutron numbers (Fig. 2), surrounded on three sides by a "sea of instability."

In trying to extend the periodic table still further, it is well to understand the basic reason for the limited number of elements (5): Why are there now 105 elements rather than 2 or 3, say, or 2000 or 3000? The underlying physics responsible for the limited extent of the periodic table is the competition between the cohesive nuclear forces and the disruptive electrostatic forces due to the protons. The limit of the periodic table at $Z \approx 105$ is set by the process of nuclear fission, which takes place when electrostatic repulsion between protons overcomes nuclear cohesion.

It has been recognized for some time that this limit to the periodic table, set by electrostatic repulsion, could be extended somewhat by nuclear shell effects. Thus the presence of a closed shell of protons or neutrons, or preferably both, beyond the end of the periodic table would provide extra binding and extra stability to the nucleus (6). With suitable techniques it might then be possible to reach an island of superheavy nuclei, centered about the magic nucleus (one with a closed shell of neutrons or protons), with relatively long half-lives against fission.

No progress was made in this direction for several years, principally because it was assumed that the next closed proton shell, that is, the next proton magic number, would be at $Z = 126$, in analogy with the known neutron magic number $N = 126$; $Z = 126$ was too far beyond the present periodic table to be reached with any kind of nuclear reaction available at that time.

The picture began to change in 1966 when it was pointed out (7) that $Z = 114$, rather than $Z = 126$, was the next magic number. The reason the neutron

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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	58 Hf	59 Ta	60 W	61 Re	62 Os	63 Ir	64 Pt	65 Au	66 Hg	67 Tl	68 Pb	69 Bi	70 Po	71 At	72 Rn
87 Fr	88 Ra	89 Ac	104	105	(106)	(107)	(108)	(109)	(110)	(111)	(112)	(113)	(114)	(115)	(116)	(117)	(118)
(119) (120) (121) (154) (155) (156) (157) (158) (159) (160) (161) (162) (163) (164) (165) (166) (167) (168)																	
Lanthanides																	
58 Ce		59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu			
Actinides																	
90 Th		91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr			
Superactinides																	
(122) (123) (124)															(153)		

and proton closed shells do not occur at the same numbers for heavy elements may be traced to the influence of the electrostatic energy, which plays an increasingly important role toward the end of the present periodic table.

calculations. A way was needed of making rather precise calculations of nuclear masses and of deformation energies so that decay modes and half-lives could be estimated. Two major approaches have been frequently applied to make such calculations. The first approach is based on a microscopic model in which the total energy is assumed to be the sum of the single-particle quantum mechanical energies. The second approach is based on a macroscopic model [the so-called liquid-drop model (10)] in which the total energy is considered to be mainly composed of a volume energy, a surface energy, and a coulomb energy. Microscopic models, for example, Nilsson's shell model [see (9)], while giving local changes in nuclear masses very well, are subject to large errors when they are used to predict the absolute values or general trends. On the other hand, the liquid-drop model gives the absolute magnitude and general trends very well but cannot be used to reproduce local fluctuations caused by shell effects.

A significant advance in the calculation of masses occurred with the merger of the shell model and the liquid-drop model. The shell effects or local fluctuations [of the order of a few million electron volts (Mev)] are extracted from the results of shell model calculations and combined with the liquid-drop binding energies (about 2000 Mev for a heavy nucleus). The philosophy of this two-part approach was proposed by Swiatecki (11) and others, but the prescription for merging was developed by Strutinsky about 1966 (12) and came to be known as the Strutinsky method. The physical basis of the method is still under extensive investigation by various groups (13, 14), but the method itself has been successfully applied to a host of phenomena. Besides reproducing experimental nuclear masses (15, 16) to within 2 Mev, the method also makes it possible to explain the existence of a large number of fission isomers (17) and provides a basis for the quantitative understanding of asymmetric fission (18) (that is, the tendency of a heavy nucleus to split into two unequal rather than equal parts), which has been one of the outstanding problems in fission for more than 30 years.

Strutinsky's method was then employed by Nilsson, Tsang, and their co-workers (15) to make the first comprehensive predictions of the properties of superheavy nuclei. These results, which became available in 1968, indi-

cated that some superheavies might have half-lives long enough for them to exist in nature and immediately "triggered" experimental searches for them at Berkeley and elsewhere, as will be discussed later. Detailed calculations were also made by Strutinsky and his co-workers, and several other groups (14, 19, 20), the most recent of which are those of Nix and his associates (20) which will be discussed in more detail below. It is interesting to note the international nature of the theoretical efforts. The nationalities of the many investigators include: American, Chinese, Danish, German, Polish, Soviet, Swedish, and Swiss. Great caution should be exercised in considering the theoretical results. These calculations involve great uncertainties. Thus the prediction of a half-life of 10^9 years may be uncertain by a factor of 10^6 either way; that is, the half-life may well be anything between 10^3 and 10^{15} years.

