

Present at the Creation

When they synthesized elements 114 and 116, Russian and Livermore scientists confirmed decades-old predictions of the existence of superheavy elements with comparatively long lifetimes.

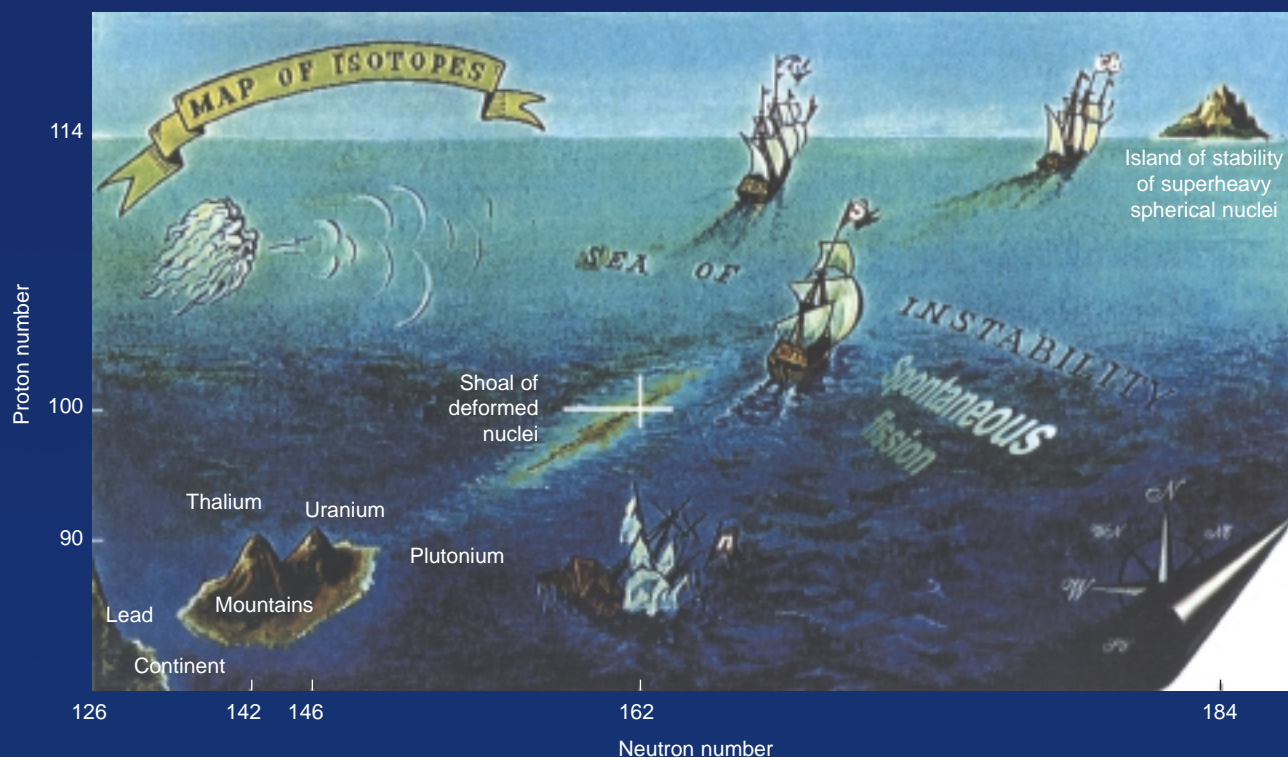
ELEMENTS that do not exist in nature—that have been created in a laboratory—are unstable. After hours or days of one element bombarding another with enough energy for both to fuse, the resulting new element typically is born and begins to decay instantly.

Neptunium and plutonium (elements 93 and 94) were the first elements created in a laboratory, at the University of California at Berkeley in 1940. Scientists have since fabricated many more elements, each one heavier and with a shorter half-life than the one before it.

In the 1960s, a few physicists predicted that some elements around element 114 would survive longer than any of their synthesized predecessors. Early estimates for the half-lives of these more stable elements were as high as billions of years. Later computer modeling reduced the anticipated half-lives to seconds or minutes before the element began to decay.

