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FISSION-FRAGMENT MASS DISTRIBUTION  
AND ESTIMATION  
OF THE CLUSTER EMISSION PROBABILITY  
IN THE  $\gamma + {}^{232}\text{Th}$  AND  ${}^{181}\text{Ta}$  REACTIONS

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1997

## 1 Mass-distribution in the $^{232}\text{Th}(\gamma, f)$ reaction

Fission fragments detection by their activity using radiochemistry and  $\gamma$ -spectroscopy methods is proved to be the best in accuracy for mass-distribution measurements. In the  $^{232}\text{Th}(\gamma, f)$  photofission reaction the individual mass yields were measured in ref.<sup>1,2</sup> The  $E_1 \times E_2$  method was applied too<sup>1,3</sup> and the growth of the mass-symmetric yield with the excitation energy increase was observed. However, mass-distribution behaviour near the symmetry was studied with the accuracy limited due to low enough yield values.

In the present experiment mass-distributions of the thorium photofission were measured at two energies by the activation technique using the Ge HP  $\gamma$ -spectrometer with the energy resolution of 1.8 keV and a 20% relative efficiency (with respect to the standard NaI) by the  $^{60}\text{Co}$   $\gamma$ -lines. A gram-weight target made of purified thorium oxide was irradiated by the bremsstrahlung at the FLNR JINR microtron beam.  $\gamma$ -spectra of induced activity were measured during two weeks after the irradiation, thus, short and long enough lived nuclides were detected. Their relative yields were determined by the  $\gamma$ -lines intensities taking into account the  $\gamma$ -line abundance, efficiency values and decay factors. Many of the detected radioisotopes accumulate the total yield of the individual isobaric  $\beta$ -decay chain. Then their cumulative yields (35 values) give one the possibility to plot the final fragment mass-distribution. Corrections on the incomplete cumulativity as well as on the yield redistribution due to the delayed neutron emission were introduced

basing on the detailed information available in the fragment yield Handbook <sup>4</sup> for the thermal neutron induced fission of <sup>235</sup>U. One can assume close enough charge-distribution parameters for the latter reaction and the studied one. For the absolute calibration of the mass-distribution the maximum yield in the asymmetric mode peaks was taken to be 8% in accordance with ref.<sup>2</sup>

The final fragment mass-distributions are demonstrated in Fig. 1 for the two values of the bremsstrahlung end-point energy  $E_e = 12$  and 24 MeV. The behaviour of the points near asymmetric maxima shows fine structure irregularities in accordance with ref.<sup>2</sup> The random errors do not exceed the size of the points, however, some systematical errors on the level of 10-15% can take place due to inaccuracies in the tabular values of  $\gamma$ -line abundance and in the detector efficiency as well as due to the possible contribution of weak non-resolved  $\gamma$ -lines. It seems to impede the conclusive discussion of the fine structure.

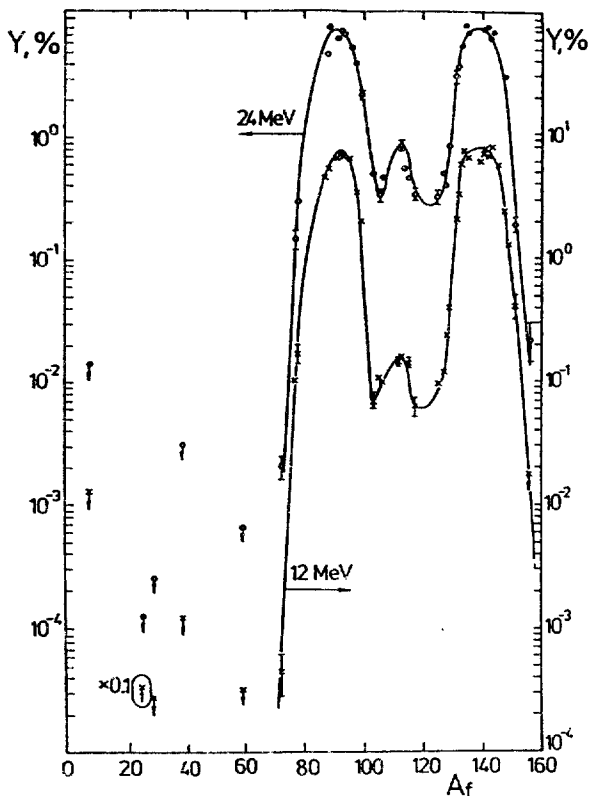


Figure 1: Final fragment mass-distributions of the bremsstrahlung-induced <sup>232</sup>Th fission.

