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Review Article

Super-heavy element research

Yu Ts Oganessian and V K Utyonkov

Joint Institute for Nuclear Research (JINR), Joliot-Curie 6, RU-141980 Dubna, Russian Federation

E-mail: oganessian@jinr.ru

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Abstract

A review of the discovery and investigation of the 'island of stability' of super-heavy nuclei at the separator DGFRS (FLNR, JINR) in the fusion reactions of 48 Ca projectiles with target nuclei 238 U- 249 Cf is presented. The synthesis of the heaviest nuclei, their decay properties, and methods of identification are discussed. The role of shell effects in the stability of super-heavy nuclei is demonstrated by comparison of the experimental data and results of theoretical calculations. The radioactive properties of the new nuclei, the isotopes of elements 112–118 as well as of their decay products, give evidence of the significant increase of the stability of the heavy nuclei with rise of their neutron number and approaching magic number N = 184.

Keywords: super-heavy elements, alpha decay, spontaneous fission

1. Introduction

The limits of nuclear stability, as it is known, are determined by interaction of nucleons in the nucleus. The neutron excess in nuclei leads to a decrease in the neutron separation energy. The limit comes at $B_n = 0 \,\text{MeV}$ (the neutron drip line). Similarly, $B_p = 0 \text{ MeV}$ (the proton drip line) determines the limit of existence of proton-rich nuclei. Another limitation arises for the extreme nuclear mass, which is defined by the probability of spontaneous fission (SF). In fact, when the fission barrier vanishes ($B_f = 0 \,\text{MeV}$), the nucleus loses its stability against fission ($T_{\rm SF} \sim 10^{-19}\,{\rm s}$). In the macroscopic approach (e.g. the liquid-drop model) the limit of existence of nuclei appears practically immediately after $Z \approx 100$ [1]. At the same time, it has been observed that in the binding energy of nuclei in their ground and highly deformed states there are variations depending on the proton or neutron number (nuclear shells). Also, the macroscopic models could not explain the variations of the fission barriers of the heavy nuclei: two times higher barrier in 208 Pb ($B_{\rm f} \approx 27\,{\rm MeV}$), practically unchanged fission barrier heights in the isotopes of the actinides from U to Fm and the nuclear shape isomerism [2], which manifests itself in heavy nuclei as 35 SF isomers [3] and many other effects, which have been seen in various experiments (figure 1). It had become obvious that the macroscopic approaches needed some corrections caused by structure of individual nucleus. The main achievements of the microscopic theory are connected with the development of the method of calculating such corrections for the ground and highly deformed states [9] based on the Nilsson or Woods–Saxon single particle potential to the smooth, macroscopic part of the energy.

The main structure corrections are governed by the nuclear shell effect. The concept of nuclear shells is here defined as a large-scale non-uniformity in the energy distribution of the individual particle states near the Fermi energy [10] directly connected to the nuclear binding energy. In many publications (e.g. see the reviews [10–13] and references therein), a number of the existing disagreements with experiment were explained by taking into account the shell effect when calculating the nuclear energy. One important consequence of these calculations was the disclosure of a significant gap in the spectrum of low lying levels in the region of the hypothetical super-heavy nuclei (SHN), viz. of a new (following N = 126) closed spherical neutron shell at N = 184 [14–17].

It was also shown that the considerable variations in the binding energy of spherical nuclei were due to the nuclear shells and that shell effects might be present also in deformed 'magic nuclei' (deformed shells) [9, 10, 18]. And finally, at further and quite significant increase of the deformation

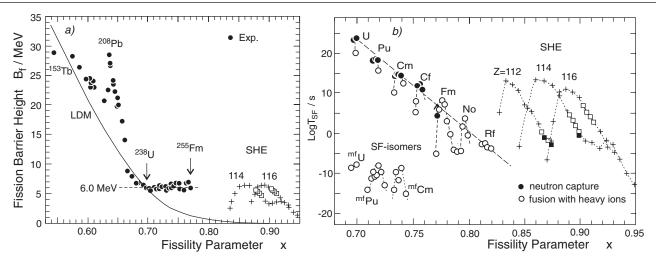


Figure 1. (a) Fission barrier heights as a function of the fissility parameter $x = (Z^2/A)/(Z^2/A)_{crit}$ at $(Z^2/A)_{crit} = 50.883$ [4]. Black points show experimental data, solid line—calculations in the liquid drop model [5, 6], crosses—calculated fission barrier heights in the macroscopic—microscopic model [7, 8] for the isotopes of elements 114 and 116, open squares—the same for the nuclei produced in the $^{242, 244}$ Pu, $^{245, 248}$ Cm + 48 Ca reactions. (b) Half-lives with respect to SF as a function of the fissility parameter x. Black points and open circles denote experimental values $T_{SF}(\exp)$ for SF of even—even nuclei from the ground and isomeric states. Dashed line is drawn to guide the eye though the maximum $T_{SF}(\exp)$ values and is extrapolated into the transactinide region according to macroscopic concept. Crosses show calculated values $T_{SF}(th)$ in the macroscopic—microscopic model [7, 8] for even—even isotopes of elements 112, 114 and 116, open squares—the same for the nuclei produced in the $^{242, 244}$ Pu, $^{245, 248}$ Cm + 48 Ca reactions, black squares—the same for the isotopes for which the SF half-lives have been measured.

arising in fission, *the shell effects continued to play an important role* in defining the potential energy and the nuclear inertial masses [7, 10].

The predictions of the nuclear properties change significantly depending on the effect of the new shells. The sum of the smooth part of the deformation energy $E_d(LD)$ and the shell correction $\Delta E_d(Shell)$ bring forth the appearance of a fission barrier.

For the heaviest nuclei with Z=112–114 and N=180–184 (see figure 2), the fission barrier height may amount to $B_{\rm f} > 6\,{\rm MeV}$ (higher than for $^{238}{\rm U}$). Therefore, the partial SF half-lives, as shown in the calculations of [7, 8], increase up to $\sim 10^5\,{\rm year}$ (figure1(b)). Then, $T_{\rm SF}$ exceeds the estimates of the macroscopic models by a factor of 10^{30} ! Less striking, but also quite strong, is the effect expected for deformed nuclei with Z=106–108 and $N\approx 162$, the effect of the deformed shells suppresses the probability of SF by a factor of $>10^{20}$. Here, an interesting situation arises.

Because of the high stability with respect to SF, the heaviest nuclei will undergo α - or β -decay (T_{α} , T_{β} << T_{SF}). From the calculated partial half-lives T_{α} , T_{EC} , β and T_{SF} , shown in figure 3, we may get an impression of the scenarios and the decay properties of the heaviest nuclei depending on Z and N. The consecutive α decays will follow until the shell effect weakens and SF becomes the main decay mode. In the region of N < 162 this is observed for even—even isotopes [20]. In the case of odd nuclei, due to the large hindrances to SF, α decay may occur down to long-living nuclei without competition from SF. In fact, this is observed in the experiments with Pb-target based reactions [20, 21]. For the heavier neutron-rich nuclei, the decay sequences of both even and odd isotopes will end by SF. The total decay time will be then determined to a great extent by the neutron number of the parent nucleus.

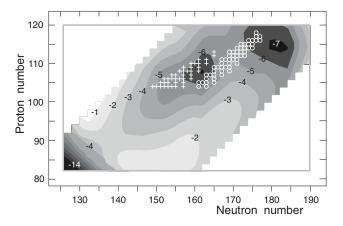


Figure 2. The map of the shell corrections $\Delta E_{\rm shell}$ to the nuclear macroscopic potential energy [18, 19]. The numbers at the contour curves correspond to the amplitude of the shell correction (in MeV). Crosses denote nuclei with $Z \ge 104$, obtained in cold-fusion reactions, open circles—in actinides (Act) + 48 Ca reactions.

When approaching the N = 184 shell, we may expect a strong increase in the decay time.

Above we gave an example of the predictions of the macroscopic—microscopic models (MM). Other, purely microscopic self-consistent approaches as the Hartree–Fock–Bogoliubov (HFB) model and the relativistic mean field (RMF) theory also predict significant increase of the binding energy of heavy nuclei at N=162 and N=184. In spite of differences in these models, the maximum shell effect corresponds to N=184 (like in MM calculations) but the proton shell closure with a higher number of protons Z=120, 122, 124 or even 126 is expected (see review [22] and references therein). Meantime, uncertainties in the quantitative estimations of the

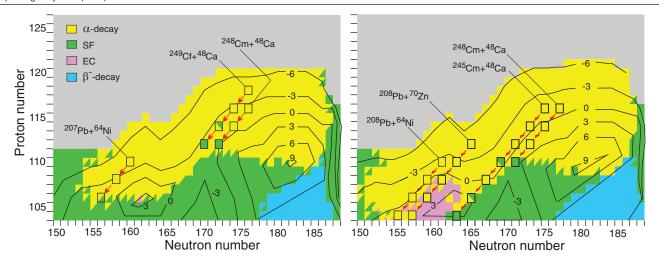


Figure 3. Contour map of the calculated half-lives as $Log(T_{1/2})$ (in seconds) and decay modes of the nuclei with $Z \ge 104$ and $N \ge 150$ (taken from [20]). The left graph refers to even—even nuclei, the right one—to odd-A nuclei. The regions corresponding to different decay modes are shown in different color. The consecutive decay of nuclei, produced in cold fusion and in actinides + 48 Ca reactions are also shown (see text for details).

nuclear shell effects do not change the general conclusion of the theory that in the large interval of masses from 250 to 320 'islands of stability' may arise, considerably changing the limits of existence of atomic nuclei.

2. Reactions of synthesis of super-heavy nuclei

As it was shown in various experiments, the methods of consecutive neutron capture previously used for production of nuclei with $Z \le 100$ or transfer reactions of nuclei, even so heavy as U + U, cannot be applied for synthesizing super-heavy elements¹. The only way for producing the heaviest nuclei remains making use of complete-fusion reactions which were applied for the synthesis of elements of the second hundred.

In theory, the process of formation of the evaporation residue (ER) consists of three consecutive stages. At the first stage, colliding nuclei overcome the Coulomb barrier and approach the point of contact. Quasi-elastic and deep-inelastic reaction channels dominate at this stage, leading to formation of projectile-like and target-like fragments in the exit channel. Then, the composite system can evolve into the configuration of an almost spherical compound nucleus (CN). After dynamical deformation and exchange by nucleons, two touching nuclei can re-separate into fragments similar to colliding nuclei or can go directly to fission channels without formation of spherical compound nucleus which is called quasi-fission. Finally, the compound nucleus cools down by the emission of neutrons and γ rays surviving fission and forming ER in its ground state. This process takes place in strong competition with fission of excited nucleus.

For each angular momentum l the partial ER cross section $\sigma_{xn}(E^*, l)$ for production of the final nucleus in its ground state at projectile energy E and corresponding excitation energy E^* of CN is factorized as the product of the partial

capture cross section $\sigma_{\text{capt}}(E, l)$, the fusion probability $P_{\text{fus}}(E, l)$ and the survival probability $P_{\text{surv}}(E^*, l)$,

$$\sigma_{xn}(E^*, l) = \sigma_{\text{capt}}(E, l) \cdot P_{\text{fus}}(E, l) \cdot P_{\text{surv}}(E^*, l). \tag{1}$$

The difference in mass of the initial nuclei A_P , A_T and the final nucleus $A_{CN} = A_P + A_T$ defines the excitation energy of the CN E^* . The minimum value of E^* at the reaction Coulomb barrier B_C is:

$$E^*_{\min} = B_{\rm C} - Q \text{ with } Q = M_{\rm CN} - (M_{\rm P} + M_{\rm T}).$$
 (2)

Energy $E^*_{\rm min}$ depends on the masses (in eV) of the interacting nuclei. When the mass ratio $A_{\rm P}/A_{\rm T}$ increases (from $A_{\rm P}=4$ to 20–30), with fixed $Z_{\rm CN}$, the excitation energy increases also (rise of the Coulomb barrier), but then becomes lower due to increasing Q. The advance to the nuclei with closed proton or neutron shells leads to an additional reduction of $E^*_{\rm min}$ [23].

Since 1974, the cold fusion reactions of 208 Pb, 209 Bi with massive projectiles ($A_P \ge 50$) have been used in the synthesis of the heaviest elements. With practically fixed mass of the target (208 Pb or 209 Bi), the rise of the atomic and mass numbers of the evaporation products is entirely connected with the increase in the mass (and charge) of the projectile. When the projectile becomes more and more heavy, the excitation energy of the CN decreases down to $E^* \approx 15-10\,\text{MeV}$ (cold fusion). Transition to the ground state takes place by the emission of only one neutron [20, 21]. As a result, the survivability of the CN $P_{xn}(E^*)$ significantly increases, this being the main advantage of cold-fusion reactions.