Half-lives of seconds or minutes may seem brief. But consider that various atoms of element 110 created in the laboratory have had half-lives ranging from 100 microseconds to 1.1 milliseconds. The only atom of element 112 that had been created before 1998 had a lifetime of 480 microseconds. As described further in the [box on p. 21](#), the long-lived nuclei of elements around element 114 would comprise an “island of stability” in a “sea” of highly unstable elements.

When a collaboration of Russian and Livermore scientists at the Joint Institute for Nuclear Research in



Map of the voyage to the island of stability.

Dubna, Russia, created element 114 in 1998, the first atom survived for 30 seconds before it began to decay, a spontaneous process that leads to the creation of another element with a lower number on the periodic table. (See the [box on pp. 18–19](#) for more information on stability and instability.) A total of 34 minutes elapsed before the final decay product fissioned, splitting in two the surviving nucleus. These lifetimes may seem brief, but they are millions of times longer than those of other recently synthesized heavy elements.

Since that groundbreaking effort in 1998, the team has created another atom of element 114. This one has a different number of neutrons and thus a

different mass, thereby making it a different isotope of element 114. The team has also created several previously undiscovered isotopes of elements 112, 110, and 108 to which element 114 decayed. More recently, the team added element 116 to the periodic table with the creation of three atoms of the element in a series of experiments.

Nuclear chemist Ken Moody leads the Livermore portion of the international collaboration. “In 1998, we proved that there really was an island of stability,” he said. “We proved that years of nuclear theory actually worked.”

The collaboration began in 1989, with heavy element chemist Ken Hulet

representing Livermore and Yuri Oganessian, scientific director of the Flerov Laboratory of Nuclear Reactions at the Joint Institute, leading the Russians. In the early 1990s, the U.S.–Russian team discovered two isotopes of element 106, one isotope of 108, and one of 110 at the Dubna institute.

“In 1990, when Ron Lougheed, who has since retired, and I went to Dubna, we were the first U.S. scientists to perform experiments at that institute,” adds Moody. “Remember what was happening then. The Berlin Wall had just fallen, and Eastern Europe was in turmoil. The early days of the collaboration were definitely interesting.”

Photo of Russian–Livermore team.



Forty Days and Nights

Noah's flood could have come and gone in the time it took the collaboration to create the first atom of element 114. For 40 days of virtually continuous operation, calcium ions bombarded a spinning target of plutonium in Dubna's U400 cyclotron. While the first atom of element 114 was actually created on November 22, 1998, Russian researchers discovered it in data analysis and communicated the news to Livermore on December 25, 1998—quite the Christmas present.

A Primer on Stability and Instability

Why should element 114 be so much more stable and long-lived than so many of its synthesized predecessors? The answer lies in basic chemistry.

The nucleus of an atom is surrounded by one or more orbital shells of electrons. The electron configurations of atoms of the many elements vary periodically with their atomic number, hence “the periodic table of the elements.”

Elements with unfilled shells seek out electrons in other elements to fill them. These include carbon, oxygen, and all of the “reactive” elements that want to react with other elements. This is the basis of covalent bonding. The noble gases (on the far right column of the periodic table) have a completely filled outer electron shell and hence are highly stable. They are termed noble because they are “aloof,” with no desire to react with other elements.

Protons and neutrons are in analogous shells within the nucleus. The proton shells of helium, oxygen, calcium, nickel, tin, and lead are completely filled and arranged such that the nucleus has achieved extra stability. The atomic numbers of these elements—2, 8, 20, 28, 50, and 82—are known as “magic numbers.” These same numbers plus 126 are magic numbers for neutrons. Notice that the magic numbers are all even. No truly stable element heavier than nitrogen has an odd number of both protons and neutrons. Elements with even numbers of protons and neutrons make up about 90 percent of Earth's crust.

A nucleus is “doubly magic” when the shells of both the protons and neutrons are

filled. Lead-208 has 82 protons and 126 neutrons, both of which are magic numbers. Lead-208 is thus doubly magic and seems to be virtually eternal.

A long-lived, stable element such as lead does not decay. However, all elements with an atomic number greater than 83 (bismuth) exhibit radioactive decay. Decay may happen in several ways. For heavy elements, an unstable or radioactive isotope usually decays by emitting helium nuclei (alpha particles) or electrons (beta particles), leaving a daughter nucleus of an element with a different number of protons. This process typically continues until a stable nucleus is reached. Plutonium, for example, decays ultimately to lead.