One can see in Fig. 1 that the symmetric mode peak manifests itself more clearly than it was shown in ref.<sup>1,3</sup> after measurements by the  $E_1 \times E_2$  method. Assuming there are three Gaussians the mass-distribution was decomposed and the mass-variance parameters were estimated as  $\sigma_s = 5.1 \pm 0.5$  for the symmetric peak and  $\sigma_a = 6.2 \pm 0.5$  a.m.u. for the asymmetric ones. If one assumes the existence of three fission modes (five peaks), the mass-distribution unfolding gives lower values for the standard I and standard II mass-variance parameters. The same as in other cases the unfolding procedure could not be absolutely reliable.

The relative abundance of the symmetric-to-asymmetric modes,  $Y_{sym}/Y_{asym}$ , (in bimodal decomposition) can be evaluated accurately because of the distinctive maxima in Fig. 1. The result in comparison with ref.<sup>1,3</sup> data is given in Fig. 2 as a function of the bremsstrahlung end-point energy,  $E_c$ . Good agreement is observed. The spectrum of excitation for the fissioning compound nuclei was calculated using the known spectrum of the bremsstrahlung radiation as well as literature data on the giant resonance cross-section and fission probability.<sup>5,6</sup> The mean excitation energy values were found to be 8.0 and 13.5 MeV at  $E_c = 12$  and 24 MeV, respectively. Thus, our experiment confirms previous measurements of the symmetric mode probability and deduced value of the symmetric fission barrier,  $B_f = 7.6$  MeV for  $^{232}\text{Th}$ .

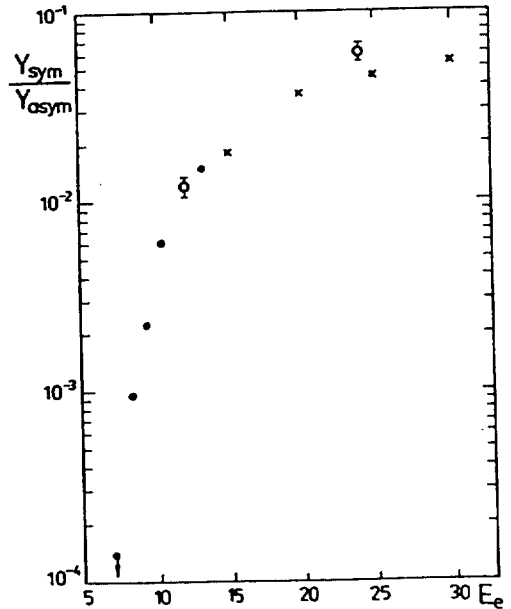


Figure 2: Symmetric-to-asymmetric modes abundance ratio as a function of  $E_c$  as it was measured in ref.<sup>1</sup> - •, ref.<sup>3</sup> - × and in present experiment - o.

## 2 Cluster emission probability

Spontaneous cluster decay of heavy nuclei from ground states as well as cluster emission from highly excited ( $E^* \geq 70$  MeV) compound-nuclei (c.n.) were described in literature. Thus, emission of light nuclei at moderate excitation energies of about 10-20 MeV is actual for the experimental studies. One can assume the emission

from a spherical c.n. and from a strongly-deformed fissioning system. In the present work the yield of such light nuclei as  ${}^7\text{Be}$ ,  ${}^{24}\text{Na}$ ,  ${}^{28}\text{Mg}$ ,  ${}^{38}\text{S}$ ,  ${}^{48}\text{Ca}$  and  ${}^{59}\text{Fe}$  is searched for in products of photon-induced activation of Ta and Th targets at the bremsstrahlung end-point energy  $E_c = 24$  MeV.

