Pure Appl. Chem., Vol. 78, No. 5, pp. 889–904, 2006.
doi:10.1351/pac200678050889

Synthesis and decay properties of superheavy elements*

Yuri Oganessian

Flerov Laboratory of Nuclear Reactions, Joint Institute for Nuclear Research, 141980 Dubna, Moscow, Russia

Abstract: A fundamental outcome of modern nuclear theory is the prediction of the "island of stability" in the region of hypothetical superheavy elements. A significant enhancement in nuclear stability at approaching the closed shells with $Z = 114$ (possibly 120 and 122) and $N = 184$ is expected for the nuclei with large neutron excess. For this reason, for the synthesis of nuclei with $Z = 112$–116 and 118, we chose the reactions ^{238}U, ^{242,244}Pu, ^{243}Am, ^{245,248}Cm, and ^{249}Cf + ^{48}Ca, which are characterized by fusion products with a maximal neutron excess. The formation and decay properties of the heaviest nuclei were registered with the use of a gas-filled recoil separator installed at a ^{48}Ca-beam of the heavy-ion cyclotron. The new nuclides mainly undergo sequential α-decay, which ends with spontaneous fission (SF). The total time of decay ranges from 0.5 ms to ~1 d, depending on the proton and neutron numbers in the synthesized nuclei. The atomic number of the new elements 115 and 113 was confirmed also by an independent radiochemical experiment based on the identification of the neutron-rich isotope ^{268}Db (T_{SF} ~ 30 h), the final product in the chain of α-decays of the odd–odd parent nucleus ^{288}115. The comparison of the decay properties of 29 new nuclides with $Z = 104$–118 and $N = 162$–177 gives evidence of the decisive influence of the structure of superheavy elements on their stability with respect to different modes of radioactive decay. The investigations connected with the search for superheavy elements in Nature are also presented.

The experiments were carried out at the Flerov Laboratory of Nuclear Reactions (JINR, Dubna) in collaboration with the Analytical and Nuclear Chemistry Division of the Lawrence Livermore National Laboratory (USA).

Keywords: superheavy elements; decay properties; α-decay; spontaneous fission; stability.

INTRODUCTION

How many chemical elements could exist?

According to quantum electrodynamics, the well-known concept of an atom (E. Rutherford, 1932) as a system consisting of a nucleus that carries positive charge and practically all the atomic mass, and electrons that move at large distance from the nucleus, is correct up to atomic number Z ~ 170 or even higher. However, the existence of atoms (elements) is limited by the instability of the nucleus itself.

Formally, the limiting mass of a nucleus is determined by the probability of its fission into two (or more) parts of smaller mass. This type of nuclear transmutation—spontaneous fission (SF) into two

*Paper presented at the 40th IUPAC Congress, Beijing, China, 14–19 August 2005. Other presentations are published in this issue, pp. 889–1090.

fragments of approximately equal mass—was observed for the first time for uranium in 1940 by G. N. Flerov and K. A. Petrzhak [1]. For the nucleus ^{238}U, the partial SF half-life appeared to be $T_{SF} = 10^{16}$ years.

N. Bohr and J. Wheeler gave a physical description of the process within a liquid-drop model of nuclear fission [2]. The model is based on an assumption of nuclear matter being a structureless (amorphous) matter similar to a drop of charged liquid. For the nuclear drop, the forces of surface tension that oppose the forces of electric (Coulomb) repulsion of protons, define its spherical shape. The situation changes with increasing the nuclear deformation. As soon as the drop reaches some critical deformation in the process of counteraction of these two forces, it divides into two parts. Evidently, to reach the critical deformation, the initial energy of the nucleus should be increased. The difference of energy of spherical and deformed shapes of the ^{238}U nucleus or the height of its fission barrier is about 6 MeV. Therefore, if one brings an extra energy of about 6 MeV or more in the uranium nucleus, it divides into two fragments. However, the uranium nucleus can divide into parts by itself (spontaneously) by "tunneling" through the fission barrier with a half-life of $T_{SF} = 10^{16}$ years. With the increase of the atomic number, the fission barrier height considerably decreases, which results in strong decrease of the nuclear lifetimes with respect to spontaneous fission; the californium isotope ^{252}Cf ($Z = 98$) already has SF half-life of $T_{SF} \approx 80$ years, and the isotope ^{256}Fm ($Z = 100$) has $T_{SF} \approx 2.9$ h. For the heavier elements, at some critical value of the nuclear charge, when the fission barrier height approaches zero, the nucleus becomes fully unstable to spontaneous fission ($T_{SF} \sim 10^{-19}$ s). This is the limit of the existence of heavy nuclei. According to estimates of N. Bohr and J. Wheeler, this situation appears for the nuclei with $Z > 100$.

However, 22 years later in our laboratory [3] and then in many other laboratories [4], it was found that the nuclei of uranium and transuranium elements can undergo spontaneous fission with two strongly different half-lives. For instance, for ^{238}U, the half-lives are 10^{16} years and $3 \cdot 10^{-7}$ s, the difference reaches an astronomic value—a factor 10^{30}! The two half-life values mean that the decay occurs from two states of the system, i.e., from the "ground state" ($T_{SF1} = 10^{16}$ years) and from the "isomeric" state ($T_{SF2} = 3 \cdot 10^{-7}$ s). The existence of spontaneously fissioning isomers contradicts the liquid-drop model, as no stable intermediate states can appear in deformation of the classic charged drop. Contradictions with the model were observed also in other regularities of SF half-lives. This means that the structure of the nucleus is of great importance in the complex process of nuclear deformation. A more detailed analysis has shown that experimental values of nuclear binding energies differ from calculated values. This difference has a regular character: it reaches a maximum (the highest binding energy) at definite, the so-called "magic" numbers of protons and neutrons. These are called closed proton and neutron shells.

By the end of the 1960s, through the efforts of many theoreticians, the microscopic theory of atomic nuclei was created, and it brought all the above contradictions to a harmonious system of physical regularities. As any theory, it had certain predictive power, in particular, in predicting the properties of very heavy still-unknown nuclei. It appeared that the stabilizing effect of nuclear shells would work beyond the limits set by the liquid-drop nuclear model (in the domain of $Z > 100$) resulting in the so–called "stability islands" around magic numbers $Z = 108$, $N = 162$ and $Z = 114$, $N = 184$. The lifetimes of the superheavy elements that belong to these stability islands can be significantly higher. This is especially relevant to the most heavy, the superheavy elements, in which the effect of the closed shells $Z = 114$ (maybe 120 or 122) and $N = 184$ increases the half-lives up to tens and hundreds of thousand of years, i.e., 30 to 33 orders of magnitude higher than without nuclear shell effects (Fig. 1). Thus, there appeared an intriguing hypothesis of the possible existence of superheavy elements that considerably expanded the borders of the material world. A direct verification of the theoretical predictions would be provided by the synthesis of superheavy nuclides and the determination of their decay properties.

