

Superheavy Elements—The Quest in Perspective

Since about 1940, the peninsula of known nuclei shown in Figure 1 has been extended again and again. The number of known elements has been increased from 92 to 106, and the number of known isotopes has been increased from about 300 to 1500. New nuclei are still being discovered, but with increasing difficulty because nuclei along the edges of the peninsula are unstable against beta decay (including electron capture), and those near its tip are unstable against nuclear fission and alpha decay. The latter two instabilities are both caused by the rapid growth of the disruptive Coulomb force as we go to heavier nuclei.

However, beyond the tip of the peninsula of known nuclei, an island of relatively stable superheavy nuclei is predicted to exist. The reason for this island is the extra stability given to a nucleus by the closing of a proton or neutron shell. As the particle numbers of nuclei are increased, successive protons and neutrons go into definite single-particle orbits. When a given shell of protons or neutrons is completely filled, that nucleus has relatively lower energy, that is, extra binding and hence increased stability.

For the past 10 years we have struggled to reach this island, both by searching for superheavy elements in nature and by attempting to produce them in various nuclear reactions at accelerators throughout the world. As is true with most endeavors worthy of pursuit, the quest for superheavy elements has not been without misfortune. Countless explorers have returned without even a glimpse of the island. Some, perhaps ill-prepared to make the journey, have shouted 'Land ho!' only to have it slowly fade before their eyes. None of these attempts have ever resulted in any conclusive evidence for the existence of superheavy elements. As of this date, there have been a minimum of 15 claims to the discovery of superheavy elements, or at the very least, suggestions that superheavy elements have been responsible for experimental effects. Most, but not all, claims have been retracted or dismissed. Yet the quest must go on!

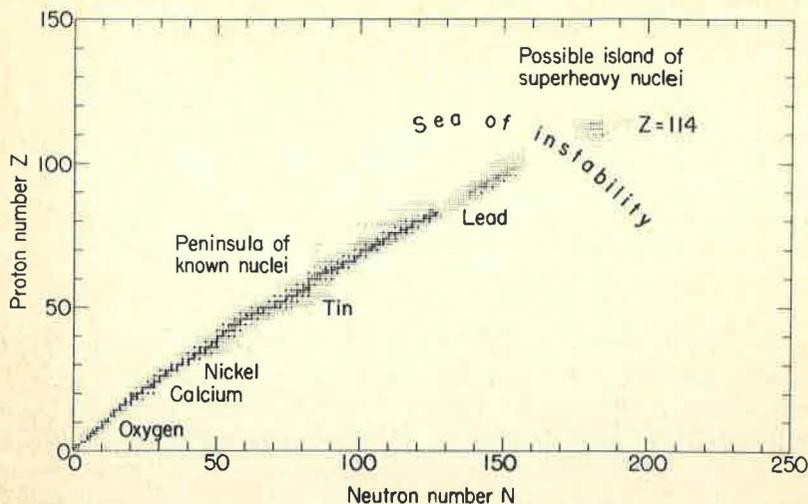


FIGURE 1. Location of the sought-for island of superheavy nuclei relative to the peninsula of known nuclei. The heaviest points represent naturally occurring nuclei, the medium-weight points represent nuclei with half-lives greater than 1 yr, and the lightest points represent nuclei with half-lives less than 1 yr. The labeled elements correspond to nuclei with closed proton shells.

Why are we caught up in this frantic search? Partly because of the same instinct to explore the unknown that drove Columbus to the New World, Hillary to the top of Everest, and Armstrong and Aldrin to the moon. Most importantly, the interest in the search for superheavy elements is because of what their discovery would teach us about the universe in which we live. Many nuclear and atomic theories yield similar results for known elements and nuclei but yield entirely different results when extrapolated into new regions. The discovery of superheavy elements would help decide between competing theories, as well as suggest entirely new ones. Serendipitous discoveries should not be excluded, as entirely new physical phenomena may be revealed if superheavy elements are ever discovered. The impact of such a discovery would be felt not only in nuclear physics and chemistry, but also in fields ranging from astrophysics and cosmology to atomic physics and quantum electrodynamics.

