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Superheavy elements in D I Mendeleev's Periodic Table

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Abstract. The results on the synthesis of new superheavy elements, synthesized in complete fusion reactions of ^{48}Ca ions with actinide targets, are summarized and analyzed. The perspectives for the synthesis of element 117, as well as of elements with $Z \geqslant 118$ are also considered. The bibliography includes 37 references.

I. Introduction

The Periodic Law and the Periodic Table of chemical elements, proposed by D I Mendeleev about 140 years ago, played a decisive role in the discovery of new stable and radioactive elements. However, the limits of the Periodic Table still remain an open question.

According to the QED-theory, the well-known concept of the atom as a system consisting of a nucleus, carrying almost entirely the atomic mass, and electron orbits lying at a long distance from the charge centre, is valid for very heavy atoms ($Z \sim 170$ or even more). Nevertheless, the limit of existence of atoms (elements) is reached much earlier because of the instability of the nucleus itself.

Only 287 nuclides out of about 2700 hitherto known are found in nature after their nucleosynthesis. The change in the proton-to-neutron ratio generates, as is well known, their radioactive decay. The neutron excess in nuclei leads to a decrease in the neutron separation energy; the limit of existence of neutron-rich nuclei is reached at $E_n = 0$ (the neutron drip line). Similarly, the zero proton separation energy, $E_p = 0$ (the proton drip line), determines the limit of existence of proton-rich nuclei.

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Another limitation is due to the maximum number of nucleons which a nucleus can hold. Formally, the extreme mass of a nucleus in the vicinity of stability, even at the most favourable proton-to-neutron ratio (nuclei in the vicinity of the beta-stability line) is determined by the probability of spontaneous fission (SF). This type of nuclear transformation of heavy nuclides was first observed for the isotope ^{238}U ($T_{SF} = 10^{16}$ years) by Petrzhak and Flerov in 1940.[1] By that time, Hahn and Strassman[2] had already discovered the induced fission of uranium. For the description of this phenomenon Bohr and Wheeler[3] put forward the liquid-drop model of nuclear fission.

This remarkable theoretical model, which is essentially a classic one, is based on the assumption that the nucleus consists of an amorphous (structureless) matter similar to a drop of a charged liquid. The deformation of the drop due to Coulomb forces eventually leads to the fission of the drop into two fragments with approximately equal masses. The fission takes place upon overcoming the potential barrier, which tends to hinder the deformation of the nucleus. For the ^{238}U nucleus, the height of the fission barrier is about 6 MeV. With increasing the nuclear charge Z, the height of the fission barrier rapidly decreases and at some critical value of Z, the nucleus becomes absolutely unstable with respect to spontaneous fission ($T_{SF} \approx 10^{-19}$ s). According to the estimates of Bohr and Wheeler, this critical situation is reached as soon as the charge number reaches values of $Z \geqslant 100$.

The radioactive properties of the transuranium elements synthesized in the 1940-50s by Seaborg and co-workers in neutron capture reactions at high flux nuclear reactors and in nuclear explosions quantitatively confirmed the similarity between nuclear matter and a liquid drop: namely, on going from ^{238}U ($Z = 92$) to ^{257}Fm ($Z = 100$) the probability of spontaneous fission increased by more than 13 orders of magnitude. However, the discovery in the 1960s of actinide isomers that can undergo spontaneous fission (spontaneously-fissioning isomers) and the detection of considerable variations in the partial spontaneous fission half-lives when changing the neutron number, which are most pronounced in the isotopes of Cf, Fm and No, produced unavoidable

contradictions with the predictions of the liquid-drop nuclear model.[4]

It was found that the deviations of the experimental nuclear binding energies from the predictions of the classical theory demonstrate a quite regular character, namely, they are maximum (highest binding energy) at specific, so-called 'magic' numbers† of protons and neutrons in a nucleus.

As a rule, in practice the shell effects are described in terms of the shell correction to the smooth changes in the nuclear masses with the number of nucleons in the nucleus. The phenomenological description of nuclear masses with inclusion of the nuclear shell effect was first performed by Swiatecki and Myers.[5] More recently (in 1967), Strutinsky developed an original method for calculating the shell corrections to the liquid-drop nuclear energy.[6] They make a basis for the macro-microscopic model, according to which the total energy of a nucleus is given by

$$E_{\text{tot}} = E_{\text{ld}} + \Delta E_{\text{shell}},$$

where E_{ld} is the macroscopic (liquid-drop) energy of the nucleus and ΔE_{shell} is the correction that reflects the shell structure of the nucleus and the nucleon pair correlations. Calculations carried out in the framework of the macro-microscopic model predict significant shell effects not only in spherical, but in deformed nuclei as well. This made it possible to improve the precision in determining the nuclear mass and shape in the ground state. In contrast to the widely existing opinion that shell effects vanish with the increase in nuclear deformation, it turned out that in highly deformed nuclei the shell effects do not vanish (rearrangement of nucleons takes place). They still introduce a large correction to the total potential energy of the nucleus. A fundamental consequences of the macro-microscopic theory was the prediction[7–9] of the existence of the so-called 'islands of stability' in the region of the hypothetical superheavy elements (SHE), situated in the vicinity of the assumed new closed shells $Z = 114$ and $N = 184$. These predictions gave an impetus to research on the synthesis of SHE, the forecasting and study of their chemical properties, the search for SHE in nature, *etc*.

II. The synthesis of the first actinides

Many of the first artificial chemical elements heavier than uranium were discovered in reactions of successive neutron capture, *viz*., in (n,γ)-reactions, during long-term irradiations at high-flux reactors. Even at present, this method continues to be the main one for producing isotopes of elements with $Z = 93-100$. However, it is not feasible when the aim is to advance into the region of the heavier nuclei because of very short half-lives of isotopes of the elements with $Z > 100$.

All nowadays known isotopes of the elements with $Z > 100$ were synthesized in reactions induced by accelerated heavy ions. In this method, in the process of fusion with the heavy particle (projectile) its total mass is imported into the target nucleus.

† Using the terminology of atomic physics, the 'magic' numbers in the nucleus correspond to the closed proton and neutron shells.

The excitation energy of the compound nucleus is given by:

$$E_x = E_{\text{P}} - [M_{\text{CN}} - (M_{\text{T}} + M_{\text{P}})] = E_{\text{P}} - Q,$$

where E_{P} is the energy of the projectile and M_{CN}, M_{T} and M_{P} are the masses of the compound nucleus, the target nucleus and the projectile, respectively.

The minimum excitation energy is obtained at the fusion reaction threshold energy corresponding, to a first approximation, to the Coulomb barrier (B_{c}):

$$E_x^{\min} = B_{\text{c}} - Q.$$

In the case of heavy target nuclei one has $B_{\text{c}} \approx 5$ MeV nucleon^{-1}.

