Journal of Alloys and Compounds, 213/214 (1994) 50–60
JALCOM 4018

Synthesis reactions and radioactive properties of transactinoid elements*

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Abstract

It is well known that the heaviest elements of the periodic table have been synthesized in the cold fusion of magic nuclei of Pb with $Z \leqslant 26$ ions. Because of dynamic limitations for fusion under strong Coulomb interaction of nuclei, the cross-sections of cold fusion reactions diminish exponentially with growing compound nucleus atomic number. For element $Z=110$ produced in the reaction $^{208}Pb(^{62}Ni,n)^{271}110$, the expected cross-section is 10^{-36} cm^2. In still more asymmetric reactions, when isotopes of actinoid elements irradiated with relatively light ions ($Z \leqslant 12$) are used as the target material, the compound nuclei possess an excitation energy of approx. 50 MeV. At this energy the nuclear shell effects are strongly suppressed and, as a result, in the case of hot compound nuclei of transactinoid elements the fission barrier is practically absent. The transition of these nuclei into the ground state depends strongly on the dynamic properties of the system with respect to the fission degree of freedom. Experimental studies were going on in two directions: (1) determination of the fission time by measuring the prefission neutrons (of Cf–Fm nuclei) in a wide interval of excitation energies; (2) direct synthesis of known nuclides with $Z=102$–105 in reactions with ions of ^{22}Ne, ^{26}Mg, ^{27}Al and ^{31}P when final nuclei are produced in the ground state after the evaporation of five or six neutrons from the excited compound nuclei ($E_x=50$–60 MeV). The dependence of the reaction cross-section (HI, 5–6n) on the atomic number of the compound nucleus in different target–ion combinations points to the possibility of synthesizing new elements in hot fusion reactions. The advantage of these reactions arises from the use of neutron-rich nuclei like ^{248}Cm and ^{249}Cf which allows us to synthesize nuclei close to the deformed shell $N=162$, for which a considerable growth of stability against spontaneous fission is predicted. Experimental set-ups and methods of detecting rare events of formation and decay of transactinide nuclei are described.

1. Neutron shells and stability of transactinoid elements

Theoretical predictions of the existence of superheavy elements' islands of stability are based on fundamental properties of nuclear matter undergoing a large-scale collective motion. Calculations of the nuclei potential energy within the liquid drop model made with account taken of their structure indicate the existence of shell effects not only in the ground but also in the strongly deformed states. Shell corrections with an amplitude of just several megaelectronvolts (at a nucleus total energy of about 2 GeV) play an important role in the stability of superheavy nuclei undergoing, with a high probability, spontaneous fission (SF).

Macromicroscopic investigations of the potential surface of nuclei at large deformations were carried out already in the early papers by Myers and Swiatecki [1, 2]. A significant breakthrough was achieved by Brack [3] 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 [4], Nix [5] and many other researchers it has been established that microscopic corrections drastically change the potential energy surface associated with fusion.

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

*During the preparation of this report we heard the sad news of the untimely death of Professor Vilen Strutinsky. The fundamental research by V. Strutinsky in the nuclear shell theory and particularly in the field of heavy and superheavy elements is well known to the scientific community and I dedicate this paper to this memory.

SSDI 0925-8388(93)04018-L

papers. We are presenting here recent data from papers by Patyk and coworkers [6, 7] where there have been calculated masses and fission barriers as well as half-lives T_α and $T_{s.f.}$ of even–even nuclei with $Z=100$–112 and $N=140$–166.

Figure 1 presents a contour map of spontaneous fission half-lives as a function of proton and neutron numbers [6]. 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 $T_{s.f.}\approx 30$ years.

The calculations describe satisfactorily the spontaneous fission half-lives of even–even nuclei with $Z=102$, 104, 106 and 108 with N-$Z=48$ produced in experiments at Dubna and Darmstadt [8, 9]. This is one of the outcomes of the theory.

The hypothesis of the existence of the island of stability is to be tested only in direct experiments on the synthesis and investigation of radioactive properties of neutron-rich nuclei with $Z\geqslant 104$ close to $N=162$.

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 ^{248}Cm type and projectiles such as ^{18}O, ^{22}Ne, ^{26}Mg.

For even–even isotopes of transactinoid elements ($Z\geqslant 106$) the increase in the number of neutrons by 6 units will lead to the increase in T_α by 3–4 orders of magnitude [7] which widens substantially the field of measurements of partial spontaneous fission half-lives at $T_{s.f.}<T_\alpha$.

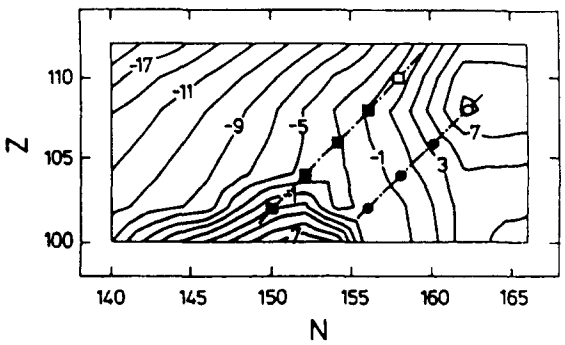


Fig. 1. Contour map of the logarithm of spontaneous fission half-life $T_{s.f.}$ (s) calculated by Patyk *et al.* [6]. The difference in the values of log $T_{s.f.}$ between neighbouring lines is 2. □, ■, even–even isotopes with N-$Z=48$ produced in cold fusion reactions; ○, ●, isotopes with N-$Z=54$ produced in hot fusion reactions. ●, ■, $T_{s.f.}$ values defined or estimated experimentally.

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, xn). 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 [10].

