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TERNARY FISSION OF PLUTONIUM

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The energy spectra of long-range α particles produced in the spontaneous fission of Pu^{238} and Pu^{240} and in thermal-neutron fission of Pu^{239} are studied by the nuclear emulsion method. The spectrum shapes are discussed and are compared with the results for complex uranium fission.

THE emission of α particles from fissioning heavy nuclei has become of increasing interest; it is considered that this phenomenon must be studied to improve our understanding of the fission mechanism. The most complete information is available regarding the thermal-neutron fission of U^{235},[1] for which the relative probabilities of ternary and binary fission, the α-particle energy spectrum, and several other properties are known. The complex fission of other nuclei induced by both thermal and fast neutrons, has also been investigated, and, more recently, complex spontaneous fission. In these studies attention was directed mainly toward determining the probability of α-particle emission; sometimes the energy distribution and certain other properties have also been determined. Thus we have the α-particle energy spectra from U^{235} and U^{238} fission induced by 14-MeV neutrons,[2] and from the spontaneous fission of Cm^{242} and Pu^{240},[3] together with somewhat inconsistent data for Cf^{252}.[4-7] We believe that it is of interest to compare both the fission probabilities and the fission α-particle energy spectra of different isotopes. The present work is a study of the complex fission of certain plutonium isotopes.

EXPERIMENTAL PROCEDURE

In our study of spontaneous fission we used electrolytic films of Pu^{238} and Pu^{240}, containing 78.2 ± 4 and 450 ± 25 μg respectively. We also irradiated a Pu^{239} film with a neutron beam from the reactor of the Physico-technical Institute of the U.S.S.R. Academy of Sciences in order to obtain the α-particle energy spectrum from thermal-neutron fission. In order to register only long-range α particles emitted by fissioning nuclei, between the type P-9-0 photographic emulsion and the film of active material we placed a foil of a heavy element (Pt, Sn, or Pb) thick enough to absorb fission fragments and the α particles of natural radioactivity. Because of possible track regression in the emulsion and because of the appreciable x-ray background, in the experiments on spontaneous fission the plates were exposed one to two days at most.

Following the photographic processing the plates were scanned microscopically. Tracks of long-range α particles with residual ranges exceeding 50 μ were recorded, thus excluding tracks resulting from the accidental contamination of plate surfaces by α activity. We retained for further examination tracks with dip angles not exceeding 60° to the undeveloped emulsion surface, thus obviating the scanning of highly inclined tracks as well as the registration of protons, which cannot easily be distinguished from α particles at large dip angles. As a control we scanned areas of the photographic plates that lay to the side of the active material during irradiation. In this way only long-range fission α particles were registered.

The energies of α particles having tracks terminating in the emulsion were calculated, the stopping powers being taken from Rybakov's data.[8] When necessary, the total thickness of the active film was taken into account. The energy distribution begins at a limit of 13 MeV determined by the experimental conditions. We introduced corrections for the experimental geometry, using the equation

$$N(E)\,dE = \frac{n(E)\,dE}{\sin\varphi_1(E) - \sin\varphi_2(E)},$$

where N(E) is the actual energy spectrum, n(E) is the observed spectrum $\varphi_1(E)$ is the limiting dip angle at which an α particle of a given energy traverses the entire emulsion thickness and emerges, and $\varphi_2(E)$ is the limiting dip angle at which an α particle traverses the filter and leaves a track of not less than 50 μ in the emulsion. As a test of the procedure a film of U^{235} for which the α-particle energy spectrum can be considered known, was irradiated at the reactor, in addition to Pu^{239}.

EXPERIMENTAL RESULTS AND DISCUSSION

Table I contains the results of the scanning and treatment of the data. Figure 1 shows the energy spectra of fission α particles; as already stated, the histograms begin at 13 MeV.

Table I

Fissioning nucleus	No. of observed α particles	No. of α particles in hemisphere after corrections for geometry
Pu^{239}+ thermal neutrons	121	246
U^{235}+ thermal neutrons	194	404
Pu^{238}(spontaneous)	77	161.5
Pu^{240}(spontaneous)	108	237.6

In [9] the energy distribution of α particles from thermal-neutron U^{235} fission, based on large statistics, showed good agreement with a Gaussian curve having its maximum at 15.15 MeV and a 10-MeV half-width. Similar spectra were obtained for α particles from the fast-neutron fission of uranium isotopes.[2] For the purpose of comparison the histograms in Fig. 1 were also put into the form of Gaussian curves having parameters determined by least squares in such a way that Pearson's test yielded the highest probability of agreement, and not merely compatibility, with the experimental distribution. These parameters are given in Table II.

