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The Discovery of Element 117: Super-Heavy Elements and the "Island of Stability"*

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Element 117 (tennessine) joined the periodic table in November 2016. Two tennessine isotopes were synthesized by bombarding ^{249}Bk from Oak Ridge National Laboratory with ^{48}Ca ions at the Joint Institute of Nuclear Research, Russia, and eleven new heaviest isotopes of odd-Z elements were observed in subsequent decay chains. These isotopes exhibit increasing lifetimes as the closed nuclear shell at neutron number N=184 is approached, providing evidence for the "island of stability" for super-heavy elements. This paper summarizes recent super-heavy element research with a focus on element 117, the role of actinide targets, and opportunities to synthesize elements 119 and 120.

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Introduction

The term super-heavy element (SHE) usually refers to the transactinides, elements with atomic number Z of 104 or greater. Originally predicted by Glenn T. Seaborg in the 1960s, these elements are located in row seven of the periodic table, immediately after element 89 as the main body of the periodic table resumes following completion of the lanthanide and actinide series. The existence of SHEs depends on the details of proton and neutron quantum states, and is therefore critical to the fundamental understanding of nuclear structure [1]. Since 1964, 15 super-heavy elements through Z=118 have been discovered, completing row seven of the periodic table.

In the late 1990s, progress in super-heavy element discovery was limited by ever decreasing cross-sections for the synthesis of higher Z elements using conventional "cold fusion" reactions [2]. A new approach, "hot fusion," was successfully developed and implemented [2] at the Joint Institute for Nuclear Research (JINR) at Dubna, Russia, based on the irradiation of actinide targets with ^{48}Ca ions. ^{48}Ca is a stable isotope (natural abundance 0.2%) with 28 neutrons, 8 more than 97% abundant ^{40}Ca. Hot fusion takes advantage of increasing fission barriers and lower neutron separation energies for nuclei as the closed shell at N=184 is approached. Compound nuclei created by actinide + ^{48}Ca reactions have large neutron numbers and are influenced by this shell effect. These nuclei stabilize by shedding excess neutrons, increasing their survivability. Compared to cold fusion for Z=113, actinide + ^{48}Ca hot fusion increases SHE production cross sections by one or more orders of magnitude for Z=113-118 [2].

Since 2000, five new super-heavy elements (Z=114-118) have been discovered using actinide + ^{28}Ca reactions at JINR [1]. These discoveries have been confirmed at GSI in Germany (elements 114, 115, 116, and 117), Lawrence Berkeley National Laboratory (elements 114 and 115), and RIKEN (element 116). The numbers of nuclei produced directly for each element is shown in Table 1, as well as the total number of nuclei including those observed in subsequent decay chains.

These new elements expand the periodic table and provide evidence for the existence of the proposed "island of stability" for super-heavy elements, a region beyond the existing periodic table where closed shells of neutrons and protons are expected to confer exceptionally long lifetimes for SHE [2,3]. ^{208}Pb, the heaviest stable element, has closed shells of neutrons and protons, a property referred to as "doubly magic." Theoretical estimates place the next doubly magic nucleus in the vicinity of N=184 and Z between 114 and 126, the presumed center of the island of stability [3]. Lifetime enhancements could be substantial, perhaps hundreds or even millions of years [1]. Approaching the island may enable chemistry studies of previously unknown elements with extreme numbers of electrons. The island also presents an opportunity to advance nuclear physics by exploring the properties of nuclei with extreme numbers of neutrons and protons, nuclei that may not currently exist in nature [4].

Actinide materials for super-heavy element research

Nine of the 15 super-heavy elements observed to date have been discovered using actinide targets. These discoveries have involved actinides from Oak Ridge National

Laboratory (ORNL) and the Research Institute of Advanced Reactors (RIAR) in Dimitrovgrad, Russia. The High Flux Isotope Reactor (HFIR) and Radiochemical Engineering Development Center (REDC) at ORNL provide a unique resource for the production, chemical separation, and purification of actinide materials, and for the fabrication of actinide targets [20]. ORNL is the repository for actinide materials for the Isotope Development and Production for Research and Applications Program of the U.S. Department of Energy (DOE) Office of Nuclear Physics.

