ОБЪЕДИНЕННЫЙ
ИНСТИТУТ
ЯДЕРНЫХ
ИССЛЕДОВАНИЙ
ДУБНА

E7 - 10750

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Submitted to "Nuclear Physics"

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Эксперименты по синтезу изотопов сверхтяжелых элементов с атомными номерами 114-116 в реакциях с ионами ^{48}Ca

Выполнены эксперименты по синтезу изотопов 114-116 элементов в реакции слияния ионов ^{48}Ca с изотопами $^{246,248}Cm$, ^{243}Am и ^{242}Pu. Облучения ионами ^{48}Ca с энергией 267 МэВ производились на внутреннем пучке циклотрона У-300 ЛЯР ОИЯИ, средний ток ионов ^{48}Ca был 10^{12} част/сек. Так как, в соответствии с теоретическими предсказаниями, сверхтяжелые элементы с Z = 112-116 должны обладать повышенной летучестью, а элементы с Z = 108-111 должны быть аналогами тяжелых металлов с Z = 76-79, после облучения из мишени химическими методами выделялись фракции летучих элементов, тяжелых благородных газов, сульфидов тяжелых металлов, а также фракция Cf-Fm.

Полученные образцы (80%) были помещены в контакт с фосфатными стеклами для детектирования спонтанного деления сверхтяжелых элементов с периодом 2 часа $\leq T_{sf} <$ 1 год. В то же время часть образцов (20%) была помещена в альфа-спектрометр. Никаких неизвестных спонтанноделящихся и альфа-активных излучений обнаружено не было. Получены предельные сечения образования сверхтяжелых элементов в изучавшихся реакциях: для спонтанно-делящейся активности $\sigma < 5 \cdot 10^{-35}$ см^2, для альфа-активных изотопов с периодом 2 часа $\leq T_\alpha <$ 10 дней $\sigma < 5 \cdot 10^{-34}$ см^2.

Препринт Объединенного института ядерных исследований. Дубна 1977

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E7 - 10750

Experiments to Produce Isotopes of Superheavy Elements with Atomic Numbers 114-116 in ^{48}Ca Ion Reactions

Experiments have been performed to synthesize isotopes of elements 114-116 in fusion reactions between ^{48}Ca ions and $^{246, 248}Cm$, ^{243}Am and ^{242}Pu target nuclei. The detection of reaction products by recording spontaneous fission and α-decay events allowed one to set the upper limits of production cross sections for superheavy elements with 2 h $\leq T_{sf} \leq$ 1 y and 2 h $\leq T_\alpha \leq$ 10 d at 5×10^{-35} cm^2 and about 2×10^{-34} cm^2, respectively.

The investigation has been performed at the Laboratory of Nuclear Reactions, JINR.

Preprint of the Joint Institute for Nuclear Research. Dubna 1977

INTRODUCTION

In recent years, intensive experimental studies have been conducted to verify the hypothesis concerning the existence of the theoretically predicted[1-3] region of enhanced stability of superheavy nuclei.

At present the use of heavy-ion reactions seems to be the only effective means of synthesizing the Z > 110 nuclei. As known, all transfermium elements with Z ≤ 105 have been produced in complete fusion reactions between the target nucleus and the heavy ion with the subsequent evaporation of several neutrons. The isotopes of Pu, Cm and Cf were used as target nuclei, and the isotopes of comparatively light elements such as O and Ne were employed as projectiles.

The production of superheavy elements by this method requires heavier ion beams. However, the results of the experiments involving nuclear reactions with Ar[4], Zn[5], Ge[6] and Kr[7] ions allowed one to set only the upper limits of the production cross sections for isotopes with Z > 112.

The negative result of these experiments may be explained either by a low production probability for fusion reaction products

or by the decay properties of the superheavy elements isotopes produced.

