NOTAS DE FÍSICA

VOLUME XI

Nº 5

^{232}Th AND ^{238}U FISSION INDUCED BY LOW-ENERGY MONOCHROMATIC γ-RAYS

by

A. Manfredini, M. Muchnik, M. Severi,
H. Bösch, J. Lang, R. Müller, W. Wölfli,
and H. G. de Carvalho

CENTRO BRASILEIRO DE PESQUISAS FÍSICAS

Av. Wenceslau Braz, 71

RIO DE JANEIRO

1963

Notas de Física - Volume XI- Nº 5

^{232}Th AND ^{238}U FISSION INDUCED BY LOW-ENERGY MONOCHROMATIC γ-RAYS +

A. Manfredini, M. Muchnik* M. Severi
Istituto di Fisica dell'Università - Roma
Istituto Nazionale di Fisica Nucleare - Sezione di Roma

H. Bösch, J. Lang, R. Müller, W. Wölfli
Laboratorium fur Kernphysik, Eidg. Tech. Hochschule - Zurich**

H. G. de Carvalho***
Centro Brasileiro de Pesquisas Físicas, Rio de Janeiro

(Received September 26, 1962)

Summary - The photofission cross-sections of ^{232}Th and ^{238}U have been measured with nuclear emulsions at 6.61 MeV. Very good statistics were obtained combining a new type of monochromatic γ-rays source with some new developments in the technique for leading and processing nuclear emulsions.

+ Published in Nuovo Cimento, vol. 25, 534 (1962).

* Present address: Istituto di Fisica Nucleare, Sottosezione di Napoli.

** Part of this work has been supported by the Schweiz. Nationalfond zur Forderung der wissenschaftlichen Forschung, Kommission fur Atomwissenschaft.

*** This work was done while this author was at Istituto di Fisica dell'Università - Roma and Istituto Nazionale di Fisica Nucleare - Sezione di Roma.

1. INTRODUCTION

Almost all the results at present available on the photofission cross-sections and fissionabilities of ^{238}U and ^{232}Th relate to the energy interval from threshold (i.e. about 5 MeV) up to 20 or 30 MeV. The general shape of these cross-sections is that of the typical photonuclear process, with a large resonance at about 14 MeV. Several authors (1,2), using radiochemical techniques, have found a minimum lying a few MeV above the thresholds. On the other hand, KATZ et al (3), using ionization chambers, found two minima between the threshold and 8 MeV in the cross-section for ^{238}U. If the radiochemical results are normalized to the same value of the cross-section given by Katz at 8 MeV, they widely disagree at lower energies.

The study of the angular distribution of the fission fragments led Baz et al (4) to conclude that some quadrupole absorption was involved in the case of ^{238}U at 9.4 MeV maximum bremsstrahlung energy. To explain the overall picture it was recalled that this reaction may, in general, be due both to electric dipole and electric quadrupole interactions. However, the role played by the quadrupole absorption is unimportant at usual energies and the main contribution comes from dipole interaction. The situation changes just above the neutron emission threshold, since the ^{238}U (γ, n) reaction is excited essentially by E1 interaction. As was pointed out by BAZ and SMORODINSKY (5), the onset of the (γ, n) reaction must lead to a very low minimum in the dipole partial cross-section, lying somewhat above the (γ, n) threshold: neutron emission, proceeding mainly through dipole absorption "eats up" the dipole part of the photofission cross-section. Furthermore, since the quadrupole partial cross-section grows monotonically in the absence of any competing reaction, at a particular energy it must reach a predominant role in the value of the total photofission cross-section.

It is possible to evaluate the ratio of dipole to quadrupole partial cross-sections by studying the angular distribution at various energies [4]. From this, and the measurement of the total cross-section, it is possible to obtain the actual dipole and quadrupole cross-sections as functions of energy. This necessitates very precise measurements of both the angular distributions and the cross-section.

