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Cross sections of the (HI, α n) channel in the cold-fusion-type reactions ²⁰⁹Bi + ⁴⁰Ar and ²⁰⁸Pb + ³⁷Cl

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Abstract

By applying an off-line radiochemistry technique, cross sections for the production of the isotope ²⁴⁰Cm ($T_{1/2} = 27$ d) in the cold-fusion-type reactions ²⁰⁹Bi + ⁴⁰Ar and ²⁰⁸Pb + ³⁷Cl at the bombarding energy $E_{lab} \leq 230$ MeV were determined to be 0.5 ± 0.2 nb and 0.6 ± 0.3 nb, respectively. The production of ²⁴⁰Cm was attributed to the 1n-deexcitation channel of the composite systems ²⁴⁹Md and ²⁴⁵Es. At the same time, the measured ²⁴⁰Cm production cross sections represent upper cross-section limits for the (HI, α n) channel of the reactions under study. These upper limits are about 100 times lower than the cross-section values reported recently by Nomura et al. for the (⁴⁰Ar, αxn) channels with x = 1,2 of the ²⁰⁹Bi + ⁴⁰Ar reaction at $E_{lab} = 208$ MeV. In the context of the above measurements, presented and discussed is the up-to-date summary of the available evidence on cross sections of the (HI, αxn) channels in the cold-fusion-type reactions induced by projectiles ranging from ³⁷Cl to ⁵⁰Ti on targets of ^{203,205}Tl, ²⁰⁸Pb, and ²⁰⁹Bi nuclei.

Appreciable EC(β^+)-delayed or/and spontaneous fission effects were detected in the ²⁰⁹Bi + ⁴⁰Ar, ²⁰⁸Pb + ³⁷Cl, and ²⁰⁶Pb + ³⁷Cl reactions. The obtained data point, in particular, to the EC(β^+)-delayed fission occurring in the decay chains ²⁴²Es $\xrightarrow{EC} \approx 7 \text{ s}^{242}$ Cf and ²³⁸Bk $\xrightarrow{EC} \approx 238$ Cm.

Keywords: NUCLEAR REACTIONS ²⁰⁹Bi(⁴⁰Ar, X), ²⁰⁸Pb(³⁷Cl, X); E = 230 MeV; measured production σ for ²⁴⁰Cm; deduced σ upper limits for (⁴⁰Ar, α n), (³⁷Cl, α n); other

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reactions; enriched targets; radiochemistry technique. RADIOACTIVITY ²⁴²Es, ²³⁸Bk (EC); measured $T_{1/2}$, EC-delayed fission yields; deduced delayed fission branching in ²³⁸Bk.

1. Introduction

It is a widely accepted fact that heavy-ion-induced complete fusion reactions followed by evaporation of several neutrons from the composite system, i.e., (HI, xn) processes, were the most prolific means for producing progressively heavier nuclei in the region of transfermium elements, Z > 100 (see, e.g., Refs. [1-5]). Most of the transfermium nuclei known to exist to date were produced by the (HI, xn) reactions using as targets both spherical nuclei near ²⁰⁸Pb and deformed actinide nuclei. In this way the chart of the nuclides was extended by now up to the borderline Z = 109. At the same time, a drastic decrease in production cross sections is observed with increasing Z, from several microbarns at Z = 102 down to some ten picobarns at Z = 109. In fusion reactions between actinide targets and beams such as ¹²C, ¹⁸O or ²²Ne, which were used in the sixties and seventies to discover elements 102 through 106, the decreases in production cross sections are thought to be due to the severe depletion from prompt fission of the composite systems formed at excitation energies of 40 to 50 MeV already at the Coulomb barrier [1-3]. When making the heaviest nuclides by fusing projec-tiles ranging from 48 Ca to 58 Fe with targets of 208 Pb or 209 Bi nuclei, the rapid decrease in cross sections with Z is believed to be associated with a progressively stronger hindrance to fusion, where the repulsive Coulomb forces more and more dominate the fusion dynamics [2,4,5]. Whatever the nature of the limitations, the cross sections for producing nuclides with Z = 110 are expected to be certainly below 10 pb for any kind of (HI, xn) reactions.

In view of the above limitations known for the (HI, xn) reactions for a long time, many attempts were made in the past on exploring alternative possibilities of synthesizing the heaviest nuclei. These attempts involved a variety of heavy-ion reactions collectively referred to as "transfer reactions" of which prominent examples are deep-inelastic collisions of the $^{238}U + ^{238}U$ and $^{238}U + ^{248}Cm$ type [6] or ¹⁸O- and ²²Ne-induced multinucleon transfer reactions using ²⁵⁴Es as a target [7]. The latter turned out to be quite prolific and have resulted in the recent discovery of a number of new neutron-rich nuclides such as ²⁶⁰Md, ²⁶²102, ²⁶¹103, and ²⁶²103 [7]. An interesting suggestion for the production of new elements was formulated recently in Ref. [8] by considering cold quasi-elastic multiproton transfer reactions induced by proton-rich projectiles near to closed nucleon shells (for example ¹⁴⁴Sm) on very heavy targets (like ²⁴⁸Cm).

Also, there were attempts to consider the productivity of heavy-ion synthesis reactions accompanied by the "direct" emission of light charged particles, in particular (HI, $\alpha x n$) reactions in which the light particle was supposed to carry off a major portion of the intrinsic energy available for the reaction intermediate. In this way, one could hope to produce relatively colder intermediate systems showing increased probabilities of survival against prompt fission. In particular, the potentialities of the αxn reactions ¹ were discussed at some length a decade ago in Ref. [1]. Some fragmentary measurements were also attempted at that time [9–13]. However, no indications in favour of the αxn reactions were found. Later on, a significant set of data was reported [14] regarding the cross sections of the pxn and αxn channels with $x \le 2$ in the reactions of ⁴⁸Ti, ⁴⁹Ti, and ⁵⁰Ti projectiles with ²⁰⁸Pb target nuclei. For these Ti-induced reactions, the upper cross-section limits have been determined to be at the level of one hundredth of the xn cross sections in the bombarding-energy range from the reaction thresholds to a maximum value of 5.5 MeV/u [14]. It seemed that these data hold out little hope of any advantages of the αxn reactions in producing the heaviest nuclei, at least for the reaction systems of the so-called cold-fusion type.