In cold-fusion reactions, ERs are some 10–15 mass units shifted from the β -stability line which leads to a considerable decrease in their half-lives. Furthermore, the cross section of ERs produced in cold-fusion reactions exponentially decreases with the increase of $Z_{\rm CN}$ because of rise of the Coulomb repulsion forces. When $Z_{\rm CN}$ changes from 102 to 113, the cross section decreases by a factor of 10^8 . Because of small neutron excess in the evaporation products and further considerable decrease of the cross section with increase of the projectile mass ($A_{\rm P} > 70$), it is impossible to reach the region

 $^{^{1}}$ In this paper, we will use term 'super-heavy elements' for the heaviest nuclei (atoms) which stability is governed by existence of the new spherical shells at Z = 114 and N = 184 predicted for the first time within the macroscopic–microscopic nuclear theory.

of SHN. Obviously, for the synthesis of these nuclei ($Z \ge 114$) with high neutron excess, it was necessary to look for other reactions.

In order to decrease the factors hindering fusion, it is desirable to make use of more asymmetric reactions and to obtain an increase in the neutron number of the ERs by using both target and projectile nuclei with maximum neutron excess. As a target material, it is reasonable to use neutron-rich isotopes of the actinides produced in high-flux reactors and thus have the largest neutron excess. Among the projectiles, the undoubted advantage is possessed by the doubly magic nucleus of the rare isotope 48 Ca. The CN 292 114, produced, for example, in the fusion of 244 Pu and 48 Ca ($Z_P \cdot Z_T = 1880$), acquires 8 additional neutrons compared to the problematic 208 Pb + 76 Ge ($Z_P \cdot Z_T = 2624$) reaction. Coulomb repulsion in the reaction 244 Pu + 48 Ca decreases by almost 40%, which in turn should lead to a strong decrease in the factors hindering the formation of the CN.

The last stage—the survival of the CN—is the decisive one in the given method of synthesis of the heaviest nuclei. The estimations of $E^*_{\rm min}$ and experiments aimed towards measuring the excitation functions for evaporation products, have shown that the CN with $Z_{\rm CN}=112$ –118, when formed in actinides + 48 Ca reactions, may attain excitation energy from 30 to 55 MeV (see section 4). This energy will be released by a cascade emission of 2 to 5 neutrons (the evaporation of charged particles is significantly less probable) and γ -rays. At each step, neutron evaporation and fission compete strongly. The survival probability can be expressed simply as:

$$P_{xn}(E^*) \sim \prod_{i=1}^{x} (\Gamma_n / \Gamma_f)_i \sim \prod_{i=1}^{x} \exp[(B_f - B_n / T)]_i.,$$
 (3)

where $B_{\rm f}$ and $B_{\rm n}$ are the fission barrier height and the binding energy of the neutron, respectively, T is the temperature of the CN and x—the number of emitted neutrons.

According to calculations, the fission-barrier height of nuclei with Z > 104 is entirely determined by the amplitude of shell correction. The large decrease of production cross sections for increasingly heavy nuclei, observed in asymmetrical hot-fusion reactions, is connected with their lower survivability caused by reduce of fission barriers for CN with an increase in Z and N. However, if the predictions of the theoretical models about the existence of the next closed shell N = 184 are justified, the fission barrier height will again increase when advancing the region where $N_{\rm CN} \ge 174$ and $Z_{\rm CN} \ge 112$. In turn, the nuclear survivability will increase too and as a result, one can expect even a rise in the cross section for heavy nuclei with higher Z and large neutron excess.

3. Experiments

The half-lives of nuclei, products of the complete-fusion reactions of neutron-rich isotopes of heavy actinide elements with ⁴⁸Ca projectiles, were expected to vary in a wide range: from microseconds (for the even–even isotopes of the heavy elements, see, e.g. [7, 22]) up to days (for the products of sequential decay of the odd nuclei). Their calculated predominant

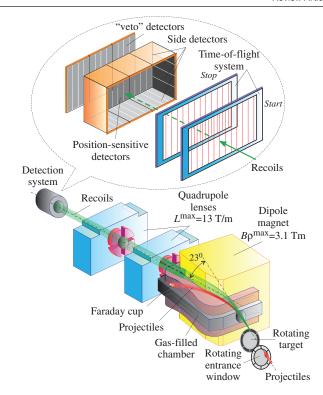


Figure 4. Layout of the DGFRS and detection system. $L^{\rm max}$ and $B\rho^{\rm max}$ are maximum field gradient and magnetic rigidity, respectively.

decay modes were α decay and SF. Production cross sections of the ERs could be at the level of a picobarn $(10^{-36}\,\mathrm{cm^2})$ or even lower. The recoiling nuclei, formed with full momentum transfer from the projectile to the CN, leave the target layer in the beam direction. Therefore, experimental equipment should separate them from the beam particles, scattered nuclei and transfer-reaction products.

The Dubna gas-filled recoil separator (DGFRS) is shown schematically in figure 4. The DGFRS has a DQ_hQ_v magnetic configuration: a flat-field dipole magnet with inclined poles for horizontal focusing (D) followed by horizontally (Q_h) and vertically (Q_v) focusing quadrupole magnets.

The ERs recoil out of a thin target with the momentum of the beam particle and enter the dipole-magnet chamber filled by gas at the pressure of about 1 Torr [24]. For keeping gas inside the DGFRS at the beam intensities of ⁴⁸Ca up to 10^{13} /s delivered by the U400 cyclotron, a rotating 1.6 μ m Ti window is mounted at its entrance as well as a $0.5-1.5 \mu m$ Mylar fixed window separating the detection system. After emerging from the target layer, the heavy atoms have a large ion charge $(q \approx 20^+)$ with broad distribution. Due to charge exchange in consecutive collisions with the gas atoms, the distribution of the ERs charges rapidly becomes narrower and the mean charge decreases to the equilibrium value $(q \approx 6^+)$ [25] whereas average ion charge of projectiles is $q \approx 18^+$ due to their high velocity. In sequential collisions with the atoms of the medium, the heavy atoms slow down and move along some average trajectory. Ions with mass (m), average charge (q) and velocity (v), will be deflected in a magnetic field strength (B) following a trajectory with curvature radius (ρ) . The ER trajectory in the field of a gas-filled

dipole magnet is determined by momentum and charge as follows:

$$B\rho = m v_0 Z^{-1/3} e^{-1} = 0.0227A Z^{-1/3} [T m],$$
 (4)

where A is the mass number and v_0 is the velocity of the electron (e) in Bohr's hydrogen atom $(2.19 \times 10^6 \,\mathrm{m\,s^{-1}})$.

Among all the reaction products, the complete-fusion reaction products have the largest possible mass. Accordingly, the other products—target-like or projectile-like ions—will be spatially separated in flight from the ERs following the trajectories with lower curvature radii. Thus, in experiments on the synthesis of SHN, the DGFRS suppresses the full-energy 48 Ca projectiles, projectile-like ions and target-like nuclei by factors of about 3×10^{15} – 10^{17} , 3×10^{13} – 6×10^{14} and 10^4 – 10^6 , respectively [26, 27]. The transmission efficiency of the separator for Z = 112–118 nuclei was estimated to be about 35–40% [24] for the size of the focal-plane detector of $120 \times 40 \,\mathrm{mm}^2$.

A specific characteristic of the DGFRS is the hydrogen gas used in the separator, which enables better suppression of projectile- and target-like recoils at the focal plane than the helium gas [28, 29] which is used in other separators (e.g. BGS [29, 30], TASCA [31, 32]). Due to the approximately linear dependence of the ion charge on velocity, transmission of ERs produced in the same reaction at different projectile energies varies weakly. Three isotopes of Fl (Z = 114) were observed in this reaction with masses 287, 288 and 289 which differ by only about 0.3%; the change of the DGFRS setting was not needed. After separation in the DGFRS dipole magnet from the beam particles and products of unwanted reactions, the ERs are focused by the quadrupole doublet (see figure 4) onto the separator focal plane located about 4m downstream. The heaviest nuclei, leaving the target with energy of 35–40 MeV, pass over this distance in about 1μ s. Due to kinematics of 252No close to that of the SHN produced in the ⁴⁸Ca-induced reactions, the ²⁰⁶Pb(⁴⁸Ca,2n)²⁵²No reaction can be used for testing and calibration of the separator and detection system. This reaction was used for determining the optimal thickness of the target also. The total yield of ²⁵²No increases with growth of the target thickness up to approximately 0.5 mg cm⁻². For thicker targets, the yield remains the same because of the lower transmission caused by multiple scattering of the recoils [33].

In the experiments on the synthesis of the heaviest nuclei performed at the DGFRS, targets of actinide oxides were used with thickness of about 0.4 mg cm⁻² electrodeposited on a 1.6 μ m Ti foil. Enriched isotopes of ²³⁸U, ²³⁷Np, ^{242,244}Pu, ²⁴³Am, ^{245,248}Cm, ²⁴⁹Bk and ²⁴⁹Cf were used as target material. Each individual target had an area of 5.4–6.0 cm² in the shape of an arc segment with an angular extension of 60° and an average radius of 60 mm. The six segments were mounted on a disc that was rotated at 2000 rpm in plane perpendicular to the beam direction. The total area of the target was 32–36 cm², the total weight of the target material was about 10–15 mg. Before implantation into the detector, the separated ERs passed through a time-of-flight (TOF) measuring system that consists of two (start and stop) multiwire proportional chambers placed within 6.5 cm from each other and filled with

pentane at ≈ 1.5 Torr. The TOF system allows distinguishing recoils coming from the separator and passing through the TOF system from signals, arising from α decay or SF of the implanted nuclei (without a TOF signal). In order to eliminate the background from the fast light charged particles (protons, α 's, etc produced from direct reactions of projectiles with the DGFRS media) with signal amplitudes lower than the registration threshold of the TOF detector, a 'veto' silicon detector was placed behind the front detector (figure 4).

ERs are finally implanted in the focal-plane detector consisting of 12 vertical position-sensitive strips (three $4 \times 4 \text{ cm}^2$ 0.3 mm-thick chips, each with four strips) providing horizontal resolution. The vertical position is determined by the resistive charge division within each strip. The implantation depth of ERs in the Si detector is lower by several times than α -particle range of SHN; thus, α particles can escape the focal-plane detector. To detect these particles as well as fission fragments, eight similar detectors without position sensitivity were located upstream and perpendicular to the focal-plane detector forming a five-sided box configuration. This results in increase of detection efficiency for full-energy α particles from about 52% for focal-plane detector only to approximately 87% after reconstruction of their energies deposited in the focal-plane and side detectors. The detection efficiency for SF of the implanted nuclei is close to 100%. Since 2012, to increase the position granularity of the detectors, which reduces the probability of observing sequences of random events that could imitate decay chains of synthesized nuclei, the new focal-plane detectors have been used. These consiste of two $6 \times 6 \text{ cm}^2$ detectors each having 16 strips with position sensitivity and six similar side detectors without position sensitivity. The detection system of the DGFRS was calibrated by registering the recoil nuclei and decays (α or SF) of known isotopes of No and Th and their descendants produced in the reactions 206 Pb(48 Ca,2n) and nat Yb(48 Ca,3-5n), respectively. Using known energies of ²¹⁵Ra and ²¹⁷Th the energy resolutions (full width at half maximum (FWHM)) were determined separately for α particles completely absorbed in the focalplane detector, for α particles that escaped this detector with a low energy release and were registered by a side detector and for α particles detected only by a side detector (without a focal-plane position signal). For instance, in a recent experiment with ²⁴⁹Bk target [34] these values were 34–73 keV, 83– 120 keV and 0.73-0.98 MeV, respectively. Fission fragments from the decay of ²⁵²No implants produced in the ²⁰⁶Pb + ⁴⁸Ca reaction were used for the total kinetic energy (TKE) calibration. The typical position resolutions of correlated ER- α and ER-SF signals were 1.1-1.8 and 0.5-1.2 mm [34], respectively. For α particles detected by both the focal-plane and side detectors, the ER- α position resolution depends on the energy deposited in the focal-plane detector and is generally inferior to that obtained for the full-energy signal.

From theoretical calculations and the available experimental data, one can estimate the expected α -particle energies of the ERs and their descendant nuclei that could be produced in a specific reaction of synthesis. For α particles emitted by the parent or daughter nuclei, it is possible to choose wide enough energy and time gates $\delta E_{\alpha l}$, $\delta t_{\alpha l}$, $\delta E_{\alpha 2}$, $\delta t_{\alpha 2}$, etc (accounting for

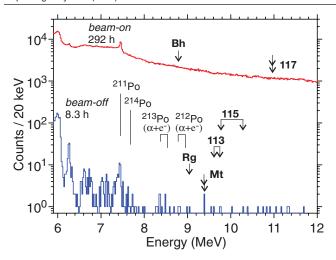


Figure 5. Total energy spectra of beam-on α -like signals (all events detected by the focal-plane detector or both the focal-plane and side detectors without registering TOF signal) and beam-off α particles recorded during the 247-MeV ²⁴⁹Bk+⁴⁸Ca run. Durations of the beam-on and beam-off intervals are given. The arrows show the energies of events observed in two correlated decay chains of ²⁹⁴117 [36].

all the uncertainties in the estimation of the expected energies and half-lives of the synthesized nuclei) and employ a special low-background detection scheme [35]. For instance, in [34] during the irradiation of the target, the beam was switched off after a recoil signal was detected with parameters of implantation energy and TOF expected for ERs, followed by an α -like signal within energy interval $\delta E_{\alpha l} = 10.7 - 11.3$ MeV in the same strip within some position window (3.2 mm) and time interval $\delta t_{\alpha l}$ (0.4 s). If the first α particle escaped the focal-plane detector and a position signal was not detected, then switching off the beam could be done when the second α particle in the corresponding $\delta E_{\alpha 2}$ (9.6–10.7 MeV) and $\delta t_{\alpha 2}$ (2 s) intervals was detected. If, during the first preset beam-off time interval, an α particle with energy expected for daughter nuclei was registered in any position of the same strip, the beam-off interval could be automatically extended to any time. This operating mode of the DGFRS is illustrated by detection of long decay chains of the odd-odd isotope of element 117 produced in the 249 Bk(48 Ca, ^{3}n) 294 117 reaction [36]. Figure 5 shows the spectrum of α -like signals (all events detected by the focal-plane detector or both the focal-plane and side detectors without a registered TOF signal) in all strips accumulated during the 247 MeV 249 Bk + 48 Ca experiment. The α -particle spectrum detected during beam-off time intervals is also shown. In the high-energy part of the α -particle spectrum, where the decays of daughter nuclei ²⁷⁴Bh to ²⁹⁰115 ($E_{\alpha} = 8.5-10.5 \,\text{MeV}$) are expected, 16 events were detected with average counting rate of about 2/h. This demonstrates very low random probability for detection of 7α particles (shown by arrows) which belong to the decays of the daughter isotopes of ²⁹⁴117 and occur within about a 3 min time interval after the decay of the parent nucleus. The calculated numbers of random sequences imitating each of the observed decay chains ranged from 2×10^{-4} to 3×10^{-20} depending on number of registered nuclei in the chain and counting rate of random events in the focal-plane and side detectors [34].

