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SYNTHESIS AND PROPERTIES OF SUPERHEAVY NUCLEI

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Introduction

The problem of synthesizing new elements has a long history. At a recent conference on nuclear-nuclear collisions (Kanazawa, 1991) Prof. P.Kienle presented in his talk the results of a research on synthesizing new elements up to Z=109 (Mt) [1].

Based on the few atoms produced in "cold fusion" nuclear reactions it was demonstrated that the radioactive properties of heavy nuclei confirm the main prediction of the macro-microscopic theory regarding a huge effect of nuclear shells on the spontaneous fission probability. As a result of the fission barrier emergence, determined by the nuclear structure, partial s.f. half-lives of heavy nuclei turn to be by 12-15 orders of magnitude larger than the values predicted by the classical liquid drop model of nuclei (fig.1)

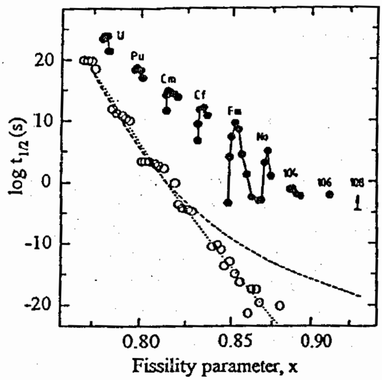

Fig.1. Partial fission half-lives on a logarithmic scale plotted sagainst the fissility parameter. •: experiment; o: corrected with Swiatecki's prescription (1969); ----: liquid drop half-life (P.Armbruster et al.) [2]. Taken from G.Muenzenberg [3].

Additional slowing down of the spontaneous fission of nuclei with the odd number of protons and/or neutrons make this effect still more important.

As a result of high stability to spontaneous fission, isotopes of heaviest elements with Z=107-109 and N=155-157 undergo α-decay with a half life of several ms.

What was the progress of these investigations within the last four years and what are the prospects?

1. Nuclear Shells and Stability of Heavy Nuclei

Macro-microscopic investigations of the potential energy surface of nuclei at large deformations were carried out already in the early papers by Myers and Swiatecki [4,5]. A significant breakthrough was achieved by Strutinsky and coworkers [6] who had created a method of calculating the total potential energy of a nucleus as a function of nuclear shape and of particle number. Both in these papers and in the later research by Pashkevich et al. [7], Nix et al. [8] and many other researchers it has been established that microscopic corrections drastically change the potential energy surface associated with fission.

Theory explains in general a number of experimental facts, fission barrier heights, shape isomerism in actinoid nuclei, spontaneous fission half-lives $T_{s.f.}$ of transactinoids, substantial variations in $T_{s.f}$ in the region N=152 etc., which have not found any explanation in the classical liquid drop models.

Similarly to any other theory it possesses a certain predictive power, in particular for prediction of masses and radioactive properties of yet unknown superheavy nuclei. Such predictions were made in a number of papers. We are presenting here recent data from papers by Patyk and coworkers [9,10] where there have been calculated masses and fission barriers as well as partial half-lives T_α and $T_{s.f.}$ of even-even nuclei with Z=100-112 and N=140-166.

Figure 2 presents a contour map of spontaneous fission half-lives as a function of proton and neutron numbers. Significant changes in $T_{s.f.}$ of nuclei far from the N=152 shell are determined to a great extent by another shell with N=162. It should be noted that both neutron shells are referred to deformed nuclei in contrast to double-magic nuclei such as ^{208}Pb (Z=82, N=126) possessing a spherical shape in the ground state. The maximum stabilization against spontaneous fission is expected for the nucleus ^{270}108 (Z=108, N=162) for which the predicted $T_{s.f.}$ may reach 10^4-10^9 s.

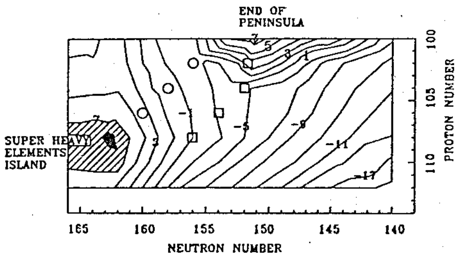

Fig. 2. Contour map of the logarithm of spontaneous fission half-life $T_{s.f}$ (s) calculated by Patyk et al. [9]. The different in the values of log $T_{s.f}$ between neighbouring lines is 2. □, even-even isotopes with N-Z=48 produced in cold fusion reactions; ○, isotopes with N-Z=54 produced in hot fusion reactions.