The experimental searches for superheavy elements both in nature and at accelerators have relied most heavily upon theoretical predictions. Because of this strong reliance upon theoretical predictions the superheavy-element quest may be likened to the search for magnetic monopoles, quarks, anti-particles, black holes, and superdense nuclear matter. Among these endeavors only the predictions and subsequent searches for antiparticles have been

successful. Of course, Hillary knew that Mt. Everest had a top, and Armstrong and Aldrin knew that the moon was accessible. We do not know that superheavy elements can even exist, apart from the questions of how to produce them in the laboratory or where and how to look for them in nature. The expected location and shape of the island, the half-lives, the atomic and chemical properties of the elements in it, and the properties associated with their radioactive decay have thus far guided our choices concerning both where and how to look and which detection methods would be applicable. We must therefore review such predictions before proceeding to the searches themselves.

Early History

The modern widespread interest in superheavy elements began in Berkeley in 1965 as a result of two independent developments.¹ The first of these was the estimate by Myers and Swiatecki that the fission barrier of a superheavy nucleus should be several MeV high, and the second was the suggestion by Meldner that the next closed proton shell after 82 is 114. It had always been assumed before, in analogy with the case for neutrons, that 126 would be the next closed proton shell. As early as 1957, for example, Scharff-Goldhaber² had suggested the possibility of another region of relative stability at the doubly magic nucleus ${}_{184}^{310}126$. However, this nucleus is far removed from the line of beta stability, and it should be highly unstable against decay by electron capture. It should also decay rapidly by alpha emission. The repulsive Coulomb force, which becomes increasingly important for heavier nuclei, is responsible for shifting the proton shell closure from 126 to 114.

There were also several other early suggestions concerning the possibility of superheavy nuclei (see Ref. 3 for a list of references). Although some of these suggestions were made even prior to 1957, the required stability was, in general, not associated with single-particle shell closures.

In 1966 Strutinsky developed an improved method for calculating the potential energy of a nucleus as a function of its shape,⁴ and he and his co-workers used this method to calculate the fission barriers of several superheavy nuclei.⁵ Subsequently, Nilsson and his co-workers applied Strutinsky's method to a modified harmonic-oscillator single-particle potential to make the first systematic survey of the expected stability of superheavy nuclei.⁶ Since then, several other groups have made detailed calculations with improved computational techniques and with improved single-particle potentials. The status of such calculations is reviewed in Refs. 3 and 7.

Nuclear Stability

Superheavy nuclei may decay by spontaneous fission, alpha decay, or beta decay (including electron capture). To calculate the stability of a nucleus against these possible decay modes, one must know the nuclear potential energy as a function of both proton and neutron number and the nuclear shape. The most fundamental way to calculate this would be to start with a basic nucleon–nucleon interaction derived from scattering data and solve the appropriate many-body equations in some approximation, for example, the Hartree–Fock approximation. Unfortunately, such basic calculations are both extremely time-consuming and have not yet achieved the accuracy of the alternative method developed by Strutinsky.⁴

Strutinsky's method is a two-part approach, with the smooth trends of the potential energy taken from a macroscopic model and the local fluctuations from a microscopic model. A macroscopic approach such as the liquid-drop model describes quantitatively such smooth trends of the nuclear potential energy but not the local fluctuations, whereas a microscopic approach, such as the single-particle model, describes the local fluctuations but not the smooth trends. So, why not synthesize the two? This combined macroscopic-microscopic method should then hopefully reproduce both the smooth trends and the local fluctuations. This method is described in detail in Ref. 3.

After the fission barrier of a superheavy nucleus has been calculated by use of the above method, its half-life against spontaneous fission is determined by calculating the penetrability through the barrier. This can be done by use of the WKB (quasi-classical) approximation once the inertia, or effective mass, associated with the deformation is known. The inertia can be determined semi-empirically by adjusting its value to reproduce known spontaneous-fission half-lives of actinide nuclei, or alternatively it can be calculated microscopically.

The half-lives with respect to alpha decay and beta decay are determined, by means of fairly standard approximations, from the energy released in the decays. This energy is found from the calculated ground-state masses of the nuclei involved.

As summarized in Refs. 3 and 7, many groups have used the methods outlined above to calculate the stability of superheavy nuclei. Most of the results are in general agreement that nuclei in the vicinity of 114 protons and 184 neutrons should be relatively stable against spontaneous fission, alpha decay, and beta decay. However, in a few calculations, either the 114 proton shell closure or the 184 neutron shell closure is totally absent. Also, because the accuracy with which such predictions can be made is only about $10^{\pm 10}$ for spontaneous-fission half-lives and about $10^{\pm 3}$ for alpha-decay and beta-decay half-lives, the detailed results differ somewhat between the various

groups performing such calculations.

