Nuclei in the “Island of Stability” of Superheavy Elements

Yuri Oganessian
Flerov Laboratory of Nuclear Reactions (Joint Institute for Nuclear Research)
141980 Dubna, Moscow region, Russia
oganessian@jinr.ru

Abstract. The observation of atomic numbers $Z$ that are 40% larger than that of Bi, the heaviest stable element, is an impressive extension of nuclear survival. Although the super heavy nuclei (SHN) are at the limits of Coulomb stability, shell stabilization lowers the ground-state energy, creates a fission barrier, and thereby enables the SHE to exist. The fundamentals of the modern theory concerning the mass limits of nuclear matter have been experimentally verified.

We are going to consider how big a nucleus may be, what is the limit of atomic nuclei mass and how it is determined.

The nuclei shown in Fig.1 having different numbers of protons and neutrons, have different binding energies and, therefore, different decay probabilities (half-lives).

In the region of transuranium elements, a sharp decrease in the stability of nuclei with increasing atomic number is observed. Indeed adding 10 protons and 4 neutrons to $^{238}\text{U}$ lead to a decrease in the period of $\alpha$-decay of $^{252}\text{No}$ by a factor of $10^{16}$ with respect to uranium. An even stronger decrease is expected for another type of decay - the spontaneous fission. Here the drop is 23 orders of magnitude! A simple extrapolation into the region of heavier nuclei shows that the limits of existence of elements will be determined by spontaneous fission. When the half-life is reduced to $10^{-14}$ s, the nuclei will decay long before they obtain its atomic structure. According to a simple extrapolation, this situation arises even for nuclei with atomic numbers 106-108.

Such a limit of atomic nuclei was predicted by N. Bohr and J. A. Wheeler [1], long before the first artificial elements were obtained. The fission of heavy nuclei was simulated by the process of deformation of a drop of charged liquid (the nuclear liquid drop model). According to Bohr and Wheeler, in the case of uranium a potential barrier about 6 MeV high prevents the fission of the nucleus (Fig.2 upper panel). If one adds an energy exceeding 6 MeV to the uranium nucleus, it will split into two fragments within about $10^{-19}$ sec. However, fission can occur spontaneously, by
tunneling through the fission barrier. The half-life of uranium relative to fission is $10^{16}$ years. Increasing the number of protons in the nucleus (advancing to the transuranium elements) reduces the fission barrier and greatly increases the probability of spontaneous fission. When the fission barrier drops, the nucleus will split into two fragments for about $10^{19}$ s. According to the calculations of the liquid drop model, this situation occurs immediately after $Z > 100$. It should be noted that when, using nuclear reactors, artificial elements up to Fermium ($Z = 100$) were synthesized, it seemed that the theoretical predictions have been fully confirmed. However, in 1962, in our Laboratory in Dubna, an unexpected effect was observed, which caused great doubts in the analogy of nuclear fission and a liquid drop. The isotope of $^{242}$Am, obtained in a nuclear reaction, experienced spontaneous fission with two, very different half-lives: $10^{14}$ a, and 0.014 s [2]. Subsequently, a similar phenomenon was observed in more than 31 nuclei with $Z = 92$-97 [3]. Definitely, fission in these nuclei occurs from two states - the ground and isomeric states (Fig.2 lower panel). But the shape isomerism of nuclei is completely incompatible with describing the fission process as a split of a liquid drop. In subsequent experiments to measure the heights of fission barriers of nuclei with $Z = 80$-100, significant differences from the predictions of the liquid drop model of fission were also found.

