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Эксперименты по синтезу нечетных нейтронодефицитных изотопов сверхтяжелых элементов в реакциях с ионами ^{48}Ca

Проведены эксперименты по синтезу нейтронодефицитных изотопов сверхтяжелых элементов в реакциях $^{233}U(^{48}Ca,2n)$, $^{231}Pa(^{48}Ca,3n)$ и $^{232}Th(^{48}Ca, 2,3n)$.

Получены верхние пределы сечений образования и периодов полураспада сверхтяжелых элементов

$^{279}112$ - 7.10^{-35} см^2, 50 мс,
$^{276}111$ - 5.10^{-35} см^2, 3 мс,
$^{277-276}110$ - 2.10^{-35} см^2, 3 мс, соответственно.

Работа выполнена в Лаборатории ядерных реакций ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна 1977

Ter-Akopian G.M. et al. **E7 - 10722**

Experiments to Produce Odd-Mass Neutron-Deficient Isotopes of Superheavy Elements by ^{48}Ca Ion-Induced Reactions

Experiments to produce neutron-deficient isotopes of superheavy elements in the reactions $^{233}U(^{48}Ca, 2n)^{279}112$, $^{231}Pa(^{48}Ca,3n)^{276}111$ and $^{232}Th(^{48}Ca,2-3n)^{277-276}110$ have been carried out using a heavy ion beam of the U-300 accelerator. In these reactions, the upper limits of production cross sections and spontaneous-fission half-lives have been determined to be $7x10^{-35}$cm^2, $5x10^{-35}$ cm^2 and $2x10^{-35}$ cm^2, and 0.05 s, 0.003 s and 0,003 s, respectively.

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

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1. INTRODUCTION

Many attempts are currently being made to verify the hypothesis concerning the existence of superheavy elements. In recent years, several groups have tried to synthesize them in complete fusion reactions induced by ^{40}Ar /1/, ^{68}Zn /2/, ^{76}Ge /3/ and ^{84}Kr /4/ ions. However, none of these experiments have proved the existence of superheavy elements.

Different authors /3,5-10/ gave possible reasons for the failure of these experiments. Based on the analysis of the data presented in refs. /1-4/ one can note that practically all the reactions investigated are characterized by relatively high excitation energies ($E^* \geq 50$ MeV). It is natural that in such highly excited states shell effects are strongly suppressed and, as a result, the cross sections for the ground-state production of final nuclei may be too small to be detected by the experimental apparatus.

On the other hand, as shown in ref. /11/, the excitation energy of compound nuclei depends substantially on the target-projectile combination chosen. This circumstance has been employed to produce isotopes of elements with atomic numbers ranging from 100 to 107 in complete fusion reactions between Tl, Pb and

Bi targets and bombarding ions from ^{40}Ar to ^{58}Fe (refs. [12-15]). In tackling the problem of synthesis of superheavy elements one should bear in mind the advantages of ^{48}Ca-induced fusion reactions. These advantages have been demonstrated for the reactions $^{206-208}Pb(^{48}Ca, xn)^{252}102$ in ref. [16].

Naturally the possibility of producing by ^{48}Ca reactions compound nuclei lying in the vicinity of the island of stability (Z=114, N=184) looks very attractive. However, in the present paper we would like to draw one's attention to a circumstance that might be helpful in synthesizing neutron-deficient isotopes of superheavy elements. If one assumes that these isotopes and their daughter products undergo α-decay with higher probability than spontaneous fission ($T_\alpha < T_{s.f.}$), then the sequential α-decay will lead to the formation of nuclei with well known properties. These nuclei may be detected with a good efficiency against the background of an enormous number of reaction by-products.

For instance, in the reactions we used, $^{233}U(^{48}Ca, 2n)^{279}112$ and $^{231}Pa(^{48}Ca, 3n)^{276}111$, the sequential α-decay of the nuclei $^{279}112$ and $^{276}111$ might lead to the daughter nuclides ^{255}Fm (T_α = 20.1 h, E_α = 7.01 MeV) and $^{256}Md \xrightarrow{E.C.} {}^{256}Fm$ (T_{sf} = 2.6 h) (see fig. 1). The detection of the ^{255}Fm and ^{256}Md decay or its absence would enable one to establish the relation between the spontaneous fission and α-decay probabilities for the Z = 111, 112 nuclei. The latter probability can be estimated using semiempirical mass formulae with an accuracy considerably exceeding that of the estimate spontaneous fission probability.

