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Synthesis and decay properties of superheavy atoms in nuclear reactions induced by stable and radioactive ion beams

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Abstract. This talk consists of two parts. The first one presents the results of investigations performed in 1998-2000 in Dubna on the synthesis of superheavy nuclei in reactions induced by ^{48}Ca ions. The radioactive decay properties of the nuclei, indicating a considerable increase in the α-decay and spontaneous fission half-lives of the isotopes of elements 110–116 when approaching the closed neutron shell at $N = 184$, are given. In the second part the possible ways of advancing into the region of more neutron-rich nuclei, using stable and radioactive ion beams, are discussed. Since so far no intense radioactive ion beams are available, some experiments with stable beams are considered as a test for the suggested ideas.

PACS. 25.70.Jj Fusion and fusion-fission reactions – 25.70.Gh Compound nucleus – 25.85.Ca Spontaneous fission – 23.60.+e Alpha decay

During the last three years in Dubna fusion reactions induced by ^{48}Ca ions have been investigated. The very rare isotope ^{48}Ca was chosen among others for several reasons.

In order to synthesize superheavy elements, lying close to the neutron shell $N = 184$ and expected according to theoretical predictions to be highly stable, it is necessary to provide for a large neutron excess in the target as well as in the projectile nucleus. In general, as targets one can use the heaviest isotopes of the artificial elements ^{244}Pu, ^{248}Cm that are accumulated in high-flux reactors in perceptible quantities. The largest neutron excess for the projectile is reached in the isotope ^{48}Ca ($N-Z = 8$). Besides, the ^{48}Ca nucleus has closed neutron and proton shells. Due to the fusion with a deformed target-nucleus there is a gain in the Q-value of the reaction; at the Coulomb barrier the produced compound nucleus has excitation energy $E_x^{\min} \sim 30$ MeV. This value is about 20 MeV lower than the one obtained in reactions of hot fusion, which we have formerly used for the synthesis of isotopes with $Z = 106$, 108 and 110. Assuming that the compound nucleus deexcites by emission of 3 or 4 neutrons and γ-rays, it may be expected that its structure (*i.e.* the shell effects) still manifests itself in the energy interval $E_x = 10$–30 MeV. This should increase the survival probability of the heavy nucleus during its transition to the ground state. In addition, the relatively low neutron binding energy ($B_n \sim 5.7$–7.4 MeV) in the neutron-rich nucleus considerably increases the probability of neutron evaporation.

In spite of these obvious advantages, the expected cross-sections of evaporation residues (EVR) in reactions of the type ^{244}Pu, ^{248}Cm + ^{48}Ca remain rather low. The studies that we have carried out of the fission of superheavy nuclei, produced in the ^{244}Pu, ^{248}Cm + ^{48}Ca reactions, have shown that the main mechanism of formation of fission fragments is connected to the quasifission process, which proceeds without the formation of a compound nucleus [1]. As can be seen in fig. 1 only a small part of the fission fragments in the region of symmetric masses can be attributed to the fission of the compound nucleus. This region of the fission fragment mass distribution has its own structure, typical for the fission of actinide nuclei, but differing in that the doubly magic ^{132}Sn nucleus is in this case in the light-fragments group.

Assuming the symmetric part to be the result of the fission of a superheavy compound nucleus and calculating its survival probability at different excitation energies, one can come to the conclusion that the expected cross-section for EVR is of the order of picobarns, perhaps even parts of a picobarn.

The cross-section $\sigma_{\rm EVR}$ strongly depends on the height of the fission barrier of the superheavy nucleus. The general conclusion is very simple: the more stable the nuclei are, the better they survive. From this it also follows that negative results in the previous attempts to synthesize new heavy elements in ^{48}Ca-induced reactions were due to the low sensitivity of the experiments.

Coming back to this problem about 15 years later, it was of major importance to increase the sensitivity of the experiments by at least 2-3 orders of magnitude.