Contrary to the (n, γ)-reactions, where the excitation energy amounts to about 6–8 MeV, in the fusion reactions induced even by such light ions as ^{4}He, one has $E_x^{\min} \approx 20$ MeV. Increasing the mass of the projectile brings forth an increase in the excitation energy of the compound nucleus due to the increase in the Coulomb barrier. The de-excitation of the hot nucleus, *i.e*., the transition to its ground state ($E_x = 0$), will take place mainly by emission of neutrons and γ-rays.

The formation cross section of the products after the evaporation of neutrons (evaporation residues) is given by:

$$\sigma_{xn}(E_x) = \sigma_{\text{CN}}(E_x) P_{xn} \prod_{i=1}^{x} \left[\frac{\Gamma_{\text{n}}}{\Gamma_{\text{tot}}}(E_x)\right]_i,$$

where $\sigma_{\text{CN}}(E_x)$ is the cross section of the formation of the compound nucleus with the excitation energy E_x, P_{xn} is the probability of its de-excitation *via* neutron emission, and $\Gamma_{\text{n}}/\Gamma_{\text{tot}}$ is the ratio of the widths (the probabilities) of neutron emission to the total decay width at each stage of the cascade of neutron emission.

The ratio $\Gamma_{\text{n}}/\Gamma_{\text{tot}}$ can be calculated within the framework of the statistical theory under certain assumptions of the thermodynamical properties of the heated nucleus. The quantity $\sigma_{xn}(E_x)$, which characterizes the survival probability of the evaporation residues, dramatically decreases with the rise of E_x (this is equivalent to the increase in the number of evaporated neutrons). The situation gets complicated due to the fact that the amplitude of the shell correction, which hinders the fission of the nucleus from the ground state, quickly decreases with the increase in the nuclear excitation energy. Both these factors lead to an extremely low survival probability of the heavy compound nuclei. Compared to the neutron capture reactions, leading to the formation of actinides with cross sections amounting to tens and hundreds barn, the formation cross section of transactinides in heavy-ion induced reactions is in the range of only $10^{-6}-10^{-4}$ barn and exponentially decreases when advancing into the region of the SHE. However, in spite of the very small cross sections, heavy-ion induced reactions are, in principle, the only way to synthesize elements with $Z > 100$.

Since the formation cross section is maximum at the lowest value of $E_x^{\min}$, for a long time it seemed preferable to use most asymmetric reactions, *i.e*., the maximum possible heavy target and a light projectile. Then, the $E_x^{\min}$ amounted to $\geqslant 50$ MeV and these reactions were called 'hot fusion reactions'. Here, main efforts are spent on obtaining at

high-flux reactors of considerable quantities of the heaviest isotopes of the transuranium elements from Pu to Fm to be used as target material. All activities carried out in this research avenue in the 1960s and in the beginning of the 1970s at the Lawrence Berkeley National Laboratory (USA) and at the Flerov Laboratory of Nuclear Reactions (JINR, Dubna) led to the discovery of isotopes of the last three actinides ($_{101}$Md, $_{102}$No, $_{103}$Lr) and of isotopes of the first three transactinides ($_{104}$Rf, $_{105}$Db, $_{106}$Sg). The first results on the synthesis of isotopes of the elements 104–106 were obtained by Flerov *et al.* (Dubna) and Seaborg *et al.* (Berkeley) in the reactions ^{242}Pu(^{22}Ne,*x*n)^{259,260}Rf, ^{249}Cf(^{12}C,4n)^{257}Rf, ^{249}Cf(^{13}C,3n)^{259}Rf, ^{243}Am(^{22}Ne,*x*n)^{260,261}Db, ^{249}Cf(^{15}N,4n)^{260}Db and ^{249}Cf(^{18}O,4n)^{263}Sg.

In the early 1970s researchers from Dubna revealed certain advantages of the fusion reactions between magic nuclei over asymmetric reactions.[10] When the doubly magic ^{208}Pb nucleus is used as a target while ions with mass $A \geqslant 50$ are used as projectiles, $E_x^{\min}$ decreases to 10–20 MeV, which in turn significantly increases the survival probability of the compound nucleus. These reactions were called 'cold fusion' reactions and used in the 1980–1990s in Darmstadt (Germany) and in Dubna to synthesize light neutron-deficient isotopes of the elements 107–112.

By the end of the 20th century (1999), almost 60 years after the discovery of the first artificial transuranium elements, neptunium and plutonium, it became possible to reach the element 112. By and large 10 isotopes of element 104 ($^{253-262}$Rf), 8 isotopes of element 105 ($^{255-258}$Db and $^{260-263}$Db), 6 isotopes of element 106 ($^{259-261}$Sg and ^{263,265,266}Sg), 6 isotopes of element 107 (^{260,261,262,264,266,267}Bh), 4 isotopes of element 108 ($^{263-265}$Hs), 2 isotopes of element 109 (^{266,268}Mt), 3 isotopes of element 110 (^{267,269,271}Ds), 1 isotope of element 111 (^{272}Rg) and 1 isotope of element 112 (^{277}112) were obtained by that time. For the majority of these isotopes, the decay mode and lifetime were determined with high confidence; for some isotopes, the mass excess and quantum characteristics of the low-lying states were calculated.

Of special interest were the relatively long-lived isotopes. Their synthesis made it possible not only to determine the nuclear physical properties, but also to study their chemical behaviour in comparison with that of lighter homologues. The lifetimes, the synthesis reactions and the main decay modes of the isotopes of elements 104–108 are given in Table 1.

The properties of the transactinides were studied by thermochromatography (for elements 104–108) and by different versions of ion-exchange and extraction chromatography (for elements 104–106) were used.

Peculiarities of the chemistry experiments with such short-lived isotopes were reported in detail by Schaeddel,[11] Guseva [12] and Zvara.[13] Here we shall only mention that these experiments revealed no significant difference between the chemical properties of elements 104–108 and their lighter homologues.

Table 1. Transactinide isotopes for chemical research.

Isotope	$T_{1/2}$/s	Synthesis reaction	Main decay modes
^{261m}Rf	81	^{248}Cm (^{18}O,5n)	α
		^{244}Pu (^{22}Ne,5n)	α
^{262}Db	35	^{249}Bk (^{18}O,5n)	α, SF
		^{248}Cm (^{19}F,5n)	α, SF
^{263}Db	29	^{249}Bk (^{18}O,4n)	α
^{265}Sg	7.4	^{248}Cm (^{22}Ne,5n)	α, SF
^{266}Sg	21	^{248}Cm (^{22}Ne,4n)	SF, α
^{267}Bh	22	^{249}Bk (^{22}Ne,5n)	α
^{269}Hs	27	^{248}Cm (^{26}Mg,5n)	α, SF

III. Isotopes of the superheavy elements

As mentioned above, up to the year 2000 the heaviest elements including element 112 were synthesized in 'cold fusion' reactions. Unfortunately, these reactions can not be used for the synthesis of nuclei, which are within the range of the theoretically predicted 'island of stability' of SHE. In the fusion of the nuclei ^{208}Pb and ^{76}Ge, the compound nucleus ^{284}114 ($N = 170$) is 14 neutrons away from the closed neutron shell $N = 184$. At such a large neutron deficiency in the compound nucleus, the evaporation residues would be positioned far from the boundary of the predicted 'island of stability'. It is noteworthy that no combination of stable and even long-lived isotopes can make it possible to advance to the nuclei at the 'peak' of the 'island of stability'. For this reason, it is necessary to advance as close as possible to the limits of this unexplored region so as to fall within the influence of the spherical shell $N = 184$. The nuclides with a larger number of neutrons can, in principle, be produced using isotopes of the heavy actinides with $Z = 94–98$ as the target material and the rare ^{48}Ca isotope as the bombarding particle.

