# *RADIOCHEMICAL STUDY OF THE PHOTOFISSION OF* $Th^{232}$

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Twelve types of fragments from the photofission of $Th^{232}$ at a peak bremsstrahlung energy of 10 Mev were identified by a radiochemical method. The dependence of the fission product yield on the mass number was determined. The dependence of the peak-to-valley ratio of the $Th^{232}$ and $U^{238}$ photofission yield curves on the peak bremsstrahlung energy was determined in the energy range from 8 to 14 Mev. Results of calculations by Fong's method relating to the photofission of thorium and uranium are presented.

THE photofission of heavy nuclei at low excitation energies provides several important items of information on nuclear properties. The items investigated are the photofission thresholds, the differential cross sections, the distribution of fission fragments in mass, energy and charge, the average number of neutrons per fission and, finally, the angular anisotropy in the distribution of fission fragments.

The overall picture of the mass distribution of fission fragments enables us to determine the most probable type of fission, the dependence of the symmetric fission on excitation energy, the mean number of neutrons per fission, and also the role played by the shell structure in the yield of the primary fission products and the role played by the collective oscillations of nuclear matter at low excitation energies. From published work it can be seen that the mass distribution of fission fragments depends on the structure of the nucleus undergoing fission and on the type and energy of the particles producing fission.

At the present time there are two papers devoted to the experimental study of the mass distribution of the products of photofission of $Th^{232}$. Hiller and Martin[1] identified thirteen fragments by a radiochemical method and used the data so obtained to plot the dependence of the yields on the mass number for the photofission of $Th^{232}$ at a bremsstrahlung peak energy of 69 Mev. In Avdonin and Petrzhak's paper[2] the determination of the mass distribution of fission products was not the main problem, and at a maximum $\gamma$-quantum energy of 12 Mev essentially only the position of the minimum has been determined for the photofission of $Th^{232}$.

However, photofission at low excitation energies has a number of advantages over other forms of fission. Only in the case of photofission is it possible to observe the fission of the nucleus under investigation, since the absorption of a $\gamma$ quantum does not change its charge and mass. Low excitation energy also eliminates the indefiniteness in the mass and in the charge of the nucleus undergoing fission, since at such energies the emission of nucleons before fission is energetically impossible. The study of the characteristics of photofission near the threshhold and below the threshhold permits us to elucidate the nature of spontaneous fission. For example, spontaneous fission may be regarded as a special case of photofission at a $\gamma$-quantum energy equal to zero. It therefore appeared to be of interest to study in greater detail the shape of the yield curve for the photofission of $Th^{232}$ at low excitation energies.

The literature already contains a number of papers[3-5] on the low energy photofission of $U^{238}$, To be able to compare the yield curves for thorium and uranium, the peak bremsstrahlung energy for the study of $Th^{232}$ was therefore taken to be 10 Mev. The work was carried out with the betatron of the Leningrad Technological Institute, with a maximum energy of 15 Mev. To obtain greater fission-fragment activities, the samples were irradiated inside the betatron accelerating chamber. The accelerated electrons were stopped directly in the substance of the sample undergoing irradiation, and this increased the betatron-radiation utilization factor. The fraction of fissions produced by electrons is assumed to be insignificant in view of the small interaction cross section for the electron and of the large surface density of the sample in the direction of the electron beam (of the order of 10 g/cm$^2$). The number of fissions induced by the neutrons produced in the sample itself and in the surrounding materials by fission or by the photonuclear effect can be estimated from the known neutron cross sections and geometrical conditions. In our case they were calculated to be less than 0.2% of the total effect for thorium and uranium. This question has been investigated experimentally by Richter and Coryell[4] and by Schmitt and Duffield.[6] Their results also lead to the conclusion that the number of fissions due to neutrons is less than 1%.

The sample for irradiation was prepared by pressing baked thorium oxide (in the case when uranium was irradiated it was prepared from uranium oxides). The weight of the sample was 0.5 to 1.0 gm. The time of irradiation was usually ~ 10 hours.