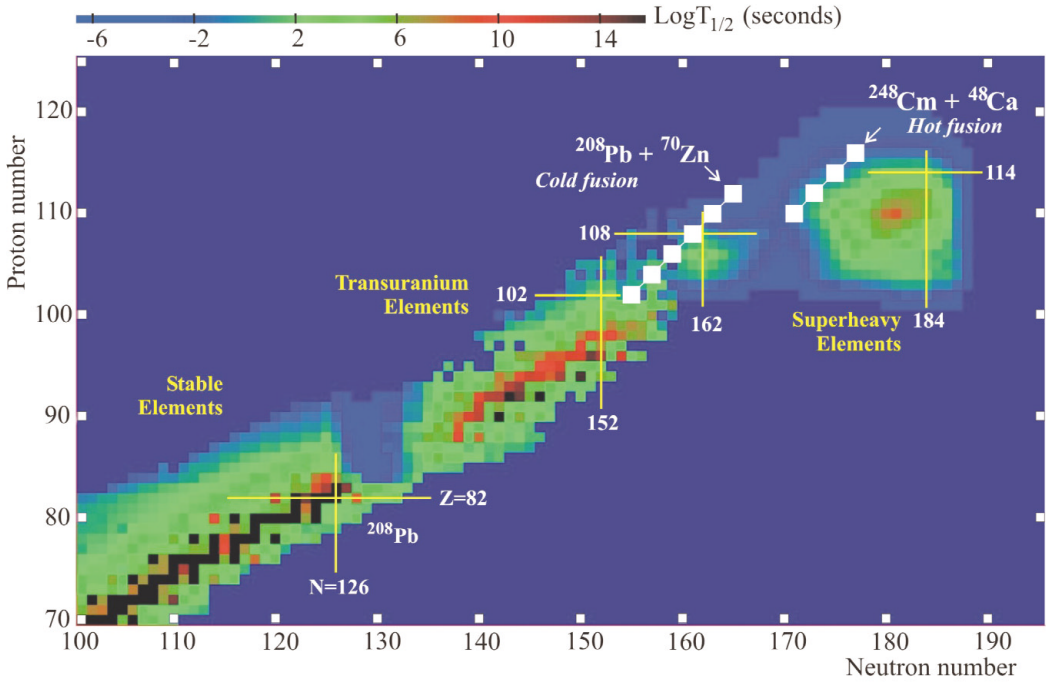

Fig. 1 Chart of the nuclides; the domain of the heavy and superheavy elements. White squares show the chains of the sequential α-decay of nuclei of element 112 produced in the cold-fusion reaction ^{208}Pb + ^{70}Zn → ^{277}112 + n and element 116—in the hot fusion reaction ^{248}Cm + ^{48}Ca → ^{293}116 + 3n. Eight extra neutrons should increase the half-life of ^{277}112 by 4 or 5 orders of magnitude.

REACTIONS OF THE SYNTHESIS OF SUPERHEAVY ELEMENTS

It is well known that the first transuranium elements were synthesized in proton-induced reactions or in sequential capture of neutrons in long-term irradiations in high-flux nuclear reactors. Long lifetimes of the new nuclides allow separation from other reaction products by radiochemical methods and then measurement of their radioactive decay properties. These pioneering works of G. T. Seaborg and colleagues [5] resulted in the discovery of eight artificial elements with Z = 93–100, the heaviest isotope being ^{257}Fm ($T_{1/2}$ ~ 100 days). Advance to the domain of still-heavier nuclei was blocked by the very short half-life of the next Fm isotope ^{258}Fm (T_{SF} = 0.3 ms). Attempts to bypass this limitation by using high-pulse neutron flux from nuclear explosions did not yield the desirable result, as well.

Elements heavier than Fm (Z = 100) were synthesized in the reactions induced by heavy ions, when a complex of nucleons, protons and neutrons, is brought into the target nucleus. This type of reaction is different from the previous case. After capturing a neutron that has no electric charge, the excitation energy of the new nucleus (compound nucleus) is only about 6–8 MeV, while fusion even with light ions like helium (^{4}He) or carbon (^{12}C) leads to heating of the compound nuclei to E_x = 20–40 MeV. With further increase of the atomic number of the projectile, it will need even higher energy to overcome the Coulomb barrier of the reaction. This results in heating of the compound nucleus; its cooling down (transition to the ground state, E_x = 0) goes via emission of neutrons and gamma rays. Here arises the first obstacle.

The heated heavy nucleus emits a neutron roughly in only one case in a hundred, while generally it undergoes fission into two fragments. It is easy to understand that increasing the excitation energy of the heavy nucleus is destructive. The survival probability of the heated nucleus drops drastically with an increase of its temperature (or excitation energy E_x) because of the higher number of neutrons to be

evaporated in strong competition with fission. In order to cool down a nucleus heated to 40–50 MeV, four or five neutrons are to be evaporated; therefore, the survival probability is only 10^{-8}–10^{-10}. The situation is aggravated by the fact that with the increase of nuclear temperature, the stabilizing effect of nuclear shells decreases. Accordingly, the fissionability of the nucleus sharply increases. Both factors lead to exceptionally low probability of formation of superheavy elements.

Advance to the domain of elements heavier than 106 became possible after the discovery in 1974 of the so-called "cold-fusion reactions" [6]. In these reactions, magic nuclei of the stable isotopes ^{208}Pb (Z = 82, N = 126) or ^{209}Bi (Z = 83, N = 126) are used as target material, and are bombarded by ions heavier than argon. In the process of fusion, the high binding energy of nucleons in the magic target nucleus leads to absorption of energy in rearrangement of the two interacting nuclei into a heavy nucleus of the summed mass. This difference in the "packing" energy of nucleons in the interacting nuclei and the resulting nucleus compensate considerably the energy necessary to overcome the high Coulomb barrier of the reaction. As a result, the compound nucleus has excitation energy of only 10–20 MeV. Therefore, in the synthesis of heavy elements in cold-fusion reactions, the heavy nucleus needs to emit only one or two neutrons to reach the ground state. Cold-fusion reactions with massive nuclei were successfully applied for synthesizing six new elements with Z = 107–112 at GSI (Darmstadt) [7]. Recently, the group of RIKEN (Tokyo) repeated the GSI experiments on the synthesis of elements 110–112 and observed two decay events of element 113 [8].