In this case, while losing the possibility to use the magic ^{208}Pb nuclei as targets in cold fusion reaction, we make again use of proton and neutron magic numbers, but this time in the ^{48}Ca projectile.

In the fusion isotopes of heavy actinides with the magic ^{48}Ca nucleus, the excitation energy of the compound nucleus is about 30 MeV. One can expect that at such an excitation energy the shell effects are still noticeable in the excited nucleus and the survival probability of the evaporation residues is higher than in conventional 'hot fusion' reactions ($E_x \geqslant 50$ MeV). At the same time, the mass and charge asymmetry of the nuclei in the entrance channel ($Z_1 \times Z_2 \leqslant 2000$) must lead to weaker Coulomb repulsion and, therefore, to an increase in the formation cross section of the compound nucleus ^{284}114 as compared to the cold fusion reaction ^{208}Pb + ^{76}Ge ($Z_1 \times Z_2 = 2624$).

In spite of the obvious advantages, all attempts to synthesize new elements in reactions with ^{48}Ca ions in 1977–1985 in different laboratories [14,15] resulted only in obtaining upper limits of their formation cross section. At the same time, recent progress in experimental techniques and the possibility to accelerate intense ^{48}Ca beams at new-generation heavy ion accelerators made it possible to make a significant step ahead, *viz.*, to increase the sensitivity of the experiments by two and even three orders of magnitude.

The experiments were carried out at the accelerator complex of the Flerov Laboratory of Nuclear Reactions (FLNR), JINR (Fig. 1). Reactions of ^{48}Ca with unique neutron-rich isotopes of the actinides (Act): ^{242,244}Pu, ^{243}Am, ^{245,248}Cm and ^{249}Cf were investigated. The target Act material (as an oxide of thickness 0.35–0.40 mg cm^{-2}) was deposited on a rotating (2000 rpm) disk made of a thin (1.5 μm) titanium foil of total surface area 32 cm^2 and



Figure 1. Accelerator complex at the Flerov Laboratory of Nuclear Reactions, JINR.

irradiated by ^{48}Ca ions of a given energy. The average intensity of the ion beam on the target amounted to 6×10^{12} s^{-1}.

The recoil nuclei are products of the reaction, left the target and were separated from the beam and other by-products by a gas-filled separator and then implanted into a 4×12 cm^2 semiconductor detector with 12 vertical position-sensitive strips, which was placed in the focal plane of the separator at a distance of 4 m from the target. The front detector was additionally surrounded by 8 (4×4 cm^2) detectors for the registration of α-particles and spontaneous fission fragments from the decay of the implanted nuclei. The efficiency of registration of the α-particles was 87% of 4π. A time-of-flight (TOF) detector was placed in front of the focal detector. The time of flight of the superheavy nuclei from the target to the focal detector was about 1 μs. Behind the front detector, a 'veto' detector was placed to exclude signals from weakly ionizing particles.

The efficiency of registration of one spontaneous fission fragment was 100%, for two fragments — about 40%. The layout of the experiment is shown in Fig. 2. The experimental conditions (reactions, target characteristics, energy and integrated dose of the ^{48}Ca ions) and the number of decay chains of the new nuclides registered in different reaction channels (2n – 5n) are given in Table 2.

In the reactions listed in the first Table 2, 86 decay chains were registered. Most all decay chains are characterized by consecutive α-decays of the parent and then of the daughter nuclides (from one to six) and are terminated by spontaneous fission. In some cases spontaneous fission was observed immediately for the parent nuclides. Chains with identical characteristics (decay modes, energy and lifetime) were attributed to the decay of the same nuclide, independently of the way of production — either in a direct reaction or as daughter products of heavier nuclides.

The identification of the atomic numbers of the new nuclides was done by different independent methods based on:

— the mechanism of fusion reactions (excitation);

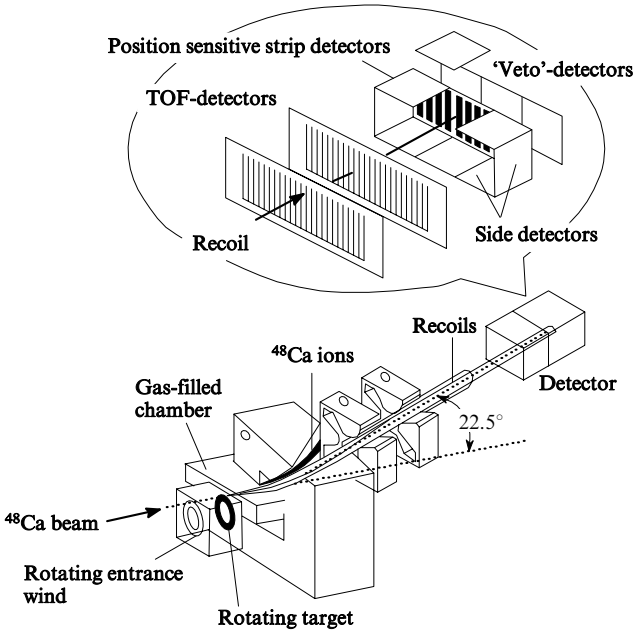


Figure 2. Layout of the experimental setup for the synthesis of SHE with the gas-filled separator.

— the decay properties of the nuclei (half-lives T_α and α-decay energies Q_α of even – even and many even – odd ‡ isotopes;

— the radiochemical identification of the atomic number of the nuclides ^{268}Db and ^{283}112 in correlated α-decay chains of the nuclei $115 \rightarrow 113 \rightarrow \text{Rg} \rightarrow \text{Mt} \rightarrow \text{Bh} \rightarrow (\text{Db})$ and $116 \rightarrow 114 \rightarrow (112) \rightarrow \text{Ds} \rightarrow \text{Hs} \rightarrow \text{Sg} \rightarrow \text{Rf}$.

As a result of these experiments, 13 isotopes of the new superheavy elements with atomic numbers 113 – 116 and 118, as well as 21 new isotopes of the formerly known transactinides with $Z = 104 - 112$, were discovered. The properties of the new nuclides are presented in Table 3.

