

Lawrence Berkeley National Laboratory

Recent Work

Title

SUPERHEAVY ELEMENTS - A CROSSROADS

Permalink

<https://escholarship.org/uc/item/6hg0t2d4>

Author

Seaborg, G.T.

Publication Date

1978-07-01

Submitted to SCIENCE

LBL-7761
Preprint

c.2

SUPERHEAVY ELEMENTS - A CROSSROADS

G. T. Seaborg, W. Loveland and D. J. Morrissey

July 1978

RECEIVED
LAWRENCE
BERKELEY LABORATORY

JAN 29 1979

LIBRARY AND
DOCUMENTS SECTION

Prepared for the U. S. Department of Energy
under Contract W-7405-ENG-48

TWO-WEEK LOAN COPY

This is a Library Circulating Copy
which may be borrowed for two weeks.
For a personal retention copy, call
Tech. Info. Division, Ext. 6782



LBL-7761

c.2

DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

SUPERHEAVY ELEMENTS — A CROSSROADS

New experimental data provide both hope and discouragement for the effort to synthesize these elements

G. T. Seaborg, W. Loveland and D. J. Morrissey

The authors are nuclear chemists working in the Department of Chemistry and the Lawrence Berkeley Laboratory of the University of California at Berkeley, California. W. Loveland is in the Department of Chemistry, Oregon State University, Corvallis, Oregon.

Introduction

For the past twelve years many nuclear scientists from around the world have devoted a considerable fraction of their time and resources to an attempt to synthesize superheavy elements (elements with atomic numbers $Z \geq 110$). To date, the results of this quest have been negative and the time appears ripe for a careful examination of the synthetic routes which have been explored and the prospects of future success along untested paths.

In 1972 Thompson and Tsang outlined in this journal¹ the reasons for believing that a massive extension of the Periodic Table of the Elements was possible through the production of superheavy elements. We shall comment briefly on current views of these expectations and summarize the results of attempts to synthesize superheavy elements (SHE's) by scientists in the United States, Europe and the Soviet Union. This will be followed by an examination of some of the reasons why these attempts have failed. Finally, a brief discussion will be made of exciting new prospects for success in this quest which have been stimulated by recent experiments at the Gesellschaft für Schwerionenforschung (GSI) at Darmstadt, West Germany. We close this survey by commenting on the impact, past and future, of this effort on nuclear chemistry and physics. Highly technical details will not be discussed, nor will the fascinating aspect of whether such elements or their decay products have been found in nature. For those wishing further information on this latter subject or a more detailed discussion of the subject of this article, a number of excellent review articles and conference proceedings^{2,3} are available.

Background

For many years nuclear scientists believed that the Periodic Table of Elements had been extended nearly to its limit (defined as the point where the number of protons in the nucleus, and the consequent repulsion between them, became so large that the cohesive nuclear forces could not hold the nucleus together and the nucleus would then undergo very rapid spontaneous fission decay). This idea was based on the observation of shorter and shorter spontaneous fission half-lives as the Z of the nucleus increased.

In the period from 1966 to 1972, a number of calculations² based upon modern theories of nuclear structure showed that in the region of proton number $Z = 114$, and neutron number $N = 184$, the ground states of nuclei were stabilized against fission. This stabilization was due to the complete filling of major proton and neutron shells in this region and is analogous to the stabilization of chemical elements, such as the noble gases, due to the filling of electronic shells in these atoms. Even more interesting, some of these detailed calculations suggested that the predicted half-lives for some of these "superheavy nuclei" might be on the order of the age of the universe, thus stimulating a great effort to observe these "missing elements" in nature. The superheavy elements were predicted to form an island of relative stability extending both above and below $Z = 114$ and $N = 184$ and separated from the peninsula of known nuclei by a sea of instability (see Fig. 1).

Some more recent calculations,⁵ based upon a careful consideration of the effect of mass asymmetry on the fission barrier and a reduced spin-orbit coupling strength, have indicated that the $Z = 114$ shell effect is

not very large. These calculations do confirm the existence of a shell at $N = 184$ but also suggest a lesser stability for species with $N < 184$, i.e., the island of stability has a cliff with a sharp drop-off for $N < 184$ as shown in Fig. 2. If these considerations are correct, it would become considerably more difficult to synthesize and detect the SHE's.

During the period following the initial optimistic predictions, efforts began at Berkeley, Orsay, Dubna, and later in Darmstadt, to "jump the gap" between the peninsula of known nuclei and the predicted island of stability by fusing two heavy nuclei together in a nuclear reaction, thus synthesizing the superheavy elements in the laboratory. As we shall show, these investigations, while failing to synthesize SHE's, appear to provide insight as to the relative stability of the SHE's and provide guidance for future research.

Predicted Properties of the Superheavy Elements

Nuclear Properties

As discussed previously, theoretical calculations have indicated that nuclei around $Z = 114$ and $N = 184$ should be relatively stable, although some estimates have attached more importance to the neutron shell at $N = 184$, and have indicated a lesser importance for the $Z = 114$ proton shell.⁵ Some calculations⁶ point to a shell closure at $Z = 126$, and not at $Z = 114$, but the general consensus of such calculations² has supported the idea of a shell closure at $Z = 114$. (As our considerations here will show, the synthesis of nuclei with Z as high as 126 seems to be beyond experimental reach.) These shells affect the synthesis of SHE's in two ways:

(1) by determining whether any excited superheavy nucleus formed in a nuclear reaction will survive destruction by fission during its deexcitation process (by controlling the height of the fission barrier);

(2) by determining if any "cold" superheavy nucleus that survived its deexcitation will live long enough to be detected through its alpha or spontaneous fission decay. Contours showing the half-lives for decay by spontaneous fission and α -particle emission as calculated by Randrup et al.⁵ (i.e., the more recent "pessimistic" estimate) are shown in Fig. 2. As one can see from examining Fig. 2, several nuclides in this island are predicted to have total decay half-lives substantially greater than 10^{-7} year (~ 3 sec). But note the precipitous decrease in spontaneous fission half-life (implying a decrease in the effective fission barrier height) as the neutron number decreases from $N = 184$, at constant proton number. This trend in the fission barriers gives one a feel for the importance of forming superheavy nuclei with the lowest excitation energy and the largest value of N possible.

The greater instability of elements with $Z \sim 114$ toward α -particle decay (compared to decay by spontaneous fission) leads to the prediction that nuclei near $Z = 110$, $N = 184$ should have the longest overall half lives. According to the predictions summarized in Fig. 2, the total decay half life of $^{294}_{110}$ is $\sim 10^5$ years. The older, more optimistic prognostications⁴ estimated the total decay half life of this nucleus to be $\sim 10^9$ years.

As a general summary of the uncertainty in these calculations, Bemis and Nix² have asserted that the accuracy of these half-life predictions is $\sim 10^{\pm 10}$ for spontaneous fission half-lives and $\sim 10^{\pm 3}$ for alpha-decay and beta-decay half-lives. Because of the very long half-

lives predicted for the most stable residents of the island of stability, a large error in the calculated half-lives could occur and still leave the possibility of forming detectable superheavy nuclei. However this uncertainty also indicates that we may have to use techniques capable of detecting very short lived nuclei in searching for SHE's. On the optimistic side, we should note that all the predicted nuclear properties refer to nuclei with even values of Z and N, while it is well known that nuclei with odd values of Z and/or N have higher fission barriers, longer spontaneous fission and α -decay half lives.

Once formed, it is important that a SHE give a unique signal in its decay in order to be easily distinguished from the many other products of the synthesis reactions. The high atomic number of the superheavy element might lead⁷ to increased fission fragment kinetic energies (235 MeV for Z = 114 as compared to 172 MeV for Z = 92), higher α -particle energies (7 MeV for Z = 114 compared to 4 MeV for Z = 92), and a very large number of neutrons emitted per fission event (10 for Z = 114 compared to 2.4 for ²³⁵U). An international group of scientists has proposed criteria for the discovery of chemical elements⁸ in which they insist that any claim to detection of a SHE must involve some proof concerning the atomic number of the new element. The aforementioned decay properties are general indicators of the formation of an element in the SHE category; detailed claims for the discovery of a particular SHE would have to be predicated on clear-cut establishment of the atomic number by chemical separations, observations of the characteristic x-rays, etc.

Chemical properties.