At the same time, in the region of transactinoid elements the necessity to use heavy projectiles with $A\geqslant 50$ leads to a strong dynamic limitation for fusion owing to a substantial growth of Coulomb forces ($Z_1Z_2\geqslant 2000$) [11, 12]. Thus, for example, the expected cross-section of element 110's production in the reaction ^{208}Pb(^{62}Ni, n)^{270}110 is estimated to be about 1 pb (10^{-36} cm^2) [13]. Unfortunately, even at these limitations, cold fusion reactions produce nuclides which are far away from the top of the predicted island of stability of super heavy elements.

It has been mentioned above that only reactions with relatively light ions and heavier targets, isotopes of actinoid elements, can lead to EVRs near $Z=108$ and $N\approx 162$. There are also limitations here for EVR formation but they are of a completely different nature.

For strongly asymmetric combinations ($Z_1Z_2\approx 1000$) 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 are practically absent at such a high excitation energy (Fig. 2); their fission barrier is determined only by the macroscopic (liquid drop) component of the nucleus deformation energy $B_f\approx B_f(\mathrm{LD})$. It is well known that for transactinide nuclei $B_f(\mathrm{LD})$ 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 ($x\geqslant 4$). Under these conditions the survival of EVRs totally depends on the dynamic properties of the excited compound nucleus.

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 not only extremely complicated but also rather ambiguous.

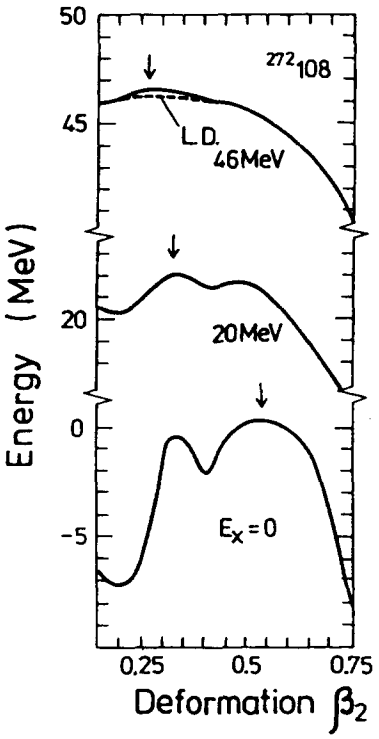


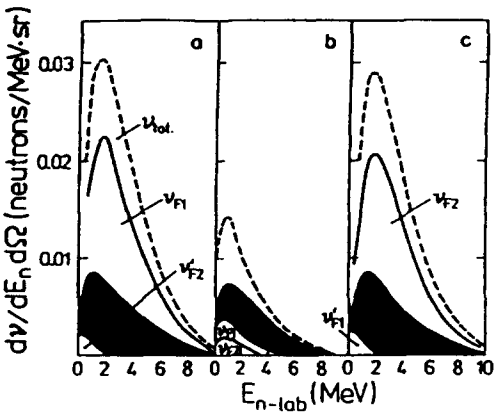
Fig. 2. Energy of ^{272}108 nucleus *vs.* its deformation at different excitation energies [14] (indicated in the figure): – – –, liquid drop component E_{LD}.

3. Fission time of excited nuclei of Cf–Fm

Investigations of the fission dynamics of hot nuclei were started already in the research of Harding and Farley [15], who measured the pre- and post-fission neutrons in the bombardment of ^{238}U with 147 MeV protons. In more recent papers (see overviews by Newton [16] and Hilscher and Rossner [17]) not only neutrons but light particle emissions of very hot nuclei were also measured. We are most interested here in the region of extremely heavy nuclei: $B_f(\mathrm{LD}) \to 0$ with $E_x \approx 40$–50 MeV.

In collaboration with HMI (Berlin) the fission neutrons of Cf–Fm nuclei with $E_x \approx 40$–50 MeV have been experimentally investigated [18, 19].

I would like to remark here that the essence of this experiment is in the measurement of neutron spectra at different angles to the fission axis. The fragments' masses and kinetic energies are also measured in the experiment. From neutron spectra one can separate the so-called post-fission neutrons (emitted by the fragments). Their space and energy distribution is in a strict correlation with the momentum of fission fragments. For pre-fission neutrons the spectra and angular distribution are independent of the fission fragments' kinematic and are related to the movement of the compound nucleus (Fig. 3).



(a) (b) (c)

Fig. 3. The neutron spectra of ^{250}Cf ($E_x = 75$ MeV) in fusion–fission reactions measured at different angles to the fission axis: (a), (c) ν_{F1}, ν_{F2}, neutrons emitted along the direction of fission fragments momentum ($\Theta_{f,n} = 0°$); ν'_{F1}, ν'_{F2}, in the opposite direction ($\Theta_{f,n} = 180°$); (b) the same at $\Theta_{f,n} = 90°$. The shaded region represents pre-fission neutrons; – – –, summary spectra. The data are taken from the paper by Kozulin *et al.* [19].

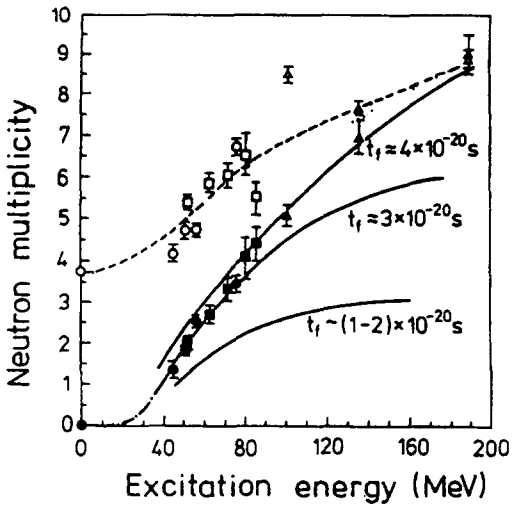


Fig. 4. The number of post-fission (○, □, △) and pre-fission (●, ■, ▲) neutrons *vs.* the excitation energy of the fissioning nucleus: ○, ●, ^{238}U(^{12}C,f) [19]; □, ■, for ^{232}Th(^{14}F,f) [18]; △, ▲, for ^{232}Th(^{20}Ne,f) [20]. Points at $E_x = 0$ are referred to spontaneous fission of ^{252}Cf. ——, calculated dependence $\nu_{pre}(E_x)$ at different values of τ_f (presented in the figure); – – –, given for guidance, $\nu_{post}(E_x)$.

