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FISSION CROSS SECTIONS OF ^{232}Th, ^{233}U, ^{235}U, ^{237}Np AND ^{238}U FOR 5–37 MeV NEUTRONS*†

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Abstract—Results are presented of fission cross section measurements on ^{232}Th, ^{233}U, ^{235}U, ^{237}Np and ^{238}U for neutrons in the energy range 5–37 MeV. The measurements were made using the 1·5 m cyclotron of the I. V. Kurchatov Order of Lenin Institute of Atomic Energy. The neutrons were produced by the D(d, n)^{3}He and T(d, n)^{4}He reactions and the time-of-flight method was used to effect the neutron energy selection. Gas scintillation fission counters were employed as detectors.

IN TWO earlier papers[1,2] results were presented on some measurements of the fission cross sections of ^{232}Th, ^{233}U, ^{235}U, ^{237}Np, ^{238}U and ^{239}Pu for neutrons in the energy ranges 3–10 and 10–22 MeV. The first of these papers describes measurements carried out using conventional fission chambers of the ionization type. The second describes work in which gas scintillation fission counters were employed in conjunction with a fast neutron time-of-flight spectrometer.

The work reported here presents the results of fission cross-section measurements on ^{232}Th and ^{238}U with 22–37 MeV neutrons, on ^{235}U and ^{237}Np with neutrons in the energy range 22–27 MeV and on ^{233}U with neutrons in the energy range 10–22 MeV. In addition, results of some additional measurements in the energy range 5–10·5 MeV are presented, these measurements being made with the aim of improving the accuracy of the cross-section curve in this region. We also give the results of some measurements of absolute cross sections in the energy range 10–22 MeV, a region in which only relative curves normalized to published values (mainly 14 MeV neutron data) have so far been obtained.

The neutrons were produced by the reactions D(d, n)^{3}He and T(d, n)^{4}He. The measurements were carried out on the 1·5 m cyclotron of the I. V. Kurchatov Order of Lenin Institute of Atomic Energy. The accelerator was operated in two regimes: using deuterons with an energy $E_d = 9{\cdot}9$ MeV, measurements were made over the energy ranges 22–27 MeV [T(d, n)^{4}He reaction] and 5–10·5 MeV (D(d, n)^{3}He reaction); using deuterons with an energy $E_d = 19{\cdot}5$ MeV, measurements were made in the energy ranges 37–27 MeV (T(d, n)^{4}He reaction) and 10–22 MeV (D(d, n)^{3}He reaction).

Within these ranges the neutron energies were varied by slowing down the deuterons with calibrated platinum foils placed in front of the target. All the measurements were made at an angle of 0° to the direction of the deuteron beam. A tritium–zirconium target was used for the measurements with d–T neutrons and a gaseous deuterium target for the measurements with d–d neutrons.

Over most of the energy range covered by these experiments, in addition to the neutrons from the reactions D(d, n)^{3}He and T(d, n)^{4}He, a large number of neutrons

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were produced also by the reactions D(d, pn), T(d, pn) and by other reactions. For the purpose of separating the monokinetic neutrons, a multichannel fast neutron time-of-flight spectrometer was used.[3] The resolving time of this spectrometer was 3 nsec. To separate out satisfactorily the effects caused by these monokinetic neutrons with this degree of resolution, a fast neutron fission detector possessing a comparatively high efficiency was needed so that measurements could be made over flight paths of 2·5–3 m. The conventional kinds of ionization fission chambers and proportional counters are unsuitable here because the rise time of the pulses which they produce is greater than 10^{-7} sec. The use of solid scintillators must also be excluded because, for these, the specific light yield falls off as the ionization density of the particles being counted rises. This effect makes it virtually impossible to discriminate between fission fragments and α-particles.