Most of the work done in this field used either bremsstrahlung beams from betatrons or linear accelerators, or the discrete spectrum of some proton induced nuclear reactions. The use of a continuous γ-ray spectrum introduces two main sources of uncertainty in the results. Firstly, the mixing of quanta of various energies obscures the nature of the angular distribution since many channels will contribute simultaneously and their anisotropies may tend to cancel out. Secondly, the cross-section will have to be obtained through the photon difference method which, as is well known, requires extremely good statistics in order to give reasonably accurate results.

For many years it was throught that the only monochromatic γ-ray source of practical importance in the energy region between

5 and 10 MeV was the F(p,γ) reaction. The main drawbacks of this source are its rather low intensity and the fact that actually three lines are emitted, namely at 6,14 6,93 and 7,14 MeV, with intensities which vary with the incident proton energy and with the thickness of the target. The thickness itself may vary during the experiment due to temperature effects and thus further uncertainties are introduced.

The study of the photofission of uranium and thorium at low energy would be best carried out by means of well-defined monochromatic γ-ray beams of high intensity, whose energy could be varied with precision between 5 and 10 MeV. The present work is the first result of a research making use of such beams, and concerns solely the unambiguous determination of the value of the total photofission cross-section for both ^{238}U and ^{232}Th at an energy of 6.61 MeV.

2. EXPERIMENTAL METHOD

Two new techniques have made possible the present work, namely a loading and processing technique of nuclear emulsions, and a way of obtaining a high intensity source for discrete γ-rays up to 11 MeV. The loading of emulsions with uranium and thorium is based on the mixing of nuclear emulsion in gel form with a known quantity of a solution of uranium or thorium citrate whose composition is accurately determined. This mixture is subsequently diluted with water, melted and poured onto suitable circular containers where it dries up to form easily stripped pellicles of very uniform thickness. Pellicles of up to 500 μm thickness have been obtained, with a heavy element content of up to 2.10^{20} nuclei per cm^3 of dry emulsion. This content may be known to better than 1% by just weighing the single pellicles cut out from the whole circular emulsion, and comparing the weights with the total weight.

For this particular work the emulsion used was Ilford's KO. The uranium loaded pellicle was 416 μm thick with a load of $6.38\ 10^{18}$ nuclei/cm^2; the thorium pellicle was 375 μm thick with a load of $6.79\ 10^{18}$ nuclei/cm^2.

The processing technique, described elsewhere [6], is such that now low-charge tracks are developed, in particular no α-particle tracks are to be seen. It furthermore eliminates any electron background due to soft γ-rays, which in these experiments is usually quite heavy. Only the fission fragment tracks are recorded, and these show up with typical length and ionization.

The monochromatic γ-ray source, described elsewhere as well[7] is based on (n,γ) reactions using the high thermal neutron flux of a nuclear reactor. The γ-rays produced by this radiative capture in different elements are limited to the narrow range from 3 to 11 MeV, and the energies are generally known with an uncertainty smaller than 8 keV. A suitable target element is placed near the core of the reactor, preferably within a tangential beam hole so that the collimated γ-ray beam will not see the core itself. In this way both the fast neutron and the γ-ray backgrounds are considerably reduced. The detector - in our case the nuclear emulsions - are placed right at the exit of the beam hole (Fig. 1). This affords an excellent collimation, since the distance from the target to the emulsions is of the order of 4 m, corresponding to a semiaperture angle of about 6 milliradians. One of the most important advantages of this type of monochromatic source is its very high intensity, about a thousand-fold that of the (p,γ) sources in practice. The intensity of the beam may be furthermore measured to within 10%, by means of a NaI(Tl) scintillator [8]. In our case the crystal dimensions were 3 in. × 3 in., and its axis was adjusted exactly parallel to the beam axis. Due to the strong and isolated γ-ray lines of all the used (n,γ)-spectra, the dose is measured as follows: in the photpeak of the line a Gaussian is fitted; with the area of the

