Chapter 8 The Search for "Heavy" Elements

The known chemical elements are arranged (Fig. 8-1) in a pattern that relates their chemical properties. That arrangement is called the "Periodic Table", and is the fundamental tool for physicists and chemists. The elements are arranged in groups (columns) indicative of similar chemical properties and periods (rows) indicative of electronic shell occupation. To a person who understands how to read the periodic table, an enormous amount of elementary properties can be discerned – for example, which element might bond with which other elements and in what ratios to form chemical compounds, which elements are metallic and which are not. The first periodic table was constructed in 1869 by Dmitri Mendeleev. Other scientists had noticed repeating patterns or periodicity in the various known elements (at that time only 63 elements were known), but Mendeleev was the first to boldly use his table to predict the existence of as yet unknown elements and actually leave spaces in his chart for them. Several elements (Ga and Ge for example) where found in subsequent years, based on his predictions of their expected masses and chemical properties, cementing the form and usefulness of the periodic table. UNESCO declared 2019 the International Year of the Periodic Table in honor of the 150th anniversary of the first periodic table.

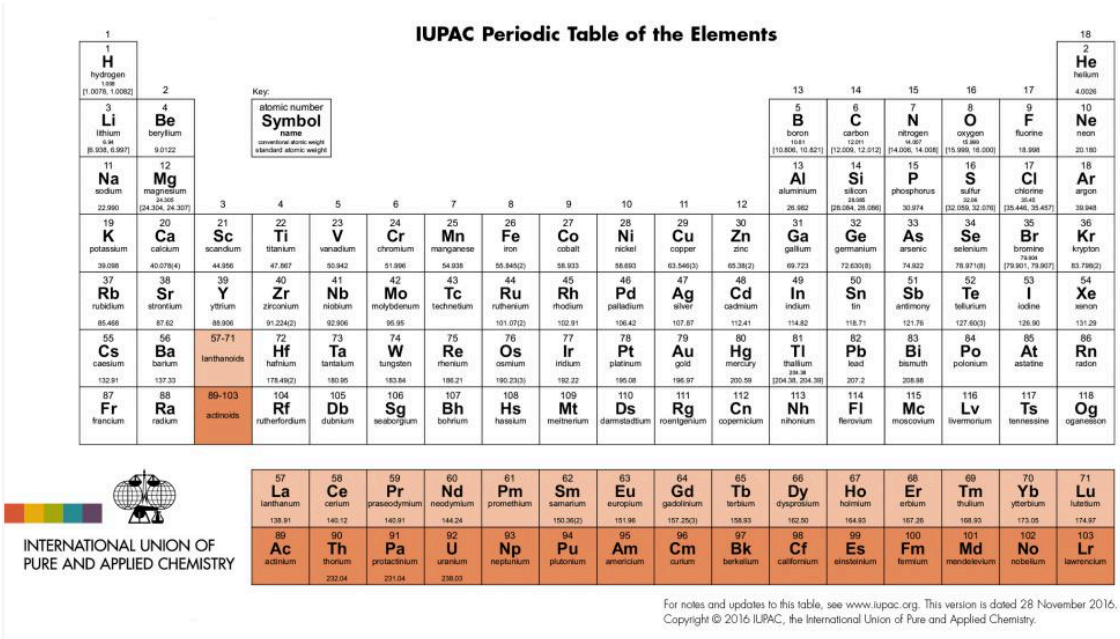

Fig. 8-1. The Periodic Table. Reference https://iupac.org/what-we-do/periodic-table-of-elements/

Each element is uniquely defined by proton number, Z. Changing the number of neutrons, N, does not changes the chemical properties significantly, but can make a **major** difference in the nuclear properties. This arrangement showing the number of neutrons and protons can be displayed in a chart of nuclides, which also conveys

important nuclear properties such as half-life and decay modes. The uppermost end of the chart of nuclides as known in 2019 is shown in Fig. 8-2. Elements through oganesson (Og, Z = 118) have been discovered, confirmed[1], and named (see Table 8-1).