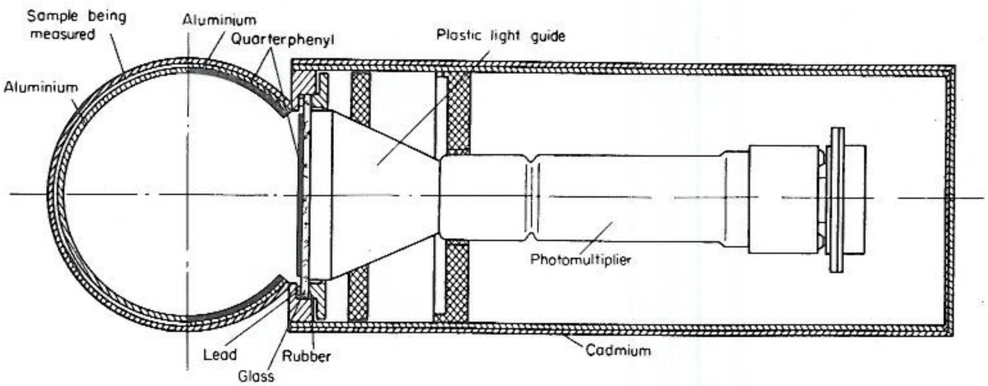

FIG. 1.—Gas scintillation fission counter.

In our experiments we used gas scintillation counters as fission detectors. A schematic diagram of one of these counters is shown in Fig. 1. It is a spherical chamber, dia. 10 cm, with a circular glass window joined by a light guide to a photomultiplier. Onto the internal surface of the hemisphere opposite the window is deposited by vacuum deposition a layer of fissile material with a thickness of 1–1·5 mg/cm² and having a total weight of 150–180 mg. Onto this layer is deposited, also by vacuum deposition a thin layer of aluminium with a thickness of 20 μg/cm². An aluminium layer with a thickness of several mg/cm² is deposited all over the remaining surface of the sphere. On top of the aluminium layer on the inside surface of the chamber (right-hand hemisphere in Fig. 1), is deposited a layer of 4-terphenyl with a thickness of 30 μg/cm². 4-terphenyl is deposited also on the inside surface of the window ($\sim$10 μg/cm²). The counter chamber was filled with xenon to a pressure a little above 1 atm and the scintillations produced within it were detected by a type FEU-33 photomultiplier tube operating in the usual time regime. However, because of the heavy α-particle loading, the voltage on some of the lower dinodes of this tube was applied through a low resistance divider.

Figure 2 shows the differential pulse height spectrum due to the fission fragments from ^{232}Th. These measurements were made with about 150 mg of thorium in the counter. Figures 3 and 4 show the time spectra of the pulses due to fission fragments from ^{233}U and ^{237}Np. The narrow peaks are caused by the monokinetic groups of neutrons from the reactions d–T or d–d. The broad continuous distribution

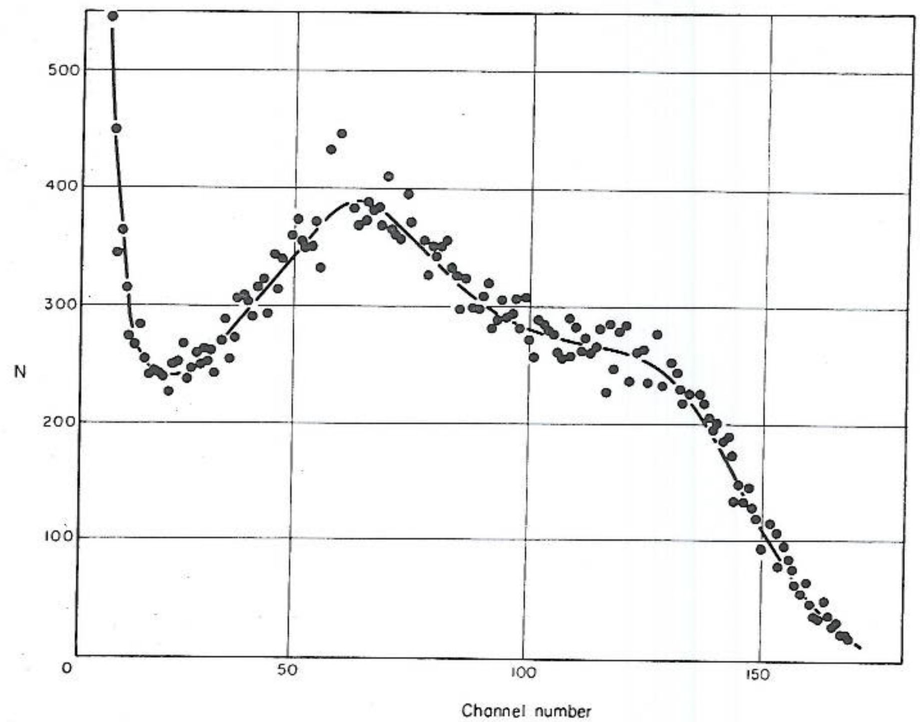

FIG. 2.—Pulse height spectrum due to ^{232}Th fission fragments.