However, with increasing the atomic charge of the projectiles, the probability of their fusion with the ^{208}Pb or ^{209}Bi target nuclei significantly drops because of the increase of the Coulomb repulsion forces that are proportional to the nuclear charges. From element 104, which can be produced in the reaction ^{208}Pb + ^{50}Ti ($Z_1 \cdot Z_2$ = 1804) to element 112 in the reaction ^{208}Pb + ^{70}Zn ($Z_1 \cdot Z_2$ = 2460), the fusion probability decreases by a factor of more than 10^4. There exists yet another limitation.

Compound nuclei produced in cold-fusion reactions have a relatively low number of neutrons. In the above case of the formation of element 112, the resulting nucleus with Z = 112 has only 165 neutrons, while an increase of stability is expected for neutron number N > 170 (Fig. 1). As it will be shown further, a higher neutron number can be reached by using targets of radioactive elements—heavy isotopes of actinides and ions of ^{48}Ca as projectiles.

EXPECTED PROPERTIES

If the theoretical hypothesis is correct, the superheavy nucleus should be stable to spontaneous fission. Thus, it will undergo α-decay, which will lead to the daughter nucleus, which is two protons and two neutrons lighter than the parent one. If the daughter nucleus has low probability of spontaneous fission, another α-decay will follow and then a third one. Alpha-decays will follow until spontaneous fission occurs. As a result, we expect to observe a "radioactive family", i.e., a chain of sequential α-decays, rather long lasting (on nuclear scale) that compete with, but finally are terminated by spontaneous fission. In principle, such a decay scenario already gives evidence of the formation of a very heavy and rather stable nucleus on the stability island.

In order to observe the expected increase of stability in full scale, one has to approach as close as possible to the shells Z = 114 and N = 184. It is very hard to synthesize such neutron-rich nuclei in nuclear reactions, since in the fusion of stable nuclei with definite ratio of protons and neutrons, it is impossible to produce the doubly magic nucleus—^{298}114. Therefore, one has to attempt using nuclei that initially have a maximum neutron number. This was the principal ground of choosing the accelerated ions of ^{48}Ca [9].

The natural calcium is known to consist of 97 % of the isotope ^{40}Ca (Z = 20, N = 20). Yet it contains 0.187 % of the heavier isotope—^{48}Ca (Z = 20, N = 28—both values correspond to closed shells), which has eight extra neutrons. In fusion reactions with ^{48}Ca-projectiles, their magic structure will also work (in cold-fusion reactions, this part was taken by the magic ^{208}Pb target nuclei); the resulting excitation energy will be about 30–35 MeV. The transition of the compound nuclei to the ground state

will occur by emission of three or four neutrons and gamma rays. Upon approaching to the closed shells $Z = 114$ and $N = 184$, the survivability of nuclei will increase because of the increase of the fission barriers. On the other hand, the mass-asymmetry in the input channel of the reaction ($Z_1 \cdot Z_2 \leq 2000$) should decrease the Coulomb repulsion and thus increase the fusion probability.

Despite the advantages that look evident, all previous attempts of synthesizing superheavy elements in ^{48}Ca-induced reactions, undertaken in different laboratories in 1977–1985, gave no result. Only after the increase of the experimental sensitivity by up to three orders of magnitude a factor of 1000, the advantages of this type of reactions for the synthesis of superheavy elements were revealed.

In the experiments that are performed in our laboratory, the beam intensity of the ^{48}Ca ions is about $8 \cdot 10^{12}$/s with the isotope consumption of ~0.5 mg/h. As target material, we use the long-lived enriched isotopes of actinides: Pu, Am, Cm, and Cf with the maximum number of neutrons. Fusion reactions of ^{48}Ca-nuclei with the nuclei of these isotopes were chosen for the synthesis of elements with $Z = 114$–118. Presentation of all the results would take too much time; therefore, we limit ourselves by describing only one experiment on the synthesis of elements 113 and 115. All the other reactions were studied by a similar technique.

EXPERIMENTAL TECHNIQUE

The heavy atom, produced in the fusion of target and projectile nuclei, moves in the beam direction to a detector mounted at a distance of about 4 m from the target. Between the target and the detector, a gas-filled separator is located, which suppresses the beam particles and by-products of the reaction. The working principle of the separator (Fig. 2) is based on the difference of the ionic charges of the atoms moving in the gas media with different velocity; in our case, the media is hydrogen gas at pressure of ≈1 Torr. This allows them to separate "in flight" in a time interval of about 10^{-6} s. Atoms that have passed through the separator are implanted into the sensitive layer of the semiconductor detector, generating signals of the implantation time of the recoil, its energy, and position of implantation. The detector with a total surface of about 50 cm^2 has 12 "strips", each with longitudinal position sensitivity.

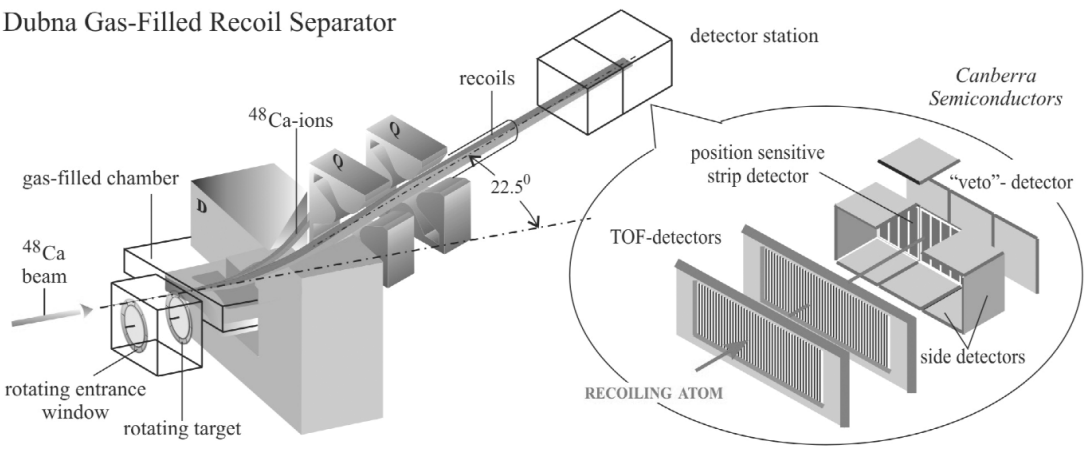

Fig. 2 Set-up used in the experiments on the synthesis of superheavy elements.

