FISSION OF Th^{232} *INDUCED BY THERMAL NEUTRONS*

E. I. KORNEEV, V. S. SKOBKIN, and G. N. FLEROV

Submitted to JETP editor February 9, 1959

J. Exptl. Theoret. Phys. (U.S.S.R.) **37**, 41-45 (July, 1959)

It is shown that the fission effect which occurs when thorium is irradiated with slow neutrons is in fact due to the fission of Th^{232} by thermal neutrons. The fission cross section is (0.06 ± 0.02) mbn. The results obtained are compared with available experimental data on the fission of even-even nuclei by thermal neutrons.

THE study of the fission of heavy nuclei from energy states lying below the fission barrier has made a considerable contribution both to the understanding of the fission process itself and also to the improvement in our understanding of the structure of atomic nuclei. At the present time investigations with a sufficiently great degree of detail have been carried out only with respect to fission from the ground state or spontaneous fission. A large number of experimental and theoretical papers have been published on the subject, and the most important regularities of spontaneous fission have consequently been established.

In addition to spontaneous fission (fission from states lying 5 or 6 Mev below the barrier) it is also of considerable interest to study fission at excitation energies near the barrier (approximately 1 Mev or less below the activation energy); such energy states are usually produced by the capture of slow neutrons by the so called "nonfissionable" nuclei. In such nuclei the energy of the added neutron is lower than the activation energy, i.e., the energy of excitation for which the lifetime with respect to fission is equal to the lifetime with respect to γ emission. An experimental determination of the probability of nuclear fission at such excitation energies will apparently allow us to obtain interesting information with respect to the shape of the fission barrier.

Some experimental results[1-3] in this direction have been obtained only comparatively recently. This is due to the specific difficulties of the experiment associated with the small values of the cross section. Principally, the results refer to thermal neutron fission; in the majority of cases only upper limits on the cross sections have been obtained. The value $2 \times 10^{-28} cm^2$ was obtained for the upper limit on the thermal neutron fission cross section of Th^{232}.

The first indication that in all likelihood the observed phenomenon is fission of thorium by thermal neutrons is contained in reference 4. The object of the present work was to obtain more definite data on the fission of Th^{232} by thermal neutrons. Since the fission cross section of Th^{232} was expected to be very small (in any case $< 2 \times 10^{-28} cm^2$), we have paid particular attention in our experiments to the elimination of possible background fission effects. This background may be due, for example, to the presence in thorium of fissionable impurities, to fission by fast neutrons and by γ-quanta with energies above the fission threshold (if such are emitted by the neutron source), or possibly to the hard γ radiation arising in the capture of thermal neutrons. Even preliminary experiments showed that the thermal column of a reactor could not be used as a source of thermal neutrons, because of the considerable intensity of fast neutrons and of hard γ radiation. In this respect, a photo neutron source is free from the above deficiencies. In our experiments we have used a photo neutron source (Sb^{124} + Be). The main part of the neutrons from such a source has an energy of ≈ 24 kev; approximately 7% of the intensity is due to neutrons of energy near 400 kev.

Evidently the only fissionable impurity (under the action of thermal neutrons) in thorium may be natural uranium, or more exactly the isotope U^{235}. Uranium concentrations of $\sim 10^{-6}$ are already dangerous. The thorium was purified by sorption of the uranium in an ion-exchange column (the method is described in detail in reference 4). The concentration of uranium in the thorium we used amounted to less than 10^{-7}.

A diagram of the experiment is shown in Fig. 1. The neutron source consisted of a beryllium cylinder of 90-mm diameter and 80-mm length; along the axis of the cylinder there was an opening into which the γ-emitter (Sb^{124}) was inserted in the form of a sphere of 19-mm diameter. The activity of the γ emitter amounted to ≈ 6 Curies. The intensity of the photo-neutron source was 10^8 sec^{-1}.

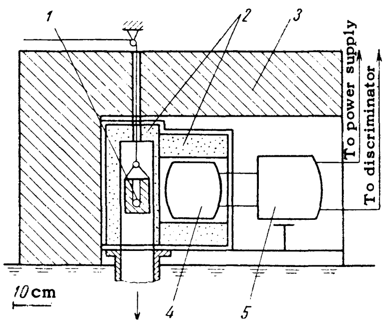

FIG. 1. Schematic experimental arrangement. 1) source, 2) paraffin, 3) shield (lead), 4) chamber, 5) amplifier.

The source was lifted by remote control from its container and placed into a paraffin cavity. The chamber was surrounded by a paraffin layer of 6-cm thickness. The paraffin blocks were surrounded on the outside by a lead screen 25 — 15 cm thick.

