RANGE AND ENERGY DISPERSION OF U²³³ FISSION FRAGMENTS

K. A. PETRZHAK, YU. G. PETROV, and É. A. SHLYAMIN

Radium Institute, Academy of Sciences, U.S.S.R.

Submitted to JETP editor January 28, 1960

J. Exptl. Theoret. Phys. (U.S.S.R.) 38, 1723-1728 (June, 1960)

The range distribution of U^{233} fission fragments (Sr⁹¹⁻⁹², Y⁹²⁻⁹³, Zr⁹⁷, Ba¹⁴⁰, and Ce¹⁴³) in various gases is studied. The dependence of the range dispersion on the nature of the gas is established. From the range dispersion the kinetic energy due exclusively to fission processes is determined. The results are compared with available experimental data and Fong's statistical theory.

INTRODUCTION

HROM the data on the distributions of the fragment energies and velocities, obtained with the aid of ionization chambers and by the time-of-flight method, it is only possible to determine the dispersion of the total kinetic energy for a given mass ratio of the fragments indirectly.

A direct determination of the kinetic energy dispersion of fragments of given mass and charge in the fission of U^{233} and U^{235} was carried out by Good and Wollan.¹ From the range distribution of fragments which emitted delayed neutrons the authors obtained the value $\Delta E/E = 5\%$ for I^{137} and 8%for Br^{87} . In reference 2 the energy distribution of Zr^{97} in the fission of U^{235} by thermal neutrons was obtained with the aid of a magnetic spectrometer. The half-width of the distribution was 11.4%.

In connection with the fact that no exhaustive data on the kinetic-energy dispersion of U^{233} fission fragments is available, it is of interest to determine its value for a series of fragments of different masses.

It is known that the measurement of the dispersion of fission-fragment ranges makes it possible to determine the kinetic-energy dispersion of the fragments with given M and Z. To determine the kinetic-energy dispersion of U^{233} fission fragments, we therefore used in this paper fission-fragment ranges in various gases obtained by collecting the fragments on thin films and subsequently analyzing these radiochemically.

EXPERIMENTAL METHOD

The apparatus (Fig. 1) consisted of an airtight aluminum container, in which a holder for rings with collodion films and a uranium target were placed. The films were inserted behind FIG. 1. Section of the container in which the irradiation was carried out. $1 - U^{233}$ target, 2 - holder for rings with films, 3 - aluminum airtight container, $R_0 -$ the distance between the target and the first film.



each other within a space of 2.5 mm: the distance $R_{\rm O}$ between the first film and the target was 136 mm. In all there were thirty films, each ~ 6 $\mu g/cm^2$ thick. The container was filled with various gases: hydrogen, helium, nitrogen, air, neon, and argon. The gas pressure was chosen such that the range of the heaviest fragment group should retain the same value in all the gases and that it should be within the first ten films. U²³³ targets of 76, 110, 145, 228, 284 $\mu g/cm^2$ with a spot diameter of 22 mm, obtained by plating platinum in an alcohol solution of uranyl nitrate, were employed in the experiments. The container was irradiated in the reactor of the U.S.S.R. Academy of Sciences during 1-2 hours at a constant temperature with a neutron flux of 10^{12} neutrons/cm²-sec. The activity of each film was then measured with a torsion β counter. In order to get rid of the activity



FIG. 2. Range distribution of Ba¹⁴⁰ nuclei in hydrogen. $O - experimental points, \bullet - Gaussian distribution.$

induced in the aluminum rings by the irradiation, the films were transferred into new rings. The two-hump curve of the activity distribution as the function of the film position obtained from these measurements made it possible to ascertain the reproducability of the experimental results, and to choose the necessary group of films for subsequent radiochemical analysis for this or that element. The chemical analysis was carried out by the usual methods,³ except for such simplifications as the preliminary separation of the fragments with the aid of films⁴ permits. The elements Sr, Y, Zr, Ba, and Ce were separated, and the identification of the isotopes by mass number was carried out by measuring their half-lives.