Theoretical Predictions of Superheavy Nuclei

The above-mentioned calculations (20) indicate that the region centered around $Z = 114$ and $N = 184$ should be very stable. These nuclei form an island somewhat beyond the tip of the peninsula of known elements in a plane of proton and neutron numbers as shown in Fig. 2. Contours of total half-lives involving all major modes of decay, namely, spontaneous fission, alpha decay, and beta decay, are shown in Fig. 3. Note that the island centers around $Z = 110$ (instead of $Z = 114$) and $N = 184$. The shift from $Z = 114$ to $Z = 110$ is mainly due to the competition between spontaneous fission (where $Z = 114$ is the most stable) and alpha decay (where nuclei with lower Z are more stable).

The island may be divided into four regions: the top, where the dominant mode of decay is alpha-particle emission; the bottom, where the dominant mode of decay is beta decay; and the two regions on the sides, where the dominant decay is spontaneous fission. In the alpha-decay region the half-lives increase as Z decreases. These contours show a characteristic kink near the magic number. In the spontaneous-fission regions, the contours have a "diamond-like" shape centered around the magic numbers. (In this calculation, the neutron magic number is $N = 184$,

but the proton magic region extends from $Z = 114$ to $Z = 120$.) In the beta-decay region the half-lives decrease with decreasing Z and distance from the island of stability. The beta-stable nuclei form a belt extending diagonally across the island through the nucleus $Z = 110$ and $N = 184$. The longest-lived nucleus in the island appears to be $^{110}_{184}$ (or could be an adjacent odd A or odd Z nucleus), which has a half-life as long as 10^9 years. Such a half-life is nearly as long as the age of the solar system. If one considers only nuclei with half-lives of 1 minute or longer, one is confined to an island with Z between 106 and 116 and N between 174 and 192. These are the nuclei experimentalists are attempting to produce.

The results of the estimates concerning the properties of superheavy nuclei raised a number of new questions. Do these elements exist in nature? Could they have been formed by astrophysical processes during the formation of the solar system? Can they be produced in nuclear reactions, for example, by neutron capture or by heavy-ion reactions? What are their chemical properties? We shall first discuss their predicted chemical properties.

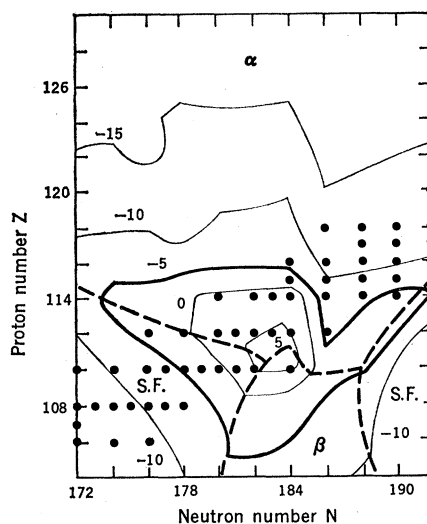


Fig. 3. Total half-lives for the decay of even Z -even N superheavy nuclei given as contours, which are labeled by the logarithm (to the base 10) of the half-lives in years. The points indicate nuclei that are calculated to be beta-stable. The heavy black line encloses a region of nuclei whose half-lives are longer than approximately 5 minutes. The broken lines separate regions whose dominant decay is by beta decay (β), alpha decay (α), or spontaneous fission (S.F.) as indicated [from Nix (1)]. [Courtesy Annual Reviews, Inc., Palo Alto, Calif.]

Predicted Chemical Properties

If the superheavy elements do become available, experimental studies of their chemical and physical properties will be a subject of considerable interest. The study of such superheavy nuclei and atoms will present a new frontier for nuclear and inorganic chemists. For example, the study of the chemical properties of the superheavy elements should give some indication of how far the periodic system of the elements can be extended and, at the same time, should shed new light on the electronic properties underlying the periodic system.

The expected properties of these elements had already received significant attention as early as 1958 when Seaborg (21) predicted properties of the elements up to $Z = 118$, eka-radon. These predictions in general have been borne out by more recent studies.

As a starting point for a discussion of the chemistry, let us consider some experimental information concerning the most recently discovered elements. Element 103 was confirmed (22) as being the last member of the actinide series in which the $5f$ electron subshell is filled. Element 104 was found (23) to resemble hafnium in its chemical properties, confirming the expectation that the $6d$ electron subshell is being filled. Experimental studies of the chemistry of element 105 have not yet been carried out.

Detailed predictions concerning the chemical properties of superheavy elements are not easy to make. Nevertheless, some progress in this direction has been made by the use of two different methods. The first is an extension of Mendeleev's method, in which the behavior of the well-known elements as a function of their chemical group and period is extrapolated into unknown regions. In the second, one studies the order in which the electrons fill their orbits by doing self-consistent field calculations for the electrons surrounding the nuclei, for example, by relativistic Hartree-Fock-Slater calculations. Such calculations have been carried out by several groups (24) at Los Alamos Scientific Laboratory, Oak Ridge National Laboratory, Northwestern University, the University of Frankfurt, and elsewhere. The predicted properties of some of the superheavy elements are shown in Table 1.

