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PHOTOFISSION CROSS SECTIONS OF ^{233}U AND ^{239}Pu NEAR THRESHOLD INDUCED BY GAMMA RAYS FROM THERMAL NEUTRON CAPTURE

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The photofission cross sections of ^{233}U and ^{239}Pu have been studied by using monochromatic and discrete photons, in the energy interval from 5.43 to 9.72 MeV, produced by thermal neutron capture. The photofission data are presented and compared with those of other authors. A possible structure was observed in the ^{233}U cross section near 7.23 MeV. According to the liquid drop model the heights of the simple fission barriers were determined: (5.6 ± 0.2) MeV and (5.7 ± 0.2) MeV for ^{233}U and ^{239}Pu respectively. The relative fissionability of the samples was also determined at each excitation energy and shown to be energy independent: (2.12 ± 0.25) for ^{233}U and (3.32 ± 0.41) for ^{239}Pu.

1. Introduction

Photonuclear studies for nuclei in the actinide region are still being performed by several laboratories due to the few data available for those nuclei in the literature. The main objective of these studies has been to obtain nuclear information at excitation energies in the region of the giant dipole resonance (GDR, between 10 and 20 MeV) and in the region of low energy, near the photofission and photoneutron thresholds (5–10 MeV) [1].

Photofission experiments can be performed by using several kinds of gamma ray sources, with resolutions ranging from a few eV to several keV [2]. With these markedly different energy resolutions it is only possible to do a qualitative comparison between the results obtained with different gamma ray sources. These comparisons are still useful because of the significance of such measurements and the lack of data available in the literature. However, they are always questionable and of limited value.

In the present work measurements of the photofission cross sections for the ^{233}U and ^{239}Pu nuclei were made in the energy region near threshold (5.43–9.72 MeV), using discrete and high resolution photons produced by neutron capture.

The data published previously for these nuclei are: (a) low energy region: Huizenga et al. [3] for ^{233}U, Ostapenko et al. [4] for ^{233}U and ^{239}Pu, Shapiro et al. [5] for ^{239}Pu and Dragnev et al. [6] for ^{239}Pu; (b) the GDR region: there is an old paper of Katz et al. [7] for ^{233}U and ^{239}Pu, Gurevich et al. [8] for ^{239}Pu, and the recent paper of Bermann et al. [9] for ^{233}U and ^{239}Pu. Only Dragnev [6] has used a neutron capture gamma ray source.

2. Experimental procedures

The experimental procedures are the same as used by Mafra [10] and Geraldo [11]. Briefly, a collimated gamma radiation beam is produced by thermal neutron capture in several critically chosen targets placed near the core of the IEA-R1 research reactor. This beam passes through several filters to minimize the neutron beam contribution and, after leaving the beam hole, impinges on the sample under study located at a distance of about 550 cm from the core of the reactor. The arrangement may be seen in fig. 1.

The gamma fluxes incident on the samples were measured by means of a 3 in. × 3 in. NaI(Tl) crystal and their intensities were obtained by the following equation:

$$I(\gamma) = \frac{\text{photopeak area}}{p(E) \times (1 - \exp(-u(E)/L))}, \tag{1}$$

Table 1
The targets, their principal (γ) line energy and fluxes used in this work

Target	E [MeV]	[γ/cm^2 s]
^{32}S	5.43	$(6.89 \pm 0.73) \times 10^4$
^{48}Ti	6.73	$(2.89 \pm 0.32) \times 10^5$
^{55}Mn	7.23	$(1.10 \pm 0.13) \times 10^5$
^{207}Pb	7.38	$(1.49 \pm 0.16) \times 10^5$
^{56}Fe	7.64	$(1.86 \pm 0.22) \times 10^5$
^{27}Al	7.72	$(1.63 \pm 0.21) \times 10^5$
^{63}Zn	7.88	$(1.17 \pm 0.13) \times 10^5$
^{64}Cu	7.91	$(1.89 \pm 0.23) \times 10^5$
^{84}Ni	9.00	$(1.74 \pm 0.20) \times 10^5$
^{52}Cr	9.72	$(8.38 \pm 1.06) \times 10^4$