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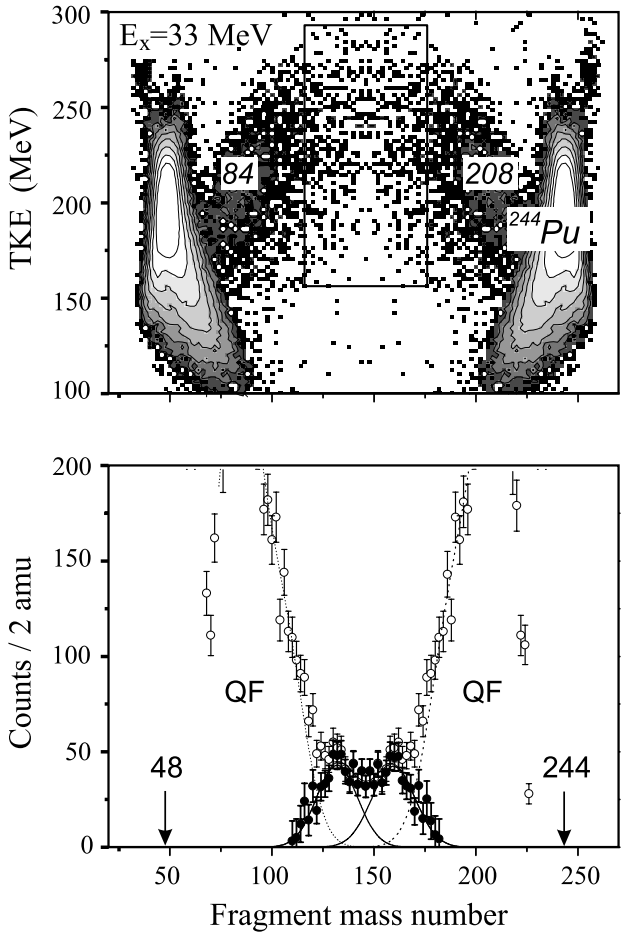

Fig. 1. Correlated fragments from the ^{48}Ca + ^{244}Pu reaction at $E_x = 33$ MeV [1]. The upper panel is a two-dimensional plot of the fragment total kinetic energy *vs.* the fragment mass. The lower panel presents the fission fragment mass distribution: the black symbols denote the symmetric part, the open symbols — quasifission events.

This was mainly achieved by the development of the accelerator techniques: at the heavy-ion cyclotron U-400 at Dubna a beam of ^{48}Ca-ions was accelerated with an intensity of about 0.7 pμA at a material consumption rate of about 0.3 mg/h.

The experiments were carried out at the kinematical separators VASSILISSA (an analogue of the set-up SHIP in GSI) and the DGFRS (the Dubna Gas Filled Recoil Separator). A detector array was installed in the focal plane of the separators, registering the time of flight of the recoil nuclei and their sequential decay by time and position correlation measurements. For a beam dose of $\sim 10^{19}$, transmission of the separators 30–40% and detection efficiency of 80–90% , the observation of one event would correspond to the formation and decay of a superheavy atom with a cross-section of ∼0.5 pb. As targets the enriched isotopes ^{238}U, ^{242,244}Pu and ^{248}Cm were used. Combined with the ^{48}Ca-ions they led to the formation of isotopes of elements with $Z = 112$, 114 and 116. In these reactions the nuclei with $Z = 112$ and 114 could be produced directly as evaporation residues in 3n- and 4n-evaporation channels, as well as daughter products of the α-decay of a heavier nucleus. In such an approach additional information could be obtained on the correlated decays in the new radioactive families.

In total, three experiments were carried out with a total dose of $2.2{\cdot}10^{19}$ ions. A detailed description of the experiments, the data analysis and their interpretation are given in the original publications [2–5]. Here we shall present only a short account of the results.

In the ^{48}Ca + ^{238}U reaction at $E_x = 33$ MeV and with a cross-section of about 5 pb we observed new spontaneously fissioning nuclei with $T_{\mathrm{s.f.}} = 1.4^{+3.3}_{-0.6}$ min. At a higher excitation energy there was no effect; only the upper limit of the cross-section was determined as $\sigma_{\mathrm{EVR}} \leqslant 7$ pb [2]. In the experiment with the ^{242}Pu-target two decay chains α-SF were detected. The decay of the nucleus by emission of an α-particle with an energy of 10.29 MeV and the following spontaneous fission were strictly correlated in the position-sensitive detector. From the analysis of the data it followed that in going from ^{238}U to ^{242}Pu a spontaneously fissioning nuclide was formed with $T_{\mathrm{s.f.}} = 3^{+3}_{-1}$ min as a daughter nucleus after the α-decay of the parent nucleus. Since the two experiments were performed in a narrow excitation energy range $E_x = 31$–33 MeV and without any background from spontaneous fission, most probably the experimentally observed α-SF decay chains are connected with the decay of the parent nucleus ^{287}114, formed in the 3n-evaporation channel with a cross-section of about 2 pb [3].