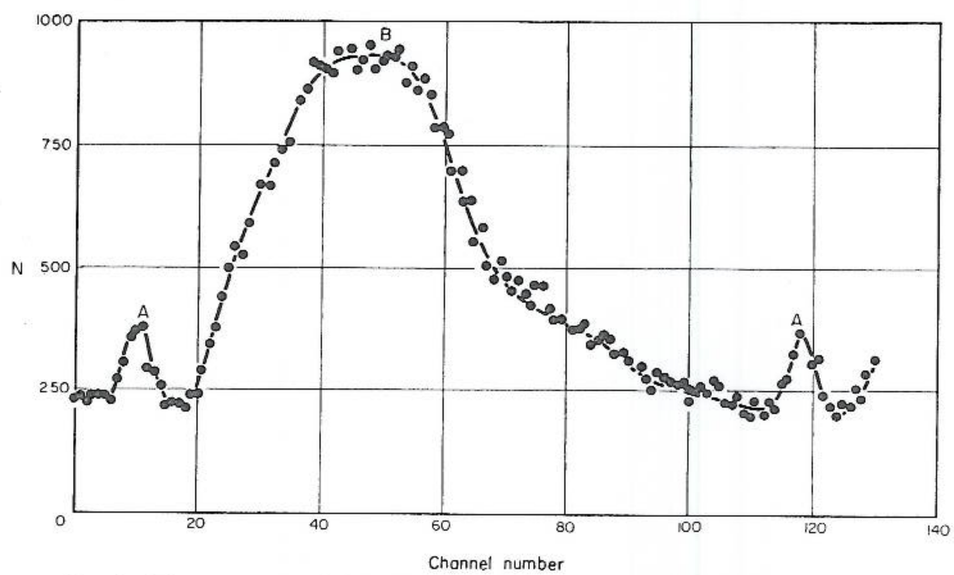

FIG. 3.—Time spectrum of fission fragment pulses due to fissions in ^{233}U induced by neutrons from the reaction D(d, n)^{3}He. Deuteron energy 19·5 MeV; flight path 2·7 m; *A*—fission induced by neutrons from the reaction D(d, n)^{3}He; *B*—fission due to neutrons from deuteron break-up.

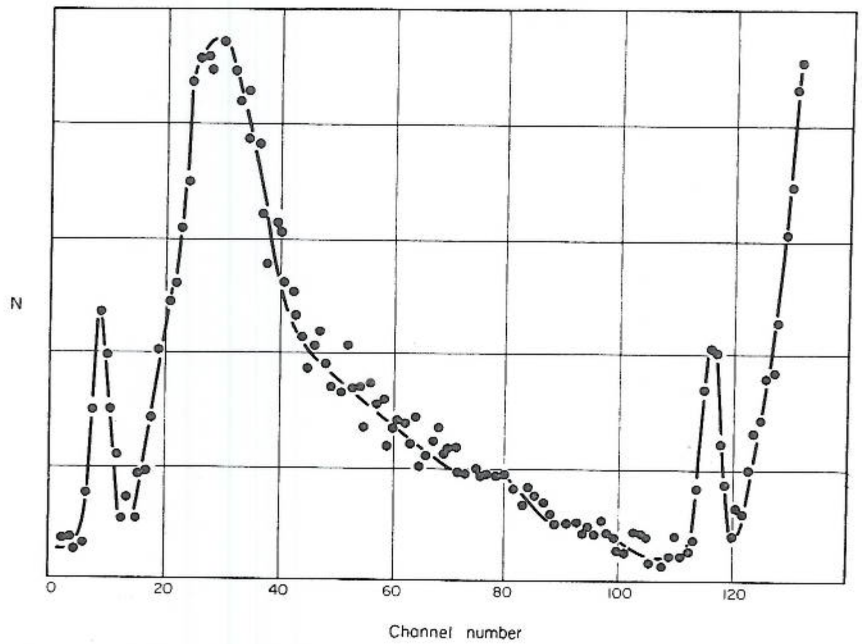

FIG. 4.—Time spectrum of fission fragment pulses due to fissions induced in ^{237}Np by neutrons from the reaction T(d, n)^4He. Deuteron energy 9·9 MeV; flight path 1·5 m.

corresponds to fissions induced by neutrons arising from the break-up of the deuteron; the pedestal beneath this peak is caused by scattered and slowed-down neutrons. Similar distributions were obtained for all the isotopes studied for the energy ranges indicated above. The area of the peak corresponds to the total number of fissions induced in the layer by neutrons belonging to the mono-energetic group.