The heavy elements that have been created in the laboratory are so unstable that they decay almost immediately and have extremely

Periodic table

1 H Hydrogen																	2 He Helium
3 Li Lithium	4 Be Beryllium											5 B Boron	6 C Carbon	7 N Nitrogen	8 O Oxygen	9 F Fluorine	10 Ne Neon
11 Na Sodium	12 Mg Magnesium											13 Al Aluminum	14 Si Silicon	15 P Phosphorus	16 S Sulfur	17 Cl Chlorine	18 Ar Argon
19 K Potassium	20 Ca Calcium	21 Sc Scandium	22 Ti Titanium	23 V Vanadium	24 Cr Chromium	25 Mn Manganese	26 Fe Iron	27 Co Cobalt	28 Ni Nickel	29 Cu Copper	30 Zn Zinc	31 Ga Gallium	32 Ge Germanium	33 As Arsenic	34 Se Selenium	35 Br Bromine	36 Kr Krypton
37 Rb Rubidium	38 Sr Strontium	39 Y Yttrium	40 Zr Zirconium	41 Nb Niobium	42 Mo Molybdenum	43 Tc Technetium	44 Ru Ruthenium	45 Rh Rhodium	46 Pd Palladium	47 Ag Silver	48 Cd Cadmium	49 In Indium	50 Sn Tin	51 Sb Antimony	52 Te Tellurium	53 I Iodine	54 Xe Xenon
55 Cs Cesium	56 Ba Barium	See Lanthanides	72 Hf Hafnium	73 Ta Tantalum	74 W Tungsten	75 Re Rhenium	76 Os Osmium	77 Ir Iridium	78 Pt Platinum	79 Au Gold	80 Hg Mercury	81 Tl Thallium	82 Pb Lead	83 Bi Bismuth	84 Po Polonium	85 At Astatine	86 Rn Radon
87 Fr Francium	88 Ra Radium	See Actinides	104 Rf Rutherfordium	105 Ha Hassium	106 Sg Seaborgium	107 Bh Bohrium	108 Hs Hassium	109 Mt Meitnerium	110	111	112		114		116		
Lanthanides		57-71 Ln* Lanthanides	57 La Lanthanum	58 Ce Cerium	59 Pr Praseodymium	60 Nd Neodymium	61 Pm Promethium	62 Sm Samarium	63 Eu Europium	64 Gd Gadolinium	65 Tb Terbium	66 Dy Dysprosium	67 Ho Holmium	68 Er Erbium	69 Tm Thulium	70 Yb Ytterbium	71 Lu Lutetium
Actinides		89-103 An* Actinides	89 Ac Actinium	90 Th Thorium	91 Pa Protactinium	92 U Uranium	93 Np Neptunium	94 Pu Plutonium	95 Am Americium	96 Cm Curium	97 Bk Berkelium	98 Cf Californium	99 Es Einsteinium	100 Fm Fermium	101 Md Mendelevium	102 No Nobelium	103 Lr Lawrencium

The box on p. 22 shows the “recipe” for the early Dubna experiments that created isotopes of element 114. Plutonium, with an atomic number, or Z , of 94, and calcium, $Z = 20$, add up to the necessary $Z = 114$. By fusing plutonium-244, an isotope of plutonium with 150 neutrons, and calcium-48, a neutron-rich isotope with 28 neutrons, a compound nucleus with 114 protons and 178 neutrons ($150 + 28$) would in theory be possible. In fact, however, when the plutonium-244 and calcium-48 nuclei

collide with enough energy to overcome their mutual electrostatic repulsion, the compound nucleus has excess energy. A few neutrons evaporate to de-excite the nucleus and produce an isotope with 175 neutrons.