The energy of the betatron radiation was calibrated by making use of the known values for the thresholds of photonuclear reactions in a number of elements. In the course of the work the constancy of the calibration was checked by means of the threshold of the reaction $Cu^{63}$ (γ; n) $Cu^{62}$. The stability of the energy during irradiation was monitored by a voltage induced in a loop situated in the magnetic field of the betatron. The error in the determination of the peak bremsstrahlung energy is estimated to be ± 0.3 Mev. The constancy of the radiation intensity was monitored by means of an ionization chamber.

The irradiated sample was dissolved in acid and a certain amount of isotopic carriers was added to the solution so obtained. Usually from two to five fission fragments were separated out from a mixture of products after one irradiation. The methods described in reference 7 were taken as the basis for the chemical procedures for the separation and the radiochemical purification of fission products. Certain changes were introduced into them associated with the necessity of opening up the irradiated oxide samples, and of separating the fragments from thorium. At the end of the radiochemical operations the carrier was precipitated for quantitative analysis. The preparation for β counting was made by transferring this precipitate onto a disk of filter paper.

The β activity of the preparations was measured by an end counter for several half-lives. The decay curves for each element were analyzed and the fact that the resultant activity belonged to the isotope under investigation was established by means of the half-life.

Twelve different kinds of fragments were separated in the course of the work. Fission fragments with different mass numbers from 83 to 143 and having short half-lives were selected. Relative activities of the fission products were determined, i.e., one fission fragment was chosen as a standard and was separated in each experiment, and the intensity of the β radiation of the others was determined relative to the intensity for the standard one. $Ce^{143}$ was chosen as the standard. The relative activity for each kind of fission fragment was determined by taking the average of the results of from 2 to 6 irradiations.

To convert from the relative activity to the relative yield for a pair of fission fragments it is necessary to know the efficiency of the end counter for the β radiation from these fragments. To obtain the required conversion coefficients, uranium samples were irradiated under similar conditions. The same fission fragments were separated from the fission products of $U^{238}$, as in the case of thorium, and their relative activities were found. By utilizing data available in the literature[3,4] on the photofission of $U^{238}$ at 10 Mev, we determined the coefficients needed for the conversion from relative activity to relative yields. These coefficients were then used to obtain the relative yields for the photofission of $Th^{232}$.

Along with determining the yield curve for the photofission of $Th^{232}$ at 10 Mev, we also investigated, for both thorium and uranium, the dependence on the bremsstrahlung peak energy in the energy range from 8 to 14 Mev of the ratio of the most probable type of fission to symmetric fission. For this purpose three fragment elements (silver, cadmium, and cerium) were separated from samples irradiated at different energies. The relative yields of $Ag^{113}$, $Cd^{115}$ and $Cd^{117}$ determined the position of the minimum of the yield curve, while the yield of $Ce^{143}$ corresponded within experimental error to the maximum yield. Experiments on the determination of the peak-to-valley ratio carried out at the peak energy of 10 Mev demonstrated good reproducibility of the results. We found it therefore possible to determine this ratio at other energies by means of a single irradiation.

The results of determining yields for the photofission of $Th^{232}$ at 10 Mev are given in Table I. In this table column 3 gives the yields of the fission products relative to the yield of $Ce^{143}$. We assumed that the total yields of the fragments shown in the table are equal to the total yield of the chain of a given mass number. An examination of the independent yields of the subsequent members of the chain shows that this assumption holds with a high degree of accuracy.

In the fission process to each mass number indicated in the table there corresponds a com-