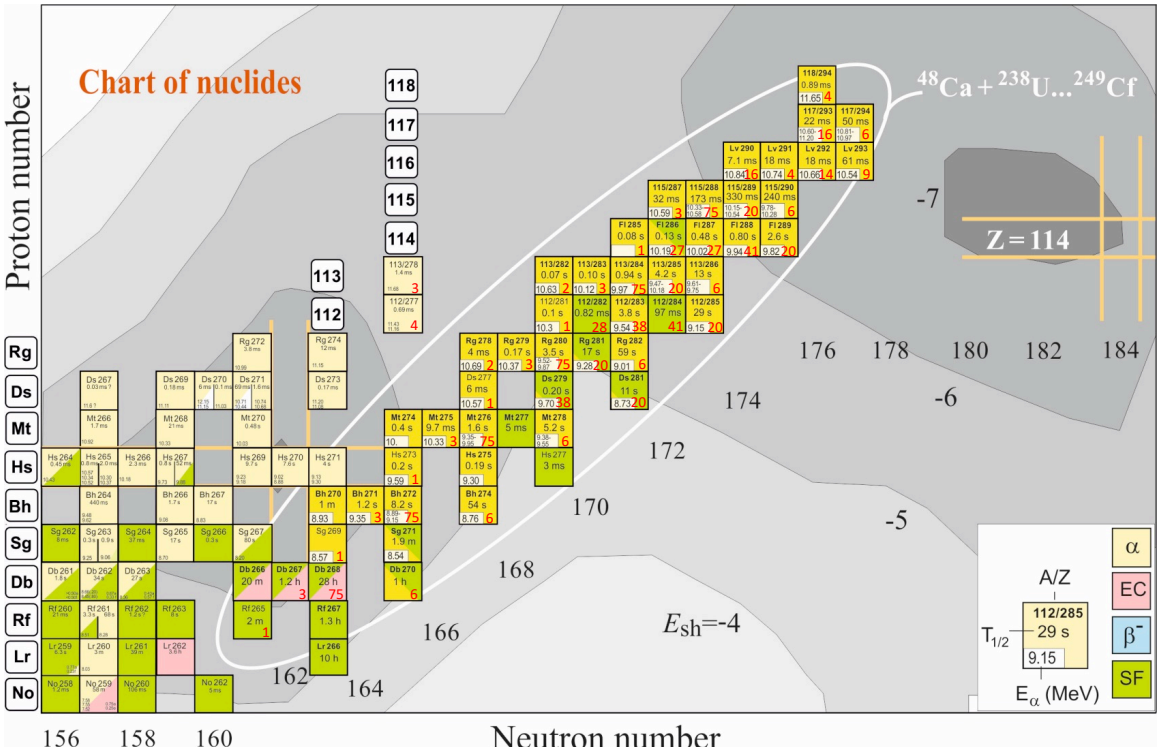

Fig. 8-2. The uppermost part of the chart of nuclides in June 2018. The color of the boxes indicates decay mode and text in the boxes indicate half-life and decay energies, The red numbers are the total number of atoms of that particular isotope that have been produced worldwide by June 2018. The grey shaded background is the magnitude of the shell corrections in a nuclear model and indicate one possible location for the next doubly magic "Island of Stability" or region of enhanced stablility near Z = 114 and N = 184.

When a nucleus captures a neutron, it often tries to correct for its neutron excess by beta decay, turning a neutron into a proton and thus creating an atom with atomic number *Z* increased by one unit. This commonly observed phenomenon suggests a way to create new elements of increased atomic number and thus to create ever more massive elements that are not found on Earth. Most of these elements are radioactive, with very short half-lives. However, theories of nuclear structure predict that at a certain atomic number, which is currently beyond present experimental limits, new long-lived nuclei can be created.

The most massive naturally occurring element on Earth is uranium (U), with a nucleus of 92 protons. In 1934, scientists started the search for more massive elements with 93 or more protons. They succeeded in 1940 when neptunium (Np, Z = 93) was synthesized at the University of California, Berkeley. Edwin McMillan and Philip Abelson observed Np while studying fission products produced in the bombardment of

[1] Oganesson has only been produced at the Flerov Laboratory of Nuclear Reactions in Dubna, Russia.

^{238}U with thermal neutrons. They found a radioactive reaction product that was not a fission product. This product was formed by the capture of a neutron to produce ^{239}U, which subsequently β^- decayed to ^{239}Np via the reaction

$$^{239}\text{U} + {}^{1}\text{n} \rightarrow {}^{239}\text{U (23.5 min)} \rightarrow {}^{239}\text{Np (2.36 d)},$$

where the half life of the nucleus is indicated in the parenthesis. McMillan and Abelson chemically separated this new element, Np, from the interfering fission products and chemically identified it as neptunium. Since this breakthrough discovery, scientists from all over the world have been trying to discover ever more massive artificially produced elements.

Plutonium (Pu, Z = 94) was discovered in 1941 by bombarding a uranium target with deuterons (a hydrogen nucleus with one proton and one neutron) in the 60-Inch Cyclotron at Berkeley. Glenn Seaborg, Arthur Wahl and Joseph Kennedy chemically separated neptunium from the target and detected alpha particles from the plutonium daughter nuclei, as:

$$^{238}\text{U} + {}^{2}\text{H} \rightarrow 2\ {}^{1}\text{n} + {}^{238}\text{Np (2.1 d)} \rightarrow {}^{238}\text{Pu (88 yr)}.$$

They chemically identified the isotope ^{238}Pu. Then, joined by Emilio Segre, they identified ^{239}Pu and showed that it was fissionable with thermal neutrons.

Once ^{239}Pu was discovered, there was the potential for using it as a new target to produce more massive elements because of its long half-life of 24,100 years. Because its half life is so long, scientists can keep the target for a long number of years.