To discover whether new elements were created by the bombardment of plutonium, the team was interested in finding “events” comprising a series of alpha decays ending with spontaneous

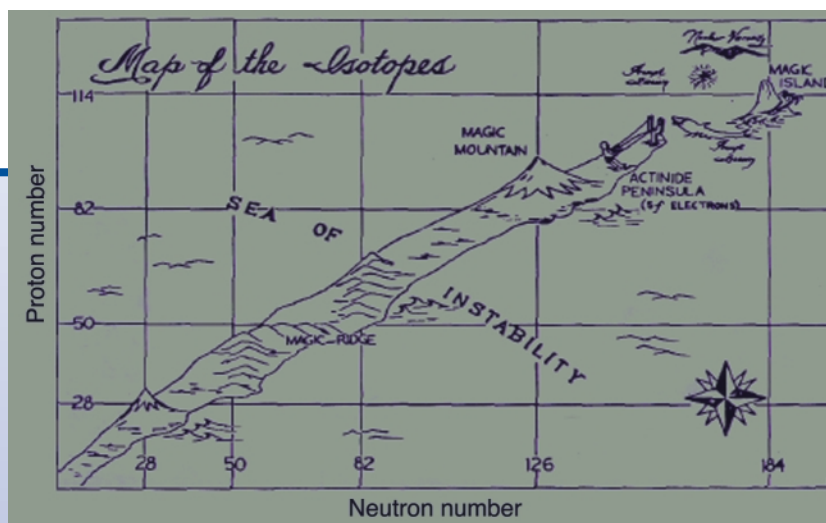
fission. In alpha decay, an isotope loses an alpha particle, which is two protons and two neutrons (or a helium nucleus). For example, an atom of element 114 with 175 neutrons (described as isotope 114-289) would emit an alpha particle, thereby becoming isotope 112-285, having lost 2 protons and 2 neutrons. The atom of 112-285 would become 110-281, which would become 108-277. At some point, fission would occur, ending the process.

At the same time, however, unwanted nuclei generated by

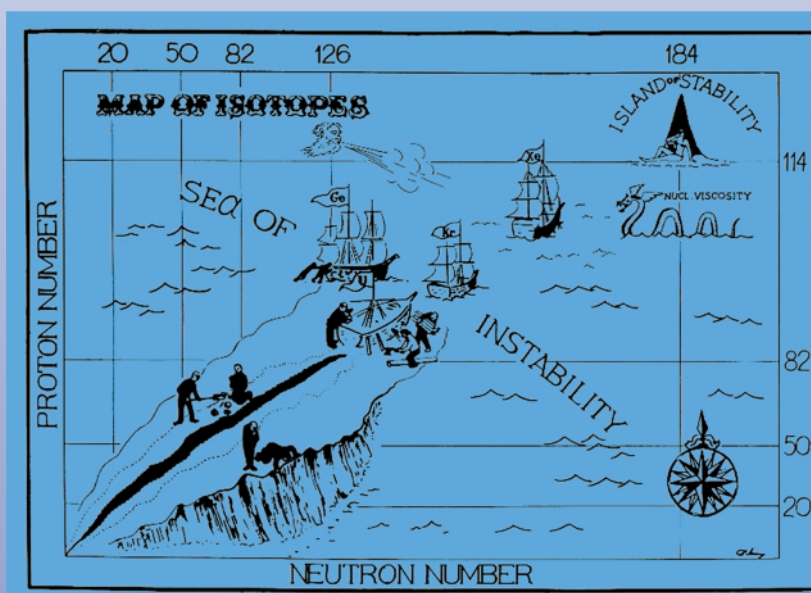
short half-lives and thus lifetimes. How quickly a particular isotope decays is measured by its half-life. Plutonium-239, which decays very slowly, has a half-life of about 24,000 years, while plutonium-238's half-life is just 88 years. Half-lives are a result of a statistical process. If an experiment produces only one atom, then a half-life cannot be determined. Thus, with one or a few atoms, scientists talk instead about lifetimes.

