Heaviest Nuclei from $^{48}$Ca-induced Reactions

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Abstract. The observation of atomic numbers $Z$ by 40% larger than that of Bi, the heaviest stable element, is an impressive extension in 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 SHN to exist. The fundamentals of the modern theory concerning the mass limits of nuclear matter have thus obtained experimental verification.

1. Introduction
A fundamental outcome of modern nuclear microscopic theory is the prediction of the “islands of stability” in the region of hypothetical superheavy elements. One important consequence of these calculations [1-8] was the disclosure of a significant gap in the spectrum of low lying levels in the region of the deformed nuclei around $N = 162$ (deformed shell) and of the hypothetical superheavy nuclei, viz. of a new (following $N = 126$) closed spherical neutron shell $N = 184$. And finally, at further and quite significant increase of the deformation arising in fission, the shell effects continued to play an important role in defining the potential energy and the nuclear inertial masses. The theoretical predictions for the new shells push far away the limits of nuclear masses and extend the region of existing heavy nuclei (and chemical elements) at least as far as $Z \sim 120$ and even more.

2. Reactions of Synthesis
For the synthesis of superheavy nuclei with $Z = 112-116$ and 118 with large neutron excess we chose the fusion reactions: $^{238}$U, $^{237}$Np, $^{242,244}$Pu, $^{243}$Am, $^{245,248}$Cm, $^{249}$Cf + $^{48}$Ca, which are characterized by evaporation residues with a maximal number of neutrons [9].

Until now for the synthesis of all heavy nuclei with $Z = 107-112$, fusion reactions of the magic nuclei $^{208}$Pb and $^{209}$Bi with a massive projectile ($A_P \geq 50$) were used [10]. In this kind of reactions the compound nucleus has an excitation energy about $E_x \approx 12-15$ MeV (cold fusion). The transition to the ground state takes place by the emission of only one neutron and $\gamma$-rays [11-15]. As a result, the survivability of the compound nucleus significantly increases, this being the main advantage of the cold fusion reactions. However, as can be seen from figure 1a, the production cross section of the evaporation products strongly decreases with the growth of $Z_{CN}$. This indicates the breaking of fusion, leading to the formation of a compound nucleus.

In reactions such as Actinides + $^{48}$Ca at excitation energies $E_x \approx 35-40$ MeV (hot fusion), the survivability of the compound nuclei is significantly lower. The two factors, the formation and survival of compound nuclei, lead to decreasing the cross section for the evaporation residues, formed
in the reactions of cold and hot fusion (figure 1). However, the restrictions themselves arise because of different reasons.

At fixed of mass/charge values of the target ($^{208}\text{Pb}$ or $^{209}\text{Bi}$) the increase of the atomic number of the projectile causes an increase in the Coulomb repulsion along the path of the collective motion of the system from the point of contact to the final configuration of the compound nucleus. For this reason the cross section $\sigma_{1n}(Z_{\text{CN}})$ will strongly decrease with increasing the atomic number of the compound nucleus (see figure 1a).

Figure 1.

a) Production cross section of the 1n-evaporation channel in cold fusion of the target nuclei $^{208}\text{Pb}$, $^{209}\text{Bi}$ and $^{50}\text{Ti}$, $^{54}\text{Cr}$, . . . $^{70}\text{Zn}$ projectiles (indicated in the figure).

b) Upper panel: production cross section of the 4n-evaporation channel in reactions of hot fusion of the actinide target nuclei and $^{22}\text{Ne}$, $^{26}\text{Mg}$, $^{34}\text{S}$-projectiles as a function of the neutron number in the compound nucleus. Lower panel: calculated values of the difference ($B_n-B_f$), which determines the survivability of the hot nucleus in the process of its de-excitation by emission of neutrons.