However, the calculation of spontaneous fission half-life $T_{s.f.}$ in the dynamical way consists in the search for one-dimensional fission trajectory in a multi-dimentional deformation space, which minimizes the action integral corresponding to the penetration of the fission barrier. Although the calculated static barrier heights are about equal, differences in half-life estimates can be attributed to varying assumptions regarding the dynamical path through the fission barrier and the consequent inertial mass. For example, Moller et al. [11], taking ^{258}Fm as model for heavier nuclei, assume that the path after the first barrier is short with the emerging fragments being nearly spherical and close to the doubly magic ^{132}Sn. On the other hand, Patyk, Sobiczewski, et al. [9,10] calculate dynamical barriers that show a different path, higher inertial mass, and consequently much longer SF half-lives. This competition between static and dynamical features of the SF process which leads to so large differences in stability makes experiments that explore ground-state decay properties of nuclei around N=162 and Z=108 one of the most important tasks in heavy element research.

2. Reactions of synthesis

It is known that the heaviest elements of the periodic table have been synthesized in the cold fusion reaction ^{208}Pb(HI,n). One of the main advantages of cold fusion is the relatively low excitation energy of the compound nucleus (E_x=18-20 MeV). At such a small excitation the nuclear shell effects disappear, although not completely, which gives a certain stability to the system with respect to fission. The transition into the ground state occurs by emission of just one or two neutrons and γ rays [12].

At the same time, in the region of transactinoid elements the necessity to use heavy projectiles with A≥50 leads to a strong dynamic limitation for fusion owing to a substantial growth of Coulomb forces (Z_1Z_2≥20000) [13,14]. A consequence of this is a strong decrease of fusion probability with the growth of bombarding ion mass. The cross sections of production of the known most heavy nuclei are in the region of picobarns. Unfortunately, even at these limitations, cold fusion reactions produce nuclides (N-Z)≤48 which are far away from the top of the predicted island of stability of super heavy elements (Fig.2).

In principle a significant growth in the number of neutrons in evaporation residues (EVRs) up to N-Z=54 can be obtained in fusion reactions between heavy actinide nuclei of the ^{244}Pu, ^{248}Cm type and projectiles such as ^{18}O, ^{22}Ne, ^{26}Mg.

There are also limitations here for EVR formation but they are of a completely different nature.

For strongly asymmetric combinations (Z_1Z_2≈1000) the dynamic limitations for fusion are small, which eliminates the fusion problem. Nevertheless, the excitation energy E_x^{min} of a compound nucleus even at the Coulomb barrier is about 40 MeV.

Structural effects practically disappear at such a high excitation energy (Fig.3); their fission barrier is determined only by the macroscopic (liquid drop) component of the nucleus deformation energy $B_f \approx B_f(LD)$. It is well known that for transactinide nuclei $B_f(LD)$ is practically equal to zero. In the absence of a fission barrier the excited nucleus becomes totally unstable to fission which should lead to a strong decrease in the probability of its transition to the ground state via cascade evaporation of neutrons (x≥4). Under these conditions the survival of EVRs totally depends on the dynamic properties of the excited compound nucleus.

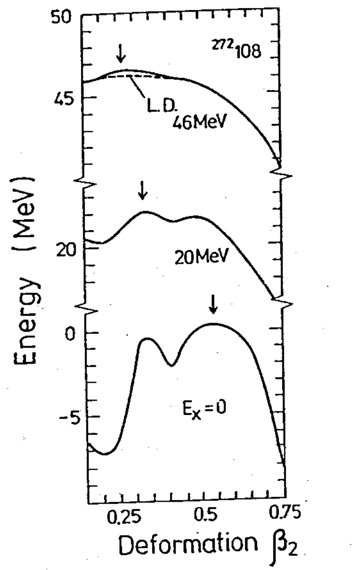

Fig. 3. Energy of ^{272}108 nucleus vs. its deformation at different excitation energies [15] (indicated in the figure): —, liquid drop component E_{LD}

Also, really, the probability of neutron evaporation at a set value of E_x is determined by the single-particle excitations and can be calculated within the statistical theory of hot nuclei decay. The time to reach the critical deformation irreversibly leading to disintegration of the nucleus into two fragments is determined by the dynamics of the collective motion of the system, the calculation of which seems to be extremely complicated.

Investigation of excited nuclei fission dynamics by measuring the characteristics of pre-fission emission of gamma-quanta, neutrons and light charged particles was performed in numerous papers (see for example the overviews by Newton [16], Hilcher and Rossner [17].