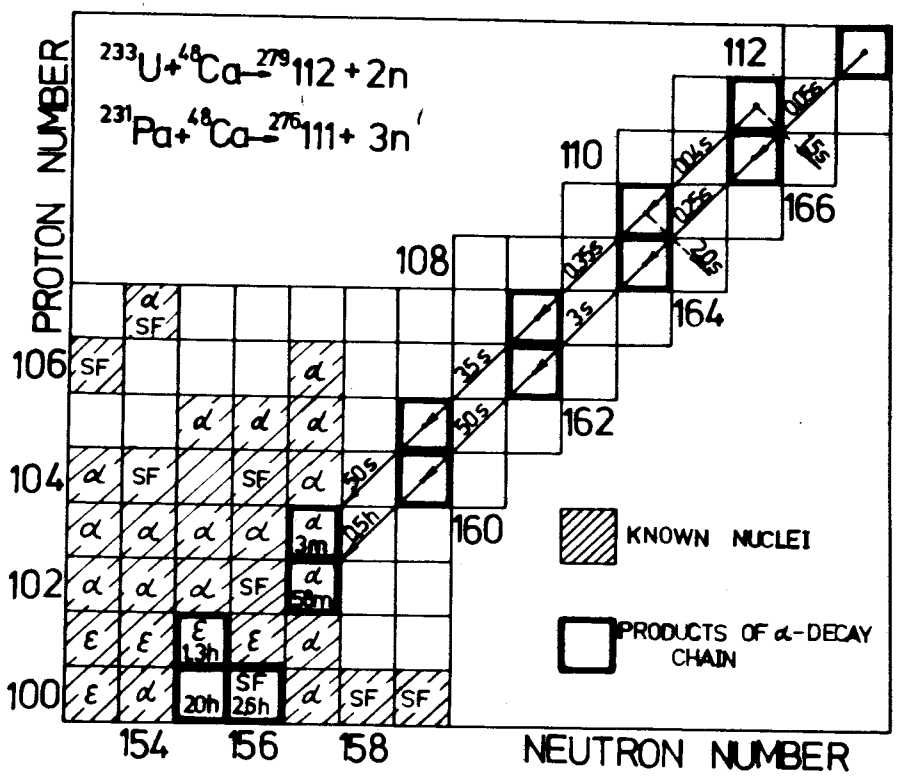


Fig. 1. Chart of isotopes of transfermium elements. The sequential α-decays of the isotopes $^{276}111$ and $^{279}112$ that might be produced by the reactions $^{231}Pa + ^{48}Ca$ and $^{233}U + ^{48}Ca$ are shown. The decay products are shown by solid lines. The calculated α-decay and electron capture half-lives are indicated above the arrows. For known isotopes the main decay modes are shown.

In particular, to estimate T_α we used the semiempirical formula given in ref. [17]. The corresponding α-decay energies were calcu-

lated by the mass formula [18]. The T_α values obtained for all products of the sequential decays of the mother nuclei $^{276}111$ and $^{279}112$ are listed in fig. 1. As seen in this figure, the α-decay half-lives of the initial nuclei $^{276}111$ and $^{279}112$ are calculated to be $T_\alpha \sim 0.05$ s.

2. EXPERIMENTAL

Targets made of ^{233}U and ^{231}Pa were bombarded in an internal beam from the U-300 heavy ion cyclotron. The maximum energy of the accelerated $^{48}Ca^{7+}$ ions was 255 MeV. The experiments to study the reaction $^{233}U+^{48}Ca$ were performed using a target prepared by depositing a 2 mg/cm^2 U_3O_8 layer onto a 1.4 mg/cm^2 aluminium backing. The energy loss of the ^{48}Ca ions in the target was about 15 MeV. Behind the target was placed an aluminium recoil catcher 2.7 mg/cm thick. The target and catcher temperature did not exceed 60°C during the bombardment. The bombardment duration was about 20 hours the integral ^{48}Ca ion flux amounted to 2×10^{16} particles.