In 1944 a ^{239}Pu target was bombarded with alpha particles at the 60-Inch Cyclotron to produce curium (Cm, Z = 96) through the following nuclear reaction

$$^{239}\text{Pu (24,110 yr)} + {}^{4}\text{He} \rightarrow {}^{242}\text{Cm (162.8 d)} + {}^{1}\text{n}.$$

After bombardment the material was sent to the Metallurgical Laboratory at The University of Chicago for chemical separation and identification of the new element. The element ^{242}Cm decays to ^{238}Pu by emitting alpha particles. The identification of curium was possible because the alpha decay of the daughter nucleus, ^{238}Pu, was already known and could be used as a signature for the identification of the curium precursor.

The discovery of americium (Am, Z = 95) soon followed when a ^{239}Pu target was bombarded with thermal neutrons in a nuclear reactor. Plutonium captured several neutrons and ultimately became americium (^{241}Am):

$$^{239}\text{Pu (24,110 yr)} + {}^{1}\text{n} \rightarrow {}^{240}\text{Pu (6,564 yr)} + {}^{1}\text{n} \rightarrow {}^{241}\text{Pu (14.4 yr)}$$

and

$$^{241}\text{Pu (14.4 yr)} \rightarrow {}^{241}\text{Am (432.7 yr)} + {}^{1}\text{n} \rightarrow {}^{242}\text{Am (16.0 h)} \rightarrow {}^{242}\text{Cm (162.8 d)}.$$

Americium was chemically separated from plutonium and further identified by observing its beta decay to the known ^{242}Cm isotope.

Once americium and curium were found and isolated in macroscopic amounts, they were used as targets to produce more massive elements through particle

bombardments. Berkelium (Bk, Z = 97) was produced by bombarding milligram quantities of americium with helium ions,

$$^{241}\text{Am}\ (432.7\ \text{yr}) + {}^{4}\text{He} \rightarrow {}^{243}\text{Bk}\ (4.5\ \text{h}) + 2\ {}^{1}\text{n}.$$

Rapid chemical techniques were developed in order to separate and identify this new short-lived element. Likewise, californium (Cf, Z = 98) was produced in a helium bombardment of a target made of microgram amounts of curium by

$$^{242}\text{Cm}\ (162.8\ \text{d}) + {}^{4}\text{He} \rightarrow {}^{245}\text{Cf}\ (45\ \text{min}) + {}^{1}\text{n}.$$

The identification of this element was accomplished with only the 5000 atoms produced in this experiment. The next two elements, einsteinium (Es, Z = 99) and fermium (Fm, Z = 100), were unexpectedly found in the debris from the "Mike" thermonuclear explosion that took place in the Pacific Ocean in 1952. Debris from the explosion was collected and analyzed at several laboratories, and the new elements were discovered in chemical separations of the material. Scientists explained the production of einsteinium and fermium through multiple neutron captures by the uranium used in the thermonuclear device followed by several successive beta decays, which ultimately resulted in atoms with atomic numbers 99 and 100.

The last three elements in the actinide series are mendelevium (Md, Z = 101), nobelium (No, Z = 102) and lawrencium (Lr, Z = 103). Mendelevium was truly a unique discovery because the new element was produced and identified virtually one atom at a time. Einsteinium was bombarded with helium ions to produce mendelevium through:

$$^{253}\text{Es}\ (20.5\ \text{d}) + {}^{4}\text{He} \rightarrow {}^{256}\text{Md}\ (78.1\ \text{min}) + {}^{1}\text{n}.$$

The production of mendelevium was estimated to be only a few atoms per experiment. The reaction products from the bombardment were collected on thin gold foils that were dissolved in an acid solution, and then chemically treated in order to separate and identify the Md atoms. This is commonly called the recoil method, and is used when small numbers of atoms are produced.

The discovery of nobelium was controversial. A team of scientists from several different laboratories claimed discovery in 1957. However, scientists from the United States and the Soviet Union could not confirm their findings. The original claim was proven to be false; the product that was thought to be nobelium was actually something completely different. Nobelium was finally produced and positively identified in 1958 through the following reaction:

$$^{246}\text{Cm}\ (4730\ \text{yr}) + {}^{12}\text{C} \rightarrow {}^{254}\text{No}\ (55\ \text{s}) + 4\ {}^{1}\text{n}.$$

The first identification of lawrencium was made at the Berkeley Laboratory's Heavy Ion Linear Accelerator (HILAC) in 1961. Several targets of californium isotopes were bombarded with beams of boron. The reaction products were collected on a Mylar tape and moved past a series of alpha detectors. The element lawrencium was identified on the basis of the known alpha decays of its descendant nuclei.