As known, the yield of the (HI,xn) reaction products depends substantially on the excitation energy of the compound nucleus. As the excitation energy increases, the probability of production of the final nucleus as a result of the successive neutron evaporation decreases sharply. At the same time, the compound nucleus minimum excitation energy, which is equal to the sum of the Coulomb barrier height, B_c, and the Q value of the fusion reaction, was 40-50 MeV for practically all the reactions investigated. This is the possible reason why the yield of complete fusion reaction products was so low that superheavy nuclides could not be detected experimentally.

On the other hand, it is not excluded that the isotopes that might be produced in these experiments have very short lifetimes in their ground states due to the fact that they are removed by several mass units from the centre of the island of stability ($Z = 114$, $N = 184$).

If one considers different target-projectile combinations in terms of the features of the reaction mechanism and the properties of final products, the use of the ^{48}Ca projectile [8-10] to synthesize superheavy elements seems most advantageous. The fusion of ^{48}Ca ions with the heavy isotopes of Pu, Am, and Cm may in principle lead to the synthesis of nuclides with the proton number $Z = 114$-116 and neutron number $N = 172$-178. Mere calculations show that the minimum value of the compound nucleus excitation energy is expected to be as low as 20-25 MeV [11].

Despite such vivid advantages, the ^{48}Ca ions were not accelerated until 1975. The reason for this was the complicated separation of the isotope ^{48}Ca from the naturally occuring isotopic mixture and its high cost. In addition, conventional ion sources operated with gas cannot be used to produce multiply-charged ^{48}Ca ions.

After the development of a new type of multiply-charged ion source [12], at the 310-cm heavy ion cyclotron of the JINR Laboratory of Nuclear Reactions a $^{48}Ca^{7+}$ ion beam with an intensity of 10^{12} part/s has been produced, the ^{48}Ca consumption rate being about 4 mg/h.

In initial experiments with ^{48}Ca ions, the yields of the products of the reactions $^{206-208}Pb(^{48}Ca,xn)^{252}102$ have been determined [12]. It has been shown that the maximum cross section characterizes the reaction involving the emission of two neutrons. Note that the cross section of the reaction $^{206}Pb(^{48}Ca,2n)^{252}102$, equal to 5×10^{-31} cm^2, is nearly a factor of 40 larger than those of the reactions $^{239}Pu(^{18}O, 5n)^{252}102$ and $^{235}U(^{22}Ne,5n)^{252}102$ leading to the same isotope [13].

These experimental data indicate that the use of ^{48}Ca ions as projectiles provides in fact unique possibilities for synthesis of transfermium elements. It should however be born in mind that the synthesis of superheavy elements in this way requires deformed target nuclei with $Z \gtrsim 92$. It is not excluded that the mechanism of the reaction involved in this case may have some specific features compared with reactions on the magic Pb nuclei.

As known, the cross section for the formation of fusion reaction products can be defined by the expression [14]

$$\sigma_{xn} = \sigma_c P_{xn}(E) \left(\frac{\Gamma_n}{\Gamma_f}\right)^{X(E)},$$

where σ_c is the compound nucleus cross section, P_{xn} is the probability of emission of X neutrons at a given excitation energy, Γ_n/Γ_f is the neutron-to-fission ratio averaged over the whole evaporation cascade.

The value of $\sigma_c(E)$ for a given target-projectile combination depends on many factors influencing the mechanism of fusion between the colliding nuclei. In a number of theoretical studies it has been shown that for a certain ratio between the masses of the projectile and target nucleus the compound nucleus formation probability should be very small compared with the total reaction cross section [15-19]. The results of experiments with Kr [20,21] and Ge [32] ions to study the fission products of compound systems are usually quoted as evidence for the validity of this point of view.

At the same time the measurements of the energy and mass distributions of correlated fission fragments from the U+Ar reaction [25,26] have shown that the magnitude of the compound nucleus cross section for this reaction makes up 50-70% of the total reaction cross section.