As a specific example, we consider the results calculated with a diffuse-surface single-particle potential of the folded Yukawa type,⁸ which leads to the island shown in Figure 1. The doubly magic nucleus ${}_{184}^{298}114$ has the longest calculated spontaneous-fission half-life of 10^{19} yr. As we move away from this nucleus, the spontaneous-fission half-lives decrease, with the decrease most gentle in the direction of increasing proton number and decreasing neutron number. However, nuclei with proton numbers in excess of 114 decay rapidly by the emission of high-energy alpha particles. When all decay modes are taken into account (alpha, beta, and spontaneous-fission decay) the nucleus ${}_{184}^{294}110$ has the longest calculated *total* half-life of 10^9 yr.

In addition to the above island of superheavy nuclei, there may be other regions of near-stability⁹ associated with the closure of the 228, 308, and 406 neutron shells and with the 164 proton shell. Fairly weak proton shell closures also occur in some calculations at 126 (or 124), 154, and 204 protons. For these higher shell closures, only three combinations of proton and neutron numbers lead to nuclei that are close to the extrapolated line of beta stability. These regions of possible stability are centered around ${}^{354}126$, ${}^{472}164$, and ${}^{610}204$.

Because of the weak proton shell closure and the extremely large repulsive Coulomb force, nuclei in the vicinity of ${}^{610}204$ should have very short half-lives with respect to both spontaneous fission and alpha decay. The calculated spontaneous-fission half-life of ${}^{472}164$ exceeds 10^{60} yr, but its calculated alpha-decay half-life is only about 10 yr.¹⁰ For nuclei close to ${}^{472}164$, but with somewhat fewer protons, total half-lives as long as 10^5 to 10^7 yr are obtained.¹⁰ Although there is some disagreement¹¹⁻¹⁶ concerning the stability of nuclei in the vicinity of ${}^{354}126$, one calculation¹³ gives 39 ms for the spontaneous-fission half-life of ${}^{354}126$ and 18 yr for its alpha-decay half-life. For even-even beta-stable nuclei in this region, the nucleus ${}^{352}124$ has the longest total calculated half-life of 67 s.¹³

Chemical, Physical, and Nuclear Properties Unique to Superheavy Elements

The chemical properties of superheavy elements may be predicted by performing self-consistent calculations for the electrons surrounding the nuclei, for example, relativistic Hartree-Fock or Dirac-Fock calculations.¹⁷⁻¹⁹ The results indicate that the order of filling electronic orbits is somewhat more complicated than had been supposed. For example, we had always thought that elements 103 through 120 would fill their electronic orbits in a way analogous to those in the preceding row in the periodic table and thus would have similar ground-state configurations, but there are exceptions at elements

103, 110, and 111. Also, beginning with element 121, the order of filling the shells is different from the order in the actinide elements. These differences arise from relativistic effects associated with the higher charge of these elements. Therefore, whereas element 114 and lead, for example, should have analogous chemical properties, the chemical properties of some of the other superheavy elements and their lighter homologs should be somewhat less similar.

Guided by the relativistic atomic calculations, semi-empirical extrapolations may be made to predict various thermodynamic properties, ionic radii, ionization potentials, etc., and to estimate the relative stabilities of various oxidation states. Fricke²⁰ has summarized the atomic-structure calculational techniques together with most of the predictions of chemical and physical properties in his recent review article.

Atomic binding energies for inner-shell electrons, and hence characteristic K- and L-series x-ray transition energies, have also been predicted for the superheavy elements using the relativistic atomic calculations.²¹⁻²⁴ These predictions may be made with a high degree of accuracy and after minor semi-empirical adjustments have been made to account for higher-order effects not directly included in the calculations; it is generally felt that uncertainties in the K- and L-x-ray transition energies are ≈ 100 eV. Taken together with predictions of x-ray transition rates,^{23,25,26} inner-shell x-rays may be used with a high degree of confidence in searches for superheavy elements. The recent calculations of Carlson and Nestor²⁴ support this contention.

The alpha decay and fission of superheavy nuclei are predicted to be substantially different from those of known nuclei. This is because of the increased Coulomb energy associated with the higher charge of superheavy nuclei. For example, the energy of an emitted alpha particle^{6,8} should increase from about 4 MeV for uranium to 7 MeV for element 114 and to 10 MeV for element 116. Similarly, in going from uranium to element 114, the average fission-fragment total kinetic energy²⁷ should increase from about 172 MeV to about 235 MeV, and the average number of neutrons released per fission²⁷ should increase from about 2.4 to over 10.