Certain effects that are negligible in the light elements become very im-

Table 1. Some physical and chemical properties of elements 110 through 115 according to Fricke and Waber (24).

Element	Electronic ground-state configuration	Chemical group	Most favorable oxidation state	First ionization potential (ev)	Metallic radius (Å)	Ionic radius (Å)	Density (g/cm ³)	Melting point (°C)	Boiling point (°C)	K α_1 x-ray energy* (kev)
110	6d ⁸ 7s ²	VIII	+ 6	9.4	1.4		27.4			157
111	6d ⁹ 7s ²	IB	+ 1, + 3	10.3	1.5		24.4			161
112	6d ¹⁰ 7s ²	IIB	+ 1, + 2	11.2	1.6		16.8			165
113	7p ¹ 7s ²	IIIA	+ 1	7.4	1.7	1.5	16	430	1130	169
114	7p ² 7s ²	IVA	+ 2	8.5	1.8	1.3	14	67	147	173
115	7p ³ 7s ²	VA	+ 1, + 3	5.9	1.9		13.5			177

* From (30, 34).

portant in superheavy atoms; for example, relativistic effects will be quite large, and the spin-orbit splitting of levels becomes a dominant feature. These relativistic effects are likely to produce unexpected chemistry in certain superheavy elements. The physical limit of the periodic system, as we know it, may occur approximately at $Z = 170$ (25). At about this point the inner electron shells might undergo a critical change because of the very large electrostatic field that exists in the atom. This change may, for example, involve the production of positrons.

The expected positions in the periodic table of the superheavy elements is indicated in Fig. 1. Elements 104 to 112 result from the filling of the 6d electron subshell, which makes them homologs of the elements hafnium through mercury. It is expected that for these elements the 7s electrons will be more strongly bound than the 6s electrons in hafnium through mercury, but the atomic radii of the superheavies should be slightly larger. The elements up to the middle of the series should tend to reach their maximum oxidation states, but near the end of the series the ionization energies should be very large and these elements should be good noble metals. The metallic state should be predominant.

Elements 113 through 118 are characterized by the filling of the 7p subshell and are thus homologous with the elements thallium through radon. Element 114 can be called eka-lead, if we use Mendeleev's terminology. These elements will tend to have lower oxidation states than their homologs. The calculations (24) indicate that element 115 will be monovalent, thus precluding the extrapolation of its properties from those of arsenic, antimony, and bismuth, which show oxidation states of only +3, +5, and -3. Element 115 therefore might exhibit a radically new chemistry differing

from that of other monovalent elements as much as the chemistries of rubidium and silver or cesium and thallium differ from each other. Elements 119 and 120 should be very similar to their homologs, francium and radium.

The elements in the vicinity of $Z = 120$ to 125 present another interesting problem. At about this point a new inner 5g transition series with a maximum of 18 electrons is expected to begin. However, it seems possible that the 5g and 6f subshells may be filled more or less simultaneously and it might be impossible to distinguish between the two subshells. If this occurred, it would give rise to a series of 32 elements, for which Seaborg proposes the name "superactinides."

The magnitude of the effort made to determine the chemical properties of superheavy elements will depend to a large extent on the number of elements produced and the range of half-lives and decay modes of the various isotopes. At Berkeley, where it is hoped the new elements can be made using the Superhilac, it is anticipated that there will be collaboration among the groups at the Lawrence Livermore Laboratory, the Argonne National Laboratory, the Oak Ridge National Laboratory, and the Los Alamos Scientific Laboratory in studying the chemistry. Preparations are under way to carry out many different experiments. For example, very efficient extraction chromatographic separations based on the behavior of homologous elements have been developed already by Horwitz and his associates at Argonne (26).

It is not within the scope of this article to discuss the detailed procedures that might be used to make separations and study chemical properties. However, many different types of separations based on homologous elements have been considered, some of

which, for example, are derived from an excellent series of monographs on radiochemical separations of the elements published by the National Academy of Sciences (Nuclear Science Series). A variety of separations are included, based on ion exchange, volatility, oxidation-reduction, solvent extraction, and precipitation methods.

The chemists who look forward to investigations of the chemical properties of the superheavies are faced with many difficulties. First of all, because the production of these new elements is likely to come about as a result of the heavy-ion reactions (discussed later), the yields are likely to be small and many of the half-lives will be very short. Second, deductions of chemical properties are likely to be made by application of tracer methods involving rapid separations of very small amounts (even a few atoms) of radioactive materials and comparisons with homologous tracer elements.