We are most interested here in the region of extremely heavy nuclei: $B_f(LD) \to 0$ with $E_x \approx$ 40-50 MeV and we are presenting here the experimental data obtained in collaboration with HMI (Berlin) for excited nuclei of Cf-Fm ($B_{L.D.} \approx 1.5$-MeV) [18,19].

As is seen in Fig.4 the contribution of pre-fission neutrons increases with the increase in excitation energy. The probability of pre-fission neutron emission can be calculated for the whole time interval of nucleus existence up to the moment of its splitting into two fragments. This time can be chosen as a parameter to obtain the best agreement with the experimental dependence $\nu_{pre}(E_x)$. For Cf-Fm nuclei $\bar{\tau}_f \sim (3\text{-}4) \times 10^{-20}$s.

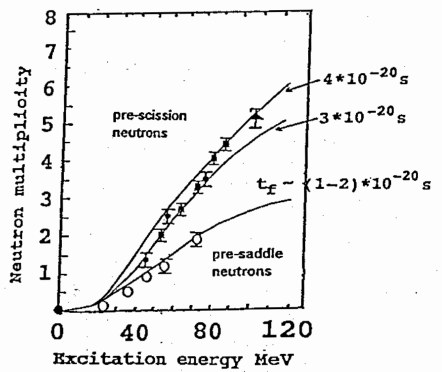

Fig. 4. The number of pre-scission neutrons vs. the excitation energy of the fissioning nucleus: ●, *$^{238}U(^{12}C,f)$ [19];* ■, *for $^{232}Th(^{14}F,f)$ [18];* ▲, *for $^{232}Th(^{20}Ne,f)$ [20]. Points at E_x=0 are reffered to spontaneous fission of ^{252}Cf. ——, calculated dependence $\nu_{pre}(E_x)$ at different values of τ_f. Open circles: pre-saddle neutrons obtained from excitation function of xn-reactions.*

Such experiments are used to measure the total number of neutrons emitted prior to reaching the scission point (pre-scission neutrons). A part of them had been emitted before the moment the nucleus reached the saddle point (pre-saddle neutrons). The number of pre-saddle neutrons can be defined from the excitation functions of the reaction $\sigma_{xn}(E_x)$ which determine the ratio of the widths Γ_n/Γ_f on each stage of compound nucleus de-excitation.

The values $\Gamma_n/\Gamma_{tot}(E_x)$ for nuclei with Z=98 and 102 presented in fig.5 and cited from the data of Sikkeland et al. [21] and Andreev et al. [22] testify to the fact that at $E_x \geq 40$ MeV the fission and neutron evaporation probabilities are comparable.

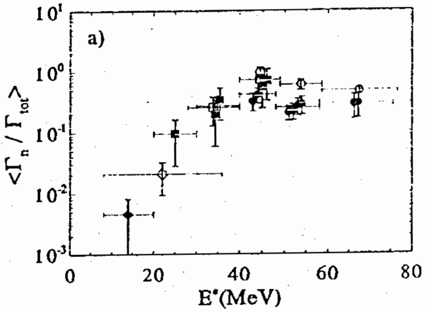

Fig. 5. The ratio Γ_n/Γ_{tot} vs. the excitation energy of compound nuclei; open symbols Z=98 [21], black points Z=102 [22].

Note that for ^{250}Cf (E_x = 80 MeV) approximately half of the neutrons is emitted before reaching the saddle point.

At such a slow progress of deformation in the fission channel (viscous regime) even the heaviest nuclei with $B_{L.D} \approx 0$ will have a finite probability of transition to the ground state through evaporation of neutrons. Quantitative data can be obtained only in direct experimental measurements of the evaporation residues formation cross sections for the heaviest excited nuclei.

The presented in fig.6 cross sections of reactions in the region Z=102-105 obtained by Andreev et al. [23] together with the previously available data for Cf and Fm and new results obtained by Lazarev et al. [24] for Z=106 make it possible to compare the possibilities of synthesizing superheavy elements in cold and hot fusion reactions.