Searches in Nature

If superheavy elements exist in nature, they most probably were produced in supernovae by the multiple capture of neutrons (the r-process). In this process, a given nucleus successively increases its mass by capturing one or more neutrons and increases its proton number by emitting a beta particle, and so on. Many naturally occurring nuclei were made in this way. Because

the surface tension of a nucleus decreases as neutrons are added to it, the heavy neutron-rich nuclei that must be formed in order to reach the island can end the process by undergoing fission. Although this conclusion is not definitely established, it appears unlikely that superheavy nuclei can be made in nature by means of the r-process.²⁸ Other methods by which superheavy elements conceivably could have been produced in nature include ejection from a rotating neutron star^{29,30} and reactions between two heavy nuclei.³¹

Terrestrial Samples

The first search for superheavy elements in nature was made in 1968 by Thompson and his co-workers.³² They used the predicted large number of neutrons per fission of a superheavy nucleus to search for eka-platinum, element 110, in a variety of samples. They also used activation analysis, mass spectrometry, x-ray fluorescence, and direct spontaneous-fission counting, but all of their results were negative.

Since spontaneous fission is relatively rare among nuclides of the known elements, observation of fission events is a very sensitive analytical tool (sensitivity $\approx 10^{-14}$ g/g for a half-life of 10^8 yr). These events can be observed either directly in suitable counters or by visualizing the intense ionization-damage tracks due to the fragments in dielectric track detectors. The first claim to the discovery of superheavy elements was made in 1969 by Flerov and Perelygin using these methods in their search for element 114 in various Pb-bearing samples.³³ Subsequent investigations by many experimenters were negative,³⁴⁻³⁶ and it is now generally conceded, even by the original authors, that the fission events were due to cosmic-ray-induced fission of Pb in the samples.

Many similar studies based on spontaneous-fission detection methods, but covering a broad spectrum of samples and superheavy elements, have been conducted since then.³⁴⁻³⁶ Malý and his co-workers have attributed excess fission activity over that expected from uranium to the presence of superheavy elements in Bi_2O_4 , PbO, Au, and Pt samples,³⁷ as well as in HfO_2 samples.³⁸ These workers also claim chemical enrichment in thermally fractionated HfCl_4 at 425°C and postulate radioactive superheavy-element growth-decay chains to explain the time-variant fission activity.

A somewhat different approach involving isotope-separation methods has been used by Stéphan and his co-workers.³⁹ From a variety of natural samples, they collected mass fractions in the range 293-314 in an electromagnetic isotope separation and followed this by neutron irradiation to induce fission which is detected using dielectric track detectors. Induced fissions were observed in these experiments but it was later shown to arise

from a collection of molecular ions of Th and of U.⁴⁰ Similar experiments have been reported by Forsling, who isotope separated samples of neutron-irradiated radiolead.⁴¹ Alpha activity was reported at mass number 292 and could not be attributed to any known impurity, but the experiments were discontinued.

Older reports of the enigmatic ≈ 4.5 -MeV alpha emitter and others in natural samples led Cherdyn'tsev to claim discovery of element 108, which he named sergenium, from his studies of sulphide ores, osmiridium, volcanic, and other samples.⁴² This work could not be repeated by others and is generally discounted.

Very recently, Gentry *et al.* reported the observation of characteristic L-x-ray lines which were ascribed to elements 116, 124, 126, and 127 in proton bombardments of the inclusions of giant pleochroic halos from Madagascar monazite.⁴³ These halos, a natural phenomenon, are attributed to the radiation-damage sphere surrounding an inclusion of alpha-active isotopes in such minerals as mica. The identifications were based on the observation of only one supposed L-x-ray emission line per superheavy element in the samples. Although great excitement was generated in the scientific community, this report was ill-founded, as subsequent workers were quick to demonstrate interferences from proton-induced nuclear reactions on the normal constituents of the sample.^{44,45} In particular, a gamma ray produced in the $^{140}\text{Ce}(p, n)$ reaction, nearly identical in energy as that expected for the $L_{\alpha 1}$ line of element 126, was shown to quantitatively account⁴⁵ for the effect in Ref. 43.