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 $\bar{\tau}_f$ is about $(3\text{–}4)\times10^{-20}$ s. The experimental value of $\bar{\tau}_f$ exceeds

noticeably the indirect estimations of fission time based on the DIC collision data [21] and the dynamic calculations performed in the frame of diffusion models with a two-body viscosity [22].

The measured value $\bar{\tau}_f$ is the average time of collective motion of the nucleus from the moment of its formation to the scission point. It is natural that to reach critical deformation one requires the time $\tau_{cr} < \bar{\tau}_f$. One can assume, nevertheless, that for heavy nuclei under a high viscosity deformation mode τ_{cr} has a finite and a small but not negligible value as compared with the characteristic time $\tau_n(E_x)$ of neutron emission.

Slow motion along the relatively flat potential surface of the nucleus increases the probability of neutron emission which should lead in its turn to a significant change in the EVR yield. Thus, nuclei have a finite probability of transition into the ground state even at $B_f(\mathrm{LD}) \approx 0$.

Unfortunately, in the statistical models of nuclei deexcitation the dynamic properties of the system are definitely not considered. For the region of strongly fissioning nuclei they are taken into account indirectly by fitting the parameters of calculation with experimental data [23, 24]. This procedure works well for the estimation of cross-section near the known data region which exists in reality. It seems, nevertheless, that the collective dynamics will play a still more important if not the main role in the production of distant transactinoid nuclei and the quantitative estimations of the cross-sections require further experimental research.

4. Hot fusion reactions

During the last two years measurements of the cross sections of the reactions (HI, xn) at $x = 4$, 5, 6 in the region $Z = 102$–105 have been carried out. The experiments were carried out on recoil separators using targets of ^{232}Th, ^{236}U, ^{242}Pu and projectiles of ^{22}Ne, ^{26}Mg, ^{27}Al and ^{31}P leading to production of isotopes ($Z \geqslant 102$) with known radioactive properties [25, 26]. From the analysis of the data of these papers it follows that in all the cases the maximum cross-section of EVR production is observed in the channel (HI, 5n) which corresponds to an excitation energy E_x of compound nuclei of about 50 MeV. Figure 5 presents the experimental values of cross-sections for (HI, 5n) in the region 102–105 together with the previously known data for Cf and Fm and with the new results for ^{248}Cm(^{22}Ne, 5n).

Even without the correction for N-Z of the compound nuclei produced in these reactions it is seen that $\sigma_{5n}(Z)$ values in the range $Z = 102$–105 are within two orders of magnitude: from 10 to 0.1 nb. Here the dependence of σ_{5n} on the fissility parameter is noticeably weaker

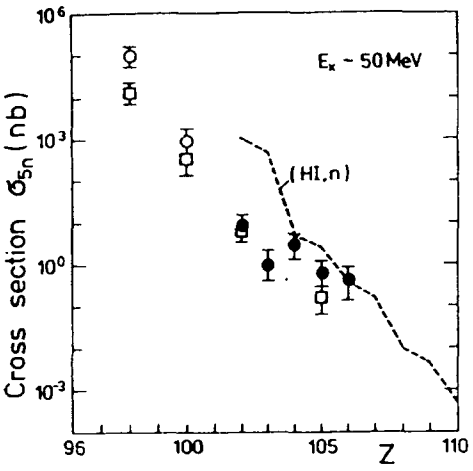


Fig. 5. The cross-sections σ_{5n} (○, ●) and σ_{6n} (□) *vs.* atomic number of EVRs. Nuclei of Cf and Fm were produced in the fusion reactions with ions of ^{16}O and nuclei with $Z = 102$–106 with ions of ^{22}Ne, ^{26}Mg, ^{27}Al. – – –, drawn through the experimental cross-section values of (HI, n)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

than in the region of light actinoids where $B_f(\mathrm{LD})$ reaches several megaelectronvolts [27].

The cross-section σ_{5n} reflects the integral probability of nuclei survival in the whole energy range up to 50 MeV. At the same time, the increase in the excitation energy E_x to about 60 MeV as is demonstrated in Fig. 5 practically does not lead to a decrease in the cross-section of EVR production in the channel (HI, 6n).

Skipping a detailed analysis, which we shall come back to later, it is possible to draw some conclusions. When dealing with hot nuclei one has no reasons to expect a dramatic decrease in the EVR cross-section on moving more deeply into the region of heavier elements in the way that occurs in cold fusion reactions. Hot fusion reactions can turn out to be the most efficient method of synthesis of superheavy nuclei in the new region of stability.

We have to note, nevertheless, that the expected cross-sections are evaluated within the picobarn scale. This produces for high requirements on the experimental method capable of registering and identifying extremely rare events of production and decay of superheavy nuclides.

5. The experimental facilities

In a complete-fusion reaction neutron evaporation slightly changes the compound nucleus momentum. As a result, the EVRs retain their strict orientation along the direction of the ion beam. Also, vice versa, in incomplete fusion reactions the initial momentum is distributed in the exit channel between two fragments that scatter at large angles.

Hence it follows that a recoil separator according to the angle and momentum can result in a substantial suppression of the background from the nuclei formed in the incomplete-fusion channels. This principle underlies all kinematic separators of recoil nuclei. As the separator should be placed at the angle $\theta_L = 0°$ it is necessary to carry out the separation of recoil nuclei from the primary beam. The recoil nuclei can be separated only in an electric field by using their difference in energy (velocity) because the magnetic rigidities of EVRs and the ion beam are practically equal. As is known, a similar method is employed in velocity and energy selectors [28, 29].