As a result of the radiochemical analysis of the films, range-distribution curves were obtained for the U^{233} fission fragments Sr^{91-92} , Y^{91-93} , Zr^{97} , Ba¹⁴⁰, and Ce¹⁴³ in hydrogen, helium, nitrogen, air, neon, and argon. From these data we determined the mean ranges and the range dispersion.

EXPERIMENTAL RESULTS

Figure 2 shows the experimental range-distribution curve of Ba¹⁴⁰ nuclei in hydrogen. The Ba¹⁴⁰ activity of different films, reduced to the solid angle of the first film relative to the target, is plotted in relative units along the ordinate axis; the fragment range in centimeters for a hydrogen pressure of 760 mm Hg and $t = 15^{\circ}C$ is plotted on the abscissa. For comparison, points correspond-

ing to a Gaussian distribution with a half-width equal to the experimental one are plotted in the figure. As is evident from the figure, good agreement between the experimental and Gaussian curve is observed. Such agreement was also obtained for different fragments in other gases.

These results differ from those of reference 5, where the differential curves are steeper on the far side from the source. The authors of reference 5 assume that the deviation of these curves from the Gaussian curves can be explained either by the nonuniform thickness of the uranium layer, or by the imperfect collimation of the fragments.

The observed range spread of fragments of a given mass can be due to a number of reasons: 1) the kinetic-energy fluctuation of the fragment, caused by the diversity of the nuclear deformation at the instant of fragment separation; 2) the change in the kinetic energy resulting from the nuclearcharge fluctuation during the formation of a fragment of given mass; 3) the statistical fluctuation of the number of collisions with electrons and nuclei during the slowing down of the fragments in the gas; 4) the change in the kinetic energy when a neutron is emitted by the fragment; 5) the slowing down in the target material; 6) the geometry of the apparatus.

To determine the effect of the target, we recorded the dependence of the magnitude of the range dispersion of Ba¹⁴⁰ in helium on the source thickness. Extrapolation of the curve to zero thickness made it possible to exclude the dispersion Δ S_{source} due to the slowing down of the fragments in the uranium layer. The contribution of the source to the dispersion for fragments of different mass was determined from the relation

$$[\Delta S_{\text{source}}]_{M} = [\Delta S_{\text{source}}]_{Ba} R_{Ba} / S_{M^{*}}$$
(1)

The correction for the geometry, ΔS_{geom} , due to the fact that the beam is not parallel, amounted according to calculations to 1.3% for the group of light fragments, and 1.6% for the group of heavy fragments. The magnitudes of the range dispersions with account of the effect of the source thick-

Gas	Sr 91-92		Y 92-93		Zr ⁹⁷		Ba140		Ce ¹⁴⁸	
	R. cm	S, %	R, cm	s, %	R, cm	S, %	R, cm	S, %	R, cm	S, %
Hydrogen Helium Nitrogen Air Neon Argon	10.05 15.75 2.58 2.54 4.80 2,60	7.377.099.518.049.8610.59	$ \begin{array}{r} 10.05 \\ 15.68 \\ 2.52 \\ 2.51 \\ 4.84 \\ 2.58 \\ \end{array} $	$6.66 \\ 6.84 \\ 9.41 \\ 7.61 \\ 8.69 \\ 9.88$	$9.61 \\ 15.61 \\ 2.50 \\ 2.44 \\ 4.66 \\ 2.49$	7.926.9910.278.209.609.38	7.5811.93 $-1.85-1.85$	6.13 7.03 9.87 11.38	7.68 12.02 1.86 1.84 	5.12 5.86 9.26 8,71

TABLE I. The ranges and the range dispersion of U^{233} fission fragments

ness and corrected for the geometry, i.e., the values

$$S = [(S_{exp} - \Delta S_{source})^2 - \Delta S_{geom}^2]^{1/2},$$

are listed in Table I (R is the mean fragment range in centimeters in the investigated gas for p = 760 mm Hg and $t = 15^{\circ}$ C, S is the relative range dispersion in percents).