One of the crucial questions in the synthesis of SHN lies in identification of the new isotopes or experimental determination of their atomic (Z) and mass (Z + N) numbers. Note, all the decay chains of SHNs were terminated by SF of previously unknown nuclei. Moreover, these descendant neutron-rich nuclides with lower Z cannot be synthesized and identified in direct reactions because of lack of reactions with stable projectiles leading to these nuclei. That is why the method of genetic α -particle correlations between first observed and well-known nuclei, widely used during last decades (see, e.g. [20, 21]), can be applied after independent identification of one of the members of the decay chain.

At the same time, the super-heavy nuclei and their daughters can be identified by other methods [37]. One of these is connected with mechanism of the complete-fusion reaction which essentially differs from other reactions of ⁴⁸Ca projectiles with heavy target nuclei. The de-excitation of CN occurs via evaporation of neutrons; their number depends on the excitation energy. The shape of excitation functions $\sigma_{xn}(E^*)$ looks like assimetric pseudo-Gaussian curve with FWHM of about 10 MeV and with maximum located at the energy corresponding to the number of evaporated neutrons (see, e.g. figures 7 and 9 below). The probability of evaporation of charged particles (protons or α particles) is suppressed due to high Coulomb barrier in heavy CN. The DGFRS strongly separates forward-peaked ERs with huge suppression of the scattered beam particles and the products of incomplete fusion (like $\alpha x n$) or transfer reactions.

The measurement of excitation function of the reaction allows determining the number of evaporated neutrons and thus, the masses of ERs when two or even three different isotopes are registered and difference of their decay properties is in agreement with expectations for the neighbouring isotopes of the same element. Practically all the investigated reactions which resulted in observation of SHN with Z = 112-118 at the DGFRS were studied at several bombarding energies [4, 26, 27, 34, 36].

An additional approach to the identification of nuclei is connected with variation of mass and charge of target in the cross bombardments with ⁴⁸Ca projectiles. In this method, one and the same nucleus can be observed either as the product of evaporation of a different number of neutrons from CN produced in the reactions with different target isotopes or as α -decay product of parent nuclide synthesized in the reaction with a heavier target nucleus (with higher Z). As an example, figure 6 shows cross reactions used in experiments on the synthesis of Fl and Lv. Finally, the α decay properties and SF half-lives of synthesized nuclei can evidently demonstrate their origin. For α decay of the even–even nuclei, one may expect with large probability transitions through ground states of the parent and daughter nuclei. In this case, measured α -particle energies E_{α} and partial half-lives T_{α} should reflect energy (Q_{α}) and probability of α transitions, which can be compared with predictions of different theoretical models of α decay and systematics of Q_{α} values for numerous known

Comparison of α -particle energies of even-Z nuclei with those observed for neighbouring odd-Z nuclei can provide

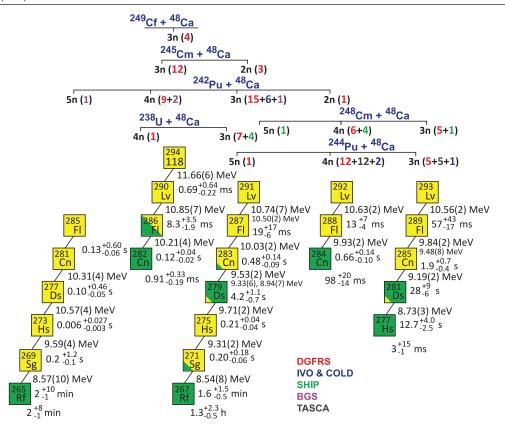


Figure 6. Summary decay properties of the isotopes of even-*Z* elements observed among the products of 48 Ca beam and 238 U, 242,244 Pu, 245,248 Cm and 249 Cf target reactions. The numbers of the decay chains of the given isotopes, the products of corresponding *xn*-evaporation channels, observed in experiments with use of the DGFRS (in red) [39–45], IVO setup with COLD detector (in blue) [46–49], SHIP (in green) [50, 51], BGS (in magenta) [52, 53] and TASCA (in grey) [32, 54] are shown. The average energies of α particles and half-lives are given for all α emitters observed in these experiments (yellow squares). The energies of rare α lines are given by smaller font. The energy uncertainties given in brackets correspond to the data with the best energy resolution. For spontaneously fissioning nuclei marked by green squares the half-lives are listed.

valuable information for identification. For unhindered α decays, from Geiger–Nuttall relation $T_{\alpha}(Z, Q_{\alpha})$, one can estimate atomic numbers of all nuclei in the α -decay chains of the new elements. For SF nuclides, the half-lives of nuclei with an odd number of neutrons and/or protons exceed those for even–even nuclei by several orders of magnitude [38].

In approaching neutron shell N=184, a considerable increase in the stability of SHN is expected. Super-heavy nuclei—neutron-rich isotopes of elements 112, 113 and 114 produced in the Act+⁴⁸Ca reactions—have half-lives from tenths to tens of seconds (figures 6 and 8 below). In the course of consecutive α decays, the partial half-life T_{α} increases. Half-lives of the isotopes of Rf and Db at the end of decay chains of SHN for which SF was observed reach hours and days. For identification of atomic numbers of SHN the methods of fast chemistry may be applied. In contrast to short-lived products of the cold-fusion reactions, the new region of chemistry of the heaviest elements becomes available.

Most of these methods were applied in experiments on the synthesis of SHN and will be discussed in the following section. Even use of combination of a few methods of identification for the full set of results obtained in the series of experiments aimed at the investigation of SHN allows us to determine unambiguously atomic and mass numbers of synthesized nuclei.

4. Results

4.1. Even-Z nuclei

4.1.1. Synthesis and identification of element 114 flerovium. The largest neutron excess in the CN with 114 protons can be achieved in the 244 Pu + 48 Ca complete-fusion reaction. The first superheavy nucleus was discovered on June 25, 1999, in experiments performed at the DGFRS by Dubna (FLNR)-Livermore (LLNL) collaboration. Two identical decay chains were observed. Each consisted of two consecutive α decays terminated by SF of the third nucleus (see figure 6). In this experiment, carried out at the lowest projectile energy only, the parent nucleus was assigned to 288 Fl [39]. Later on, the cross-section measurement in combination with decay properties of produced nuclei gave correct identification of the mass number of 289 for this isotope of element 114.

In 2003 the study of the 244 Pu + 48 Ca reaction was continued at higher 48 Ca energies [42] (see figure 7). The same isotope was observed at two higher excitation energies. At increased energies, another isotope, 288 Fl, with different decay properties was produced also. Its decay chain consisted of α decay

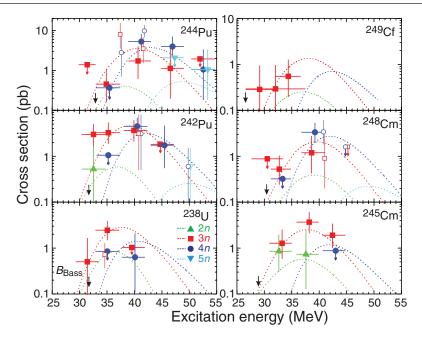


Figure 7. Excitation functions for the 2n (green triangle up), 3n (red square), 4n (blue circle) and 5n (cyan triangle down) evaporation channels from the complete-fusion reactions 238 U, 242,248 Cm, 245,248 Cm, 249 Cf + 48 Ca measured at the DGFRS [39–45] (solid symbols) and the SHIP [50, 51], BGS [52, 53] and TASCA [32, 54] (open symbols). For reference purposes, the Bass barrier [55] is shown by a black arrow in each panel.in the left bottom panel it is labeled with B_{Bass} . Lines show the results of calculations [56–59]. Vertical error bars correspond to statistical uncertainties [60] for the DGFRS experiments and available data for other setups. Horizontal error bars represent the range of excitation energies populated at given beam energy. Position of some symbols were shifted from centers of energy intervals for avoidance of their mixing. Upper cross-section values are shown by colored arrows.

of parent nucleus and SF of daughter nuclide which half-life is lower by factor of 300 than that for daughter isotope in the first decay chain. Finally, at the highest energy the third isotope, 287 Fl, was observed. The consideration of decay properties of daughters in the three chains evidently indicated that the second chain should originate from even-N nucleus and neighbouring isotopes should have odd number of neutrons. This is explained by the hindrance factor increasing SF half-lives of odd-N and odd-N nuclei by a few orders of magnitude that was established for many nuclei (see, e.g. [38]). Such assignment is in agreement with fine structure in α decays of synthesized nuclei, which will be discussed below (see figures 6 and 11).

The measured production cross sections of all the three isotopes agree well with expectations for complete-fusion reactions with evaporation of three, four and five neutrons from CN ²⁹²Fl [56–59, 61, 62]. The decay properties of ²⁸⁷Fl were investigated in more detail and the new lighter isotope ²⁸⁶Fl was synthesized in the cross bombardment of ²⁴²Pu by ⁴⁸Ca [44] in 2003. The isotope ²⁸⁷Fl, product of the 5*n*-evaporation channel of the 244 Pu + 48 Ca reaction, was observed also in the 242 Pu(48 Ca,3n) reaction at the three lowest excitation energies of the CN ²⁹⁰Fl [44] (see figures 6 and 7). Further increase of projectile energy allowed synthesis of the new isotope ²⁸⁶Fl. Its decay chain was similar to that of its heavier even-even neighbour 288 Fl, an α decay of ²⁸⁸Fl and SF of daughter nuclide ²⁸⁴Cn. In agreement with theoretical expectations, the stability of nuclei, especially against SF, decreases with receding from the neutron magic number N = 184: even–even isotope ²⁸⁶Fl with approximately equal probabilities undergoes α decay and SF.

In the $^{238}\text{U} + ^{48}\text{Ca}$ reaction, investigated in 2003–2004 [44], one event of SF was attributed to the decay of ^{282}Cn and

seven α -decay chains of ²⁸³Cn were detected at three projectile energies (see figures 6 and 7). In one case ²⁷⁹Ds underwent α decay, instead of the more probable SF mode, then two more α decays of ²⁷⁵Hs and ²⁷¹Sg and SF of ²⁶⁷Rf were registered. A similar long chain was observed in the ²⁴²Pu(⁴⁸Ca, 3n)²⁸⁷Fl reaction also [44]. Once again this long decay chain has been registered in the third cross bombardment ²⁴⁵Cm(⁴⁸Ca, 2n)²⁹¹Lv [42, 45] (see below).

Experiments on the synthesis of Fl isotopes were repeated in other laboratories. A study of the ²⁴²Pu + ⁴⁸Ca fusion reaction at the BGS was published in 2009–2010. One and two decay chains of ²⁸⁷Fl and ²⁸⁶Fl, respectively, were observed [52, 53] with decay modes, half-lives and decay energies in agreement with results published by the DGFRS group [42–45]. Besides, one more decay chain of the lightest isotope ²⁸⁵Fl, product of the 5*n*-evaporation channel of the ²⁴²Pu + ⁴⁸Ca reaction, was found to consist of five consecutive α decays terminated by SF of 265 Rf (see figures 6 and 7). In addition, the 238 U + ⁴⁸Ca reaction was studied at one ⁴⁸Ca energy at the SHIP [50] in 2005-2007. Here two decay chains were measured, which fully confirm data that were previously assigned to the isotope ²⁸³Cn in experiments at the DGFRS. Two ER-SF chains were assigned to a 50% SF branch of this isotope; this, however, was not evident from the data where ²⁸³Cn was observed as daughter nucleus after α decay of ²⁸⁷Fl [42–45, 52] and the upper limit of 0.1 was set for a SF branch of ²⁸³Cn [44]. And finally in 2009, the decay properties of ^{288,289}Fl were also confirmed in experiments [32, 54] performed with use of the TASCA (see figures 6 and 7). In addition, in this work a rare α -decay branch for ²⁸¹Ds and SF of ²⁷⁷Hs were detected in one decay chain of ²⁸⁹Fl.