We used a multilayered ionization chamber (18-cm diameter, 15-cm height) to record the fission. Thorium in the form of the oxide ThO_2 was deposited on aluminum plates of total area $\approx 2300\ cm^2$. The total amount of actively utilized material was 2.5 g. The chamber was filled with industrially pure argon at a pressure of 1 atmos. Pulses from the chamber were applied to an amplifier and from there to a recording device with an amplitude discriminator. The amplitude of pulses from the fission fragments at the output of the amplifier amounted to several tens of volts.

To estimate the flux of thermal neutrons, we made use of a similar chamber containing 2.4 mg of natural uranium.

Upon irradiation of the thorium by thermal neutrons, a definite fission effect was observed (see table). To estimate the thermal-neutron fission we initially utilized a cadmium absorber (0.4-mm thick). The counting rate when the chamber was surrounded by cadmium amounted to approximately one half the counting rate without the absorber. Fission in the presence of the cadmium absorber could be ascribed both to neutrons of energy above the absorption limit in cadmium, and possibly also to the hard capture γ radiation from cadmium. Subsequent experiments showed the fission observed in the case of a cadmium absorber due principally to the γ radiation produced in the cadmium. The chamber was surrounded by a thick ($\approx 0.6\ g/cm^2$) boron absorber, and by more cadmium above that. The boron layer effectively absorbed neutrons of a wider energy spectrum than cadmium. The counting rate was reduced by a factor of ~ 20 when the chamber containing the thorium was surrounded by boron. However, fission in the case of two absorbers (cadmium outside and boron inside) corresponded approximately to that in the case of cadmium alone. Therefore, the cadmium absorber (owing to the production of hard γ-radiation accompanying neutron capture) cannot be utilized to separate out the effect due to thermal neutrons. In subsequent work, a boron absorber was used for this purpose.

Experimental conditions	Absorber	Fission effect	
		Chamber with thorium (counts/hr)	Chamber with uranium (counts/hr)
Source and chamber in paraffin (chamber with copper walls)	–	122 ± 5	99 ± 3
	Cd	60 ± 5	5 ± 0.3
	B (600 mg/cm²)	6 ± 1	2 ± 0.2
	Cd + B (600 mg/cm²)	60 ± 8	–
Source and chamber in paraffin surrounded by boron – 100 mg/cm² (aluminum chamber)	–	123 ± 5	98 ± 3
	B (600 mg/cm²)	27 ± 2	5 ± 0.2
Source and chamber in paraffin, additional piece of Be inside paraffin cavity.	–	121 ± 5	101 ± 3
Without paraffin.	B (600 mg/cm²)	15 ± 3	

The essential role played by photofission in the experiments involving cadmium required the performance of a number of control experiments to estimate the photofission due to the capture γ radiation produced in the materials of the chamber and the shielding. For this purpose we compared thorium fission in two chambers containing the same amount of thorium: a heavy one (copper walls, thick aluminum plates), and a light one (thin aluminum walls, thin plates). The yield of hard capture γ radiation with $E_\gamma > 5.4$ Mev in the case of the light chamber ought to be approximately 20 times lower than for the heavy one. A comparison showed that fission due to the capture γ radiation arising in the materials of the chamber was insignificant. Further, it was shown that γ rays arising in the lead shield due to neutron capture also played an insignificant role. The observed fission effect did not change when the paraffin was surrounded on the outside by a layer of boron ($100\ mg/cm^2 B_4C$). The results of these experiments are also presented in the table.

We have also estimated the possibility of the fission of thorium by fast neutrons, arising in beryllium through capture γ radiation (the cross section for the fission of Th^{232} by fast neutrons is $\approx 10^{-25} cm^2$). However, the contribution due to fast neutron fission is apparently very small. When an additional piece of beryllium of approximately the same size as was used in the photoneutron source was introduced into the paraffin cavity, no increase in the number of fissions was observed.

Thus, the foregoing control experiments show that fission due to various side effects makes a very insignificant contribution to the observed effect. The negligibly small content of uranium in thorium ($< 10^{-7}$) excludes the possible explanation of the effect by means of uranium fission (U^{235}). Therefore, the only acceptable explanation is apparently the fission of Th^{232} by thermal neutrons. To estimate the fission of thorium by thermal neutrons, we made use of a thin (100 $mg/cm^2 B_4C$) boron absorber in the form of a layer of boron carbide held together by zapon lacquer. The counting rate of a standard (uranium) chamber with such an absorber was reduced by a factor of 20. As can be seen from the table, the effect due to the thermal neutron fission of thorium amounted to 96 fissions/hour.

Thus, it may be considered established that thorium (Th^{232}) undergoes fission by thermal neutrons. The fission cross section in this case amounts to (0.06 + 0.02) mbn.