One may assume that as one goes to somewhat heavier ions, such as ^{48}Ca, the cross section for the nuclear fusion reaction should form a considerable portion of the total reaction cross section. An indirect substantiation of this assumption is provided

by a large amount of the experimental data obtained recently in experiments to synthesize isotopes with Z =100-107 in reactions induced by ^{40}Ar, ^{48,49,50}Ti, ^{52,53,54}Cr, ^{55}Mn and ^{56,58}Fe on Tl, Pb and Bi [26-29] as well as in experiments to study fission of U and Th by ^{40}Ar [30] and ^{48}Ca [31] ions.

Another important factor determining the yield of fusion products in the ground state is the Γ_n/Γ_f ratio which depends on the mass and charge of the compound nucleus and also on its excitation energy. According to theoretical calculations [32] the value of the Γ_n/Γ_f ratio for nuclei in the vicinity of the island of stability may amount to $10^{-1} - 10^{-2}$ at excitation energies of 30-40 MeV. However, as the isotopes of interest lie far beyond the region of known nuclei, it is difficult to evaluate the accuracy of such calculations. Nevertheless it is natural to assume this value to have a certain limit determined by the ratio of the characteristic nuclear fission time, τ_f, to the time of neutron emission, τ_n. We have estimated this limit to be $\tau_f/\tau_n \sim 10^{-3}$. Taking into account these qualitative arguments the cross section for the fusion reaction involving the subsequent evaporation of two or three neutrons is expected to be equal to $\sim 10^{-33} - 10^{-34}$ cm^2.

Now we consider the possible radioactive decay properties of superheavy nuclei. For isotopes with Z=114-116 producible in the fusion of ^{48}Ca ions with ^{242}Pu, ^{243}Am and ^{246,248}Cm, the main decay mode may be α-particle emission or electron capture, or spontaneous fission.

At present the estimates exist for α-decay and β-decay half-lives based on different nuclear mass formulae. For the nucleides of interest the α-decay and β-decay half-lives are predicted to lie between several seconds and several days [1,2]. One can expect that the errors involved in these estimates do not exceed one or two orders of magnitude. At the same time, theoretical predictions concerning the spontaneous-fission half-lives of superheavy nuclei may differ by a factor of 10^3 - 10^8.

It is necessary to note that as one goes farther from the centre of the island of stability due to α-decay or electron capture, the spontaneous fission probability will increase sharply. Therefore, irrespective of the accuracy of the theoretical estimates, the nuclei produced in fusion reactions, or the products of their α- or β-decay should undergo predominantly spontaneous fission. Thus, to observe superheavy elements in reactions of ^{48}Ca with Pu, Am and Cm isotopes we have employed the efficient and selective method of detecting spontaneous fission fragments.

EXPERIMENTAL

According to theoretical estimates [2,3] spontaneous-fission or α-decay half-lives of the isotopes produced in the fusion reaction between ^{48}Ca ions and Pu, Am and Cm target nuclei may range from 1 hour to 1 year. Therefore the experiments were designed to detect long-lived nuclides.

Each experiment consisted of three successive stages, namely, the accumulation of reaction products during the irradiation, the chemical isolation of superheavy elements and, finally, the measurements of the α- and spontaneous-fission activities of the samples obtained. Such a procedure provided a maximum experimental sensitivity due to the nearly 100% efficiency of detecting spontaneous fission events and the removal of practically all spontaneously fissioning background activities.

The irradiations were carried out using an internal beam of the 310-cm heavy ion cyclotron of the JINR Laboratory of Nuclear Reactions. The experimental conditions are given in table 1. The target was prepared by depositing a Pu, Am and Cm oxide layer onto a golden backing serving also as a recoil catcher. The ^{48}Ca ion energy at the entrance into the target was 267 MeV, and the energy spread did not exceed 1.5%. The energy loss in the target material was 10-15 MeV. The beam intensity was measured continuously during the irradiation and was equal, on the average, to 10^{12} part/s. The power released on the target was 40 W. As the superheavy nuclei producible by reactions may be highly volatile, special measures were taken to provide a reliable cooling of the target. The target golden backing supplied with a water-cooling system was placed at an angle of 20° with respect to the beam direction, which increased the surface irradiated by a factor of 3. The target temperature during the irradiations did not exceed 40°. After each irradiation the target substance was