One of the most interesting aspects of the superheavy elements is their predicted chemical properties.⁹ The electronic properties of the elements are fairly well understood as the result of relativistic Hartree-Fock and Hartree-Fock-Slater calculations. The prediction of chemical properties based upon these electron configurations usually includes the judicious use of Mendeleev-like extrapolations of the smooth trends in the variation of a property such as the heat of vaporization amongst the members of a given Periodic Table group (Fig. 3 shows the predicted position of the SHE's in the Periodic Table). Not surprisingly, most calculations predict chemical properties for the SHE's to have easily recognizable similarities to those of their homologues, i.e., element 114 chemistry is characterized by a +2 oxidation state like its homologue lead. Pitzer¹⁰ has pointed out, though, that due to relativistic effects, the elements 112 (eka-mercury) and 114 (eka-lead) may, in fact, be very noble, i.e., volatile gases or liquids.

Thus one must be cautious in predicting SHE chemical properties due to the importance of relativistic effects in determining their electron configurations. For example the six 7p electrons are predicted to be split into two groups, four $7p_{3/2}$ and two $7p_{1/2}$ electrons, with the splitting between these electron energies being such that the filled $7p_{1/2}^2$ orbital will act as a closed shell and additional $7p_{3/2}$ electrons will act as electrons outside of a closed shell. As an example of this effect, element 115 (eka-bismuth) is predicted to have its valence electrons in the configuration $7p_{1/2}^2 7p_{3/2}$ with a consequent stable +1 oxidation state in contrast to the stable +3 oxidation state of its

homologue bismuth. Thus chemists are excited about this possibility of studying "relativity in a test tube."

Based upon the assumption that the half lives of any superheavy nuclei produced in laboratory syntheses might be sufficiently long (>1 sec), chemical separation methods for identifying the atomic number of these nuclei have been devised using these predicted chemical properties. Separations based upon the ion exchange behavior of the bromide complexes of the elements,¹¹ the tendency of the elements to co-precipitate with CuS,¹² and their possible volatility and ease of reduction¹³ have been applied to attempts to synthesize and chemically identify superheavy elements.

Summary of Reported Attempts to Synthesize Superheavy Elements

Table 1 contains a summary of recent attempts to synthesize superheavy elements in nuclear reactions utilizing the complete fusion of two heavy ions. The energetics of the reactions, fission barrier heights and neutron binding energies were taken from appropriate recent calculations.²¹ Since the sought after superheavy element is initially produced as an excited compound nucleus, its survival requires the loss of its excitation energy by the emission of neutrons in competition with the much more probable fission process (which will destroy the superheavy nucleus if it occurs). A simple estimate of the survival rate of the superheavy nuclei formed in these reactions was made using Fermi gas level density expressions which included consideration of the effect of angular momentum on the SHE survival.²² (When two heavy nuclei collide, large amounts (30 to 100 h) of rotational angular momentum are introduced into the system. The centrifugal forces which arise

increase the probability of nuclear fission.)

In examining the data in Table 1, one should remember that the probability of producing a detectable superheavy nucleus is equal to the product of two factors, (a) the probability of initially getting the reacting heavy ions to fuse, i.e. form a composite superheavy system, and (b) the probability of the excited superheavy system formed in the nuclear reaction surviving its de-excitation process. There are three general classes of results shown in Table 1. They are:

- 1) An attempt to fuse a heavy nucleus with a light ion to form a composite system near $Z = 114$, wherein the survival rate (factor (b) above) was so low as to preclude production and observation of superheavy nuclei.
- 2) An attempt to fuse a heavy target nucleus with a heavy ion projectile to form a composite system that "overshoots" the center of the island of stability and then, after deexcitation, decays by α and β decay towards the center of the island of stability. Due to the large numbers of neutrons in the composite system (190 neutrons in the $^{76}\text{Ge} + ^{238}\text{U}$ reaction) in these reactions the overall predicted survival rates of these species are very good. Despite extensive searches over a wide range of bombarding energies, projectile-target combinations and product half-lives by scientists in the Soviet Union, no successful SHE syntheses have been achieved and rather low upper limits on SHE production have been set. There are very strong indications²³ that the initial fusion probability (factor (a) above) rapidly approaches zero as the

Z of the heavy ion exceeds ~ 26 . Thus no SHE's appear to be formed by these "overshoot" reactions. (In fact, if SHE's exist, the experimental upper limits on SHE production may serve as upper limits on the extent of complete fusion in these systems.)

- 3) The intriguing case of the $^{48}\text{Ca} + ^{248}\text{Cm}$ system, wherein both the fusion probability and the survival probability up to the poorly known last step in the deexcitation process are such as to possibly allow detectable quantities of superheavy nuclei to be formed. Unfortunately a "fission catastrophe" in the last step of the deexcitation process leads to a prediction of a low overall survival rate.

Because of the promising character of the $^{248}\text{Cm} + ^{48}\text{Ca}$ reaction for synthesizing superheavy nuclei and the apparent failure to do so using this reaction, it behooves us to examine this system in greater detail to see why the production of SHE's was not observed.

Why Weren't SHE's Seen in the $^{48}\text{Ca} + ^{248}\text{Cm}$ Reaction?

The reaction of $^{48}\text{Ca} + ^{248}\text{Cm}$ to produce SHE's has been extensively studied^{13,18,20} by groups at the Lawrence Berkeley Laboratory, the Lawrence Livermore Laboratory and the Joint Institute for Nuclear Research (Dubna). The reacting heavy ion and target nucleus were brought together at the minimum energy (about 20 MeV above the interaction barrier) thought to be necessary to cause complete fusion, hopefully producing a composite system with some 40+ MeV of excitation energy. In the course of many carefully planned and executed experiments, upper

limits for the production of SHE's (expressed as cross sections) were measured and are summarized in Fig. 4. Superheavy products of these reactions were searched for by a variety of techniques, including:

- a) Spontaneous fission decay in flight of the recoil super-heavy nuclei (labeled DIF in Fig. 4).
- b) Gas jet collection of the recoils followed by α -particle and spontaneous fission counting (W).
- c) Direct counting of the stopped recoils for spontaneous fission activity (FOILS).
- d) Chemical separations of product nuclei based upon their projected chemical properties followed by spontaneous fission and α -particle counting. CHEM represents the work described in Ref. 20, DUBNA- α and DUBNA-SF represents the work described in Ref. 18, and GAS the work of Ref. 13 (in which volatile products were examined).

What would we have expected the formation cross section for super-heavy nuclei to be in this reaction? An estimate of the cross section for the fusion of ^{48}Ca and ^{248}Cm might be $\sigma_F \geq 10^{-27} \text{ cm}^2$ based upon the observation²⁴ of the production of the complete fusion product $^{254}_{102}\text{No}$ with a cross section of $3 \times 10^{-30} \text{ cm}^2$ from the similar $^{48}\text{Ca} + ^{208}\text{Pb}$ fusion reaction. If one uses the same method of estimating survival probabilities used in Table 1, one calculates a survival probability of $\sim 10^{-5}$ for the ^{254}No nuclei, thus implying a complete fusion cross section of $\sim 300 \times 10^{-27} \text{ cm}^2$. From this number and the systematics of complete fusion cross sections, we extrapolate a value of $\sigma_{CF} \geq 10^{-27} \text{ cm}^2$ for the $^{48}\text{Ca} + ^{248}\text{Cm}$ reaction. In addition, we note that in the reaction of ^{40}Ar and ^{48}Ca

with ^{238}U , products were observed^{25,26} (with a production cross section of $\geq 60 \times 10^{-27} \text{ cm}^2$) that appear to have resulted from the fusion of the ^{48}Ca or ^{40}Ar and the ^{238}U nucleus followed by fission. (These products have excitation functions and angular distributions characteristic of the fusion-fission process.) Since the only definitive signature of the complete fusion process in the $^{48}\text{Ca} + ^{248}\text{Cm}$ reaction is the detection of SHE's, it is possible that the reacting ions did not actually fuse (a possibility suggested by some calculations²⁷), but in view of the evidence cited above, we shall proceed under the assumption that some fusion ($\sigma \geq 10^{-27} \text{ cm}^2$) did take place in the $^{48}\text{Ca} + ^{248}\text{Cm}$ reaction.