More neutron-rich nuclei were produced in the ^{48}Ca + ^{244}Pu reaction. In this reaction at a beam dose of $2.0 \cdot 10^{19}$ ions two identical sequences were observed, each consisting of 2 consequent α-decays and spontaneous fission (fig. 2a). The total decay time was about 0.5 min. In these sequences all measured parameters were in good agreement within the limits of position and energy resolution of the detector array and the time distribution of the decays [4].

The excitation energy of the compound nucleus at which the mentioned decay sequences were registered was about $E_x = 36$ MeV, a value close to the maximum of the cross-section for the 4n-evaporation channel.

In the same experiment one more long decay chain was also observed (lasting about 35 min), consisting of 3 consequent α-decays and spontaneous fission [5]. It could be attributed to the decay of the neighbouring odd isotope, formed in the 3n-evaporation channel (the 5n-channel being excluded by the projectile energy, corresponding to $E_x \leqslant 39$ MeV).

If the identification of the nuclides —isotopes of element 114— is correct, then in the reaction ^{48}Ca + ^{248}Cm after the decay of the isotopes of element 116 with mass numbers 292 and 293, formed in the fusion reaction with 4n- and 3n-evaporation, decay of the daughter nuclei (the isotopes of element 114) should be also observed in the experiment with the same characteristics as in the ^{48}Ca + ^{244}Pu reaction.

In order to improve the experimental conditions, the algorithm of the measurements was changed. After the

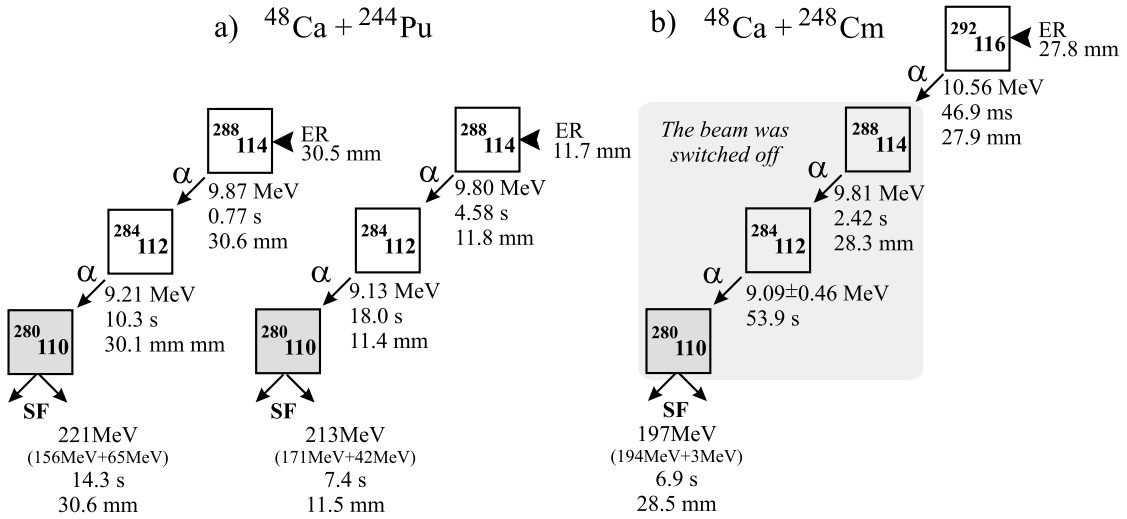

Fig. 2. Decay sequences registered by the detector array in the focal plane of the gas-filled recoil separator: a) the reaction $^{48}Ca + ^{244}Pu$; b) the reaction $^{48}Ca + ^{248}Cm$. For each event the particle energy, time and position in the strip are shown.