Heavy actinides are produced at ORNL by irradiation of mixed americium and curium targets in the intense thermal neutron flux (~3×10^{15} n/s-cm^2) of HFIR [20]. Approximately 40 g of Am/Cm is converted to oxide microspheres, blended with aluminum powder, and pressed into cermet pellets. The pellets are loaded into specially-designed aluminum tubes and inserted into the flux trap of HFIR, where the Am/Cm is transmuted to heavier isotopes in a series of neutron captures and beta decays. A typical campaign includes 3-5 24-day reactor cycles and can produce up to 200 mg of ^{252}Cf, 20 mg of ^{249}Bk, 1-2 micrograms of ^{254}Es, and about a picogram of ^{257}Fm.

After irradiation, the Am/Cm targets are allowed to decay for several months to eliminate short-lived fission and activation products. The targets are then transferred to REDC for chemical separation and purification of the transcurium products. This process, illustrated in Figure 1, includes multiple dissolution and ion exchange steps, performed remotely over several months in heavily shielded hot cells [20-25]. The aluminum matrix is dissolved in a heated $NaOH/NaNO_3$ solution, and the precipitated metal hydroxides are dissolved in nitric acid [21]. A solvent extraction process using di-2-ethylhexylphosphoric acid (HDEHP) extracts the lanthanides and actinides [22], and a LiCl anion exchange process separates the lanthanides from the actinides [23]. The separation of the transcurium actinides (Fm, Es, Bk, and Cf) from the remaining Am/Cm is enhanced using a second LiCl anion exchange. The heavy actinides are separated from each other using alpha-hydroxyisobutyric (AHIB) acid and cation exchange [24], and the Cf fraction is packaged for future use. Approximately 75% of the Am/Cm starting material remains at the end of the process and can be recycled for the next campaign.

For the synthesis of element 117 using ^{48}Ca ions, a ^{249}Bk target is required—the fusion of Ca (Z=20) with Bk (Z=97) produces a compound nucleus with Z=117. Berkelium is typically recovered at HFIR/REDC as a byproduct of californium production using additional separation and purification steps [25]. The Bk fraction from the above AHIB step is concentrated by passing it through a cation exchange column followed by solvent extraction in HDEHP to remove residual ^{252}Cf. At this point, the Bk can be safely transported to a glove box for further purification. A total of ~22 mg of high purity ^{249}Bk was produced and separated for the original element 117 experiment, with $<10^{-9}$ g of ^{252}Cf contamination and overall decontamination factors of 10^7 or greater for all radionuclides. ^{249}Bk beta decays to ^{249}Cf with a half-life of 327 days and must be produced fresh for each experiment.

The discovery of element 117

The discovery of element 117 was reported in 2010 [15] by an international team from Russia and the U.S. using the ^{249}Bk + ^{48}Ca reaction at the Dubna Gas-Filled Recoil Separator (DGFRS) at JINR. This experiment utilized ~15mg of ^{249}Bk from ORNL painted onto thin titanium foils at RIAR. The foils, containing ~0.31mg/cm^2 of ^{249}Bk, were mounted on a rotating wheel at the DGFRS and exposed to an intense beam of ^{48}Ca ions of ~1 particle microamp in the U-400 cyclotron. Beam energies of 252 and 247 MeV in the center of the Bk target produced compound nuclei of ^{297}117 with excitation energies of 39 and 35 MeV, respectively. These excitation energies were chosen to match the expected peaks in the cross section for production of ^{293}117 and ^{294}117 nuclei stabilized by the emission of 4 and 3 neutrons, respectively.