By observing the decay chain of their descendant nuclei, scientists discovered elements 104 through 118. Only a few atoms of these new elements were produced in

each experiment. The atoms were isolated from the target and beam material by using a particle separator, which separates atoms, based on their different masses. The atoms were then allowed to decay and the subsequent alpha particle decay products from the descendant nuclei were correlated to identify the unknown parent nucleus. The heaviest The decay chains used to identify Og and Sg are shown in Fig. 8-3. Different combinations of targets and projectiles were used in accelerators to produce these elements (see Table 8-2).

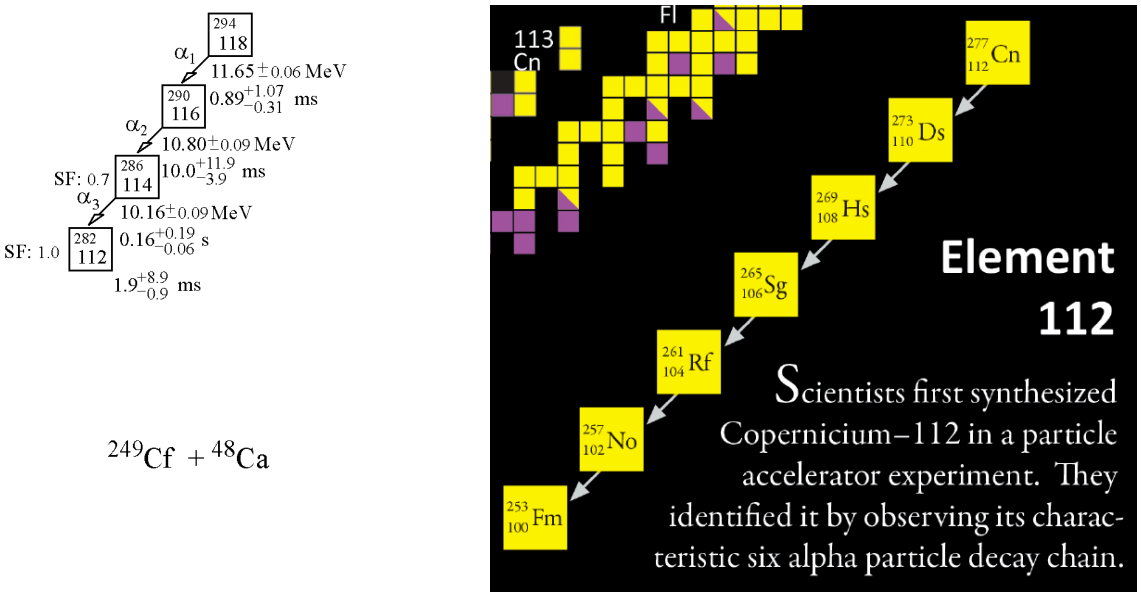

Fig. 8-3. The decay chain for identifying Og(118) and the decay chain for Sg(112) from the Nuclear Wall Chart.

There are two methods for producing new elements. First, cold-fusion reactions typically use more symmetric beam and target nuclei to produce a compound nucleus. Secondly, hot-fusion reactions use a more asymmetric beam and target nuclei to produce a compound nucleus with generally higher excitation energy. Both of these types of reactions utilize doubly magic nuclei, as either target or projectile in anattempt to increase the stability of the compound nucleus. Cold-fusion reactions have produced elements 104-112 and hot-fusion reactions have produced elements 113-118. Element 113 has also been produced with a cold-fusion reaction.

Rutherfordium (104), dubnium (105), and seaborgium (106) were synthesized and identified at Berkeley. Bohrium (107), hassium (108), and meitnerium (109) were synthesized and identified in the early 1980s at the Gesellschaft für Schwerionenforschung (GSI) laboratory near Darmstadt, Germany via cold-fusion nuclear reactions.

The decade of the 1990's brought three more new elements to the periodic table, elements named darmstadtium (110), roentgenium (111) and copernicium (112), all produced first via cold-fusion reactions at GSI (see Table 8-2). The half-lives of the produced isotopes were all in the milliseconds to microseconds range, and the production

rates continued to plummet.

In 1998, the Dubna/LLNL collaboration began to investigate superheavy element production using the hot-fusion reaction of beams of ^{48}Ca with various actinide targets, in an attempt to reach the Island of Stability long predicted to be at Z = 114 and N = 184. The first reaction was ^{48}Ca + ^{244}Pu → ^{292}114* and yielded element 114, now named flerovium. While the cross-sections for production of superheavy elements via cold-fusion reactions continued to plummet, those for hot-fusion reactions were unusually high, around a picobarn (see Fig. 8-4).