If the nucleus of the implanted atom undergoes α-decay, then the detector, with measuring all the above-mentioned parameters, would register the emitted α-particle (with an expected energy of some 10–11 MeV): time, energy, and coordinates. If a second one followed the first decay, then similar in-

formation would be obtained for the second α-particle, and so on, until spontaneous fission occurs. The last decay will be detected as two coinciding in time signals with high amplitude ($E_1 + E_2 \sim 200$ MeV). In order to increase the detection efficiency for the α-particles and pairs of fission fragments, the front detector was surrounded by side detectors, thus composing a "box" open from the side of the separator. Two time-of-flight (TOF) detectors mounted in front of the detector array measure the velocity of the recoils. Thus, the first signal generated by a recoil has a TOF-mark. Subsequent signals from the decay of nuclei do not have such a mark. Decays may be of various length and be characterized by emission of one or a few α-particles of different energy. However, if they originate from the same nucleus and represent a radioactive family (mother–daughter–granddaughter nucleus, etc.), the coordinates of all the signals: of the recoil nucleus, of α-particles, and of fission fragments, should coincide within the position resolution of the detector. Our detectors measure the α-particle energy with accuracy of ~0.5 % and have position resolution of about 0.6 mm.

EXPERIMENTAL RESULTS

Synthesis of elements 113 and 115

(Physical experiment)

Synthesis of odd-Z nuclei is attractive since odd proton or neutron number means significantly lower probability of spontaneous fission, and the number of sequential α-decays will be larger (longer decay chains) than in the decay of even–even nuclei. To overcome the Coulomb barrier in the reaction ^{48}Ca + ^{243}Am, the ^{48}Ca ions should have energy $E \geq 236$ MeV. On the other hand, if one limits the beam energy by 248 MeV, the thermal energy of the compound nucleus ^{291}115 will be about 40 MeV; it will be cooled down via emission of three neutrons and gamma rays. Then the reaction product will be the isotope of element 115 with neutron number $N = 173$. After emerging from the target layer, the atom of the new element in about 1 μs traverses the separator, which is accordingly tuned, and reaches the detector. Note that information on registering the recoil nucleus has a TOF-mark, which means that the nucleus has arrived from the separator. If, in some time, another signal arrived from the same position on the detector's surface, but without a TOF-mark (i.e., from the decay of the implanted atom), the beam was switched off and all the subsequent decays were registered with practically zero background. In the experiment ^{48}Ca + ^{243}Am, during ~20 s after switching the beam off, four other signals were detected. Their positions in the given strip coincided with the preceding signals within less than 0.7 mm. Spontaneous fission, in the same strip and in the same position, was detected only on the next day, 28.7 h later, as two signals from fission fragments with the sum energy of 205 MeV (Fig. 3a).

Such chains were detected three times [10]. All of them look the same (six generations of nuclei in a radioactive family) and agree with each other both in α-decay energies and in decay times, taking into account the exponential decay law of nuclei. If the observed effect is related, as expected, to the decay of the isotope of element 115 with mass 288, which is formed after emission of three neutrons from the compound nucleus, increasing the energy of the ^{48}Ca beam by only 5 MeV should decrease it by a factor of 5–6. Indeed, at the energy of 253 MeV the effect was not observed. However, here a new shorter decay chain was found; this one included four α-particles (we consider that indeed, there were five of them, but the last α-particle escaped through the open part of the detector array) and lasted for only 0.4 s. The sequential emission of α-particles ended ~1.8 h later in spontaneous fission. It is evidently the decay of a different nucleus, quite probably of the neighboring isotope of element 115 with mass 287, produced in the fusion reaction with emission of four neutrons. The decay of the nucleus with $Z = 115$, $N = 173$ is shown in Fig. 3b, where the contour plot presents the calculated half-lives of the superheavy nuclides with different proton and neutron number. Here is also shown the decay of the lighter isotope of element 111 with neutron number $N = 161$, which was synthesized in the reaction ^{209}Bi + ^{64}Ni [12,13].