Table 1

Reaction	Target effective thickness	Target isotopic composition	B_{Coul}	Total ion flux on target	Irradiation time
Cm+ ^{48}Ca	2 mg/cm^2	^{248}Cm 8 % ^{246}Cm 67 % ^{244}Cm 15 %	231 MeV 232 MeV	2.5x10^{16}	10 h
Am + ^{48}Ca	35 mg/cm^2	^{243}Am 94 % ^{241}Am 7 %	230 MeV	2.3x10^{16}	12 h
Pu + ^{48}Ca	2 mg/cm^2	^{242}Pu 95 % ^{239}Pu 5 %	228 MeV	5.3x10^{16}	20 h

washed off from the golden backing with nitric acid. Then the upper gold layer 15 μm thick was removed mechanically. The effective thickness of the target was chosen so that practically all superheavy nuclei produced in fusion reactions were stopped in the layer to be removed. Then this layer was treated chemically with the purpose of isolating superheavy elements.

It is commonly accepted [33,34,35] that elements 112-116 should be rather volatile, even more volatile than their chemical homologs Hg-Po. It is not excluded that elements 112-114 may be close in volatility to noble gases [36]. This characteristic feature of elements 112-116 was used to extract them from the golden catcher.

Based on a series of experiments with the homologs of the elements mentioned, we have chosen the following scheme of experiments (fig. 1).

The gold was placed for 60 minutes into a quartz tube heated to a temperature of 1020±30° in hydrogen flow. The gas containing evaporated products was passed through quartzose sand for a best retention of the less volatile elements. In the next section the tube walls were covered with a copper alloy foil 1.7 μm thick. Here the gas was gradually cooled from 800°C to the room temperature. The next section has the walls covered with silver foil. Then the gas passed through two traps cooled with liquid nitrogen. The walls of the first trap in which heavy noble gases condensed were coated with mylar film to detect spontaneous fission events. After the experiment this trap was disconnected and hermetically sealed so that

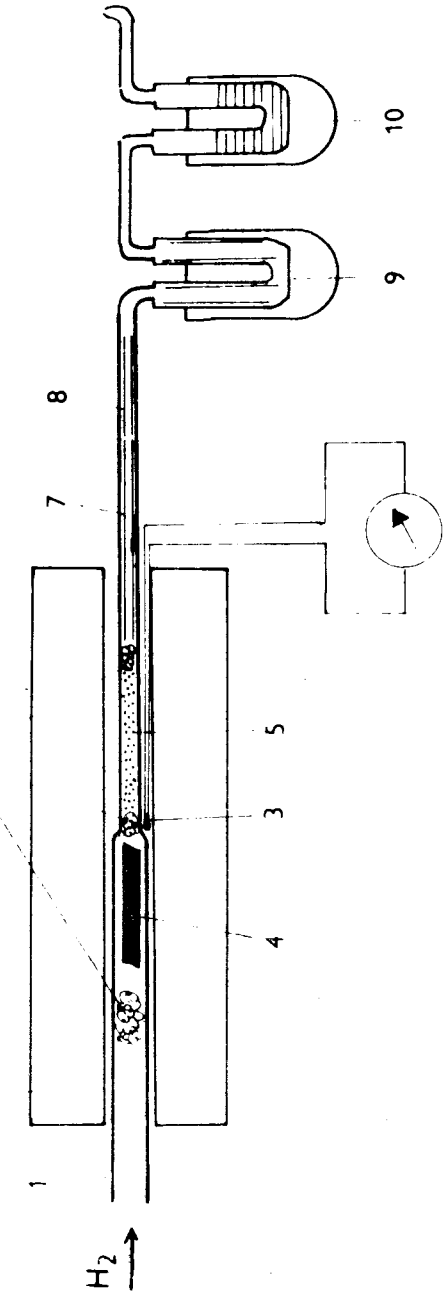

Fig. 1. Scheme of the experimental arrangement for the chemical separation of superheavy elements: (1) quartz tube, (2) heater, (3) thermocouple, (4) gold catcher foil, (5) quartzose sand, (6) quartz wadding, (7) copper alloy foil, (8) silver foil, (9) mylar trap, (10) activated carbon trap.

one could record the spontaneous fission activity of the gas products accumulated during a long period of time. The second trap contained activated carbon for the final purification of hydrogen.