The resulting recoils of ^{293}117 and ^{294}117 tennessine (Ts) nuclei were separated from beam particles and other reaction products in a magnetic field and implanted in a position-sensitive detector where their energies were detected. Suppression factors were ~10^{15} for beam particles and 10^4 for target-like particles at the detector. The embedded element 117 nuclei, ~ 1/month, decayed by sequential alpha emissions in decay chains (Z=117 to 115 to 113, etc.), with the lifetimes of the resulting nuclei and the associated alpha energies providing a unique signature for the originating isotope. After observation of a compound nucleus implantation followed by an alpha emission at the same location with the expected energy, the accelerator was temporarily shut down to allow subsequent alpha emissions and the fission that terminates the chain to be detected under very low background conditions. The beam separation and detection system is illustrated in Figure

2. During 140 days of almost continuous irradiation (70 days at each at excitation energy), six element 117 decay chains were observed, five from the ^{293}117 and one from the ^{294}117 isotope. ^{294}117, together with the previously reported ^{294}118 [14], are the two heaviest nuclei observed to date.

The experiment produced thirteen new heaviest isotopes including two isotopes of element 117 and eleven in the related alpha decay chains. These decay chains are shown in Figure 3, with all nuclei closer to the presumed center of the island of stability than previously observed for odd-Z isotopes. For ^{293}117, the decay chain includes up to four sequential alpha emissions followed by a fission event. For ^{294}117, there are up to seven alpha decays in the chain. These results were confirmed in Dubna in 2012 [14] and independently at GSI in 2014 [13], again in ^{249}Bk + ^{48}Ca reactions using berkelium produced at ORNL. To date, 22 atoms of element 117 have been produced, 16 of ^{293}117 and 6 of ^{294}117. During the course of the second Dubna experiment [16], a decay chain from element 118 was also observed. The ^{249}Bk (327-day half-life) had partially decayed to ^{249}Cf, allowing the ^{249}Cf + ^{48}Ca reaction to occur producing element 118.

In December 2015 a joint committee of the International Union of Pure and Applied Chemistry and the International Union of Pure and Applied Physics officially recognized the discovery of element 117 as well as its decay product element 115, assigning priority for the discoveries to an international collaboration involving JINR, Lawrence Livermore National Laboratory, ORNL, and Vanderbilt University [26]. The provisional name tennessine (Ts) was recommended for element 117, recognizing the contribution of the

Tennessee region, including ORNL, Vanderbilt, and the University of Tennessee, to super-heavy element research, including the production and separation of unique actinide target materials at ORNL. Tennessine (Ts) was formally approved in November 2016 after the required five-month public comment period [27].

In addition to the element 117 (Ts) decay chains, Figure 3 shows the nuclear chart as it exists today for elements with Z=105 and above. Including nuclei descendent from Ts isotopes, these data include a particularly wide range of Z=113 isotopes spanning 8 neutron numbers, from the cold fusion results for ^{278}113 (N=165) [28] obtained in multi-year experiments at RIKEN to the hot fusion results from Dubna for $^{282\text{-}286}$113 (N=169 through 173) [1]. The lifetimes of these nuclei steadily increase with N, from ~1.4 ms for ^{278}113 to 9.5 seconds for ^{285}113, as shown in Table 2. This increase of a factor of roughly 6,000 over 8 neutron numbers provides evidence for the existence of the island of stability, and for the influence of the shell closure at N=184. A similar range of neutron numbers has been observed for Z=112 (Cn) isotopes, with half-lives increasing by tens of thousands as neutron number increases from N=165 to N=173 [1,29]. Half-lives increase with neutron number for all observed isotopes with $Z \geq 110$, with the rate of increase slowing for elements with $Z > 113$, consistent with theoretical predictions [30].

The alpha decay chains and lifetimes for isotopes observed in the element 117 experiments are compared to other odd-Z isotopes in Figure 4, including isotopes of new elements 115 and 113. These data show a continuing trend for increased stability with higher neutron numbers, with lifetimes increasing and alpha decay energies decreasing as the shell closure at N=184 is approached. This behavior is consistent with the predictions of nuclear models [30] and with the existence of the island of stability for super-heavy elements. Similar consistent trends are observed for even-Z nuclei in the synthesis of elements 118, 116, and 114 (and the resulting decay chains) in other actinide + ^{48}Ca reactions [1].