A schematic representation of the deexcitation of any $^{296}_{116}$ compound nuclei formed in the $^{48}\text{Ca} + ^{248}\text{Cm}$ reaction is shown in Fig. 5 where we have used two different estimates of the reaction energetics and fission barrier heights to calculate the survival rates of the superheavy nuclei. The estimates used are those of Fiset and Nix⁴ (which in turn are similar to most theoretical calculations done in the period from 1966-72) and those of Randrup et al.⁵ (which represent a more recent, "pessimistic", approach). The "experimental" upper limit on the SHE survival rate in this reaction can be calculated as the ratio (SHE production cross section upper limit) / (complete fusion cross section), i.e., $5 \times 10^{-35} / 10^{-27} \leq 5 \times 10^{-8}$. Clearly, calculations based upon the older, more "optimistic" barriers of Fiset and Nix grossly overestimate the survival probabilities in this reaction, giving values approaching unity. Calculations based upon the more recent "pessimistic" barriers and energetics of Randrup et al. are consistent with the data. The calculations based upon the barriers and energetics of Fiset and Nix may be brought into agreement

with the observed upper limit cross sections for SHE production by using values for the fission barrier heights that are 4-5 MeV lower than originally predicted. The overall cross section for the production of detectable superheavy nuclei would be predicted to be $10^{-27} \times 10^{-11} \lesssim 10^{-38} \text{ cm}^2$, using the barriers of Randrup et al. An appreciation of the miniscule magnitude of these cross sections can be obtained by realizing that under the most favorable experimental conditions available today, a production cross section of 10^{-35} cm^2 corresponds to the production of 1-3 SHE atoms per day of irradiation.

Thus the failure to observe SHE's in this laboratory synthesis reaction seems to indicate that the fission barriers of these elements are considerably lower than those reported earlier.^{2,4} This observation has certain qualitative consequences. If one accepts the calculations of Randrup et al.⁴ as correctly describing the properties of the superheavy nuclei (which is consistent with the experimental data for the $^{48}\text{Ca} + ^{248}\text{Cm}$ reaction), then, as noted previously, one concludes that the longest total half-life of a superheavy nucleus is $\sim 10^5$ years, a fact which precludes their observation in terrestrial matter or any object whose age significantly exceeds 10^5 years, such as cosmic radiation. (This, of course, does not preclude observation of fossil remnants of extinct SHE's, such as decay products or fission tracks.) At the same time, one must be careful to note that the experimental results only test the cumulative survival probabilities, not the topology of the superheavy island. Thus we do not know whether the island of stability has a structure like the Matterhorn, steeply falling into the sea of instability, as N decreases from 184 as suggested by the calculations

of Randrup et al, or whether it is a lesser peak with a broad base extending to significantly lower values of N, thus resembling the legendary home of Satan in the San Francisco Bay Area, Mt. Diablo, as would be suggested by the Fiset and Nix topology appropriately lowered to fit experimental data.

Some Future Possibilities

Have we learned anything that might aid us in future searches for superheavy elements using complete fusion reactions?

From examining the estimates (in Fig. 5) of survival probabilities based upon the barriers of Randrup et al, one concludes that in the $^{48}\text{Ca} + ^{248}\text{Cm}$ reaction, the survival of superheavy nuclei is quite good until the last step(s) in the deexcitation chain, at which time a "fission catastrophe" is estimated to occur, wherein one "rolls off the island of stability." An obvious improvement in the yield of SHE's produced in this reaction would result if the compound nucleus $^{296}_{116}$ could be produced at an excitation energy less than 44 MeV. For example, if the initial excitation energy of the $^{296}_{116}$ species were 37 MeV instead of the value of 44 MeV used in the experiments, the overall SHE survival probability would be estimated to increase by $10^2 - 10^3$, giving a SHE production cross section of $10^{-36} - 10^{-35} \text{ cm}^2$ or less.

Sierk²⁷ and others, however, have argued on the basis of hydrodynamical calculations that complete fusion of ^{48}Ca and ^{248}Cm will not occur unless the projectile energy is such that the $^{296}_{116}$ species is produced with an excitation energy of 55 - 70 MeV. According to our calculations, such an excitation energy would cause all the SHE precursors to fission, leaving no SHE survivors. *Thus we appear to be caught on the horns of a dilemma. If the bombarding energy is low, the reacting*

nuclei don't fuse; if the bombarding energy is high enough to get fusion, the product nuclei don't survive.

However, an investigation²⁶ of a similar reaction, $^{40}\text{Ar} + ^{238}\text{U}$ has shown that the fusion reaction begins to occur when the energy of the projectile is 8-12 MeV above the Coulomb barrier, in agreement with other theoretical considerations.²⁸ The bombardments of ^{248}Cm with ^{48}Ca were performed at an average ^{48}Ca laboratory energy (in the target) of 255 MeV, which is 22 MeV higher than the simple Coulomb barrier for this reaction. Thus it appears possible to lower the ^{48}Ca energy to the region $241-245 \leq E_{\text{Ca}} \leq 248$ MeV (increasing the SHE survival probability) and yet still allow some complete fusion to occur.

Another possibility for improving the survival probability for superheavy nuclei formed in complete fusion reactions is to begin with a more neutron-rich target, such as ^{250}Cm . Using the same estimation procedures employed in constructing Table 1 and similar values of the excitation energy, we predict that in the $^{48}\text{Ca} + ^{250}\text{Cm}$ reaction, the survival probability of the superheavy species will increase by a factor of $\sim 10^4$ compared to the survival probability in the $^{48}\text{Ca} + ^{248}\text{Cm}$ reaction. If the complete fusion cross section for the $^{48}\text{Ca} + ^{250}\text{Cm}$ reaction is $\sim 10^{-27} \text{ cm}^2$, then we would predict a superheavy production cross section of $\sim 10^{-34} \text{ cm}^2$ or less, a conceivably detectable level.

In any case, the results of the $^{48}\text{Ca} + ^{248}\text{Cm}$ experiments serve as a valuable benchmark for any other attempts to produce superheavy nuclei. They tell us present detection methods were not adequate to detect the superheavy survivors from a process producing superheavy precursors with a cross section of 10^{-27} cm^2 and an excitation energy of ~ 40 MeV.

Deep Inelastic Pathways to the Superheavy Elements - Hope for the Future?

A new mechanism for the interaction of heavy ions was discovered²⁹ some five years ago and has been investigated extensively.³⁰ Termed "deep inelastic scattering," it is an inelastic scattering in which there is massive energy and nucleon transfer between projectile and target. It soon became apparent this reaction might offer another pathway to the SHE's. A preliminary report of the production of superheavy elements using the deep inelastic mechanism in the $^{136}\text{Xe} + ^{238}\text{U}$ reaction has appeared,³¹ but attempts to duplicate these observations have failed.^{32,33} However, recent experiments performed at the GSI in Darmstadt³⁴ have encouraged those who believe that it may be possible to make the superheavy elements using this new reaction pathway. The product atomic number distribution resulting from the reaction of 1785 MeV ^{238}U ions with a thick ^{238}U target is shown in Fig. 6. For the heavy mass products, one sees a broad distribution of products with atomic numbers near that of U. These products are the survivors of the deep inelastic scattering process. A detailed examination of the data represented in Fig. 6 reveals the production (with a cross section of 10^{-33} cm^2) of ^{255}Fm from ^{238}U (a net transfer of 8 protons and 9 neutrons to the target with *survival of this product*). Preliminary indications³⁵ are that more nucleons are transferred per MeV of excitation energy in the U + U reactions compared with deep inelastic scattering reactions involving heavy targets and lighter projectiles, thus allowing the production of "colder" products in the U + U system. Thus, on paper at least, one might think of reactions involving heavy target nuclei in which massive nucleon transfers could lead to the production and survival of superheavy nuclei.

The proper question to be asked is whether one can put a quantitative base under such extrapolations. For the $^{238}\text{U} + ^{238}\text{U}$ reaction, studied by Schädel et al.,³³ the yield of products with $Z = 70$ from the starting point of $Z = 92$ corresponds to a production cross section of 10^{-28} cm^2 . Assuming the number of $Z = 70$ products has not changed during the deexcitation process, the symmetric character of the $\text{U} + \text{U}$ system dictates that the yield of primary products with $Z = 114$ corresponds to a cross section of 10^{-28} cm^2 (in rough agreement with the predictions of Ayik et al.³⁶). The excitation energy of the $Z = 114$ species is not well known. If one believes that in the $^{48}\text{Ca} + ^{248}\text{Cm}$ reaction complete fusion occurred to an extent such that $\sigma_{\text{CF}} \cong 10^{-27} \text{ cm}^2$, then the $\text{U} + \text{U}$ deep inelastic reaction offers no improvement over this system unless the excitation energy of the $Z = 114$ species is ≤ 40 MeV or they are very neutron-rich.