The experimental values of the half-lives and the energies of α-decays for the Z-even nuclides with $Z \geqslant 100$ are shown in Fig. 3 along with the values calculated using the Viola – Seaborg relation.

$$\log T_\alpha = (aZ + b)Q_\alpha^{-1/2} + cZ + d.$$

It can be seen that the experimental data obtained confirm the main theoretical predictions for the considerable increase in the stability of nuclei close to the deformed shell $Z = 108$, $N = 162$. A much stronger rise of stability is observed for the superheavy nuclides in the region $Z = 112 - 118$ and $N \geqslant 170$. This effect is due to the influence of the spherical shell $N = 184$. The experimental values Q_α for the isotopes $Z = 110 - 118$ are 0.1 – 0.5 MeV lower than the calculated ones; consequently, the lifetimes are 2 – 10 times longer than the theoretically predicted ones.

‡ Even – even, even – odd and odd – odd isotopes are isotopes with corresponding numbers of protons and neutrons.

Table 2. Experimental conditions and the number of registered decay events from the reactions Act + ^{48}Ca.

Reaction	Target thickness /mg cm^{-2}	Isotopic enrichment (%)	E_{LAB} /MeV	E_x /MeV	Dose of ^{48}Ca-ions on target	Number of events				Ref.
						2n	3n	4n	5n	
^{233}U + ^{48}Ca	0.44	99.97	240	32.7 – 37.1	0.70×10^{19}	–	–	–	–	16
^{238}U + ^{48}Ca	0.35	99.3	230	29.3 – 33.5	0.58×10^{19}	–	1	–	–	16
			234	32.9 – 37.2	0.71×10^{19}	–	6	–	–	16
			240	37.7 – 41.9	0.52×10^{19}	–	–	1	–	16
^{237}Np + ^{48}Ca	0.35	99.3	244	36.9 – 41.2	1.10×10^{19}	–	2	–	–	17
^{242}Pu + ^{48}Ca	0.40	99.98	235	30.4 – 34.7	0.50×10^{19}	1	5	–	–	16
			238	33.1 – 37.4	0.49×10^{19}	–	5	–	–	16
			244	38.0 – 42.4	0.47×10^{19}	–	5	7	–	16
			250	43.0 – 47.2	0.32×10^{19}	–	–	2	–	16
^{244}Pu + ^{48}Ca	0.38	98.6	231	29.8 – 32.9	0.46×10^{19}	–	–	–	–	18
			236	32.9 – 37.4	1.50×10^{19}	–	2	–	–	18
			243	38.9 – 43.0	0.40×10^{19}	–	2	7	–	19
			250	44.9 – 49.0	0.31×10^{19}	–	1	4	–	19
			257	50.4 – 54.7	0.29×10^{19}	–	–	1	1	19
^{243}Am + ^{48}Ca	0.36	99.9	248	38.0 – 42.3	0.43×10^{19}	–	3	–	–	20, 21
			253	42.4 – 46.5	0.43×10^{19}	–	–	1	–	20, 21
^{245}Cm + ^{48}Ca	0.35	98.7	243	30.9 – 35.0	1.20×10^{19}	2	3	–	–	19
			249	35.9 – 39.9	0.54×10^{19}	1	5	–	–	22
			255	40.7 – 44.8	0.83×10^{19}	–	4	–	–	22
^{248}Cm + ^{48}Ca	0.32	96.3	240	30.4 – 35.8	0.66×10^{19}	–	3	–	–	23
	0.35	97.4	247	36.8 – 41.1	0.70×10^{19}	–	2	6	–	16
^{249}Cf + ^{48}Ca	0.23	97.3	245	26.6 – 31.7	2.50×10^{19}	–	1	–	–	24
	0.34	> 98	251	32.1 – 36.6	1.60×10^{19}	–	2	–	–	22

Table 3. Decay properties of the new nuclides.

Z	A	No. observations	Decay mode (%)	$T_{1/2}$ (see [a])	Calculated value $T_{1/2}$	E_α /MeV	Q_α /MeV
118	294	3	α	$0.89^{+1.07}_{-0.31}$ ms	0.4 ms	11.65 ± 0.06	11.81 ± 0.06
116	293	4	α	61^{+57}_{-20} ms	80 ms	10.54 ± 0.06	10.69 ± 0.06
	292	5	α	18^{+16}_{-6} ms	40 ms	10.66 ± 0.07	10.80 ± 0.07
	291	3	α	18^{+22}_{-6} ms	20 ms	10.74 ± 0.07	10.89 ± 0.07
	290	10	α	$7.1^{+3.2}_{-1.7}$ ms	10 ms	10.84 ± 0.08	11.00 ± 0.08
115	288	3	α	87^{+105}_{-30} ms	60 ms	10.46 ± 0.06	10.61 ± 0.06
	287	1	α	32^{+155}_{-14} ms	30 ms	10.59 ± 0.09	10.74 ± 0.09
114	289	10	α	$2.6^{+1.2}_{-0.7}$ s	2 s	9.82 ± 0.05	9.96 ± 0.05
	288	18	α	$0.80^{+0.27}_{-0.16}$ s	0.9 s	9.94 ± 0.06	10.08 ± 0.06
	287	16	α	$0.48^{+0.16}_{-0.09}$ s	0.5 s	10.02 ± 0.06	10.16 ± 0.06
	286	24	α (50)/SF (50)	$0.13^{+0.04}_{-0.02}$ s	0.2 s	10.19 ± 0.06	10.33 ± 0.06
113	284	3	α	$0.48^{+0.58}_{-0.17}$ s	0.3 s	10.00 ± 0.06	10.15 ± 0.06
	283	1	α	100^{+490}_{-45} ms	140 ms	10.12 ± 0.09	10.26 ± 0.09
	282	2	α	73^{+134}_{-29} ms	6 ms	10.63 ± 0.08	10.78 ± 0.08
112	285	10	α	29^{+13}_{-7} s	50 s	9.15 ± 0.05	9.28 ± 0.05
	284	19	SF	97^{+31}_{-19} ms			9.80
	283	22	α (⩽100)/SF (⩽10)	$3.8^{+1.2}_{-0.7}$ s	3 s	9.54 ± 0.06	9.67 ± 0.06
	282	12	SF	$0.82^{+0.30}_{-0.18}$ ms			10.69

Table 3 (continued).