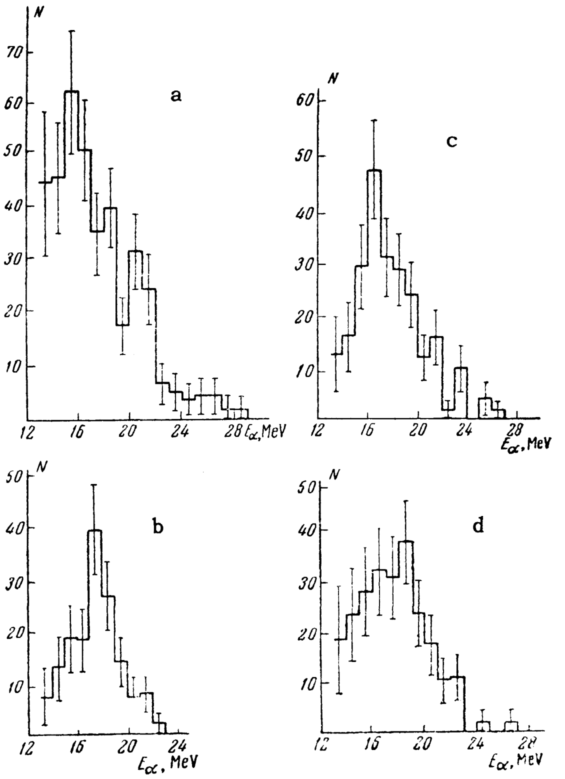

FIG. 1. Energy spectra of long-range α particles from ternary fission. a – thermal-neutron fission of U^{235}; b – thermal-neutron fission of Pu^{239}; c – spontaneous fission of Pu^{238}; d – spontaneous fission of Pu^{290}.

Table II

Fissioning isotope	E_{max}, MeV	ΔE, MeV
1) U^{235}+thermal neutrons	15.5±0.5	10.0±1.0
2)Pu^{239}+thermal neutrons	17.1±0.6	7.5±1.0
3)Pu^{238} (spontaneous)	17.3±0.4	5.5±1.0
4) Pu^{240}(spontaneous)	17.0±0.5	7.5±1.0

It is seen from Table II that our distribution parameters for thermal-neutron U^{235} fission are in good agreement with those generally accepted. It can also be noted that by comparison with U^{235} the maximum of the distribution for all the investigated plutonium nuclei is shifted toward greater energies but has a smaller half-width. Pearson's test showed complete incompatibility of distributions 2) and 3) with the Gaussian curve for uranium. The closeness of the parameters of curves 2), 3), and 4) indicates either that there is no difference between them or that the difference lies outside our experimental error. On this basis we combined the results of 2), 3), and 4) and represented the energy spectrum of α particles from plutonium fission by a single Gaussian curve having the parameters $E_{max} = 17.2$ MeV and $\Delta E = 7.5$ MeV.[1] The spectrum is shown in Fig. 2 together with the α-particle spectrum from uranium fission.

We believe that there is also a logical reason for representing the α-particle spectra as

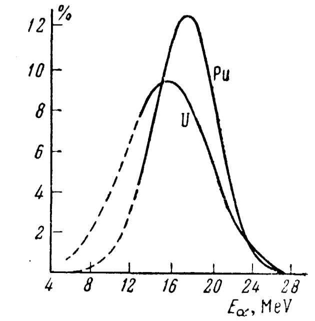

FIG. 2. Energy spectra of long-range α particles from the ternary fission of uranium and plutonium.

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[1] The appreciable x radiation from Pu^{238} resulted in a large spot background that probably impaired the registration efficiency for α particles having higher energies. The energy spectrum of Pu^{238} α particles may therefore be too narrow.

Gaussian or near-Gaussian distributions. The energy spectrum of binary-fission fragments is Gaussian because of the statistical character of scission of the fission neck.[10] In ternary fission a nucleus, before breaking apart, passes through the same deformation stages as in binary fission.[11,12] We can therefore expect a Gaussian distribution of ternary-fission fragment energies, as has been observed in the thermal-neutron fission of U^{235}.[11] It follows from the experiments[11,12] that the kinetic energies of fragments from binary and ternary fissions are related by $E_{\alpha} = E_b - E_t$, where E_b and E_t are the total kinetic energies of the fragments and E_{α} is the ternary-fission α-particle energy. From general statistical laws the distribution of the difference between two quantities obeying Gaussian distributions is also Gaussian in form. We therefore believe that the shape of the fission α-particle spectrum is determined by the purely statistical character of the instants of neck scission. The relation proposed by Dmitriev et al[11] between the dispersions of the energy curves is not absolutely required, because it was derived for statistically independent quantities and is thus not directly applicable to the kinetic energies of particles formed in ternary fission.

We still lack sufficient data to determine the causes of the shift of E_{max}; further work for this purpose is therefore highly desirable.

In conclusion we now return to the probability of spontaneous ternary fission. From the data in Table I, knowing the amount of fissioning matter and the exposure time, we computed the probability of spontaneous ternary Pu^{238} and Pu^{240} fission ($E_{\alpha} > 13$ MeV), obtaining 1:(395 ± 90) for Pu^{238} and 1:(410 ± 65) for Pu^{240}. In these calculations we used the half-lives $T_f = 4.9 \times 10^{10}$ years[13] and $T_f = 1.340 \times 10^{11}$ years[14] for Pu^{238} and Pu^{240} respectively. When the spectrum is extrapolated to the region $E_{\alpha} < 13$ MeV the probabilities become 1:(355 ± 80) for Pu^{238} and 1:(370 ± 60) for Pu^{240}. The errors included in these results are large; however, a further enlargement of the statistics would not essentially improve the accuracy, because the half-lives for spontaneous fissions have been determined very inaccurately. However, it must be noted that our results for the probability of spontaneous Pu^{240} fission agree very well with Nobles' data.[7]

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