implantation of the heavy nucleus into the detector and the emission of an α-particle with an energy from 10.25 to 11.5 MeV (both signals should be correlated in time and position), the accelerator was switched off. The entire decay that followed (in this case, the decay of isotopes of element 114) took place in "background-free" conditions. Such an event occurred on July 19, on the 35th day of the experiment [6]. Following the emission of the 10.56 MeV α-particle 47 ms after the registration of the recoil nucleus by the detector, the beam was switched off and, then, during the next 60 min the detector array registered the sequential decay. A sequence α-α-SF, whose parameters (times and decay energies) completely agree with those formerly obtained in the decay of the nucleus $^{288}114$, was observed in the same strip and position window, where the recoil nucleus and the first α-particle were registered. The overall decay time amounted to about 1 min (fig. 2b).

It is noteworthy that in this reaction the background due to spontaneous fission fragments (the decay of $^{252,254}Cf$) is $7 \cdot 10^{-7}\,\mathrm{min}^{-1}$; the background due to α-particles in the interval $8.0 \leqslant E_\alpha \leqslant 11$ MeV is not more than $6 \cdot 10^{-5}\,\mathrm{min}^{-1}$. At such a low rate, the probability for the observed correlation to be a random coincidence of signals imitating the decay chain can be totally excluded. Therefore, the α-decay with $E_\alpha = 10.56$ MeV should be attributed to the isotope of element 116 with mass number 292.

In the same experiment a long decay chain was observed (lasting 46 min), consisting of three consequent α-decays and spontaneous fission. It agreed well, for what concerns energies and decay times, with the long decay sequence in the reaction $^{48}Ca + ^{244}Pu$. Unfortunately, in this sequence the first α-particle from the decay of the odd nucleus $^{293}116$ was missing and the only observable was the consequent decay of the daughter nucleus. Because of the absence of the first α-particle, such a picture was considered only as an indication of the formation and decay

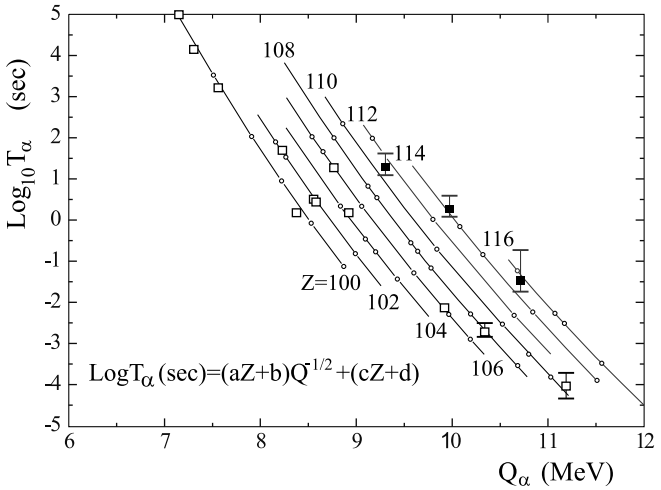

Fig. 3. $\mathrm{Log}_{10}T_\alpha$ *vs.* Q_α for the even-even isotopes with $Z \geqslant 100$. The solid lines denote calculations [7,8], squares —experimental data, the black squares— experimental data, obtained in the ^{48}Ca-induced reactions.

of the neighbouring isotope of element 116. Experiments are now in progress and we hope to get some more events.

A few remarks are here necessary.

The advent into a new region of nuclei inevitably means that all experimentally observed decays correspond to unknown nuclei. The identification of new nuclides — the determination of their atomic and mass numbers— becomes very difficult. This is the price of the unknown. At the same time, even-even α-radioactive nuclei obey the basic relation of radioactive α-decay, known as the Geiger-Nuttall rule, which connects the energy of α-decay, Q_α and the probability of decay, T_α. The mentioned relationship in the version of the Viola and Seaborg formula, according to refs. [7,8] reproduces with high accuracy the atomic number of all 58 known α-radioactive nuclei, for which the

Table 1. Experimental and calculated Q_α values for the α-decay chain of $^{292}116$.