The experimental range values listed in the table are corrected for absorption in the collodion films and in the target material.

After correcting for the source thickness and the geometry of the apparatus, the range dispersion can be represented by the sum:

$$S^2 = S^2_{\text{fiss}} + S^2_{\text{braking}}, \qquad (2)$$

where the first term is determined only by processes connected with the fission and does not depend on the subsequent interaction of the fragment with the material, while the second term is a function of the mass and charge of the stopping gas. This was indeed the basis for the division of these quantities.

The fragment-range spread of fragments passing through a substance is due to the statistical fluctuation of the number of collisions with electrons and nuclei. The relative spread due to electron collisions is 0.1%, which is negligibly small by comparison with the fraction of the spread due to nuclear collisions.

The dispersion due to nuclear collisions can be calculated from the formula:⁶

$$S_{\nu}^2 = \text{const} \left(R_{\nu}/R \right)^2 M_1 M_2 / (M_1 + M_2)^2,$$

where R_{ν} is the end part of the range where braking is exclusively due to nuclear collisions, R is the total fragment range, M_1 and M_2 are the nuclear masses of the fragment and of the stopping gas. The ratio R_{ν}/R was calculated after Bohr.⁷ It turned out that it is approximately equal for heavy and light fragments and varies within 10%, depending on the nature of the gas. Owing to this, by substituting (3) in (2), the latter can be written in the form

$$S^2 = S^2_{fiss} + \text{const} \cdot M_1 M_2 / (M_1 + M_2)^2.$$
 (4)

From this it is evident that S^2 is a linear function of the parameter $M_1M_2/(M_1 + M_2)^2$. The dependence of S^2 on $M_1M_2/(M_1 + M_2)^2$

The dependence of S² on $M_1M_2/(M_1 + M_2)^2$ was plotted from the experimental data for all investigated fragments. As an example, Fig. 3 shows the results for Sr⁹¹⁻⁹² and Ba¹⁴⁰. The intersection of the straight lines with the ordinate axis determines the value of S²_{fiss} for each fragment.



According to Bohr,⁶ the fragment range varies as \sqrt{E} . The range dispersion is, therefore, connected with the dispersion of the kinetic energy by the relation

$$2\Delta R/R = \Delta E/E.$$

Consequently, in Fig. 3 the intercepts with the ordinate axis characterize the kinetic-energy dispersion of fragments with a given mass.

To obtain the dispersion of the kinetic energy connected with the diversity of the deformations at the instant of fragment separation, it is necessary to exclude the dispersion caused by the recoil during the emission of a prompt neutron, and the fluctuation of the kinetic energy of the fragments due to the charge distribution during the fission into fragments of a given mass.

Assuming that the neutrons are emitted by the moving fragment isotropically in the c.m.s., we obtain a value of 3% for $(\Delta E/E)_n$ for the groups of heavy and light fragments. Account of the charge distribution during the formation of fragments of a given mass add to the dispersion a contribution $(\Delta E/E)_Z$ amounting to between 1 and 2% for all fragments. The final values of $(\Delta E/E)_0$, listed in Table II, are found from the relation

$$[\Delta E/E]^{2} = [\Delta E/E]_{0}^{2} + [\Delta E/E]_{n}^{2} + [\Delta E/E]_{Z}^{2}$$

From the equality of the fragment momenta at the instant of fission, it is possible to assume that the kinetic energy of the complementary fragment and the total kinetic energy of the two fragments will have the same relative dispersion, i.e.

$$[\Delta E/E]_l = [\Delta E/E]_h = [\Delta E/E]_t$$

for a given M_h/M_l ratio.