It is noteworthy that by this time it was already known that nuclei in their ground states differ in form and energy and at certain «magic» numbers of protons and/or neutrons the nuclear binding energy considerably increases. However, it was always assumed that reaching strong deformation on the way to splitting into two fragments, the structure of the nucleus disappears and the fission process can still be considered in the liquid drop approximation. In fact, as was shown in [4], nuclear structure does not disappear with increasing deformation, but is modified and continues to play a significant role in the process of nuclear fission. Calculation of the potential energy of the nucleus at large deformations, taking into account structural effects, explained the experimentally observed high fission barrier of the «doubly magic» nucleus $^{208}$Pb ($Z = 82$, $N = 126$), as well as the shape isomerism of uranium and transuranic nuclei, as a consequence of the double-humped fission barrier of actinide nuclei.

It was natural to extend this approach to the region of nuclei with $Z \geq 104$, where, according to the droplet model, nuclei may not exist. A formal calculation led to unexpected results. It turned out that in the deformed nucleus with mass 270 ($Z = 108$ and $N = 162$) a fission barrier occurs, which leads to an increase in its half-life to seconds (instead of $10^{19}$ sec). But even higher stability appears in heavier (superheavy) nuclei with proton number $Z = 114$ and a large number of neutrons $N = 184$. Here the strong effect of new nuclear shells works, similarly to the shells $Z = 82$ and $N = 126$ in the «doubly magic» spherical nucleus $^{208}$Pb. Some new calculations indicate that on the map of nuclei-heavyweights form a large enough area, called the «Island of Stability» of superheavy elements (see Fig.1). In the region where the liquid-drop model predicted that the nuclei should decay within $10^{19}$ sec, now the half-lives of «long-lived» nuclei at the peak of the island of stability are expected to reach thousands and even millions of years! (see, for example, review [5]).
Hypothetical superheavy elements radically change our ideas about the limits of the material world, and they indicate very significant expansion of the boundaries of existence of atomic nuclei and chemical elements. Here we have a problem how to check this extravagant theoretical hypothesis experimentally.

The known method of obtaining transuranic elements in successive neutron capture reactions with powerful reactors and even in nuclear explosions, confined to the observation of nuclei only up to $^{257}$Fm. In reactions induced by light ions ($Z_i = 6-10$) it was advance to elements with $Z = 106$ only. Entry into the area of $Z > 106$ occurred after the discovery the so-called «cold fusion» reactions when the target nuclei - $^{208}$Pb or $^{209}$Bi and massive projectiles with $Z_a = 22-30$ [6] were used. In the reactions of «cold fusion» the formed compound nuclei are slightly heated, they are cooled by the emission of one neutron, and therefore have a relatively high survival rate. This was their main advantage, which allowed in the years 1978-1998 to synthesize 6 new elements up to 112 [7]. But unfortunately, as will follow later, this method cannot be used for the synthesis of nuclei located on the «island of stability» of superheavy elements. The problem is that with increasing the mass and charge of the bombarding ion the Coulomb repulsion grows, and thus greatly decreases the probability of formation of compound nuclei with increased charge numbers. There is another fundamental limitation. In cold fusion reactions, due to lack of neutrons, the produced heavy nuclei are outside the predicted «island of stability». Even for the most heavy isotopes of elements 110, 111, and 112 obtained in this way, the half-lives are estimated to be only tens or hundreds of microseconds. It seemed that using more asymmetric masses of the interacting nuclei (the target nucleus is heavier than the mass of projectile) might reduce the strength of the Coulomb repulsion and thus raise the estimated probability of fusion by 4-5 orders of magnitude. But the compound nuclei formed in these reactions are very hot; the losses during their cooling are even greater. Unsuccessful attempts to synthesize superheavy elements in reactions of «hot fusion», undertaken in various laboratories in the years 1977-1985, led to the pessimistic view that the elements heavier than 112 might be quite stable, but it would be practically impossible to produce them.

In this conclusion, in our opinion, there was, however, an inherent contradiction. If the theoretical predictions were correct, the nuclei near the closed shells should have high fission barriers. In fact, it is this circumstance that causes the enormous stability of superheavy nuclei against spontaneous fission. But the high fission barrier must also prevent the fission of the heated compound nucleus and enhance its survival in the process of neutron emission during its cooling. In other words, the nuclei lying in the «island of stability» should fission less strongly and

Figure 3. A schematic view of gas-filled recoil-separator, which was used in the experiments on the synthesis of superheavy elements.