Recently, however, the $\alpha x n$ reaction as a "possible new approach to superheavy elements" was put forward again by Nomura et al. [15,16]. It was stated [15] that the αxn reactions, in which a precompound α -particle emission takes place, occur significantly even near the Coulomb barrier. This statement was supported by cross-section measurements made for the αn and $\alpha 2n$ channels of the ²⁰⁹Bi + ⁴⁰Ar reaction. These measurements were performed at an ⁴⁰Ar bombarding energy of 5.21 MeV/u by using the gas-filled recoil separator GARIS [17]. The observation of the α -particle-decay lines of ²⁴⁴Cf and ²⁴³Es formed via the (⁴⁰Ar, α n) and (⁴⁰Ar, α 2n) channels was reported and on this basis it was stressed that the αx n reactions occur with a significant probability even in the cold-fusion-type combination of target and projectile if these are allowed energetically. The cross sections for the ²⁰⁹Bi(⁴⁰Ar, α n) and ²⁰⁹Bi(⁴⁰Ar, α 2n) reactions were determined to be surprisingly large, 44 ± 15 nb and 36 ± 15 nb, respectively [16]. By involving these results into a series of oversimplified considerations, Nomura et al. have arrived at the conclusion [15,16] that the productivity of the $\alpha 2n$ reactions in the region of the heaviest elements can be orders of magnitude higher as compared to that known for xn reactions. For instance, a possible cross section for the 209 Bi(65 Cu, $\alpha 2n$) 268 110 reaction was estimated to be as high as 1 nb [15], in a dramatic contrast with picobarn values expected [2,4,5,14] for xn reactions leading to Z = 110 nuclides.

Thus, both the data and the conclusions of Refs. [15,16] turned out to be in strong contradictions with what was measured or accepted as being true before.

In the present paper we report on experiments designed to clarify these contradictions by studying cross sections of the α n channel in the cold-fusion-type reactions ²⁰⁹Bi + ⁴⁰Ar and ²⁰⁸Pb + ³⁷Cl.

¹ To designate heavy-ion reaction channels, here and below we use shortened notations, i.e., xn or αxn instead of (HI, xn) or (HI, αxn), etc.

2. Experimental procedure

2.1. Experimental method

To perform the measurements with the highest possible sensitivity and reliability, in the present experiments, in contrast to the work by Nomura et al. [15,16], we used the off-line radiochemistry technique instead of employing, e.g., the Dubna gas-filled recoil separator [18]. The recoil separators like those operated at RIKEN [17] or JINR [18] possess a certain (generally small) angular acceptance and are powerful means for studying products from complete fusion reactions followed by neutron evaporation, which are featured by the well-known, clearly defined and strongly forward-peaked kinematics. This is not the case for fusion-evaporation processes accompanied by a "direct" or "precompound" emission of particles. Kinematic features of such processes are not well known and can be predicted only with a limited degree of confidence. At the same time, the heavy residues produced in these processes are anticipated to show significantly broader, sidepeaked angular distributions and thus are expected to be suppressed by the kinematic separators by an unknown factor essentially dependent on both reaction system and bombarding energy. Therefore, to avoid uncertainties associated with kinematics, we applied the catcher technique with off-line chemical separations, which allowed all the products of interest to be collected irrespective of their unknown angular distributions.

For our off-line α -decay measurements, the nuclide indicative of the α n channel was chosen to be the long-lived isotope ²⁴⁰Cm with $T_{1/2} = 27$ d, $b_{\alpha} > 99.5\%$, $E_{\alpha} = 6290.5$ keV (71.1%) and 6247.7 keV (28.9%) [19]. As is seen from Fig. 1, the immediate products of the α n channel in the ²⁰⁹Bi + ⁴⁰Ar and ²⁰⁸Pb + ³⁷Cl reactions, ²⁴⁴Es and ²⁴⁰Bk, are represented by ²⁴⁰Cm with the 100% probability. Evidently, a number of other reaction channels can also contribute to the measured yield of ²⁴⁰Cm. These are 1n, 1p, α p, 2α n, and 2α p channels in the ²⁰⁹Bi + ⁴⁰Ar system and 1n, 1p, and α p channels in the ²⁰⁸Pb + ³⁷Cl case. Thus, the measured cumulative yield of ²⁴⁰Cm will represent an upper cross-section limit for the α n channel.

For the reactions under study, the off-line technique does not allow the αxn channels with x = 2 and 3 to be probed with an appropriately high sensitivity since there are no suitable α -emitters in the corresponding decay chains. Yet a special remark should be made regarding xn and αxn channels with x = 3. These channels lead to nuclides which can undergo spontaneous or/and electron-capture (EC)-delayed fission, see Fig. 1. The detection of the fission events can thus give useful additional information, and, to obtain it, we have extended our experimental technique to a possibility of fission-fragment counting.

2.2. Targets and irradiations

The irradiations were performed at the U400 cyclotron of the JINR Laboratory of Nuclear Reactions (Dubna) by using the wheel system described in Ref. [23]. A



Fig. 1. Radioactive decay chains of the products from the $^{209}Bi + {}^{40}Ar$ and $^{208}Pb + {}^{37}Cl$ reaction channels associated with x = 1 and x = 3. The decay properties indicated were taken from Refs. [20-22].

beam of ⁴⁰Ar or ³⁷Cl projectiles with an incident energy of 230 MeV and an average intensity of about $5 \cdot 10^{12}$ particles/s tangentially struck the lateral surface of a cooled copper cylinder onto which 2 to 3 mg/cm² of the metallic target material was deposited. This cylindrical target (serving simultaneously as a recoil

catcher) rotated with a constant velocity. Mica fission-fragment detectors arranged all around the rotating target cylinder (except for the beam-input zone) were used for the detection of EC-delayed or spontaneous fission events.

The metallic layers of Bi or isotopically enriched ²⁰⁸Pb (99.0%) were deposited onto the target cylinder by evaporation in a vacuum; a target of ²⁰⁸Pb enriched to 94.9% was also used in one of bombardments. After the irradiation, the entire layer of the target material was mechanically removed from the target cylinder and then was treated radiochemically to achieve element separation and prepare a thin source for α -decay measurements.

Earlier this technique was extensively used in experiments aimed at synthesizing transfermium elements (see, e.g., Refs. [14,23]) where it permitted the detection of nuclides produced with picobarn cross sections. More recently, it was employed in the experiments which led to the discovery of a new region of $EC(\beta^+)$ -delayed fission around ¹⁸⁰Hg-¹⁸⁸Pb [24,25], as well as in the studies of the stability of the K-isomeric states of ²⁵⁰Fm and ²⁵⁴102 against spontaneous fission [26].