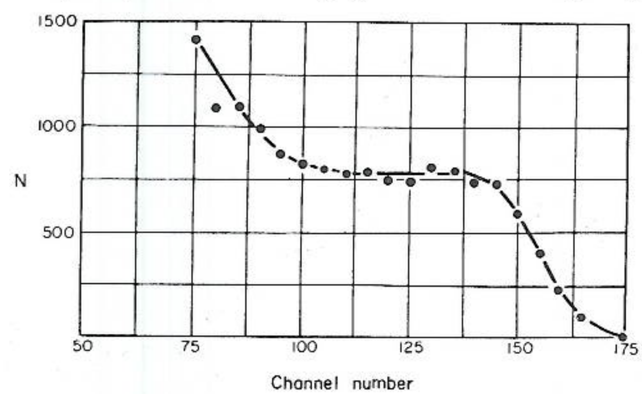

FIG. 5.—Pulse height spectrum of recoils in stilbene crystal.

The intensity of the flux of neutrons produced in the D(d, n)^3He and T(d, n)^4He reactions at various deuteron energies was measured by a stilbene scintillation counter connected into the time-of-flight spectrometer circuit. The pulse height spectrum of proton recoils in the crystal was recorded at the same time as the neutron time spectrum. With the help of a supplementary single-channel time analyser, which gated the pulse height analyser, only those events were selected which were associated with the counting of monokinetic neutrons from the d–T or d–d reactions. As an example, Fig. 5 presents the pulse height spectrum of recoils in a stilbene crystal taken at a neutron energy $E_n = 36{\cdot}5$ MeV. The plateau on this graph corresponds to the recoil protons; the sharp rise in the low energy part of the spectrum is due to pulses from carbon recoils and from the products of other reactions. From these data the neutron flux was determined using a procedure similar to that described by RIBAKOV and SIDOROV[3].

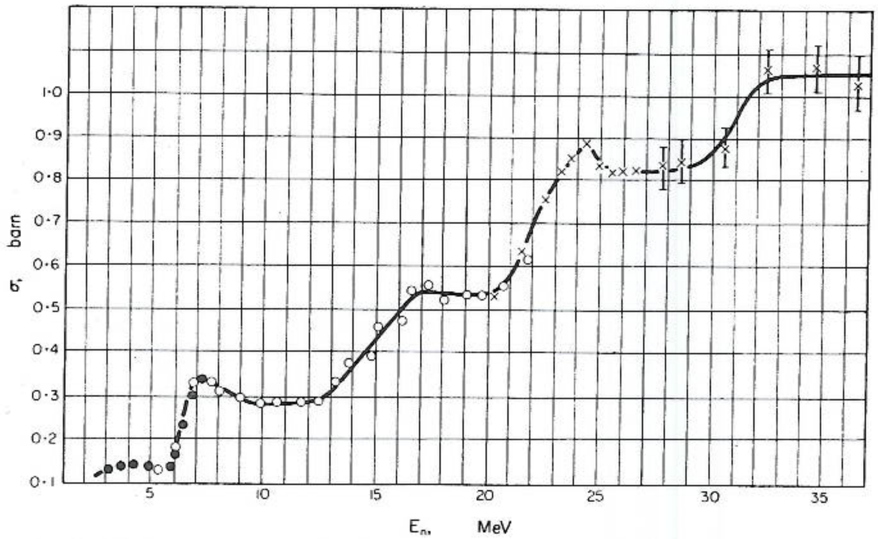

FIG. 6.—Fission cross section of ^{232}Th for neutrons with energies 3–37 MeV: ●—T(p, n)^{3}He neutrons[1]; ○—D(d, n)^{3}He neutrons; ×—T(d, n)^{4}He neutrons.