Z	A	$Q_{\rm exp}$	$Q_{\rm theor.}$				
			YPE + WS [7,8]	FRDM + FY [9]	SHFB [10]	RMF [11]	TFM [12]
110	280	$\leqslant 9.4$[a)] (SF)	9.84	9.05	9.8	8.98	9.245
112	284	9.30 ± 0.05	9.80	8.69	9.4	9.30	8.885
114	288	9.98 ± 0.05	10.32	9.16	9.4	9.83	9.385
116	292	10.71 ± 0.06	11.07	10.82	10.43	11.647	11.025

[a)] The Q_α limit was calculated from the experimental $T_{1/2}$ value by using the formula from [7].

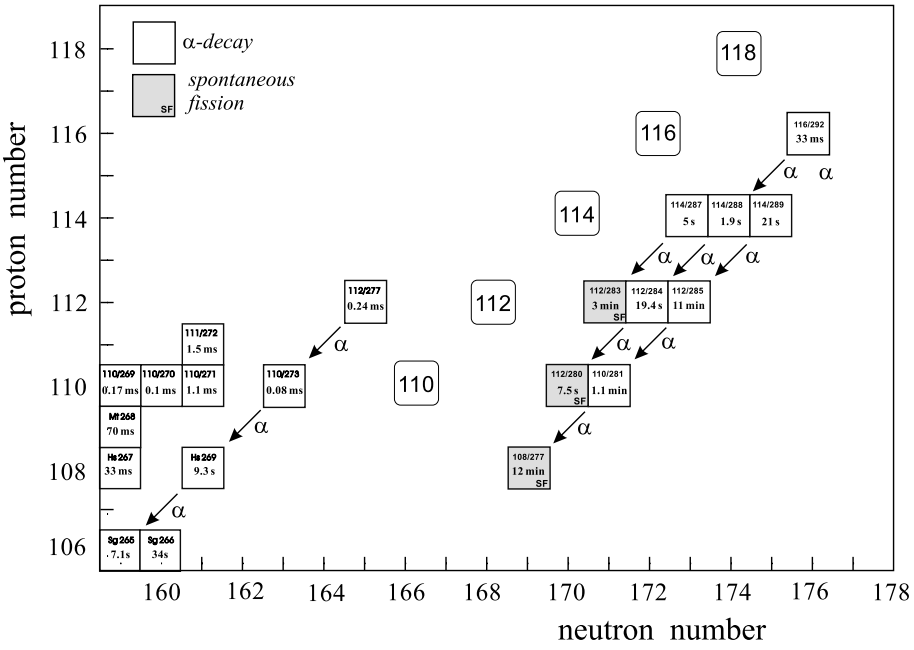

Fig. 4. The nuclide chart for the region of heavy elements with $A > 265$.

energy and decay times have been measured simultaneously. In fig. 3 the calculated and all experimental data on Q_α and T_α are shown for nuclei with $Z = 100$–116.

From this relationship it follows that for even-even nuclei in the decay chain 116→114→112, the atomic numbers are $115.4^{+2.7}_{-1.0}$ (one event) → $114.4^{+1.2}_{-0.8}$ (three events) → $111.2^{+1.2}_{-0.7}$ (three events). These values are in agreement with the identification in the decay sequence of even-even nuclei shown in fig. 2.

The decay properties of the new nuclides strongly differ from the properties of isotopes lying in the region of more neutron-deficient nuclei (fig. 4).

For the even-even isotopes of element 110, the transition from $^{270}110$ to $^{280}110$ shows an increase in the lifetime by about 5 orders of magnitude. For the odd isotopes of element 112 adding 8 units to the neutron number brings forth an increase of the α-decay lifetime by approximately 6 orders of magnitude. This is due to the significant decrease of the decay energy ($\Delta Q_\alpha \approx 3$ MeV), as was predicted by theory for the appearance of spherical closed shells defining the "island of stability" of superheavy elements.

The comparison of the experimental and calculated values of Q_α, obtained in different theoretical models [7–12], is presented in table 1. Such a comparison is of great value, because the main part of the calculations of mass and decay energies was done before any experimental data became available.