Figure 4. Decay chains of isotopes with $Z=114$ and 116, synthesized in the fusion reactions with calcium-48 projectiles.
should survive easier. Moreover, as we approach the boundaries of the „Island of Stability“, the rise of the formation cross sections of superheavy nuclei would be a direct indication of the inclusion of the effect of the new nuclear shells with \( Z = 114 \) (probably 120-122) and \( N = 184 \). Based on these assumptions, we chose a different reaction of synthesis of superheavy elements. Neutron-rich nuclei of SHE can in principle be synthesized if the heaviest actinides: \(^{244}\text{Pu}, \, ^{243}\text{Am}, \, ^{248}\text{Cm}, \, ^{249}\text{Bk} \) and \(^{251}\text{Cf} \), obtained in powerful nuclear reactors, were used as targets. As the projectile, we have chosen the rare isotope of calcium with mass number 48. As a result of two years work using an ECR-source and the U-400 accelerator, a stable beam of \(^{48}\text{Ca} \) ions was obtained with high intensity: \( 5.10^{12} -1.10^{13} \) pps. This allowed conducting long-term experiments (6000 hours/year) over the past 10 years. The registration of rare events of formation and decay of superheavy atoms was performed by the gas-filled recoil separator (Fig.3). In this setup, the nuclei emitted from the target, enter a hydrogen atmosphere (pressure about 1 Torre) and are separated by mass and ionic charge in a magnetic field and focus on the focal plane detector, located at a distance of 4 m from the target. The reaction products pass the distance between the target and the detector for 1 microsecond. The separator has a transmission for evaporation residues about 35-40%; the by-products of the reaction are suppressed by factor of \( 10^4 \). The focal plane detector registers the energy, position and time when the recoil nucleus stops in it. Then similar information follows concerning the charged particles (alpha particles or fission fragments) emitted in the decay of the implanted nucleus.

We recall that the theoretical predictions of the nuclei within the «Island of Stability» are stable against spontaneous fission. In the process of successive alpha-decay, the daughter nuclei will move away from the closed shells and will approach the boundaries of the island. The likelihood of spontaneous fission increases rapidly and eventually the chain will be interrupted by spontaneous fission. In other words, we expect that the decay of a superheavy nucleus will have the form of a chain (radioactive family), consisting of successive alpha transitions, relatively long in time, terminated by spontaneous fission. Our focal plane detectors have efficiency of 87% for the registration of alpha particles, almost 100% - for a single spontaneous fission fragment, 42% - for both fragments. The location of the stopped recoil nucleus at the working surface of the detector is determined with an accuracy of better than 0.2%.

![Figure 5](image_url)

**Figure 5.** Nuclei with \( Z=112-118 \) and products of their radioactive decay - the isotopes of elements 111, obtained in the fusion reactions of the Actinides and \(^{48}\text{Ca} \). The experimental values of half-lives are shown in the squares.
The location of the stopped recoil nucleus at the working surface of the detector is determined with an accuracy of better than 0.2%.

The coordinates of the follow-up signals can be used to establish the genetic link between the recoil nucleus and its decay. The probability of random coincidences of signals, simulating a correlated decay, can be significantly reduced by switching off the accelerator beam between the registration of the recoil nucleus and its subsequent first alpha-particle decay. For example, the probability of random coincidences in the registration of one event of formation and decay of the nucleus of the isotope of element 117 by the chain $^{209}117-\alpha\rightarrow^{209}114-\alpha\rightarrow^{205}111-\alpha\rightarrow^{201}Rg(SF)$, is $3.10^{-11}$ [9].