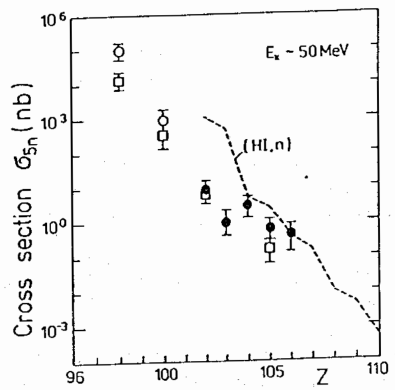

Fig. 6. The cross sections σ_{5n} (○, •) and σ_{6n} (□) vs. atomic number of evaporation residues. Nuclei of Cf and Fm were produced in the fusion reactions with ions of ^{16}O and nuclei with Z=102-106 with ions of ^{22}Ne, ^{26}Mg, ^{27}Al. —, drawn through the experimental cross-section values of (HI,n) in cold fusion reactions.

In the synthesis of isotopes with Z≥104 the EVR production cross section in the reactions Pb(HI,n) and the cross sections of more asymmetric hot fusion reactions of the type Pu,Cm(^{22}Ne,4-6n) are practically comparable within the experimental error.

But, like it has been indicated above, asymmetric reactions allow to penetrate deeper into the region of still more neutron-rich nuclei and present in this way a unique opportunity of testing different theoretical models in the region of deformed shells Z=108, N=162.

3. Observations of Enhanced Stability of Nuclei Z=104 and 106 near Closed Shells

Essentially, this was the underlying idea of a joint JINR (Dubna) - LLNL (Livermore) experiment on synthesis of 106 element heavy isotopes [24].

The ground-state decay properties of ^{266}106 should be a quite sensitive probe of the theoretical predictions shown in fig.2. If there is increased stability near N=162 and Z=108, the isotope ^{266}106 should have a SF- or α-decay half-life of tens of seconds. Otherwise, ^{266}106 should decay by SF with a half-life of ~100 μs, a T_{sf} difference of ~10^5 or more. Thus a distinct signature for enhanced nuclear stability near N=162 and Z=108 would be the observation of the α decay of ^{266}106 followed by the SF decay of the daughter nucleus ^{262}104. A signature for the odd-A isotope ^{265}106 would be the observation of its decay followed by decays of the known nuclides ^{261}104 and ^{257}102.

To produce ^{265}106 and ^{266}106 we used the complete fusion reaction ^{248}Cm+^{22}Ne at bombarding energies which are expected to provide maximum cross sections for the 4n and 5n evaporation channels.

In a 360-hour irradiation of the ^{248}Cm target with a ^{22}Ne total ion beam dose of $1.6 \cdot 10^{19}$ produced on the U-400 accelerator (FLNR) by means of the gas-filled separator of recoils there have been synthesized two new most neutron-rich isotopes of element 106 with masses 265 and 266 (fig.7).

Both the isotopes ^{265}106 (N=159) and ^{266}106 (N = 160) undergo mostly the α-decay with energies E_α = 8.71 ÷ 8.91 and

8.63±0.05 MeV correspondingly. The energy of α-decay of the even-even nucleus $^{266}106$ ($Q_\alpha = 8.76$ MeV) determines its half-life T_α=10-30 s.

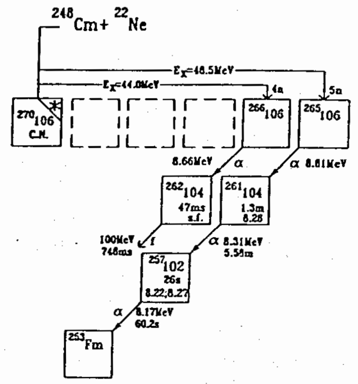

Fig. 7. Two out of the ten events demonstrating the decay of $^{265}106$ and $^{266}106$ isotopes produced in the reaction of $^{248}Cm+^{22}Ne$. The excitation energy values for the compound nucleus of $^{270}106$ at which these events have been registered are shown in figure.

Based on the six registered (α,sf) correlations referring to the α-decay of $^{266}106$ nucleus there was also determined the partial spontaneous fission half-life of the daughter nucleus $^{262}104$ (N=158) $T_{s.f.} = 1.2^{+1.0}_{-0.5}$ s.

Radioactive properties of even-even isotopes of $^{262}104$ and $^{266}106$ give an indication of a substantial growth of heavy nuclei stability to spontaneous fission when approaching the closed shells Z=108 and N=162 (fig.8).

And really, with decreasing number of neutrons in the nucleus of $^{262}104$ by 6 units ($^{256}104$) or of protons - by 4 units (^{258}Fm), the spontaneous fission half-life gets correspondingly 250 and 2500 times shorter.