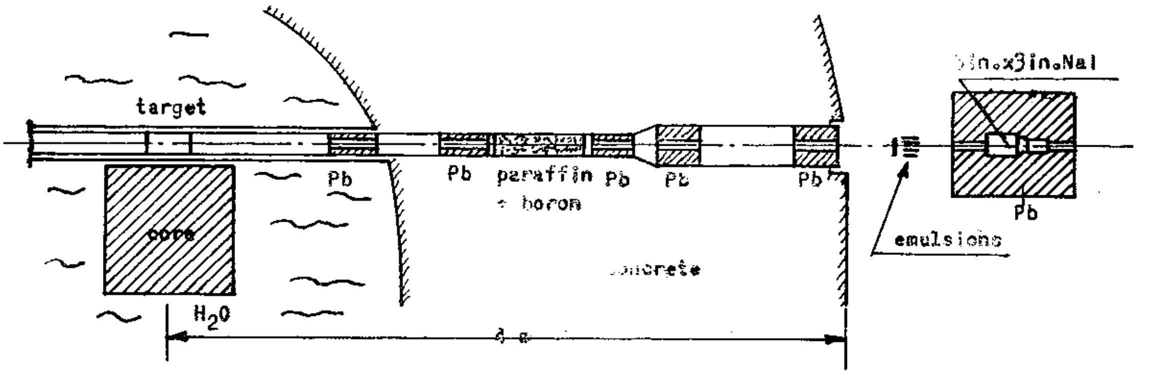

Fig. 1. Experimental lay out. Note the horizontal exposed emulsions. The measurement of the angular distribution of the fission fragments, carried out by means of these emulsions, will constitute the subject of Part II (to be published) of the present paper. For a description of the collimation assembly of the γ-ray source see ref. (7).

Gaussian and the experimentally determined photon efficiency, the intensity (i.e. the number of quanta per second) can be easily calculated. Since an exactly adjusted collimation in front of the crystal determines the beam diameter, the desired γ-ray flux is also thus obtained. The flux is measured at the beginning and at the end of each irradiation. During the irradiation the thermal neutron flux of the reactor is contolled by means of a monitor.

The present exposure was carried out at the 1 MW swimming--pool type reactor "SAPHIR", where these findings were obtained. A schematic diagram of the experimental set-up is shown in Fig. 2. The exposure was carried out with a weighted mean energy of 6.61 MeV. The emulsions were exposed at normal incidence.