A further problem is the experimental observation that in the deep Δ inelastic scattering reactions involving heavy targets (such as the reactions of $\text{Xe} + \text{Ta}$, $\text{Ca} + \text{Cm}$, and $\text{U} + \text{U}$), the heaviest survivors of the deep inelastic transfer process correspond to a net transfer of roughly equal numbers of neutrons and protons, giving rise to n-deficient products. This can be seen as a consequence of the transfer of increasing excitation energy with increasing numbers of nucleons. (The excitation energy causes the emission of more neutrons thus leading to n-deficient survivors.) This is clearly not desirable for superheavy element synthesis where one needs to make as neutron-rich a species as possible (see Fig. 2). For example, to go from ^{238}U to $^{298}_{114}$ requires an increase of ~ 1.7 neutrons for every proton added, thus implying an initial transfer of more than 1.7 neutrons per proton. Using the reaction $^{160}\text{Gd} + ^{136}\text{Xe}$

as a test for the increase of 18 protons and 34 neutrons (to produce ^{212}Pb), Otto et al.³³ set an upper limit for the cross section for this reaction of 10^{-33} cm². However, Schädel et al.³⁴ have pointed to evidence that in the deep inelastic process, the maximum primary product yield is for N/Z ratios near the valley of β^- stability, thus leading to predictions of more n-rich SHE precursors.

In any case, if one starts with a *very heavy* target nucleus then the probability of transferring the proper number of nucleons at a low enough excitation energy to form a surviving SHE should increase dramatically. There are possible modifications of the $^{238}\text{U} + ^{238}\text{U}$ experiment that could significantly improve the survival rates of the SHE's. For example, the bombardment of a ^{248}Cm target with a heavier projectile such as ^{244}Pu should allow the primary yield of the SHE precursors to increase (due to the need to transfer fewer nucleons compared to the $^{238}\text{U} + ^{238}\text{U}$ reaction) and the excitation energy of the superheavy precursors to decrease, increasing the survival rate of the secondary products. The decrease in excitation energy of the SHE precursors is a consequence of the fact that excitation energy of the deep inelastic products divides as the mass; thus a heavier projectile will carry away more excitation energy leaving less excitation in the superheavy precursor. Also, as hinted at in the considerations of the U + U reaction,^{34,37} the special stability of the "magic" superheavy nucleus could lead to a minimum excitation of this deep inelastic transfer product. Using the calculational framework suggested by Ayik et al.,³⁶ the yields of superheavy products from the $^{248}\text{Cm} + ^{244}\text{Pu}$ reaction should be at least 10 times greater than the yields from the $^{238}\text{U} + ^{238}\text{U}$ reaction. The use of even heavier

targets, such as ^{254}Es , has the advantage that a smaller number of nucleons needs to be added to synthesize SHE's, but this advantage may be offset by the small quantity of available target material. For example, the formalism of Ayik et al. would predict a 40-fold increase in SHE yield from the $^{254}\text{Es} + ^{244}\text{Pu}$ reaction compared to the $^{238}\text{U} + ^{238}\text{U}$ reaction, but this increase is completely negated by the 400-fold decrease in achievable target thickness.

Since the exact details of the superheavy element production process within the deep inelastic transfer mechanism depend so critically on the poorly characterized "tails" of the distributions of product mass, charge and excitation energy, it is very difficult to make meaningful quantitative estimates of the SHE production probabilities, and the estimates cited above should be viewed with caution. Once one has determined that one is "in the ballpark" of producing detectable numbers of SHE nuclei, as appears to be the case for various postulated heavy target-heavy projectile deep-inelastic transfer processes, then the path is clear for a continuation of the program to attempt to synthesize and identify these elusive elements using this reaction path.

Outlook for the Future

Clearly the effort to synthesize superheavy elements is at a crossroads. We have been deeply disappointed by the failure of apparently promising approaches. Yet, as our discussion indicates, there is still significant hope, and sufficient possibilities to sustain future effort. What does the future hold for the quest to synthesize superheavy elements? Hopefully, "in the best of all possible worlds" all of the following items might be part of our future:

1. *A general improvement of the methods used to detect superheavy elements.* With no further changes in much of the detector apparatus, an increase of 10^1 - 10^2 in detection sensitivity could be obtained by irradiating target nuclei with higher intensity particle beams for longer times. More research is needed into the problems of running these high intensity, high energy beams of heavy ions through thin foils of heavy elements. Such research may be crucial to future experiments with exotic beams and targets, especially when one realizes that due to these "targetry" problems, current experiments only utilize a small fraction of the total ion beams available from modern accelerators.

Better means need to be developed also for detecting short (i.e. $t_{1/2} < 1$ sec) half-life superheavy activities. More emphasis needs to be placed on purely physical methods of superheavy element detection, such as magnetic spectrometers, velocity separators, etc., which can identify the product atomic number without the use of chemical separations.

2. *A further extension of the complete fusion approach to SHE synthesis using the $^{48}\text{Ca} + ^{250}\text{Cm}$ reaction and the reaction of ^{48}Ca with ^{248}Cm at a lower bombarding energy.* The addition of two more neutrons to the target (^{250}Cm in place of ^{248}Cm) will, by the estimation procedures used in Table 1, increase the survival probability of the superheavy species by a factor of $\sim 10^4$. The availability of ^{250}Cm is very limited, unfortunately, and probably quantities sufficient to undertake an experiment could only become available after recovery from the debris of an old nuclear weapons test. As discussed earlier, further studies of the

$^{48}\text{Ca} + ^{248}\text{Cm}$ reaction at lower bombarding energies could also lead to an increase in SHE production of $10^2 - 10^3$.

3. *The ultimate extension of the deep inelastic transfer approach to SHE synthesis using an exotic target (such as ^{248}Cm or possibly ^{254}Es) and an exotic projectile (^{244}Pu). For the favorable case of the $^{248}\text{Cm} + ^{244}\text{Pu}$ reaction, the production cross section for SHE's might increase dramatically, thus allowing detection of any SHE formed.*

The reader may ask himself why one should bother with such unusual and expensive projects as outlined above. Why not just give up and turn from this crossroads to an easier task? Many of the original reasons for embarking on this attempt are still valid and compel us to further effort. The opportunity to uniquely test so much of modern nuclear science in this dramatic extension to a new and unknown region and the probable serious impact on chemistry of opening up a vista of many new chemical elements whose behavior and properties might be governed by rules (i.e. relativistic ones) not used in describing today's experiments help keep the quest alive. Also we know the new experiments, like the old ones, should have a significant "fallout" on other areas of nuclear science and chemistry. For even if we fail to make superheavy elements, the chances of greatly enhancing our knowledge of the nuclear structure and chemistry of the actinides and transactinides by the production of new isotopes of existing elements or the production of new non-superheavy chemical elements by such efforts seem good.

Summary

In summary, it would appear to us that:

- 1) The failure to synthesize SHE's with a complete fusion reaction is most likely understandable in terms of the low survival probabilities of the SHE precursors formed in these reactions or (in some cases) the failure to achieve complete fusion.
- 2) An additional approach to the synthesis of SHE's is through the use of the deep inelastic transfer reaction, using the heaviest available targets and projectiles.

