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OBSERVATION OF NUCLEON CLUSTERS IN THE SPONTANEOUS DECAY OF ^{234}U

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1. Introduction

Since the work of Rose and Jones /1/ the study of the spontaneous decay of heavy element nuclei involving emission of nucleon clusters, such as ^{14}C and ^{24}Ne, continues to arouse considerable interest.

The new mode of radioactive decay was detected in 9 isotopes of Ra, Th, Pa, and U /2/. The experimental data accumulated permit the study of the dependence of the cluster emission probability on the atomic number Z and mass number A of both the initial nucleus and the cluster emitted. It can also be possible to reveal the influence of the nuclear parity effect, and determine the ratio of decay probabilities for one nucleus emitting different clusters. The semiempirical dependences of the Geiger-Nuttall type for α-decay obtained from the set of data allow one to give more precise estimates of cluster decay probabilities for as yet unstudied nuclei.

The experimental data on the spontaneous emission of particles with masses intermediate between those of α-particles and fission fragments give an impetus to an analysis of the difference and similarity of the α-decay and fission of nuclei.

In this context it is interesting to investigate cluster emission from nuclei that undergo both α-decay and spontaneous fission. Bearing in mind the experimental possibilities and the theoretical estimates of the cluster emission probability /3-6/, the studies of uranium isotopes look most promising /7/. The emission of Ne nuclei was established to occur in ^{232}U /8/ and ^{233}U /9/ decay. For ^{234}U some 11 decay events accompanied by Ne emission and 3 events involving Mg emission were detected /10/.

The presently known results about cluster emission from nuclei heavier than radium were obtained using the solid-state track detector technique at Berkeley /2/ and at Dubna /11/. The results obtained by the two groups are complementary to one another since they used different detector materials and different processing methods. This fact is of great importance in investigating processes which can be recorded at the limiting sensitivity of the modern techniques. The present paper deals with the study of cluster emission in the spontaneous decay of ^{234}U, ^{235}U, and ^{236}U.

2. Experimental technique

Experiments were carried out using three radioactive samples the characteristics of which are given in table 1. The isotopic composition of the samples was determined by mass-spectrometric and α-spectrometric methods.

As in our earlier work /9,11/, the products of uranium decay were recorded by lavsan (polyethyleneterephthalate) track detectors sensitive to particles with charges $Z \geqslant 6$. The detectors were exposed in the air in a geometry close to 2π. The track detection was carried out in the angular range 15° - 70° with respect to the detector plane. This yielded a cluster detection efficiency equal to 0.33 of 4π.

For obtaining the visible (under a microscope) optical image of a particle track the detectors were etched by a 20% NaOH solution at a temperature of 60°C. The etching procedure included several stages. The revelation of the full Ne track length of about 30 μm took about 4 hours. The whole etching process was divided into several 30-min steps. After each step there were

Table I. Characteristics of the uranium samples and the numbers of the clusters detected

Isotope	Isotope concentration, %		
	sample 1	sample 2	sample 3
Uranium-232	-	0.0012	-
234	0.85	98.05	0.001
235	89.80	0.13	0.047
236	-	0.24	99.845
238	9.35	1.58	0.107
Total amount of uranium, mg	1350	18.5	25.3
Square, cm^2	1940	256	46
Exposure duration, days	233	420	160
Number of Ne tracks	7	24	0
Number of Mg tracks	3	13	0

carried out the search and measurement of the parameters of the tracks revealed in scanning under an optical microscope. Such a procedure makes it possible to obtain some data on the kinetics of etching.

To identify the particle producing a track use was made of the dependence of the etching selectivity V_T/V_M (V_T is the velocity of etching along the track in detector material, and V_M is the etching velocity outside the damage zone produced by the particle detected) upon the value of the residual particle range R which is the difference between the length of the fully etched track and that of its part etched by the moment of terminating a given etching step. The detectors were calibrated using 1.6 - 3.0 MeV/amu ^{20}Ne and ^{26}Mg ion beams from the U-300 cyclotron of JINR. It was established that the ratio V_T/V_M was affected by the magnitude of the integrated α-particle flux to which the detector material was exposed in the air on a radioactive sample. This dependence can be approximated in our measurements by the following expression

$$V_T/V_M = a(dE/dx)^b, \qquad (1)$$

where a = 0.0063, b = 1.83 + 0.81×($\Phi_\alpha \times 10^{-9})^{0.04}$, dE/dx is the