For more asymmetric in mass/charge hot fusion reactions, type of Act.$^{48}\text{Ca}$, the forces hindering fusion are weaker. The main losses are due to fission of the heavy excited nucleus in the process of its de-excitation by means of neutron evaporation. Thus, the survival probability of the nucleus is determined to a large extent by its fission, which depends on the height of the fission barrier. It should be noted that in nuclei with $Z>100$ the height of the fission barrier practically totally is defined by the contribution of the shell corrections to the deformation energy of the nucleus. How fissility of the compound nuclei changes as one goes away from the closed deformed shells $N=152$ and $N=162$ is well seen on the lower part of figure 1b. This is the reason for the strong decrease of the cross sections of hot fusion reactions, which are shown in the upper part of the figure. However, as one approaches the closed neutron shell $N=184$, the height of the fission barrier of the very heavy (superheavy) nuclei
increases almost by a factor of 2 (calculation) and, as a result of this, the cross sections of the evaporation residues from the Act.+ \(^{48}\)Ca reactions also increase (experiment). Such a correlation between the calculated \(B_f(Z,N)\) values and the experimental cross sections \(\sigma_{\alpha}(Z,N)\) gives direct evidence of the existence and the strong effect of a neutron shell located at \(N \geq 180\).

3. Setting the Experiments

The Gas-Filled Recoil Separator (DGFRS) used in the experiments with \(^{48}\)Ca-projectiles is schematically presented in figure 2. The typical beam intensity of \(^{48}\)Ca ions at the target was 1.0-1.2 \(\mu\)A. In the experiments, targets of actinide oxides of the highly enriched isotopes of U, Np, Pu, Am, Cm and Cf were used.

![Figure 2. Layout of the Gas-filled Recoil Separator.](image)

Evaporation residues passing through the separator were implanted in a \(4 \times 12\)-cm\(^2\) semiconductor detector with 12 vertical position-sensitive strips. The detection efficiency of the focal-plane detector array for \(\alpha\)-particles is 87% of 4\(\pi\); for detection of one fission fragment - close to 100%, for two fission fragments – about 40%. The setup allows investigation of nuclei in a range of half-lives from \(10^5\) s to more than \(10^9\) s.

From the characteristics of the DGFRS, which are given above, it follows that with a \(^{48}\)Ca-beam intensity of 1.2 \(\mu\)A, 0.35 mg/cm\(^2\) target thickness and a beam dose \(5 \times 10^{18}\) (realized for 200 hours of operation) the observation of one decay event corresponds to the production cross section of about 0.7 pb.

4. Experimental Results

For the synthesis of superheavy nuclei at DGFRS, the fusion reactions of \(^{48}\)Ca with target nuclei, the isotopes of Ra, U, Np, Pu, Am, Cm, Bk and Cf (11 isotopes of 8 elements), were used. The decay chains are presented in figure 3. In the investigations carried out at different \(^{48}\)Ca energies, 48 new nuclides were detected, all of them being evaporation products and their daughter nuclei in the region of \(Z = 104\) to \(118\) and \(A = 266\) to \(294\) [16-18]. The nuclei produced in cold fusion reactions are also presented in figure 3. In cold fusion reactions sequential \(\alpha\)-decays of neutron-deficient nuclei led to daughter products in a known nuclear region. The identification of the mass and charge number of a new nuclide is determined here by the decay properties of the known daughter nucleus and by
establishing correlations in sequential $\alpha$-transitions leading to its formation. Evaporation products of the reactions Act. + $^{48}$Ca have a larger neutron number; their sequential $\alpha$-decays take place among unknown nuclei and are terminated by spontaneous fission. In this case some additional measurements are needed.

![Diagram of nuclear masses](image)

**Figure 3.** Chart of the nuclides for $Z \geq 104$. The left and right shaded parts of the graphic present two regions of nuclei, which have been synthesized in cold fusion and in Act. + $^{48}$Ca reactions. The nuclei, which are located between these two regions, have been produced in fusion reactions of actinide target nuclei with light projectiles ($A \leq 26$). The half-lives (without experimental errors) for the main decay modes are given in the squares.

The atomic numbers for the odd nuclides (taking into account their long lifetimes) and for all nuclei in the decay chain can be, in principle, determined by means of the chemical isolation of the most long-lived nuclide. Such an experiment was performed including the identification of the odd-odd SF-nuclide $^{260}$Db ($T_{1/2} \approx 32h$), which terminated the 5-step chain of sequential decay of the mother nucleus $^{260}$115, synthesized in the $^{243}$Am + $^{48}$Ca reaction [19]. Independently, the charge and mass of the nuclei were determined by the excitation functions of the evaporation reaction channels. With this purpose, for all the studied reactions, the production cross sections of a given evaporation product were measured as a function of the energy of the bombarding ion (the excitation energies of the compound nucleus), varying the target isotope (or the mass of the compound nucleus). All the above-mentioned data give a consistent picture of the charges and masses of the nuclei, obtained in the experiments with the Act. + $^{48}$Ca reactions. The production cross section, the identification, as well as the decay properties of the $Z = 112, 114$ and now 116 were recently confirmed in several independent experiments [20-22].