TABLE 1

Attempts to synthesize superheavy elements using complete fusion reactions

Reaction studied	Compound nucleus	Mean excitation energy of compound nucleus (MeV)	Predicted survival probability of compound nucleus	Observed upper limit cross section (cm ²) for SHE production (for indicated t _{1/2})	Ref.
Class 1 — Compound nuclei with low survival probabilities					
²³² Th + ⁴⁸ Ca	²⁸⁰ ₁₁₀	44.5	10 ⁻²¹	4 × 10 ⁻³⁵ (>3 ms)	15
²³¹ Pa + ⁴⁸ Ca	²⁷⁹ ₁₁₁	34	10 ⁻¹⁷	5 × 10 ⁻³⁵ (76 m)	15
²³³ U + ⁴⁸ Ca	²⁸¹ ₁₁₂	33	0 ^a	7 × 10 ⁻³⁵ (20 hr)	15
²⁴⁸ Cm + ⁴⁰ Ar	²⁸⁸ ₁₁₄	43	0 ^a	10 ⁻³⁰ (10 ⁻⁸ -10 ⁻¹ sec)	19
²⁴² Pu + ⁴⁸ Ca	²⁹⁰ ₁₁₄	43.5	0 ^a	10 ⁻³⁵ (6 hr-1 yr)	18
²⁴³ Am + ⁴⁸ Cm	²⁹¹ ₁₁₅	41	0 ^a	2 × 10 ⁻³⁵ (6 hr-1 yr)	18
Class 2 — Small probability of forming compound nuclei					
²⁰⁸ Pb + ⁸⁴ Kr	²⁹² ₁₁₈	25.5	~0(10 ⁻⁹) ^b	10 ⁻³⁰ (>6 × 10 ⁻⁷ sec)	14
²³⁸ U + ⁶⁸ Zn	³⁰⁶ ₁₂₂	47	1.0	10 ⁻³⁰ (10 ⁻⁹ sec-1 yr)	17
²³² Th + ⁷⁶ Ge	³⁰⁸ ₁₂₂	32	1.0	10 ⁻³⁴ (5 ms-1 yr)	16
²⁴² Pu + ⁶⁸ Zn	³¹⁰ ₁₂₄	45	0.9	10 ⁻³⁰ (10 ⁻⁹ sec-1 yr)	17
²³⁸ U + ⁷⁶ Ge	³¹⁴ ₁₂₄	68	3 × 10 ⁻²	10 ⁻³³ (5 ms-1 yr)	16
²⁴³ Am + ⁶⁸ Zn	³¹¹ ₁₂₅	39	0.9	2 × 10 ⁻³² (10 ⁻⁹ sec-1 day)	17
²⁴⁶ Cm + ⁶⁸ Zn	³¹⁴ ₁₂₆	34	0.3	10 ⁻³⁰ (10 ⁻⁹ sec-1 yr)	17
²³² Th + ⁸⁴ Kr	³¹⁶ ₁₂₆	51	< 10 ⁻¹⁴	5 × 10 ⁻³⁰ (>6 × 10 ⁻⁷ sec)	14

TABLE 1 (continued)

Reaction studied	Compound nucleus	Mean excitation energy of compound nucleus (MeV)	Predicted survival probability of compound nucleus	Observed upper limit cross section for SHE production ($t_{1/2}$)	Ref.
Class 3 - Compound nuclei with possible survival					
$^{246}\text{Cm} + ^{48}\text{Ca}$	$^{294}_{116}$	40	$< 5 \times 10^{-16} (10^{-11})^b$	2×10^{-35} (6 hr-1 yr)	18
$^{248}\text{Cm} + ^{48}\text{Ca}$	$^{296}_{116}$	44	$< 4 \times 10^{-11} (10^{-5})^{b,c}$	5×10^{-35} (6 hr-1 yr)	13,18,20

^a Nuclei whose survival rate is exactly zero are cases in which some member of the neutron emission chain has a non-existent fission barrier.

^b The cumulative survival rate for these nuclei up to the last step in the deexcitation process is given in parentheses. In the last step of the deexcitation process, the excitation energy is at or below the neutron binding energy and well above the fission barrier. The result of this circumstance is a "fission catastrophe" in which nearly all the nuclei fission.

^c See text for discussion.

REFERENCES

1. S.G.Thompson and C.F.Tsang, *Science* 178, 1047 (1972).
2. C.E.Bemis,Jr. and J.R.Nix, *Comments on Nuclear and Particle Physics* 7, 65 (1977).
3. Proceedings of the International Symposium on Superheavy Elements, Lubbock, Texas, March 9-11, 1978.
4. E.O.Fiset and J.R.Nix, *Nucl. Phys.* A193, 647 (1972).
5. J.Randrup, S.E.Larsson, P.Möller, A.Sobiczewski, and A.Lukasiak, *Phys. Scr.* 10A, 60 (1974).
6. F.Petrovich, R.J.Philpott, D.Robson, J.J.Bevelacqua, M.Golin, and D.Stanley, *Phys. Rev. Lett.* 37, 558 (1976); see also J.M.Moss, *Phys. Rev.* C17, 813 (1978).
7. J.R.Nix, *Phys. Letters* 30B, 1 (1969).
8. B.G.Harvey, G.Herrmann, R.W.Hoff, D.C.Hoffman, E.K.Hyde, J.J.Katz, O.L.Keller,Jr., M.Lefort, and G.T.Seaborg, *Science* 193, 1271 (1976).
9. O.L.Keller,Jr. and G.T.Seaborg, *Ann. Rev. Nucl. Sci.* 27, 139 (1977).
10. K.S.Pitzer, *J. Chem. Phys.* 63, 1032 (1975).
11. J.V.Kratz, J.O.Liljenzin, and G.T.Seaborg, *Inorg. Nucl. Chem. Letters* 10, 951 (1974).
12. G.N.Flerov, *Proceedings Int. Conf. Reac. Complex Nucl.* 2, 459 (1974).
13. J.D.Illige, E.K.Hulet, J.M.Nitschke, R.T.Dougan, R.W.Lougheed, A.Ghiorso, and J.H.Landrum, *Phys. Lett.* 78B, 209 (1978).
14. P.Colombani, B.Gatty, J.C.Jacmart, M.Lefort, J.Peter, M.Riou, C.Stephen, and X.Tarrago, *Phys. Letters* 42B, 208 (1972).

15. G.M.Ter-Akopian, H.Bruchertseifer, G.V.Buklanov, L.P.Chelnokov, V.I.Chepigin, Choi Vai Sek, V.B.Fefilov, K.A.Kavrilov, Y.P.Kharitonov, N.N.Kolesnikov, B.N.Markov, O.A.Orlova, A.A.Pleve, and T.Y.Oganessian, JINR Preprint E7-10722.
16. G.N.Flerov, Y.T.Oganessian, Y.V.Lobanov, A.A.Pleve, G.M.Ter'Akopyan, A.G.Demin, S.P.Tretyakova, V.I.Chepigin, and Y.P.Tretyakov, Sov. J. Nucl. Phys. 19, 247 (1974).
17. A.Pleve, A.G.Demin, V.Kush, M.B.Miller, and N.A.Danilov, Sov. J. Nucl. Phys. 19, 123 (1974).
18. Y.T.Oganessian, H.Bruchertseifer, G.V.Buklanov, V.I.Chepigin, Choi Val Sek, B.Eichler, K.A.Gavrilov, H.Gaeggler, Y.S.Korotkin, O.A.Orlova, T.Reetz, W.Seidel, G.M.Ter-Akopian, S.P.Tretyakova, and I.Zvara, Nucl. Phys. A294, 213 (1978).
19. H.R.Bowman, R.C.Gatti, R.C.Jared, L.G.Moretto, W.J.Swiatecki and S.G.Thompson, Lawrence Berkeley Laboratory Report UCRL-17989, 1967.
20. E.K.Hulet, R.W.Lougheed, J.F.Wild, J.H.Landrum, P.C.Stevenson, A.Ghiorso, J.M.Nitschke, R.J.Otto, D.J.Morrissey, P.A.Baisden, B.F.Gavin, D.Lee, R.J.Silva, M.M.Fowler, and G.T.Seaborg, Phys. Rev. Lett. 39, 385 (1977); R.J.Otto, D.J.Morrissey, D.Lee, A.Ghiorso, J.M.Nitschke, and G.T.Seaborg, J. Inorg. Nucl. Chem. 40, 589 (1978).
21. For nuclei with $Z \leq 120$, the droplet model masses [W.D.Myers, The Droplet Model of Atomic Nuclei, Plenum Press, 1978)] were corrected using the shell corrections of Ref. 5 to give reaction energetics and neutron binding energies. For $Z > 120$, the energetics and barriers were taken from Ref. 4.