Finally, it should be emphasized that it may be essential to apply chemical methods to the identification of the atomic numbers of some of the new elements. Although observation of alpha-particle and spontaneous-fission events having higher energies than those associated with the decay of previously known isotopes would be an almost certain indicator of superheavies (20), such an indication, however, would not be sufficient to reveal the atomic number and identity of the element. Nor would measurements of the mass numbers of the nuclei in question be sufficient. An element assignment could be made on the basis of measurements of the energies of the characteristic x-rays, but, if the yields of the products formed in the reactions are small, it may be difficult or even impossible to employ this method successfully. Another method commonly used to make element assignments [for example, elements 102, 103, 104, and 105 (27)] is the observa-

tion of decay to daughters having well-known characteristics. But this method may not be useful for superheavies because, as indicated in Fig. 2, the decay chains are expected to be terminated by spontaneous fission in the "sea of stability." Thus methods of chemical separation would be necessary. Even in this case proof of the proton number may not be simple and straightforward. It might be necessary to decide on the basis of separations of tracer amounts of the element, without the benefit of prior knowledge about whether element 110, for example, is more like platinum than it is like iridium. On the other hand, if it were similar to mercury with respect to the volatility of its metallic state, the identification might not be so difficult. Such problems will be very challenging, especially if some of the elements exhibit unexpected behavior.

Early Experimental Work

The first attempts (28) to produce superheavy elements by means of heavy-ion reactions were carried out at Berkeley (29–31) in 1967 in response to the early suggestions of Myers and Swiatecki. Bombardments of Cm^{248} with Ar^{40} were carried out at the Berkeley heavy-ion linear accelerator, with the use of very sensitive apparatus for detecting spontaneous-fission events. The results were negative, in agreement with the results of the first comprehensive calculations by Nilsson *et al.* (15) which became available in 1968. The results of these calculations, however, stimulated an extensive search for superheavy elements in nature, as discussed in the following section.

Search for Superheavy Elements in Nature

The prediction by Nilsson *et al.* that the half-life of the nucleus 110^{294}_{184} (eka-platinum) should be in the neighborhood of 10^8 years suggested that small amounts of superheavy elements might be present in nature. The presence of these elements on the earth could have resulted from their formation along with the other elements at the time the earth was formed. If some of the nuclei have half-lives near 2×10^8 years, small fractions could have survived the period ($\sim 4.5 \times 10^9$ years) since the earth was formed.

Since it is possible that there are

large uncertainties in these estimates of half-lives and since odd A and odd Z -odd N nuclides generally exhibit retardation in decay by comparison with even Z -even N nuclei, the surviving nuclides could extend over several proton numbers, for example, 108 through 115. In addition, a search for very heavy particles in cosmic rays at high altitudes by Fowler *et al.* (32) indicates the presence of uranium and adjacent elements and even suggests a possible contribution from elements with Z greater than 100. Thus the possibility exists that superheavy nuclei might have been produced in more recent cosmic events (33), and nuclei with shorter half-lives (10^4 to 10^5 years) could conceivably have been deposited in small amounts on the surface of the earth.

A search for the new elements on the earth depends on suitable choices of the most promising minerals and ores containing elements having chemical properties most closely resembling those of the elements being sought. Therefore, certain assumptions had to be made about the chemical properties of the new elements (see the section on chemical properties). The search for eka-platinum (element 110) in nature was undertaken (30, 34, 35) on natural platinum ore containing significant amounts of the neighboring elements. This selection was made on the basis that the most prominent chemical characteristic of elements 108 to 114 is expected to be their predicted nobility, and one might expect them to be found with the noble metals. Even so, it is not certain that the behavior of the superheavy elements will be completely analogous to that of their homologs. Obviously a pure or purified metal might not be an ideal source since a high degree of selectivity in refining might eliminate homologous elements if they are not almost identical. In the search at Berkeley for element 110 in natural platinum ores a variety of techniques were used (30, 34), for example, the low-background counting of neutrons, gamma rays, and spontaneous fissions. The Berkeley group also used x-ray fluorescence, mass spectrometric, and activation analysis techniques. The results were negative and corresponded to a limit of less than 10^{-11} gram of superheavy element per gram of platinum.

The most sensitive methods employed to search for the new elements are those involving the detection of spontaneous-fission events. In this case

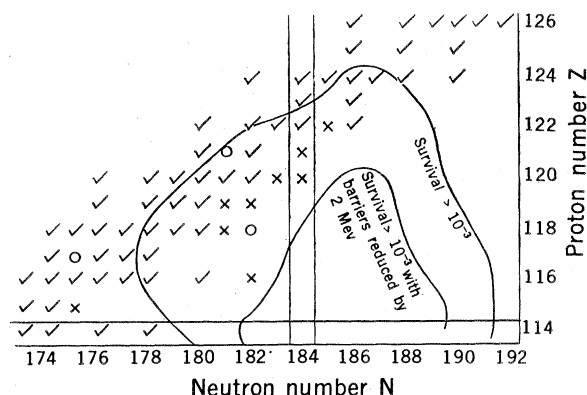
the assumption is made that the elements are not completely stable and that spontaneous fission always occurs in the termination of a decay chain. This condition seems to hold for all the theoretical calculations and is well illustrated by the results in Fig. 3. One can detect spontaneous-fission events either by observing fission tracks or by counting the neutrons emitted when spontaneous fission occurs.