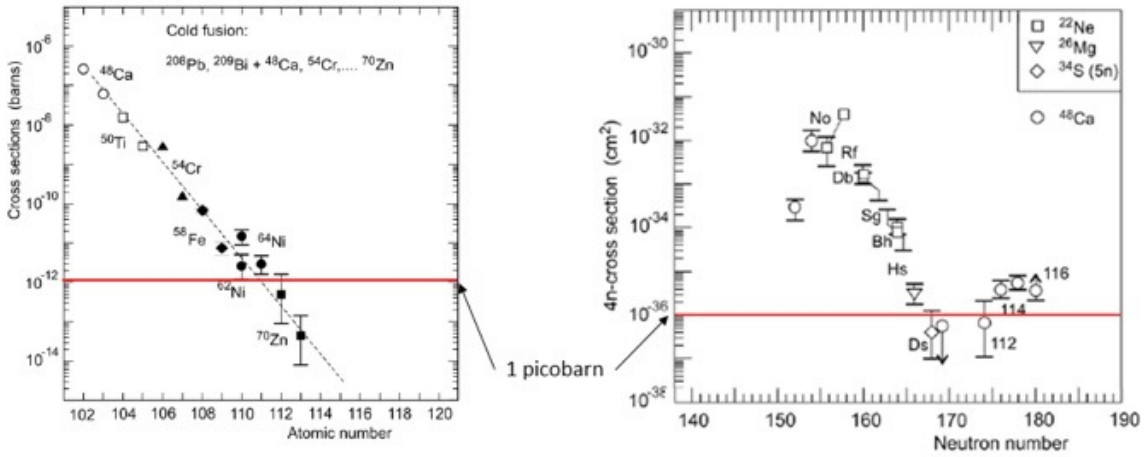

Fig. 8-4. Comparison of production rates of nuclei between cold-fusion reactions (left) and hot-fusion reactions (right). Red lines indicate one picobarn cross-sections.

During the decade of 2000-2010, six new chemical elements were produced by the Dubna/LLNL collaboration (and later collaborators from ORNL and Vanderbilt University), and one element, element 113 (nihonium), was also produced via heroic experiments at RIKEN laboratory in Japan using the cold-fusion reaction ^{70}Zn + ^{209}Bi. Hot-fusion reactions produced flerovium (114), livermorium (116), moscovium (115) and nihonium (113), oganesson (118) and finally tennessine (117). These elements were confirmed by other laboratories and accepted by the IUPAC, thus completing the seventh row or period of the periodic table.

It should be noted that the half-lives of some isotopes produced via these reactions are significantly longer than those of prior isotopes produced via cold-fusion reactions. Using Cn isotopes as an example, ^{285}Cn ($t_{1/2}$ = 29 s) lives nearly 5 orders of magnitude longer than ^{277}Cn ($t_{1/2}$ = 0.69 ms). This, combined with increased production rates, provides strong evidence that the newly produced isotopes exist on the shores of the Island of Stability, near the N = 184 and Z = 114 closed shells. Theorists now predict a region of enhanced stability centered around Z = 114, 120, 124 or 126, perhaps a broader region of isotopes with longer half-lives. The heaviest produced isotopes are still 7 neutrons short of N = 184.

There have been multiple attempts, by multiple laboratories, to produce elements 119 and 120 using different hot-fusion reactions. Because targets of Es and Fm cannot be

made due to small amounts of target material exisiting on the planet at any given time, beams of ^{48}Ca cannot be used, and any advantage obtained using a doubly magic nucleus as beam is lost. Nevertheless, attempts to produce the next elements have been made, and have even captured the imagination of the public at large.

The popular TV show, "The Big Bang Theory" even had several story lines where the genius character, Sheldon Cooper, postulated the production of element 120 using various reactions (see Fig. 8-5). All of the reactions on the whiteboard have been tried, with no success so far, except for the reaction containing Md as a target because a target of Md cannot be made due to scarcity of the material.

Weare in a time period where development of better equipment is occuring to continue the searches for new superheavy elements. The Super Heavy Element Factory (SHEF) in Dubna, Russia is currently nearing completion and will be dedicated to super heavy element research.