In 2011 a joint IUPAC/IUPAP Working Party (JWP) recommended that the Dubna–Livermore collaboration be credited with discovery of the new element 114 produced in the ²⁴²Pu(⁴⁸Ca, 3*n*)²⁸⁷114 reaction [63]. The element with atomic number 114 was named flerovium (FI) to honour the Flerov Laboratory of Nuclear Reactions which was founded by Flerov and where super-heavy elements were synthesized [64].

The relatively high stability of 283 Cn $(T_{1/2} \approx 4 \text{ s})$ allowed researchers to investigate for the first time, the chemical properties of element 112. In 2006-2007, the IVO setup was applied for collecting the ²⁴²Pu + ⁴⁸Ca reaction products in the chamber filled by a He/Ar carrier gas which delivered volatile reaction products (including short-lived isotopes of Hg and Rn) to detection system COLD. Here, atoms were deposited according to their interaction with the detector surfaces. The COLD consisted of an array of 32 pairs silicon detectors, with the active surfaces facing each other. The surface of one detector in each of 32 pairs was covered by gold layer. The temperature gradient was established along detectors by a thermostat heating at the inlet and a liquid-nitrogen cryostat cooling near the outlet. The primary fusion-evaporation reaction product ²⁸⁷Fl has a half-life of about 0.5 s which was too short compared with the average transport time from the reaction chamber to the detector. Thus, only the daughter nucleus ²⁸³Cn could reach the detector. In these experiments, five decay chains of ²⁸³Cn were registered [46, 47]. By directly comparing the adsorption characteristics of ²⁸³Cn to that of mercury and the noble gas radon, it was found that the element Cn was more volatile than Hg and unlike radon, reveals a metallic interaction with the gold surface. These adsorption characteristics establish element Cn as a typical element of group 12.

The results of the study of the chemical properties of Fl are less definite. In the first experiment [49] performed in 2007, three decay chains of ^{287,288}Fl were observed in the reactions of ⁴⁸Ca with ^{242,244}Pu. Their deposition on the detectors with temperature –90, –88 and –4 C indicates that element Fl is at least as volatile as element 112 (noble-gas-like behaviour). In the second work [65] published in 2014, two decay chains were attributed to the ²⁴⁴Pu + ⁴⁸Ca reaction products ^{288,289}Fl. But both events were observed at room temperature (+21 C) indicating volatile-metalic behaviour of Fl.

4.1.2. Synthesis of element 116 livermorium. The first nucleus of element 116 was discovered at the DGFRS on July 19, 2000 [40, 41]. Like in 1999 experiment 244 Pu + 48 Ca, the first study of the 248 Cm + 48 Ca reaction in 2000–2001 was performed at low-energy side of the excitation function [43, 44]. In this series of experiments, three similar decay chains of parent nucleus were observed which were followed by a sequence of two α decays and SF in each case. The decay properties of these descendants were in full agreement with those measured in 1999 in the 244 Pu + 48 Ca reaction [39]. After measuring the excitation function of the 244 Pu + 48 Ca reaction [42], ERs observed in the 248 Cm + 48 Ca reaction were identified as the product of the 3 n-evaporation channel, 293 Lv.

Investigation of this reaction was continued in 2004 at higher ⁴⁸Ca energy [43, 44]. Here, like in the case of the ²⁴⁴Pu + ⁴⁸Ca reaction, increase of the excitation energy allowed us to

observe simultaneously two isotopes: 293 Lv and a new lighter isotope 292 Lv. If the same type of nuclear reaction occurred in both cases, then after α decay of the parent nuclei in the reaction with 248 Cm the descendant isotopes should be the same as those observed in primary reaction with 244 Pu. Indeed, decay properties of the daughter nuclei in the 248 Cm + 48 Ca reaction were identical to those registered directly in the reaction with 244 Pu [39, 42] (see figures 6 and 7).

Experiments were continued with a ²⁴⁵Cm target. In 2003, at the first (lowest) bombarding energy two new isotopes of element 116 were synthesized [42]. The decay properties of daughter nuclei in one of these were in agreement with those observed in the 244 Pu(48 Ca, 5n) 287 Fl and 242 Pu(48 Ca, 3n) 287 Fl [42–44] as well as in the 238 U(48 Ca, 3n) 283 Cn [44] reactions. The α decay of the second isotope lead to decay chains seen in the 242 Pu(48 Ca, 4 n) 286 Fl [44] and 238 U(48 Ca, 4 n) 282 Cn [44] reactions. This indicates observation of the 2n- and 3n-evaporation channels of the ²⁴⁵Cm + ⁴⁸Ca reaction, i.e. ²⁹¹Lv and ²⁹⁰Lv, respectively (see figures 6 and 7). In 2005, further increase of ⁴⁸Ca energy resulted in a reduction in the yield of ²⁹¹Lv and increase of that of ²⁹⁰Lv [45]. Finally, at the highest energy, only 3n-evaporation channel was observed. Such variation of production cross sections of two isotopes is in agreement with what it should be expected for the behaviour of the xn channels of the complete-fusion reactions. The decrease of the neutron number in the target nuclei, e.g. from ²⁴⁴Pu to $^{242}\mathrm{Pu}$ or from $^{248}\mathrm{Cm}$ to $^{245}\mathrm{Cm}$, results in rise of the neutron binding energy and somewhat lower excitation energy of CN at the fusion barrier (figure 7). This leads to a relative increase in the yields of lower xn channels. Note, the 2n-evaporation channel was also observed in two other reactions with somewhat lower-N target nuclei. A single decay chain was assigned to the 2*n*-reaction product in the experiment with ²⁴²Pu [44] (figure 7) and four chains were observed in the ²⁴³Am(⁴⁸Ca, $(2n)^{289}$ 115 reaction [66] (see below).

Owing to strong correlations in three α -decay chains of ^{291}Lv with following ^{287}Fl and lighter descendants (even up to ^{267}Rf in one chain), the IUPAC/IUPAP JWP recommended that the Dubna–Livermore collaboration be credited with discovery of element 116 [63]. The element with atomic number 116 was named livermorium (Lv) in honour of the Lawrence Livermore National Laboratory because of experiments on the synthesis of super-heavy elements, including element 116, were performed in collaboration with group of researchers from this laboratory [64].

The independent confirmation of the results obtained at the DGFRS in 2000, 2001 and 2004 in the $^{248}\text{Cm} + ^{48}\text{Ca}$ reaction [40, 41, 43, 44], where element 116 was observed for the first time, followed in 2007 in the SHIP experiments [51]. Four and one decay chains of ^{292}Lv and ^{293}Lv , respectively, were registered at the excitation energy of 40.9 MeV (see figures 6 and 7). The decay properties of the parent and all the descendant nuclei are in agreement with those measured at the DGFRS. One more chain was not definitely assigned. The energy and lifetime of the first α decay agree with data determined for ^{293}Lv but energies of the next three α particles are larger than those measured for ^{289}Fl , ^{285}Cn [39–42] and ^{281}Ds [32, 54] as well as much longer decay time was observed for the terminating

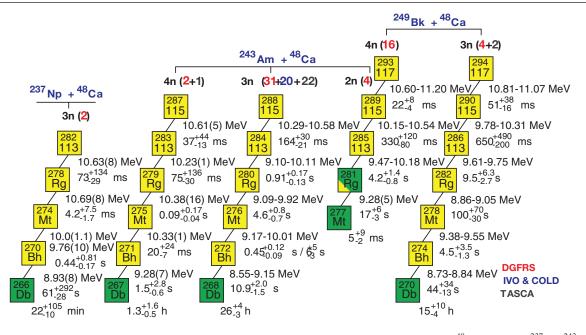


Figure 8. Summary decay properties of the isotopes of odd-Z elements observed among the products of ⁴⁸Ca beam and ²³⁷Np, ²⁴³Am and ²⁴⁹Bk target reactions. The numbers of the decay chains of the given isotopes, the products of corresponding xn-evaporation channels, observed in experiments with use of the DGFRS [26, 27, 34, 36, 66–70] (in red), chemical setup (in blue) [26, 71, 72] and TASCA (in grey) [73, 74] are shown. The average energies or energy intervals of α particles and half-lives are given for all α emitters observed in these experiments (yellow squares). The energy uncertainties given in brackets correspond to the data with the best energy resolution. For spontaneously fissioning nuclei marked by green squares the half-lives are listed.

fission event than that seen at the TASCA experiment [32, 54]. The decay properties of detected nuclei are in good agreement with those observed in long decay chains originating from 291 Lv which could be produced in the 5n-evaporation channel of the reaction with 248 Cm or in the reaction with lighter Cm isotope, e.g. 246 Cm (percentage of 3.1%) (private communication by Hofmann). Indeed, α -particle energies and lifetimes of the second to fourth detected nuclei coincide well with data for 287 Fl, 279 Ds and 275 Hs as well as decay time of SF nucleus is in agreement with half-life for 271 Sg (see figure 6). Somewhat lower α -particle energy for the parent even-odd 291 Lv and missing of one of five decays (283 Cn) are quite possible.

4.1.3. Synthesis of element 118. For the first time, the decay chain of the element 118 was synthesized on March 19, 2002, in the reaction 249 Cf(48 Ca, 294 118 studied at the DGFRS [45]. Two more decay chains of 294 118 were observed in 2005 at higher projectile energy [45]. Finally, the fourth decay chain of the same isotope was registered during two experiments aimed at the synthesis of element 117 in the 249 Bk + 48 Ca reaction performed in 2009–2010 and 2012 [36] 2 . Identification of the isotope 294 118 was based on the results of the four cross reactions: 249 Cf(48 Ca, 290 Lv \rightarrow ... \rightarrow 282 Cn [42, 45], 242 Pu(48 Ca, 4n) 286 Fl \rightarrow 282 Cn [44] and 238 U(48 Ca, 4n) 282 Cn [44] (see figure 6). Among the daughter nuclei in the decay chain of 294 118, decay properties of two isotopes 286 Fl and 282 Cn were

confirmed in independent experiments at the BGS [52, 53] (see subsection 4.1.1. above).

4.2. Odd-Z nuclei

4.2.1. Synthesis and identification of elements 113 and 115. The discoveries of elements 113 and 115 as the products of the complete-fusion reaction ²⁴³Am + ⁴⁸Ca that led to the synthesis of isotopes of element 115 and their α decay product, element 113 that was also unknown at that time, were reported in 2004 [67]. The first decay chain was observed at the DGFRS on 24 July, 2003. This involved two new elements at once, $^{288}115$ and $^{284}113$, followed by α decays of three new neutron-rich isotopes of the known elements ²⁸⁰Rg, ²⁷⁶Mt and ²⁷²Bh and SF of ²⁶⁸Db (see figure 8). The electron-capture (EC) of ²⁶⁸Db leading to presumably rapid SF of 268 Rf ($T_{SF} \sim 1$ s [7]) could not be excluded as well. The run was performed at two projectile energies, which resulted in observation of two isotopes ²⁸⁸115 and ²⁸⁷115 as well as their descendant nuclides down to ²⁶⁸Db and ²⁶⁷Db (figure 8). In the ²⁴³Am + ⁴⁸Ca reaction, the energies of the bombarding particles and reaction cross sections were comparable with results of experiments where excitation functions for the reactions ^{242,244}Pu, ^{245,248}Cm + ⁴⁸Ca have been measured (see figures 7 and 9). Two neighbouring isotopes of the new element were detected at different ⁴⁸Ca energies, in agreement with expectations for the fusion-evaporation reactions. After observing decays of the three nuclei ²⁸⁸115 at the excitation energy $E^* \approx 40 \,\text{MeV}$, with the increase of energy to $E^* \approx 45 \,\mathrm{MeV}$ a new decay chain originating from different isotope was registered [26, 67] (see, e.g. the difference in

 $^{^2}$ In the course of the long-term work, $^{249}\mathrm{Cf}$ —the product of β^- decay of $^{249}\mathrm{Bk}$ (330 d)— is being accumulated in the target. The gradual ingrowth of $^{249}\mathrm{Cf}$ in the $^{249}\mathrm{Bk}$ target material during both series of experiments resulted in production of $^{294}118$ with cross section comparable with values measured for the $^{249}\mathrm{Cf}(^{48}\mathrm{Ca},3n)$ reaction at close excitation energies (see figures 6 and 7).