At the same time, as has been shown in refs. 30–32, it is possible to revert to nuclear separation in a magnetic field if one creates a low pressure gaseous medium in which the ion and recoil charges depend on velocity and, therefore, greatly differ from each other. Gas-filled recoil separators are based on this principle.

We use both types of kinematic separators of beams available from the U400 accelerator. As the characteristics of the recoil energy selector (the facility "VASSILISSA") were published earlier [29], we shall give here the parameters of the gas-filled magnetic separator (Fig. 6).

The separator was put into operation in 1989. Its D–Q–Q design and main parameter values as well as first tests at the U400 cyclotron were described in ref. 33. Since then many significant improvements and numerous model experiments have been carried out to develop the separator into a facility for heavy element research. Owing to its underlying principle, the separator exhibits excellent qualities for fusion–evaporation reactions induced by ^{40}Ar and similar projectiles. Therefore the main goal of our recent developments was to achieve a reliable application of the separator to highly asymmetric fusion–evaporation reactions induced by much lighter projectiles such as ^{18}O or ^{22}Ne on targets of the transuranium nuclides [34, 35]. Special emphasis was laid on the possibility of applying very intense beams of these lighter projectiles to strongly radioactive and rather exotic target species such as ^{242}Pu or ^{248}Cm.

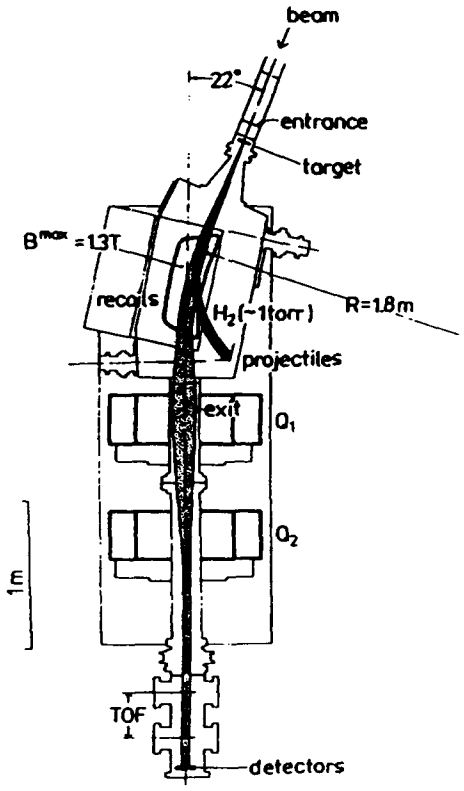


Fig. 6. Lay-out of the Dubna gas-filled recoil separator.

Being very attractive from the viewpoint of heavy element research, the highly asymmetric fusion–evaporation reactions represent the most difficult case for study with recoil separators. In fact, as compared, for example, with ^{40}Ar-induced reactions, the asymmetric reactions are characterized by much broader angular distributions of EVRs. The large angular divergence of the EVRs restricts effective target thicknesses to values of about 0.2 mg cm^{-2} or less. Other complications originate from low kinetic energies $\langle E_R \rangle$ of EVRs recoiling out of the target (typically $\langle E_R \rangle$ are below 10 MeV) as well as from large dispersions of E_R. Perceptible $\langle E_R \rangle$ losses occur in the 3.7 m long path of EVRs through the gas filling the separator (usually H_2 at 0.7–1 Torr), as well as in the exit window and in pentane gas filling the detection module. The energy decrease leads to a significant decrease in the EVR transmission through the separator. Furthermore, a decrease in $\langle E_R \rangle$, *i.e.* in the average EVR velocity, causes a lowering of the average charge state $\langle q \rangle$ of EVRs in the gas media and seems to give rise to significant fluctuations of q. Appreciable q fluctuations at the low $\langle q \rangle$ values characteristic of EVRs from very asymmetric reactions are supposed to give additional reasons for fairly low EVR collection efficiencies ϵ_c in these cases as well as for extended image sizes at the focal plane (Fig. 7).

A summary of measured or estimated characteristics of separation for a number of highly asymmetric fusion–evaporation reactions is presented in Table 1. It is seen here that collection efficiencies ϵ_c for heavy EVRs produced by these reactions range between 3% and 10%. However, despite the fairly low ϵ_c values, the net sensitivity of experiments can be significantly improved by applying very intense beams of ^{18}O, ^{22}Ne and other lighter projectiles, up to $(2–4)\times10^{13}$ pps, which are provided by the Dubna U400 cyclotron. To accept such intense beams, the separator is equipped with beam-wobbling systems, rapidly rotating entrance windows, rotating target wheels etc. We stress also that for asymmetric reactions a very favourable separation

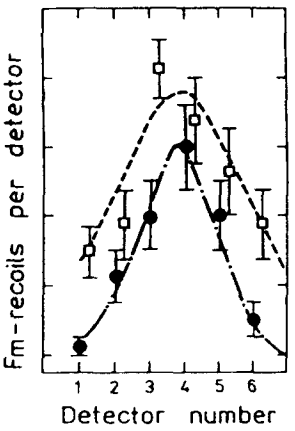


Fig. 7. Distribution of recoil nuclei Fm over the six detectors in the focal plane of the separator: □, $^{235}U(^{18}O, 5n)^{248}Fm$ reaction; ●, $^{206}Pb(^{40}Ar, 2n)^{244}Fm$ reaction.

quality lies in the extremely strong suppression of both full-energy projectiles and scattered-beam particles.

After passing through the 0.5 μm thick Mylar window and a time-of-flight ΔE system consisting of two multiwire proportional chambers in pentane at a pressure of 1 Torr, the EVR is implanted into the array of six separate Si(Au) detectors (20×30 mm^2) covering the area of 120×30 mm^2 in the focal plane.

Semiconductor detectors register the residual energy of the EVRs and are used for the subsequent observation of α and spontaneous fission decays of the implants.