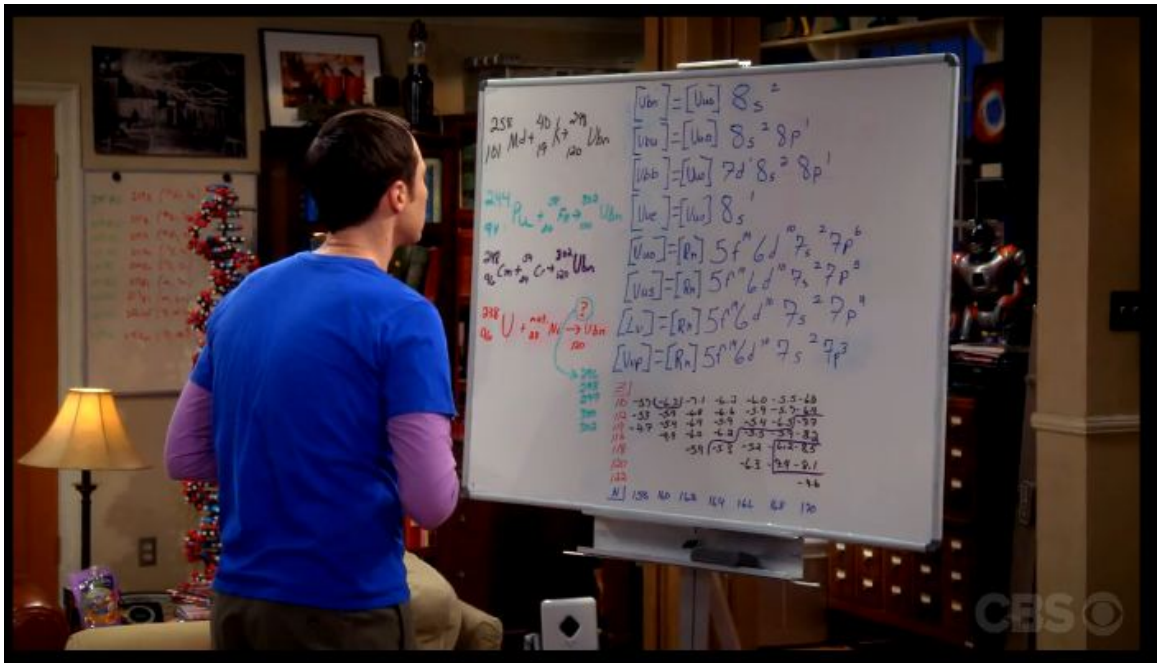

Figure 8-5. Season 7 (2013) – "The Romance Resonance" of the popular TV series "The Big Bang Theory" in which various nuclear reactions are suggested to produce element 120. From top to bottom, the reactions ^{258}Md + ^{40}K, ^{244}Pu + ^{58}Fe, ^{248}Cm + ^{54}Cr and ^{238}U + Ni are suggested. All of these reactions have been attempted, except for the ^{258}Md reaction because a target cannot be constructed of this scarce material.

Traditionally, the discoverers of a new element chose its name, and then the International Union of Pure and Applied Chemistry (IUPAC) officially approves it. Table 8-1 lists the currently approved IUPAC names.

Table 8-1. "Heavy" element names that are approved by IUPAC.

Element Number	IUPAC Proposal	Symbol
101	Mendelevium	Md
102	Nobelium	No
103	Lawrencium	Lr
104	Rutherfordium	Rf
105	Dubnium	Db
106	Seaborgium	Sg
107	Bohrium	Bh
108	Hassium	Hs
109	Meitnerium	Mt
110	Darmstadtium	Ds
111	Roentgenium	Rg
112	Copernicium	Cn
113	Nihonium	Nh
114	Flerovium	Fl
115	Moscovium	Mc
116	Livermorium	Lv
117	Tennessine	Ts
118	Oganesson	Og

New experimental techniques and apparatus have been developed for scientists to extend the periodic table to even more massive elements. A more efficient particle separator uses magnetic fields to separate atoms based on their mass and charge. This equipment can detect nuclides with low production rates and extremely short half-lives. The present limits for discovering new elements are based on the low production rates and short half-lives. The hope is that new development in detection equipment will increase the sensitivity for detecting fewer atoms (or even a single atom) with very short half-lives.

As can be seen from the prior discussion, a variety of techniques have been used to synthesize new chemical elements. Modern "nuclear alchemists" working on the synthesis of superheavy elements "transmute" one element into another by using particle accelerators to smash one element into another. The currently preferred method of producing new elements is fusion-evaporation reactions. This process uses the fusion of two nuclei to form a compound nucleus which then emits few or several neutrons and produces the brand new element.

It has been possible to study the chemical properties on the macroscopic scale for elements as massive as einsteinium (99) and on the tracer scale for elements as massive as seaborgium (106). The elements beyond the actinides in the Periodic Table are termed the "transactinides" and are shown in a Modern Periodic Table, Fig. 8-1, in their expected places. The yields of the most massive elements produced in bombardments of target nuclei with "heavy" ions become extremely small with increasing atomic number,

dropping to as little as one atom per week of bombardment for elements as massive as atomic number 112. The production of one isotope of element 113 with cold fusion reactions was at a production rate of about one atom per half-year. The half-lives decrease into the millisecond and the microsecond range so that identification of the new nuclei becomes increasingly difficult. Their half-lives would be impossibly short were it not for the presence of closed shells of nucleons to increase the nuclear stability.

Table 8-2. Summary of the reactions and methods used in the discovery of the actinide and transactinide elements. See Chapter 7 for an explanation of the reaction notation.