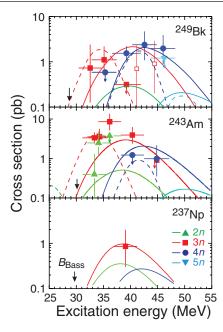


Figure 9. Excitation functions for the 2n (green triangle up), 3n (red square), 4n (blue circle) and 5n (cyan triangle down) evaporation channels from the complete-fusion reactions 237 Np, 243 Am, 249 Bk + 48 Ca measured at the DGFRS [26, 27, 34, 36, 66–70] (solid symbols) and TASCA [74] (open symbols). For reference purposes, the Bass barrier [55] is shown by black arrow in each panel; in the bottom panel it is labeled with $B_{\rm Bass}$. Vertical error bars correspond to statistical uncertainties [60] for the DGFRS experiments and available data from TASCA. Horizontal error bars represent the range of excitation energies populated at given beam energy. Symbols with arrows show upper cross-section limits. The results of theoretical calculations are shown by solid [56–59] and dashed [61] lines.

half-lives of the neighbouring ²⁸⁰·Rg and ²⁷⁹Rg, ²⁷⁶Mt – ²⁷⁵Mt and other descendant nuclei in figure 8). The decay properties of SHN will be discussed in the following section but here we emphasize that the decay properties of nuclei observed in the ²⁴³Am + ⁴⁸Ca reaction evidently differ from those produced in the reactions with even-*Z* target nuclei. One can compare figures 6 and 8 where isotopes of odd-*Z* elements show decay properties intermediate between those of neighboring even-*Z* nuclei. Their decay chains are longer which is caused by unpaired protons increasing stability of nuclei against SF by orders of magnitude, etc.

In addition to the above-mentioned measurements, a chemical experiment was performed for identification of the long-living SF isotope 268 Db that was observed after five sequential α decays of 288 115 nuclei [71]. The 243 Am target was bombarded by 48 Ca ions with an energy corresponding to 247 MeV in the middle of the target that means $E^* = 39$ MeV for the CN 291 115 (compare with the DGFRS data in figure 9). On leaving the target, the recoiling nuclei passed through a collimator suppressing the yield of transfer-reaction products (capture angle of $\pm 12.5^{\circ}$) and were stopped in a copper catcher. Each one–two days the front layer of catcher with a thickness exceeding range of ERs was mechanically cut from the surface. This part of the catcher could contain 268 Db ($T_{SF} \sim 1$ d) atoms. Then elements of group 4 and 5 were isolated from actinides [71].

The test experiments [75] have shown that the factor of separation of transactinides from actinides was more than 10^4 . On the other hand, from two possible transactinides with Z =104 and 105, the element of group 4 (Rf) preceded by five α decays could be produced in the ²⁴³Am + ⁴⁸Ca reaction only in the pxn channel or as a result of EC/ β^+ decay of one of elements 115-Bh. However, the pxn channel for x = 1-5 or EC/ β^+ decay of ²⁸⁸115 or ²⁸⁴113 would lead to the now known isotopes of Fl or Cn whose decay chains strongly differ from the observed ones (see figures 6 and 8). The EC/ β ⁺ decay of lighter descendants $^{280}\text{Rg}-^{272}\text{Bh}$ leads to even-even isotopes $^{280}\text{Ds}-$ ²⁷²Sg for which SF is expected with half-lives less than 1 s (see [7] and figure 13 below). Therefore, the ~1-d SF, if observed in transactinides fraction, could originate from the element of group 5(Db) only (direct SF or SF with short half-life following EC). In 2004, in the chemical experiment [71] 15 SF events of the nuclei of a transactinide element were detected. These showed a half-life of 32^{+11}_{-7} h, high total kinetic energy of SF fragments (~235 MeV) and average neutron multiplicity per fission act (4.2) and were produced with a cross section of about 4.2 pb. All the values, within experimental errors, are in agreement with those $(T_{1/2}, \text{TKE}, \sigma_{3n})$ measured for ²⁶⁸Db at the DGFRS in 2003 [67] and later experiments [66, 70] (figures 8 and 9). These results were later (in 2005 [72]) corroborated in another chemical experiment that attempted studying more delicate chemical properties of Db within group 5.

In chemistry experiments [71, 72] the SF activity was produced in the same 243 Am + 48 Ca reaction (I) at the same projectile energy (II) with the *same* decay properties (namely, decay mode (III), half-life (IV), total kinetic energy (V)) and the same cross section (VI) as it was observed in the experiment performed at the DGFRS [67]. All the factors allowed one to conclude that one and the same isotope has been observed in both the physical and chemical experiments [26, 67, 71, 72]. Simultaneously, all the precursors (Z = 107, 109,111, 113 and 115) discovered in [67] were identified by the method of genetic relation between ancestor and descendant [37]. In our further investigation of the region of odd-Z SHN we studied neutron-deficient isotopes and continued, as well, the ²⁴³Am + ⁴⁸Ca experiment in a more extended range of projectile energies for observation of more nuclei and detailed measurement of the excitation function of this reaction. In 2006 in the 237 Np(48 Ca, ^{3}n) reaction, we observed two decay chains originating from the lighter odd-odd isotope ²⁸²113 [68] (figure 8). Decay properties of ²⁸²113 and its descendant nuclei ²⁷⁸Rg, ²⁷⁴Mt, ²⁷⁰Bh and ²⁶⁶Db were in full agreement with those following from decay properties of heavier isotopes ^{283,284}113 and other descendants previously produced in the 243 Am + 48 Ca reaction.

In contrast to even-Z target nuclei, application of the cross-bombardment method for odd-Z nuclei is limited by the number of target nuclei available for experiments, that is 237 Np, 241,243 Am and 249 Bk. The 249 Bk(48 Ca, 3-4n) reactions lead to heavier isotopes of element 115 and their descendants; therefore only the isotope 289 115, the product of the 2n-evaporation channel of the reaction 243 Am + 48 Ca, seems to be useful for cross-bombardment experiments. In 2010–2012, with aim of synthesizing this isotope as well as measuring excitation

function in a wider energy range, we performed a new series of experiments with 243 Am [66, 70]. The results of all these experiments are presented in figures 8 and 9. Indeed, at the two lowest 48 Ca energies, we detected the product of the 2 *n*-reaction channel, 289 115, undergoing two consecutive α decays and terminated mainly by SF of 281 Rg, as it was observed for the same nuclei synthesized in the reaction 249 Bk + 48 Ca (see below). These chains were not detected at higher 48 Ca energies.

In sum with results of 2003, at four energies, 31 decay chains of $^{288}115$, product of the evaporation of three neutrons, with maximum yield at the excitation energy of about 36 MeV, were registered. At the energy $E^* \approx 45$ MeV, we detected two decay chains of $^{287}115$ that were not found at lower 48 Ca energies. Such a behaviour of $\sigma_{xn}(E^*)$ is expected for cooling process of exited CN $^{291}115$ and is in full agreement with numerous similar observations for fusion-evaporation reactions³. The radioactive decay properties of $^{288}115$, $^{287}115$ and their daughter nuclei discovered in 2003 [26, 67] were completely confirmed by registration of 28 new decay chains in the new series of experiments [66, 70].

In four decay chains of $^{288}115$, α decay of 268 Db was searched for within beam-off intervals from 2.7h up to 1–3 d following decay of 272 Bh (see discussion in [34]). No events were found with $E_{\alpha} = 7.7-8.2$ MeV which could be expected for 268 Db. This result is in agreement with chemistry experiments [71, 72] where SF activity was found in the fraction of transactinide elements. Thus, an upper limit of 7% can be set for α -decay branch b_{α} for 268 Db.

In 2012, the same 243 Am + 48 Ca reaction was studied at two close beam energies of 242.1 and 245.0 MeV ($E^* = 35.1$ and 37.5 MeV) at the TASCA [73] applying a high-resolution α , x-ray and γ -ray coincidence spectroscopy technique. In total, thirty correlated α -decay chains were detected which contain five ER- α - α -SF and two ER- α -SF events 'compatible with decay chains proposed to originate from either ²⁸⁹115 or ²⁸⁸115 [73]'. The excitation function of the reaction was not measured in this experiment; short decay chains and reasons for doubts about their assignment are not given in [73]. One long chain of five α decays was clearly compatible with the characteristics of the decay chain attributed to start from the isotope ²⁸⁷115 [66, 67]. The remaining 22 chains are compatible with the 31 chains previously assigned to the decay of $^{288}115$ [26, 66, 67, 70]. Here α decay of 268 Db was not observed as well. An upper limit $b_{\alpha} \le 4\%$ follows from results of all these experiments. The cross-section values are not given in [73]. The summary decay properties of ^{287,288}115 are shown in figure 8.

In both experiments at the DGFRS [26, 66, 67, 70] and TASCA [73], complex spectra of the α -particle energy were measured (see figure 12 below). In decay of 276 Mt, that shows the most complex α -particle spectrum, we could not exclude observation of two states with different lifetimes [66]. Despite the single half-life for 276 Mt proposed in [73], its decay time

in one chain was $t = 8.95 \,\mathrm{s}$ whereas the average lifetime for other 15 events was $\tau = 0.55 \,\mathrm{s}$ (probability to decay with $t \ge 8.95 \,\mathrm{s}$ for 16 events with $\tau = 0.55 \,\mathrm{s}$ is only 1.5×10^{-6}). Thus, this new long decay time just could confirm the assumption of [66]. In figure 8 we present result of a two-exponential fit [76] of all the available data for $^{276}\mathrm{Mt}$ which suggests two half-lives. However, more statistics is still needed for definite conclusion.

In addition to studying the formation and radioactive properties of the products of the 243 Am + 48 Ca reaction, the TASCA experiment [73] was aimed at the measuring x- and γ -rays in coincidence with α particles of nuclei starting from element 115. In one decay chain the escape event with $E=0.825\,\text{MeV}$ was 'firmly attributed' to 276 Mt (probability of its random origin is not given in [73]) and was coincident with two Ge-detector entries at 136 and 167 keV, which are consistent with $Z=107\,$ K $_{\alpha 2}$ and $Z=107\,$ K $_{\alpha 3}$ and $Z=107\,$ Recause of 10–15% probability of random $Z=107\,$ Recause of 10–15% compton scattered or background events. Besides, several $Z=107\,$ Recoincidences were observed for $Z=107\,$ Re and $Z=107\,$ Recoincidences were observed for $Z=107\,$ Re and $Z=107\,$ Recoincidences were observed for $Z=107\,$ Recause of $Z=107\,$ Recau

In spite of the fact that registration of x-rays in coincidence with α particles of $^{276}\mathrm{Mt}$ was not definitely established, the experiment demonstrated the prospects for investigation of nuclear structure of SHN. However, these studies call for considerable increase of production rate of nuclei which could be reached at the new experimental facilities which are planned to be put into operation in a few years (see section 5).

4.2.2. Synthesis of element 117. The synthesis of heavier element 117 became feasible when 22.2 mg of the 249 Bk target material was produced at the High Flux Isotope Reactor (HFIR) at the Oak Ridge National Laboratory (ORNL) [27] and sent to Dubna. For the first time, element 117 was observed at the DGFRS on August 20, 2009, in the complete-fusion reaction 249 Bk(48 Ca, 49 Ca

In 2009-2010, at two projectile energies corresponding to the excitation energies of the CN ²⁹⁷117 of 39 and 35 MeV, we synthesized two isotopes of element 117 [27, 69] (see figures 8 and 9). At the excitation energy of 39 MeV that corresponds to the expected maximum for production yield of the 4n-evaporation channel we have registered five decay chains of the odd-even isotope ²⁹³117. From the well established behavior of the excitation functions measured for numerous reactions, it followed that a reduction of the projectile energy should result in a decrease of the cross section for the 4n channel and increase of the cross section of a heavier odd-odd isotope with lower α -particle energy and longer lifetime. Indeed, at 35 MeV excitation energy, we produced one longer decay chain of the isotope ²⁹⁴117. As was expected for an odd-odd nucleus, fission of ²⁸²Rg and ²⁷⁸Mt is suppressed because of the unpaired neutrons compared to α emission and gives a longer α chain.

Identification of isotopes of the new element 117 was made similarly to that of the isotopes with Z = 115 produced in the ²⁴³Am(⁴⁸Ca, 2-4n)²⁸⁷⁻²⁸⁹115 reaction. Neutron-rich ²⁹⁰115 and ²⁸⁹115 that are daughters of α decay of the isotopes of element 117, produced in the ²⁴⁹Bk(⁴⁸Ca,3-4n)^{294,293}117 reaction should have lower α -particle energies and respectively, longer lifetimes

 $^{^3}$ The products of the 2n-evaporation channel of the 242 Pu, 245 Cm + 48 Ca reactions, isotopes 288 Fl and 291 Lv, were observed in our experiments with comparable cross sections at $E^* = 32 - 38$ MeV [42–45] (figure 7).

compared with the isotopes $^{288}115$ and $^{287}115$ produced in the reaction with 243 Am. Indeed, α -decay energies of all the descendant nuclei $^{289,290}115$, $^{285,286}113$, 282 Rg, 278 Mt and 274 Bh (products of the 249 Bk + 48 Ca reaction) have smaller E_{α} and longer T_{α} when compared with neighboring isotopes $^{287,288}115$, $^{282-284}113$, $^{278-280}$ Rg, $^{274-276}$ Mt and $^{270-272}$ Bh observed in the reactions with 243 Am and 237 Np (see figures 8 and 12 below). Moreover, analogously to lighter isotopes of odd-Z nuclei with $Z \le 111$ and $N \le 163$, the α -decay energies of all the products of the reaction with 249 Bk including parents $^{293}117$ and $^{294}117$ have intermediate values between those measured for neighboring even-Z nuclei (see figures 6, 8, 11 and 12 below).