The typical distribution of the chemical elements produced during the irradiation was obtained by measuring the α- and γ-spectra and is shown in table II. It should be emphasized that a very high factor of decontanimation from involatile elements, $>10^9$, reached at such one-stage treatment, leads to the complete removal of the background due to actinides in searches for α-decay and spontaneous fission events of elements 112-116. The behaviour of these elements is expected to be similar to that of their homologs precipitated on the copper, silver and mylar foils.

As it cannot be excluded that isotopes with Z=112-116 may appear to be short-lived with respect to α-decay and electron capture, the target may contain their daughter isotopes with Z=108-111, the chemical homologs of noble metals [33]. One of their common features that we used to isolate them is the ability of forming stable bromide complexes in solutions and coprecipitating with insoluble sulphides. This property should also be characteristic of elements 112-116. Therefore even if their volatility is insufficient to evaporate them during the heating of the golden catcher, these elements can be isolated at the second stage of the treatment.

The gold was dissolved and transformed to a solution of bromide in dilute hydrobromic acid. From this solution we isolated

Table II

Distribution of the elements produced in nuclear reactions after heating

Gold	platinum group metals 100%; actinides 40-60%; Sc , K , Rb : 20%
Quartz tube	actinides 60...40%; Sc , K , Rb 80%
Quartz sand	traces of K , Rb , Pb , Bi , and Tl
Copper alloy and silver foils	Hg , Tl , Pb , Bi , Po , At > 90%; actinides: < 10 %
Mylar trap	Hg < 10%; Xe > 90%
Activated carbon trap	Ar, Kr > 90%

actinides by use of cation exchange resin, which allowed one to obtain the Cf–Fm fraction, and by using electrolysis on a backing, to prepare sources with a layer thickness corresponding to the absorption of not more than 20 keV for E_α = 5.8 MeV. By extracting tributylphosphate from the remaining solution we removed gold and thallium. Then, after the adding of microquantities of Cu and Pb sulphides were precipitated by hydrogen sulphide. The chemical yield of the VI Perion elements in the precipitate was not less than 90%. The required decontamination from actinides has been achieved. Sources for α-ray spectrometry have been manufactured from 20% of the deposit, while the remaining substance was deposited on phosphate glasses.

The activity of the samples with respect to spontaneous fission and their α-spectra have been measured. The samples obtained by sublimation methods were measured in two hours, and the samples obtained at the second stage of the treatment were analysed in 10 hours after the irradiation. To detect spontaneous fission fragments the samples were put in contact with phosphate glasses. The detection efficiency was not less than 90%. After certain intervals of time the detectors were replaced, which permitted, in principle, the measurement of half-lives from 2 hours to a year.

To record α-radiation, 25% of the area of the copper and silver catcher foils, the sample containing 20% of the activity of the sulphide fraction and the whole Cf–Fm fraction were placed in an α-ray spectrometer with Si(Au) semiconductor detectors. The effi-

ciency of the detectors was 30%, and the energy resolution was not worse than 50 KeV. The device intrinsic background at $E_\alpha > 6$ MeV did not exceed 2 pulses per day.

In measuring spontaneously fissioning activity during the exposure time of, on the average, 200 days, no tracks due to fission fragments have been detected. In the mylar of the trap containing the fraction of heavy noble gases, no tracks of spontaneous fission fragments have been recorded either. As a result, we estimate the upper limit of the production cross section to lie near 5×10^{-35} cm^2 (table III).

A spontaneously fissioning activity has been observed only in the Cf-Fm fraction. In the reaction ^{246,248}Cm+^{48}Ca the total number of the fission events detected during 150 days in this fraction was 2×10^3. On the basis of the time distribution of fission fragments this activity and the effect observed in the actinide fractions in other experiments could be fully assigned to spontaneous fission of ^{254}Cf ($T_{1/2}$ = 60.5 days), produced in transfer reactions.