2.3. Chemical separations

Three steps of chemical treatment were applied to isolate the fraction of Cf, Cm, and Am from the irradiated Bi or Pb target material, the total amount of which was about 200 mg per one target wheel.

The first step was to separate lanthanide and actinide elements from the target material. With this aim in view, the latter was dissolved in concentrated HNO₃, with adding 10 μ g of La as a carrier. Also added were tracers of ¹⁶⁰Tb and ¹⁴⁴Ce as well as markers of ¹⁴⁸Gd, ²⁴¹Am, and ²⁴⁴Cm used for the chemical yield determination. The prepared solution was evaporated to a small volume and then 50 ml of 0.002 M ethylene–diamine–tetraacetic acid (EDTA) buffered at pH = 1.2 were added. In the case of Bi, which is known to form, in acid media, stable complexes with EDTA [27], a solution of 0.01 M EDTA was used. The solution was heated up to 50°C and passed through a cation-exchange column filled with the resin Dowex-50, at the same temperature, to avoid crystallization of EDTA. Under these conditions, the lanthanides and actinides were absorbed on the column, whereas Bi and Pb were quantitatively eluted. After sorption, the column was washed with 100 ml of 0.002 M EDTA solution buffered at pH = 1.2 and then, successively, with water, with 0.2 M HF, with water again, and with 2 M HCl.

The second step was to purify the lanthanides and actinides absorbed on the Dowex-50 column. To this end, these elements were eluted with 6 M HCl, evaporated to dryness, and dissolved in 0.1 M HCl. The obtained solution was purified on a column with di(2-ethylhexyl) phosphoric acid (HDEHP) from which the absorbed lanthanides and actinides were eluted with 6 M HCl and subsequently passed through an anion-exchange Dowex-1 column. The eluate from this column was dried and the residue, after dissolving in 0.1 M HCl, was passed through a cation-exchange Aminex A-5 (~ 13 μ m) column from which the fraction of Cf, Cm, and Am was eluted with α -hydroxyisobutyrate solution at pH = 4. The above procedure allows decontamination factors higher than 10⁸ to be obtained in

purifying trivalent Am and heavier actinides from Ra, Ac, Th, U, Np, Pu, and other elements.

In the third step, thin sources were prepared for α -decay measurements. The fraction of Cf, Cm, and Am from the Aminex column was finally purified using HDEHP and Dowex-1 columns and then electrodeposited onto a polished disk of stainless steel. Typically, the measured chemical yield of Cm was equal to about 85%.

2.4. Alpha-particle spectrometry

The radiochemically prepared sources with the fraction of Cf, Cm, and Am were measured in the low-background α -activity spectrometer described in Ref. [28]. Along with accumulating α -particle-energy spectra, the spectrometer records also detection times of individual α -decay events. Thus, analyzing the energy-time correlations of the detected α -events makes it possible to reveal and rule out background α -events which could originate, e.g., from natural decay series involving α -emitters of the Po-Th region.

The measurements were performed in a nearly 2π counting geometry; typically, the activity spot of a 12 mm diameter was placed at a 0.7 mm distance in front of a circular 20 mm surface-barrier Si detector. The close counting geometry explains the appearance of visible low-energy tails in the profile of the recorded α -lines. For each particular case, the spectrometric detection efficiency was determined by considering the actual shape of the reference α -lines from ¹⁴⁸Gd and ²⁴¹Am. These reference α -lines were used also to determine the net efficiency of the chemical procedures as well as to check the stability of the spectrometer in the course of the measurements which continued (albeit with breaks) for approximately two months.

3. Measurements and results

3.1. Observation of ²⁴⁰Cm

To study the production of ²⁴⁰Cm, we have performed three ²⁰⁹Bi + ⁴⁰Ar bombardments and a bombardment of ²⁰⁸Pb + ³⁷Cl. Each of these bombardments was followed by the radiochemical treatment of the irradiated target material with subsequent α -decay measurements. A summary of experimental results is given in Table 1.

Typical α -particle-energy spectra resulting from the fraction of Cf, Cm, and Am are shown in Fig. 2 for both reactions under study. The two energy spectra are quite similar and show, with a sufficient statistics, a clear peak at ≈ 6.3 MeV, which should be assigned to ²⁴⁰Cm. Apart from the coincidence with the α -particle energy known for ²⁴⁰Cm, this assignment of the peak is strongly supported by a number of other arguments. The argument of prime importance is provided by the fact of the preceding chemical isolation of the Cf–Cm–Am fraction. Second, the measurements performed over a period of some 70 days allow us to follow the

Experimental results on the production of ²⁴⁰Cm in the ²⁰⁹Bi+⁴⁰Ar and ²⁰⁸Pb+³⁷Cl reactions at $E_{lab} \leq 230$ MeV

Reaction	I ^a	N _a ^b	Y(²⁴⁰ Cm) ^c	$\sigma(^{240}\text{Cm}) (\text{nb})^{d}$	
²⁰⁹ Bi + ⁴⁰ Ar	0.8 0.7 2.0	40 80 176	$\begin{array}{r} 2.3 \substack{+ \ 0.9 \\ - \ 0.9 \\ - \ 0.7 \\ 1.7 \substack{+ \ 0.8 \\ - \ 0.7 \\ - \ 0.4 \end{array}}$	$0.6 \pm 0.3 \\ 0.5^{+0.3}_{-0.2} \\ 0.5 \pm 0.2$	
²⁰⁸ Pb + ³⁷ Cl	2.1	130	2.5 ± 0.8	0.6±0.3	

^a Beam dose in units of 10¹⁸ incident particles.

^b Number of α -particles detected in the ≈ 6.3 MeV α -group.

^c Thick-target yield of ²⁴⁰Cm (i.e., yield per one beam particle) in units of 10^{-15} ; the total detection efficiency is taken into account in the determination of Y.

^d Estimated assuming $\Delta E_{FWHM} = 8 \pm 2$ MeV for the corresponding excitation function.