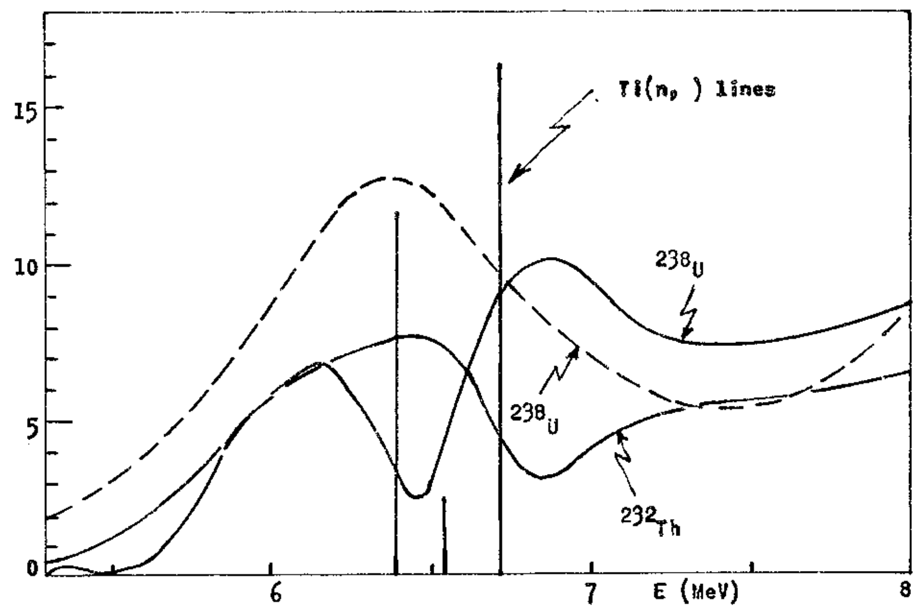

Fig. 2. Sketch of preceeding experimental results for the photofission cross-section of ^{238}U (— Katz et al. (3)) Schmitt's results have been normalized to those of Katz at 8 MeV. Shown in the figure also the three main lines of the Ti(n,γ) process with their corresponding intensities in arbitrary units.

Since the angular distribution of the fission fragments is peaked at 90° with respect to the incident γ-ray, and since the shrinking of the emulsion after processing tends to flatten the tracks on the plane of the pellicle, most of the tracks are very easily seen at the microscope and extremely few will be lost on account of large dip angles. The scanning consisted in counting the fission events over an accurately known area. An eyepiece with a calibrated square cell which defined the scanning field, allowed the determination of this area to better than 1%. A reticule placed on the emulsion and made up of points 50 μm in diameter evenly spaced at every 500 μm was later used to accurately identify a sample of fission events in order to control the scanning efficiency by the method of double scanning. It was found to be (95 ± 1)%.

3. RESULTS

The measured intensities of the three lines involved in our experiment and due to the (n, γ) process on titanium are shown in Table I.

TABLE I

Energy (MeV)	Measured intensity (quanta/100 radiative captures)
6.753	41.0
6.550	6.2
6.413	29.4

The present exposure, thus, took place at a weighted mean energy of $6.61 ^{+.14}_{-.20}$ MeV. Table II summarizes the data necessary for the calculation of the photofission cross-sections, which are given by

$$\sigma_f = \frac{f}{\varphi N} ,$$

where N_f is number of fissions per unit area, φ is the γ-ray dose (quanta/cm^2), and N is the heavy element load of the particular pellicle (nuclei/cm^2).

TABLE II

Element	Number of fissions	Area (cm^2)	N_f	Flux (quanta/cm^2)10^{11}	(nuclei/cm^2)10^{18}	Scanning efficiency
Thorium	2.054	0.33±0.01	6.245	2.0±0.2	6.79±0.07	(95±1)%
Uranium	3.324	0.33±0.01	9.996	2.0±0.2	6.38±0.06	(95±1)%

There are three sources of spurious fission events. In the first place titanium emits a considerable number of quanta at energies other than those of Table I. Being, many of these, above the threshold for photofission, and being their integral intensity 1.5 quanta/100 radiative neutron captures (9), due account must be taken in the calculation of the cross-sections. Taking as good the photofission cross-section curves given by previous authors (3), the weighted contribution of these lines, according to their relative intensities, is 5.7% in the case of uranium and 4.3% in the case of thorium.

Secondly there is also a small contribution of ^{235}U fissions. Taking .07% as this isotope's contamination of the uranium load in the pellicles, and considering that its fissionability is 1.5 times greater than that of ^{238}U, it is easily seen that the cross-section has to be diminished by about 0.3%. The given cross-sections have been corrected for these effects.

Lastly, the contribution due to the fast neutron contamination of the beam should be examined. This contamination was $1.7 \cdot 10^{-5}$ neutron/γ-ray, while the spectrum of these neutrons should be the usual fission spectrum coming from the reactor, except for a slight hardening. Since, however, both the uranium and thorium cross-sections for fission by fast neutrons are quite small under 1 MeV - in fact the thorium cross-section has the threshold at about 1.5 MeV - one may neglect the hardening. Making use of the fission cross-sections given in the literature[10-11], it is easily calculated that the contribution to the number of uranium fissions due to the fast neutrons in our case should be of the order of 0.06%, and in the case of thorium, 0.02%. This spurious effect is thus entirely negligible.