Similar to this at the decrease of the number of neutrons in the nucleus of $^{266}106$

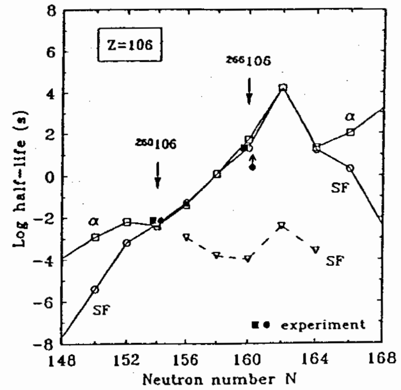

Fig. 8. Predicted partial half-lives [9,10] for SF and α decay of the even-even 106 isotopes shown by the lines connecting circles and squares, respectively. The dashed line connecting the triangular points shows SF half-life predictions [11]. The experimental values for $^{260}106$ [25,26] and the results for $^{266}106$ [24] are shown for comparison.

by 6 units ($^{260}106$) or of the number of protons by 4 units ($^{262}102$) the half-life of spontaneous fission grow over 5000 times smaller. Moreover, a heavier nucleus has a larger half-life: $T_{s.f.}$ (Z=106, N=160) >> $T_{s.f}$(Z=104, N=158).

This means that the nuclei obtained in this experiment are in the process of an abrupt increase of stability to spontaneous fission like it has been predicted by macro-microscopic calculations by Patyk, Sobiszewski et al. [9,10], (fig.3). Another spontaneous fission mode, characterized by a short way of tunneling through the fission barrier [11] and leading to a sharp decrease of $T_{s.f.}$ for $^{266}106$ is prohibited by more than 10^4 times.

The expected significant growth of T_α with the growing number of neutrons makes possible experiments studying the chemical properties of element 106 - EkaW.

Note that when using an exotic target of isotope ^{250}Cm ($T_{1/2}$~10^4 years) produced in a nuclear explosion, in a similar way there can be produced some heavier alpha-active isotopes of element 106 with N=161, 162 and with half lives of several hours.

Another way - synthesis of heavier elements in the maximum effects area of nuclear shells.

4. Experiments on Synthesizing New Isotopes with Z=108 (Hs)

Among all possible target-ion combinations leading to the production of a $^{270}108$ nucleus with closed shells Z=108 and N=160, the reaction $^{238}U(^{36}S,4n)^{270}108$ seems to be the most promising one.

Investigations of fusion reactions $^{206-208}Pb(^{34,36}S,2\text{-}4n)$ in the course of which there were synthesized new neutron-deficient isotopes of Cf demonstrated high sensitivity and selectivity of the kinematic separator to detection of evaporation residues [27].

Along with that, because of a great expenditure of a rare isotope ^{36}S (the natural abundance - 0.015%) by a PIG-type ion source it would be most difficult to carry out such an experiment*.

That is why in March-April 1994 experimenters in Dubna were using a beam of a more abundant isotope ^{34}S enriched up to 90%.

The bulk of data is being processed yet but the preliminary results are of certain interest.

At the irradiation of a ^{238}U target with a total ^{34}S-beam dose of $1.7 \cdot 10^{19}$, the

* - this does not exclude, nevertheless, the production of a ^{36}S beam using a more economical ECR ion source in future.

position sensitive strip detectors of recoils located in the focal plane of the recoil separator registered 3 (α-α) correlation events clearly pointing to the production of a new isotope of element 108 with a mass of 267.

The energy of bombarding ions was E_L=185 MeV(E_x≈50 MeV) near the maximum of the 5n-evaporation channel which did not exclude, nevertheless, the observation of a heavier isotope $^{268}108$ in the 4n - channel. As is seen in Table I, 20-30 ms after the recoil nucleus entry into the detector there was observed its decay with emission of an α-particle with an energy E_α = 9.74-9.88 MeV followed by the subsequent decay of $^{259}104$ or $^{255}102$ daughter nuclei the decay properties of which are well known (fig.9).

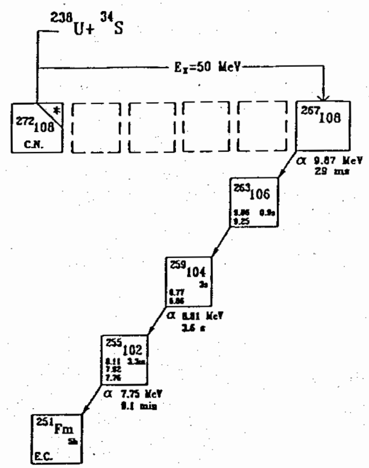

Fig. 9. Decay properties of isotope $^{267}108$ produced in the $^{238}U(^{34}S,5n)$ reaction.