Fig. 2. Alpha-particle-energy spectra from the fraction of Cf, Cm, and Am isolated after ²⁰⁹Bi + ⁴⁰Ar and ²⁰⁸Pb + ³⁷Cl irradiations. The α -peak at ≈ 5.5 MeV is due to the marking activity of ²⁴¹Am, $E_{\alpha} = 5486$ keV (85.1%) and 5443 keV (13.3%) [19], whereas the 3183 keV α -line of the ¹⁴⁸Gd marker is not included in the figure. The α -peaks at ≈ 5.8 MeV and ≈ 6.1 MeV are due to the marking activity of ²⁴⁴Cm, $E_{\alpha} = 5805$ keV (76.4%) and 5762 keV (23.6%), involving an admixture of ²⁴²Cm, $E_{\alpha} = 6113$ keV (74.1%) and 6069 keV (25.9%) [19].



Fig. 3. Decay of the ≈ 6.3 MeV α -activity from the fraction of Cf, Cm, and Am isolated after ²⁰⁹Bi+⁴⁰Ar and ²⁰⁸Pb+³⁷Cl irradiations. Statistical errors are shown by vertical bars. The indicated half-life values were determined by the maximum-likelihood method [29].

decay of ≈ 6.3 MeV α -activity and thus to estimate its half-life. The corresponding decay data are presented in Fig. 3 and show a reasonably good agreement with the half-life value known for ²⁴⁰Cm. Third, no time correlations were revealed between the ≈ 6.3 MeV α -decays and subsequent α -events detected within a time interval of some tens of seconds; moreover, no α -events were detected in the α -energy region of ≈ 6.35 to ≈ 10 MeV. This allows us to eliminate a possible contribution from ²²⁰Rn ($E_{\alpha} = 6288$ keV, $T_{1/2} \approx 56$ s [19]) appearing in the decay series of natural thorium (note that the α -decay of ²²⁰Rn should be followed by the α -decay of the 0.15 s isotope ²¹⁶Po with $E_{\alpha} = 6778$ keV [19]). Finally, with reference to Table 1, we stress a good reproducibility of the results obtained from the repeated bombardments. All in all, there are good grounds to conclude that the ≈ 6.3 MeV α -group belongs to the decay of ²⁴⁰Cm.

The measured "thick" target yields of the ≈ 6.3 MeV α -activity were used to estimate the effective production cross sections of ²⁴⁰Cm in the reactions under study. To obtain these estimates, one needs to make an assumption regarding the

Reaction	I ^a	N _{df} ^b	T _{1/2} ^c	σ_{df} (nb) ^d	
209 Bi + 40 Ar	4.0	1301	$ \begin{array}{r} 0.8^{+1.5}_{-0.7} \text{ s} \\ 7^{+3}_{-2} \text{ s} \\ \sim 2 \min \end{array} $	$\begin{array}{c} 0.04 \substack{+3.0 \\ -0.02} \\ 0.14 \pm 0.04 \\ 0.03 \pm 0.02 \end{array}$	
²⁰⁸ Pb + ³⁷ Cl	2.7	1588	8^{+13}_{-3} s	0.19 ± 0.06 0.08 ± 0.03	
206 Pb + 37 Cl	0.9	561	0.5 mm	0.2	

Estimated half-lifes $T_{1/2}$ and production cross sections σ_{df} of the delayed fission activities observed in the reactions under study

^a Beam dose in units of 10¹⁸ incident particles.

^b Total number of detected fission events.

^c Deduced by using the maximum-likelihood method [29]; the indicated errors of $T_{1/2}$ reflect statistical uncertainties only.

^d Estimated assuming $\Delta E_{FWHM} = 10 \pm 2$ MeV for the corresponding excitation function.

width(s) of the corresponding excitation function(s), $\Delta E_{\rm FWHM}$. With a $\Delta E_{\rm FWHM}$ value of 8 ± 2 MeV, the effective ²⁴⁰Cm production cross sections were estimated to be 0.5 \pm 0.2 nb and 0.6 \pm 0.3 nb for the ²⁰⁹Bi + ⁴⁰Ar and ²⁰⁸Pb + ³⁷Cl reactions, respectively. Using larger $\Delta E_{\rm FWHM}$ values will evidently lower down the above cross-section values.

3.2. EC-delayed and spontaneous fission activities

An interesting finding of the present experiments is the observation of relatively strong EC-delayed and spontaneous fission activities in the reactions under study. The production of fissioning nuclides in these reactions has never been studied before; moreover, this is true for the production of any evaporation residues by these reactions, with the exception of experiments [30] where the ²⁰⁹Bi + ⁴⁰Ar reaction was used to produce the isotope ²⁴⁷Md. Therefore to characterize the fission effects in more detail, we carried out a number of additional bombardments without chemical separations. Specifically, we performed three ²⁰⁹Bi + ⁴⁰Ar bombardments at different periods of the target wheel revolution, $T_{rev} = 9$, 48, and 110 s, as well as three ²⁰⁸Pb + ³⁷Cl bombardments, with $T_{rev} = 48$, 110, and 770 s; there was also one bombardment ²⁰⁶Pb + ³⁷Cl performed at $T_{rev} = 105$ s. Table 2 presents summarized results from these bombardments made at $E_{lab} \leq 230$ MeV.

A general pattern of the fission effects observed is seen from Table 2 and Fig. 4. With good statistics, fission events were detected in all the three reactions in a time range extending from about 1 s to some 10 min. Evidently, several activities should contribute to form the observed pattern of fission events. Under given experimental conditions, the fission events appearing in the reactions we study should be associated predominantly with the x = 3 channel which is expected to have the largest cross section, as can be inferred from careful studies of the very similar reaction systems 206,207,208 Pb + 40 Ar [30–34]. Besides, several particular

Table 2



Fig. 4. Time distributions of delayed fission events detected in some particular bombardments $^{209}Bi + {}^{40}Ar$, $^{208}Pb + {}^{37}Cl$, and $^{206}Pb + {}^{37}Cl$. See also Table 2.

products of the x = 3 channel are known or can be expected to have fission branches at the percent level (see Fig. 1), which is not the case for the channels of x = 1, 2, and 4. However, in view of the extremely high sensitivity of the present experiments, some contributions from the x = 5 channel products of the $^{209}\text{Bi} + ^{40}\text{Ar}$ and $^{208}\text{Pb} + ^{37}\text{Cl}$ reactions should not be ruled out completely. This is hinted at

also by the observations from the 206 Pb + 37 Cl bombardment. All in all, there is quite a number of potential sources of the fission events detected. This makes the problem of disentangling the fission activities rather complicated.