The same method was applied as described above for the fission fragments. In addition, the  ${}^{48}\text{Ca}$  doubly magic cluster yield was estimated by the complementary product ( ${}^{133}\text{I}$ ) yield using the radiochemical isolation of the iodine fraction from a gram-weight metal Ta target. One could expect an enhanced probability of the c.n. decay in the case of both fragments being spherical, near-magic nuclei. However, as low as  $\leq 10^{-11}$  limit was reached for the probability of the  ${}^{181}\text{Ta}^* \rightarrow {}^{48}\text{Ca} + {}^{133}\text{I}$  reaction. For lighter nuclei,  ${}^{24}\text{Na}$  etc., the probability limits are presented in the Table. The symmetric fission fragments in the Ta target irradiation were not detected, and it was explained by a high-enough fission barrier,  $B_f \approx 25$  MeV for Ta. The sensitivity level reached for any individual product varied from  $10^{-8}$  to  $10^{-10}$  in dependence on its individual radioactive properties. A significant background in the  $\gamma$ -detector is due to the  ${}^{182}\text{Ta}$  nuclide produced in the  $(n,\gamma)$ -reaction after the Ta irradiation on the microtron beam.

In the case of the  ${}^{232}\text{Th}$  target an intensive  $\gamma$ -radiation of the fission fragments creates a much higher counting rate in the spectrometer and disturbs the weak  $\gamma$ -lines detection. The  ${}^{24}\text{Na}$  and  ${}^{28}\text{Mg}$  nuclides have half-lives of 15 and 21 h and abundant high-energy  $\gamma$ -lines of 2754 and 1779 keV, respectively. These properties are optimal for their revealing in the presence of other radionuclides. However, the analysis of the fission fragment  $\gamma$ -spectra showed that very weak  $\gamma$ -lines of 2752.8 and 1778.6 keV were emitted in the decay chains of  ${}^{112}\text{Pd} \rightarrow {}^{112}\text{Ag} \rightarrow {}^{112}\text{Cd}$  and  ${}^{132}\text{Te} \rightarrow {}^{132}\text{I} \rightarrow {}^{132}\text{Xe}$ , respectively (see Nucl. Data Sheets). These lines disturb the detection of the  ${}^{24}\text{Na}$  and  ${}^{28}\text{Mg}$  nuclides. Only upper limits of the emission probabilities were finally deduced and they were not as deep as in the case of the Ta target. The results are given in the Table.

Table: Relative yields of light nuclei normalized to the  $(\gamma,n)$ -reaction yield at  $E_c = 24$  MeV.

Product	$T_{1/2}$	${}^{181}\text{Ta}$ target		${}^{232}\text{Th}$ target	
		Q, MeV	Yield	Q, MeV	Yield
${}^7\text{Be}$	53.3 d	-10.3	$\leq 1.7 \cdot 10^{-7}$	-4.2	$\leq 1.2 \cdot 10^{-5}$
${}^{24}\text{Na}$	15.0 h	28.0	$\leq 0.7 \cdot 10^{-10}$	50.9	$\leq 1.1 \cdot 10^{-7}$
${}^{28}\text{Mg}$	20.9 h	37.3	$\leq 1.2 \cdot 10^{-10}$	69.0	$\leq 2.3 \cdot 10^{-7}$
${}^{38}\text{S}$	2.84 h	56.7	$\leq 1 \cdot 10^{-10}$	87.0	$\leq 2.8 \cdot 10^{-6}$
${}^{48}\text{Ca}({}^{133}\text{I})$	(20.8 h)	81.7	$\leq 1.5 \cdot 10^{-11}$	-	-
${}^{59}\text{Fe}$	44.5 d	84.8	$\leq 3.1 \cdot 10^{-8}$	125.5	$\leq 5.8 \cdot 10^{-7}$

Emission of light nuclei in the ternary fission of  $^{242}\text{Pu}$  c.n. excited to  $E^* \geq 20$  MeV was discussed in ref.<sup>7</sup> Emission from a fissioning system is not a subbarrier process because of a large elongation of the scission point and, respectively, a decreased barrier. Therefore, the emission probability is regulated by the formation and partition probabilities for the third fragment, rather than the barrier penetration factor. Thus, one can expect a weak energy dependence for the ternary fission probability, and, indeed, the probability of the long-range  $\alpha$ -particle emission shown in Fig. 3a demonstrates a surprising stability with the excitation energy. In Fig. 3b the  $^{24}\text{Na}$  nuclei emission probability is plotted by the results of ref.<sup>7,8</sup> and our values of the upper limit. For the bremsstrahlung induced reactions the horizontal bars show the width of energy distribution for the fissioning nuclei. From Fig. 3b one can conclude that the cluster emission can be detected in photofission of  $^{232}\text{Th}$  at  $E_\gamma = 24$  MeV if the sensitivity of the experiment is improved by one or two orders of magnitude. Being measured, the cluster emission probability may throw some light on the potential energy and dynamics of the fissioning system on the way deviated from the standard fission valleys in deformation coordinates. This is because some special shape should be precursive to the ternary fission with a massive enough third fragment.