The potentialities of the separator for heavy element research with asymmetric fusion–evaporation reactions can be exemplified by our recent experiments on the production of isotopes of element 104 in the $^{242}Pu+^{22}Ne$ reaction. In these experiments, the ^{22}Ne beam with a typical intensity of 1.5×10^{13} particles s^{-1} applied to a rotating target of ^{242}Pu allowed us to reach, in several days, a total beam dose of 3.6×10^{18} and to detect between cyclotron beam pulses many tens of α and spontaneous-fission decays of the isotopes $^{259}104$ and $^{260}104$ which are produced in the reaction under study with cross-sections of the order of 1 nb. A remarkable long-term stability of the separator operation was revealed in the 106 experiments.

One of the future main directions of development of our techniques is related to the updating of the Si(Au) detector array in the focal plane.

In the $^{248}Cm+^{22}Ne$ experiments instead of six separate detectors an array with an area of 120×40 mm^2 consisting of 24 position-sensitive strip detectors was used. The detectors together with their electronics and data acquisition system were developed in Berkeley and Livermore as a part of preparatory work for the joint Dubna–Livermore experiment on the synthesis of new isotopes of element 106.

We are planning to use in future an array of high resolution position-sensitive detectors (PIPS) manufactured by Canberra Semiconductor. It also consists of three 40×40 mm^2 detectors including four 10 mm strips (total, 12 strips) with position sensitivity in the vertical direction.

As compared with the initial variant (six separate detectors) which has been used in $^{242}Pu+^{22}Ne$ experiments ($\sigma_{5n}\approx1$ nb) the new system allows us to decrease

TABLE 1. Characteristics of the Dubna gas-filled recoil separator

Target	^{nat}W	$^{235,238}U$	^{235}U	^{242}Pu	^{238}U	$^{206,207}Pb$	^{207}Pb
Projectile	^{22}Ne	^{18}O	^{22}Ne	^{22}Ne	^{26}Mg	^{34}S	^{40}Ar
Z of EVRs	Po	Fm	102	104	104	Cf	Fm
Beam energy (MeV)	112	93	122	114	134	170	196
$\langle E_R\rangle$ (MeV)	11.0	5.8	8.7	7.0	11.5	21.8	28.2
$\langle q\rangle$	3.3 ± 0.1	2.1 ± 0.1	2.3 ± 0.1			4.8 ± 0.2	5.9 ± 0.2
ϵ_c (%)	16 ± 3	3 ± 1	6 ± 2[a]	6 ± 2[a]	10[a]	35 ± 10	45 ± 10
Suppression of full energy projectiles	$>10^{15}$ [a]	$>5\times10^{16}$	$>10^{17}$	$>2\times10^{18}$	$>4\times10^{17}$	$>3\times10^{15}$	
scattered projectiles with $E>35$ MeV			2×10^{16}	7×10^{16}	5×10^{15}	10^{14}	5×10^{12}
target-like products	$>10^3$			$\geqslant10^4$			5×10^4 [a]
Image size (full width at half-maximum)							
horizontal	6.8 ± 0.2	8.7 ± 1.4	12 ± 3			7 ± 1	
vertical	2.1 ± 0.1	3.5 ± 0.2					

[a]Calculated values.

the background by 2 orders of magnitude and to improve the efficiency and energy resolution for the registration and identification of extremely rare chains of superheavy nuclei decay.

6. Synthesis of nuclei near the top of the stability island

The preparation for the joint FLNR (Dubna)–LLNL (Livermore) experiment on the synthesis of neutron-rich isotopes of element 106 in the reaction $^{248}Cm + ^{22}Ne$ was started in 1990 [36]. After the technical preparation, creation of a synchronous target, detector system, selection of calibration reactions and performance of test experiments on the production of the known isotopes of element 104 in the reactions $^{242}Pu + ^{22}Ne$ and $^{238}U + ^{26}Mg$ [26], in April of 1993 there was carried out a 20-day irradiation of a ^{248}Cm target with a ^{22}Ne beam of 116 MeV and 121 MeV corresponding to the maxima of excitation functions σ_{4n} and σ_{5n} respectively.

On the basis of the correlations of events (position, time and energy of decays) the chains of sequential decays of two new isotopes of element 106 with masses 265 and 266 (Fig. 8) were restored.

It was found that the main mode of radioactive decay of these new nuclei is α decay [37]. Such properties signify that there exists a new region of superheavy elements which are extraordinarily stable against spontaneous fission.

The experimental procedure and the analysis of experimental results will be presented in a separate report by Dr. R. Lougheed at this conference and will be published later. I shall limit myself to a consideration of the results of this experiment for the discussion of possibilities of further investigations on the synthesis and investigation of properties of superheavy elements.

Figure 9 presents the calculated and experimental data on T_α and $T_{s.f.}$ of even–even isotopes of element 106. The observation of the α decay of $^{266}106$ is evidence of a significant increase in $T_{s.f.}$ (by more than 3 orders of magnitude) at the transition from the known isotope $^{206}106$ ($N=154$) to the new isotope, $^{266}106$ ($N=160$). The spontaneous fission half-lives of even–even isotopes of 102–108 obtained in cold and hot fusion reactions are presented with the general systematics of $T_{s.f.}$ (see Fig. 10). The great difference in the behaviour of $T_{s.f.}(N)$ for N-$Z=48$ and 54 is a consequence of a sharp increase in nuclei stability against spontaneous fission near deformed shells with $N=152$ and $N=162$. Thus, the experimental result confirms the main conclusions of theory about a significant growth of transactinoid nuclei stability near the shell $N=162$.

Two other circumstances referring to nuclear fission should also be noted.