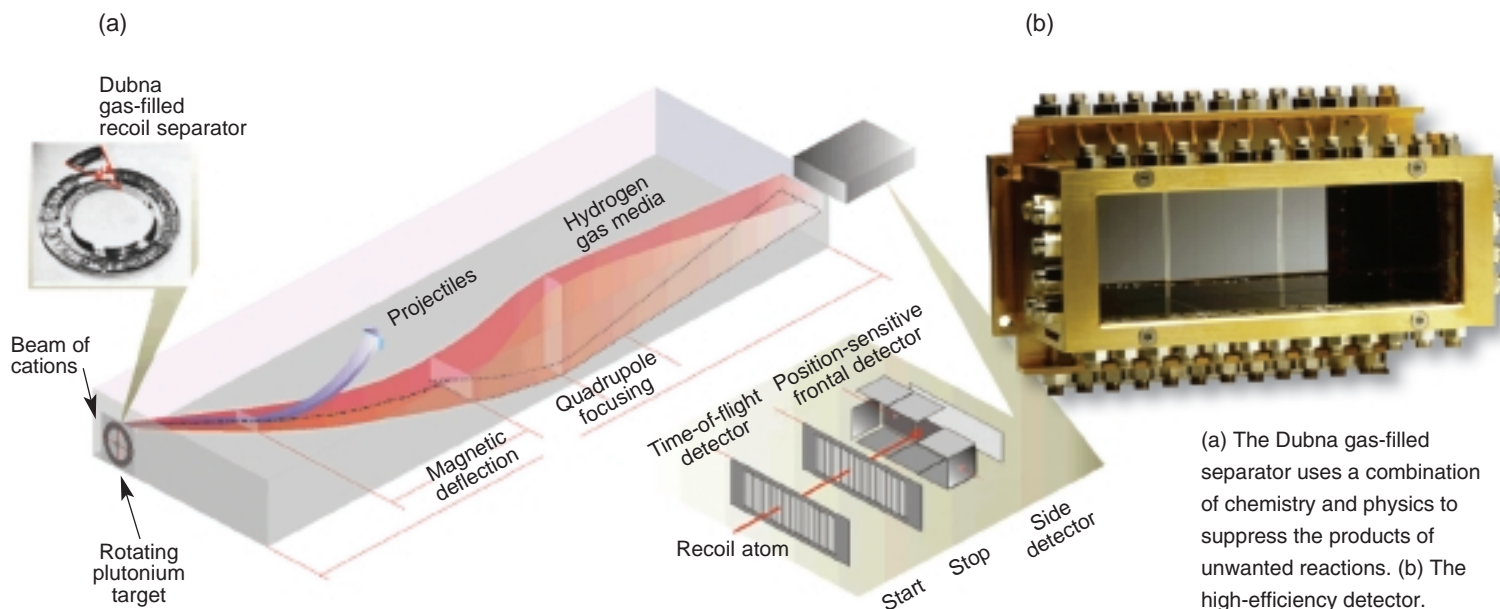
In the mid-1960s, a physicist in the U.S. predicted that the next magic proton number above 82 would be 114, not 126, and that an atom with a doubly magic nucleus of 114 protons and 184 neutrons should be the peak of an island of stability. Russian scientists had come to the same conclusion at about the same time.

In the years since, increasingly sophisticated computer models have indicated that element 114 would exhibit significant nuclear stability even with neutron numbers as low as 175. Note that element 114 is expected to lie in the same column (or group) of the periodic table as lead. The two elements are expected to share many properties.



Nuclear theory (top) in the U.S. and (bottom) in Russia, in about 1969.





(a) The Dubna gas-filled separator uses a combination of chemistry and physics to suppress the products of unwanted reactions. (b) The high-efficiency detector.

the experiment also undergo alpha decay and fission, mimicking the decay sequence of element 114. Trillions of these unwanted nuclei are produced every day, whereas the expected production rate for an element 114 isotope was much less than one atom per day. To deal with the problem of unwanted nuclei in earlier experiments, Dubna scientists had developed a gas-filled mass separator to separate unwanted nuclei from the desired ones. "It worked marvelously," says Moody.

Heavy-element reaction products recoil from the spinning plutonium target wheel and enter the mass separator, a chamber filled with low-pressure hydrogen gas confined between the pole faces of a dipole magnet. The magnetic field is adjusted so that, for the most part, only the nuclei of interest pass through to the detector array.

The desired nuclei are focused with a set of magnetic quadrupoles, pass through a time-of-flight counter, and are captured by a position-sensitive detector. A signal from the time-of-flight counter allows the team to distinguish between the effect of

products passing through the separator and the radioactive decay of products that are already implanted in the detector. The flight time through the counter is also used to discriminate between low- and high-Z products, because heavier elements travel more slowly. The position-sensitive detector lowers the rate of background interference, allowing scientists to identify and ignore unwanted products.