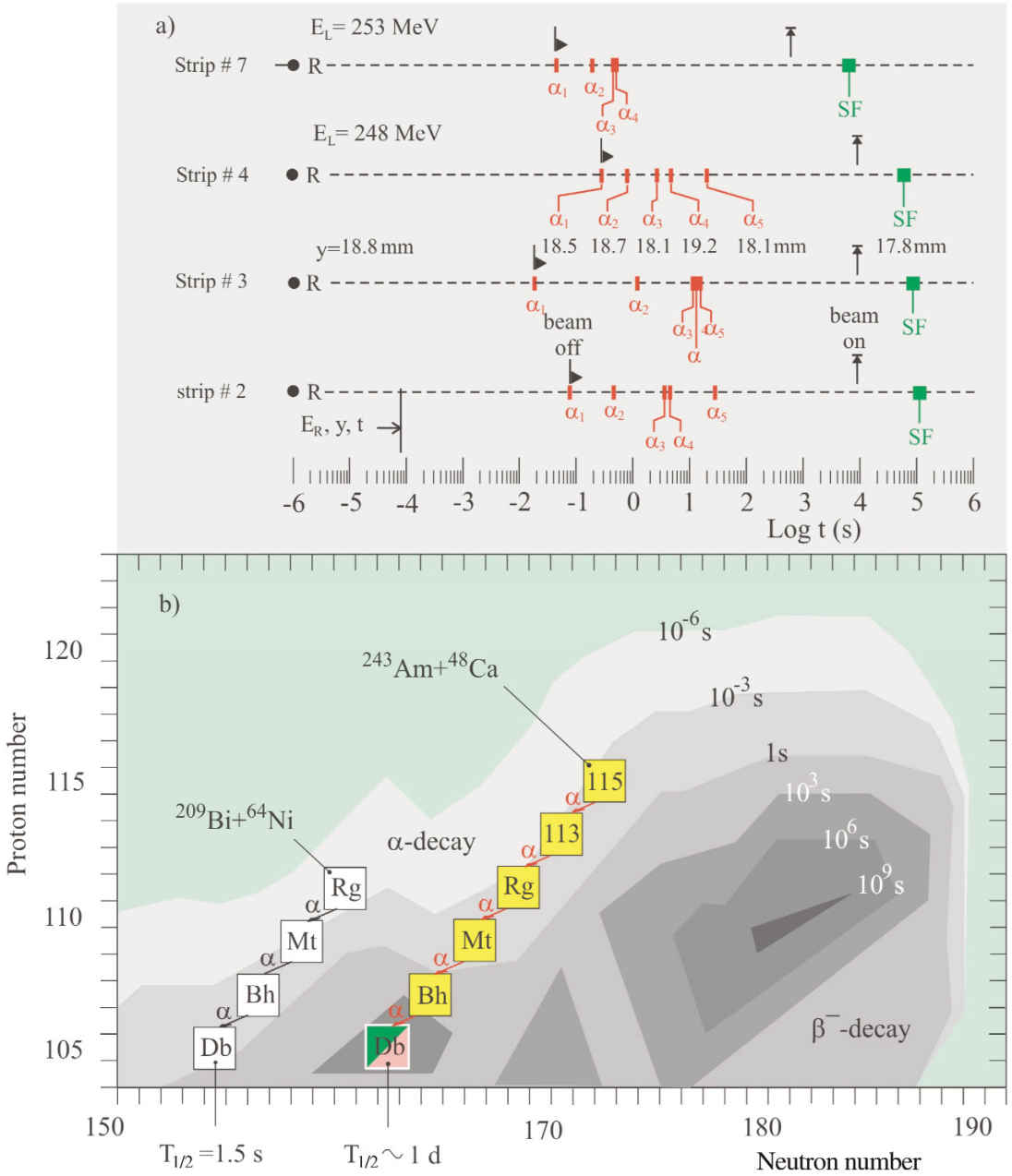

Fig. 3 Detector signals originating from implantation and sequential decay of nuclei of element 115. Decay chains of the nuclei of elements 111 and 115, synthesized in the reactions ^{209}Bi + ^{64}Ni and ^{243}Am + ^{48}Ca, respectively. The contour plot presents nuclear half-lives, calculated within the framework of microscopic theory [11].

First, one should note that the decay energies and times in both cases agree well with theoretical predictions. Despite the fact that the isotope ^{288}115 is 11 neutrons far from $N = 184$, the isotopes of elements 113 and 115 have relatively long lifetimes ($T_{1/2}$ ~ 0.1 and 0.5 s, respectively).

Following five α-decays, an isotope of element 105 dubnium (Db) is formed with $N = 163$, whose stability is determined by the closed neutron shell $N = 162$. The strength of this shell is demonstrated by the great difference (~10^5) in half-lives of the two Db isotopes that differ by only eight neutrons.

(Chemical experiment)

In the above instance, the properties of the long-lived isotope ^{268}Db that terminates the decay chain of element 115 are interesting by themselves.

Because of the long half-life, atoms of element 105 can be separated by the classic *off-line radiochemical technique* of ion-exchange chromatography with subsequent measurement of their decay—spontaneous fission. This experiment gives independent and unambiguous identification of the atomic number of the final nucleus ($Z = 105$) and of all the nuclides formed in correlated α-decays of element 115 [14].

According to the Periodic Table of the Elements, an atom with $Z = 105$ belongs to Group V and is the chemical homolog of niobium (Nb) and tantalum (Ta).

The chemical properties of Db are well known, these were studied in detail with short-lived isotopes $^{261–263}$Db in [15–17], using on-line express techniques. In our experiment, reaction products were knocked into a copper catcher. After a 20 ÷ 30-h irradiation, the catcher was dissolved and the fraction of transactinides—elements with $Z \geq 104$—was separated from this solution, and then, from this fraction elements of Group V, Db together with its chemical homologs, Nb and Ta, was separated. The products of the chemical separation were deposited in a thin layer on a 0.4-μm Mylar backing that was put between two semiconductor detectors in order to register both fragments of spontaneous fission. The whole array, in turn, was positioned into a neutron detector, to determine the number of neutrons emitted in spontaneous fission of Db nuclei. In eight identical experimental runs, we detected 15 events of spontaneous fission of Db ($T_{1/2} = 1.2^{+4}_{-2}$ d). The sum kinetic energy of the fragments is about 230 MeV; at average, about four neutrons are emitted per fission. Such characteristics could be expected for spontaneous fission of nuclei with $Z = 104–105$, $N = 163–164$.

The chemical run supports the results of the physical experiment: the nuclei of element 115, produced in the reaction ^{243}Am + ^{48}Ca via five sequential α-decays, 115 → 113 → 111 → 109 → 107 → 105, indeed, led to the formation of the long-lived spontaneously fissioning nucleus with atomic number 105 (Fig. 4). In these experiments, another previously unknown element with atomic number 113 was also synthesized, as the α-decay daughter of element 115.

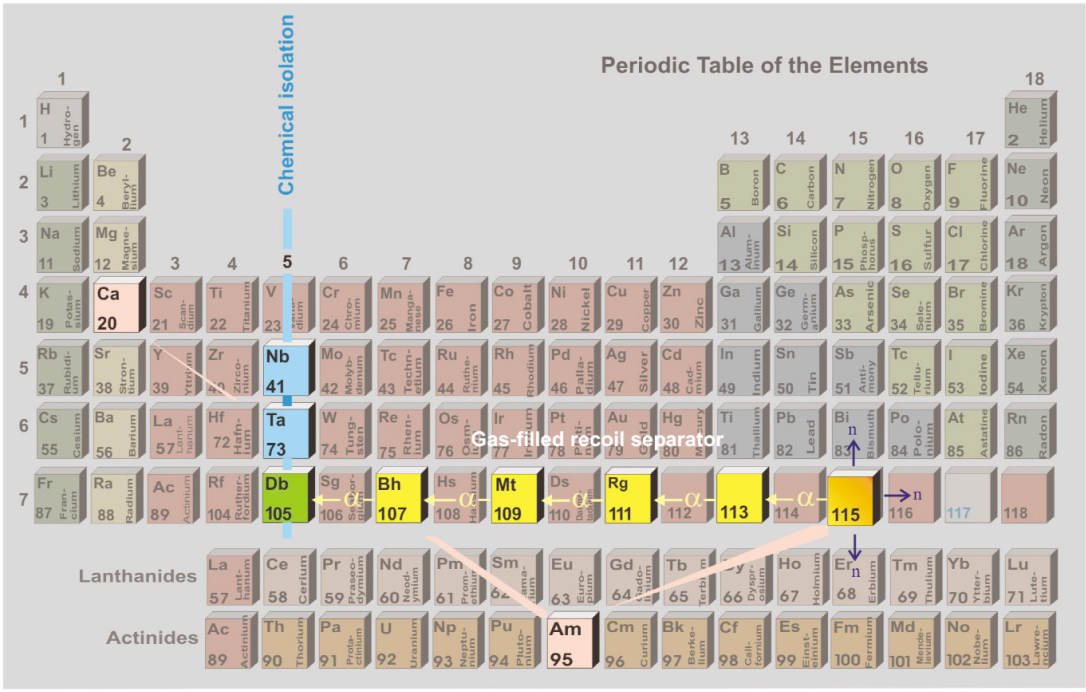

Fig. 4 Synthesis of elements 113 and 115.

Even-Z neutron-rich isotopes of elements 104–116

Results obtained in the reaction ^{243}Am + ^{48}Ca are not an isolated observation. When synthesizing even-Z nuclei—the isotopes of elements 112, 114, and 116—we also observed lengthy decay chains ending in spontaneous fission of nuclei with Z = 104–114. Half-lives of the new nuclides ranged from milliseconds to hours, depending on the atomic number and number of neutrons in the nuclei [18–21]. In the near future, we intend to run an on-line experiment to study the chemical properties of relatively long-lived isotopes of elements 112 and 114, produced in the reactions ^{238}U, ^{242}Pu + ^{48}Ca.

