Cluster decay of ^{236}U

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Six magnesium tracks were detected by solid-state track detectors over 390 days of measurements. The probability for this channel for ^{236}U decay is $\sim 2\times 10^{-13}$ of that of α decay. The partial decay half-life is $\sim 1.2\times 10^{20}$ yr.

Each new observation of cluster decay is an important supplement to the existing information.[1] Studies of the cluster decay of isotopes of the same element are particularly valuable, since in this case it is possible to identify shell and deformation effects, which can be seen clearly in α decay and spontaneous fission.[2] We have accordingly carried out an experimental search for a cluster decay of ^{236}U. Before this study was undertaken, a cluster decay of $^{232,233,234}U$ had been detected, and lower limits had been found on the decay half-lives for $^{235,236}U$ (Refs. 1 and 3–7).

The clusters were detected by the well-developed method of solid-state track detectors.[1,8] We had at our disposal 1296 mg of uranium (in terms of the metal), in the form of 40 radioactive sources. Each source consisted of a layer of U_3O_8, with a thickness of 4.25 mg/cm^2 and an area of 3×3 cm^2, on an aluminum substrate 0.2 mm thick. The isotope composition of the source was

$$<0.001\%\ ^{234}U,\quad <0.047\%\ ^{235}U,\quad 99.845\%\ ^{236}U,\quad 0.107\%\ ^{238}U.$$

As the track detector we used polyethylene terephthalate (polyester) 200 μm thick. The calibration of the detectors required for identifying tracks was carried out in ^{20}Ne, ^{24}Mg, ^{27}Al, and ^{28}Si beams with an energy of 0.5–3.0 MeV/nucleon, accelerated at the U-400 cyclotron of the G. N. Flerov Nuclear Reactions Laboratory of the Joint Institute for Nuclear Research. The detectors were in contact with the source material for 390 days. The method used to measure the properties of the tracks, involving an etching in several time steps,[1,8] makes it possible to reliably determine the atomic number of the particle which forms the track, but this method is relatively insensitive to the mass number A (the accuracy is $\sim\pm 1$). However, in determining the mass number of a cluster, we can generally rely on the value of the energy balance Q. Since cluster decay is a deep subbarrier process, the isotope corresponding to the largest value of Q is the most likely to be emitted. In the case of cluster decay of ^{236}U, the most favorable values of Q are those of ^{30}Mg (Refs. 9 and 10).

At the detectors, six magnesium tracks with a range of 13–21 μm in the polyester were detected. According to range–energy curves,[11] this range corresponds to ^{30}Mg

energies of 24–45 MeV. These energies are in the range we would expect them to lie for the two-body decay $^{236}U \to {}^{30}Mg$ with $Q = 72.5$ MeV, when we allow for the thickness of the sources at our disposal.

The efficiency of the cluster detection in our experiments was 0.3 of 4π . Consequently, the data we found lead to a probability for the cluster decay of ^{236}U involving the emission of Mg which amounts to $\sim 2 \times 10^{-13}$ of the probability for α decay. The corresponding partial decay half-life is $\sim 1.2 \times 10^{20}$ yr. The observed effect is lower by a factor ~ 20 than the boundary established in Ref. 7. The most dangerous background source is knockout of recoil nuclei with atomic numbers $Z \geqslant 12$ from light impurities in the material of our sources by fragments of the spontaneous fission of ^{236}U (their total number, over the entire exposure time, was $\sim 1 \times 10^5$). According to the technical specifications of our sources, the concentration of impurities from Na to Si did not exceed 0.01%. To explain the observed effect, their concentration in the source would have to have been $\sim 1\%$. The conditions under which the detectors were etched were selected to develop only those tracks which were formed by particles with $Z \geqslant 10$. This approach prevented the detection of oxygen recoil nuclei formed by fission fragments in the U_3O_8 layer and in the detector material.

Detection of recoil nuclei from the substrate was eliminated by the kinematic limiting angle of scattering of fission fragments by particles of smaller mass and also by the fairly thick layer of U_3O_8. Only in the surface region of the detectors, corresponding to the geometric boundaries of the U_3O_8 layer on the aluminum substrates, did we detect five tracks with properties corresponding to $Z = 13$. We believe that these tracks were probably formed by recoil nuclei from the aluminum substrate.