The results obtained by us (see Fig. 2) on the fission of Th^{232} may be compared with the available experimental data on the fission of even-even nuclei by thermal neutrons. Figure 2 shows the ratio of the fission cross section to the cross section for the formation of the compound nucleus σ_f/σ_c which is proportional to the probability of fission as a function of the difference $B_n - E_a$, where B_n is the neutron binding energy and E_a is the activation energy; σ_c for thermal neutrons is close to the value for the radiation capture cross section. The calculated threshold values are taken from the work of Vandenbosch and Seaborg.[5] The available experimental data are far from complete (in fact, only upper limits on fission cross sections are available); nevertheless it is apparently possible to speak of the existence of a definite dependence of the probability of fission on excitation energy, namely that the probability of fission decreases exponentially with an increase in $B_n - E_a$. This agrees with the expression provided by theory for the quantum mechanical barrier penetration factor $G \sim \exp\{-\Delta E/\epsilon\}$.[6] For purposes of comparison we have drawn an exponential curve through the point we obtained for fission of Th^{232}, taking the value of the coefficient ϵ in the exponent from theory ($\epsilon = 100$ kev).

Interesting results were recently obtained by Lamphere,[7] who measured the excitation curve for the fission of U^{238} (solid curve in Fig. 2) upon irradiation with neutrons of energy below threshold down to 400 kev. Although these results, strictly speaking, are not completely comparable with the data on the fission of even-even nuclei by thermal neutrons (we here are dealing with the fission of one nucleus at different excitation energies), generally speaking they do not contradict the regularity noted above. The excitation curve has exponential segments with the slope corresponding to $\epsilon \approx 100$ kev. However, the over-all picture is complicated by the presence of horizontal segments which appear near levels corresponding to inelastic scattering.

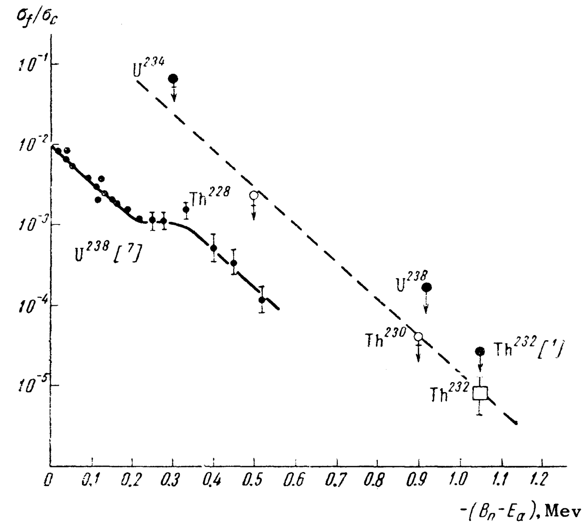

FIG. 2. Thermal neutron fission of even-even nuclei. □ – result of present work; ● – threshold determined experimentally, ○ – threshold calculated; arrow indicates that the value is known to be too high ($\sigma_f <$), crossed arrow indicates upper limit ($\sigma_f \leq$).

The probability of fission is apparently determined not so much by the excitation energy as by the difference between the excitation energy and the activation energy, irrespective of whether only the excitation energy varies, as in the case of a single nucleus, or whether E_a also varies for different nuclei. Thus, it would be interesting to accumulate further experimental data on the fission of even-even nuclei by thermal neutrons. There is also no doubt that the difficulties of carrying out an experiment on the fission by thermal neutrons are considerably less than in the case of fission by neutrons of other energies. In particular, it is of interest to determine the cross sections for thermal neutron fission of the uranium isotopes U^{234} and U^{236} for which fission thresholds have been obtained experimentally.

[1] Neutron Cross Sections, US AEC, 1955.
[2] R. L. Macklin and J. H. Lykins, J. Chem. Phys. **19**, 844 (1951).
[3] Ghiorso, Osborne, and Magnusson, The Transuranium Elements, New York, **14B**, 22 – 26, 1949.
[4] Flerov, Klochkov, Skobkin, and Terent'ev, Dokl. Akad. Nauk SSSR **118**, 69 (1958), Soviet Phys. "Doklady" **3**, 79 (1958).
[5] R. Vandenbosch and G. T. Seaborg, Phys. Rev. **110**, 507 (1958).
[6] V. G. Nosov, J. Exptl. Theoret. Phys. (U.S.S.R.) **31**, 335 (1956), Soviet Phys. JETP **4**, 263 (1957).
[7] L. Granberg and J. S. Levin, Phys. Rev. **109**, 2063 (1958).

Translated by G. Volkoff
7