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ON THE SPONTANEOUS EMISSION OF CLUSTERS BY THE ^{230}Th , ^{237}Np and ^{241}Am NUCLEI

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New results concerning the exotic decay modes of the ^{230}Th , ^{237}Np and ^{241}Am nuclei are presented. For studying the emitted clusters we used polyethylene-terephthalate detectors which record only particles with $Z \geq 6$. The radioactive decay of ^{230}Th by ^{24}Ne nuclei emission was detected with a branching ratio relative to alpha decay $\lambda_{cl}/\lambda_\alpha = (5.6 \pm 1.0) \times 10^{-13}$. For the ^{237}Np and ^{241}Am decays by heavy cluster emission the upper limits for the branching ratio were set at about 4×10^{-14} and 3×10^{-15}, respectively.

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

О спонтанном испускании кластеров ядрами ^{230}Th , ^{237}Np , ^{241}Am

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Сообщаются новые результаты по изучению экзотического распада ядер ^{230}Th , ^{237}Np и ^{241}Am с вылетом кластеров и регистрации их полиэтилентерефталатом, чувствительным к частицам с $Z \geq 6$. Открыт радиоактивный распад ^{230}Th с испусканием ^{24}Ne; относительная вероятность процесса по отношению к α-распаду $\lambda_{кл}/\lambda_\alpha = (5{,}6 \pm 1{,}0) \cdot 10^{-13}$. Для распада ядер ^{237}Np и ^{241}Am с вылетом тяжелых кластеров установлен верхний предел вероятности процесса на уровне $4 \cdot 10^{-14}$ и $3 \cdot 10^{-15}$, соответственно.

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During the last few years intensive theoretical and experimental studies were carried out concerning the natural decay modes of the very heavy nuclei involving the emission of heavy clusters [1-5]. In this paper the new decay modes of the ^{230}Th, ^{237}Np and ^{241}Am nuclei are studied.

This phenomenon was investigated using the technique described earlier [1].

Plane sources were prepared on a 0.1 mm nickel backing in the form of the oxides of the isotopes being investigated. The special-purpose technology used for the preparation of the sources eliminated the contamination of the detectors by the very active specimens under study during long exposures. The quantity, composition and area of the radioactive sources used are given in Table 1.

Table 1

Source	Composition		Isotope weight (mg)	Surface area (cm^2)	Alpha-activity $\alpha/cm^2 day(2\pi)$
Th	^{230}Th	40.6%	210	1040	6.7×10^9
	^{232}Th	59.4%	310		5.3×10^4
Np	$^{237}Np^*$		320	500	8.3×10^8 (total) 7.2×10^8 (only ^{237}Np)
Am	^{241}Am		3,7	36	5.6×10^{11}

* This source contained an admixture of $^{238-240}Pu$ (13% according to alpha-activity).

For studying the emitted clusters the 175 μm polyethyleneterephthalate detectors were irradiated in air using Th and Np sources at a distance of 1 mm and with an Am source in vacuum at a distance of 1 cm. The detectors allowed one to detect clusters with atomic numbers $6 \leq Z \leq 20$ and separate them from fission fragments in the calculated energy range from 35 to 85 MeV [1,6].

The exposure time was determined taking into account the alpha-activity of each source. As was previously shown [1], for accurate spectrometric measurements in polyethyleneterephthalate detectors the integral alpha particle flux has to be lower than 5×10^{11} α/cm^2.

Inside the polymer material irradiated in air, some processes occur along the particle trajectory leading to a change in the material structure and this affecting the measured geometrical parameters of the track [6]. It was experimentally shown that this process, i.e., the change in the etched track length, in the case of 46 MeV ^{20}Ne ions, is finished at most one month after the irradiation. Moreover, the high alpha particle flux makes this process faster. Nevertheless, in order to ensure that all the recorded cluster tracks, in the case of a very long exposure time, have the same etching characteristics, the detectors were stored 30 days in air prior to the chemical processing, which was carried out in a 20% NaOH solution at a temperature of 60°C.

Together with the detector exposed to the radioactive sources, there were etched the calibration samples irradiated with ^{16}O, ^{20}Ne, and ^{26}Mg ions with energies of 1.6-3.0 MeV/nucleon and with dip angles of 30° and 45°. It has to be mentioned that the calibration samples were exposed to sources together with the detectors.