(1) At the spontaneous fission of $^{266}106$ one could not exclude *a priori* the symmetric fission mode ($Z_{f1}=Z_{f2}=53$, $N_{f1}=N_{f2}=80$) leading to the formation of fragments near the magic numbers $Z=50$, $N=82$. The symmetric fission in the region of heavy isotopes of $Z=100$–102 discovered in the experiments by Hulet *et al.* [38] has been interpreted in theory by a shorter way of reaching the scission point on the potential energy surface. According to calculations made by Möller *et al.* [39] this should lead to a substantial increase in the spontaneous fission probability (Fig. 9). However, at the spontaneous fission of $^{266}106$ as well as of $^{262}104$ similar effects have not been observed and this ensures their high stability.

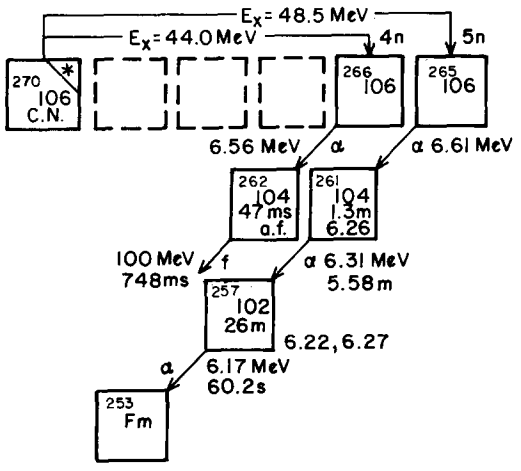


Fig. 8. Two out of the ten events demonstrating the decay of $^{265}106$ and $^{266}106$ isotopes produced in the reaction of $^{248}Cm + ^{22}Ne$. The excitation energy values for the compound nucleus of $^{270}106$ at which these events have been registered are shown in the figure.

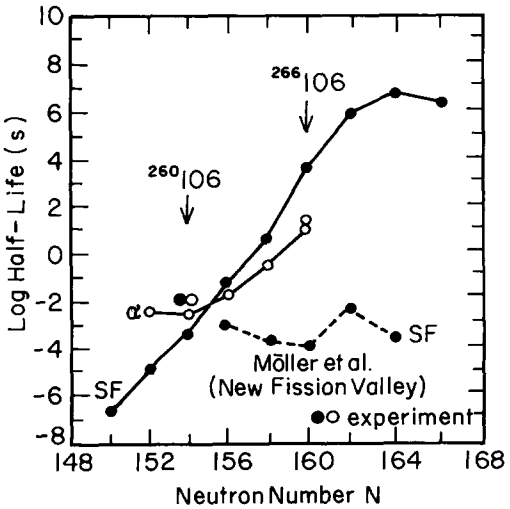


Fig. 9. The half-lives of isotopes of element 106: O, α decay half-life (seconds); ●, spontaneous fission half-life (seconds); —, drawn through calculated values of T_α and $T_{s.f.}$ [6, 7]; – – –, calculations of $T_{s.f.}$ in the symmetric fission valley [35].

(2) According to our data the daughter nucleus of $^{262}104$ produced after the α decay of $^{266}106$ has $T_{s.f.} = 1.3^{+1.6}_{-0.8}$ s. This value is approximately 30 times greater than the value $T_{s.f.} = 47$ ms obtained by Somerville *et al.* [40] in the experiments with $^{248}Cm + ^{18}O$ and tentatively referred to the decay of the nucleus of $^{262}104$. (It should be noted that in these experiments there has also been observed an activity with $T_{s.f.} = 1.3 \pm 0.1$ s.) If the identification of $T_{s.f.} \approx 47$ ms is correct then for the $^{262}104$ nucleus observed two spontaneous fission half-lives have been. This would mean that the spontaneous fission takes place from two states: the ground and the isomeric states. In spontaneously fissioning even–even isotopes of transactinoid elements such an effect could be related to the breaking of nucleon pairs leading to the formation of quasi-particle isomeric states with a strongly forbidden γ transition into the ground state. Similar isomers in the region of actinoid nuclei as well known [41, 42]. The theory predicts them also for $Z = 104$ [43].

The problem of spontaneous fission from quasi-particle states was investigated by us theoretically and experimentally [44, 45]. If the structure of the $^{262}104$ nucleus has an isomeric state of this type, there is a high probability that in the reaction $^{248}Cm(^{18}O, 4n)^{262}104$ after the emission of the last neutron in the γ transitions the quasi-particle isomeric levels ($E_x \approx 1.3$ MeV, $K^{\pi} \approx 9^-$) will be populated. At the decay of the isomer with $T_{s.f.}(is) < T_{\gamma}(is)$ in the experiment two spontaneous fission activities with $T_{s.f.}(is) \neq T_{s.f.}(g)$ will be observed; if $T_{s.f.}(is) > T_{\gamma}(is) > T_{s.f.}(g)$ then the γ decay of the isomer into the ground state will lead again to two spontaneous fission activities with $T_{s.f.}(is) > T_{s.f.}(g)$. In this situation the real value of $T_{s.f.}(g)$ can be estimated only if the nucleus is produced by the decay of the mother nucleus which takes place in the chain $^{266}106 \xrightarrow{\alpha} {}^{262}104$. So, the value of $T_{s.f.} \approx 1.3$ s obtained after the α decay of $^{266}106$ nuclei is unambiguously related to the half-life of spontaneous fission of the nucleus $^{262}104$ from the ground state.

7. Possibilities of investigating the chemical properties of element 106

Relatively large half-lives of the new nuclei and the known chains of their decay open a possibility of setting up direct experiments studying the chemical properties of element 106 (EkaW). The two approaches developed in Dubna, thermochromatography and fast continuous chemical isolation of transactinoid elements in solutions, may be used for the purpose.