However, even-Z nuclei can be identified in a different way. In their synthesis reactions we can use as targets various isotopes of U, Pu, Cm, and Cf. The nucleus of interest may be produced from compound nuclei with $Z = Z_{CN}$, but with different mass A_{CN} (depending on the mass of the target nucleus), which emit correspondingly a different number of neutrons—x.

The same result may be obtained for a given compound nucleus Z_{CN}, A_{CN} (with fixed target mass), by accurate variation of the ^{48}Ca beam energy that leads to varying of the number of evaporated neutrons. Finally, a given nucleus may be synthesized either directly as a product of neutron evaporation with $Z = Z_{CN}$ and $A = A_{CN}$–x, or resulting from one or more α-decays of heavier nuclei with Z + 2, A + 4 or Z + 4, A + 8, etc. We used all three ways to identify isotopes with Z = 112, 114, and 116. For instance, in Fig. 5 we exemplify various ways of synthesizing the isotope ^{287}114 that was produced in three different reactions. All the 19 chains of decay of the nuclide ^{287}114, detected in these experiments, agree in α-decay energies (Q_α) and half-lives (T_α and T_{SF}).

Below, we present another example that refers to our recent work—synthesis and study of decay properties of the heaviest nuclide, the isotope of element 118 with mass 294. These results are presented for the first time.

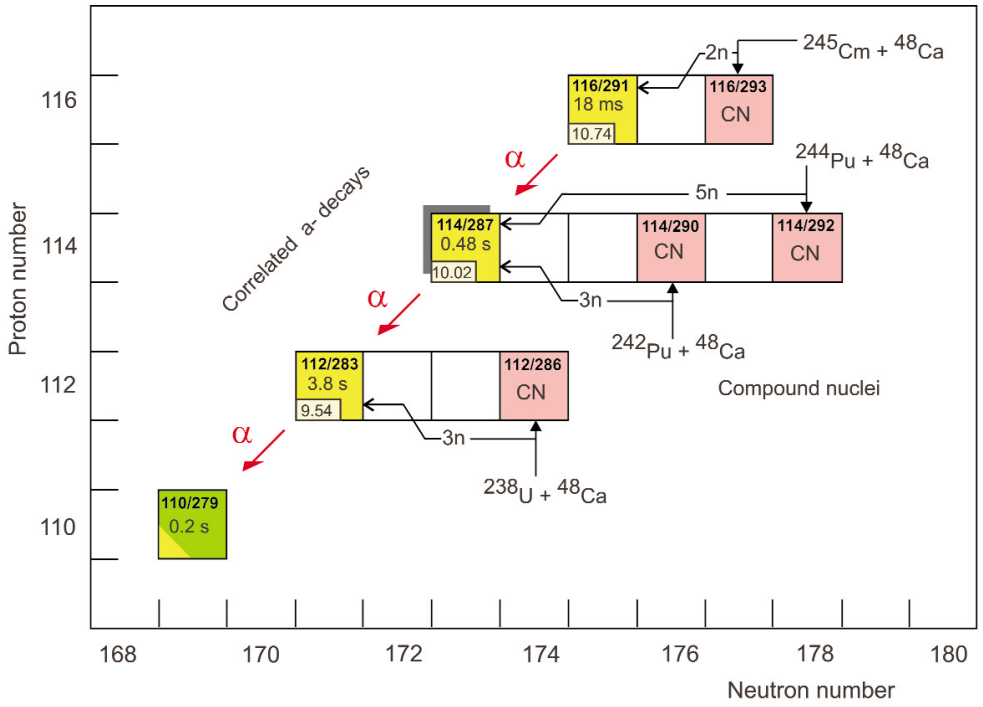

Fig. 5 The isotope $^{287}114$ produced in various fusion reactions with ^{48}Ca.

Synthesis of element 118

In spring 2002, when running the experiment with the reaction $^{48}Ca + ^{249}Cf$, with high beam dose $2.5 \cdot 10^{19}$ ions of ^{48}Ca (the experiment lasted about four months), we detected a single event of production of a superheavy nucleus, that followed a decay pattern: $R \rightarrow \alpha_1 \rightarrow \alpha_2 \rightarrow SF$, with energies and decay times expected for the even–even isotope $^{294}118$ [22]. In the following three years, we synthesized a series of isotopes of elements 112, 114, and 116 and determined their decay properties. These included the isotopes $^{290,291}116$, $^{286,287}114$, and $^{282,283}112$ (the daughter products of sequential α-decays of the mother nuclei $^{295}118$ and $^{294}118$), which could be produced in the most probable reaction channels with the emission of two and three neutrons, respectively. From this data, one could construct the decay scenario of the α-radioactive isotopes $^{294}118$, $^{295}118$ and determine the decay characteristics of their daughter nuclei (presented in Fig. 6a). From the decay properties of the isotopes $^{291}116$ and $^{290}116$, it followed that the above-mentioned single event, observed in the reaction $^{48}Ca + ^{249}Cf$, refers to the formation and decay of $^{294}118$.

In February 2005, we started, with higher sensitivity, the second experiment $^{48}Ca + ^{249}Cf$, which was finished in March. Here, with the beam dose of $1.6 \cdot 10^{19}$, we detected two more decay events of the isotope $^{294}118$ that completely agreed with the expected energies and half-lives of the daughter nuclei: $^{290}116(\alpha) \rightarrow ^{286}114(SF/\alpha) \rightarrow ^{282}112(SF)$. Finally, in the reaction $^{48}Ca + ^{245}Cm$, we measured the dependencies of the production cross-sections of the isotopes of element 116 on the ^{48}Ca beam energy (excitation functions). From this experiment, it also follows that the daughter nuclide of the α-decay of nuclei with $Z = 118$ is the isotope of element 116 with mass 290 that was produced in the 3n-evaporation channel of the reaction $^{48}Ca + ^{245}Cm$.