This experiment consisting of a single exposure to what amounts to be a single γ-ray energy, it cannot be taken as a basis for deciding on the shape of the cross-section curves. Since titanium emits essentially three lines whose intensities are known, it is however possible to take as good the cross-sections given by previous authors at different energies, and to weigh each value of σ_f according to the relative intensity of the three lines. A weighted mean cross-section is thus obtained with which to compare the results of the present paper. In doing so, however, it is important to bear in mind that the cross-section curves given by previous authors are obtained by the photon difference method, using yield curves from bremsstrahlung beams. The necessarily large uncertainties (of the order of 30%) in the resulting cross-sections render the comparison with the present data not very significant. Figure 2 shows the differentiated yield curves for uranium and thorium given by KATZ[3] as well as the one for uranium given by SCHMITT[2]. The latter one has been normalized to the same value of KATZ's curve at 8 MeV. Shown in the same figure are the three lines emitted by titanium. The weighted mean cross-sections are displayed in Table III showing a fair agreement both with the results of KATZ and with those of SCHMITT. Further experiments at other energies (exposures with sulphur and lead targets have been already carried out) will provide more information concerning the actual shape of the cross-section curves.

TABLE III

	σ_{Th}(mb)	σ_U (mb)	$\sigma_{f(Th)}/\sigma_{f(U)}$
Present work	4.73±0.50	7.89±0.83	0.600±0.005
Katz et al.[3]	5.6	6.6	0.92
Schmitt et al.[2]	-	10.7	-

One further experimental result is obtained by dividing the photofission cross-section for thorium by that of uranium. This is the definition of relative fissionability given by HUIZENGA et al.[12], based on the fact that the total anelastic cross-sections for photon absorption corresponding to these two elements are practically equal. Our result is included in Table III, and may be compared with that of KATZ. It is to be noticed that since the uranium-loaded and the thorium-loaded pellicles were exposed together, the measurement of the γ-ray dose - which is the main source of error - is not relevant as far as the relative fissionability is concerned.

* * *

References

1. E. J. WINHOLD and I. HALPERN: Phys. Rev., 103, 990 (1956).
2. R. A. SCHMITT and R. B. DUFFIELD: Phys. Rev., 105, 1277 (1957).
3. L. KATZ, A. P. BAERG and F. BROWN: Second U. N. International Conference on the Peaceful Uses of Atomic Energy, vol. 45 (1958), p. 188.
4. A. I. BAZ, N. M. KULIKOVA, L. E. LAZAREVA, N. V. NIKITINA and V. A. SEMENOV: Second U. N. International Conference on the Peaceful Uses of Atomic Energy, vol. 15 (1958), p. 184.
5. A. I. BAZ and Y. A. SMORODINSKY: Quoted in ref.[4] as a private communication.
6. H. G. DE CARVALHO and A. G. da SILVA: Suppl. Nuovo Cimento, 19, 24 (1961).
7. L. JARCZYK, H. KNOEPFEL, J. LANG, R. MÜLLER and W. WÖLFLI: Nucl. Insts., 13, 287 (1961).

8. L. JARCZYK, J. LANG, R. MULLER and W. WOLFLI: Helv. Phys. Acta, 34, 488 (1961) and Nucl. Instr. (to be published).

9. E. TROUBETZKOY and H. GOLDSTEIN: Nucleonics (Nov. 1960), p. 171.

10. D. J. HUGHES: Second U. N. International Conference on the Peaceful Uses of Atomic Energy, vol. 16 (1958), p. 8.

11. S. P. KALININ and V. M. PANKRATOV: Second U. N. International Conference on the Peaceful Uses of Atomic Energy, vol. 16 (1958), p. 136.

12. J. R. HUIZENGA, J. E. GINDLER and R. B. DUFFIELD: Phys. Rev., 95, 1009 (1954).

* * *