The latter method has been employed by Grimm, Herrmann, and Schüssler (36), who used a He^3 -filled proportional-type neutron counter. They examined multikilogram samples of pure and ore-grade minerals. Samples were selected on the basis of both homologous chemical behavior and geochemical rules, and covered the range from eka-osmium to eka-bismuth. They found no evidence for superheavy nuclei.

The method of observing fission tracks has been used by Flerov and his collaborators (37), who reported results from spontaneous-fission measurements on lead-bearing samples—in particular, lead glass—which they felt could be explained as due to the presence of superheavy nuclei. These measurements were made by scanning plastic track detectors (Mylar foils each about 1 square meter in area) that had been in contact with lead foils for 100 days. Further investigations on samples of lead glass, including a fragment from an 18th-century glass vase, showed a number of spontaneous-fission events in excess of those expected from the small amounts of uranium and thorium present in the samples. However, similar experiments on other lead-bearing minerals were inconclusive. The results on samples of lead glass appear to have been confirmed by the fission events counted in proportional counters with a large area (1.96 square meters) (38). If we assume a half-life of 10^9 years for the spontaneously fissioning nuclei, the concentration would be $\sim 10^{-12}$ gram per gram of lead.

Price *et al.* (39) used a very sensitive method to obtain results which seem to be in conflict with those of Flerov and his associates. Price *et al.* searched for fission tracks accumulated over millions of years in ancient minerals (more than 10^8 years old). Their results for lead-bearing hardystone and gold-bearing quartz gave no evidence for the presence of superheavy elements, and concentration limits of less than 10^{-15} gram per gram of lead and less than 10^{-17} gram per gram of gold

Fig. 4. Available landing places near the island of stability in heavy-ion reactions. These are designated by $\checkmark$, $\times$, or $\circ$. Those designated by $\checkmark$ denote that naturally occurring targets such as U^{238} or Th^{232} can be used to reach the product indicated; the latter two symbols denote landing places that cannot be reached without Cm^{260} or Cf^{262} targets, respectively. The longer curve marks out the region where the probability of the compound nucleus (after it has been formed in its ground state well) surviving four successive neutron-fission competitions is expected to be in excess of 10^{-3} . This region will be decreased to the area indicated by the shorter curve if the calculated fission barrier is arbitrarily cut down by 2 Mev. [Figure prepared in collaboration with W. J. Swiatecki.]



were assigned. However, since the samples were of different composition and origin, it might be possible for superheavy elements to be present in the lead samples of Flerov and his associates and not in the lead minerals of Price *et al.* Price (40) and Lal and his co-workers (41) studied fission tracks in moon rock in search of the fission of superheavy nuclei, but the results were inconclusive.

Advantage can also be taken of the fact that the spontaneous fission of superheavy elements is expected to be different from the fission of well-known elements. Rather simple theoretical considerations strongly indicate that, when these new elements undergo fission, the fragments should have significantly higher energies and should involve the emission of a larger number of neutrons per fission—probably about ten, rather than about two as in the case of uranium (42). By measuring the number of neutrons emitted in each fission event one should be able to distinguish superheavy elements from other elements. This approach was employed by the Berkeley group (43), who have recently concluded an extensive search for superheavy elements in nature. They used a large liquid scintillator to measure the number of neutrons per fission event in large samples of minerals and ores. Their counter was located in a tunnel about 260 meters below the surface of the earth to minimize the influence of interfering cosmic radiation. More than 40 samples of ores, minerals, and rocks were examined, including manganese nodules, moon rocks, and large samples of gold and platinum in their natural states. The

selection of samples was also made to include the range of elements from eka-platinum to eka-bismuth. No evidence for superheavy elements was found in any of the samples. Groups at Oak Ridge (44) and Dubna, U.S.S.R. (45), are also employing He^3 counter systems to detect events in which large numbers of neutrons are emitted.

A number of other searches for superheavy elements have been made, but none has given conclusive evidence of their presence. Although the results up to now do not definitely rule out the presence of these elements in nature, the weight of evidence is such as to suggest strongly that they do not exist on the earth. Thus the question arises, How can we account for their absence? If the half-lives are much less than 2×10^8 years, they would have disappeared by radioactive decay during the 4.5×10^9 years since the earth was formed. On the other hand, if the half-lives are long enough, the conclusion would be that they probably were not formed during nucleogenesis in which the other elements were formed. Although some investigators have suggested that there should be no difficulty in producing the new elements in the neutron-capture process, an examination of some of the most reliable mass formulas indicates that there may be difficulties in binding neutrons to nuclei with such necessarily large neutron excess. Even if there were no difficulty in binding neutrons, neutron-induced fission or spontaneous fission might prevent the formation of the products of interest. Stability against fission is certain to be very small in the case of the intermediate nuclei in the formation process.