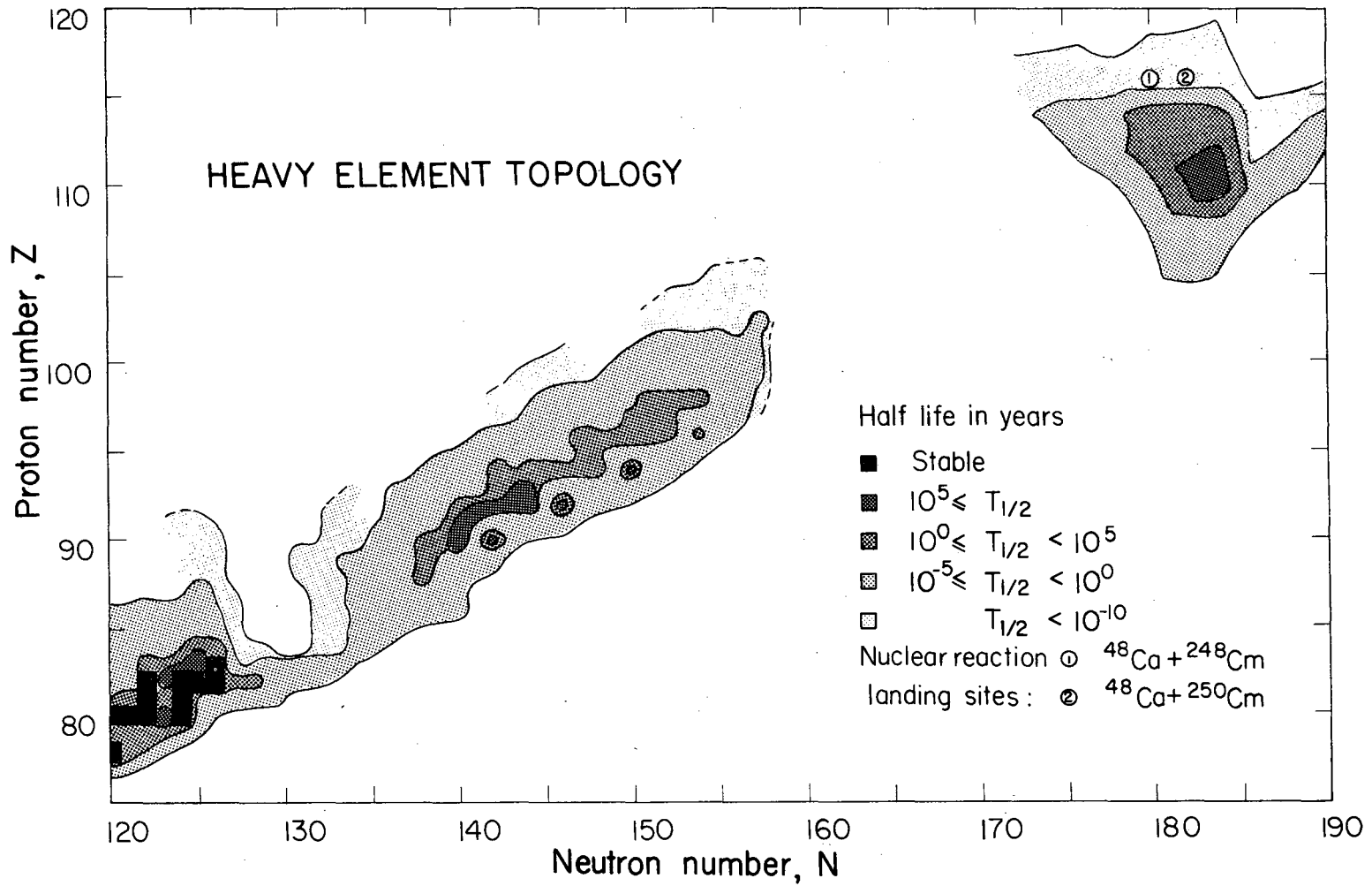
22. An outline of the formalism used is contained in: R.Vandenbosch and J.R.Huizenga, Nuclear Fission (Academic Press, New York, 1973), pp.233-250. Details of its application can be found in: W.Loveland, Oregon State University Report RLO-2227-TA35 (1978). Each emitted neutron was assumed to remove an excitation energy equal to $B_n + 2T$. Values of K_0 were taken from typical values for transuranium nuclei (p.202 of Vandenbosch and Huizenga); the fission level density parameter, a_f , was set equal to $1.1 a_n = 1.1(A/8)$.
23. D.J.Morrissey, Ph.D. Thesis, University of California, Berkeley, Lawrence Berkeley Laboratory Report LBL-7713 (1978).
24. J.M.Nitschke, R.E.Leber, M.J.Nurmia, and A.Ghiorso, Lawrence Berkeley Laboratory Report LBL-6534 Rev. (1978).
25. R.J.Otto, D.J.Morrissey, G.T.Seaborg, and W.D.Loveland, Z. für Physik A287, 97 (1978).
26. M.de Saint Simon, R.J.Otto, and G.T.Seaborg, Phys. Rev. C 18, 1651 (1978).
27. A.J.Sierk, in Proc. Int. Symp. on Superheavy Elements, Lubbock Texas, 1978, March 9-11, 1978.
28. J.Blocki, J.Randrup, W.J.Swiatecki and C.F.Tsang, Ann. Phys. 105, 427 (1977).
29. M.Lefort, C.Ngo, J.Peter and B.Tamain, Nucl. Phys. A216, 166 (1973).
30. See, for example, M.Lefort and C.Ngo, Ann. Phys. 3, 5 (1978); W.U.Schröder and J.R.Huizenga, Ann. Rev. Nucl. Sci. 27, 465 (1977).
31. G.N.Flerov, in Proc. Int. Conf. on Reactions Between Complex Nuclei, Nashville, June 1974, p.459.
32. H.Gäggeler, W.Brüchle, H.Ahrens, H.Folger, G.Franz, J.V.Kratz, M.Schädel, I.Warnecke, G.Wirth, N.Trautmann, G.Herrmann, N.Kaffrell,