Without favouring any of the existing models, we have to note that with an accuracy of $\Delta Q_\alpha = \pm 0.5$ MeV the experimental values of Q_α coincide with the theoretical calculations. Therefore, it is with the same accuracy that experiment (in some models perhaps with higher accuracy) confirms the theoretical concept of the existence of an "island of stability" in the region of the hypothetical superheavy elements.

The combinations that we have investigated using ^{48}Ca-ions lead to the formation of nuclei with $N = 173$–176, lying 7–10 a.m.u. away from the closed neutron shell with $N = 184$. It seemed natural, that we should expect a further rise in cross-section when approaching $N = 184$. Unfortunately, in reactions like ^{48}Ca + ^{244}Pu, ^{248}Cm one reaches already the maximum possible neutron excess ($N - Z = 59$–61). A further increase of the number of neutrons may be achieved only using more neutron-rich

partner nuclei. However, here the possibilities are rather limited.

The maximum possible neutron excess in the target nucleus is defined by the capability of high-flux reactors to produce neutron-rich isotopes. On the other hand, the production of radioactive beams of Ca isotopes with mass numbers $A > 48$ with an intensity of ~ 0.1–1 pμA is also a difficult task due to the low formation cross-sections of such nuclei in different reactions. The situation becomes even more complicated for cold fusion reactions, where radioactive beams of the superexotic nuclei such as ^{82}Ni, ^{84}Zn or ^{86}Ge are necessary.

Another way to synthesize heavier elements with $Z =$ 124–126 is provided for by reactions, where the increase in the neutron number is achieved by means of increasing the nuclear charge and mass, *i.e.* by means of using heavier projectiles.

The synthesis of such heavy nuclei is of special interest. According to the calculations performed in the Hartree-Fock-Bogoliubov (HFB) and the relativistic mean-field (RMF) models, these nuclides may demonstrate a significant shell effect; they may have an unusual density ("semi-bubble nuclei") and as a result higher stability. The question is: what is the cross-section for the formation of such nuclei in fusion reactions?

Due to the increase in the Coulomb repulsion of the nuclei with raising of the projectile mass, the compound nucleus formation probability should strongly diminish. For instance, for the compound nucleus ^{312}124, which could be obtained in the ^{64}Ni + ^{248}Cm reaction, it will most probably be considerably less than the one observed in the case of the ^{48}Ca + ^{248}Cm$\rightarrow^{296}$116 reaction. However, the survival probability of such a nucleus will be much higher than the one for the isotope ^{296}116, due to the decrease in $E_x^{\min}$ by almost 15 MeV.

As a result, the obtained cross-sections $\sigma_{\rm EVR}$ to a great extent depend on the competition between two large quantities (fusion and survival probabilities); the resulting value of $\sigma_{\rm EVR}$ can be determined only experimentally. If the synthesis of this nucleus is feasible in the experiment, then probably its decay products will be nuclei with neutron numbers closest to the $N = 184$ shell and living long enough so as to allow detecting them at the existing setups.

This is a difficult way, but it is worthwhile testing it.

Another possibility is offered by symmetric fusion reactions between nuclei close to the fission fragments of the superheavy nucleus.

It is well known that the fission fragments possess considerable neutron excess. The fusion of such nuclei might in principle lead to the formation of superheavy compound nuclei with low enough excitation energy and with $N \sim 184$.

Indeed, this is an extreme case of cold fusion reactions, in which, as is well known, the nuclear structure plays an important role. Besides the energy balance in the fusion of magic nuclei, which is always of advantage for obtaining the minimum possible excitation energy of the heavy