Production of Superheavy Elements in Neutron-Capture Reactions

Possible difficulties in producing the new elements in reactions associated with nucleogenesis have been indicated above, but these difficulties do not necessarily rule out the production of such elements by irradiation with neutrons of targets with high proton numbers on a slow time scale in, for example, the high flux isotope reactor at Oak Ridge. However, the heaviest nucleus produced so far by this means is Fm^{257} , and this same isotope is also the heaviest one produced to date in thermonuclear explosions on a faster time scale. In this case the fermium isotopes were produced by the rapid successive capture of about 20 neutrons in targets such as U^{238} . Therefore, these methods do not appear at all promising.

A possible means of circumventing the difficulties inherent in the production by neutron-capture processes in the extreme cases of fast or slow time scales has been suggested by Meldner (46). His proposal is to utilize capture of neutrons from thermonuclear explosions that would be controlled to allow some intermediate beta decays. This approach would tend to minimize some of the difficulties associated with the attempts described above but might be extremely difficult from a technical point of view.

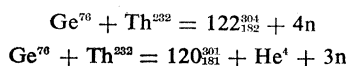
Production with Heavy Ions

Probably the most promising approach to the production of superheavy nuclei involves the use of heavy ions. One of the major differences between the processes used to make elements up to element 105 and those necessary for producing superheavy elements is that for the latter there is a gap of very unstable nuclei between the island and the peninsula (see Fig. 2). Thus it is impossible to go step by step to the island; it is apparently essential to make a big jump to the island by means of relatively heavier projectiles (such as Ca^{48} , Ge^{76} , and Kr^{86}). In this process the heavy ions are accelerated to a high energy and used to bombard a target nucleus such as Th^{232} or U^{238} . Hopefully, the projectile and target will fuse together, forming compound nuclei within the island of stability. Until recently only those projectiles up to Ar^{40} were available with sufficient energies to fuse with heavy targets, and

it was not possible to bring about a jump close to the center of the island with the use of Ar^{40} . But with the newer heavy-ion accelerators at Dubna, U.S.S.R., and Orsay, France, heavier projectiles are available. Even more intense beams of very heavy projectiles should become available soon at Berkeley, and also near the end of 1974 at the "universal linear accelerator" in Darmstadt, Germany.

In Fig. 4, we show the landing places (compound nuclei) close to the island of stability which can be reached by the target-projectile combinations with the use of known nuclei. Even with the available long-lived elements as projectiles, the center of the island cannot be reached unless we use special methods such as those suggested below. Furthermore, it does not seem advisable to use too heavy a projectile, because such a heavy projectile would overshoot the island and land in a region where the products are very unstable.

One of the most favorable target-projectile combinations, advocated by Swiatecki (47), after consideration of various effects that enter into heavy-ion reactions, is



Both final nuclei indicated would (hopefully) decay by successive alpha emission or electron capture toward the center of the island. Even with these reactions, certain difficulties are likely to be encountered, as discussed in a later section.

In a discussion of heavy-ion reactions it is necessary to recognize the need to accelerate heavy ions to a sufficiently high energy. Why not just mix two elements together and extract a product of much higher atomic number? The electrostatic energy, which, as we have seen before, becomes increasingly important at the end of the present periodic table, prevents fusion. The very large positive charges in the target and projectile nuclei prevent them from coming within the very small distances required to make them touch and fuse together. This fusion distance is about 10^{-12} centimeter. In order to make even a relatively light ion, such as argon, fuse with uranium, the argon must have its energy raised to about 200 Mev. It is rather difficult to achieve such high energies. One reason is that all accelerators require projectile atoms to have a net charge (thus the projectiles must be ions). Thus elec-

trons must be removed from the atoms in "ion sources." The heavier the atom, the larger is the number of electrons that have to be removed for acceleration to high energies. This stripping of electrons presents considerable difficulties, and, in some cases, a procedure of stripping in successive stages is used.

One of the main difficulties in reaching the island of stability by heavy-ion reactions is that neither the projectiles nor the targets have a sufficient number of neutrons to form a compound nucleus near the center of the island. Four methods may be considered for dealing with this problem.

The first is by secondary reactions after multinucleon transfer. It has been observed that, in the heavy-ion collision, a few neutrons may be transferred from the target to the projectile or protons may be transferred from the projectile to the target (48). In both cases the result is that the projectile becomes more neutron-rich. These neutron-enriched projectiles may then undergo a second reaction with the target to form superheavy nuclei. Estimates of the yields to be expected from such a process indicate there is some chance of success (49).