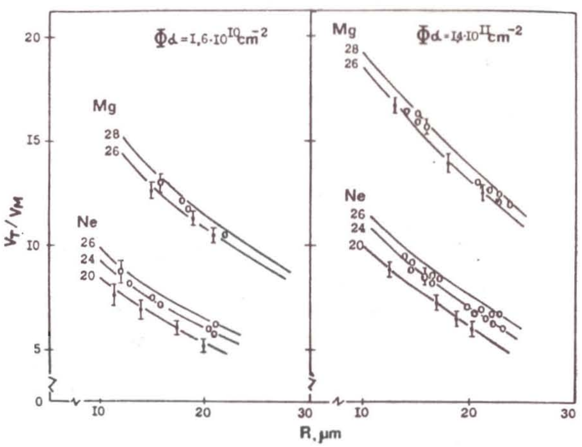

Fig. 1. Dependence of the etching selectivity V_T/V_M upon the residual range. The curves are drawn according to eq.(1). The calibration data on ^{20}Ne and ^{26}Mg are indicated by closed circles. The results of processing some of the tracks revealed in studies of ^{234}U decay are shown by open circles.

specific ionization in units of MeV cm^2/mg, and Φ_α is the number of α-particles that have passed through 1 cm^2 of the detector.

To take this effect into account the detector samples calibrated by ions at the U-300 cyclotron were exposed in the air on the uranium samples studied under the same conditions as the detectors for recording spontaneous decay.

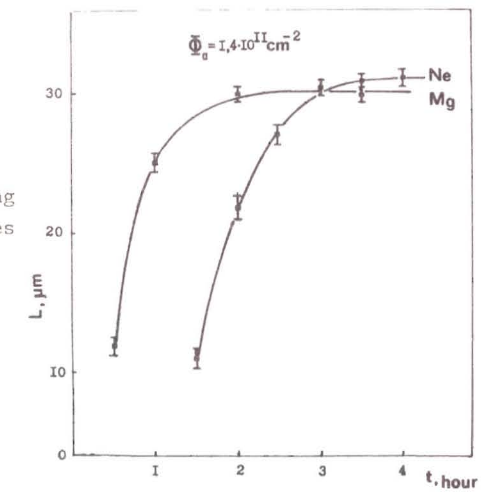

Fig.2. Dependence of the etchable track length on etching time for Ne and Mg. The curves drawn through the data are to guide the eye.

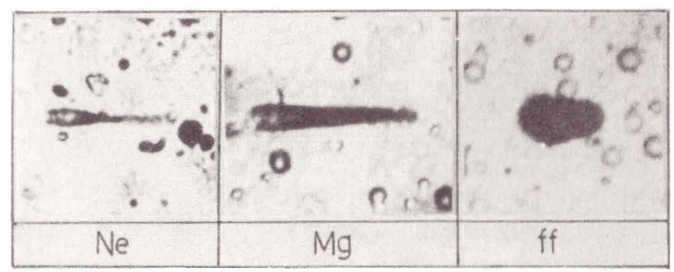

Fig. 3. Profiles of Ne, Mg and fission fragment tracks with the same magnification of x1200 after etching during 3.5 hours. The circular tracks of different diameters are due to recoil nuclei knocked out by α-particles.

The scattering of α-particles and fission fragments on light nuclei incorporated in the source and detector materials produces the secondary background due to recoil nuclei with $Z \geqslant 6$. In order not to exceed the number of recoil nuclei over the one making cluster observation difficult, the time of exposure was limited for each detector by the admissible α-particle flux ($\lesssim 10^{12}$ α-particles per cm^2).

Measurements of tracks during multi-stage etching have the advantage that one and the same track can be used repeatedly for identifying the particle which has produced it. The results of calibrations and measurements for ^{234}U are given in fig. 1. The fact that in the etching process Mg tracks were revealed considerably earlier than Ne tracks because of the higher specific ionization of the former also alleviated the search and enhanced the reliability of track identification. This is demonstrated in fig. 2. The profiles of the tracks due to Ne, Mg and a fission fragment measured with the same magnification factor after 3.5-hour etching are shown on a microphotograph (fig. 3).