In Table II new results are presented together with the earlier data obtained by Muenzenberg et al. [28] for lighter isotopes of element 108 in cold fusion reactions.

Table I

Prominent correlation chain observed in the ^{238}U + ^{34}S reaction

Particle	Particle energy MeV	Strip number	Time interval	Position deviation mm	Beam pulse
EVR	4.2	11			on
α	9.74	11	29 ms	1.0	off
α	7.75	11	9.1 min	1.0	off
EVR	6.2	6			on
α	9.87	6	32 ms	0.5	off
α	8.81	6	3.6 s	0.1	on
EVR	4.4	9			on
α	9.88	9	20 ms	0.2	on
α	8.80	9	2.0 s	0.2	off
EVR	4.0	2			on
α	8.86	2	0.68 s	0.5	off
α	7.77	2	3.7 min	0.8	off

Table II

Ground-state decay properties of the isotopes of element 108 (Hs)

Isotope	Decay mode	Alpha-particle energy *MeV*	Half-life *ms*	Number of decay sequences	Synthesis reaction
$^{264}108$	α	≈10.8	$0.076^{+0.364}_{-0.036}$	one	$^{207,208}Pb+^{58}Fe$ Münzenberg *et al.*
$^{265}108$	α	10.36	$1.8^{+2.2}_{-0.7}$	three	
$^{267}108$	α	9.83	19^{+29}_{-7}	three	$^{238}U+^{34}S$ This work

The yield of $^{267}108$ nuclei in the 5n-channel (E_x = 50 MeV) corresponds to the reaction cross section of about 2 pb (with a factor of 3 accuracy).

On the other hand the cross section of $^{265}108$ nuclei produced in the cold fusion reaction $^{208}Pb(^{58}Fe,n)^{265}108$ at E_x = 18 MeV amounts to 19^{+18}_{-11} pb [28].

A substantial decrease of the survival probability with increasing excitation energy of compound nuclei Z=108 from 18 to 50 MeV (five stages of neutron evaporation instead of one) is compensated to a substantial extent by lifting strong dynamic limitations for fusion at the transition from the reaction $^{208}Pb+^{58}Fe$ (Z_1Z_2=2132) to a more asymmetric combination $^{238}U+^{34}S$ (Z_1Z_2=1472).

This circumstance changes significantly the understanding of the character of dynamic limitations for fusion of asymmetric systems [29,30]. According to theoretical calculations and regularities obtained from dynamical models of the extra push type the fusion of as massive nuclei as ^{238}U and ^{34}S is to be strongly prohibited and this has not been observed in the experiment.

5. Prospects of Synthesizing New Elements

All the presently known data on masses and radioactive properties of transactinoid nuclei are well described by the modern macro-microscopic theory. Remaining in the frame of theoretical predictions one can assume that the yet unknown nuclei with Z>106 and N>154 will undergo mostly the α-decay.

This circumstance changes qualitatively the approach to the problem of synthesizing new elements since the periods and energies of alpha-decay determined by nuclear masses in the ground state can be predicted with good accuracy.

Both the cold and hot fusion reactions can be used as the next step for the synthesis of new elements with Z>109.

Nevertheless, in both the cases the expected cross section will be about 1pb or less, which requires a large increase of the sensitivity of experiments.

A substantial progress in this direction has been obtained at the GSI (Darmstadt) by using the new injector of the UNILAC accelerator with an ECR ion

source as well as by updating the systems of separation and registration of very rare events of nuclear decay of the new element.

In the scheduled experiment on $^{208}Pb(^{62}Ni,n)^{269}110$ we are expecting an α-decay of the $^{269}110$ nucleus leading to the production of a daughter nucleus $^{265}108$ the properties of which have been defined earlier [28].

Another possibility - the hot fusion reaction using isotopes of plutonium as the target material. In principal, in the reaction $^{242,244}Pu(^{34}S;4n5n)^{271-274}110$ there can be obtained more neutron-rich isotopes of element 110 with N = 161-164 for which the correlation chain of α-decay may be also restored based on the known properties of daughter nuclei $^{265,266}106$ or $^{265}108$. Such an experiment requires an increase in the intensity of ^{34}S ion beam on the U-400 accelerator and certain updating of the recoil separator.

Despite the complexity of such experiments they are a direct way of testing modern macro-microscopic theories describing and predicting the properties of super heavy nuclei. On the other hand, they present a unique opportunity of studying the collective motion dynamics at fusion and fission of heavy nuclear systems in extreme conditions of the Coulomb and nuclear forces.

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