To determine the absolute values of the cross sections, the layers in the counters were calibrated using 3·4 MeV monokinetic neutrons from the T(p, n)^{3}He reaction for which the fission cross sections of all the isotopes studied in this work are well known. All these calibration measurements were carried out under exactly the same conditions as the main measurements. The 3·4 MeV fission cross sections assumed for the purpose of these calibrations were ^{232}Th(0·135 barn[4]); ^{235}U(1·23 barn[2]); ^{237}Np(1·62 barn[2]) and ^{238}U(0·55 barn[2]). The chamber containing ^{233}U was not calibrated; the results were tied in with the published data for 10 MeV neutrons[5].

Figures 6–10 present the results of the fission cross-section measurements. The overall accuracy of the measurements in the range 5–27 MeV was not less than 5 per cent. In the range 27–37 MeV, the accuracy of the measurements was not worse than 10 per cent. This increase in the error is associated with increased backgrounds and with the fact that there was some uncertainty in the background measuring procedure. The energy resolution was determined mainly by the target thickness; for the various neutron energy ranges it was: 5–10·5 MeV, from 500 to 300 keV; 10–22 MeV, from 700 to 250 keV; 22–37 MeV, from 1 MeV to 300 keV. In Figs. 6–10, and in Fig. 11 for ^{239}Pu, the results in the range 3–8·5 MeV published earlier[1] are also included. Corrections have been applied to these earlier curves to allow for the improved value for the efficiency of the long counter which was used to measure the neutron flux.

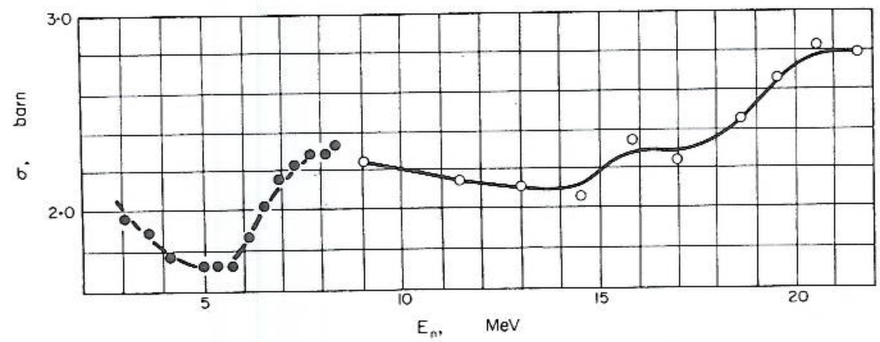

FIG. 7.—Fission cross section of ^{233}U for neutrons with energies 3–22 MeV: ●—T(p, n)^{3}He neutrons; ○—D(d, n)^{3}He neutrons.

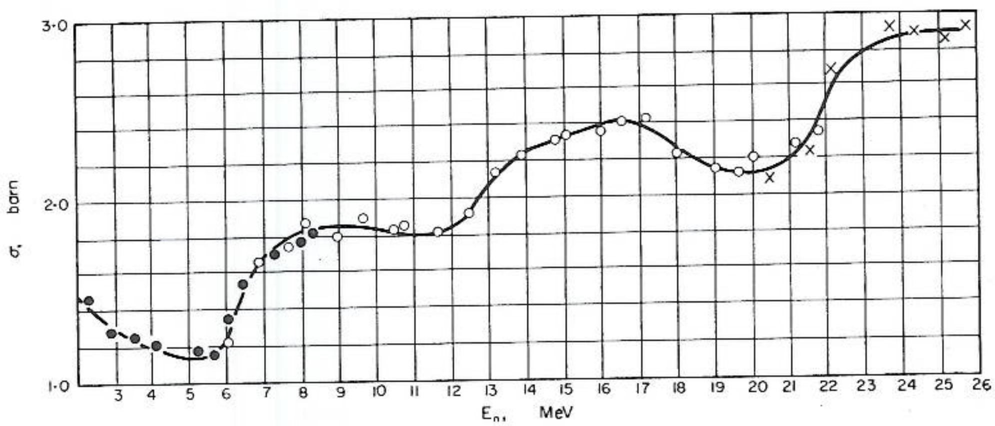

FIG. 8.—Fission cross section of ^{235}U for neutrons with energies 3–27 MeV: ●—T(p, n)^{3}He neutrons[1]; ○—D(d, n)^{3}He neutrons; ×—T(d, n)^{4}He neutrons.