It is well known that the fission of heavy nuclei such as Cf^{252} yields neutron-rich fission fragments which are then available as projectiles with enough neutrons to make favorable heavy-ion reactions. In this method it is difficult to accelerate these fission fragments to a high enough energy to cause compound-nucleus formation when the fragments bombard a target (50). The intensity of the beam of accelerated fission fragments may be too small to cause observable heavy-ion reaction products. One way to get around this difficulty is to accelerate, for example, the nucleus U^{238} to high energies first and then to allow the U^{238} particles to collide with deuterons or helium, in which case sufficiently energetic fission fragments would be produced.

Another method suggested by Hyde for producing very-neutron-rich projectiles involves the use of protons in the energy range of 10^8 Mev, which have about 100 times the energy required for ordinary nuclear reactions. The collision of the high-energy protons with heavy target nuclei causes the target to fragment into pieces, some of which would be very neutron-rich and might have sufficient energies to produce heavy-ion reactions. So far this method

has not been successful, despite some initially encouraging results (51).

Another suggested method involves accelerating very heavy nuclei such as uranium and using them to bombard another uranium nucleus to give a total $Z = 184$ and $A = 476$ (52). This combination is expected to be very unstable and to divide into smaller pieces. One of the pieces might be right in the island of stability. However, very little is known about this process, and possibly, in such a violent reaction, only many small pieces would be produced.

The methods outlined above are necessarily very speculative at this time; further studies and quantitative estimates of production probabilities are needed. However, they may all be technically possible in the next few years.

Physical Methods of Detecting

Products in Heavy-Ion Reactions

The predicted half-lives of nuclei near the center of the island of stability are very long. Thus radiochemical methods might be useful for the separation and identification of such nuclei. However, one must be prepared to allow for the large uncertainties inherent in these predictions. It may be that the theory overestimates half-lives by a large factor, and one may be confronted with the detection of products with very short half-lives. Further, most combinations of target and projectile (see Fig. 4) will yield products some distance away from the center of the island of stability, with very short half-lives. When the half-lives are below about 10 seconds, it becomes difficult, despite great improvements in recent years, to apply radiochemical methods, and the application of physical methods becomes necessary.

Fortunately, a large number of rapid and sensitive methods of detecting and identifying the products of heavy-ion reactions had already been developed in extending the periodic table upward through the transuranium elements 103, 104, and 105. Recent developments in fast electronics, solid-state counters, and reliable high-speed computers have made it possible to meet the stringent technical requirements necessary for the study of very short half-lives.

In many of the physical methods of detection, one takes advantage of one of the characteristics of the heavy-ion reactions themselves: When an energetic heavy ion strikes a target nucleus,

Table 2. Previous attempts to make superheavy elements by heavy-ion reactions (σ , cross section; τ , half-life).

Investigators	Reference	Reaction	Results
Sikkeland	(28)	$^{18}\text{Ar}^{40} + ^{92}\text{U}^{238} \rightarrow \text{fission fragments}$	Fission fragment from $^{110}\text{Zn}^{278}$ (?)
Thompson; Nurmia <i>et al.</i>	(30, 31)	$^{18}\text{Ar}^{40} + ^{90}\text{Cm}^{248} \rightarrow ^{114}\text{Zn}^{288-x} + xn$	$\sigma < 5 \times 10^{-32} \text{ cm}^2$ for $\tau > 10^{-9}$ second
Oganessian <i>et al.</i>	(55)	$^{54}\text{Xe}^{136} + ^{92}\text{U}^{238} \rightarrow \text{fission fragments}$	Fission fragments from $^{146}\text{Ba}^{374}$ (?)
Flerov <i>et al.</i>	(56)	$^{30}\text{Zn}^{66} + ^{92}\text{U}^{238} \rightarrow ^{122}\text{Zn}^{304-x} + xn$	$\sigma < 5 \times 10^{-30} \text{ cm}^2$ for $\tau > 10^{-8}$ second
Bimbot <i>et al.</i>	(54)	$^{36}\text{Kr}^{84} + ^{90}\text{Th}^{232} \rightarrow ^{126}\text{Zn}^{316-x} + xn$	High-energy α from $^{126}\text{Zn}^{316-x}$ (?)

the resulting nucleus is driven forward with relatively high energy as a consequence of the conservation of momentum. Such a high-energy product is able to escape from the target and can be transported rapidly by various means to detectors that record the properties of the subsequent radioactive decay (27). Commonly used methods of transporting such "recoil atoms" to counters involve rapidly flowing gases such as helium. Rapidly moving drums or tapes are also used to collect and transport the recoil atoms to locations near counters that can measure their radiations. Other methods involve bending the recoil atoms in a magnetic field (to separate out the products of interest) and measuring their velocities and energies simultaneously. By such means one can determine the masses of the recoil nuclei. Means are also available for observing the properties of daughters of the radioactive decay and for measuring the energies associated with the decay. It has been predicted that most of the superheavy nuclei will decay by successive alpha decays before undergoing spontaneous fission (20). The alpha energies are expected to be much larger than those from the decay of previously known nuclei. Thus a measurement of these successive high-energy alpha decays will be characteristic of the presence of superheavy nuclei. These particular methods are only a few examples of some of the approaches being taken to solve the problem.