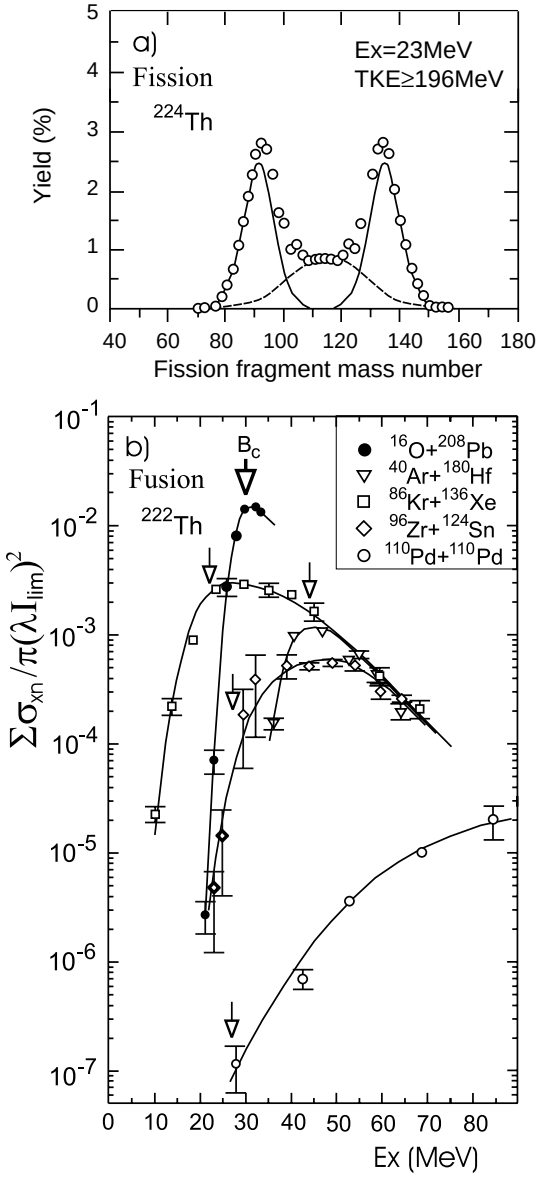

Fig. 5. a) Fission fragment mass distribution for ^{224}Th ($E_x = 23$ MeV) with TKE $\geqslant$ 196 MeV [13], obtained in the ^{16}O + ^{208}Pb reaction. b) Total evaporation-residue cross-sections for $^{220\text{-}224}$Th, obtained in different fusion reactions, indicated in the figure. The open squares are experimental data from the ^{86}Kr + ^{136}Xe reaction [15].

mono-nucleus, another important factor in its further fate is the nuclear shell effect in the fission fragments.

What may one expect, if in the fusion process nuclei close in charge and mass to the fission fragments of the heavy nucleus are used?

In order to check this idea, in 1998 we investigated the reactions of fusion and fission of the neutron-deficient Th isotopes. We shall give a short review of the obtained results.

In a first series of experiments, the mass and kinetic energy spectra of the fission fragments of the compound nuclei ^{224}Th ($E_x = 23$ MeV), formed in the ^{16}O + ^{208}Pb

reaction (fig. 5a), were measured [13]. At the highest TKE values, TKE = 196 MeV (cold fission), the fission fragment mass distribution had an asymmetric shape with maxima at $A_{\rm L} \sim 88$ and $A_{\rm H} \sim 136$. A similar picture was observed also in the charge distribution, where the maxima corresponded to $Z_{\rm L} \sim 36$ and $Z_{\rm H} \sim 54$ [14]. A conclusion could be made that one of the most probable fission channels of the weakly excited nucleus ^{224}Th is: Th $\rightarrow$ ^{86}Kr + ^{136}Xe, both fragments are stable nuclides.

In another experiment the formation cross-sections of Th isotopes in the inverse reaction ^{86}Kr + ^{136}Xe were measured [15].

In the region of low energies $E_x \leqslant 20$ MeV, as it can be seen in fig. 5b, the cross-section of evaporation residues, formed in the reaction ^{136}Xe(^{86}Kr,xn)$^{222-x}$Th, exceeds by about 3 orders of magnitude the cross-section $\sigma_{\rm EVR}$ in other reactions leading to the formation of the $^{220-224}$Th nuclei. At the same time reducing the mass of the target nucleus by 6 a.m.u. (^{130}Xe instead of ^{136}Xe) causes a decrease in $\sigma_{\rm EVR}$ by almost 3 orders of magnitude. The explanation for this can be that, in spite of the high fission barrier, no symmetric fission takes place in case of the compound nucleus ^{216}Th. This circumstance, as has been shown in the calculations of V. Pashkevich, is due to the structure of the potential energy surface of the nuclei ^{216}Th and ^{222}Th close and beyond the fission barriers [16]. One may assume that the collective nuclear motion from the contact configuration of the two touching spherical nuclei ^{86}Kr and ^{136}Xe to the saddle point of ^{222}Th follows a shorter path compared to the case of ^{216}Th, which undergoes mainly symmetric fission.