Ketelle *et al.*⁴⁶ were able to place extremely low limits on the concentration of any superheavy elements of reasonable half-life in Madagascar monazite of similar origin to Ref. 43, by using a neutron-multiplicity counting method. Stéphan *et al.*,⁴⁷ using the isotope separation methods of Refs. 39 and 40, also examined Madagascar monazite and were able to set concentration limits orders of magnitude lower than those implied by Gentry *et al.* in Ref. 43. Finally, Sparks and co-workers^{48,49} have used an elegant photon-induced x-ray fluorescence technique with intense synchrotron radiation to examine the inclusions of the giant halos directly. These experiments have the potential of being more sensitive than those performed by Gentry *et al.*⁴³ and thus are considered by many to be definitive. In a forthcoming publication,⁴⁸ the results obtained for 11 Madagascar giant-halo inclusions have shown no evidence for superheavy elements at concentration levels at least a factor of 10 less than those implied in Ref. 43. In more recent experiments by these workers,⁴⁹ a variety of halo specimens were examined, including sample #19D previously studied by Gentry *et al.*,⁴³ but rigorous analyses of these data have not yet been performed. Thus, to date, it appears that the highest atomic-number element ever identified using characteristic

x-rays is element 105.⁵⁰ The origin of the enigmatic giant halos is not established, although Holbrow⁵¹ has recently proposed an origin based on the fission of ^{244}Pu .

Extra-terrestrial Samples

Anomalous excess fissionogenic-xenon isotope abundances in meteorites of the Fe-Cr-S type (Allende) were interpreted by Anders *et al.* as evidence for a now-extinct superheavy element.⁵² From the assumed condensation history of the meteorite and the predicted chemical properties of elements in this region, element 115 (eka-bismuth) was selected as the best candidate, but elements 113 and 114 were other possibilities. Similar explanations for excess fissionogenic xenon in meteorites were proposed by Dakowski,⁵³ but fissionogenic xenon from ^{244}Pu was later shown to account for this anomaly. Fission of ^{244}Pu was also considered by Anders *et al.*,⁵² but they discounted it as the origin of their anomaly.

Fossil tracks in meteorites and in lunar soil samples have been ascribed to the fission tracks from now-extinct superheavy elements on the basis of the excess track lengths.⁵⁴ The long fission tracks would indicate an increased fission-fragment kinetic energy, as predicted for superheavy elements with $Z \gtrsim 110$. However, tracks from Fe-group cosmic rays have subsequently been shown to cause interferences.⁵⁵

Observations of the charge spectrum of primary, ultra-relativistic cosmic rays in nuclear emulsions and in plastic track detectors have revealed two events that were ascribed to elements 104 and 107.^{56,57} However, as pointed out by Fleischer, Price, and Walker,⁵⁸ large errors in these measurements do not rule out Cm, Pu, or U.

Negative results have been obtained in many additional searches for superheavy elements involving both terrestrial and extra-terrestrial samples. These experiments are summarized in Refs. 7, 34, 36, and 59.

Searches at Accelerators

There are three possible mechanisms by which superheavy elements might be produced in reactions between two heavy nuclei. These are compound-nucleus formation, transfer reactions, and fission. In the former method a target and projectile are brought together with sufficient energy that a nearly spherical compound nucleus is formed. The most favorable reaction of this type is expected to be $^{48}\text{Ca} + ^{248}\text{Cm} \rightarrow ^{296}116^*$. In most cases the system will undergo fission, either before or after a compound nucleus is formed. But

with a small probability, the compound nucleus can de-excite by the emission of neutrons or alpha particles and some gamma rays. The resulting ground-state superheavy nuclei are in turn predicted to decay successively by the emission of high-energy alpha particles and by electron capture.⁸ This could lead to some final superheavy-element products that live for years.

In a transfer reaction, a somewhat heavier projectile is used. A portion of the projectile combines with the target to form a superheavy nucleus, and the remaining portion carries away some energy and angular momentum that would otherwise remain as undesirable excitation energy and rotational energy. Although this method offers certain advantages, transfer reactions will probably suffer from very low cross-sections for transferring the desired fraction of the projectile.

In the fission mechanism, a heavy target and projectile, such as uranium plus uranium, fuse to form a massive excited nucleus. This nucleus then undergoes fission, the hope being that one of the fission fragments will be a superheavy nucleus. This method will probably not work because the fission fragments will be very elongated at birth. Because of this large distortion, any superheavy nucleus formed as a fission fragment will itself probably undergo fission rather than de-excite by neutron emission.