A number of on-line model experiments with short-lived isotopes of tungsten in the reactions $^{144,147}Sm(^{24}Mg, \alpha n)^{163-167}W$ have been carried out recently. In the investigation by Yakushev *et al.* [46] it has been shown that in certain temperature regimes of the thermochromatographic column it is possible to separate with high efficiency the volatile chlorides and oxochlorides of W from Hf, Ta and all the lanthanoids within a time comparable with or smaller than the half-lives of the investigated isotopes ^{166}W ($T_{1/2} = 16$ s) and ^{167}W (20 s). On the contrary, it has been demonstrated by Szeglowski *et al.* [47] that in HF solutions similarly to Hf the elements Ta and W can be separated rapidly enough from lanthanoids in successive precipitations on the cation exchange and anion exchange columns. According to estimations the time of W extraction is less than 20 s; in the experiment ^{164}W ($T_{1/2} = 6$ s) has been registered. Naturally, the methods presented do not limit the scope of research and in this sense they are not unique. They demonstrate a possibility of modern radiochemistry for the investigation of fundamental chemical properties of the element 106.

It should be noted that by using the radiochemical methods of separation of $^{248}Cm + ^{22}Ne$ reaction products it becomes possible to increase substantially the efficiency of collecting EVRs as compared with that on recoil separators.

8. Prospects of synthesizing the nuclei with $Z > 106$ near the shell $N = 162$

Investigations of new isotopes of element 106 in hot fusion reactions may be continued for the neighbouring nuclei in the reactions $^{248}Cm(^{20}Ne; 4n, 5n, 6n)^{262-264}106$. Surely of the greatest interest is the synthesis of nuclei on the top of the island of stability with $Z = 108$, $N = 162$. For this purpose, by following the same line, it is possible to use the reaction $^{248}Cm(^{26}Mg; 4, 5n)^{269,270}108$. The expected cross-section σ_{4n} of the reaction in this case will be smaller than in the experiments with $^{248}Cm + ^{22}Ne$ and the intensity of the ^{26}Mg beam on the U-400 accelerator is approximately 3 times smaller than the intensity of the ^{22}Ne beam which is determined by the operation modes of the plasma ion source. On the whole this is a feasible but very difficult method.

Another possibility is more symmetric combinations, specifically the reactions of fusion with bombarding ions of sulphur. For the synthesis of the $^{270}108$ nucleus the reactions $^{238}U(^{36}S, 4n)^{270}108$ or $^{244}Pu(^{34}S, 4n)$-$^{274}110 \xrightarrow{\alpha} {}^{270}108$ can be used. According to calculations [7] the isotope $^{270}108$ will undergo α decay with $T_{\alpha} \approx 1$ s, $Q_{\alpha} = 9.44$ MeV, and will transit into the known nucleus $^{266}106$. More symmetric reactions possess in our case certain technical advantages. The recoil energy of EVRs on the Coulomb barrier will be 23.5 MeV. This increases the efficiency of EVR collection by approximately one order of magnitude (Table 1) and allows us to use

TABLE 2. Cross-sections of the reactions $^{206,207,208}Pb(^{34,36}S, xn)^{238,240}Cf$

Reaction	Method[a]	Beam energy (MeV)	Excitation energy (MeV)	Beam dose ($\times 10^{17}$)	Detected fission events	xn cross-section (nb)
$^{208}Pb(^{34}S, 2n)^{240}Cf$	WT			12.0	65	1.5
$^{206}Pb(^{34}S, 2n)^{238}Cf$	WT			3.0	244	0.3
	RS	169	33.3	1.4	26	0.3
$^{207}Pb(^{34}S, 3n)^{238}Cf$	WT			1.5	425	1.5
	RS	171	35.2	0.8	46	1.5
	RS	178	41.2	0.5	5	≈ 0.2
$^{208}Pb(^{34}S, 4n)^{238}Cf$	WT			3.0	387	0.5
$^{206}Pb(^{36}S, 2n)^{240}Cf$	WT			1.5	38	5.5
$^{206}Pb(^{36}S, 4n)^{238}Cf$	WT			0.5	86	0.7

[a]WT, wheel technique; RS, recoil separator.

thicker layers of target material. The intensity of the beam of $^{34,36}S$ ions produced from compounds with a high isotopic enrichment can reach about 5×10^{12} particles s^{-1}. Thus, a total luminosity (with account taken of the separator efficiency) can be obtained at the level of about 2×10^{30} cm^{-2} s^{-1} which allows us in the course of long irradiations to reach the ultimate cross-sections $\sigma_{min} \geqslant 1$ pb. The question is, which additional limitations can arise at the transition to more symmetric reactions?

9. Synthesis of neutron-deficient isotopes of Cf in reactions with sulphur ions

Monoisotopic targets made of $^{206,207,208}Pb$ were irradiated with $^{34,36}Sn$ and ^{40}Ar beams of the U-400 accelerator to synthesize light isotopes of californium. The experiments were performed on the internal beam by using the wheel technique and on the external beam by using a gas-filled recoil separator. The experimental procedure and the complete analysis of experimental data obtained with ions of ^{34}S and ^{40}Ar are presented in the paper by Lazarev *et al.* [48]. In Table 2 these data are supplemented with the results of recent experiments with ions of ^{36}S.

In experiments with $Pb+^{34}S$ a new 23 ms spontaneously fissioning isotope ^{238}Cf was synthesized, an indication of the existence of another isotope ^{237}Cf was obtained and partial half-lives of spontaneous fission for even–even isotopes of Cf with $N=140$, 142 and 144 were defined. The position of the new data in the systematics of the spontaneous fission half-lives is presented in Fig. 10.

The tremendous effect of the deformed neutron shell $N=152$ is demonstrated again in the region $Z=98$. The decrease in the number of neutrons in Cf isotopes from $N=148$ to $N=140$ leads to the decrease in $T_{s.f.}$ by about 3×10^{12} times.