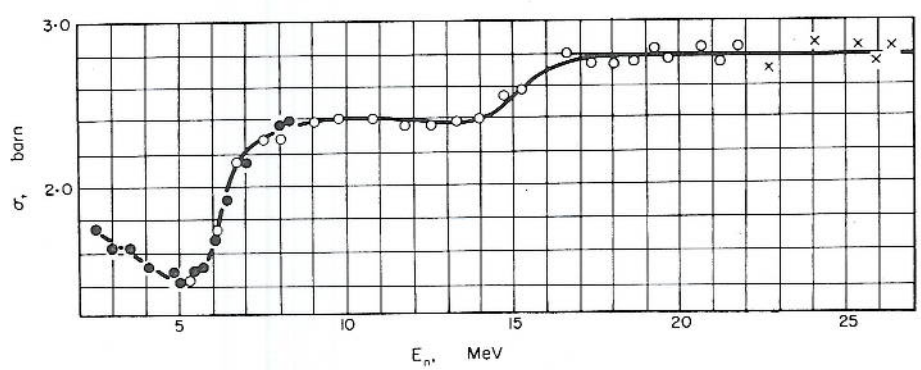

FIG. 9.—Fission cross section of ^{237}Np for neutrons with energies 2·5–27 MeV: ●—T(p, n)^{3}He neutrons[1]; ○—D(d, n)^{3}He neutrons; ×—T(d, n)^{4}He neutrons.

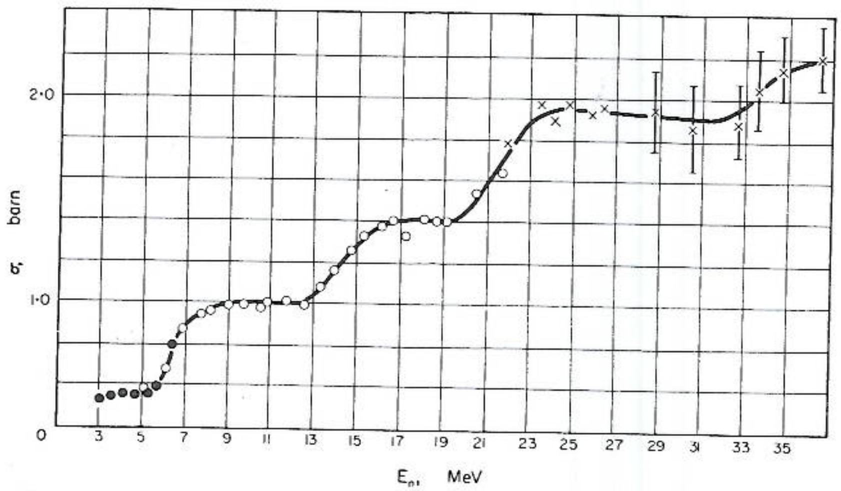

FIG. 10.—Fission cross section of ^{238}U for neutrons with energies 3–37 MeV: ●—T(p, n)^{3}He neutrons[1]; ○—D(d, n)^{3}He neutrons; ×—T(d, n)^{4}He neutrons.

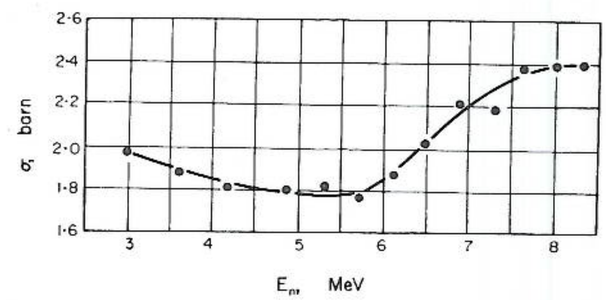

FIG. 11.—Fission cross section of ^{239}Pu for neutrons with energies 3–8·5 MeV.

A general feature common to all the isotopes in the energy region covered in this work is the fact that as the energy increases the fission cross section rises in sudden steps. Superimposed on this overall background, irregularities can be seen characterized by a comparatively sharp rise in cross section followed by a slow fall. This sort of irregularity was noticed first in the cross section of ^{232}Th at a neutron energy of 7–8 MeV. We observed similar cross-section 'overswings' for ^{235}U at 15–17 MeV, for ^{232}Th and it seems also for ^{238}U at a neutron energy of around 25 MeV. The overswings are observed whenever a step is due to the contribution from the fission of an even-even isotope, these having large neutron binding energies and depressed fission thresholds. The overswing is due to the fact that at the beginning of the step the emission of the next neutron is energetically forbidden.