The number of consecutive α decays originating from $^{289}115$ and $^{288}115$ differ. Decay chains of $^{289}115$ are terminated mainly by SF of 281 Rg but all 31 [26, 66, 67, 70] and 22 [73] observed decay chains of $^{288}115$ ends in SF of 268 Db (figure 8). Furthermore, in spite of complex α -particle spectra of odd-Z nuclei, decay properties of $^{289}115$ and $^{288}115$ and descendant nuclei are different (compare T_{α} values for nuclei in figure 8 and shape of α -particle spectra in figure 12 below, especially for isotopes of element 113). At the same time, α -particle energies, decay times and decay modes of isotopes $^{289}115$, $^{285}113$, and 281 Rg observed in the reactions with 243 Am and 249 Bk agree. Therefore, one can conclude that isotope $^{289}115$ was produced in cross reactions with two target nuclei to provide cross bombardment evidence for the discovery of the new elements 113,115 and 117.

These conclusions were confirmed also by the following experiments aimed at the measurement of excitation functions for isotopes of element 117 [34, 36]. In 2012 the ²⁴⁹Bk + ⁴⁸Ca reaction was studied at five excitation energies within interval $E^* = 30.4$ –48.3 MeV. In two campaigns with ²⁴⁹Bk target four decay chains of ²⁹⁴117 were produced at two laboratory-frame beam energies $E_{\text{lab}} = 244$ and 247 MeV ($E^* = 32.6$ and 35.1 MeV) and 16 chains of ²⁹³117 were observed at three higher energies $E_{\text{lab}} = 252$, 256 and 260 MeV ($E^* = 39.3$, 42.6 and 46.0 MeV) (see figures 8 and 9) providing additional evidence of the identification of the nuclei of element 117.

The same reaction was studied in 2012 at the TASCA at 48 Ca energies of 252, 254 and 258 MeV [74]. In total, four decay chains, two long and two shorter ones (not published yet), all terminated by SF, were observed. The decay properties of the nuclei in the long chains registered at the two largest 48 Ca energies and assigned to 294 117 are in good agreement with results reported from the DGFRS group. In addition, in both chains events with E_{α} = 7.89 and 7.90 MeV were found 1.3 h and 1.6 h after α decay of 274 Bh and assigned to 270 Db (T_{α} = 1h) followed by SF of 266 Lr ($T_{\rm SF}$ = 11h). This was not observed in experiments at the DGFRS and needs a short comment.

Consecutive α decays of nuclei ²⁸⁸115–²⁷²Bh or ²⁹⁴117–²⁷⁴Bh in all the observed decay chains occurred within 2 and 7 min, respectively. The probability of random observation of any of daughter nuclei with $E_{\alpha} = 8.5$ –10.5 MeV (see figures 5 and 12 below) within beam-off interval of 10 min at the DGFRS was about 2.5 × 10⁻³ (at $E_{\text{lab}} = 243$ MeV in [66, 70]) and 2.8 × 10⁻⁴ (at $E_{\text{lab}} = 247$ MeV in [34, 36]). But the situation differs at the end of decay chains where the time interval between the last α particle and SF increases up to hours and

tens of hours. The probability of the appearance of a random α particle within this interval increases accordingly.

However, the probability of the random origin P_{err} of events within some energy range (e.g. $E_{\alpha} = 7.7-8.2 \,\text{MeV}$ in [34]) and relatively long time interval, say 2h, was not presented in [74]. The given total numbers of beam-off α particles with $E_{\alpha} = 6-12 \,\text{MeV}$ in the detector pixels where decay chains were observed [74] results in a rather large $P_{\rm err}$ value of 0.5. Furthermore, one could expect similar decay properties for 270 Db (N = 165) and 268 Db (N = 163) which follows from close predicted Q_{α} values (8.31 and 8.26 MeV, respectively [78]) as well as from comparable α -decay half-lives of other N = 163, 165 isotopes ^{269,271}Sg and ^{270,272}Bh (see figures 6 and 8 and table 1 below). But, as mentioned above, α decay of ²⁶⁸Db was not seen in the DGFRS [26, 66, 67, 70], TASCA [73] and chemistry experiments [71, 72]. Thus, the partial T_{α} value for ²⁶⁸Db exceeding that for ²⁷⁰Db, suggested in [74], by more than two orders of magnitude (≥300 h) seems to be very unlikely. That is why we do not include results for ²⁷⁰Db in figure 8 and below until finishing detailed analysis of the data ([36] in [74]).

4.3. Attempts to produce Z = 119 and 120 nuclei

By now, ten reactions of actinide target nuclei ^{233,238}U, ²³⁷Np, ^{242,244}Pu, ²⁴³Am, ^{245,248}Cm, ²⁴⁹Bk and ²⁴⁹Cf with ⁴⁸Ca were used for investigation of the region of SHN. In the ²³³U + ⁴⁸Ca reaction no decay chains were observed with an upper cross section limit of 0.6 pb [44]. Nine other reactions resulted in synthesis of 25 even-Z and 29 odd-Z nuclei.

The decay properties of these nuclei revealed a significant increase in their stability as they approached the predicted neutron shell N = 184. The nuclides with the largest neutron and proton numbers that were synthesized in reactions with the heaviest target nuclei ²⁴⁸Cm, ²⁴⁹Bk and ²⁴⁹Cf that are 293 Lv (N = 177), 294 117 (N = 177) and 294 118 (N = 176), respectively, still possess 7–8 fewer neutrons than the predicted magic number N = 184. One can expect that increasing the number of neutrons in these nuclei would result in further increase of their stability. Unfortunately, nuclides with Z > 98 do not exist in quantities sufficient for producing targets for these types of experiments. Therefore, isotopes with larger neutron excess can be reached only if they are formed as heavier ERs. For this purpose, one needs to use completefusion reactions with projectiles heavier than ⁴⁸Ca. One should also note that increasing the atomic number brings us closer to the closed proton shell at Z = 120-126 predicted by some microscopic models, which could also increase shell effects. However, most calculations predict much lower cross sections for complete-fusion reactions with projectiles heavier than ⁴⁸Ca [59, 61, 62, 79–83].

In 2007 the 244 Pu(58 Fe, xn) $^{302-x}$ 120 reaction was studied at the DGFRS [84]. No decay chains consistent with fusion-evaporation reaction products were observed during an irradiation with a beam dose of 0.7×10^{19} 330 MeV 58 Fe projectiles. The sensitivity of the experiment corresponds to a cross section of 0.4 pb for the detecting a single decay; the upper cross-section limit was set at 1.1 pb. In 2007–2008 a more symmetric reaction 238 U + 64 Ni leading to the same CN was used at the SHIP [85].

Table 1. Decay	properties of nuclei.
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	Table 1. Decay properties of nuclei.								
\overline{Z}	N	A	No. observed ^a	Decay mode, branch b	Half-life ^c	E _α (MeV)	$Q_{\alpha}^{\text{exp}} (\text{MeV})$	$Q_{\alpha}^{\text{th}} (\text{MeV})$	
118	176	294	4 (4/4)	α	0.69 ^{+0.64} ms	11.66 ± 0.06	11.82 ± 0.06	12.11	
117	177	294	5 (5/5)	α	51 ⁺³⁸ ms	10.81-11.07	11.18 ± 0.04	11.43	
	176	293	15 (15/15)	α	22^{+8}_{-4} ms	10.60-11.20	11.32 ± 0.05	11.53	
116	177	293	5 (5/5)	α	57 ⁺⁴³ ms	10.56 ± 0.02	10.71 ± 0.02	11.09	
	176	292	9 (8/7)	α	13^{+7}_{-4} ms	10.63 ± 0.02	10.78 ± 0.02	11.06	
	175	291	4 (4/4)	α	19 ₋₄ ms 19 ⁺¹⁷ ms	10.74 ± 0.07	10.89 ± 0.07	10.91	
			, ,		1 > -6 1113	10.50 ± 0.02			
	174	290	11 (11/11)	α	$8.3^{+3.5}_{-1.9}$ ms	10.85 ± 0.07	11.00 ± 0.07	11.08	
115	175	290	6 (5/6)	α	650 ⁺⁴⁹⁰ ₋₂₀₀ ms	9.78-10.31	10.41 ± 0.04	10.65	
	174	289	16 (15/16)	α	330^{+120}_{-80} ms	10.15-10.54	10.49 ± 0.05	10.74	
	173	288	46 (41/46)	α	164^{+30}_{-21} ms	10.29-10.58	10.63 ± 0.01	10.95	
	172	287	3 (3/3)	α	37^{+44}_{-13} ms	10.61 ± 0.05	10.76 ± 0.05	11.21	
114	175	289	16 (14/16)	α	1.9 ^{+0.7} _{-0.4} s	9.84 ± 0.02	9.98 ± 0.02	10.04	
			, ,		-0.4	9.48 ± 0.08			
	174	288	35 (31/30)	α	$0.66^{+0.14}_{-0.10}$ s	9.93 ± 0.03	10.07 ± 0.03	10.32	
	173	287	19 (18/17)	α	$0.48^{+0.14}_{-0.09}$ s	10.03 ± 0.02	10.17 ± 0.02	10.56	
	172	286	27 (22/14)	α :0.6 SF:0.4	$0.12^{+0.04}_{-0.02}$ s	10.21 ± 0.04	10.35 ± 0.04	10.86	
	171	285	1 (1/0)	α	$0.13^{+0.60}_{-0.06}$ s			11.11	
113	173	286	6 (5/6)	α	9.5 ^{+6.3} _{-2.7} s	9.61–9.75	9.79 ± 0.05	9.98	
	172	285	17 (17/17)	α	$4.2^{+1.4}_{-0.8}$ s	9.47-10.18	10.01 ± 0.04	10.21	
	171	284	47 (39/47)	α	$0.91^{+0.17}_{-0.13}$ s	9.10-10.11	10.12 ± 0.01	10.68	
	170	283	2 (2/2)	α	75^{+136}_{-30} ms	10.23 ± 0.01	10.38 ± 0.01	11.12	
	169	282	2(2/2)	α	73^{+134}_{-29} ms	10.63 ± 0.08	10.78 ± 0.08	11.47	
112	173	285	17 (16/16)	α	28 ⁺⁹ ₋₆ s	9.19 ± 0.02	9.32 ± 0.02	9.49	
	172	284	37 (34/-)	SF	98^{+20}_{-14} ms			9.76	
	171	283	33 (23/31)	α :1. SF: ≤ 0.1	$4.2^{+1.1}_{-0.7}$ s	9.53 ± 0.02	9.66 ± 0.02	10.16	
					-0.7	9.33 ± 0.06			
	170	202	14 (14/)	(OE)	10.22	8.94 ± 0.07		10.60	
	170	282	1 (1/1)	SF	0.91 ^{+0.33} _{-0.19} ms	10.21 + 0.04	10.46 + 0.04	11.21	
	169	281	1 (1/1)	α	$0.10^{+0.46}_{-0.05}$ s	10.31 ± 0.04	10.46 ± 0.04	11.21	
111	171	282	6 (6/6)	α	100^{+70}_{-30} s	8.86–9.05	9.16 ± 0.03	9.85	
	170	281	20 (17/2)	α:0.1 SF:0.9	17^{+6}_{-3} s	9.28 ± 0.05	9.41 ± 0.05	10.48	
	169	280	45 (41/42)	α	$4.6^{+0.8}_{-0.7}$ s	9.09-9.92	9.91 ± 0.01	10.77	
	168	279	3 (2/2)	α	90^{+170}_{-40} ms	10.38 ± 0.16	10.53 ± 0.16	11.08	
	167	278	2 (2/2)	α	4.2 ^{+7.5} _{-1.7} ms	10.69 ± 0.08	10.85 ± 0.08	11.30	
110	171	281	17 (17/1)	α:0.07 SF:0.93	$12.7^{+4.0}_{-2.5}$ s	8.73 ± 0.03	8.85 ± 0.03	9.30	
	169	279	36 (31/4)	α:0.1 SF:0.9	$0.21^{+0.04}_{-0.04}$ s	9.71 ± 0.02	9.85 ± 0.02	10.24	
	167	277	1 (1/1)	α	$0.006^{+0.027}_{-0.003}$ s	10.57 ± 0.04	10.72 ± 0.04	10.79	
109	169	278	5 (5/5)	α	4.5 ^{+3.5} _{-1.3} s	9.38–9.55	9.58 ± 0.03	9.55	
	168	277	2 (2/-)	SF	5^{+9}_{-2} ms			9.84	
	167	276	43 (43/41)	α	$0.45^{+0.12}_{-0.09}$ s	9.17-10.01	10.03 ± 0.01	10.09	
					6+5s				
	166	275	3 (3/3)	α	20^{+24}_{-7} ms	10.33 ± 0.01	10.48 ± 0.01	10.34	
	165	274	2 (2/2)	α	440 ⁺⁸¹⁰ ₋₁₇₀ ms	10.0 ± 1.1	10.2 ± 1.1	10.63	
						9.76 ± 0.10			
108	169	277	1 (1/1)	SF	3+15ms			9.03	
108					-	0.21 + 0.02	0.45 + 0.02	0.41	
108	167	275	4 (4/4)	α	$0.20^{+0.18}_{-0.06}$ s	9.31 ± 0.02	9.45 ± 0.02	9.41	

(Continued)

	Table 1. (Continued)								
\overline{Z}	N	A	No. observed a	Decay mode, branch b	Half-life ^c	E_{α} (MeV)	$Q_{\alpha}^{\text{exp}} (\text{MeV})$	$Q_{\alpha}^{\text{th}} (\text{MeV})$	
107	167	274	6 (5/6)	α	44 ⁺³⁴ ₋₁₃ s	8.73-8.84	8.94 ± 0.03	8.83	
	165	272	44 (39/44)	α	$10.9^{+2.0}_{-1.5}$ s	8.55-9.15	9.18 ± 0.01	9.08	
	164	271	2 (2/2)	α	$1.5^{+2.8}_{-0.6}$ s	9.28 ± 0.07	9.42 ± 0.07	9.07	
	163	270	1 (1/1)	α	61^{+292}_{-28} s	8.93 ± 0.08	9.06 ± 0.08	8.63	
106	165	271	4 (4/2)	α:0.6 SF:0.4	1.6 ^{+1.5} min	8.54 ± 0.08	8.67 ± 0.08	8.71	
	163	269	1 (1/1)	α	2^{+10}_{-1} min	8.57 ± 0.10	8.70 ± 0.10	8.32	
105	165	270	6 (6/-)	SF e)	15 ⁺¹⁰ ₋₄ h			8.11	
	163	268	66 (66/-)	SF ^{e)}	$26^{+4}_{-3}h^{d}$			7.80	
	162	267	3 (3/-)	SF ^{e)}	$1.3^{+1.6}_{-0.5}$ h			7.41	
	161	266	1 (1/-)	SF e)	22^{+105}_{-10} min			7.52	
104	163	267	2 (2/-)	SF	1.3 ^{+2.3} _{-0.5} h				
	161	265	1 (1/-)	SF	2+8min			7.27	

 2^{+8}_{-1} min

No events originating from isotopes of element 120 were observed. Measured upper cross-section limit of 0.09 pb was set at a mean excitation energy of 36.4 MeV and beam dose of 2.1×10^{19} . A cross-section limit of 0.56 pb was set in the 248 Cm + 54 Cr reaction at the SHIP in 2011 [86].