It should be noted that the stability of the even-odd $\alpha$-emitters of element 112 is higher than in the case of their SF-daughter nuclei with $Z = 110$. Also, the $\alpha$-decay half-lives ($T_{\alpha}$) of the even-even isotopes of element 114 are considerably longer than the spontaneous fission half-lives ($T_{SF}$) of their daughter products – the isotopes of element 112. Such effects that indicate the increase of stability of the mother nuclei witness, most probably, of the presence and the strong effect of the theoretically predicted neutron shell at $N = 184$. In connection with this it would be interesting to perform quantitative comparison of the experimental results with calculations within different microscopic
nuclear models. It is noteworthy that the predictions of the existence of the islands of stability in the region of the hypothetical superheavy elements emerged indeed from these calculations.

5. Decay Properties of Superheavy Nuclei

5.1. Alpha - decay

As can be seen from figure 3, the odd isotopes of element 112 and all isotopes (even and odd) with \( Z \geq 113 \) predominantly undergo \( \alpha \)-decay. A comparison of the \( \alpha \)-decay energies of nuclides with mass \( 264 \leq A \leq 297 \) and atomic number \( 106 \leq Z \leq 118 \), synthesized in the cold fusion reactions \( ^{208}\text{Pb}, \, ^{209}\text{Bi} + ^{58}\text{Fe}, \, ^{64}\text{Ni}, \, ^{70}\text{Zn} \) and in the Act.\( ^{48}\text{Ca} \) reactions, are given in figure 4.

![Figure 4](image)

**Figure 4.** Comparison of the experimental values of the \( \alpha \)-decay energies \( Q_{\alpha}(\text{exp}) \) with the calculated within the framework of the macro-microscopic model [23]. The main data belong to nuclei, which have been synthesized in Act.\( ^{48}\text{Ca} \) reactions. In the left part of the graphic, the data for the light isotopes with \( Z = 107-113 \), obtained in cold fusion reactions, are also shown.

A difference of decay energy \( Q_{\alpha} \) between experiment and theory is present. The values \( \Delta Q_{\alpha} = Q_{\alpha}(\text{exp}) - Q_{\alpha}(\text{th}) \) are given for the calculation performed in the macro-microscopic model of ref. [23,24]. It can be seen that for all even-\( Z \) nuclei with \( Z = 106-118 \) and \( A = 271-294 \), and for nuclei with even as well as with odd number of neutrons, the quantity \( \Delta Q_{\alpha} \) does not exceed 0.5 MeV; for odd-\( Z \) nuclei: \( \Delta Q_{\alpha} \leq 1.0 \) MeV. In both cases, we can assume that there is quite good agreement of theory with experiment, moreover if we keep in mind that the calculation has been performed before obtaining the experimental data.

The comparison of \( Q_{\alpha}(\text{exp}) \) with the values \( Q_{\alpha}(\text{th}) \), calculated within the Skyrme-Hartree-Fock-Bogoliubov (HFB) and the Relativistic Mean Field models (RMF), was carried out, too (see [16]). In the HFB model a better agreement is obtained with masses from [25] calculated with 18 parameters. Finally, in the RMF model the agreement between theory and experiment is the least satisfactory. But it cannot be excluded that a better agreement can be achieved in this model also, if a different set of parameters is used. As a whole, the measured values of \( Q_{\alpha}(\text{exp}) \) are in agreement with theory, because the model calculations do not claim to be more precise in determining \( Q_{\alpha}(\text{th}) \) than 0.4-0.6 MeV.
5.2. Spontaneous Fission