In studying α-spectra we also observed some lines due to transfer reaction products. As an example, fig. 2(a,b,c) shows the spectra obtained in the experiment with a curium target. In the spectrum of the volatile products precipitated on the copper catcher (fig. 1a) one can see the lines corresponding to the known isotopes of At , Po , Pb and Bi , which are produced in transfer reactions on the golden backing. The study of the α-spectra of actinide fractions (fig.1c) permitted the estimation of the cross section for the production of the isotopes

TABLE III

Chemical fraction	Reaction products	Decay mode	Lifetime	Cross sections(cm^2) for different targets ^{246}Cm	^{248}Cm	^{243}Am	^{242}Pu
Volatile metals	112-116	s.f.	2h - 1y	$< 2 \times 10^{-35}$	$< 2 \times 10^{-34}$	$< 1.5 \times 10^{-35}$	$< 10^{-35}$
		α-decay 7MeV$<E_\alpha<$9MeV	2h -10d	$< 5 \times 10^{-34}$	$< 5 \times 10^{-33}$	$< 4 \times 10^{-34}$	$< 2.5 \times 10^{-34}$
Sulphides of heavy metals	108-111	s.f.	10h- 1y	$< 2 \times 10^{-35}$	$< 2 \times 10^{-34}$	$< 1.5 \times 10^{-35}$	$< 10^{-35}$
		α-decay 7MeV$<E_\alpha<$9MeV	10h-10d	$< 2 \times 10^{-34}$	$< 2 \times 10^{-33}$	$< 1.5 \times 10^{-34}$	$< 10^{-34}$
Heavy noble gases	112-116	s.f.	2h - 1y	$< 2 \times 10^{-35}$	$< 2 \times 10^{-34}$	$< 1.5 \times 10^{-35}$	$< 10^{-35}$
Cf-Fm	^{254}Cf	s.f.	60.5d	2.4×10^{-31}		7×10^{-34}	$< 2 \times 10^{-34}$
	^{246}Cf	α-decay E_α=6.7MeV	35.7h	3.6×10^{-28}		3×10^{-31}	10^{-32}
	^{252}Fm	α-decay E_α=7.04MeV	22.8h	5.1×10^{-32}		$< 10^{-33}$	$< 10^{-33}$