The fission cross-section curve for ^{233}U in the energy region 12–17 MeV is very interesting. The rise in the cross section due to the contribution of ^{232}U is not very pronounced here. This is particularly surprising because the fission threshold for ^{232}U is more than 2 MeV lower than the neutron binding energy; it indicates either that the fission probability of ^{232}U is low, which is completely at variance with the generally accepted views on the dependence of fission probabilities on, for example, the parameter Z^2/A, or it indicates a rapid decline in the fission probability of ^{233}U with energy.

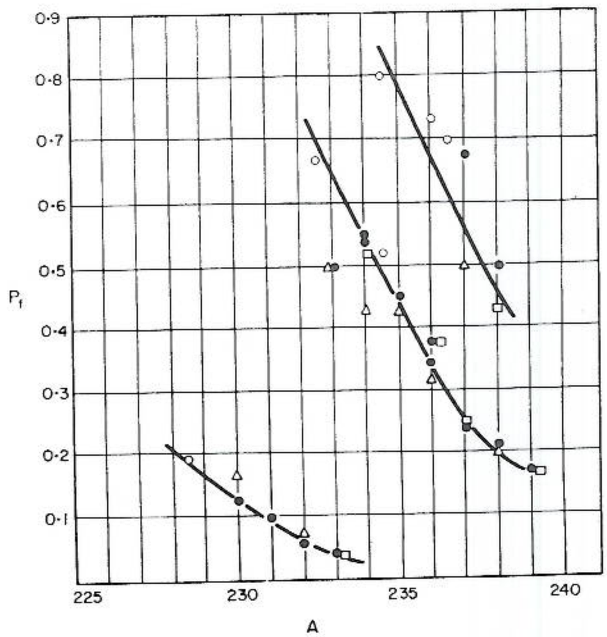

FIG. 12.—Fission probabilities for isotopes of thorium, uranium and neptunium: ● from the results of this work; □—for 3 MeV neutrons; △—photofission and photoneutrons; ○—spallation reactions.

The steps in the cross-section curves are well pronounced throughout the whole energy region studied in this work. One can therefore find the contribution to the cross section of any particular isotope to an accuracy of about 10 per cent and so determine the fission probability for that isotope. Fission-probability data are given in Table 1 and in Fig. 12, which includes also fission probabilities taken from the results of other workers.[6,7]

TABLE 1.—FISSION PROBABILITIES FOR ISOTOPES OF THORIUM, URANIUM, NEPTUNIUM AND PLUTONIUM

Isotope	E_n (MeV)	From our measurements	From other measurements
^{233}Th	3	—	0·04[6]
^{232}Th	10	0·060	—
^{231}Th	17	0·10	0·12[7]
^{230}Th	26	0·13	—
^{229}Th	34	>0·16	—
^{239}U	3	0·17	0·17[6]
^{238}U	10	0·21	—
^{237}U	17	0·24	0·25[6]
^{236}U	26	0·37	—
^{235}U	34	~0·40	—
^{236}U	3	0·39	0·38[6]
^{235}U	10	0·45	—
^{234}U	17	0·55	0·52[6]
^{234}U	3	0·55	0·52[6]
^{233}U	10	0·50	—
^{238}Np	3	0·50	0·43[6]
^{237}Np	10	0·68	—
^{240}Pu	3	0·59	0·59[6]
^{239}Pu	10	0·58	—

In working out these fission probabilities, the following values were taken for the cross sections for the formation of the compound nucleus and were applied to all the isotopes studied

E_n, MeV	3	10	18	25	35
σ_c, barn	3·3	2·9	2·8	2·8	$<$2·8

It should be pointed out that all the fission probabilities given in Table 1 are for intermediate nucleus excitation energies ~8–10 MeV. However, the close proximity of these values to the fission probabilities obtained from the measurements with 3 MeV neutrons is not trivial since, at neutron energies of the order of 20 MeV, the neutrons are captured by the nuclei with orbital angular moments of the order of 10.

Comparing the measured cross sections with the values calculated by adding up the fission probabilities, it is evident that the probability of fission is constant over the range of energies studied in this work (except, as mentioned in the above discussion, ^{233}U).

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