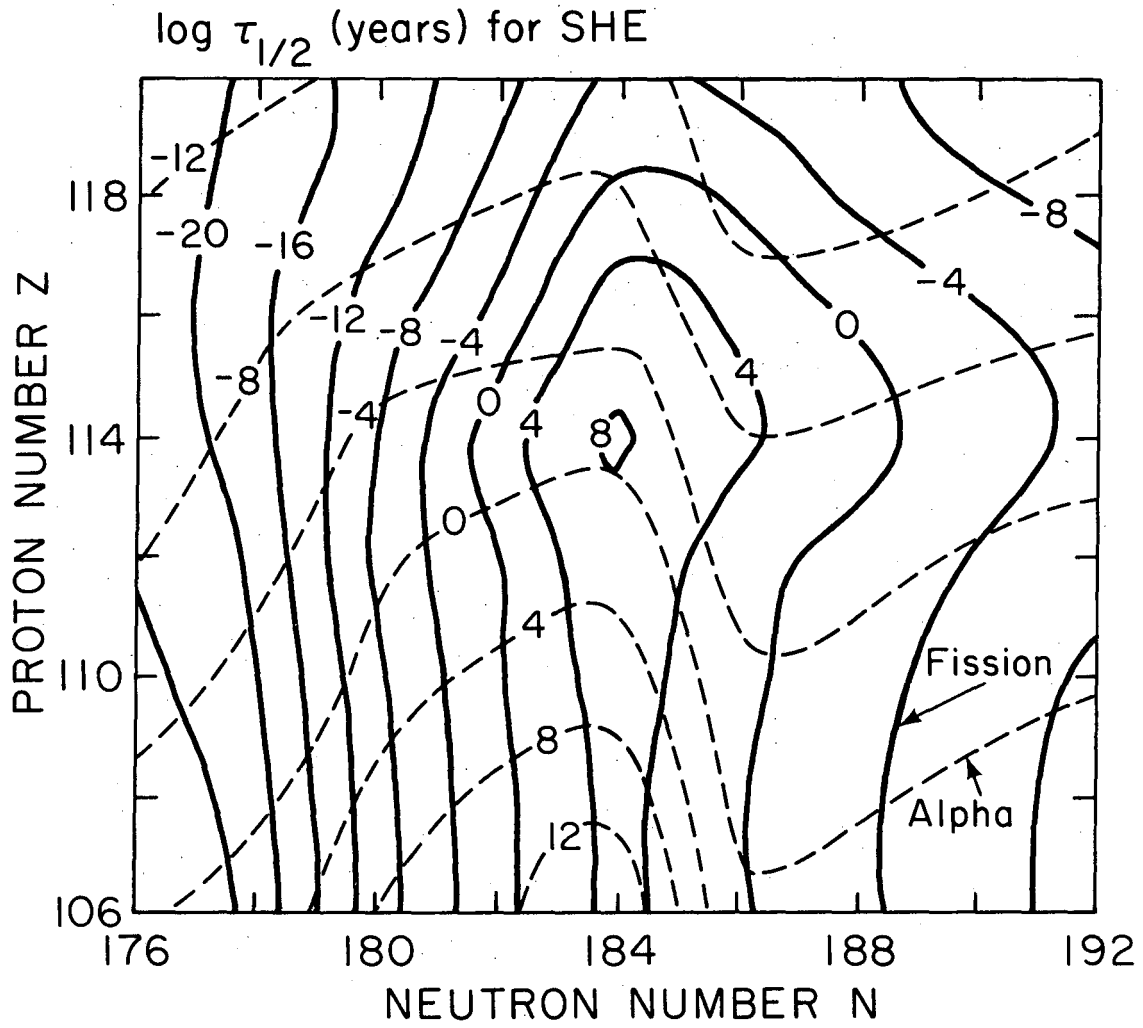
- P.Peuser, G.Tittel, M.Weber, and M.Zendel, Z. Phys. A286, 419 (1978).
33. R.J.Otto, A.Ghiorso, D.Lee, R.E.Leber, S.Yashita, and G.T.Seaborg, Radiochimica Acta 24, 3 (1977).
34. M.Schädel, J.V.Kratz, H.Ahrens, W.Brüchle, G.Franz, H.Gaggeler, I.Warnecke, G.Wirth, G.Herrmann, N.Trautman, and M.Weis, Phys. Rev. Lett. 41, 469 (1978).
35. D.D.Hildebrand, H.Freisleben, F.Puhlhofer, W.F.W.Schneider, R.Bock, D.V.Harrah, and H.J.Specht, Phys. Rev. Lett. 39, 1065 (1977).
36. S.Ayik, B.Schürmann, and W.Nörenberg, Z. Phys. A279, 145 (1976).
37. B.Schürmann, W.Nörenberg, and M.Simbel, Z. Phys. A286, 263 (1978).
38. We have greatly profited from numerous discussions with our many colleagues who have been an integral part of the research at Berkeley on superheavy elements, especially R. J. Otto, P. A. Baisden, A. Ghiorso, and J. M. Nitschke. We are indebted to J. V. Kratz, J. Randrup, and W. J. Swiatecki for critical reviews of the manuscript. This work was supported by the Nuclear Physics Division of the U.S. Department of Energy.

FIGURE CAPTIONS

- Fig. 1. A representation of the stability of nuclei (based upon known and predicted⁴ total decay half-lives) showing a peninsula of known elements and an island of predicted relative stability (nuclei near $Z = 114$ and $N = 184$) in a sea of instability. The position of the initial composite species found in the $^{48}\text{Ca} + ^{248,250}\text{Cm}$ reactions is also shown to emphasize the large number of neutrons that must be added to reach the island of stability.
- Fig. 2. Combined diagram of the predicted half-lives of the superheavy nuclei with respect to spontaneous fission (solid lines) and alpha decay (dashed lines). [From Randrup *et al.*⁵]
- Fig. 3. A modified form of the periodic table of the elements showing the predicted chemical properties of the superheavy elements.
- Fig. 4. Observed upper limits on the production cross section for superheavy elements produced in the $^{48}\text{Ca} + ^{248}\text{Cm}$ reaction.
- Fig. 5. A schematic representation of the deexcitation of SHE's formed in the $^{48}\text{Ca} + ^{248}\text{Cm}$ reaction.²²
- Fig. 6. The product distributions in the $^{238}\text{U} + ^{238}\text{U}$ reaction [from Schädel *et al.*³⁴] a) The distribution in atomic number of the products; b) Contour plot of the yields of products with given Z and A .