Z	A	No. observations	Decay mode (%)	$T_{1/2}$ (see [a])	Calculated value $T_{1/2}$	E_α/MeV	Q_α/MeV
111	280	3	α	$3.6^{+4.3}_{-1.3}$ s	0.4 s	9.75 ± 0.06	9.87 ± 0.06
	279	1	α	170^{+810}_{-80} ms	7 ms	10.37 ± 0.16	10.52 ± 0.16
	278	2	α	$4.2^{+7.5}_{-1.7}$ ms	1 ms	10.69 ± 0.08	10.85 ± 0.08
110	281	10	SF	$11.1^{+5.0}_{-2.7}$ s			9.00
	279	26	α (10)/SF (90)	$0.20^{+0.05}_{-0.04}$ s	0.2 s	9.70 ± 0.06	9.84 ± 0.06
109	276	3	α	$0.72^{+0.87}_{-0.25}$ s	0.1 s	9.71 ± 0.06	9.85 ± 0.06
	275	1	α	$9.7^{+46.0}_{-4.4}$ ms	2 ms	10.33 ± 0.09	10.48 ± 0.09
	274	2	α	445^{+810}_{-176} ms	20 ms	9.76 ± 0.10	9.90 ± 0.10
108	275	3	α	$0.19^{+0.22}_{-0.07}$ s	0.8 s	9.30 ± 0.06	9.44 ± 0.06
107	272	3	α	$9.8^{+11.7}_{-3.5}$ s	3 s	9.02 ± 0.06	9.15 ± 0.06
	270	1	α	61^{+292}_{-28} s	5 s	8.93 ± 0.08	9.06 ± 0.08
106	271	3	α (70)/SF (30)	$1.9^{+2.4}_{-0.6}$ min	0.8 min	8.54 ± 0.08	8.67 ± 0.08
105	268	18	SF/EC [b]	29^{+9}_{-6} h			7.83
	267	1	SF	$1.2^{+5.8}_{-0.6}$ h			8.15
	266	1	SF/EC [b]	22^{+105}_{-10} min			8.24
104	267	2	SF	$1.3^{+2.3}_{-0.5}$ h			8.21

[a] Hereafter, the subscript and superscript on the right-hand side refer to measurement error. [b] EC is electron capture.

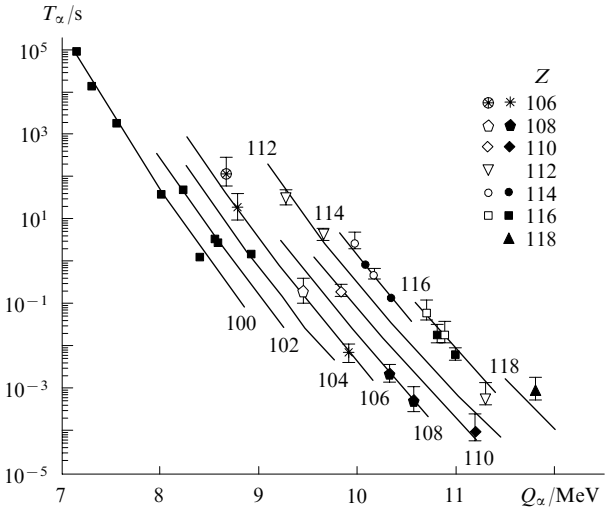


Figure 3. Experimental half-lives (T_α) and α-decay energies (Q_α) for even-Z nuclides with $Z \geqslant 100$.
The solid symbols refer to even–even nuclides and the open symbols refer to even–odd nuclides. The solid lines represent calculations using the Viola–Seaborg formula.

Unfortunately, the synthesis of neutron-rich isotopes with N close to 184 is still not feasible due to constraints caused by the use of stable isotopes with a given proton-neutron ratio as projectiles. However, the decay properties of the 34 new nuclei are evidence of the enormous effect of the new nuclear shells, which bring forth 'islands of stability' in the region of the formerly unknown superheavy elements.

IV. Chemical identification and properties of SHE

As mentioned above, chemical identification of the ^{268}Db and ^{283}112 isotopes in the correlated decay chains $115 \rightarrow 113 \rightarrow \text{Rg} \rightarrow \text{Mt} \rightarrow \text{Bh} \rightarrow (\text{Db})$ and $116 \rightarrow 114 \rightarrow (112) \rightarrow \text{Ds} \rightarrow \text{Hs} \rightarrow \text{Sg} \rightarrow \text{Rf}$ is an independent method of identification of the atomic numbers of the new nuclides. For the first time, this experiment was carried out [25] for element 115.

The isotope of element 115 with the mass number 288 was discovered in the reaction $^{243}\text{Am}(^{48}\text{Ca},3n)^{288}115$ in the experiments carried out at the gas-filled separator of FLNR JINR.[20] It was found out that the isotope $^{288}115$ undergoes 5 consecutive α-decays, which terminate by the spontaneous fission of ^{268}Db (Fig. 4). The total time of these 5 transitions was about 20 s. The half-life of the final nucleus, ^{268}Db, determined using three events observed was 16^{19}_{-6} hours. The possibility for the observed spontaneous fission of the odd–odd nucleus ^{268}Db ($Z = 105$, $N = 163$) to be due to electron capture with subsequent fast fission of the even–even nucleus ^{268}Rf ($Z = 104$, $N = 164$, $T_{SF} \approx 1.5$ s) was taken into account. This did not overthrow the conclusion that the experimentally registered long spontaneous fission half-life was determined by the decay of ^{268}Db. Because of the small number of events it could also not be excluded that the ^{268}Db isotope could in some cases undergo α-decay followed by the spontaneous fission of ^{264}Lr.

The ^{268}Db isotope cannot be produced at an energy close to the Coulomb barrier *via* the transfer to the ^{243}Am target nucleus of 25 nucleons (10 protons and 15 neutrons) from the ^{48}Ca nucleus.[26]

The new isotope of element 105 with a half-life of about 1 day is an excellent object not only for the identification of

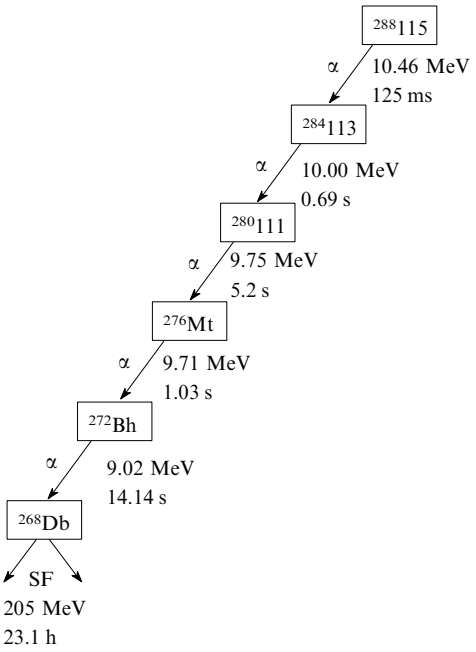


Figure 4. Decay of the isotope $^{288}115$, synthesized in the $^{243}Am + {}^{48}Ca$ reaction using the gas-filled separator for recoil nuclei.[20]

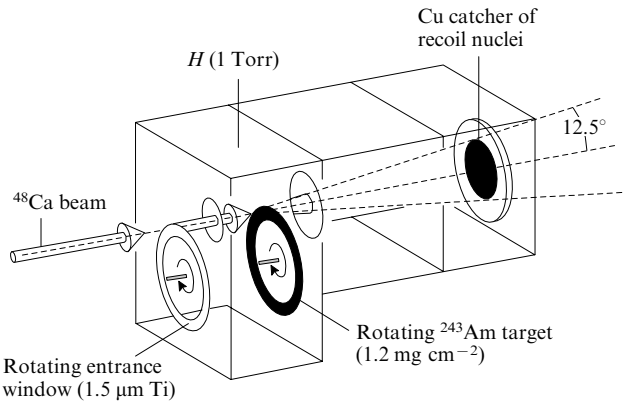


Figure 5. Schematic of experimental setup for chemical identification of ^{268}Db.[21]

the new nuclide, but also for the investigation of its chemical properties.