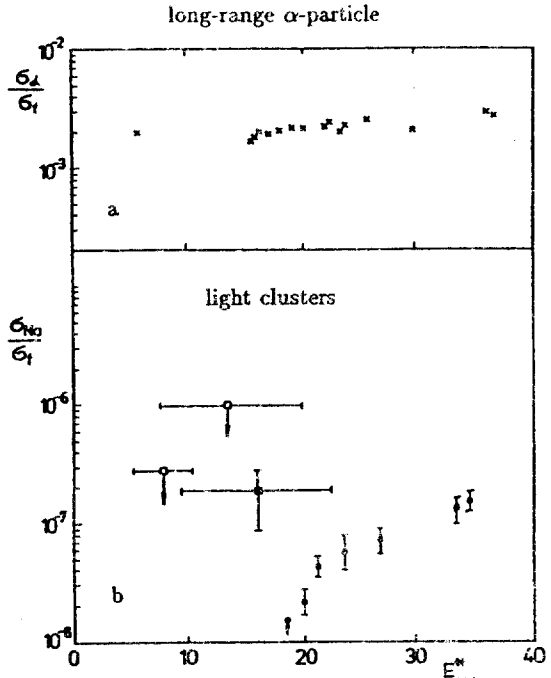


Figure 3: Excitation function for the ternary fission relative probability plotted: a) by data compiled in ref.<sup>4</sup> for long-range  $\alpha$ -particle emission and b) for  $^{24}\text{Na}$  production by the results of ref.<sup>7</sup> -  $\circ$ , ref.<sup>8</sup> -  $\blacksquare$ , and present experiment -  $\square$ .

### 3 Summary

Mass-distribution of the  $^{232}\text{Th}(\gamma, f)$ -reaction is measured and the symmetric mode relative yield and mass-variance parameters are determined at two values of the mean excitation energy. The probability of light nuclei ( $^{24}\text{Na}$ , etc.) emission from the fissioning system is estimated with a sensitivity limited on the level of  $10^{-7}$ . The reliable detection of this process seems to be important as a probe of the potential surface at nonstandard deformations. The cluster emission probability from an excited up to 24 MeV  $^{181}\text{Ta}$  nucleus is limited on the deep level of about  $10^{-10}$  -  $10^{-11}$ , which confirms that the Coulomb barrier penetration factor is very strong for the subbarrier emission.

### Acknowledgements

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Карамян С.А. и др.

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Измерение массового распределения осколков деления и оценки вероятности эмиссии кластеров в реакциях  $\gamma + {}^{232}\text{Th}$  и  ${}^{181}\text{Ta}$

Измерены кумулятивные выходы радионуклидов и построено массовое распределение осколков деления в реакции  ${}^{232}\text{Th} (\gamma, f)$  при граничной энергии тормозных квантов 12 и 24 МэВ. Для мишеней из Th и Ta, облученных тормозным излучением с энергией  $E_e = 24$  МэВ, получены верхние пределы выхода легких ядер  ${}^{24}\text{Na}$ ,  ${}^{28}\text{Mg}$ ,  ${}^{38}\text{S}$  и др. Результаты обсуждаются на основе представлений о мультимодальном механизме деления и эмиссии кластеров из деформированной делящейся системы или из составного ядра.

Работа выполнена в Лаборатории ядерных реакций им.Г.Н.Флерова ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна, 1997

Karamian S.A. et al.

E15-97-11

Fission-Fragment Mass Distribution and Estimation of the Cluster Emission Probability in the  $\gamma + {}^{232}\text{Th}$  and  ${}^{181}\text{Ta}$  Reactions

Fission-fragment mass distribution has been measured by the cumulative yields of radionuclides detected in the  ${}^{232}\text{Th} (\gamma, f)$ -reaction at the bremsstrahlung end-point energies of 12 and 24 MeV. The yield upper limits have been estimated for the light nuclei  ${}^{24}\text{Na}$ ,  ${}^{28}\text{Mg}$ ,  ${}^{38}\text{S}$  etc. at the Th and Ta targets exposure to the 24 MeV bremsstrahlung. The results are discussed in terms of the multimodal fission phenomena and cluster emission from a deformed fissioning system or from a compound nucleus.

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

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