When considering the fission data more specifically, we should point out at first a fission activity with a half-life of about 7 s appearing in the ²⁰⁹Bi + ⁴⁰Ar and ²⁰⁸Pb + ³⁷Cl reactions (see Fig. 4). The most probable explanation of this activity seems to be the EC-delayed fission in the decay chain ²⁴²Es $\frac{EC}{\approx 7_s}$ ²⁴²Cf. This explanation is in line with an observation [21] made at Darmstadt with SHIP in the ²⁰⁵Tl + ⁴⁰Ar reaction, where on the basis of three detected fission events the EC-delayed fission in the ²⁴²Es \xrightarrow{EC} ²⁴²Cf chain was reported and its probability $P_{d.f.}$ evaluated to be $(1.4 \pm 0.8) \times 10^{-2}$. The production cross section of the new isotope ²⁴²Es with $T_{1/2} = 5$ to 25 s was reported to be 40 nb, whereas that for the EC-delayed fission events was estimated to be 180 pb [21].

Again, a fission activity with $T_{1/2} = 2.0^{+1.5}_{-0.5}$ min can be singled out from our data. This is clearly seen in the results from the ²⁰⁸Pb + ³⁷Cl bombardment made at $T_{rev} = 770$ s (see Fig. 4). This is a new fission activity since it, despite the similar half-life, cannot be explained by EC-delayed fission in the decay chain ²⁴⁰Bk $\xrightarrow{EC}_{-4 \text{ min}}$ ²⁴⁰Cm, which is known to occur with a very small probability, $P_{d.f.} \approx 10^{-5}$ [35] or $(1.3^{+0.8}_{-0.7}) \times 10^{-5}$ [36]. Note that according to our measurements described in Section 3.1, the ²⁴⁰Bk production cross section in the ²⁰⁸Pb + ³⁷Cl reaction does not exceed 0.6 nb. In addition, the new fission activity cannot be due to spontaneous fission of the 3.68 min isotope ²⁴²Cf since our direct experiments [37] have shown the spontaneous-fission branch for ²⁴²Cf to be very small, $b_{s.f.} \leq 1.4 \times 10^{-4}$. With the above facts taken into account, the most probable origin of the 2 min fission activity seems to be the EC-delayed fission in the decay chain ²³⁸Bk \xrightarrow{EC}_{-238} Cm, for which a higher probability $P_{d.f.}$ can be expected as compared to the ²⁴⁰Bk \xrightarrow{EC}_{-240} Cm case. With this assignment, using the data of Ref. [21] on the production and decay properties of ²⁴²Es (the parent of ²³⁸Bk), we obtain an order-of-magnitude estimate $P_{d.f.} \approx 3 \times 10^{-3}$ for the ²³⁸Bk, we obtain an order-of-magnitude estimate $P_{d.f.} \approx 3 \times 10^{-3}$ for the ²³⁸Bk, we obtain an order-of-magnitude estimate $P_{d.f.} \approx 10^{-3}$ for the ²³⁸Bk, we obtain an order-of-magnitude estimate $P_{d.f.} \approx 3 \times 10^{-3}$ for the ²³⁸Bk, we obtain an order-of-magnitude estimate $P_{d.f.} \approx 10^{-3}$ lower a fission activity with $T_{1/2} = 2.4 \pm 0.1$ min and $P_{d.f.} = (4.8 \pm 2.0) \times 10^{-4}$ was produced in the reaction ²⁴¹Am + ⁴He (75 MeV).

In the ²⁰⁹Bi + ⁴⁰Ar reaction, there seem to be observed also fission events with $T_{1/2}$ of about 1 s. Such an activity can appear due to the 8% spontaneous-fission branch of ²⁴⁶Fm [20], see Fig. 1. A contribution to the ~1 s fission events can come also from EC-delayed fission in the ²⁴⁶Md $\xrightarrow{\text{EC}}_{-1s}$ ²⁴²FM chain. According to systematics [38], the unknown isotope ²⁴⁶Md is expected to be an α -emitter with a $T_{1/2}$ of just about 1 s and some 15% branch for EC(β^+) decay. Unfortunately, obtaining more definite information about the ~1 s fission events is hindered by

Table 3

Reaction	E_{lab}^{a} (MeV)	E ^{Bass c} (MeV)	E* ^d (MeV)	$\sigma_{\alpha x n}$ (nb)	Ref.	_
208 Pb(37 Cl, α n) 240 Bk	230	179.4	75.4	< 0.6	present study	-
209 Bi(40 Ar, α n) 244 Es	230	193.1	62.5	< 0.5	present study	
209 Bi(40 Ar, α n) 244 Es	208 ^b	193.1	44.3	44 ± 15	[16]	
209 Bi(40 Ar, $\alpha 2n$) 243 Es	208 ^ь	193.1	44.3	36 ± 15	[16]	
204 Pb(48 Ca, $\alpha 2n$) 246 Fm	235	216.5	38.0	< 2	[12,13]	
205 Tl(45 Sc, $\alpha 2n$) 244 Fm	240	224.2	50.9	≤ 0.1	[11,12]	
203 Tl(45 Sc, α) 244 Fm	240	225.1	49.6	≤ 0.2	[11,12]	
208 Pb(50 Ti, $\alpha 2n$) 252 102	273	238.2	50.2	< 0.06	[14]	
208 Pb(49 Ti, α n) 252 102	271	238.1	53.1	≤ 0.004	[14]	
208 Pb(48 Ti, α) 252 102	259	237.9	46.1	≤ 0.003	[14]	

Summary of experimental data on $\alpha x n$ cross sections for cold-fusion-type reactions on Tl, Pb, and Bi target nuclei

^a Laboratory energy of beam particles entering an "infinitely thick" target characteristic of experimental techniques used in Refs. [11–14] as well as in the present study.

^b Bombarding energy used in Ref. [16].

^c Bass fusion barrier calculated according to Ref. [39].

^d Compound-nucleus energy corresponding to the indicated E_{lab} value.

the presence of the intense ≈ 7 s fission activity attributed to EC-delayed fission in the 242 Es $\frac{EC}{\approx 7}$ 242 Cf chain.