According to the ground-state electronic configuration ($[Rn]5d^{14}6p^{3}7s^{2}$), dubnium pertains to Group 5 elements in the Periodic Table, being a heavier homologue of Nb and Ta. Obviously, the chemical identification of Db requires its isolation accordance with characteristics of this group. For this reason, the method of sorption isolation of Group 5 elements in the form of anionic fluoride complexes was used. Taking into account the fact that the ^{268}Db isotope of interest undergoes spontaneous fission, particular attention was paid to the isolation of Group 5 elements from the actinides and first of all from the long-lived spontaneously fissioning isotopes of californium ^{252}Cf ($T_{1/2} = 2.65$ year, SF 3.1%) and ^{254}Cf ($T_{1/2} = 60.5$ day, SF 99.7%). The separation from the actinides also gave an answer to the question whether the observed spontaneous fission came from transactinides, which in turn excluded another decay mode:

$$^{268}Db \xrightarrow{\alpha} {}^{264}Lr \xrightarrow{SF}$$

The experiment was performed at the U400 heavy ion accelerator (FLNR, JINR) in June 2004. The experimental layout is shown in Fig. 5.

The target, in the form of a rotating disk made of a thin (1.5 μm) titanium foil of total surface area 32 cm^2 covered with ^{243}Am (99.9%) in the form of an oxide 1.2 mg cm^{-2} thick relative to ^{243}Am, was irradiated with 247 MeV ^{48}Ca ions (the energy corresponds to the middle of the target). The recoil nuclei (reaction products), left the target into an angular range $\theta = 0-12.5^\circ$ and were stopped in a copper catcher 50 mm in diameter placed at a distance of 100 mm downstream the target. Products of the fusion reaction of ^{243}Am with ^{48}Ca (in the given geometry the angular acceptance was $\pm 12.5^\circ$) fell on the catcher with a nearly 100% efficiency. The range of the recoil nuclei in the catcher material was at most 3–4 μm. Eight identical experiments were carried out. In each experiment, the irradiation time was 20 to 45 hours.

In each experiment, at the end of irradiation, the recoil nuclei catcher was transported to the radiochemical laboratory. The Group 5 elements were separated from other reaction products (including the actinides) by co-sorption from the nitrate solution onto a Dowex 50×8 cation-exchange resin followed by selective desorption of Group 5 elements in the form of fluoro anion complexes. The separation efficiency of Group 5 elements was close to 90% with a separation coefficient from the lanthanides of the order of 10^5.

The procedure (from the end of irradiation until the beginning of the detector measurements) took 2 to 3 hours. Details are available in the literature.[25, 27]

After the radiochemical separation and preparation of thin spectroscopic sources, long-term measurements of the specra of α-particles and correlated spontaneous fission fragments with simultaneous registration of fission neutrons were performed.

In the eight irradiations of ^{243}Am targets with ^{48}Ca ions (total beam dose 3.4×10^{18}), 15 events of spontaneous fission were detected. The measurements continued for 957 hours. All 15 spontaneous fission events were registered within a time interval of 174 hours after the beginning of the measurements. During the remaining 783 hours, no spontaneous fission events were detected. Taking account of the separation factor of Group 5 elements and actinides equal to 10^5, the actinide isotopes including ^{264}Lr could produce no more than 0.002 spontaneous fission event after 174 hours of measurement.

The total kinetic energy ($\overline{TKE}$) of the fission fragments, defined as the sum of the amplitudes of coincidence signals from both detectors, was about 230 MeV after taking account the absorption in the source and backing. This result is in agreement with the data of the physical experiment ($\overline{TKE} \approx 225$ MeV). The average number of neutrons per fission event was $\bar{\nu} \sim 4.2$. Both parameters, the high value of TKE and the large number of neutrons, provide evidence of the fission of a heavy nucleus. It is noteworthy that for the spontaneous fission of ^{248}Cm one has $\overline{TKE} = 181$ MeV and $\bar{\nu} = 3.14$ while for ^{252}Cf one has $\overline{TKE} = 185$ MeV and $\bar{\nu} = 3.75$.

It should be noted that Group 4 and 5 elements were isolated together in the experiment mentioned above. For

this reason, the method employed is applicable both to isotopes, Db and Rb, the final products of decay of the parent nuclei with $Z = 115$ and 114, respectively. However, the observation of spontaneous fission of the Rf isotopes should be excluded due to the negligibly small cross section of the reaction $^{243}Am(^{48}Ca,pxn)^{290-x}114$ and the short spontaneous fission lifetime expected for an even-Z nuclide. That is why all spontaneous fission events registered by the detectors in the $^{243}Am + ^{48}Ca$ reaction were attributed only to the decay chain of element 115. The experimental conditions (separation of the actinides) also excluded the assumption of possible additional α-decay of ^{268} Db followed by spontaneous fission of ^{264}Lr. Generally, the experiment described served as an independent proof of the synthesis of element 115 and also of element 113 in the $^{243}Am + ^{48}Ca$ reaction.

A detailed comparative analysis of the 'physical' and 'chemical' experiments on the synthesis of element 115 in the $^{243}Am + ^{48}Ca$ reaction is given in Ref. 21.

More recently, the chemical experiment was repeated [28, 29] using more sophisticated methods of selection, which ensure the separation of Groups 5 and 4 elements and in some cases, the separation of the elements within Group 5 (niobium and tantalum fractions were isolated). Five spontaneous fission events were registered, including 3 events in the joint fraction of Group 5 elements and 2 events in the Ta fraction. In the Group 4 fraction, spontaneous fission events were not registered. Thus, the results of the chemical identification of element 115 were confirmed using the final decay product ^{268}Db.

As to the chemical properties of Db, the authors rightly noted that the obtained experimental statistics did not allow unambiguous conclusions about the chemical properties of Group 5 elements in the oder Nb–Ta–Db. Additional experiments are necessary, which (accounting for the ^{268}Db half-life of 29 hours) can guarantee wide possibilities of detailed studies of the chemical properties of this transactinide.