New opportunities in super-heavy element research

^{249}Bk and ^{249}Cf are the heaviest target materials used to date for super-heavy element synthesis. Using ^{48}Ca ion beams, these targets have produced elements 117 and 118. In order to reach higher Z using these targets, higher Z ion beams such as ^{50}Ti and ^{54}Cr will be needed, where production cross sections are expected to be substantially lower [1]. Higher intensity ion beams and improved detection systems will be required to compensate for these lower cross sections.

New facilities coming on line including the Super-Heavy Element Factory at Dubna, and upgrades at RIKEN and GSI, offer the potential to increase production rates for SHE by one or two orders of magnitude. This could enable experiments using ^{50}Ti beams with ^{249}Bk and ^{249}Cf targets to reach elements 119 and 120 (see Figure 3). An important precursor to these experiments is investigating the reaction mechanisms for ^{50}Ti on lighter targets, such as Cm and Pu, where cross sections are higher. These studies will provide information on ^{50}Ti + actinide reactions in order to predict beam energies and expected production rates for element 119 and 120 experiments using ^{50}Ti beams.

Heavier targets, such as ^{251}Cf, offer the potential to reach higher neutron numbers. ^{251}Cf can be obtained from old ^{252}Cf sources, where the highly radioactive ^{252}Cf decays with a half-life of 2.6 years. After several decades a mixture of ~50% ^{249}Cf, 15% ^{250}Cf, and 35% ^{251}Cf remains. Old Cf sources at REDC have been recovered and chemically separated from fission and other decay products (such as ^{248}Cm) to create a modestly radioactive mixed Cf material suitable for super-heavy element experiments [31]. This mixed Cf has been fabricated [31] into targets at ORNL, and initial experiments have been performed at Dubna using ^{48}Ca beams to search for the $^{295}118$ and $^{296}118$ isotopes of element 118. $^{296}118$ would be within 6 neutrons of N=184, one neutron closer than previously achieved and the heaviest nucleus to date (two neutrons closer if the 2n reaction channel can be accessed). Ideally, a highly enriched ^{251}Cf target would be desirable, increasing production rates for $^{48}Ca + {}^{251}Cf$ reactions by about a factor of three compared to the mixed target. This would require the development of an electromagnetic separator with appropriate shielding and contamination control for handling actinides.

If available in sufficient quantities, ^{254}Es would be an interesting target for even heavier isotopes and elements. ^{254}Es would provide a path to element 119 using ^{48}Ca and possibly to Z=121 using ^{50}Ti, both within 4 neutron numbers of N=184. Currently, ^{254}Es is available only in microgram quantities, 3 orders of magnitude lower than required for current SHE targets. In the 1980s, a dedicated HFIR campaign, the Large Einsteinium Activation Program [32], was proposed to increase ^{254}Es production by about a factor of ten using a tailored sequence of staged, multiphase irradiations in HFIR. The resulting 40 micrograms of ^{254}Es is more than two orders of magnitude less than the 10-15mg used in current SHE experiments, but it would be sufficient to cover a 0.1cm^2 area with the requisite thickness of target material of ~0.3mg/cm^2. This would enable super-heavy element experiments with ^{254}Es targets assuming a target technology were available that could survive continuous exposure to the entire ion beam in this small area.

Summary and conclusions

The use of actinide + ^{48}Ca fusion reactions has led to the discovery of five new super-heavy elements, extending the periodic table to Z=118 and producing more than 50 new heaviest isotopes of elements 105-118. These discoveries depend critically on the availability of specialized facilities for the production and separation of actinide materials, including very high-flux reactors and heavily-shielded radiochemical hot cells available in only a few locations worldwide. Separation science and technology has been essential to this progress. These discoveries, including tennessine, have extended our understanding of the properties of super-heavy nuclei and provided evidence for the existence of the "island of stability" for super-heavy elements. New accelerator facilities and target/beam combinations offer the potential to extend the nuclear chart to elements 119 and 120 and to an additional neutron number. These nuclei provide sensitive tests of nuclear models, and lifetime increases associated with a closer approach to the island offer the potential for chemistry studies on previously unknown elements.

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Table 1. New elements produced since 2000 using actinide + ^{48}Ca reactions.