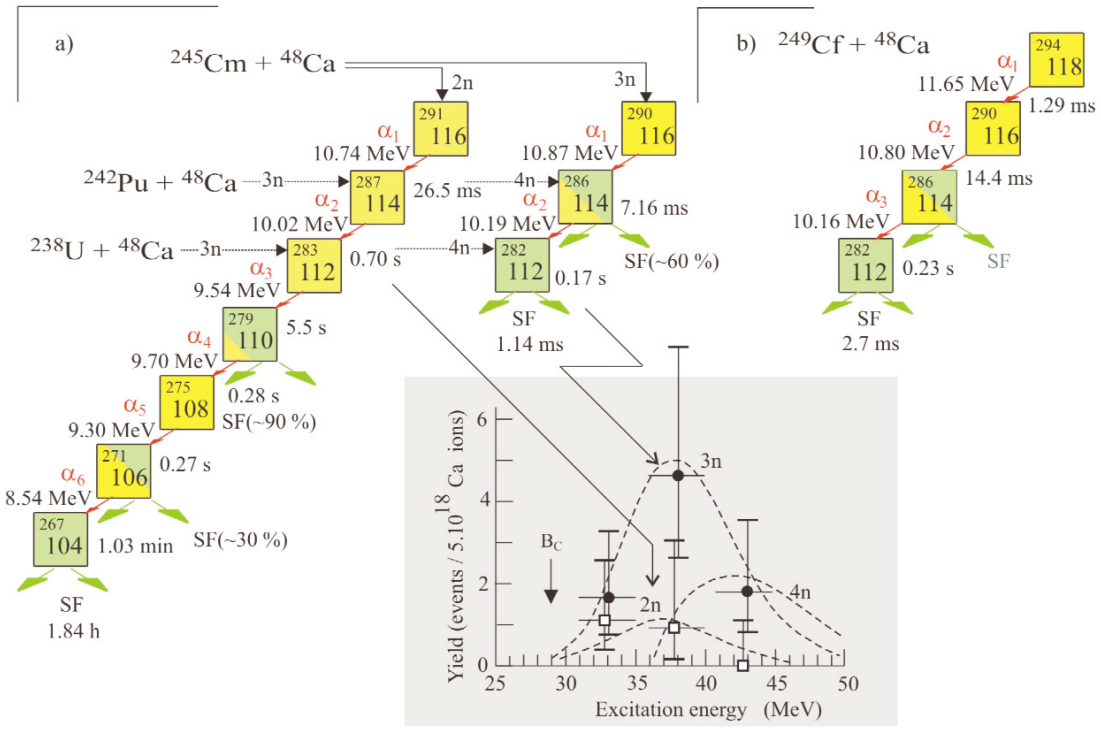

Fig. 6 (a) Decay chains of the isotopes of element 116, synthesized in 2n- and 3n-evaporation channels in the reaction ^{245}Cm + ^{48}Ca. (b) Decay of the nuclei of element 118, synthesized in the reaction ^{249}Cf + ^{48}Ca.

Now, summing the data of the two experiments with the reaction ^{48}Ca + ^{249}Cf and the decay properties of the daughter products—the isotopes ^{290}116, ^{286}114, and ^{282}112—which were synthesized in the reactions ^{238}U, ^{242}Pu and ^{245}Cm + ^{48}Ca, we can show the decay properties of the heaviest nuclide ^{294}118 (Fig. 6b).

By now, the data on the decay properties of 29 new nuclei with Z = 104–118 have become available; these are presented in the chart of nuclides (Fig. 7).

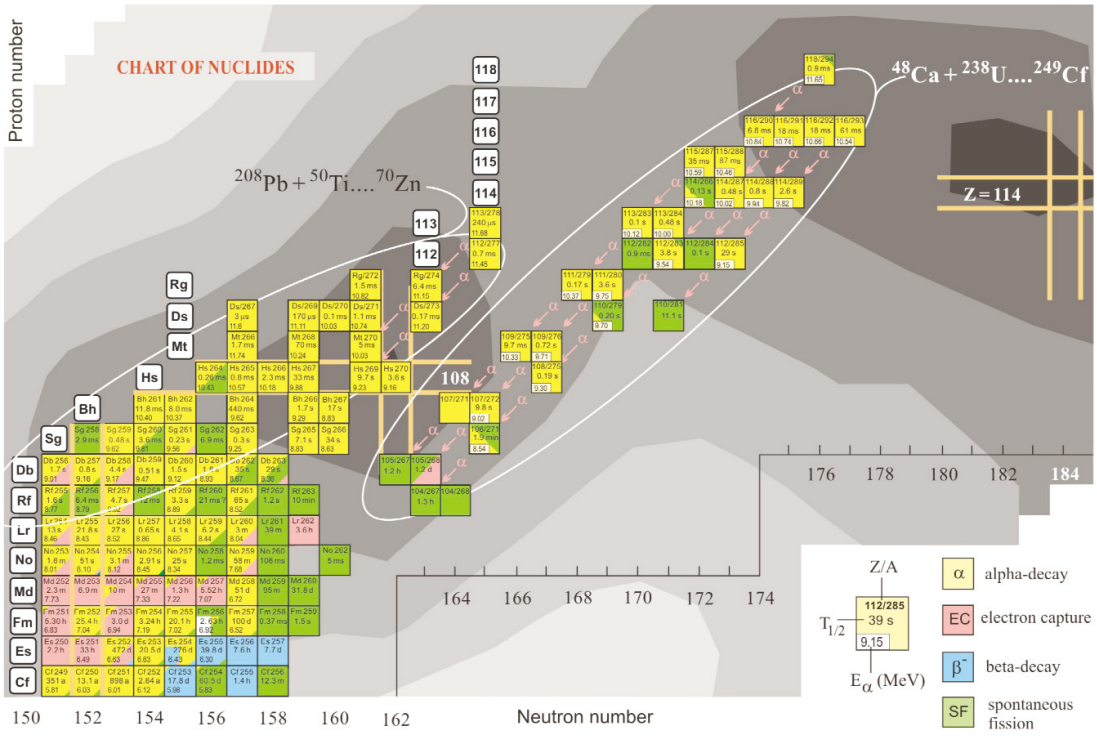

Fig. 7 Chart of the heavy nuclides with Z ≥ 98. Isotopes, produced in cold-fusion reactions with ^{208}Pb and ^{209}Bi-targets, are shown inside the oval-shaped white line on the left, while those synthesized in hot-fusion reactions with ^{48}Ca projectiles are shown inside the white line on the right. The half-lives are given for the dominant decay mode. Darker colors of the areas of the contour plot represent the strength of the nuclear shell effect, which determines the stability of heavy nuclei against spontaneous fission. In the vicinity of closed shells (dark areas), the nuclei undergo α-decay (yellow squares). As the daughter gets further away from the magic numbers Z = 108, N = 162 and Z = 114, N = 184 spontaneous fission becomes more probable. At the boundaries of the stability islands, a nuclide may undergo both decay modes. For odd–odd nuclei—such as the isotope ^{288}115 and its daughter products with Z = 113, 111, 109 and 107—α-decay dominates down to the isotope Db-268 (Z = 105), which undergoes spontaneous fission after about one day.