The study of the chemical properties of the superheavy elements is of special interest. In the region of element 114 and its neighbours, a significant role in the electronic structure of the superheavy atom, in particular of the outmost electrons, is played by the so-called relativistic effects. Elements 112 (also Zn, Cd, Hg) and 114 (Ge, Sn, Pb) belong to Groups 12 and 14 in the Periodic Table. It follows that their ground-state electronic configurations should be $[Rn]5f^{14}6d^{10}7s^2$ and $[R]5f^{14}6d^{10}7s^27p^2$, respectively. Simple extrapolations show that element 112 is similar in properties to noble metals. At the same time, the inclusion of relativistic effects§ in the calculations of the atomic properties of SHE implies compression of the spherical s- and $p_{1/2}$-electron orbitals. Consequently, the chemical stability of the elements 112 and 114 in their ground state increases. Moreover, it cannot be excluded that these elements may be noble gases similar to radon. Other calculations also confirm the strong compression of the 7s orbitals. However, it is also predicted that the relativistic spin-orbital splitting of the 6d orbitals will cause element 112 to be a noble transition metal or even a semiconductor. Note that in the case of element 112 the majority of theoretical predictions show that an increased volatility in the elementary state is expected, which contradicts the estimate of the chemical inertness (eka-Hg and eka-Rn). An answer to this question could be obtained by studying the adsorption of the element on the surface of noble metals.

These chemical experiments became possible only recently after the synthesis of the relatively long-lived isotopes of element 112 in the above-mentioned reactions of ^{48}Ca with ^{238}U and ^{242,244}Pu. In the text below we present key result of the chemical experiments with the isotopes of elements 112 and 114 obtained at the accelerator complex of FLNR JINR in 2006–2007. ^{nat}Nd (about 1% by weight) was added to the ^{242}Pu target material. The 49-s α-emitter ^{185}Hg produced in the $^{142}Nd + ^{48}Ca$ reaction was used as monitor throughout the whole experiment. In addition, incomplete fusion reactions of ^{48}Ca and ^{242}Pu produce isotopes of radon (including the 4-s α-emitter ^{219}Rn). Thus, in the experiments the behaviour of three chemical elements (element 112, Hg and Rn) was controlled simultaneously.

The ^{242}Pu target was irradiated with 270-MeV ^{48}Ca ions. The isotopes of element 114, produced as fusion products, left the target, were stopped in a He–Ar medium and transferred to the detectors through a capillary. The detector system consisted of 32 pairs of PIPS detectors, providing high efficiency of registration of α-particles and spontaneous fission fragments. One detector in each pair (the detectors faced each other) was covered with a thin gold layer. Along the detector array (downstream the He–Ar jet) the temperature on the surface of the detectors varied from 35 °C (first pair of detectors) to −184 °C (last pair). Two 'scenarios' were possible. If element 114 is volatile (at room temperature) and the system performance (time-taken by an atom from the target chamber to reach the detectors) is comparable to the lifetime of its isotopes (*e.g.* ≈ 0.5 s for ^{287}114), then the atoms of element 114 will directly be captured by the He–Ar jet and transported *via* the Teflon capillary to a distance of about 10 m into a cryogenic detector. If element 114 is not volatile or the system performance is not high enough, the He–Ar jet will capture the atoms of element 112, produced in the decay of element 114.

In the experiment carried out in the year in 2006, the system performance was to about 4 s; the aim was to study only the properties of element 112. Two decay chains ($\alpha \rightarrow f$) were registered, exactly in agreement with the available data for the isotope ^{283}112. It should be noted that in the physical experiments the isotope ^{283}112 and its daughter nucleus, the isotope ^{279}110, were registered as both a direct product of the $^{238}U(^{48}Ca,3n)$ reaction and the daughter and granddaughter nuclei in the decay chains of the radionuclides ^{287}114 and ^{291}116 in the $^{242}Pu(^{48}Ca,3n)$ and $^{245}Cm(^{48}Ca,2n)$ reactions. Thus, the results of the chemical experiments [30] were the first independent confirmation of the synthesis of the new elements 112, 114 and 116. More recently, the synthesis of element 112 in the $^{238}U(^{48}Ca,3n)$ reaction was confirmed by Hofmann's group (Germany) in a physical experiment at the kinematic separator SHIP.[31]

In a series of experiments (in 2007), 3 additional events corresponding to the formation and decay of the ^{283}112 nucleus were registered. The obtained data [32] on the chemical behaviour of element 112 completely repeated the results of the first experiment.

§ Theoretical estimates of the influence of relativistic effects on the properties of SHE, including analysis of different calculation methods, are given in the reviews of Pershina and Zaitzevskii (see this issue, p. 1153 and p. 1173).

Atoms of element 112 were found on a gold surface in the temperature range from -5 °C to -39 °C. Statistical analysis of the data performed in Ref. 33 allowed one to estimate the standard enthalpy of adsorption of element 112 on the surface of gold ($-\Delta H_{ads} = 52^{+4}_{-3}$ kJ mol^{-1}). Using the accepted empirical dependence

$$-\Delta H_{ads} = [(1.08 \pm 0.005)\Delta H_{subl} + (10.3 \pm 6.4)] \text{ kJ mol}^{-1},$$

the standard enthalpy of adsorption could be determined as 39^{+23}_{-10} kJ mol^{-1}. Taking into account the calculated entropy of sublimation, the boiling temperature of element 112 was estimated to be equal to 357 K. A conclusion was drawn that element 112 is a heavier homologue of Hg because the trend of changed in the properties of Group 12 elements also hold for it.¶

In the second experiment, the time for transportation of the recoil nuclei to the detectors was reduced to 1 s. In this experiment,[34–36] an event was registered, completely repeating the decay chain of the ^{287}114 nucleus in the experiment at the gas-filled separator (Fig. 6). The decay of the nucleus was registered by a gold-coated detector which was at a temperature of about -80 °C. This is still one event only. The future experiments are aimed to increase the number of events; this can be achieved by increasing the system performance. The reproduction of this result would mean that the behaviour of element 114 is closer to that of the inert gases rather than to its nearest homologue Pb.

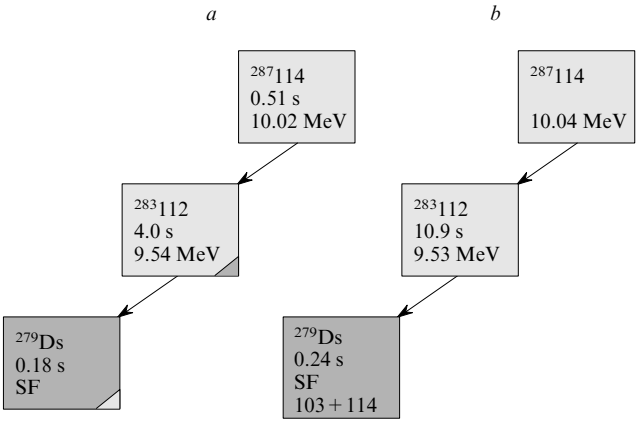


Figure 6. Decay chains of the ^{287}114 isotope: (*a*) synthesis at the gas-filled separator [16] and (*b*) chemical experiment.[35]

V. Perspectives

Reactions between the magic nucleus ^{48}Ca and the actinide targets were used to synthesize new superheavy elements in the Mendeleev's Periodic Table with atomic numbers 112–116 and 118. The synthesis of element 117 was not realized up to now due to difficulties in producing the necessary target material ^{249}Bk. The half-life of this unique isotope is only 320 days and its production (long-term irradiation of lighter and relatively long-lived isotopes of the actinides at a high-flux reactor, followed by isolation and purification) in the needed quantity (tens of mg) is a very complicated task. Far from being easy is the experiment with ^{249}Bk, because of both the high activity of the target (more than 10 Ci) and the relatively short lifetime of the isotope. However, the expected result conforms to these difficulties.