Element (Z)	Year	Target	Projectile	Nuclei produced (directly/total)	Reference
Flerovium (114)	2004	^{239,240}Pu ^{242,244}Pu	^{48}Ca	82/124	[5-9]
Moscovium(115)	2004	^{243}Am	^{48}Ca	113/155	[5,10-12]
Livermorium (116)	2004	^{245,248}Cm	^{48}Ca	37/42	[5,6,13,14]
Tennessine (117)	2010	^{249}Bk	^{48}Ca	22	[5,15-17]
Oganesson (118)	2006	^{249}Cf	^{48}Ca	5	[5,16,18,19]

Table 2. Half-lives of element 113 isotopes created directly and descendant from isotopes of element 115 and 117 [1,28]. Observed half-lives increase by ~6,000 as neutron numbers increase from N=165 to 173, consistent with the influence of the closed nuclear shell at N=184.

Originating parent nucleus			^{287}115 (Mc)	^{288}115 (Mc)	^{293}117 (Ts)	^{294}117 (Ts)
Initiating reaction	^{209}Bi+^{70}Zn	^{239}Np+^{48}Ca	^{243}Am+^{48}Ca	^{243}Am+^{48}Ca	^{249}Bk+^{48}Ca	^{249}Bk+^{48}Ca
Direct or descendant Z=113 isotope	^{278}113	^{282}113	^{283}113	^{284}113	^{285}113	^{286}113
Neutron number, N	165	169	170	171	172	173
Half-life (ms)	1.4 (+1.9–0.5)	73	75	910	4200	9500