After the irradiation the target and catcher foil were treated chemically to separate transplutonium elements. For this purpose the recoil catcher dissolved in hot concentrated nitric acid in the presence of a carrier (1 mg of lanthane). To check the separation and chemical yield, ^{169}Yb, , ^{159}Dy , ^{151}Gd and ^{141}Ce were used

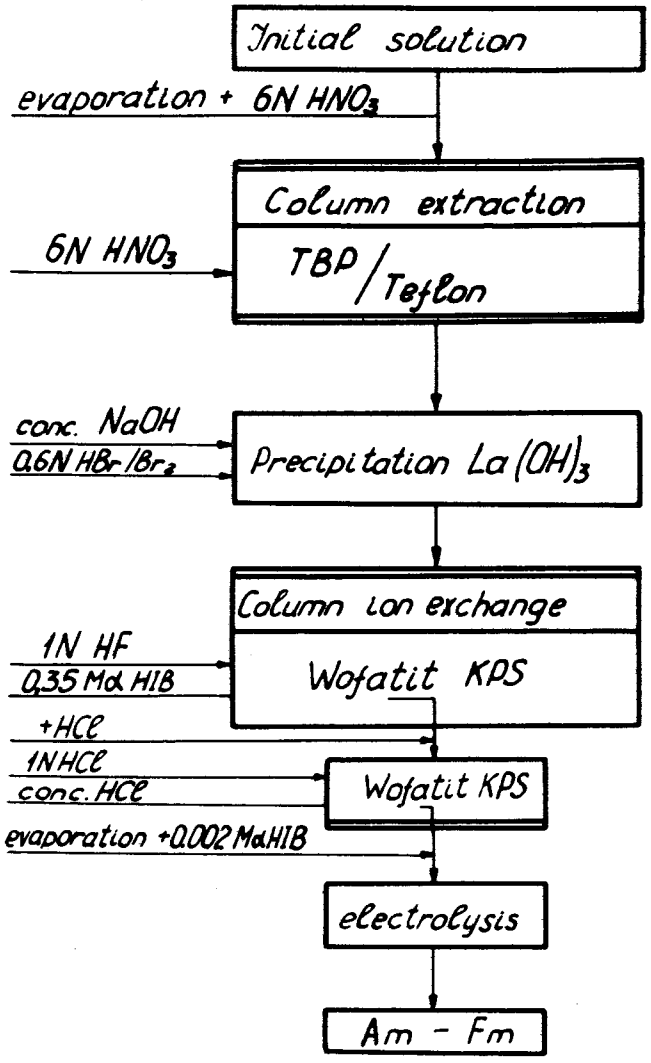
as tracers. The scheme of the further chemical treatment is presented in fig. 2. The final stage of this treatment consists of preparing by electrolysis fine specimens of the Am-Fm fraction suitable for α-spectrometry. The total chemical yield of the fraction mentioned, was not less than 70%. The thickness of the layers obtained permitted detection of α-particles with an energy resolution not worse than 50 keV.

In 10 hours after the irradiation the ion sources were placed into an α-ray spectrometer with surface-barrier detectors. The α-ray spectrum of the total fraction of transplutonium elements is presented in Fig. 3. All the spectrum lines can be identified unambiguously as due to the known isotopes of Am, Cm and Cf produced in transfer reactions.

At the same time for the 50-hour measurement no one ~ 7 MeV α-particle has been detected which might be assigned to the decay of ^{255}Fm ($E_\alpha = 7.01$ MeV). Thus the given experiment provides a possibility of obtaining the upper limit of the production cross section for ^{255}Fm, the daughter product of the decay of the initial nucleus $^{279}112$ in the reaction $^{233}U + ^{48}Ca$ (see fig. 1). The upper limit obtained is equal to 7×10^{-35} cm^2 in the ^{48}Ca ion energy range of 240-255 MeV (the calculated compound nucleus excitation energy lies between 26.5 and 39 MeV).

The subsequent experiments using ^{231}Pa were carried out under somewhat different conditions. The 0.3 mg/cm^2 target substance was deposited onto a thick aluminium backing. As the target was placed at 12° with respect to the beam direction, its effective thick-

Scheme of Chemical Separation of TPE



α HIB = α hydroxyisobutyrate PH = 4,7

Fig. 2. Scheme of the chemical treatment of the target and catcher foils.