During 40 days in November and December 1998, with ten-thousand trillion ions per hour of calcium-48 bombarding the plutonium target, the team observed the signals of just three spontaneous fission decays. Three synthesized compound nuclei had been created and passed through the separator before fissioning. Two of them lasted about 1 millisecond each and proved to be products from the decay of the nuclear isomer of americium-244.

Only one of the events involved an implant in the detector followed by three alpha decays in the detector array. This isotope of element 114 (114-289) had a lifetime of 30.4 seconds. It decayed to element 112, which, with a

lifetime of 15.4 minutes, decayed to element 110. Element 110, with a lifetime of 1.6 minutes, then decayed to element 108, which decayed by spontaneous fission.

In 2000 and 2001, the collaboration performed three experiments in which a curium-284 target was bombarded with calcium-48 ions to create element 116. The team chose this combination of isotopes because they would produce isotopes of element 116 that should decay to the previously observed isotopes of element 114.

Researchers produced the super-heavy isotope 116-292 once in each of these experiments. They also created some other isotopes repeatedly. Isotopes 114-288, 112-284, and 110-280 have been found five times, lending credibility to several experimental results. However, the first atom of 114-289 with the 30.4-second lifetime has yet to be replicated.

In the Final Analysis

The recipe for element 114 on p. 22 refers to the analysis of 7 gigabytes of data from the first experiments. The team has since accumulated another

A Stormy Voyage to the Island of Stability

As of November 2001, scientists throughout the world had synthesized 20 elements that do not exist in nature. The ones up to meitnerium (109) have been given official names. Elements 110, 111, 112, 114, and 116 will not be named until their existence has been corroborated with several experiments or by several different groups. Recall that one of the fundamental tenets of science is reproducibility.

In 1940, Ed McMillan and his team at Berkeley bombarded uranium with neutrons to create neptunium (element 93). Then Glenn Seaborg and his colleagues created plutonium-238, the first isotope of plutonium (element 94), through the decay of neptunium-238, which they produced by bombarding uranium with deuterium (heavy hydrogen). Elements 99 and 100 were discovered in the debris of the first hydrogen bomb test in 1952 from the simultaneous capture of many neutrons by uranium. The heavy, highly radioactive uranium isotopes decayed quickly by beta emission down to more stable isotopes of elements 99 (einsteinium) and 100 (fermium). Elements 95, 96, 97, 98, and 101 were created by irradiating heavy nuclei with beams of alpha particles to boost the atomic numbers two steps at a time.

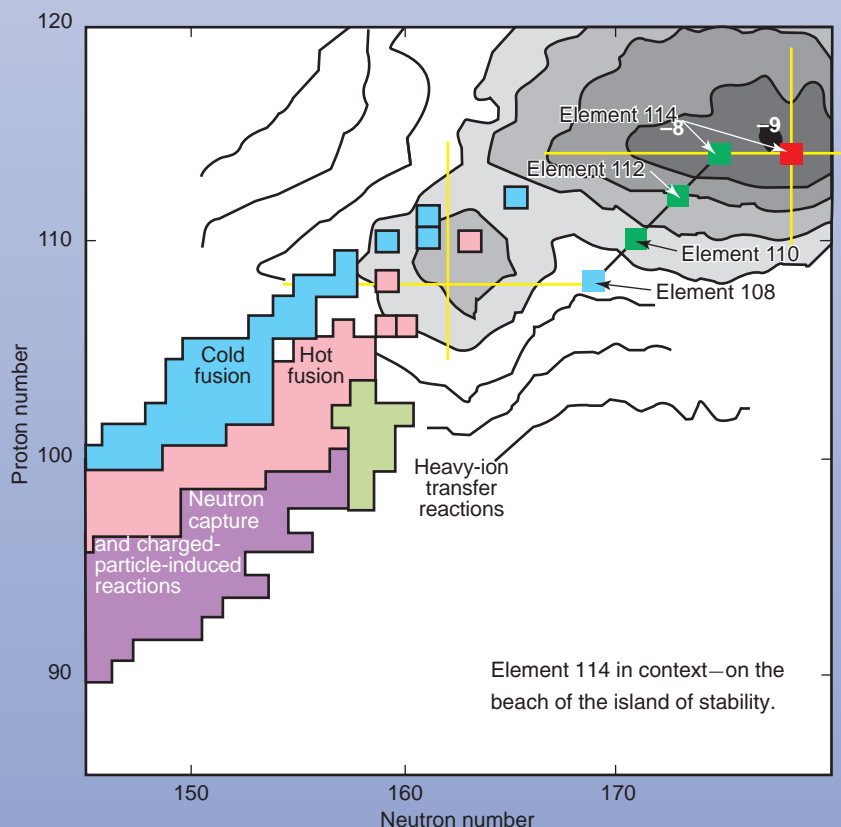
Beginning in the late 1950s, the new particle accelerators were capable of accelerating ions heavier than helium. First, ions of the lightest elements were directed at the heaviest elements. But it took excess energy to cause them to fuse, producing a very hot nucleus that tended to fission almost immediately. Known as “hot fusion,” this method yielded elements 102 through 106 by 1974. Many of these experiments included Livermore scientists.