The theoretically calculated decay chains of the isotopes of element 117, formed in the reaction ^{48}Ca + ^{249}Bk are given in Fig. 7. As can be seen, 14 new isotopes of the SHE may be synthesized in this reaction and the lifetimes of the isotopes of elements 113 and 115 will be long enough to permit experiments aimed to study their chemical properties.

At present, researchers at the FLNR, JINR in collaboration with the Oak Ridge and Livermore National Laboratories (USA) and RIAR (Dimitrovgrad, Russia) are going on for the performance of these experiments.

The synthesis of heavier isotopes in ^{48}Ca-induced fusion reactions is limited by the absence of target material. The isotope ^{249}Cf, which is used as a target for the synthesis of nuclei with $Z = 118$, may turn to be the last one in the actinide target series. At the same time, according to the microscopic models of the Hartree–Fock–Bogoliubov type or to the relativistic mean field theory, the advance to the heavier nuclei with $Z \geqslant 120$ leads to a change in the distribution of the matter density in the nuclear volume (the so-called 'bubble' nuclei). Improved stability of these nuclei is expected.

The synthesis of such nuclei requires fusion reactions with ions heavier than ^{48}Ca (*e.g.*, ^{50}Ti, ^{58}Fe, ^{64}Ni, *etc.*). Preliminary experiments [37] carried out in 2008 at the DGFS (FLNR, JINR) allowed one to determine the upper limit of the formation cross-section of the isotopes of element 120 in the ^{244}Pu + ^{58}Fe reaction equal to 0.4 pb, *i.e.*, at least an order of magnitude less than in the ^{48}Ca induced reactions. Obviously, the experiments with ions heavier than Ca need cardinal modernization of experimental equipment (accelerators, separators).

The rising stability of the heaviest nuclei with $Z \geqslant 120$ entirely changes the method of their synthesis in fusion reactions with heavy ions. The need for in-flight separation of reaction products in the synthesis of nuclei with $Z = 112-114$ in order to quickly transport the recoil atoms to the detectors is not the decisive factor because their half-lives are sufficiently long. The use of classic on-line mass-separators is preferable, because the latter, together with their high efficiency due to the possibility to use thicker targets and the higher selectivity, make it possible to determine the mass of the nuclei. This idea is being realized in Dubna with the MASHA (Mass Analyzer of Superheavy Atoms) facility, which is passing tests now.

By and large, the results obtained recently provide the first experimental confirmation of the hypothesis of the existence of 'islands of stability' for superheavy elements. This offer great prospects for further studies on the synthesis and properties of superheavy elements of Mendeleev's Periodic Table.

The experiments described in this review were performed at the accelerator complex of the Flerov Laboratory of Nuclear Reactions, JINR (Dubna, Russia) in collaboration with the Analytical and Radiochemical Division of the Lawrence Livermore National Laboratory (LLNL, Livermore, USA) and the Department of Radiochemistry of the Paul Scherrer Institute (PSI, Villigen, Switzerland).

¶ Experimemtal design is reported in more detail by Gäggeler (see this issue, p. 1139).

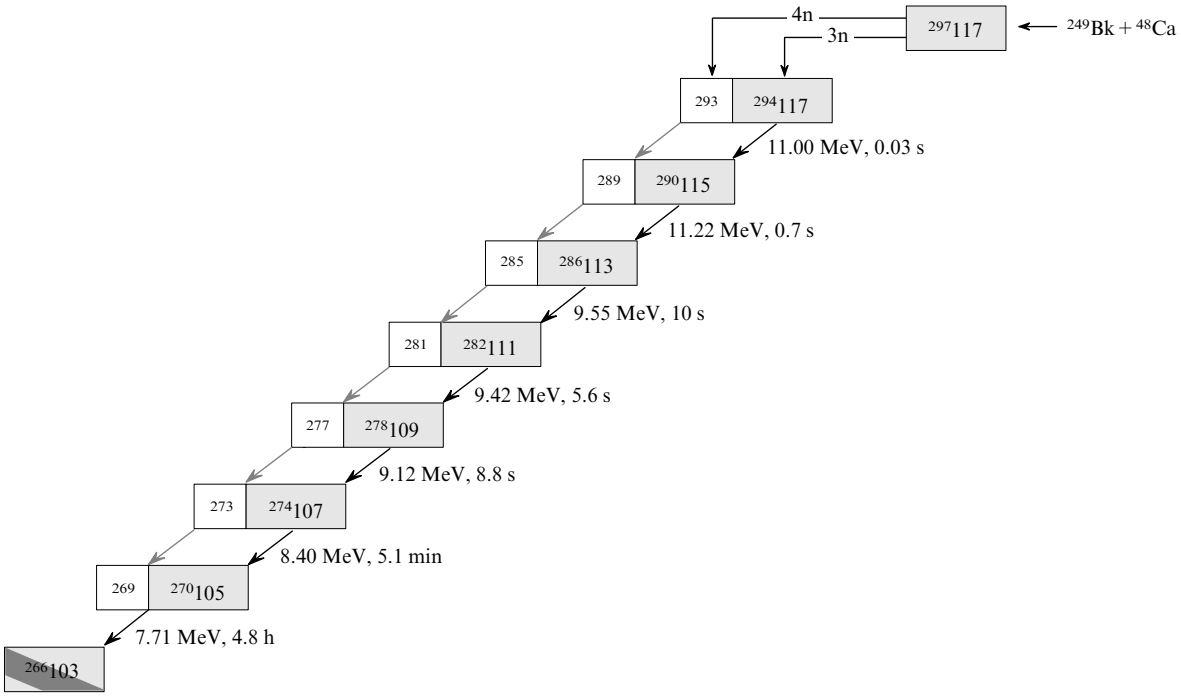


Figure 7. Expected decay scheme of the isotopes of element 117 in the $^{249}Bk + ^{48}Ca$ reaction.

Unique isotopes of Pu, Am, Cm and Cf were produced at the high-flux reactors of RIAR (Dimitrovgrad, Russia) and the Oak Ridge National Laboratory (USA). Additional isotopic enrichment was done at the mass-separator C2 at the RFNC – VNIIEF (Sarov, Russia).

Successful performance of the experiments became possible thanks to the concentration of the scientific and engineering resources of FLNR, the support of the Directorate of JINR and the wide collaboration with the scientists from the leading scientific centres working in the field.

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