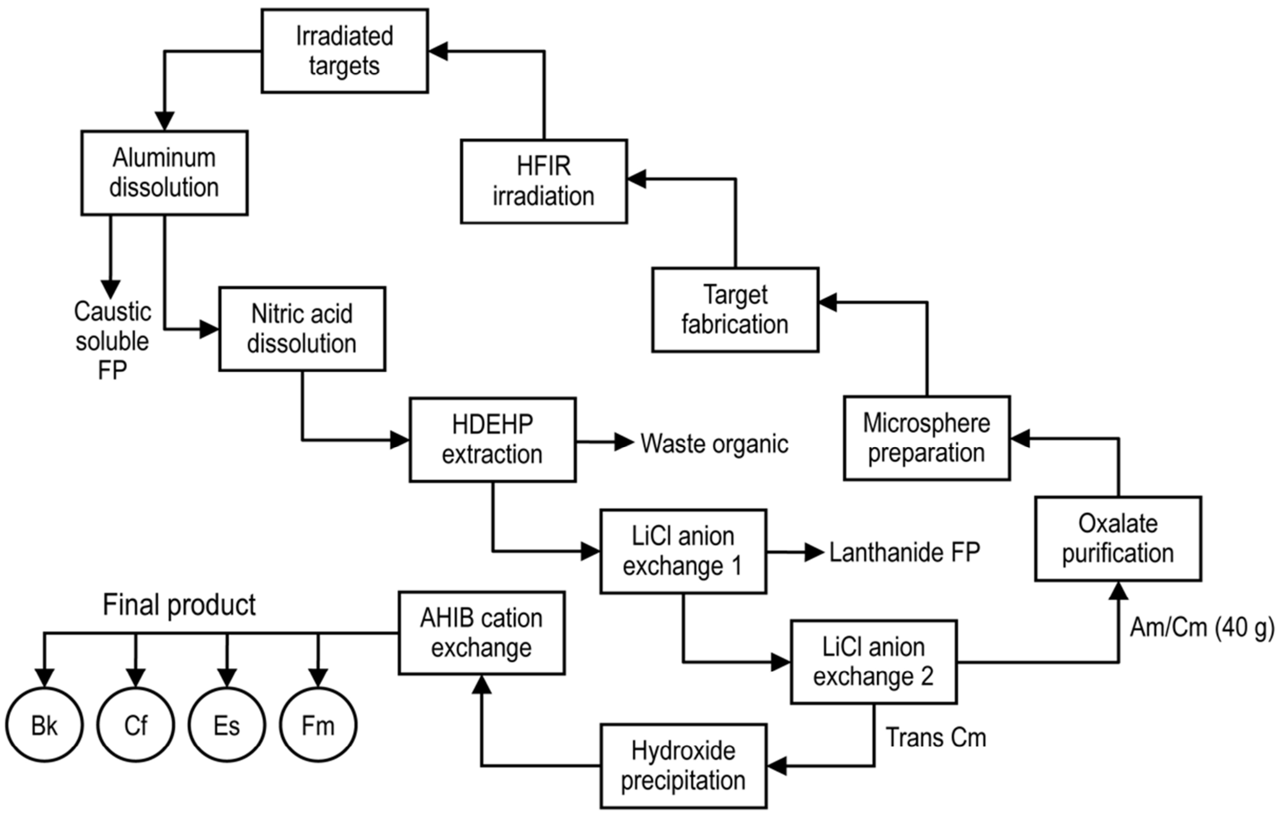

Fig. 1. Flow chart for production and chemical separation of transcurium actinides at HFIR/REDC. Typical yields are up to 200 mg of ^{252}Cf, 20 mg of ^{249}Bk, 1-2 micrograms of ^{254}Es, and about a picogram of ^{257}Fm [20].

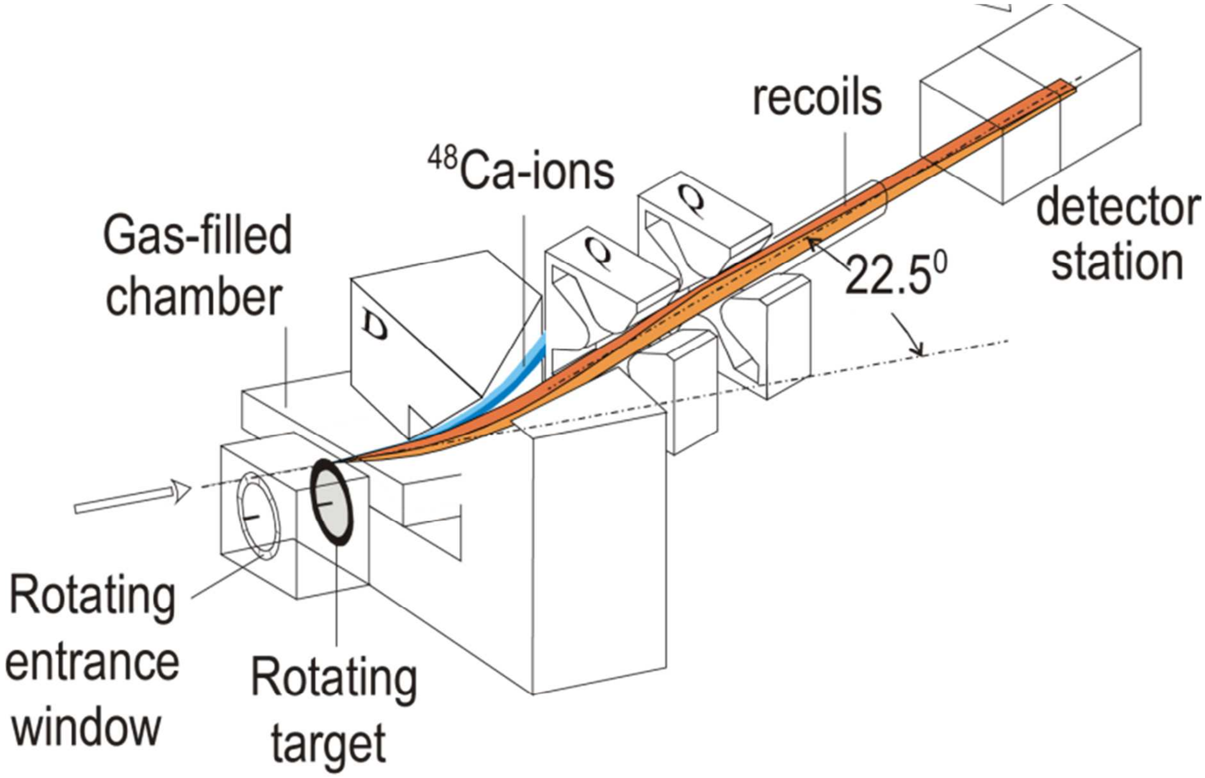

Fig. 2. Schematic of the beam separation and detection system at the Dubna Gas-Filled Recoil Separator [2].