In 1974, Yuri Oganessian at the Joint Institute at Dubna found that if heavier ions are directed at lead and bismuth, less energy was needed to create new elements. These two elements are extra-stable, and thus the resulting compound nucleus has less energy and is more likely to remain intact. This process is known as “cold fusion,” not to be confused with the discredited fusion energy process of the same name. Even with cold fusion, so few nuclei of the new element are produced during an experiment that existing detection techniques were not sensitive enough to find them.

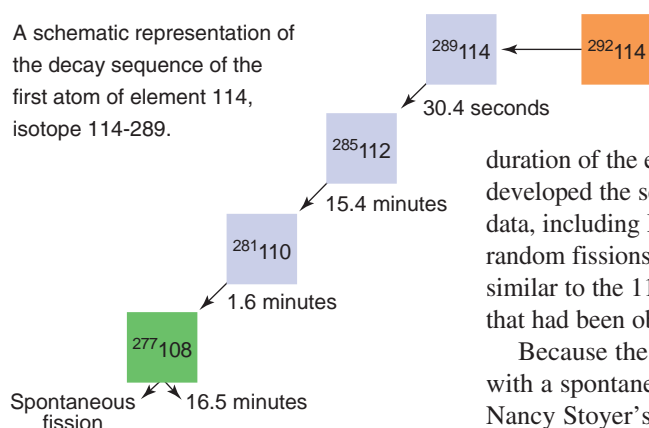
The field of synthesizing ever heavier elements went on hiatus for several years until sophisticated new separation and detection methods were developed in the early 1980s in Germany. German researchers were then able to create and detect elements 107, 108, and 109 in experiments that have since been corroborated such that these synthetic elements now have names. They also created isotopes of 110, 111, and 112, but these results have not yet been fully corroborated.

The German group, the Consortium for Heavy Ion Research at Darmstadt, Germany, has produced an isotope of element 112 that decayed into the same isotope of 110 that the Dubna–Livermore team found in 1994. This isotope had the same energy and lifetime, which is encouraging validation.

The voyage to the island of stability has been a stormy one. It took until 1998 to even reach the beach. As shown in the figure below, the island’s peak is still tantalizingly just out of reach.



A schematic representation of the decay sequence of the first atom of element 114, isotope 114-289.



number generator that places random fission events into the real data throughout the

duration of the experiment. Nancy Stoyer developed the search code that sifted the data, including Monte Carlo-generated random fissions, for decay sequences similar to the 114-289 decay sequence that had been observed experimentally.

Because the actual decay chains end with a spontaneous-fission event, Nancy Stoyer's search algorithm looks

backward from the planted fission event for candidate alpha-decay chains that match actual decay chains and end with a fission event. The number of returned "accidental" decay chains defines the probability that a decay sequence is random. For the first atom of element 114, the random probability was 0.6 percent. "If we eliminate decay chains in which all alpha events do not meet the Geiger-Nuttall relationship," says Moody, "the random probability falls to 0.06 percent. That's fantastic."