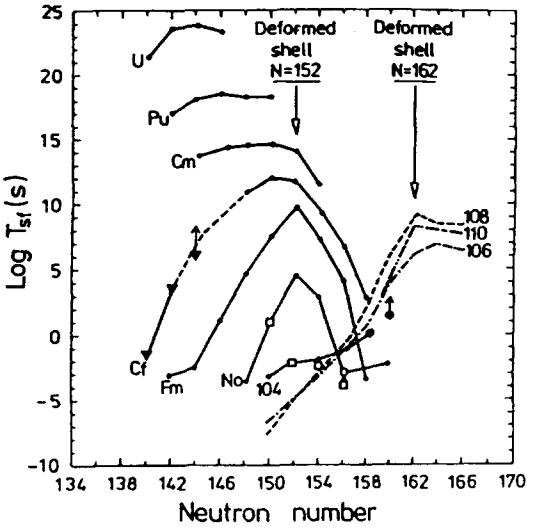


Fig. 10. Systematics of spontaneous fission half-lives $T_{s.f.}$ (s) for even–even isotopes of transuranium elements: ——, drawn through experimental points; – – –, drawn through calculated values $T_{s.f.}$ (N) for $Z=106$, 108, 110 [6]; □, $T_{s.f.}$ of isotopes with N-$Z=48$ produced in cold fusion reactions; ●, ○, isotopes with N-$Z=54$ in hot fusion reactions; ▼, data for light isotopes of Cf produced in the reaction of $Pb+^{34}S$ [43]. Arrows indicate deformed shells with $N=152$ and 162.

On the irradiation of Pb isotopes with ions of ^{34}S, $E_{x(min)} \approx 33.5$ MeV. Here the cross-section ratio σ_{xn} at $x=2$, 3, 4 is close to that for the well-studied cold fusion reaction $^{206-208}Pb+^{40}Ar$ ($E_{min}=31.5$ MeV). The summary cross-section $\Sigma\sigma_{xn}$ in the reactions $^{208}Pb(^{34}S, xn)^{242-x}Cf$ is only several nanobarns which is explained by a strong neutron deficit in this region of nuclei.

In the reactions of $^{208}Pb+^{34}S$ and $^{206}Pb+^{36}S$ leading to the production of a compound nucleus of ^{242}Cf, the growth of the yield of $^{238,240}Cf$ with ions of ^{36}S is determined by the gain in the Q reaction on the account of the ^{36}S isotope mass excess.

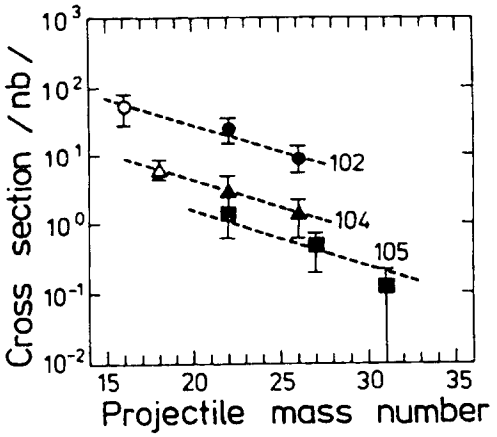


Fig. 11. The cross-sections σ_{5n} *vs.* the mass of bombarding ion: ○, ●, experimental cross-sections for (HI, 5n)^{253}102; △, ▲, (HI, 5n)^{259}104; ■, (HI, 5n)^{258}105. For $Z=105$, the cross-section of the reaction ^{243}Am(^{22}Ne, 5n)^{260}105 are also presented. ●, ▲, ■, data of ref. 25.

As expected, no obvious dynamic limitations hampering the formation of a compound nucleus in the reactions of Pb + S ($Z_1Z_2=1312$) have been found. One can assume that a similar situation will take place for U + S ($Z_1Z_2=1472$) as well. A number of experimental facts testify to the correctness of this assumption.

Figure 11 presents the results of recent experiments in Dubna on measurement of the cross-sections of ^{258}102 and ^{258}105 isotope production in the reactions (HI, 5n) by the irradiation of ^{232}Th and ^{236}U with ions of ^{22}Ne, ^{26}Mg, ^{27}Al and ^{31}P [25]. Together with the data of other experiments they give an idea about the dependence $\sigma_{5n}(A_p/A_T)$ in the region of transactinoid nuclei. The systematic decrease in the cross-section for more symmetric reactions is determined mainly by the kinematic limitations over the critical angular momentum of the compound nucleus. Simple extrapolations of the reaction cross-sections show that isotopes with $Z=108$, 110 and $N=160$–162 are still accessible for detection under the existing experimental possibilities.

10. Conclusions

The overview of data presented in this report makes possible a number of important conclusions.

(1) Asymmetric hot fusion reactions between actinide nuclei of the target and projectiles with $A \leqslant 36$ may be used for the synthesis of superheavy nuclides.

(2) Investigations of the mechanism of excited compound nuclei production and decay in the region of transactinoid elements show that the cross-sections of EVR production in hot fusion reactions are comparable and at $Z>106$ they may be higher than those in cold fusion reactions.

(3) The on-line recoil technique based on the magnetic separation of reaction products in hydrogen combined with a sophisticated detection system enables us to define the radioactive properties of new nuclei produced with cross-sections up to 1 pb.

(4) In the reactions of ^{240}Cm + ^{22}Ne two new α radioactive isotopes of element 106 with masses 265 and 266 have been synthesized and the half-life for the spontaneous fission from the ground state of the nucleus ^{262}104 has been defined. Radioactive properties of these nuclei give direct indications of the existence of the region of extraordinarily stable superheavy nuclei predicted by the macromicroscopic theory for deformed shells with $N=162$, $Z=108$.

(5) The relatively high stability of the isotopes ^{265}106 and ^{266}106 and the known chains of sequential decays make it possible to set up direct experiments studying the chemical properties of element 106 by rapid methods of modern radiochemistry.

(6) Further increase in beam intensities on heavy ion accelerators and the development of experimental methods open, it seems to me, new prospects for the synthesis of new elements of the periodic table in hot fusion reactions of nuclei.

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