A considerable fission yield was revealed also in the ²⁰⁶Pb + ³⁷Cl reaction. It is probable that two or even more fission activities contribute to the decay curve observed in this case (see Fig. 4). More bombardments are required to characterize these activities and make definite assignments. In principle, the ²⁰⁶Pb + ³⁷Cl reaction appears to be a probe for EC-delayed and spontaneous fission effects in the decay chains ²⁴⁰Es \xrightarrow{EC} $\xrightarrow{236}$ Cm.

4. Discussion

As noted in Section 2.1, our measured cross sections for the production of ²⁴⁰Cm represent upper cross-section limits for the α n channel of the ²⁰⁹Bi + ⁴⁰Ar and ²⁰⁸Pb + ³⁷Cl reactions. In Table 3 these upper limits are compared with the α n and α 2n cross-section values reported by Nomura et al. [15,16]. Also included in Table 3 are αxn cross-section data reported for a number of similar reaction systems of the cold-fusion type [11–14]. It is seen that our upper α n cross-section limit for the ²⁰⁹Bi + ⁴⁰Ar reaction is two orders of magnitude lower than the value obtained in Refs. [15,16].

Moreover, the main source of ²⁴⁰Cm detected in our measurements seems to be the 1n-evaporation channel of the reactions under study, for which the so-called minimum excitation energy E_{\min}^* of the composite systems (i.e., the excitation energy at the Bass fusion barrier B_{lab}^{Bass} [39]) is calculated to be about 32 MeV, like that for the well-studied nearby systems ^{207,208}Pb + ⁴⁰Ar. For cold-fusion-type reaction systems close to those examined in the present work, the only previous cross-section measurement for the 1n-deexcitation channel was attempted [31] in the ²⁰⁷Pb + ⁴⁰Ar reaction ($E_{\min}^* = 31.3$ MeV), with the result $\sigma_{1n}^{\max} \leq 0.1$ nb ², which is not far from our values, 0.5 ± 0.2 nb and 0.6 ± 0.3 nb for the ²⁰⁹Bi + ⁴⁰Ar and ²⁰⁸Pb + ³⁷Cl reactions with $E_{\min}^* = 31.5$ MeV and 32.4 MeV, respectively. Note that, for the cold-fusion-type reactions, even small variations in E_{\min}^* values are known to lead to appreciable changes in the cross sections of particular xn-

Table 4

Summary of maximum cross sections σ_{xn}^{\max} measured for xn-evaporation channels of the ⁴⁸Ca-induced cold-fusion reactions on Tl, Pb, and Bi target nuclei

Reaction	E _{lab} ^a (MeV)	E ^{Bass c} (MeV)	E ^{* d} (MeV)	Detected nucleus	σ_{xn}^{\max} (nb)	Ref.
²⁰⁵ Tl(⁴⁸ Ca, n) ²⁵² Md	223	213.2	31.4	²⁵² Fm	35 ± 13 ^{e,f}	[42]
²⁰⁵ Tl(⁴⁸ Ca, 3n) ²⁵⁰ Md ²⁰³ Tl(⁴⁸ Ca, n) ²⁵⁰ Md	223	213.2 214.1	31.4 31.3	²⁴⁶ Cf	145 ± 40 ^g	[42]
204 Pb(48 Ca, γ) 252 102	235	216.5	38.0	²⁵² 102	< 0.5	[13]
²⁰⁶ Pb(⁴⁸ Ca, 2n) ²⁵² 102	235 215 ^b 248	215.6	37.9 21.7 48.3	²⁵² 102 ²⁵² 102 ²⁵² 102	$500 \\ 190 \pm 32 \\ 500 \pm 200$	[13] [44,45] [26]
²⁰⁷ Pb(⁴⁸ Ca, 3n) ²⁵² 102	235	215.2	37.2	²⁵² 102	~ 100	[13]
²⁰⁸ Pb(⁴⁸ Ca, n) ²⁵⁵ 102	224 231 210 ^ь	214.7	28.2 34.2 16.7	²⁵⁵ Fm ^h ²⁵⁵ Fm ^h ²⁵⁵ Fm ^h	$420 \pm 130 e,i$ 130 ± 60 260 ± 30	[42] [26] [47]
²⁰⁸ Pb(⁴⁸ Ca, 2n) ²⁵⁴ 102	224 224 214 ^b 231 213 ^b 227 ^b 215 ^b	214.7	28.2 28.2 19.7 34.2 19.3 30.7 20.9	²⁵⁴ Fm ²⁴⁶ Cf ²⁴⁶ Cf ²⁴⁶ Cf ²⁵⁴ 102 ²⁵⁴ 102 ²⁵⁴ 102	$\begin{array}{c} 4700 \pm 1000 \ ^{e,j} \\ 4900 \pm 900 \ ^{e,j} \\ 3300 \pm 370 \ ^{k} \\ 1700 \pm 700 \\ 2340 \pm 540 \\ 3400 \pm 400 \\ 390 \pm 70 \end{array}$	[42] [42] [47] [26] [47] [33] [44,45]
²⁰⁸ Pb(⁴⁸ Ca, 3n) ²⁵³ 102	231 225 ^b	214.7	34.2 28.8	²⁵³ Es ²⁵³ Es	100^{+260}_{-50} 109 ± 33	[26] [47]
²⁰⁸ Pb(⁴⁸ Ca, 4n) ²⁵² 102	235	214.7	37.2	²⁵² 102	~ 20	[13]
²⁰⁹ Bi(⁴⁸ Ca, n) ²⁵⁶ 103 ²⁰⁹ Bi(⁴⁸ Ca, 2n) ²⁵⁵ 103	212 ^b 236 215 ^b 236	217.5	17.6 36.8 19.6 36.8	${}^{256}_{252} \text{Fm} + {}^{255}_{255} \text{Fm}$ ${}^{255}_{103} \text{Fm} + {}^{255}_{255} \text{Fm}$	61 ± 20 m 76 ± 30 ^{e,i,i} 437 ± 96 m 600 ± 360 ^{e,i,1}	[47] [42] [47] [42]
²⁰⁹ Bi(⁴⁸ Ca, 3n) ²⁵⁴ 103	227 ^ь 236		29.4 36.8	²⁵⁴ 103 ²⁴⁶ Cf	28 ± 11 $40 \pm 12^{\text{e,m}}$	[47] [42]

² Please note an evident misprint in the last line of Table 1 in Ref. [31]. This misprint was reproduced also in Tables 2 and 3 of Ref. [34], yet it was corrected in Ref. [40]. The correct result is $\sigma_{\text{in}}^{\text{max}} \leq 0.1$ nb.

evaporation channels. Thus, taking the 1n-evaporation channel into account, our upper α n cross-section limits for the reactions under study should be lowered down further compared to those given in Table 3.