Is it then possible to produce with the same or even higher probability the heavier isotope ^{232}Th using the reaction between the radioactive nuclei ^{88}Kr + ^{144}Xe? The answer is far from obvious and most probably it is negative, since the asymmetric fission of ^{232}Th or ^{236}U is due to other factors influencing the formation of the fission fragments. For the synthesis of these nuclei important may turn out to be the isotopes, which lie close to the shells $Z = 50$ and $N = 82$ (^{132}Sn).

It is well known that the shells $Z = 50$ and $N = 82$ determine the character of spontaneous and low-energy fission of all actinides, also they are responsible for the symmetric fission of ^{258}Fm, ^{260}Md, ^{260}No and manifest themselves in the light-fragment group in the fission of the superheavy elements with $Z = 112$, 114, 116.

Because of the long half-life of ^{132}Sn ($T_{1/2}$ = 39 s), ^{132}Sn-beams could be obtained by the ISOL method. For the production of ^{132}Sn it is natural to use the fission of ^{238}U. Among all possible ways of accelerating fission fragments (except the exotic ones, where as a driver a powerful reactor is used), the method based on the photofission of ^{238}U in the region of the giant dipole resonance ($E_x \sim 15$ MeV) seems to be most feasible. From calculations and test experiments, which we have performed with an electron beam at $E = 25$ MeV, it follows that using an electron accelerator as a driver at $E = 50$ MeV and intensity $I = 1$ mA in a compact ^{238}U target (20 g) it is possible to produce $\sim 5 \cdot 10^{11}$ ^{132}Sn nuclei per second. Then, about 1% of these nuclei can be accelerated to energies $\leqslant 10$ MeV/A [17].

In this way, the intensity of the ^{132}Sn-beam may reach $\sim$ 1 pnA, which is still 3 orders of magnitude less than the intensity of the ^{48}Ca-beams used at present for the synthesis of new elements. Therefore, having this intensity, we can expect to synthesize element 114 in the hypothetical reaction ^{160}Gd(^{132}Sn,n)^{291}114 only if the corresponding formation cross-section is $\sigma_{\rm EVR} \geqslant 1$ nb.

This cross-section is $3{\cdot}10^4$ times less than the cross-section of the Th isotopes in the ^{86}Kr + ^{136}Xe reaction. A question arises: to what extent will this value compensate for the reduction of the cross-section as a result of the 1.6-fold increase of the Coulomb interaction in the superheavy nucleus compared to ^{232}Th? The answer to this question is very important in the given concept on the synthesis of superheavy elements.

So far radioactive beams of ^{132}Sn ions are not available. For this reason further experiments could be carried out using stable isotopes and the next step we consider is the use of the ^{136}Xe + ^{136}Xe reaction. In the fission of the ^{272}Hs symmetric fission modes should manifest themselves due to the influence of the closed neutron shells $N = 82$ in the fission fragments. The expected radioactive characteristics of the nuclei in the decay chain of the Hs isotopes, formed in 1n- and 2n-evaporation channels, is very convenient for detection, many of them are well-known nuclides. The absence of background from α-radioactive and spontaneously fissioning nuclei should simplify our task and increase the sensitivity of the experiment.

With the present-day technical opportunities the performance of an experiment aimed at the synthesis of isotopes of element 108 in symmetric reactions with cross-sections of about 1 nb and even less is not a problem. Other reactions of the type ^{124}Sn + ^{124}Sn $\rightarrow$ $^{248-x}$Fm or ^{124}Sn + ^{136}Xe $\rightarrow$ $^{260-x}$Rf, leading to known nuclides can add to the picture of the fusion of fission-like nuclei.

This paper presents the results of investigations carried out in collaboration with colleagues from LLNL (Livermore), GSI (Darmstadt), RIKEN (Wako-shi) and others. Many of them are co-authors of the original publications. I am taking the opportunity to express my warm gratitude to all of them for their contribution to the synthesis of new elements. I am also grateful to Dr. R. Kalpakchieva for her help in preparing this manuscript and for fruitful discussions.

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