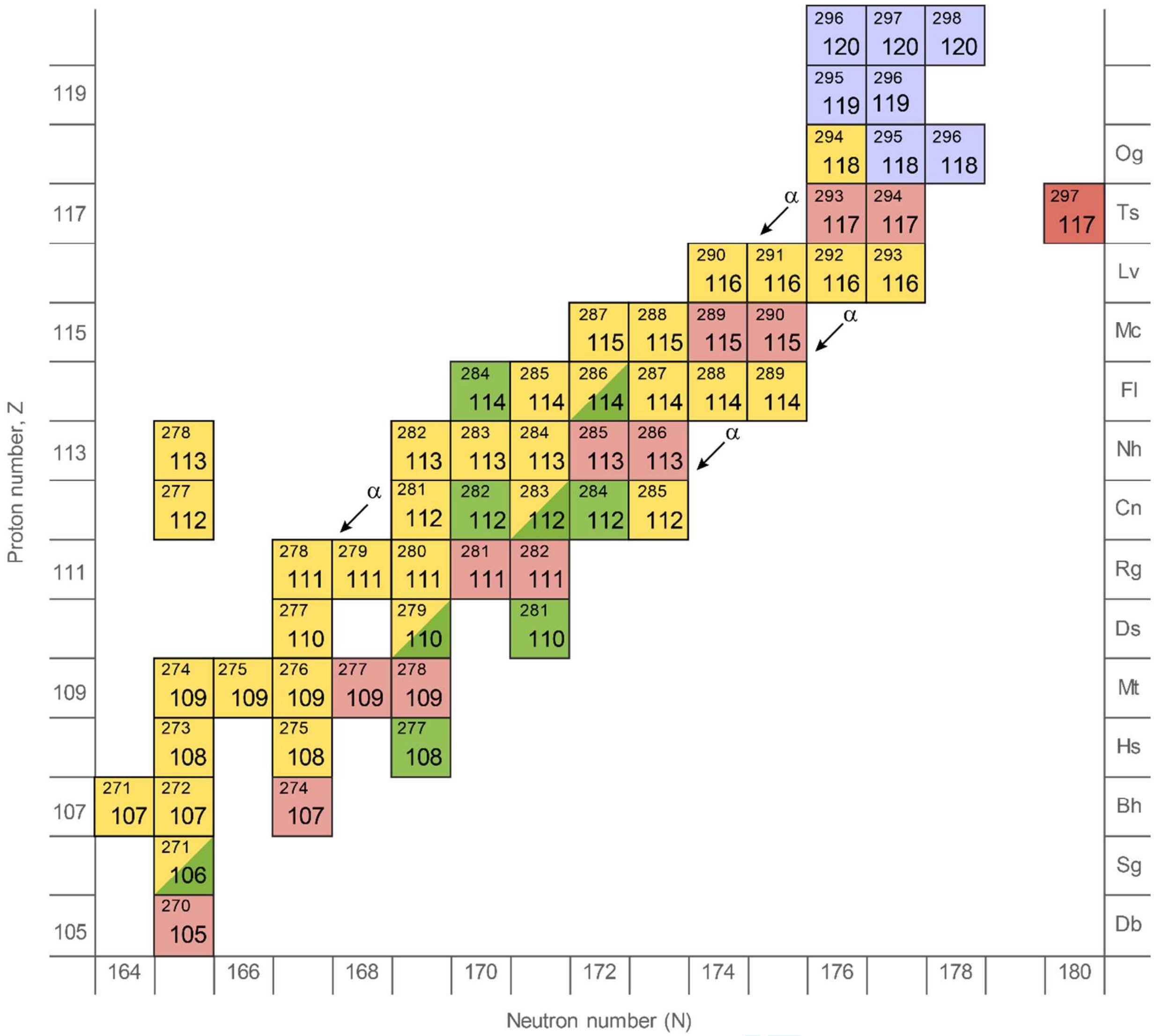

Fig. 3. Nuclear chart for super-heavy nuclei created in actinide + ^{48}Ca reactions. The "hot fusion island" includes five new heaviest elements and more than 50 new heaviest isotopes, all closer to the predicted "island of stability". Shown in red is the ^{297}117 compound nucleus created in the complete fusion reaction ^{249}Bk + ^{48}Ca. This excited nucleus stabilized by emission of 3 or 4 neutrons to ^{294}117 and ^{293}117, respectively, followed by sequential alpha decays leading to descendent isotopes of elements 115, 113, etc., as shown in lighter red. ^{278}113 and ^{277}112, resulting from cold fusion