For 11 out of the 48 synthesized nuclei spontaneous fission is the predominant mode of decay. In two more nuclei, $^{271}$Sg and $^{286}$114, spontaneous fission competes with $\alpha$-decay. For the remaining nuclides spontaneous fission was not observed. The partial SF half-lives of nuclei with $N \geq 163$, produced in fusion reactions with $^{48}$Ca, together with the half-lives of SF-nuclides with $N \leq 160$, are shown in figure 5. Four isotopes of element 112 with $N = 170$-173 are located in a region, where a steep rise of $T_{SF}(N)$ is expected. Indeed, in the even-even isotopes $^{282}$112 and $^{284}$112 the difference of two neutrons increases the partial half-life $T_{SF}$ by two orders of magnitude. The neighbouring odd isotopes $^{283}$112 and $^{285}$112 undergo $\alpha$-decay. For them, only lower limits of $T_{SF}$ can be determined (shown in the figure). Same for the even-even isotopes of element 114: the additional two neutrons in the nucleus $^{286}$114 ($T_{SF} \approx 0.13$ s) lead to increase of the stability relative to spontaneous fission. Such a picture is observed also for odd-Z isotopes: $^{281}$Rg ($T_{SF} \approx 26$ s) and $\alpha$-emitting odd-odd nucleus $^{282}$Rg [18]. It is evident that the rise of stability relative to spontaneous fission is observed for the nuclei, which are by 10-12 neutrons away from the closed neutron shell $N = 184$.

On moving to the nuclei with $Z < 110$ and $N < 170$ the probability for spontaneous fission decreases again when the close deformed shell $N = 162$ is approached. The stabilizing effect of the $N = 162$ shell manifests itself in the properties of the even-even isotopes of Rf, Sg and Hs with $N \leq 160$, which, as seen from figure 5, are well described by the mentioned model calculations. The odd SF-isotopes with $Z = 104$-110, produced in the $^{48}$Ca- induced reactions, are located in the transition region, where the larger the neutron number, the smaller the effect of the $N = 162$ shell. In this region, the $N = 184$ shell comes into effect. Such a behaviour of $T_{SF}(exp)$ as a function of $Z$ and $N$ correlates with the SHE fission barrier heights and has been predicted by all models: MM, HFB and RMF. For the isotopes of element 115, due to the strong hindrances to spontaneous fission of nuclei with odd proton (or/and neutron) number, $\alpha$-decay predominates as far as the $N = 162$ shell, where, similarly to the previous case, the sequences terminate by spontaneous fission.

Figure 5. Partial half-lives for spontaneous fission $T_{SF}$ vs. $N$ for nuclei with even $Z = 98$-114. Solid symbols and crosses denote even-even, open symbols – even-odd nuclei. Solid lines are drawn through the experimental points of even-even nuclei, the dashed lines – calculated $T_{SF}(th)$. Spontaneous fission half-lives of neutron-rich Db-isotopes and of the $^{281}$Rg are shown also.

6. Half-lives and stability of the heaviest nuclei

In figure 6 the half-lives of the isotopes of elements 110-118 are shown.
Figure 6. Half-lives of the nuclei with $Z \geq 110$ (in seconds) vs. neutron number $N$. The open symbols denote nuclei undergoing $\alpha$-decay, the solid symbols – spontaneous fission. All nuclides with $N \leq 165$ shown in the figure have been synthesized in cold fusion reactions, nuclei with $N > 165$ – in Act. + $^{48}$Ca reactions [16-19]. The curves are drawn through the experimental points to guide the eye.

It can be seen that the stability of the heavy nuclides in the region $N \geq 165$ rises considerably with the increase of $N$; the increase of the neutron number in the isotopes of elements 110-113 by $\Delta N = 8$ leads to half-lives greater by about 4-5 orders of magnitude. The rise in nuclear stability is observed also for the nuclides with $Z = 114$ and 116, in an interval $\Delta N = 4$. In the macroscopic models, such as the classical liquid drop model (or its modifications), trans-actinide nuclei in the absence of a fission barrier will undergo fission within some $10^{-19}$ s. The experimental half-lives, as it follows from figure 6, are about 17-19 orders of magnitude longer than expected from this model. Apparently, such a sharp distinction offers evidence of the strong influence of the shell structure of superheavy nuclei on their decay properties. While the relatively high stability in the region of the neutron-deficient deformed nuclei $Z = 110-112$ and $N = 160-165$ can be explained as due to the $N = 162$ shell, the further rise in the region of $N \geq 165$ definitely comes from the effect of another shell, situated at $N \geq 177$. According to the predictions of all microscopic models, this spherical shell is at $N = 184$ and comes after the lead shell at $N = 126$. We should note the strong effect of the $N = 184$ shell; it manifests itself even in nuclei that are at a distance of 12-14 neutrons (see figure 6).