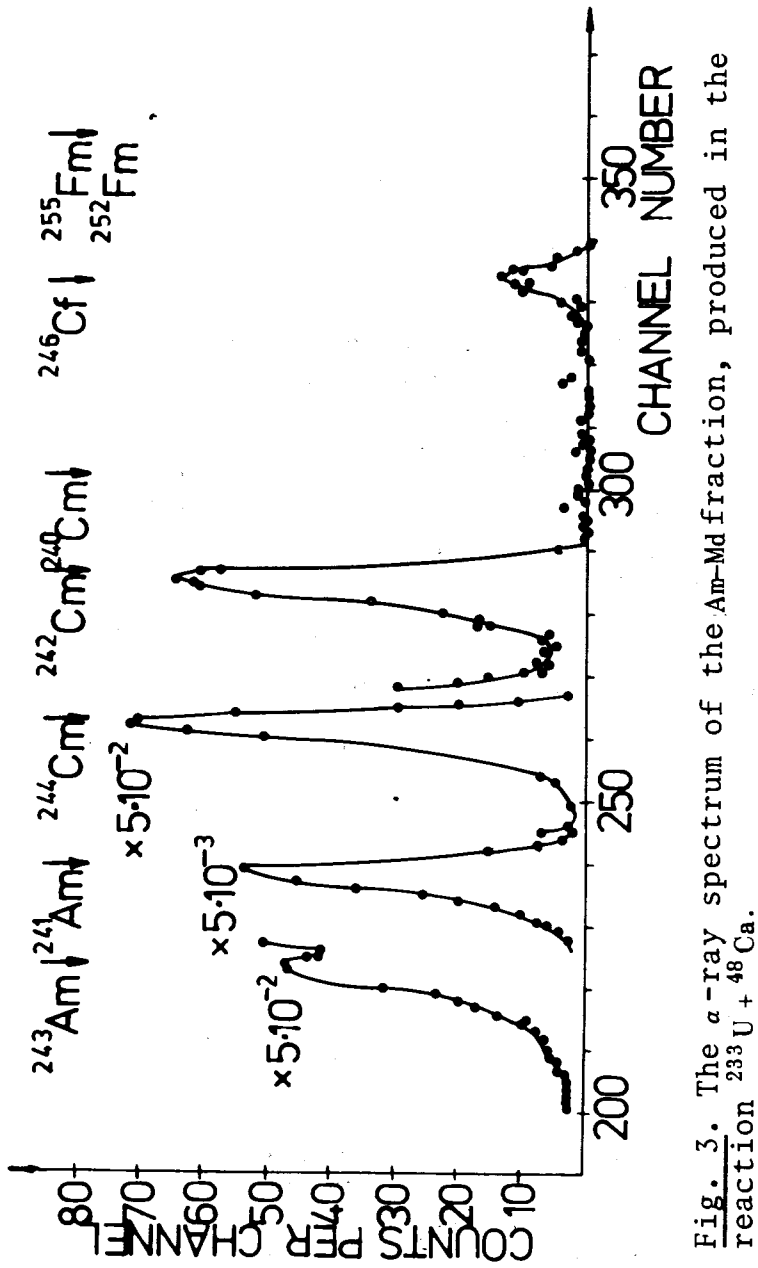


Fig. 3. The α-ray spectrum of the Am-Md fraction, produced in the reaction $^{233}U + ^{48}Ca$.

ness was 1.5 mg/cm^2. After irradiation the target was put in contact with phosphate glasses to detect fission fragments. The glasses were replaced each hour, which permitted identification of ^{256}Md according to its half-life. The detection efficiency was about 80%. The irradiation duration for the protactinium target was 5 hours, the integral ion flux at an initial energy of 255 MeV was 1.4×10^{16} and 1.8×10^{16} at 240 MeV.