3. Results of measurements and discussion

The numbers of the Ne and Mg tracks revealed in each sample are given in table I. From the data presented in fig. 1 it follows that the most probable masses of Ne and Mg clusters are 24-26 and 28, respectively. Our technique of processing solid-state track detectors in the regions of atomic numbers $Z \lesssim 15$ and mass numbers $A \lesssim 30$ provides the resolutions $\Delta Z \sim \pm 0.15$ and $\Delta A \sim \pm 1$. The comparison of the magnitude of the effect observed on samples 1 and 2, made taking into account the amount of ^{234}U

Table II. Data on the probabilities of cluster emission from the ^{234}U, ^{235}U, and ^{236}U nuclei

Nucleus		^{234}U				^{235}U				^{236}U				Ref.
Cluster		Ne		Mg		Ne		Mg		Ne		Mg		
Experiment	$T_{1/2}$, yr	$(6.3^{+2.1}_{-1.3})\times10^{17}$		$(1.1^{+0.4}_{-0.3})\times10^{18}$		$>1.4\times10^{20}$		$>9\times10^{20}$		$>6\times10^{18}$		$>6\times10^{18}$		present paper
		$(3.7^{+1.2}_{-0.9})\times10^{17}$		$(1.1^{+1.3}_{-0.6})\times10^{18}$		–		–		–		–		/10/
	$-\log(\frac{\lambda_{cl}}{\lambda_\alpha})$	$(12.41^{+0.13}_{-0.10})$		$(12.64^{+0.16}_{-0.12})$		>11.3		>12.1		>11.4		>11.4		present paper
		(12.18 ± 0.12)		(12.65 ± 0.34)		–		–		–		–		/10/
Cluster		^{24}Ne	^{26}Ne	^{28}Mg	^{30}Mg	^{25}Ne	^{26}Ne	^{28}Mg	^{30}Mg	^{24}Ne	^{26}Ne	^{28}Mg	^{30}Mg	
Theory	$-\log(\frac{\lambda_{cl}}{\lambda_\alpha})$	11.9	12.3	10.6	15.1	12.0	12.3	11.2	12.3	14.7	14.9	12.1	12.1	/3/
		13.2	12.9	11.1		13.4	12.5		11.7				12.0	/4/
		14.0	14.4	13.4		12.8	13.1	12.6	13.7			15.0	14.8	/5/
		11.8	11.1	11.0		12.9	12.0		12.3				10.9	/6/

in each sample and the exposure time, shows that the observed effect of cluster emission is likely to be related to ^{234}U alone. This conclusion ensues from the fact that the difference between the effects which could be attributed to ^{235}U is equal to 0.0 ± 3.2 for Ne and -1.6 ± 2.1 for Mg, i.e. it is within the measuring errors. Together with theoretical estimates, the obtained values of the cluster emission probability relative to the α-decay probability are listed in table II. The experimental data take into account the contribution from ^{232}U decay to the total number of the detected tracks in samples 1 and 2, which is 4 events for Ne and not more than 0.2 events for Mg. Our ^{234}U results agree fairly well with those of work /10/. Although the statistical accuracy is not very high, both our measurements and work /10/ give the lower probability for Mg emission than that for Ne whereas theory predicts the converse /3-6/.

The mere systematics of the experimental data obtained are presented in fig. 4. This figure shows the logarithms of the partial half-lives of uranium isotopes emitting helium, neon and magnesium nuclei as functions of the natural logarithm of the Coulomb potential barrier penetrability. This approach is similar to the simple theoretical justification of the Geiger-Nuttall law for α-decay /12/ and has already been used in the analysis of the cluster emission decay earlier /2,13/. The barrier penetrability was calculated using the following relation

$$P = \exp\left\{-2/\hbar \int_{R_N}^{Z_1 Z_2 e^2/Q} \left[2 \frac{A_1 A_2}{A_1 + A_2} u \left(\frac{Z_1 Z_2 e^2}{r} - Q \right) \right]^{1/2} dr \right\}, \qquad (2)$$

where A_1, A_2 and Z_1, Z_2 are the mass and atomic numbers of the cluster and the daughter nucleus, respectively; Q is the mass difference between the initial and final nuclei, $\hbar$ is the Planck constant, u is the unit mass, and e is the single charge. The R_N value was taken to be equal to $1.44\ (A_1^{1/3} + A_2^{1/3}) \times 10^{-13}$ cm. The cluster mass numbers given in brackets in fig. 4 correspond to the highest penetrability of the barrier and do not contradict the experimental data the error of which is about one mass unit. The straight lines for He and Ne are drawn through the points for the even-even isotopes of uranium. The position of the