In Fig. 4 the dependence of $\Delta E_t (M_h/M_l)$ has been plotted from the data of Table II; the values of the total kinetic energy were taken from the work of

1246

Fragment	Sr#1-#2	Y 9 8- 93	Zr**	Ba140	Ce143
ΔΕ/Ε,%	12.9	12.4	13,2	11.6	9.4
[ΔΕ/Ε] ₀ ,%	12.3	11.8	12.8	11.2	8.7
ΔΕt,Mev	19.7	19,0	21,3	17,7	13,6
M _h /M _l	1.51	1,48	1,37	1,55	1,64

TABLE II. Dispersion of the kinetic energy of U²³³ fission fragments

Fraser and Milton.⁸ For comparison, we have given the dependence of the total kinetic-energy dispersion of the two particles on the mode of fission, obtained from the topological plots of that work. We note that these topological plots do not take into account the recoil due to prompt-neutron emission.



FIG. 4. The dependence of the total kinetic energy and of the dispersion of the kinetic energy on the mode of fission: 1 - total kinetic energy, 2 - dispersion of the total kineticenergy, calculated according to the contour diagrams of Fraser and Milton,⁸ $3 - \text{dispersion of the total kinetic energy accord$ $ing to the data of this paper.}$

DISCUSSION OF RESULTS

The precision of the range determination is 2%. The main part of this error is connected with the unequal thickness of the collodion films. The film thickness was determined by allowing a collodion drop of constant weight to spread over a surface of known area. Thus the equal thickness of the films depended on the uniform spreading out of the collodion and on the constancy of the weight of the drop.

The indeterminacy in fixing the end of the range, due to the sagging of the films during irradiation, was 0.5%. The fragment range was determined from a curve plotted on the average with ten points, each of which was the result of a separate radiochemical analysis. In this connection, the contribution of the radiochemical-analysis error to the precision estimates of the range determination did not exceed 0.5%. The small temperature variations during the irradiation did not affect the value of the mean range but caused an increase in the dispersion of the ranges. The error in the determination of the range dispersion with account of all the enumerated factors amounted to 10%.

The values of the kinetic-energy dispersion obtained in the present paper are in good agreement with the data of reference 2 and with the results obtained from the contour diagrams of reference 8. As is evident from Fig. 4, the calculated dependence of the total kinetic-energy dispersion on the mode of fission has a maximum for M_h/M_l = 1.3, that is in the region of the twice closed shell with Z = 50 and N = 82 for the heavy fragment. The obtained results do not confirm Fong's theory,¹⁰ according to which the dispersion of the total kinetic energy has a maximum for the most probable mode of fission, and is about 10 Mev.

In conclusion, the authors express their gratitude to E. B. Nikol'skaya for her help in carrying out the radiochamical analysis.

¹W. M. Good and E. O. Wollan, Phys. Rev. 101, 249 (1956).

²Cohen, Cohen, and Coley, Phys. Rev. 104, 1046 (1956).

³Nat. Nucl. Energy Ser. Div. IV, Plutonium Project Record 9, Radiochemical Studies of the Fission Products, Book 3, ed. by C. D. Corryell and N. Sugarman.

⁴ Petrzhak, Nikol'skaya, Petrov, and Shlyamin, Радиохимия, (Radiochemistry) 1, 227 (1959).

⁵Katcoff, Miskel, and Stanley, Phys. Rev. 74, 631 (1948).

⁶N. Bohr, Phys. Rev. 59, 270 (1941).

⁷N. Bohr, Kgl. Danske Videnskab. Selskab,

Mat.-fys. Medd. 18, No. 8 (1948).

⁸J. S. Fraser and J. C. D. Milton, Phys Rev. 93, 818 (1954).

⁹ Protopopov, Baranov, Selitskiĭ, and Eĭsmont, JETP 36, 1932 (1959), Soviet Phys. JETP 9, 1374 (1959).

¹⁰ P. Fong, Phys. Rev. 102, 434 (1956).

Translated by Z. Barnea 334