Two experiments with 50Ti beam and target nuclei 249Cf and ²⁴⁹Bk were performed at the TASCA in 2011 and 2012. These were aimed at the synthesis of elements 120 and 119, respectively [87] (The results have not yet been published).

5. Discussion

5.1. Production cross sections

The excitation functions $\sigma_{xn}(E^*)$ of the reactions leading to production of isotopes of elements 112-118 shown in figures 7 and 9 are typical for the process of de-excitation of CN via evaporation of neutrons. The maximum cross-section values correspond to evaporation of three to four neutrons depending on neutron excess in the CN. In comparison with these reaction channels, the evaporation of two neutrons observed in the reactions with ²⁴²Pu, ²⁴³Am and ²⁴⁵Cm is suppressed by shift of maximum of cross section σ_{2n} below the Coulomb barrier ($E^* < E_{\min}$). One should note that different orientations of the deformed target nucleus at the touching point with spherical nucleus ⁴⁸Ca change location and shape of the Coulomb barrier with regard to the Bass barrier calculated for spherical nuclei. As it follows from measured cross sections $\sigma_{xn}(E^*)$ at the lowest excitation energies E^* , in the reactions Act + 48 Ca the CN is formed mainly in 'equatorial' collisions, characterized by minimum distances between the centers of the interacting nuclei, which leads to shift of the cross-section maxima by several MeV to higher values than the corresponding Bass barriers (see figures 7, 9 [56–59]).

With movement of the Coulomb barrier to higher energies $E^* > 50 \,\mathrm{MeV}$, one can expect a decrease of $\sigma_{xn}(E^*)$ because of ascending competition from fission of CN. Indeed, in the reactions of 48 Ca with 244 Pu and 242 Pu at $E^* = 53$ and $50 \,\text{MeV}$, respectively (see figure 7), the only single decay chains of ²⁸⁷Fl and ²⁸⁵Fl, products of the 5*n*-evaporation channel, could be observed. Thus, all the ERs in the Act + ⁴⁸Ca reactions were registered within relatively narrow excitation-energy interval $29 \le E^* \le 53 \,\text{MeV}$.

In this energy range, the total fusion-evaporation cross sections $\sigma_{\text{tot}} = \sum_{x} \sigma_{xn}(E^*)$ reach maxima at $E^* \approx 40 \,\text{MeV}$ (hot fusion); transition of nucleus to the ground state occurs via evaporation of 3-4 neutrons. This energy is about three times larger than excitation energy of nuclei at the cross-section maxima in the cold-fusion reactions used for the synthesis of elements 110-113. The maximum total ER cross sections measured in the experiments on the synthesis of elements with $Z \ge 102$ in cold fusion and in ⁴⁸Ca-induced reactions are shown in figure 10.

In cold-fusion reactions, as it follows from the figure 10(a), the production cross section drops by about 8 orders of magnitude when Z_{CN} increases from 102 to 113. Such an effect, as a result of growth of the Coulomb factor $k = Z_1 \cdot Z_2 / (A_1^{1/3} + A_2^{1/3})$ by 44% (figure 10(c)), is associated with potential energy surface of the colliding system which causes the hindrance of the formation of CN with stronger Coulomb interaction.

One can note that in theory, the dynamics of the CN formation in competition with the quasi-fission process is the most vague reaction stage. For calculation of the probability $P_{\rm fus}$, different models are used which suppose different mechanisms of CN formation. In the multidimensional Langevintype dynamical approach [56–59, 79], it is assumed that two touching nuclei lose their 'individualities' at the touching point and one strongly deformed heavy nucleus with summary mass evaluates in the multidimensional space of deformations

^a Number of observed decays and number of events used for calculations of half-lives / α -particle energies, respectively.

^b Branching ratio is not shown if only one decay mode was observed.

^c Error bars correspond to 68%-confidence level.

d The value obtained combining the results of physical and chemical experiments.

^e The SF mode was observed but EC/ β ⁺ or α decay is not excluded.

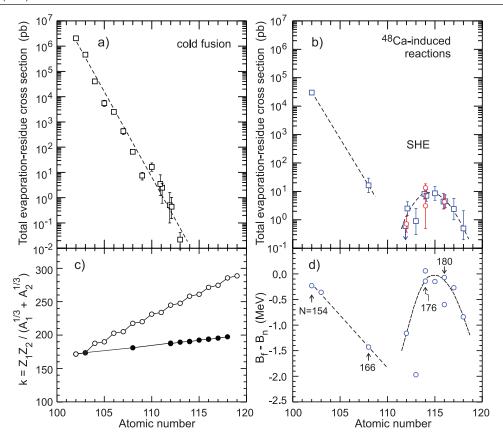


Figure 10. Maximum cross sections of the production of the isotopes of the heavy elements in (a) cold fusion reactions: 208 Pb, 209 Bi + 48 Ca, 50 Ti, 54 Cr, ... 70 Zn ($E^* = 12-20$ MeV) and (b) hot fusion reactions: 208 Pb, 226 Ra, 233,238 U, 242,244 Pu, 243 Am, 245,248 Cm, 249 Bk and 249 Cf + 48 Ca ($E^* = 35-40$ MeV) (data from DGFRS and other separators are in blue and red, respectively). In plot (c), Coulomb factors $Z_1 \cdot Z_2 / (A_1^{1/3} + A_2^{1/3})$ for nuclei in the cold (open circle) and 48 Ca-induced (closed circle) reactions are shown. (d) Difference of fission barrier heights (involving non-axial shapes) and neutron binding energies of the CN in 48 Ca-induced reactions calculated in macroscopic—microscopic nuclear model [88–90] and corrected for the odd-even effect are shown. Arrows show number of neutrons in the CN with the given atomic number.

into a spherical CN or goes into fission channels. A somewhat different scenario of this process is assumed in the 'dinuclear system model' [81, 82]. The dinuclear system stays in contact configuration and undergoes successive transfer of all nucleons from the lighter nucleus to the heavier partner in competition with the quasi-fission processes. Here two touching nuclei keep their relative distance and their 'individuality'. In still another approach, the extended versions of the 'fusion-by-diffusion' model [61, 83], the stochastic process of shape fluctuations leads to the overcoming of the saddle point. Several analytical formulas for description of the fusion probability are proposed [59, 62, 80]. This reaction stage plays a decisive role in cold-fusion reaction.

On the contrary, in more asymmetric reactions of ⁴⁸Ca with actinide nuclei the Coulomb repulsion is less. For super-heavy nuclei with Z = 112-118 the Coulomb factor changes by 6.5% only. Because the initial states of the CN $Z_{\rm CN} = 112-118$ are similar, this allows a uniform description of their transition to ground state via emission of neutrons and γ -rays. The calculated survivability of the CN, which depends on the thermodynamic characteristics of the heated nuclei in the course of their cooling down via emission of neutron(s) and on fission barriers, should correlate with ER cross sections as obtained in the experiment.

In figure 10(b), the total cross section $\sigma_{\text{tot}} = \sum_{x} \sigma_{xn}(E^*)$ measured in the experiments in all the reactions of fusion of ⁴⁸Ca with the target nuclei of Pb, Ra and actinide targets U-Cf are shown. The calculated values of $(B_f - B_n)$ are shown in figure 10(d).

Comparing these, one can see that the relatively high cross sections for production of ERs in hot fusion reactions with 48 Ca are connected with high survivability of the heated CN. This provides direct evidence of the presence of the high fission barriers in nuclei with Z < 120 which appears due to nuclear shell effects.

5.2. Decay properties

5.2.1. Alpha decay. The energies of α particles for even-Z nuclei 281 Cn, 294 118, 291 Lv, 292 Lv, 293 Lv and their descendants registered by the focal-plane detector only or together with the side one at the DGFRS [39–45] are shown in figure 11. Here we also show results of experiments carried out with the use of other facilities (see figure 6): chemical setup IVO + COLD [46–49] and separators SHIP [50, 51], BGS [52, 53] and TASCA [32, 54].

In agreement with expectations, the α decays of the eveneven nuclei 294 118, 290,292 Lv and 286,288 Fl are terminated by SF

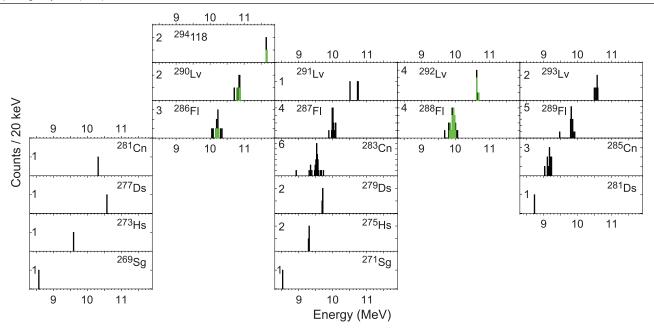


Figure 11. α -particle energy spectra for even-*Z* nuclei registered by the focal-plane detector only or together with the side one at the DGFRS [39–45], IVO + COLD [46–49], SHIP [50, 51], BGS [52, 53] and TASCA [32, 54]. Note, the energy resolution of *α* particles ΔE_{α} detected simultaneously by the focal-plane and side detectors was up to 0.2 MeV (spectra for the events with energy resolution better than 0.1 MeV shown in green). The data from the IVO + COLD are included if ΔE_{α} are published.

of short-lived isotopes 282,284 Cn (see figures 6, 11). The isotope 286 Fl has an SF branch of 40^{+11}_{-10} % (68% confidence level). The α -particle energy spectra of these nuclei are characterized mainly by one α line, which corresponds to the ground-state to ground-state transitions as the most probable for even—even nuclei. In figure 11 the spectra of events registered solely by the focal-plane detectors with energy resolution better than 0.1 MeV are shown by green histograms.

The decay chains originated from the even-odd nuclei ^{291,293}Lv are terminated mainly at later stage, by SF of $^{279,281}\mathrm{Ds}$ which have small $\alpha\text{-decay}$ branch of 11^{+6}_{-4} % and 7^{+9}_{-5} %, respectively. Two SF events of four observed in the ²³⁸U + ⁴⁸Ca reaction at the SHIP [50] were assigned to 50% SF branch of ²⁸³Cn that seemingly does not contradict results observed at the DGFRS in the same reaction [44]. Here, the three decay chains of seven were detected as ER-SF sequences with presumably missing α particles of ²⁸³Cn. However, when 283 Cn is produced after α decay of the parent nucleus 287 Fl [42-45, 52], only the upper limit of 7% for the SF branch of ²⁸³Cn can be derived. It seems that the population of isomeric and ground states, even with comparable lifetimes, in direct reaction and after α decay cannot be excluded (compare with $^{261}\mathrm{Rf}$ ([91] and references therein). Further α decay of the even-odd nuclei leads to SF of ^{265,267}Rf and ²⁷⁷Hs (SF branch for ²⁷¹Sg is not excluded with probability of 40%).