Element	Production Reaction(s)[1]	Method of Discovery	Year
neptunium (Np)	$^{238}U(n,\beta^-)^{239}Np$	Chemical Separation	1940
plutonium (Pu)	$^{238}U(^{2}H,2n)^{238}Np$ $^{238}Np(\beta^-)^{238}Pu$	Chemical Separation	1941
curium (Cm)	$^{239}Pu(^{4}He,n)^{242}Cm$	Chemical Separation	1944
americium (Am)	$^{239}Pu(n,\gamma)^{240}Pu$ $^{240}Pu(n,\gamma)^{241}Pu$ $^{241}Pu(\beta^-)^{241}Am$	Chemical Separation	1945
berkelium (Bk)	$^{241}Am(^{4}He,2n)^{243}Bk$	Chemical Separation	1949
californium (Cf)	$^{242}Cm(^{4}He,n)^{245}Cf$	Chemical Separation	1950
einsteinium (Es)	$^{238}U(15n,7\beta^-)^{253}Es$	Chemical Separation[2]	1952
fermium (Fm)	$^{238}U(17n,8\beta^-)^{255}Fm$	Chemical Separation[2]	1953
mendelevium (Md)	$^{253}Es(^{4}He,n)^{256}Md$	Recoil Method, Chemical Separation	1955
nobelium (No)	$^{246}Cm(^{12}C,4n)^{254}No$	Recoil Method, Chemical Separation	1958
lawrencium (Lr)	$^{249/250/251/252}Cf(^{10/11}B,xn)^{258}Lr$	Direct α Counting[3]	1961
rutherfordium (Rf)	$^{249}Cf(^{12}C,4n)^{257}Rf$ $^{249}Cf(^{13}C,3n)^{259}Rf$	Parent-Daughter α Correlation	1969
dubnium (Db)	$^{249}Cf(^{15}N,4n)^{260}Db$	Parent-Daughter α Correlation	1970
seaborgium (Sg)	$^{249}Cf(^{18}O,4n)^{263}Sg$	Parent-Daughter-Granddaughter α Correlation	1974
bohrium (Bh)	$^{209}Bi(^{54}Cr,n)^{262}Bh$	Velocity Separator[4]	1981
hassium (Hs)	$^{208}Pb(^{58}Fe,n)^{265}Hs$	Velocity Separator[4]	1984
meitnerium (Mt)	$^{209}Bi(^{58}Fe,n)^{266}Mt$	Velocity Separator[4]	1982
darmstadtium (Ds)	$^{209}Bi(^{59}Co,n)^{267}Ds$ $^{208}Pb(^{62,64}Ni,n)^{269,271}Ds$ $^{244}Pu(^{34}S,5n)^{273}Ds$	Mass Separator Velocity Separator[4,5] Recoil Separator	1991 1994 1995
Roentgenium (Rg)	$^{209}Bi(^{64}Ni,n)^{272}Rg$	Velocity Separator[4]	1994
Copernicium (Cn)	$^{208}Pb(^{70}Zn,n)^{277}Cn$	Velocity Separator	1996
Nihonium (Nh)	$^{243}Am(^{48}Ca,3n)^{288}Mc$ $^{237}Np(^{48}Ca,3n)^{282}Nh$ $^{209}Bi(^{70}Zn,n)^{278}Nh$	Gas-filled Separator[6]	2003
Flerovium (Fl)	$^{244}Pu(^{48}Ca,3n)^{289}Fl$	Gas-filled Separator[7]	1999
Moscovium (Mc)	$^{243}Am(^{48}Ca,3n)^{288}Mc$	Gas-filled Separator[7]	2003
Livermorium (Lv)	$^{248}Cm(^{48}Ca,3n)^{293}Lv$	Gas-filled Separator[7]	2000
Tennessine (Ts)	$^{249}Bk(^{48}Ca,3n)^{294}Ts$	Gas-filled Separator[7]	2010
Oganesson (Og)	$^{249}Cf(^{48}Ca,3n)^{294}Og$	Gas-filled Separator[7]	2002

[1]When more than one reaction is given, it means that a sequence of reactions was necessary to discover the element, or multiple reactions were used to produce different isotopes.
[2]The Es and Fm reactions were not done in a laboratory setting; multiple neutron captures, followed by successive beta decays were the result of a thermonuclear explosion.
[3]A mixture of four Cf isotopes was bombarded simultaneously with a beam containing ^{10}B and ^{11}B. The symbol "xn" means that different numbers of neutrons were emitted, depending on the actual combination of target and beam used to produce Lr.
[4]A velocity separator is used to separate reaction products based on the fact that reaction products of different masses will be emitted with different velocities.
[5]At GSI two different reactions were used to produce two different isotopes of element Ds.
[6]Nh was first produced with the ^{48}Ca + ^{243}Am reaction as a decay product of element 115, and then also with the ^{70}Zn reaction over the course of many years of beam time.
[7]Production of these elements was by a variety of cross-bombardment reactions and at various beam energies so excitation functions could be determined. Lighter elements are also decay products of heavier elements.