During a 20-hour exposure no one spontaneous fission track has been detected. Thus the given experiment makes it possible to set the upper limit on the cross section for the production of ^{256}Md, which is produced as a daugther nucleus in the decay of the parent nucleus ^{276}111 in the reaction ^{231}Pa + ^{48}Ca. This upper limit is equal to 5×10^{-35} cm^2 in the ^{48}Ca ion energy range of 255-230 MeV (23 MeV $\leq E^* \leq$ 44 MeV).

During the following 26-day exposure period 17 spontaneous fission tracks evenly distributed in time have been detected. These fragments may be due to spontaneous fission of the known U-Cm isotopes produced by transfer reactions.

Since the experiments described allowed one only to set the upper limits of cross sections for the production of ^{255}Fm and ^{256}Md, an attempt has been made to detect directly the radioactive decay of their mother nuclei ^{279}112 and ^{276}111. or their neighbours. Therefore subsequently we carried out experiments to search for short-lived spontaneously fissioning emitters among the products of the reactions ^{232}Th+^{48}Ca and ^{231}Pa+^{48}Ca. The bombardments were performed at an internal beam of the U-300 cyclotron. The nuclei recoiling from the thin target reached the rotating disk to be transported to mica fission fragment detectors. This arrangment made it possible to record spontaneous fission with a half-life of 3 ms.

The results obtained in these experiments and the data of check irradiations by ^{40}Ar ions are listed in the table to follow.

Table

Reaction	$B_{Coul.}$ lab.syst. MeV	E_0 MeV	Integral ion flux $\times 10^{16}$	Number of fragment tracks	Limit of cross section for the (HI, xn) reaction cm^2
^{232}Th+^{48}Ca	222	255	1.3	3	2×10^{-35}
^{232}Th+^{40}Ar	199	232	1.3	4	
^{231}Pa+^{48}Ca	225	246	2.0	1	4×10^{-35}
^{231}Pa+^{40}Ar	201	240	1.2	1	

Several fission tracks observed in long exposures of thorium and protactinium targets have been analysed to determine the upper limits of the production cross sections for superheavy spontaneously fissioning nuclei. These cross sections equal to $(2\text{-}4) \times 10^{-35}$ cm^2 are shown in the last column of the table. The observed spontaneous fission events seem to be due to delayed fission of transfer reaction products such as ^{234}Am or ^{227,228}Np /19/.

DISCUSSION

The absence of the daughter products of the $^{276}111$ and $^{279}112$ decay or short-lived spontaneously fissioning nuclides among the products of the reaction of ^{48}Ca with ^{232}Th and ^{231}Pa can be interpreted in terms of two different assumptions. On the one hand, it cannot be excluded that the experimental sensitivity was insufficient to observe the effect expected. On the other hand, the results obtained may indicate that the properties of the isotopes with $Z = 110 - 112$ and $N = 164 - 168$ rule out their observation in these experiments.

On the basis of the analysis of the large amount of experimental data /11-15/ obtained in the studies of complete fusion reactions between heavy ions of ^{40}Ar to ^{58}Fe and Tl, Pb and Bi targets, as well as on the basis of the data on fission of Th and U by ^{40}Ar and ^{54}Cr ions /20/ and ^{40}Ca ions /21/ one can assume that the compound nucleus cross sections of elements 110-112 in the reactions investigated by us make up a noticeable part of the geometric cross sections. The excitation energy of these nuclei near the Coulomb barrier is estimated to be equal to 20-25 MeV, corresponding to the maximum probability of reactions involving the evaporation of two or three neutrons.

By assuming the neutron-to-fission width ratio of the compound nucleus, averaged over the evaporation cascade, to be 10^{-3}, one can estimate cross sections for the reactions $^{232}Th(^{48}Ca, 2, 3n)$, $^{231}Pa(^{48}Ca, 3n)$ and $^{233}U(^{48}Ca, 2n)$ to exceed the cross section upper limits reached in our experiments.

It is noteworthy that the expected values of Γ_n/Γ_f for excited nuclei with $Z = 110-112$ and $N = 167-170$ obtained by extrapolating the known values in the region of transfermium elements lie between 5×10^{-3} and 3×10^{-2}.