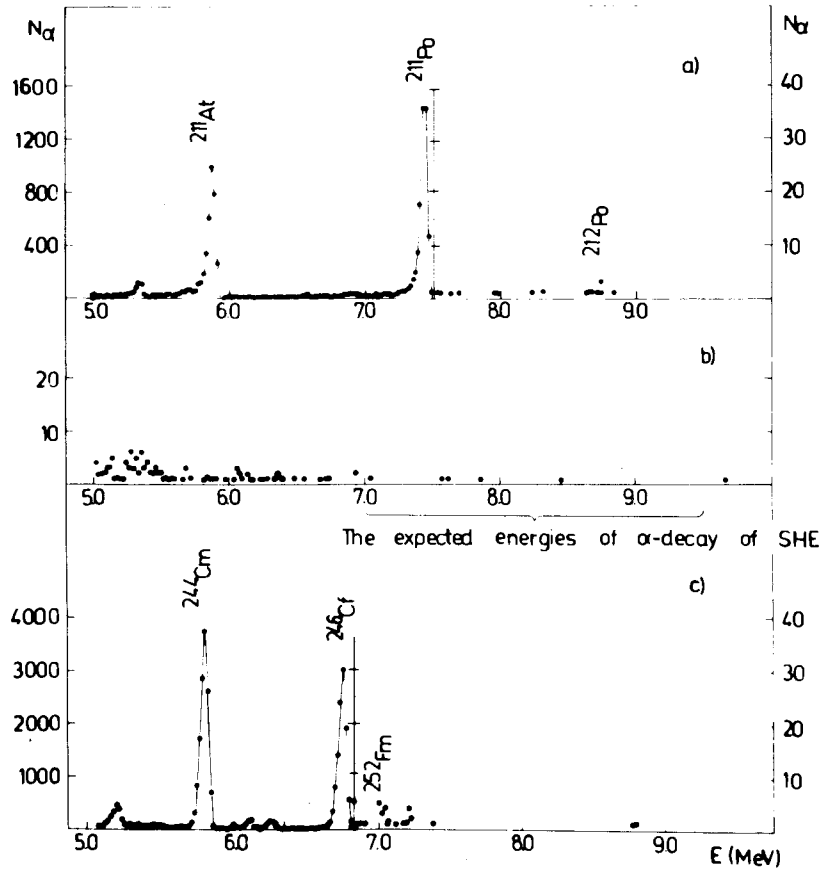

Fig. 2. The α-spectra of the gaseous (a), sulphide (b) and californium (c) fractions, measured in the experiment with a curium target.

^{246}Cf and ^{252}Fm in multi-nucleon transfer reactions. The values of the cross sections for the production of some transfer reaction products are listed in table III.

In the α-spectra of sulphide fractions (fig. 1c) no lines corresponding to the

α-particle energy of $\geq$ 7 MeV has been observed. In the spectra of gaseous and actinide fractions no unknown emitters with an energy of α-particles higher than 6.5 MeV has been recorded either. The values of the upper limits of the cross sections obtained are listed in table III.

We thought it appropriate to carry out an experiment to search among the products of the reactions studied for comparatively short-lived spontaneously fissioning emitters. Since for the reaction ^{243}Am + ^{48}Ca the spontaneous fission background due to long-lived isotopes was practically absent, in this experiment ^{243}Am was used as a target nucleus. The experimental technique permitted, in principle, the detection of nuclides with lifetimes >10 ms. The nuclei recoiling from the target were collected on a thin rotating disk. Fission fragments were recorded by means of mica detectors. The irradiation time was 20 hours. In the mica, over 200 tracks due to fission fragments have been recorded. Their time analysis has shown that only one track may correspond with a probability of 95% to spontaneous fission of a nuclide with a half-life of $\geq$100 ms. The rest of the tracks were entirely due to the formation of the spontaneously fissioning isomer ^{242}Am ($T_{1/2}$ = 13.7 ms) in the transfer reaction. Thus in this experiment we succeeded in setting the upper limit of the production cross section for spontaneously fissioning isotopes with lifetimes longer than 100 ms at 3×10^{-35} cm^2.

DISCUSSION OF RESULTS

The fact that no unknown spontaneously fissioning or α-active emitters were observed among the products of the reactions studied may be due either to a low probability of SHE production in fusion reactions compared with that assumed, or due to that the isotopes produced are too short-lived to be detected by the experimental technique chosen.

However, if our understanding of the mechanism of the fusion reaction, based on known experimental data, is valid for this region of compound nuclei, the results of the experiments allow one to set the upper limits of the lifetimes of the corresponding nuclides, i.e., $T_{1/2} \leq 2$ h in the case of the reactions $^{246}Cm + ^{48}Ca$ and $^{242}Pu + ^{48}Ca$, and $T_{1/2} \leq 100$ ms for the reaction $^{243}Am + ^{48}Ca$.

It is interesting to compare the values obtained with some theoretical predictions.

The height and shape of fission barriers for superheavy nuclei were calculated by different authors [1-3]. In particular, calculations predict that nuclei with Z near 114 and N around 184 should have increased stability with respect to spontaneous fission. It has however been indicated earlier that the absolute values of estimates may strongly differ from each other. For instance, the calculation of Fiset and Nix [2] gives the value of the spontaneous-fission half-life for the most stable doubly-even isotope $^{298}114$ to be about 10^{15} years, whereas in the calculation of Randrup et al. [37] this quantity is predicted to be about 10^{8} years. Still stronger discrepancies in the estimates

for spontaneous-fission half-lives take place for nuclei removed from N=184 by several neutrons.