**TABLE I.** Yields of products of photofission of $Th^{232}$ at a maximum bremsstrahlung energy of 10 Mev

| Separated fragment | Half-life, hours | Yield relative to $Ce^{143}$ | | Yield in % normalized to 200% | |
|---|---|---|---|---|---|
| | | Value | % error | Value | % error |
| 1 | 2 | 3 | | 4 | |
| $Br^{83}$ | 2.4 | 0.187 | 25 | 1.8 | 0.44 |
| $Sr^{91}$ | 9.7 | 0.597 | 16 | 5.7 | 0.91 |
| $Zr^{97}$ | 17.0 | 0.244 | 22 | 2.3 | 0.51 |
| $Mo^{99}$ | 68.3 | 0.119 | 18 | 1.1 | 0.2 |
| $Ag^{113}$ | 5.3 | 0.007 | 24 | 0.066 | 0.016 |
| $Cd^{115}$ | 53 | 0.0034 | 26 | 0.032 | 0.008 |
| $Cd^{117}$ | 2.83 | 0.0039 | 30 | 0.037 | 0.011 |
| $Sb^{129}$ | 4.2 | 0.052 | 48 | 0.50 | 0.25 |
| $J^{133}$ | 20.5 | 0.45 | 40 | 4.3 | 1.7 |
| $Ba^{139}$ | 1.43 | 0.53 | 15 | 5.0 | 0.75 |
| $Ba^{140}$ | 306 | 0.808 | 21 | 7.7 | 1.5 |
| $Ce^{143}$ | 33.0 | 1.0 | — | 9.5 | — |

plementary "mirror-point" fragment, which has the same yield. To determine its mass it is necessary to know the mean number of neutrons emitted per fission. By making different assumptions with respect to the mean number of neutrons we found that the fundamental and the mirror points fit a smooth curve best when $\nu = 3$ (Fig. 1). By equating the area bounded by the smooth curve of relative yields to 200% we obtained the values for the absolute yields of the fission products under investigation. The absolute yields are given in column 4.

The large values of the errors in Table I is explained in the following manner. In view of the low activities of the products of low energy photofission of $Th^{232}$, the error in the relative activities obtained directly from experiment was about 6% in the neighborhood of the peaks and about 10 to 12% in the neighborhood of the minimum. The larger errors for $Sb^{129}$ and $I^{133}$ are due to the poor reproducibility of results in the former case and to the difficulty in analyzing the complex decay curve and to the extrapolation error in the latter case. Our method of calibrating the counting apparatus requires us to take into account the error in the determination of relative activities in the photofission of $U^{238}$ and the error in the relative yields taken from references 3 and 4. The error introduced through the normalization of the area bounded by the smooth yield curve to 200% is insignificant and was not taken into account in our work.

Figure 1 shows three curves of the dependence on the mass number of the fission product yields. The main characteristics of these curves are given in Table II.

From a comparison of the yield curves for the

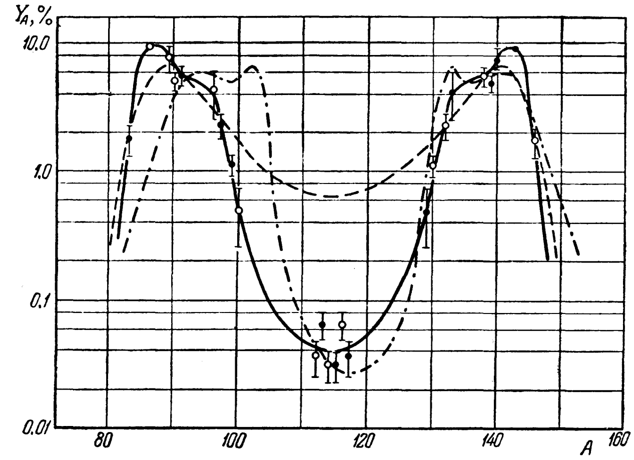


FIG. 1. The dependence of fission product yields on the mass number. Solid curve gives the results of the present work for the photofission of $Th^{232}$ at 10 Mev: ● are the basic points, o are the mirror points; the dotted curve corresponds to the photofission of $Th^{232}$ at 69 Mev;[1] the dot-dashed curve corresponds to the photofission of $U^{238}$ at 10 Mev.[3,4]

photofission of $Th^{232}$ and $U^{238}$ at 10 Mev it may be seen that the peak for the light fragments in the thorium curve is noticeably displaced towards lower mass numbers compared to the same peak in the case of uranium. At the same time the positions of the heavy fragment peaks almost coincide. Such a distribution of fission-product yields of thorium and of uranium agrees well with the mechanism of fission proposed by Hill[8] and determined by the nuclear shell effect and by the principal role played by the heavy fragment.