Because of the low ^{234}U concentration in our sources, the contribution of the cluster decay of this isotope to the effect which we observed does not exceed 0.1%. This high concentration of the isotope ^{236}U also rules out the possibility that $^{235,238}U$ are contributing to the observed effect, according to the measured limit on the probability for the cluster decay of ^{235}U (Ref. 7) and according to theoretical estimates of the probability for the cluster decay of ^{238}U (Refs. 9 and 12). No estimates for ^{238}U yield cluster-decay probabilities higher than for ^{236}U. Furthermore, the cluster

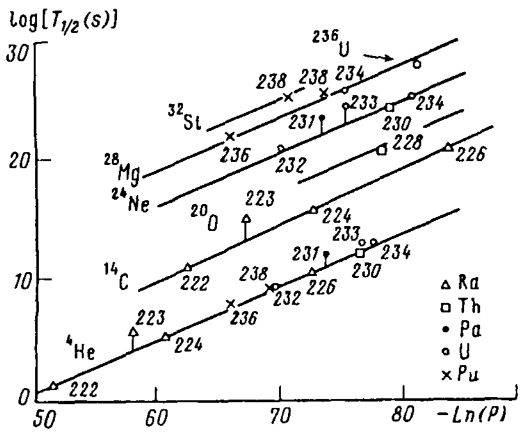

FIG. 1. The base-10 logarithm of the partial decay half-life versus the natural logarithm of the transmission of the Coulomb potential barrier, P, for emission of He, C, Ne, Mg, and Si by isotopes of Ra, Th, Pu, and U. The procedure used to calculate the transmission is the same as in Ref. 14.

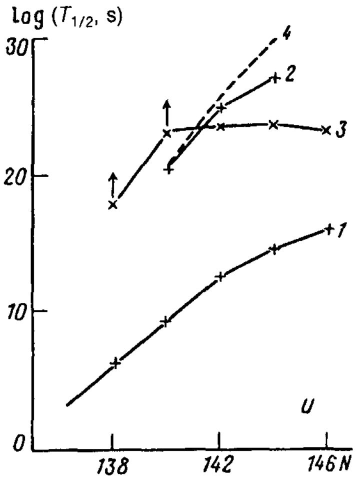

FIG. 2. Partial decay half-lives for the emission of α particles (1), for the emission of clusters (2), and for the spontaneous fission (3) of even–even uranium isotopes versus the neutron number N in the original isotope. The dashed line (4) shows estimates of the partial half-lives for cluster decay according to Refs. 9 and 12. Data from Ref. 6 were used for the spontaneous fission of ^{232}U.

most likely to be emitted would be Si, rather than Mg. With 1×10^5 as the total number of fission fragments detected, the probability for detecting even a single track of Mg formed in triple fission does not exceed 0.01 (Ref. 13).

The partial half-life for decay of ^{236}U accompanied by the emission of Mg which we found was represented in a semiempirical Geiger–Nutall systematics[14] (Fig. 1). From the standpoint of the values of spectroscopy factors,[12] the points for decays involving the formation of ^{30}Mg in this systematics should lie slightly above the straight line connecting the points for decays involving the production of ^{28}Mg. The value which we found for the partial decay half-life of ^{236}U is about an order of magnitude smaller than that expected from the systematics in Fig. 1, although the accuracy of this systematics is of course not great. The theoretical predictions of Refs. 9 and 12 also give expected decay half-lives one or two orders of magnitude greater.

The general tendency in the variation of the half-lives for the cluster decay of even–even isotopes of uranium is similar to the behavior of the α-decay and spontaneous-fission half-lives in the empirical systematics shown in Fig. 2. At $N=142$, the decay half-lives deviate somewhat from a linear extrapolation; the predictions of Refs. 9 and 12 are close to that extrapolation. In the cases of α decay and spontaneous fission, this result can be explained by shell and deformation effects on both the value of Q and the shape of the exit potential barriers.[2] The results for ^{236}U obtained by us experimentally may be taken as an indication that effects of this sort should also be taken into account in cluster-decay theory.

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