Whether or not any of the above reaction mechanisms will be successful in producing superheavy nuclei depends critically on the magnitude of nuclear dissipation, the transfer of energy of collective motion into internal excitation energy. Should it turn out that nuclear dissipation is large, it may prove impossible to bring the target and projectile from their initial configuration of two touching spheres to a final configuration that is nearly spherical. Work on this point is proceeding vigorously, but present information is inconclusive.^{60,61}

The first attempt to produce superheavy elements artificially was made by Thompson and his co-workers^{62,63} and by Ghiorso and his co-workers⁶⁴ with reactions of the type $^{40}\text{Ar} + ^{248}\text{Cm} \rightarrow ^{288}114^*$. Upper limits were placed on production cross-sections and half-lives, but the neutron numbers of the product nuclides were sufficiently low that the nuclei did not survive until detection.

In the reaction $^{84}\text{Kr} + ^{232}\text{Th} \rightarrow ^{316}126^*$, Lefort and his co-workers observed alpha particles with energies between 13 and 15 MeV from the reaction products.⁶⁵ This was taken as evidence for the production of element 126, although subsequent experiments using more direct identification techniques were negative. These techniques included simultaneous kinetic energy, time-of-flight, and magnetic rigidity measurements and were 5 to 10 times more sensitive than the initial experiments.

Ions as heavy as ^{136}Xe in combination with targets of ^{238}U have been used in attempts to produce superheavy elements. Flerov and Oganessian

observed spontaneous-fission activity in a chemically isolated sulphide fraction containing the elements Os and Bi after long bombardments.⁶⁶ The half-life is ≈ 150 days, and the production cross-section is $\approx 10^{-3.3}$ cm². However, neutron-multiplicity determinations indicate that the average number of neutrons per fission is only 1.5 to 3.5, which is substantially lower than the number expected for the fission of a superheavy nucleus. These workers plan additional, more definitive experiments. Other heavy-ion projectiles have included ^{74,76}Ge and ⁶⁵Cu in combinations with ²³²Th and ²³⁸U targets; these attempts are summarized in Ref. 36. Recent attempts to produce superheavy elements in U-U collisions at the UNILAC in Darmstadt, West Germany, have not yielded any evidence for superheavy elements⁶⁷ nor have attempts at the SuperHILAC at Berkeley in reactions of ²⁴⁸Cm with ⁴⁸Ca projectiles.⁶⁸

Secondary reactions induced by energetic heavy recoils in the interaction of high-energy protons with suitable targets have also been used in accelerator searches for superheavy elements. Marinov *et al.* claimed the production of element 112, eka-mercury, after observing spontaneous-fission events in the chemically isolated mercury fraction following a long-term 24-GeV proton irradiation of W.⁶⁹ Subsequent experiments attributed at least 70% of these events to the presence of ²⁵²Cf contaminants. A repetition of the original experiment by members of the Marinov group failed to confirm the initial results.⁷⁰ Many other attempts to reproduce the initial results of Marinov *et al.* have also been negative.³⁶ It now appears unlikely that this secondary-nuclear-reaction mechanism will produce superheavy elements, as the heaviest product ever observed has been ²⁴⁸Cf with a production cross-section of $\approx 10^{-3.6}$ cm². Although Malý has observed very energetic recoils in the bombardment of heavy elements with electrons,⁷¹ no evidence was found for the production of superheavy elements.⁷²

The predicted island of superheavy nuclei appears to have eluded discovery thus far. But with the German UNILAC now coming into operation, and with heavy-ion accelerators under construction at Oak Ridge (USA), Dubna (USSR), and Caen (France), future explorers will be better equipped than those in the past. The new accelerators will provide additional projectiles, more intense beams, and better energy resolution. Also, several new experimental techniques have been developed to assist in the identification of superheavy elements. These include the simultaneous measurement of the atomic number, mass number, and kinetic energy of recoil nuclei. Such methods as resonance-ionization spectroscopy and the observation of in-flight x-rays may also prove useful.

As we try to journey to the island of stability, we should keep in mind that we are entering a practically unexplored realm of science. We may or may not find the island that we seek. If not, we may make even more important

discoveries that are presently unforeseen. Irrespective of the outcome, we are on one of the voyages of the century.

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