The geometric efficiency of cluster detection was equal to 0.66 2π.

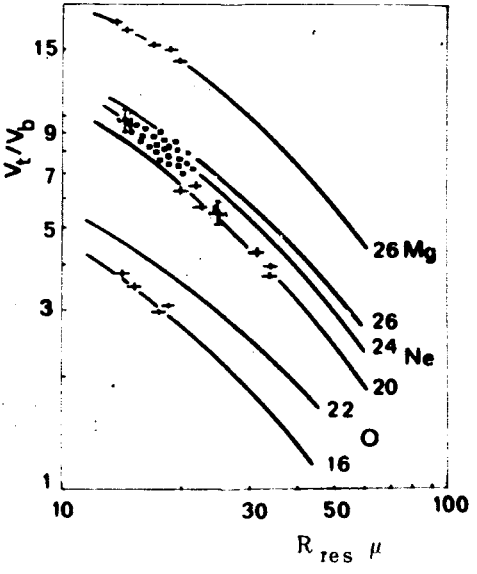
Since the identification method chosen is the variation of the V_t/V_b with the residual range, the detectors were etched 2-3 times for different time intervals. After a total time of 4 hours, the etching reached the cluster stopping point. From the cone geometrical parameters, measured under optical microscopes with a magnification of 16x25x40, the track length and V_t/V_b were calculated.

Using the calibration with ^{16}O, ^{20}Ne and ^{26}Mg ions, the $V_t/V_b = f(dE/dx)$ dependence was obtained for the given alpha particle flux of 2×10^{11} α/cm^2: $V_t/V_b = 6.3 \times 10^{-3} (dE/dx)^{2.83}$ and dE/dx is expressed in MeV cm^2/mg.

By taking into account the dE/dx - range relationship, the V_t/V_b variation with the residual range for different ions was obtained (Fig.1). As can be seen, the experimental points measured with the Th source are situated around the curve for the ^{24}Ne ions. The data treatment using the minimum method χ^2 leads to the conclusion that ^{24}Ne is the most probable type of the decay cluster. The V_t/V_b values have been obtained only for a part of the cluster tracks revealed, as the corresponding measurements are too time-consuming.

The track length distribution for the decay products of Th, given in figure 2, has a maximum at about 29 μm, which is the range of the ^{24}Ne ions corresponding to the calculated value of the kinetic energy of the emitted heavy cluster.

Fig.1. The calculated dependences of the etching selectivity V_T/V_B (where V_T and V_B are the rates of polymer etching along the track and the bulk etch rate for the detector material, respectively) on the residual range R_{res}. The dots indicate experimental data for clusters, the crosses give the results of measurements with control detectors exposed to ^{16}O, ^{20}Ne and ^{26}Mg ions with energies of 1.6 to 3.0 MeV/nucleon.



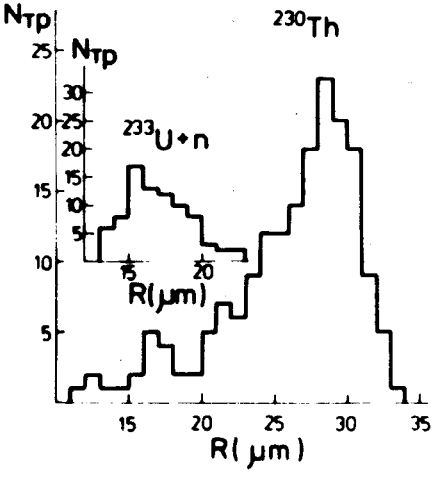


Fig.2. The distribution of the full ranges of the ^{230}Th decay products and those of fragments from the neutron-induced fission of ^{233}U in polyethyleneterephthalate. The thickness of the ^{230}Th and ^{233}U sources was equal to 0.6 and 0.3 mg/cm^2, respectively.

The distribution of the full ranges R is rather broad. This is due to some experimental shortcomings, namely a large source thickness and a rather high recoil nuclei background, which have introduced some ambiguities in the estimate of the cluster mass number because of difficulties either in the estimation of the self-absorption process in the source or in the measurements of the geometrical parameters of the track.