Therefore we are inclined to believe that the absence of any effects in the experiments described is conditioned by the properties of the isotopes with $Z = 110 - 112$ and $N = 164 - 168$ to be produced. Hence it follows that either the initial nuclei $^{276}111$ and $^{279}112$ or the daughter products of their α-decay mostly undergo spontaneous fission ($T_{sf} \ll T_\alpha$). This assumption seems to be most realistic for the initial nuclei $^{276}111$ and $^{279}112$ having $T_{sf} \ll 0.05$s. The direct experiments to search for short-lived spontaneous fission activities in the reactions $^{232}Th + ^{48}Ca$ and $^{231}Pa + ^{48}Ca$ have led to still lower limiting values, $T_{sf} \leq 0.003$s.

Fig. 4 shows the systematics of spontaneous-fission half-lives of doubly-even isotopes of transfermium elements, as well as the half-life limits for elements 110-112 obtained in the present paper.

It is necessary to note that these limits apply to even-odd and doubly-odd isotopes. As known, the presence of an odd nucleon in nuclei with $Z \geq 100$ leads to a spontaneous-fission hindrance factor of $10^2 - 10^5$. We think that this hindrance factor should be taken into account in estimating the T_{sf} limits for odd-mass nuclei. If one assumes that the hindrance factor for an odd numbers of protons or neutrons is equal to 10^{-3} (the average value for all known odd-mass isotopes of transfermium elements), whereas in the case of doubly-odd isotopes this factor

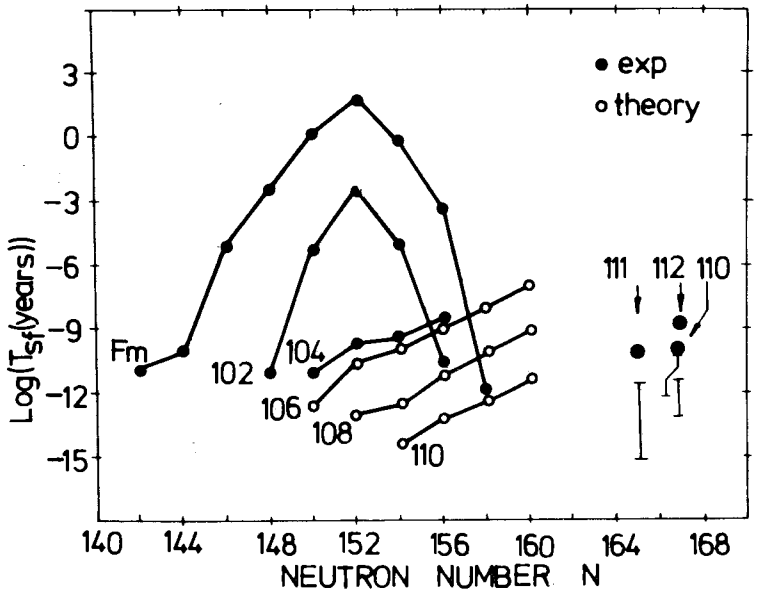


Fig. 4. Systematics of spontaneous-fission half-lives of doubly-even isotopes of transfermium elements. The closed circles connected by intercepts are experimental data taken from ref./13/ open circles are calculation/22/. The spontaneous-fission half-life limits for the isotopes $^{277}110$, $^{276}111$ and $^{279}112$ are shown by closed circles. The vertical lines drawn under these circles indicate the limits of spontaneous-fission half-lives of adjacent doubly-even isotopes.

is about 10^{-5} one can set more realistic limits and compare them with theoretical calculations made, as a rule, for doubly-even nuclei. Therefore fig. 4 presents two half-life limits for each nucleus, i.e.,

the upper limit obtained directly in experiment (without taking the hindrance factor into account) and the lower limit including the hindrance factor.

The results of the comparison of the obtained T_{sf} limits with theoretical calculations/22/ do not exclude that the lifetimes of neutron-rich isotopes with N = 165-168 may prove to be smaller than those of neutron-deficient nuclei with N=158-160. If so, the stability enhancement will be expected to take place only for a large neutron excess, N ≥ 180. This has been qualitatively predicted in a number of theoretical papers/25-27/. The spontaneous-fission half-life estimates made in the present paper may be helpful in making the fission barrier calculations more precise.

From an experimental point of view it would be appropriate to increase the performance rate of the experimental technique in order to determine the spontaneous-fission half-lives of these nuclei.

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