As to the surprisingly large cross-section values reported in Refs. [15,16] for the α n and α 2n channels of the ²⁰⁹Bi + ⁴⁰Ar reaction, a reason for this seems to lie in the misinterpretation of the α -particle-energy spectra recorded with poor statistics by using the gas-filled recoil separator GARIS. In these measurements, the α -decay of the primary α n product, the 37 s isotope ²⁴⁴Es with $b_{\rm EC} = 0.96^{+0.02}_{-0.03}$ [41], could not be observed because of the very small α -decay branch. The identification of the α n channel was made on the basis of approximately six α -counts with energies of 7.16 and 7.21 MeV, taken, with a delay of 0.8 s, during the beam-off periods; note that pulsed ⁴⁰Ar beams were used in the experiments [15,16], with time structures of 40 s (or 10 s) beam-on and 40 s (or 10 s) beam-off periods. The above counts were tentatively assigned to the α -decay of the 19.7 min daughter nucleus ²⁴⁴Cf. This assignment was based on the measured α -particle energies only, without any time information. Besides, in the measured α -spectrum there is present the very strong 7.27 MeV α -line due to the 25 s transfer product ^{211m} Po, which produces a background comparable with the number of the α -counts attributed to ²⁴⁴Cf (see Fig. 4 in Ref. [15] and Fig. 6 in Ref. [16]). In a similar way, the identification of the α 2n channel was made on the basis of approximately six 7.89 MeV α -counts collected during the beam-off periods. The half-life was not measured, but it was noted that three 7.89 MeV counts fell in the region of t > 5 s

Notes to table 4:

^a Laboratory energy of beam particles entering an "infinitely thick" target characteristic of experimental techniques used in Refs. [13,26,42].

Bombarding energy corresponding to the maximum of the excitation function.

^c Bass fusion barrier calculated according to Ref. [39].

^d Compound-nucleus excitation energy corresponding to the indicated E_{lab} value.

^e Our estimate obtained on the basis of the experimental data of Ref. [42] by assuming $\Delta E_{\text{FWHM}} = 8 \pm 2$, 9 ± 1 , and 10 ± 2 MeV for 1n-, 2n-, and 3n-evaporation channels, respectively, according to excitation function measurements performed in Refs. [32,33,44-46].

^f Obtained by taking into account the 70.5% abundance of ²⁰⁵Tl in the natural thallium.

^g Our estimate obtained on the basis of the experimental data of Ref. [42] by using the value 0.994 [20] for the total probability of the decay chain leading from ²⁵⁰Md to ²⁴⁶Cf and by assuming $\Delta E_{\text{FWHM}} = 9 \pm 2$ MeV for the effective excitation function. Due to the presence of two isotopes in the natural thallium used as target in Ref. [42], the given cross-section estimate represente of two isotopps in the initial matrix in used as target in Ref. [42], the given cross-section estimate represents in fact the quantity $(0.705\sigma_{3n}^{max} + 0.295\sigma_{1n}^{max})$. If we assume that σ_{1n}^{max} for the ²⁰³Tl(⁴⁸Ca, n)²⁵⁰Md reaction does not exceed 35 ± 13 nb, we obtain an estimate of $\sigma_{3n}^{max} = 170^{+60}_{-40}$ nb for the reaction ²⁰⁵Tl(⁴⁸Ca, 3n)²⁵⁰Md. ^h As argued in Ref. [26], yields of the (⁴⁸Ca, γ) and (⁴⁸Ca, α) reactions leading to ²⁵²Fm are expected

to be much lower compared to that of the reaction (⁴⁸Ca, n); see also Ref. [47]. ⁱ Obtained by using the empirical values [20] for $b_{\rm EC}$ of ²⁵⁵Md and ²⁵⁵102, 0.93 and 0.384, respectively. ^j Obtained by using the branching ratios $b_{\rm EC}(^{254}102) = 0.1$ and $b_{\alpha}(^{254}102) = 0.9$ [42,48].

^k Average of two measurements performed in Ref. [47].

¹ Obtained by using $b_{\rm EC}(^{255}103) = 0.23$ from systematics [38] as well as measured cross sections $\sigma_{1n}^{\rm max}$ and $\sigma_{2n}^{\rm max}$ from Ref. [47] in estimating $\sigma_{2n}^{\rm max}$ and $\sigma_{1n}^{\rm max}$, respectively. ^m Obtained by using the value 0.97 [20,45,48,49] for the total probability of the decay chain leading from $^{254}103$ to 246 Cf.

after the end of the beam-on period, which probably rules out a contribution from the 0.12 s transfer product ^{212m}At with $E_{\alpha} \simeq 7.84$ and $\simeq 7.90$ MeV. These α -counts were attributed to the 21 s isotope ²⁴³Es which is known to have $E_{\alpha} = 7.89 \pm 0.02$ MeV and $b_{\alpha} > 30\%$ [41]. In turn, the production of ²⁴³Es was considered to be a signature of the $\alpha 2n$ channel. However, this identification of the $\alpha 2n$ channel does not seem to be unambiguous again. First, at the ⁴⁰Ar bombarding energy of 208 MeV, the isotope ²⁴³Es could be produced, with a non-negligible cross section, via the 2n-evaporation channel. Second, apart from possibilities associated with a variety of transfer products, one very probable possibility would be to attribute the 7.89 MeV α -counts to the lighter isotopes of einsteinium, ²⁴²Es and ²⁴¹Es, which were shown to be 5 to 25 s α -emitters with $E_{\alpha} \approx 7.85$ and ≈ 7.93 MeV [21] and could be formed via the 3n- and 4n-evaporation channels of the ²⁰⁹Bi + ⁴⁰Ar reaction. At the ⁴⁰Ar bombarding energy of 208 MeV, the 3n channel of the 209 Bi + 40 Ar reaction is quite probable, as can be inferred, e.g., from the excitation functions measured for the 3n channel of the 207,208 Pb + 40 Ar reactions [32,33]. The more so, the collection efficiency of the recoil separator GARIS for the xnevaporation residues was measured to be 2.5 times larger than that for the αxn products, 30% instead of 12% [15,16]. Summing up, we conclude that there are no good grounds to interpret the above experimental data in favour of the αn and $\alpha 2n$ reaction channels.