It has been possible to study the chemical properties of rutherfordium, hahnium and seaborgium using the advanced techniques of one-atom-at-a-time chemistry. These experiments show that these properties generally are consistent with those expected on the basis of extrapolation from those of their lower-mass *homologues* in the Periodic Table, hafnium, tantalum, and tungsten. Recent studies have shown that the properties of bohrium (107) and hassium (108) are the homologues of rhenium and osmium. Some initial chemical studies of Cn (Z = 112) and Fl (Z = 114) have been performed which indicate that Cn is likely a homologue of Hg. So far the studies have been inconclusive on Fl. However, the chemical properties cannot be determined reliably in detail from trends exhibited by the lighter homologues, because of the important role played by relativistic effects (the fact that some electrons are moving at velocities near the speed of light) in these more massive elements. Elements higher than hassium are placed in their expected place in the Periodic Table.

Just how many elements are there? Eventually, it will not be possible to add another proton to the nucleus even with the addition of many neutrons The repulsion of the protons will be so high, that no number of neutrons could keep the nucleus together. Because of the complex interplay between the nucleons, via the short-range attractive strong nuclear force and the longer range repulsive Coulomb force, forces that are poorly understood, it is extremely difficult to predict at what proton number the nucleus will rapidly fall apart. Additionally, as the nucleus becomes more massive, the orbiting electrons become more relatavistic, and at some point the innermost electrons will actually spend the majority of the time inside the nucleus. This will increase the probability of "forced" beta-decay, where a proton is converted into a neutron, and will also preclude any increase in the number of protons in the nucleus. This effect has been estimated with the best relatavistic calculations, by P. Pyykkö, to occur at Z = 172, and the resultant periodic table is shown in Fig. 8-6. It is exciting to consider that perhaps a third of the elements in the periodic table have yet to be discovered.

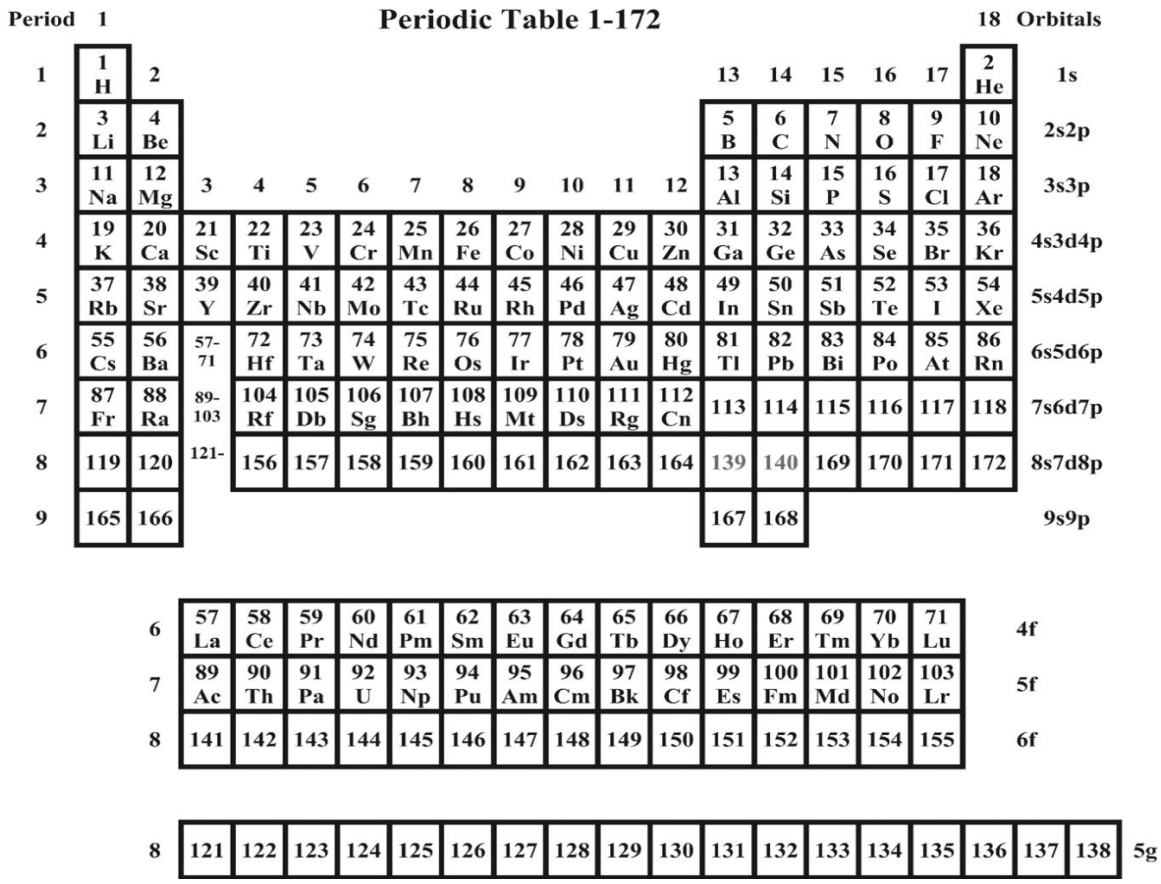

Figure 8-6. Periodic table extrapolated to element 172 using the best relatavistic calculations to predict chemical behavior beyond the known 118 elements.

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