23 gigabytes of data, all requiring extensive analysis to verify the times and energies of the alpha decays. Valid decay sequences must fall within the alpha decay time and energy parameters of what is known as the Geiger-Nuttall relationship.

Scientists at Livermore and Dubna analyzed the data in parallel. Livermore gave the Dubna institute a computer workstation for the Russian scientists to use on that mountain of information. Nuclear chemists John Wild and Nancy Stoyer analyzed the data at Livermore. "These dual analyses were independent but were calibrated. In the end, our results agreed," says Wild.

The team must also confirm that the sequences they saw were not composed of random events. "The problem of randomness is real, especially for long-lived elements," adds Wild. "The longer the lifetime of a member of a decay sequence, the greater the probability that the decay could be random."

A novel Monte Carlo method to estimate the probability of whether a decay chain was random or the real thing was the brainchild of nuclear chemist Mark Stoyer. It is a pseudo-random

Recipe for a New Element

A Livermore chemist with a sense of humor developed this recipe to describe the creation of element 114.

Ingredients:

- 2 grams calcium-48, a rare neutron-rich isotope of calcium. Out of every 100,000 atoms of calcium, only 187 atoms are calcium-48.
- 30 milligrams plutonium-244, the most neutron-rich, long-lived isotope of plutonium. The world's supply of this isotope is only 3 grams.
- The U400 cyclotron at Dubna, Russia, to accelerate calcium ions to 10 percent the speed of light (236 megaelectronvolts).
- A gas-filled recoil separator for removing unwanted reaction products.
- A position-sensitive detector for capturing decays of reaction products.
- 2 computers, one for data acquisition and another for data analysis.
- Numerous Russian technicians and accelerator operators.
- 19 Russian scientists.
- 5 American scientists.

Directions: Combine the first seven ingredients, using 0.3 milligrams per hour of calcium-48. Add lots of patience, a dash of luck, and a dollop of inspiration. Simmer for about 6 months, 24 hours per day, 7 days a week. Use the last two ingredients to analyze 7 gigabytes of data for signature decay sequences of element 114. Garnish with several papers describing the results.

Serves: Very few. In two experiments, makes one atom of 114-289, the lifetime of which is 30 seconds, and two atoms of 114-288, each with a lifetime of 2 seconds.

New Elements Still to Come

The Livermore researchers are continuing its work to explore the southwest shores of the island of stability. With funding from the Laboratory Directed Research and Development program, they have begun efforts to add elements 115 and 113 to the periodic table. They are in the process of sending 22 milligrams of pure americium-243 to Dubna for the work on element 115.

Current exploration of the island of stability, or its beaches, is limited to stable targets and projectile beams. There exists no suitable combination of projectile and target to produce 114-298, the long-predicted highly stable isotope. The isotopes 114-289 and 114-288 require the most neutron-rich isotopes of plutonium and calcium. In the future, when radioactive beam accelerators are capable of producing intense beams of even more neutron-rich isotopes, researchers may venture farther toward the center of the island. For example, calcium-50 has a half-life of 14 seconds, far too short to gather material together to put into a conventional ion source. However, plans are for a radioactive beam facility to produce calcium-50 and accelerate it to energies required for the experiments well before it can decay. Thus, an isotope of element 114 with a mass of 290 or 291, two neutrons closer to the center of the island, may well be possible.

As scientists continue to explore for new elements, they expect that more spherical and longer-lived isotopes will be produced, which will most certainly require more sensitive detection schemes. Challenges abound.

Livermore researchers also want to study the chemical properties of elements 112 and 114. The combination of chemical and nuclear properties defines the usefulness of any nuclide. Most heavy elements exist in such small amounts, or for such short times, that no one has pursued practical applications for them. However, several heavy elements do have uses—americium is used in smoke detectors, curium and californium are used for neutron radiography and neutron interrogation, and plutonium is elemental in nuclear weapons. Although elements 114 and 116 have no immediate use, they do exist, and more of them can be manufactured when uses for them are found. Adds Moody, “Showing that the isotopes of element 114 produced by the collaboration have unique chemical properties will also provide proof that they are indeed a new element.”

—Katie Walter

Key Words: element 114, element 116, heavy elements, island of stability, Joint Institute for Nuclear Research in Dubna, Russia.

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