Just with this setup were synthesized isotopes of superheavy elements with atomic numbers 114 and 116 for the first time [10]. They showed a huge increase in nuclear stability by increasing the number of neutrons in the heavy nucleus (Fig.4). Adding to the previously known nuclei $^{275}110$ and $^{277}112$ 8 neutrons led to an increase in their half-lives by almost 100 000 times!

Our data on the decay characteristics of the isotopes of elements 112, 114 and 116 have been reproduced in 2007-2010 in a number of independent experiments [11-15]. Later, the heaviest element with atomic number 118 [16] was synthesized, followed by the synthesis of elements with odd atomic numbers: 113, 115 and 117 [17,9] in the reactions $^{239}Np$, $^{241}Am$ and $^{249}Bk + ^{48}Ca$. The data on the decay characteristics of the isotopes of 6 new superheavy elements, obtained up till now, together with those of the products of their radioactive decay (altogether 47 new nuclei) are shown in Fig.5. They can now be compared with the predictions of the microscopic theory [18].

Such a comparison is shown in Fig.6 for even-Z nuclei, for which the calculation can be done with the greatest accuracy. On the basis of the entire set of experimental data on the properties of superheavy nuclei produced in reactions induced by $^{48}Ca$ some general conclusions can be made.

1. The largest number of nuclei undergo alpha decay. The experimental energy of the alpha decay of superheavy nuclei differs from the calculated values by no more than 5-7%. This is a good agreement with the calculations of the nuclear macro-microscopic model. The periods of $\alpha$-decay of the new nuclides were, on the average, up to 10 times higher than predicted by the theory.

2. Spontaneous fission was observed only in 14 isotopes (see Fig.5). They belong to two groups: the isotopes of elements 110-114 with N = 169-172 and the isotopes of lighter elements with Z = 104-106 and N = 160-165. Spontaneously-fissioning nuclei of the first group are located between the closed neutron shells N = 162 and N = 184; they are away from them by 7-12 neutrons. The smallest value of $T_{SF} \approx 10^{-3}$ sec is observed for the even-even nucleus of element 112 with N = 172. As we move away from this area towards an excess or deficit of neutrons, the partial spontaneous fission half-life increases (top graph of Fig.6). For nuclei with Z $\geq$ 114 and N $\geq$ 174 the spontaneous fission was not observed. In the second group, the nuclei are located around the deformed shell N = 162; they

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure6.png}
\caption{The experimental values of the spontaneous fission half-lives and alpha-decay energies are compared with theoretical expectations. The bottom graph shows the half-lives of superheavy nuclei vs N.}
\end{figure}
have a relatively high stability. The even-odd nucleus of element 104 with N = 163 has a $T_{SF} \sim 5.10^3$ s. The long-lived odd-Z nuclei, the isotopes of element 105 ($T_{1/2} \approx 10^5$ sec), most probably undergo electron capture. Then the experimentally observed spontaneous fission refers to the decay of even-even isotopes of element 104 with N = 164 and 166. For these nuclei, the so-called fast mode of symmetric fission is expected due to the effect of the nuclear shells Z = 50 and N = 82 in the fission fragments.

**Conclusion**

Increasing the atomic number of the last stable element $^{209}$Bi ($T_{1/2} = 1.5.10^{19}$ years) by 40%, we are witnessing an impressive picture of the vitality of atomic nuclei. In the region of extreme Coulomb forces, due to the effect of new shells, the nuclear binding energy in the ground state increases, a fission barrier arises, and strongly increased stability of the heaviest nuclei leads to the existence of a large region of superheavy elements. The fundamental predictions of the modern theories concerning the mass limits of the atomic nuclei have received their first experimental confirmation.

The experiments were carried out at the U-400 heavy ion cyclotron of the Flerov Laboratory of Nuclear Reactions (FLNR, JINR) in collaboration with LLNL (Livermore, USA), ORNL (Oak Ridge, USA), Research Institute of Atomic Reactors (Dimitrovgrad, Russia) and Vanderbilt University (Nashville, USA).

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