The general outlook and the future

The properties of the heaviest nuclei that belong to this domain of transactinides, their decay modes, energies, and decay times, as a whole, are in good agreement with the predictions of modern theory. It looks as if the hypothesized existence of the stability islands of superheavy nuclei that considerably widen the element world for the first time has its experimental confirmation.

Prospects

Now, the task is to study in more detail the nuclear and atomic structure of the new elements, which represents a considerable problem, first, because of the low yield of the reaction products under study. In order to increase the number of atoms of the superheavy elements, one needs to increase the intensity of the ^{48}Ca beam and to improve the efficiency of the experimental technique. Modernization of the heavy-ion accelerator that is planned for the nearest years, using all the recent achievements in accelerator technology, could result in an increase of the beam intensity by a factor of about five. Another part of the problem needs a cardinal change in experimental technique; the solution can be found in developing new experimental methods, based on the properties of superheavy elements.

The operation principle of the employed device—the separator of recoil nuclei (Fig. 2)—is based on the difference in kinematics of various types of reaction products. The products of fusion of target nuclei and ^{48}Ca that are of interest for us are knocked forward from the target in a narrow cone (±3°) with a kinetic energy of about 40 MeV. By limiting the trajectories of recoiling nuclei, with all those parameters taken into account, we practically completely eliminate the ion beam, suppress the background from reaction by-products $10^4 \div 10^6$ times, and deliver the atoms of new elements to the detector in 1 μs with an efficiency of 30–40 %. In other words, the separation of the reaction products occurs "in flight".

Yet, here we need to use target layers not thicker than 0.2 μm, which is some three times less than needed for obtaining efficient yield of the superheavy nucleus of a given mass and five to six times less, if one considers synthesis of two isotopes of this element with neighboring masses. In order to obtain the data on mass numbers of the isotopes of the superheavy element, a lengthy and time-consuming series of experiments is needed—the measurements should be repeated at various energies of the ^{48}Ca ions.

At the same time, as our previous experiments have shown, the synthesized atoms of superheavy elements have half-lives that considerably exceed the operation time of the recoil separator. Therefore, in many cases there is no necessity to separate the reaction products in such a short time. Then, one could change the operation principle of the device and attempt the separation of reaction products with an "on-line" mass-separator. Being inferior to the recoil separator in rapidity, the new device—mass analyzer of superheavy atoms (MASHA)—will increase the efficiency 10 times, and for the nuclides with $T_{1/2} \geq 1$ s (depending on temperature regimes and the physical–chemical properties of the atoms under study) will provide direct mass measurement of the superheavy nuclei, together with determining their decay properties [23].

Search for superheavy elements in Nature

Another aspect of the problem of superheavy elements concerns obtaining even longer-lived elements. In the above-presented experiments, we reached just the edge of the "island" and noticed a steep ascent, but we are far from its peak, where nuclei might live for thousands or maybe millions of years. We lack neutrons in the synthesized nuclei, to approach the shell $N = 184$. Nowadays it is unreachable—there are no reactions to produce such neutron-rich nuclides. Probably in the far future, physicists will manage to use intense beams of radioactive ions with neutron number more than in ^{48}Ca. Such projects are widely discussed, without considering the costs of constructing those accelerator giants. Yet, one could try a different approach to this problem.

Assuming that the most long-lived superheavy nuclei have half-lives of $10^5 \div 10^6$ years (this is not far from theoretical predictions characterized by certain ambiguities), one cannot exclude that they could be found in cosmic rays—the witnesses of synthesis of elements on younger planets of the Universe. In a stronger assumption that the half-lives of the "long-livers" reach tens of millions of years or more, they might be present in tiny amounts in Earth objects. Among the possible candidates, we give preference to the isotopes of element 108 (Hs) with neutron number around 180. Chemical experiments with the short-lived isotope ^{269}Hs ($T_{1/2} \sim 10$ s) have shown that element 108, as expected, is the chemical homolog of element 76—osmium (Os) [24]. Then, a sample of metallic Os could contain in very small amounts element 108, Eka(Os). The presence of Eka(Os) in Os could be determined by its radioactive decay. Probably the superheavy long-liver could undergo spontaneous fission itself, or spontaneous fission of a lighter and shorter-lived daughter or granddaughter could occur after the preceding α- or β-decays. Thus, at the first stage, one could search for rare spontaneous fission events in the Os sample. We have started such an experiment, and it will go for 1–1.5 years. The decay of Eka(Os) isotopes will be registered by detecting a neutron blast that accompanies spontaneous fission. In order to protect the detection equipment from the neutron background generated by cosmic rays, the measurements are carried out in the underground laboratory (Modane, France), that is located at a depth corresponding to 4000 m in water equivalent. Detecting a single event of spontaneous fission during a pe-

riod of one year would correspond to the concentration of $5 \cdot 10^{-15}$ g/g of element 108 in Os, assuming its half-life is 10^9 years. This small value corresponds to 10^{-16} of the concentration of uranium in the crust of the Earth.

Despite the extreme sensitivity of the experiment, the chances of discovering the relic superheavy elements are very small. The absence of the effect would give the upper limit of the half-life of the long-liver at the level of $T_{1/2} \leq 5 \cdot 10^7$ years.

CONCLUSION AND ACKNOWLEDGMENTS

Experiments on the synthesis of new elements became fruitful owing to the considerable advances in accelerator techniques, in the physical and chemical methods of separation, and in achieving high sensitivity in the detection of rare decay events of superheavy nuclei. The results of the six years of work presented in this talk are just the first step in the problem of synthesis and study of the properties of the new chemical elements. However, they open a vast field for further investigation of the fundamental properties of the nuclides at the very limit of the existence of chemical elements.

The experiments were performed at the heavy-ion accelerator of the Laboratory of Nuclear Reactions of the Joint Institute for Nuclear Research (JINR, Dubna), in collaboration with the Analytical and Nuclear Chemistry Division of the Lawrence Livermore National Laboratory (LLNL, Livermore). It is my debt to express my deep gratitude to my colleagues and coauthors of most of the publications for their creative work and great contribution to solving this task.

In our experiments, we used unique target materials—the isotopes of Pu, Am, Cm, and Cf produced in high-flux nuclear reactors in Dimitrovgrad (Russia) and Oak Ridge (USA), and enriched at the S-2 mass-separator, Sarov (Russia). We are indebted to our colleagues for their efforts.

This work was performed with the support of the Russian Ministry of Atomic Energy and grants from the Russian Foundation for Basic Research Nos. 01-02-16486, 03-02-06236, and 04-02-17186. Much of support for the LLNL collaborators was provided through the U.S. DOE under contract No. W-7405-Eng-48.

Finally, I would also like to thank the Program Committee for the invitation to give this talk.

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