reactions [28,29], are also shown. Nuclei shown in blue are potentially reachable using ^{48}Ca and ^{50}Ti beams and existing ^{249}Bk and $^{249\text{-}251}$Cf target technology. Green denotes fission.

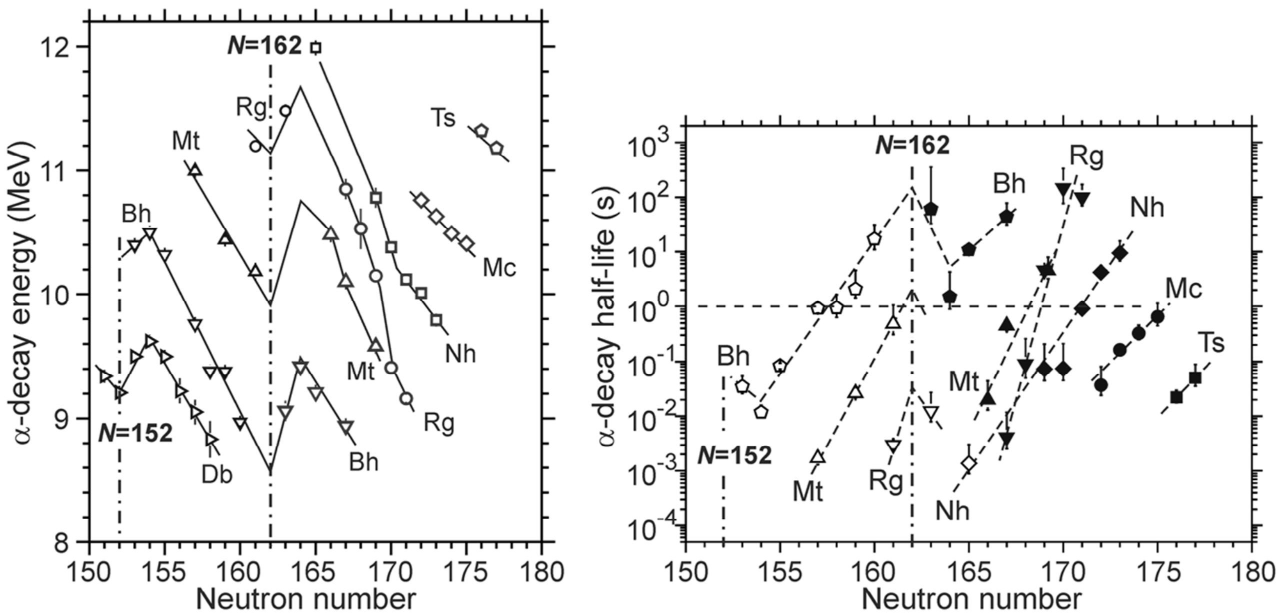

Fig. 4. Alpha-decay energies (left) and half-lives (right) for isotopes of odd-Z elements 105-117. The data show a consistent pattern for nuclear properties of odd-Z super-heavy elements. A similar pattern is observed for even-Z isotopes [1]. Note the decreasing decay energies and increasing half-lives for new elements 117 (Ts), 115 (Mc), and 113 (Nh) as neutron number increases toward the closed shell at N=184.