The theoretical predictions for nuclear lifetimes made by Fiset and Nix [2] and Rundrup et al. [37] are shown in fig. 2. The same figure shows also the upper limits of lifetimes for odd-neutron isotopes, obtained in our experiments involving reactions with $^{246,248}Cm$, ^{243}Am, ^{242}Pu as well as ^{233}U, ^{231}Pa and ^{232}Th targets.

By assuming the hindrance factor for spontaneous fission for even-odd nuclei to lie between 10 (ref. [38]) to 10^3, and between 10^2 and 10^5 for doubly-even nuclei, one can estimate the upper limits of the lifetimes of the corresponding doubly-even isotopes. These estimates are also presented in fig. 3.

If we compare the results of our experiments with the theoretical predictions mentioned (see fig. 3), one can see that the upper limits of the lifetimes of isotopes with Z =114-116 and N=176-178 are somewhat lower than the values calculated by Fiset and Nix. At the same time, the calculation of Randrup et al. predicts for nuclei in this region lifetimes 4-5 orders of magnitude smaller than the obtained limits, and this does not contradict the experimental results.

We believe that a final reply to the question concerning the existence of the island of stability may be received after performing further experiments including those with ^{48}Ca ions to synthesize superehavy elements. The technique of these experiments should be efficient enough to detect spontaneously fissioning reaction products with lifetimes of 10^{-2} - 10^{-10} s.

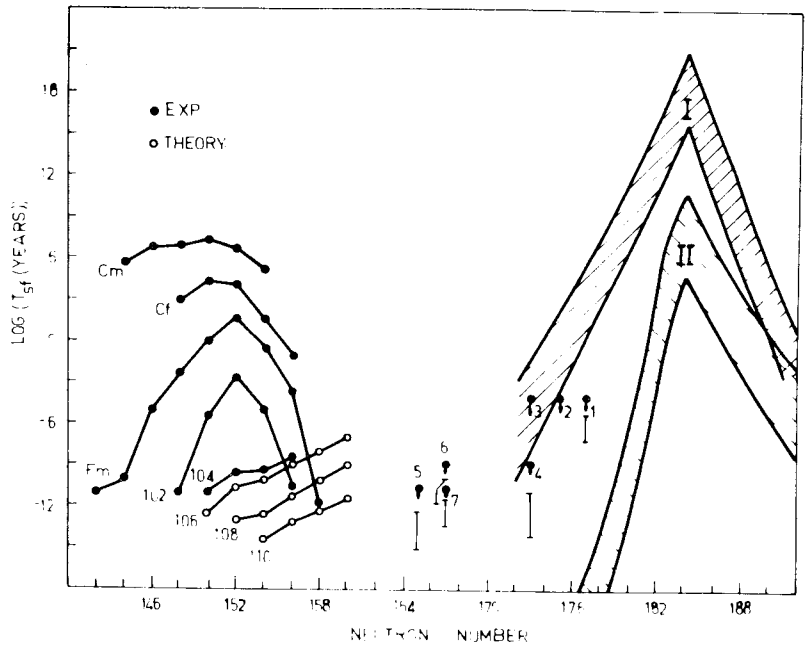

Fig. 3. Systematics of spontaneous-fission half-lives of doubly-even isotopes with Z = =100-104 and theoretical estimates for superheavy nuclei. Regions I and II correspond to the calculations of Fiset and Nix [2] and Randrup et al. [37], respectively. The upper limits of the spontaneous-fission half-lives obtained in experiments with ^{248}Cm (1), ^{246}Cm (2), ^{242}Pu (3), ^{243}Am (4), as well as ^{231}Pa (5), ^{233}U (6) and ^{232}Th (7) are also presented.

The authors are thankful to the USSR State Committee for Utilization of the Atomic Energy for providing the enriched ^{48}Ca isotope, the U-300 Cyclotron staff for the most efficient operation of the accelerator, and Professor G.N.Flerov for his constant support of the work and numerous fruitful discussions.

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Received by Publishing Department
on June 13, 1977.