In the context of the present discussion, it is important to note that Orlova et al. [42] reported on the experimental studies of the production of evaporation residues in the reactions of ⁴⁸Ca projectiles with targets made of ^{nat}Tl, ²⁰⁸Pb, and ²⁰⁹Bi. By applying an off-line radiochemical technique, thick-target yields of the long-lived, α -decaying nuclides ²⁴⁶Cf, ²⁵²Fm + ²⁵⁵Fm, and ²⁵⁴Fm were measured and used to characterize cross sections for different evaporation channels of the above reactions; these experimental data were included also in the conference report [43]. Along with the new interesting experimental information about the xn channels in the ⁴⁸Ca-induced cold-fusion reactions, the paper [42] offered also a number of speculations as well as standard statistical-model calculations made with an ALICE code. Albeit implicitly, emphasis was put on the importance of αxn - and pxn-deexcitation channels in the fusion-evaporation reactions induced by ⁴⁸Ca. However, a careful examination of the data reported in paper [42], as well as any other data relevant to the problem, shows that there is no evidence at all which could support the guesses and calculations of Ref. [42] associated with αxn and pxn channels. To demonstrate this, in Table 4 we present an up-to-date summary of the available data on xn-evaporation channels of the cold-fusion reactions induced by 48 Ca projectiles on target nuclei of 203,205 Tl, 208 Pb, and 209 Bi [13,26,33,42,44-47]. In particular, the data of Ref. [42] are included in Table 4 without any exception. It can be seen from Table 4 that there is a reasonably good agreement between the results obtained in different works. Even more important is the fact of a good agreement between the off-line radiochemical measurements performed with long-lived daughter and granddaughter products and the on-line measurements carried out with the primary reaction products. Finally, referring to Table 4, we stress that all the experimental data of Ref. [42] can be quite naturally explained without appealing to αxn and pxn channels. This is why the reaction systems studied in Ref. [42], with the particular long-lived nuclides chosen there for the off-line measurements, cannot be used for a sensitive probing of the αxn and pxn channels. A comparison of the cross-section data shown in Table 4 with the upper αxn cross-section limits collected in Table 3 clarifies this point unambiguously.

5. Conclusions

Our experiments have shown positively that the cross sections of the $\alpha x n$ channel in the 209 Bi + 40 Ar and 208 Pb + 37 Cl reactions are small, well below 0.5 nb. Our results are in accord with the bulk of the presently existing experimental data on the $\alpha x n$ cross sections for cold-fusion-type reaction systems [11-14,50]. As Table 3 shows, these existing data form in fact a collection of upper cross-section limits – the αxn cross sections are so small that no αxn product was unambiguously detected until now in fusion-evaporation reactions induced by projectiles ranging from ³⁷Cl to ⁵⁰Ti on targets of Tl, Pb, and Bi nuclei. Even if the energy available in these reaction systems allows the emission of an α -particle, the probability of the α -particle emission turns out to be very small, by at least one or two orders of magnitude smaller than that of neutron emission; this statement is true for the proton emission as well. Clearly, in such a case, there are no grounds to rely on the $\alpha x n$ reactions as processes offering a more productive way to superheavy elements, as it was suggested in Refs. [15,16]. Let us mention that in the very massive systems, where fusion of the partners is severely hindered by the dynamical ("extra-push") reasons associated with the excessively strong Coulomb forces, the loss of an α -particle during the amalgamation stage may increase the fusion probability of the remainder, as hinted to by observations [51] made in the system 110 Pd + 110 Pd \rightarrow 220 U. However, this cannot help much since the probability of an early α -particle emission is expected to be very small (it was estimated to be of the order of 10^{-4} for the neutron deficient ¹¹⁰Pd + ¹¹⁰Pd system [51]), and, besides, the α -emission reduces the Z value of the evaporation residues produced.

The extremely low cross sections of the αxn channels, with the concurrence of much more probable xn channels, cause serious difficulties in identifying the former in experiments. As is shown in Section 4, the data of Refs. [15,16] do not represent the material necessary for an unambiguous identification of the αn and $\alpha 2n$ channels and can readily be explained without appealing to the α -particle emission. The same is true for the experimental data of Ref. [42] on the ⁴⁸Ca-induced fusion-evaporation reactions. Yet, there is a need to understand better the particular reasons for the very low probabilities of the αxn channels in the cold-fusion-type reactions. Therefore, more extensive experimental and theoretical studies of the αxn channels are certainly called for.

All what has been said above on the low probabilities of αxn channels concerns the cold-fusion-type reaction systems formed of ³⁷Cl, ⁴⁰Ar, and heavier projectiles and near-magic Tl, Pb, and Bi target nuclei. However, an opposite situation had long been known to take place for the reactions induced by lighter projectiles like ¹²C or ¹⁶O on actinide and other target nuclei. In these reactions, formation cross sections of $\alpha x n$ products are often much larger compared to those of xn-evaporation residues (see, e.g., Refs. [52–54] and references therein). These observations seem to be reasonably explained by mechanisms involving transfers of α -particlelike clusters (C, Be, He) from projectile to target [53,54]. Note that this α -particle structure of projectiles tends to disappear when going from ¹²C or ¹⁶O to heavier bombarding species. Another well-known and quite understandable case of the enhanced α -particle and proton emission is that characteristic of very proton-rich composite systems (see, e.g., Refs. [55–57]). Here, αxn and pxn products generally dominate reaction yields, regardless of the projectiles used to induce these reactions. There can probably be also some other reaction types characterized by an enhanced charged-particle emission. Considering the abovementioned reaction groups is certainly beyond the scope of the present paper. The only remark we make here is that different groups of the αxn phenomena should be treated separately because the underlying physics is expected to be rather different. Just the contrary was the case in Refs. [15,16] where conclusions concerning potentialities of the αxn reactions were drawn by considering, on an equal footing, several different groups of the αxn processes mentioned above (see Fig. 2 in Ref. [15] or Fig. 4 in Ref. [16]).

Finally, we would like to stress that the cold-fusion-type reactions studied in the present work could definitely be used further to produce and study new neutrondeficient Md, Es, and Bk nuclides via the xn-evaporation channels of x = 2, 3, 4. The observation of the new EC-delayed fission activities described in Section 3.2 of the present paper is but one example of these potentialities.

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