By taking into account the theoretical predictions[2,9] as well as the experimental results (figs.1 and 2), one can conclude that the new decay mode of ^{230}Th takes place most probably by the emission of the ^{24}Ne ion.

The effect of Ne emission from the $^{230}Th + ^{232}Th$ isotopic mixture (see table 1) can practically be attributed to the ^{230}Th isotope. Theoretical and experimental data [2-5] show that ^{223}Ra has the highest probability, relative to alpha decay, to emit nuclei heavier than helium: for ^{14}C nuclei emission its relative probability is equal to about 6×10^{-10}. In our case, even if one may consider ^{232}Th to have the relative probability for Ne emission close to this maximum value, due to the difference between the alpha-decay half-lives of ^{230}Th (7.54×10^4 years) and ^{232}Th (1.4×10^{10} years) the contribution of the latter isotope to the whole effect will not exceed 1%.

No clusters have been detected from the Np and Am sources, this fact being additional evidence for the absence of a background in our experiment.

The experimental conditions and results are given in Table 2 and theoretical data are presented in Table 3.

Table 2

Isotope	Alpha-activity per exposition (α/cm^2)	Exposure			$(\lambda_{cl}/\lambda_\alpha)_{exp.}$
		Nr.	Time (days)	Nr.	
^{230}Th	2.2×10^{11}	1	33	90	$(5.6 \pm 1.0) \times 10^{-13}$
	2.1×10^{11}	2	31	75	
^{237}Np	2.6×10^{10}	1	35	0	$< 4 \times 10^{-14}$
	6.5×10^{10}	2	87	0	
^{241}Am	5.6×10^{12}	1	10	0	$< 5 \times 10^{-15}$
		2	10	0	
		3	10	0	

As is seen from Table 3, ^{24}Ne emission from ^{230}Th is energetically most favourable compared with other neon isotopes. As cluster emission is a subbarier process, even relatively little changes in the values of the mass difference between the initial and final nuclei, Q, lead to great changes in the cluster emission probability. Thus, our experimental evidence that the neon isotope emitted in ^{230}Th decay is most likely to have mass number 24 agrees with theoretical estimates.

It is worth mentioning that from the experimental data [7] as well as from the ^{226}Ra quantity contained in

the $^{230}Th + ^{232}Th$ source, one should expect about 40 tracks of ^{226}Ra decay by the emission of ^{14}C. Due to the sensitivity of the detector, the maximum etched track length for ^{14}C ions is equal to about 10 μm. Since the C and O recoil nuclei have lower energies, they are etched faster than the ^{14}C cluster tracks. After a longer etching time the background becomes so high that it is impossible to discriminate the ^{14}C cluster tracks against the recoil nuclei tracks.

Table 3

Isotope	Q, MeV	$\lambda_{cl}/\lambda_{\alpha}$ [2]	$\lambda_{cl}/\lambda_{\alpha}$ [8]	$\lambda_{cl}/\lambda_{\alpha}$ [9]
^{22}Ne	53.0	$<10^{-18}$		
^{23}Ne	53.1	$<10^{-18}$		
^{24}Ne	57.8	2.5×10^{-12}		1.6×10^{-13}
^{25}Ne	55.3	2.5×10^{-17}		
^{26}Ne	55.1	2.0×10^{-18}		
^{22}O	43.2	1.3×10^{-14}	3.5×10^{-12}	

The present experiments have not been aimed at studying spontaneous fission. Nevertheless, in experiments with thorium there have been detected 25 spontaneous fission fragments which differ from clusters both in the range (see fig.2) and shape. Bearing in mind the possible sources of the background, for example, thorium fission induced by neutrons from the (α,n) reaction on the light nuclei of the detector material, the effect obtained can be used only for estimates of the lower limit of the spontaneous-fission half-life of ^{230}Th. According to our data, this limit is set at $\geq 2\times10^{18}$ years, that is one order of magnitude higher than the previously known value of 1.5×10^{17} years [10]. This value should be attributed just to ^{230}Th since the spontaneous-fission half-life of ^{232}Th makes up $\geq 10^{21}$ years [11]. The partial half-life for ^{24}Ne-emission is $(1.3\pm0.3)\cdot10^{17}$ years.

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