Problems Associated with Heavy-Ion Reactions

Even with the most favorable target-projectile combinations, there are formidable difficulties, which are summarized as follows:

High excitation energies. When a heavy-ion projectile fuses with the target nucleus, the resulting compound nucleus has a large excitation energy. Calculations by Moretto (53) show that the very shell effects responsible for the stability of the superheavy nuclei (and,

in fact, for the existence of the island of stability) will be destroyed as the excitation energy is increased. Then the stability against fission that comes from the shell effect is lost. The excitation energy can be reduced only by the emission of several neutrons or charged particles. Thus, survival at such high excitation energies becomes a question of the competition between fission and neutron or charged-particle emission. If neutron emission is sufficiently probable, then the superheavy nuclei (minus the number of neutrons emitted) may survive. If the probability of fission is sufficiently large, then superheavy nuclei will not survive. In typical reactions the emission of four neutrons should be sufficient to reduce the excitation energy to a safe level. In Fig. 4 we show rough estimates for the survival probabilities due to these competing effects and indicate the best landing areas in the island of stability. The optimum combination, $\text{Ge}^{76} + \text{Th}^{232}$, that Swiatecki has proposed should have a survival probability of approximately 1:10,000. That is to say, the yield of the final product is expected to be reduced by the factor 10^4 because of fission competition.

Angular momentum effects. When a heavy ion collides with a target nucleus, rotational angular momentum is necessarily introduced. The centrifugal forces which arise make the system less stable. Some estimates of the result of this effect have been made which indicate that the expected yields of superheavy products should be further reduced by factors ranging from 10 to 10^3 .

Fusion probability. The largest uncertainty of all has to do with the probabilities for the projectiles and targets to fuse together (47). It is not sufficient for the projectile and the target nuclei to merely come in contact with each other: an extra push is necessary to force them to fuse together. The probability that they can be made to fuse together into a final spherical compound nucleus involves not only the nuclear inertia, which acts against the push, but also the viscosity of the flow of nuclear matter, which is

a dissipative effect converting the pushing energy into useless excitation energy. Good estimates of nuclear inertias and viscosities have not been made so far; these are important gaps in our knowledge of nuclear properties.

Present Status

Small beams of energetic krypton ions have been available at Orsay, and zinc and xenon projectiles have been available at Dubna. To date attempts by both the French (54) and the Soviet (55, 56) groups (summarized in Table 2 together with previous efforts) to produce observable amounts of superheavy nuclei have not been successful. However, a new heavy-ion linear accelerator is near completion in Berkeley that is expected to produce heavy-ion beams with much larger intensities than either the Orsay or the Dubna installation. This accelerator will make possible a major assault on the production of superheavy elements. It is a completely open question whether it will indeed be possible to synthesize these elements by heavy-ion reactions, and scientists all over the world are anxiously awaiting the outcome. Extensive searches for these elements in nature have been carried out, and it seems that the prospects for finding them in terrestrial materials are greatly diminished by the negative results obtained so far. There is still a possibility that superheavy elements might be found in cosmic radiation. The results of recent searches have not been conclusive.

If the superheavy elements are discovered, it will be the crowning success of an international effort of many years with contributions being made by many workers from many parts of the world. We would then be facing the happy prospect of confronting the comprehensive theoretical map of the island of superheavy elements with new experimental data. The confrontation will provide a new testing ground for our understanding of the chemistry of the elements and the physics of the nucleus. The possible existence of superheavy

elements on the earth in some time past or in supernovas would also have impact on other fields, such as geology and astrophysics. Practical and useful applications would be forthcoming eventually, as is always the case with basic research, although in most cases definite predictions of the direction and nature of the applications cannot be made.

On the other hand, if we were not able to produce the superheavy nuclei either because their half-lives are too short or because of difficulties associated with the fusion of projectile and target, there are still many exciting studies that can be made. These include studies of the collisions between target and projectile nuclei and of the fragments and radiations emitted as a result of the collisions. Thus information will be gained about the conditions prevailing during the very brief time when the target and projectile nuclei are together. A study of such transient systems may extend our present knowledge of the nucleus in two ways. First, these systems may have a wide variety of shapes, such as asymmetric dumbbells, triaxial ellipsoids, and cylindrical shapes. Effects of large centrifugal forces on these shapes may also be studied from observation of off-center collisions. Second, these systems are interesting because they have proton numbers up to about 200 and mass numbers up to about 500, well beyond our present periodic table. We will be confronted with the most intense electric fields occurring anywhere in the universe, specifically, the fields in the close vicinity of such heavy systems. In the neighborhood of $Z = 170$, a critical condition might occur. In this case quantum electrodynamics has to be applied to the understanding of the observed phenomena. The result would be an unusually intimate interaction between nuclear physics and quantum electrodynamics in which the theory of matter and radiation will undergo testing under unique conditions.

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