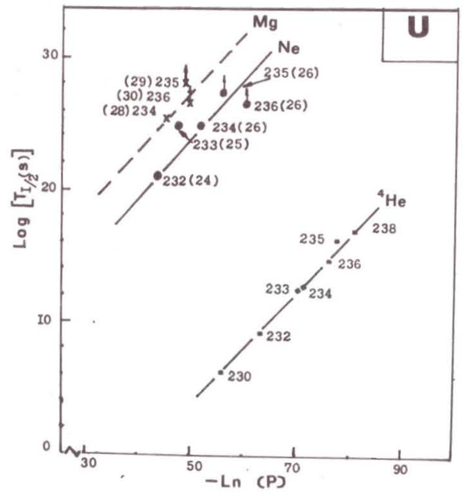

Fig. 4. Dependence of the common logarithm of the partial half-life on the natural logarithm of the Coulomb potential barrier penetrability for the emission of He, Ne, and Mg nuclei from uranium isotopes. Unbracketed figures indicate the mass numbers of the uranium isotopes, bracketed figures are the mass numbers of clusters.

straight line for Mg is determined by the experimental value of the partial half-life of ^{234}U. Its slope is chosen to be the same as that of the straight lines for He and Ne. By analogy with α-decay one can speak about the presence of the hindrance factor for the cluster-emission decay of odd-mass nuclei, which is about 40 and not less than 3 for Ne emission from ^{233}U and from ^{235}U, respectively.

A different choice of the radius parameters or the use of nuclear potential other than a square well will certainly result in a change in the position of points on the abscissa axis in fig. 4. It is however unlikely that they will change the conclusion about the presence of the hindrance factor in the cluster decay of odd-mass nuclei.

4. Conclusion

The entire set of data permits the assertion that ^{234}U decay proceeds via four channels of hadron decay, i.e. α-decay ($T_{1/2}$ = 2.45 x 10^5 years), the emission of Ne nuclei ($T_{1/2} \sim 5 \times 10^{17}$ years), the emission of Mg nuclei ($T_{1/2} \sim 1 \times 10^{18}$ years), and spontaneous fission ($T_{1/2} \sim 2 \times 10^{16}$ years). Bearing in mind the data on the yields of separate fission fragments /14/ one can state that the decay of ^{234}U through the channels involving Ne and Mg production occurs with approximately the same probability as decay in spontaneous fission accompanied by the formation of a certain nuclide near the maximum of the mass distribution. The hindrance factor value for the decay of the odd-mass nucleus ^{233}U accompanied by the formation of Ne particles ($\sim$ 40) lies between that for α-decay (equal to 1.2 for the most intensive transition /12/) and the hindrance factor for

spontaneous fission (> 250 according to the experimentally established limit for ^{233}U spontaneous fission /15/).

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Третьякова С.П. и др. E7-88-803
Наблюдение нуклонных кластеров
при спонтанном распаде ^{234}U

При спонтанном распаде ^{234}U зарегистрирован вылет ядер Ne и Mg с вероятностью по отношению к альфа-распаду $/3,9\pm1,0/\times10^{-13}$ и $/2,3\pm0,7/\times10^{-13}$, соответственно. Установлены верхние границы вероятности такого распада для ^{235}U и ^{236}U. Получена систематика периодов полураспада для эмиссии кластеров, аналогичная зависимости Гейгера-Нэттола для альфа-распада. Установлено наличие запрета на испускание кластеров нечетными ядрами ^{233}U и ^{235}U.

Работа выполнена в Лаборатории ядерных реакций ОИЯИ.

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Observation of Nucleon Clusters in the
Spontaneous Decay of ^{234}U

The emission of Ne and Mg nuclei was detected in the spontaneous decay of ^{234}U with probabilities of, respectively, $(3.9\pm1.0)\times10^{-13}$ and $(2.3\pm0.7)\times10^{-13}$ relative to α-decay. The upper limits of this kind of decay for ^{235}U and ^{236}U have been established. Systematics of half-lives for cluster emission have been obtained to be similar to the Geiger-Nuttall dependence for α-decay. Some evidence for the presence of hindrance to cluster emission from the odd-mass nuclei ^{233}U and ^{235}U is obtained.

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

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