Fig. 1

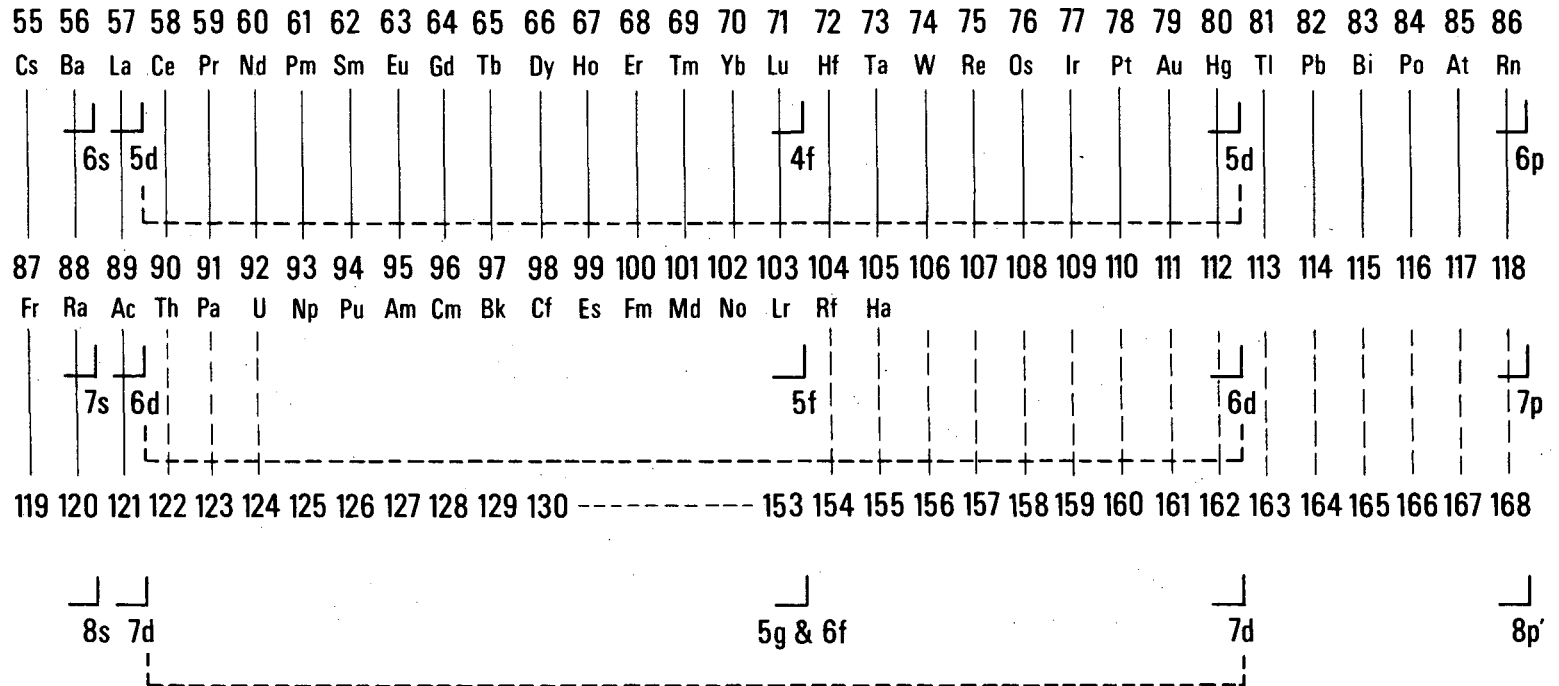




XBL 788-10626

Fig. 2

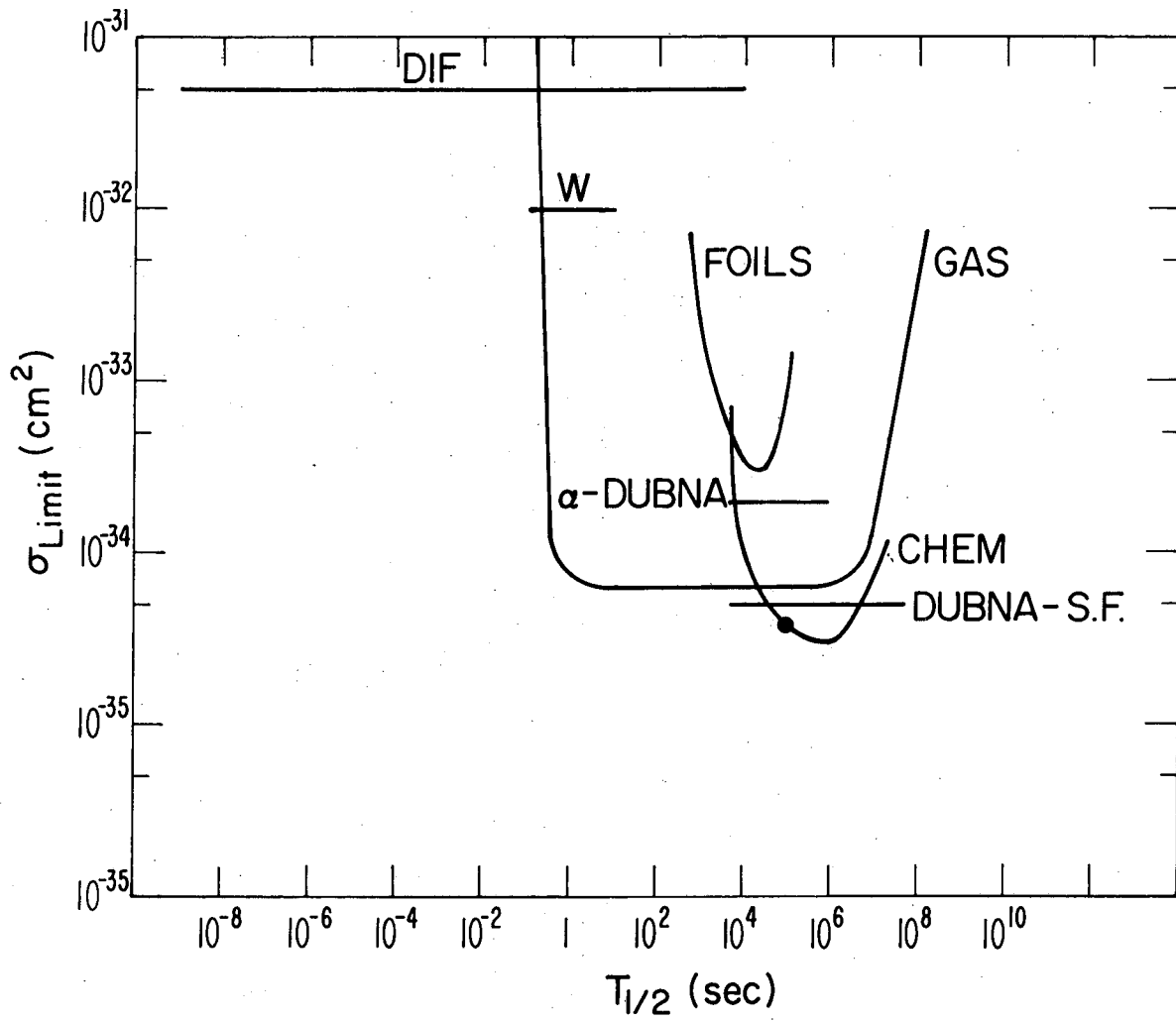
MODIFIED FORM OF PERIODIC TABLE SHOWING KNOWN AND PREDICTED ELECTRON SHELLS



-32-

Fig. 3

XBL 757-3431



XBL 783-7603

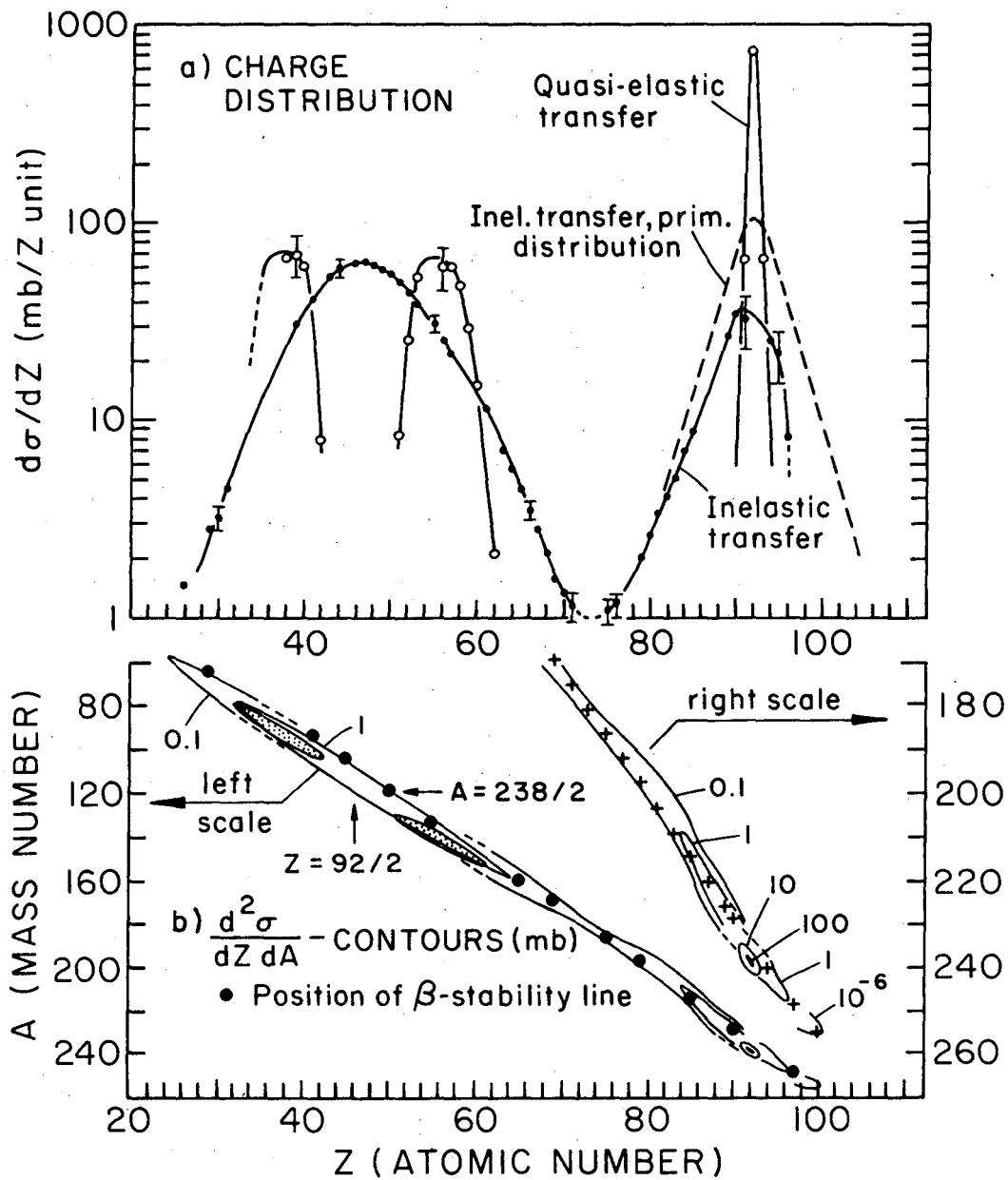
Fig. 4

Deexcitation of SHE precursors from the $^{48}\text{Ca} + ^{248}\text{Cm}$ Reaction

Nucleus	Excitation Energy (MeV)	Fission Barrier Height in MeV (Fraction that Survive Fission in percent)	
		Randrup <i>et al.</i> ⁵	Fiset and Nix ⁴
$^{296}_{116}$ → fission →	44	5.7 (18)	11.0 (98)
↓ neutron emission			
$^{295}_{116}$ → f →	34	5.9 (23)	10.5 (99)
↓ n			
$^{294}_{116}$ → f →	26	4.1 (1.4)	10.0 (99)
↓ n			
$^{293}_{116}$ → f →	16	3.5 (1.4)	9.6 (100)
↓ n			
$^{292}_{116}$ → f →	8	2.9 (5×10^{-6})	9.3 (100)
↓ γ, n			
<u>Predicted Cumulative Survival Probability</u>		$< 4 \times 10^{-11}$	0.97

XBL 7812-13760

Fig. 5



XBL 788-10627

Fig. 6

This report was done with support from the Department of Energy. Any conclusions or opinions expressed in this report represent solely those of the author(s) and not necessarily those of The Regents of the University of California, the Lawrence Berkeley Laboratory or the Department of Energy.

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