In the neighborhood of mass numbers 133 and 96 on the yield curve for the photofission of $Th^{232}$ higher yields are observed compared to the smooth decrease of the yields from the peak towards the minimum. In the heavy fragment peak this region coincides with the region of mass numbers where "fine structure" is observed in the photofission of $U^{238}$. This enables us to make the hypothesis that

**TABLE II.** Principal characteristics of the yield curves for the photofission of $Th^{232}$ and $U^{238}$

| Parameters of the yield curves | $Th^{232}$ ($\gamma_{10}$; f) present work | $Th^{232}$ ($\gamma_{69}$; f) from reference 1 | $U^{238}$ ($\gamma_{10}$; f) from references 3 and 4 |
|---|---|---|---|
| Position of the heavy fragment peak | 140 | 139 | 138 |
| Position of the light fragment peak | 89 | 90 | 97 |
| Maximum yield | 9.5% | 6.8% | 6.0% |
| Half-width of peak | 8 | 11 | 12–13 |
| Peak-to-valley ratio | 250 | 10 | 215 |

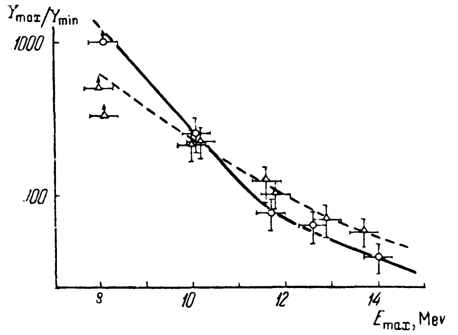


FIG. 2. Dependence of the peak-to-valley ratio on the maximum bremsstrahlung energy for photofission of: solid curve – $Th^{232}$, dotted curve – $U^{238}$.

the higher yields in the photofission of $Th^{232}$ are associated with the preferred formation among the primary fission products of nuclei with a closed shell ($N = 82$).

A comparison of the yield curves[1] for the photofission of $Th^{232}$ at 10 Mev and at 69 Mev shows that in this case the positions of the peaks almost coincide. However, in the former case the half width of the peak of the yield curve is less and the value of the maximum yield is greater than in the latter case. Thus the peak of the yield curve becomes wider and lower as the excitation energy of the nucleus undergoing fission is raised, while the proportion of symmetric fissions increases. The width of the peak also increases as the mass number of the nucleus undergoing fission is increased.

The peak-to-valley ratio is one of the basic characteristics of the dependence of the fission product yield on the mass number and is specially investigated in a number of papers.[3,5,9,10]. In our work this ratio is determined for the first time for the photofission of $Th^{232}$ in the region of low excitation energies and it is shown that the variation of this ratio in the range from 8 to 14 Mev for the peak bremsstrahlung spectrum energies is more pronounced for thorium than for uranium. Figure 2 shows the variation of this ratio with energy. The results of the present work are in good agreement with the number 80 obtained for the photofission of thorium at 12 Mev.[2]

In Fong's last paper[11] a new method was proposed for calculating the yield curve theoretically. We considered it to be of interest to utilize this method to calculate the yield curves theoretically for the photofission of thorium and of uranium. Unfortunately, final results have not been obtained due to the lack of precise values for the nuclear masses of $Th^{232}$ and $U^{238}$. As a result of these calculations we have obtained only the differences in the maximum excitation energies for a given type of fission and for symmetric fission [formula (46) of reference 11]. The dependence of this difference on the ratio of the masses of primary fission fragments enables us to show in general outline the shape of the yield curves and the position of the peaks on these curves.

The results obtained by us for the photofission of $Th^{232}$ and $U^{238}$ agree well with the experimental data both with respect to the shape of the curves and also with respect to the position of the maxima. Thus, the position of the maxima of the theoretically calculated curve for $Th^{232}$ is at mass numbers 89 and 140, while for $U^{238}$ the corresponding values are 97 and 138. The calculated curve for the photofission of $U^{238}$ has "fine structure" at mass number 132.

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