The α -particle energy spectra of the even–odd isotopes are broader. The complex spectra are clearly seen for 283 Cn, 289 Fl and 291 Lv. In addition to the main α line at E_{α} = 9.53 MeV, two lines with lower energies of 9.33 and 8.94 MeV were registered for 283 Cn [42–47] whereas a spectrum of 287 Fl is consistent with a single α transition. The hindrance factors (HF) of 1.5 $^{+0.5}_{-0.3}$, 0.3 $^{+1.5}_{-0.14}$ and 2.9 $^{+1.7}_{-1.6}$, respectively, for three energies of 283 Cn can be estimated as a ratio of partial half-lives which

are determined by numbers of observed events and half-lives expected for Z=112 nucleus with measured α -particle energies from, e.g. semi-empirical systematics (Viola–Seaborg formula [92]). Such data could be decisive for predictions of the low-lying quasi-particle states of nuclei. Predominantly single-line spectrum is seen for ²⁹³Lv but two different energies were observed for ²⁸⁹Fl with $E_{\alpha}=9.48$ and $9.84\,\text{MeV}$. Both these α decays are unhindered (HF = $1.1\,^{+0.6}_{-0.8}$ and $1.1\,^{+0.5}_{-0.2}$, respectively).

The energy spectra of α particles for odd-Z nuclei ²⁸²113, ^{287,288}115, ^{293,294}117 and their descendants registered at the DGFRS [26, 27, 34, 36, 66–70] and TASCA [73, 74] are shown in figure 12. Even in cases with relatively low statistics, one can see wider energy distributions for these nuclei than those shown in figure 11. The α -decay chains of four parent nuclei end by SF of isotopes ^{266,267,268,270}Db. Only odd–even ²⁸¹Rg undergoes SF with small branch for α decay ($b_{\alpha} = 12^{+9}_{-7}\%$) which is followed by SF of ²⁷⁷Mt (see figure 8).

The decay properties of nuclei produced in the fusion-evaporation reactions with 48 Ca are shown in figures 6 and 8 and table 1. The α -decay energies of nuclei Q_{α}^{th} calculated within macroscopic–microscopic model [88, 89] are given in column 9. For even–even nuclei with Z=114–118 and N=172–176 the values $\Delta Q_{\alpha}=Q_{\alpha}^{\text{exp}}$ – Q_{α}^{th} amount to from –0.5 to –0.1 MeV. Measured in experiments α -decay energies Q_{α}^{exp} are lower and correspondingly, the half-lives T_{α}^{exp} of superheavy nuclei are larger than theoretical values. For even–Z isotopes of elements 106–116 with odd number of neutrons N=165–177 the difference ΔQ_{α} varies within –0.8–0.4 MeV. Finally, for odd-Z nuclei of elements 107–117 the discrepancies are $\Delta Q_{\alpha}=-1.1$ –0.4 MeV in assumption that the measured maximum α -particle energy corresponds to transitions through the ground states of odd-Z nuclei which is not always

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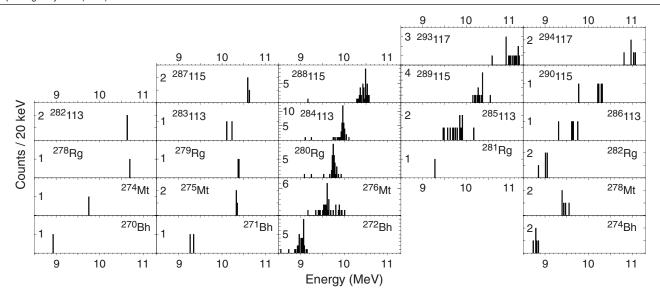


Figure 12. α -particle energy spectra for odd-Z nuclei registered by the focal-plane detector only or together with the side one at the DGFRS [26, 27, 34, 36, 66–70] and TASCA [73, 74].

valid. From comparison of decay properties of 43 α -decaying nuclei produced in the Act + 48 Ca reactions, it follows that their agreement with theoretical predictions is not only qualitative but, to some extent, quantitative. Here one should bear in mind that accuracy of predictions of masses of the known nuclei reaches 0.3–0.4 MeV [89] and 0.4–0.7 MeV for some other approaches (see, e.g. review [22] and references therein). Note, results of Q_{α}^{th} calculations within other MM models and purely microscopic approaches (HFB, RMF) also agree with experimental data ([22] and references therein). Therefore, the hypothesis of existence of super-heavy nuclei received experimental confirmation.

5.2.2. Spontaneous fission. For 16 of the 54 synthesized nuclei SF was observed. For seven even-Z nuclei SF is the predominant mode of decay (see figure 6 and table 1). In five nuclei, SF competes with α decay. As to the odd-Z nuclei, SF was registered for six nuclides (figure 8 and table 1). For the remaining nuclides SF was not observed. The partial SF halflives of even-even isotopes with $N \ge 162$, produced in fusion reactions with ⁴⁸Ca, together with the half-lives of SF nuclides with N < 162, are shown in figure 13. Two isotopes of element 112 with N = 170 and 172 are located in a region, where a steep rise of $T_{SF}(N)$ is expected. Indeed, in the even-even isotopes ²⁸²Cn and ²⁸⁴Cn the difference in two neutrons increases the partial half-life T_{SF} by two orders of magnitude⁴. A similar effect is also observed for the even-even isotopes of element 114. Addition of two neutrons to the nucleus 286 Fl ($T_{SF} \approx 0.3 \text{ s}$) leads to increase of the stability of ²⁸⁸Fl with respect to SF by at least a factor of 15. For even-even isotopes ²⁸⁸Fl, ²⁹⁰Lv, ^{292}Lv and $^{294}118$ SF was not observed.

It is significant that the rise of stability with respect to SF is observed for the heavy nuclei with $Z \ge 110$, which are 10-12 neutrons farther from the closed neutron shell N = 184. On

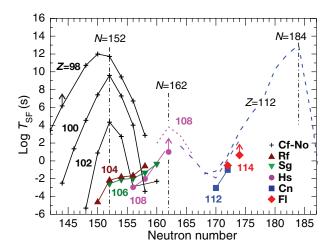


Figure 13. Common logarithm of partial spontaneous fission half-life versus. neutron number for even–even isotopes of elements with Z = 98-114 [32, 42–45, 49, 51–54, 93]. Dashed lines show the theoretical $T_{\rm SF}$ values [7, 8] for even–even Z = 108 (in magenta), 110 (in violet) and 112 (in blue) isotopes.

moving to the nuclei with Z < 110 and N < 170 the probability of SF decreases again when the closed 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.

Because of the high hindrance of SF in the nuclei with odd number of protons (and neutrons) and relatively low T_{α} , the isotopes of elements 113 and 115 produced in the reactions 237 Np + 48 Ca and 243 Am + 48 Ca with N = 169-173 undergo α decay [26, 27, 34, 36, 66–70, 73]. Spontaneous fission is observed only at the end of chain for isotopes of element 105 (or their EC/ α -decay products). For 268 Db α decay seems to be less probable (see above).

In case of the reaction ²⁴⁹Bk(⁴⁸Ca,3-4n)^{294,293}117, the daughter nuclei have one or two extra neutrons compared to the reaction with ²⁴³Am. In analogy with the neighbouring even-*Z* isotopes all the nuclei in the decay chains of ²⁹³117

 $^{^4}$ From measured SF half-lives of the even–even isotopes 282 Cn and 284 Cn it follows that the odd neutron in the 283 Cn nucleus imposes a hindrance to spontaneous fission exceeding 5000.

and $^{294}117$ with Z > 111 and $N \ge 172$ will undergo α decay. The nucleus 281 Rg (N = 170) belonging to the 'critical' region between neutron shells N = 162 and 184 might avoid SF due to the hindrance resulting from an odd proton. The hindrance of the SF in 281 Rg with respect to its even—even neighbuor 282 Cn [42–45, 52, 53] is $\sim 2 \times 10^4$. Despite this, the isotope 281 Rg undergoes SF with a probability of about 90%. Accordingly, even the high hindrance governed by oddness does not 'save' the odd nucleus (281 Rg, 277 Mt) from SF which is caused by the weakening of the stabilizing effect of the neutron shells at N = 162 and N = 184. However an extra neutron and the double effect of oddness favor the α decay in the neighboring isotopes 280 Rg and 282 Rg.

Practically for none of the odd-Z nuclei the branch for EC/ β^+ was observed. For example, isotope ²⁸⁸Fl, EC product of ²⁸⁸115 and its descendants were not seen in the ²⁴³Am + ⁴⁸Ca reaction, which indicates that for nuclei in the decay chain of ²⁸⁸115 the probability for EC/ β^+ is less than 2%. Meanwhile, nuclei at the end of decay chains, neutron-rich odd–odd isotopes ^{266,268,270}Db, could undergo EC decay leading to SF of even–even Rf isotopes for which $T_{\rm SF} = 23 \, {\rm s}$, 1.4 s and 20 ms are predicted [7]. For odd–even isotopes ²⁶⁷Db as well as ²⁸¹Rg values of the observed half-lives are somewhat lower than the bulk of data in the systematics of T_{β} versus Q_{β} for known isotopes of Np-Db (see, e.g. figure 7 in [27]). This could indicate a larger probability of undergoing SF than EC/ $_{\beta}^+$ decay leading to 1 h ²⁶⁷Rf and 13 s ²⁸¹Ds.

Spontaneously fissioning nuclei with Z = 108-112 and N = 168-170 located between N = 162 and N = 184 neutron shells have the lowest $T_{\rm SF}$ values. The α -decay chains of the heaviest nuclei both with even and odd Z, N numbers are terminated here by SF. The decay chains of nuclei outside this region end by SF of neutron-rich isotopes of Rf and Db located closer to the neutron shell at N = 162. Thus, the decay chains of the heaviest nuclei synthesized in the Act + 48 Ca reactions are not connected with a region of known nuclei. They form an isolated region of the heaviest nuclei—some 'island of superheavy nuclei'. The existence of this island and relatively high stability of SHN are determined in full by the new closed shells at N = 184 and Z = 114 (possibly 120–126).

Investigation of SF of SHN is of independent interest. Extensive information about the SF process, obtained for nuclei in the region of actinides (especially for ²⁵²Cf), can be essentially extended for heavier and super-heavy nuclei whose fission barrier is entirely determined by the influence of the new nuclear shells.

6. Perspectives

For the synthesis of super-heavy nuclei with the largest neutron excess N = 175-177, the heavy isotopes ²⁴⁴Pu, ²⁴⁸Cm, ²⁴⁹Bk and ²⁴⁹Cf produced at high flux reactors were used as target nuclei. Experiments aimed at the synthesis of the new isotopes of element 118 in the ^{249–251}Cf + ⁴⁸Ca reactions are being considered at FLNR. As a target material, there will be used the long-lived isotopes of Cf extracted from 'old' ²⁵²Cf neutron sources which were made 30–40 years ago at

the HFIR (ORNL). With such a target of mixed isotopes of californium, in the 3n- and 4n-evaporation channels, the three new isotopes - $^{293}118$, $^{295}118$ and $^{296}118$ —could be produced. Note that 251 Cf is the heaviest nuclide that can be produced at the HFIR in required amounts. For the synthesis of elements with Z > 118, projectiles of heavier than 48 Ca have to be used.

Another direction is to recede from the shell at N = 184and investigate SHN with lower neutron excess that could result in connection of region of known nuclei and 'island of SHN'. For these purposes, light isotopes of actinides, such as ^{233–236}U, ^{239,240}Pu or ²⁴¹Am, can be used in the reactions with 48 Ca which leads to isotopes of Z = 112-115 elements with N = 165-172. However, deficit of neutrons—shift to the edge of island of stability and withdrawal from the shell at N =184—can result in reduce of production cross sections caused by decrease of fission barriers and correspondingly, survivability of CN. This effect was already revealed in the experiments on the synthesis of light isotopes of elements 112-114 in the 233 U, 237 Np and 239 Pu + 48 Ca reactions [44, 68]. Further attempts in this direction, with the existing sensitivity of experiment, does not seem a trivial task. At the same time, discovery of SHN raises many questions concerning the validity of the application of the nuclear shell model and the limits of existence of the heaviest nuclei, about the sizes and nuclear density of the SHN, atomic structure and chemical properties of super-heavy elements. Still an open question is: can the SHN be produced in nucleosynthesis?

Obviously, further investigations of the heaviest nuclei and synthesis of the new elements call for considerable increase in the sensitivity of equipment used in experiments. Our knowledge of the decay properties of SHN and their production cross sections in the ⁴⁸Ca-induced reactions as well as the latest achievements in accelerator and plasma physics, reactor technologies and experimental techniques form the basis for creation of the new laboratory—type of a factory for continuous production of super-heavy nuclei (SHE Factory). Expected yield of SHN can be increased by almost two orders of magnitude compared with the existing level. Since the beginning of 2013, at FLNR (JINR) the construction of the SHE Factory is under way.

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The investigations reported in this review involve numerous experiments performed in Dubna in 1999–2014. This was preceded by the 5 year work aimed at increase of the ⁴⁸Ca beam, further development of methods of preparation of actinide targets, modernization of the DGFRS and its detection system for registration of rare events of production and decay of SHN,

etc. The experiments were carried out in collaboration with groups from US National Laboratories in Livermore and Oak Ridge, RIAR in Dimitrovgrad (Russia), Vanderbilt University in Nashville and University of Tennessee in Knoxville (USA), PSI in Villigen (Switzerland), RIEPh in Sarov (Russia). The investigation of SHN is one of the high-priority directions in the scientific programme of the JINR. During all stages of the experiments, we received support from the Committee of Plenipotentiaries of the Governments of the JINR Member States, JINR Directorate and Scientific Council.

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