7. Chemical studies of superheavy elements

As a consequence of the considerable rise of the half-lives of the isotopes of heavy and superheavy elements with the increase of the neutron number, it becomes possible to study their chemical behaviour using express radiochemical methods. As a matter of fact, for the first time it becomes possible to check the similarity or difference of the chemical properties of the heaviest elements when compared to their light homologues. In other words – there is a unique chance to determine the location of the superheavy elements in the Periodic Table.

As is seen from figure 3, most suitable for this purpose are the odd isotopes of 112 and 113 elements: $^{286}112$, $^{288}112$, $^{284}113$ and $^{286}113$ undergoing $\alpha$-SF decay. To what extent, for example element 112 is a homologue of Hg depends on the so-called “relativistic effect” in the electronic structure of the superheavy atom. According to some relativistic calculations, the chemical behaviour
of element 112 will have a noble-gas like inertness similar to radon [26]. Other calculations of the full electronic structure predicted its chemical behaviour similar to that of a noble metal [27, 28]. Will element 112 as actively as Hg interact with Au or just the opposite – will it be closer to the chemically neutral Rn? The reaction $^{242}$Pu($^{48}$Ca,3$n$)$^{287}$114($T_{1/2} \approx 0.5$ s)$-\alpha \rightarrow ^{283}$112 was used to produce the isotope $^{283}$112.

Both characteristics – the high volatility and the ability to form inter-metallic compounds for Hg, Rn and 112 atoms, defined the experimental setup for the chemical identification of element 112 shown on figure 7a [29]. The recoil nuclei leaving the $^{242}$Pu-target stopped in a He/Ar gas medium. Only nuclei with high volatility were transported to the detectors. Transport time from the stopping chamber to the detectors was 3.6 s. It was shown that all Hg atoms are registered by the first detectors with the Au coating. This can be explained by the strong adsorption on the detector Au-surface, which is due to the chemical reaction leading to the production of the Hg/Au compound. On the contrary, the decay of the chemically neutral Rn atoms is observed in the region of the last detectors, which are at the lowest temperatures (figure 7a), consistently with the adsorption enthalpy of Rn. In three experiments, carried out at different transport velocity of the recoil nuclei, 5 atoms of the isotope $^{283}$112 undergoing sequential $\alpha$-SF decay were detected with characteristics totally coinciding with the formerly obtained at the gas-filled separator.

For the atoms of element 112, as well as for the atoms of Hg, adsorption on the Au-surface of the detectors is observed [30]. This is evidence of the formation of an inter-metallic compound [112-Au].

**Figure 7.**

a) Experimental setup for simultaneous determination of the chemical adsorption of the atoms of the elements Hg, Rn and 112 onto the surface of 32 pairs of detectors at different temperatures (indicated in the figure) [29];

b) Enthalpy of sublimation and boiling point of the elements: Zn, Cd, Hg (literature data) and element 112 – according to data on the adsorption of the atoms of element 112 onto Au-surfaces at different temperatures [30].
Together with this, the distribution of the adsorbed atoms with \(Z = 112\) as a function of temperature is shifted relative to Hg in the direction of lower temperatures, which is an indication of the high volatility of element 112 (see figure 7b). Here lies the difference between the properties of Hg and element 112, which can be explained by relativistic calculations [28]. In our opinion, such investigations with heavier elements will make it possible to define the extension of the Periodic Table in its non-relativistic version. Experiments with the atoms of element 113 and 114 are presently being carried out at our Laboratory.

8. Conclusion

Decay properties of the nuclei obtained in Act. +\(^{48}\)Ca reactions show that the basic theoretical concept on the existence of closed shells in the region of the hypothetical superheavy elements and their decisive role in defining the limits of nuclear mass has received its experimental confirmation.

The experiments were carried out at the Flerov Laboratory of Nuclear Reactions (JINR, Dubna) in collaboration with the Lawrence Livermore National Laboratory (LLNL, Livermore), the Oak Ridge National Laboratory (ORNL, Oak-Ridge), the Vanderbilt University (VU, Nashville) and the Research Institute of the Atomic Reactors (RIAR, Dimitrovgrad, Russia) ; the experiments on the chemical identification of the isotopes \(^{268}\)Db and \(^{283}\)112 – within the collaboration:  Paul Scherrer Institute (PSI, Villigen) - Department for Chemistry and Biochemistry of the University of Bern – FLNR (Dubna) - LLNL (Livermore) - Institute of Electronic Technology (IET, Warsaw) with the participation of Dr. M. Hussonois from the Institute of Nuclear Physics (IPN, Orsay).

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