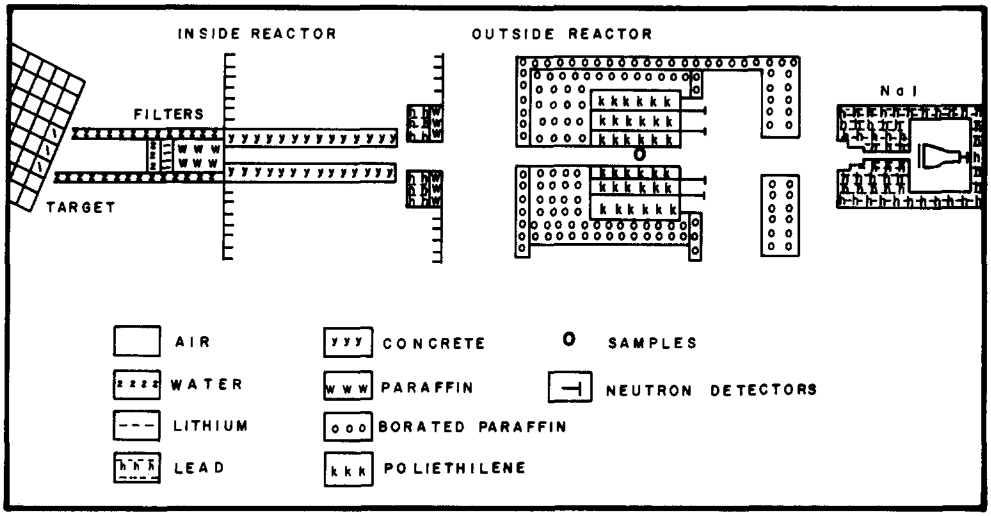

Fig. 1. Schematic experimental arrangement used in the photofission measurements. The neutron detectors that will be used in the photoneutron cross section measurements are also included.

where: $p(E)$ is the photofraction; $(1-\exp(-u(E)/L)$ is the efficiency of the scintillation counter for gamma rays at energy E.

The photopeak area was evaluated making a weighted least-squares fit of a Gaussian function on the experimental data.

The denominator of eq. (1), which represents the photopeak efficiency, was taken from ref. [12] due to the similarities of the two arrangements. The uncertainties of these values range from 5% to 10%.

The fluxes of the main line of each target used in this experiment are shown in table 1.

The reactor power was monitored by a self-powered detector.

The photofission fragments were detected by the track registration technique in Makrofol-KG (8 μm). The Makrofol foils and the ^{233}U and ^{239}Pu samples were irradiated in the form of "sandwiches" separately. After these irradiations the tracks created by the fission fragments in the Makrofol foils were etched in a (35%wt) KOH solution at 60 °C for 30 min. Finally the total number of tracks was counted by an automatic spark counting chamber [13]. The total efficiency of this technique was obtained by using a ^{252}Cf calibrated source and its value was 0.422 ± 3.1%.

In order to subtract the background contribution in the photofission measurements caused by gamma rays coming from the reactor core (mainly aluminium capture gamma rays) a blank target was used. To simulate the gamma attenuation inside the targets a replica of each one was placed inside the beam hole and outside the reactor [11].

The ^{233}U and ^{239}Pu samples were granted by the International Atomic Energy Agency (IAEA) and contained 52.4 and 51.4 mg respectively, deposited in the form of U_3O_8 and PuO_2 on four titanium disks, each with an active diameter of 40 mm.

The mass evaluation and isotopic analysis of these samples were carried out by AERE-Harwell Chemistry Division [14] using a gravimetric method and the results are shown in table 2. The masses of the samples have been experimentally confirmed by using the gamma spectrometry method [15].

The error sources were: the photon flux calibrations 11–13%, fission detector efficiency 3.1%, mass determinations 2%, self-absorption in the samples 4–5% and reproducibility and statistical uncertainties in the photofission counts 2–5%. These values resulted in an overall uncertainty of about 15% for the final photofission cross sections.

Table 2
Masses of the samples, including the isotopic percentage

Nuclei	Masses [mg]	Isotopic percentage [atoms/%]
^{233}U	(1) 13.9	^{233}U–99.702
	(2) 13.0	^{234}U– 0.236
	(3) 12.8	^{235}U– 0.012
	(4) 12.7	^{238}U– 0.050
	Total 52.4 (±2%)	
^{239}Pu	(1) 12.8	^{238}Pu–00.01
	(2) 12.6	^{239}Pu–99.01
	(3) 13.2	^{240}Pu–00.98
	(4) 12.8	
	Total 51.4 (±2%)	

3. Results and discussion

Because ^{233}U and ^{239}Pu present large fission cross sections for thermal neutrons, special care was taken to control this background source. An experimental verification of fission induced by these neutrons was made by getting some data both with and without cadmium foil wrapped around the samples. The results showed no difference between the measurements, within the experimental errors. The targets used to produce gamma radiation were critically chosen in such a way that the secondary gamma ray intensities were less than 10% of the main gamma rays for the majority of them [17].

In order to calculate the photofission cross sections, contributions due to the secondary gamma rays with energies above the fission threshold were taken into account. For this endeavour, the photofission cross sections were represented by using the following formula

$$\sum_i \sigma(\gamma, f)_i r_i = \frac{F}{eNG}, \tag{2}$$

where: $\sigma(\gamma, f)_i$ = photofission cross sections at the energy of the ith line in the gamma ray spectrum emitted by the target element; r_i = gamma ray flux of the ith line relative to the main gamma line, corrected for the attenuation in the filters of the collimation assembly; F = number of fission tracks obtained per unit of time of exposition; e = total fission detection efficiency; G = flux of the main gamma ray.

By making approximations of about 60 keV in energy, a set of ten linear equations with 40 unknown quantities may be obtained. This system of linear equations was solved using the same approximations reported in refs. [10,11].

Table 3
^{233}U and ^{239}Pu photofission cross sections as a function of gamma ray energies

Targets and energy [MeV]		^{233}U $\sigma(\gamma, f)$ [mb]	^{239}Pu $\sigma(\gamma, f)$ [mb]
S	5.43	8.25 ± 4.08	8.89 ± 4.87
Ti	6.73	13.99 ± 2.05	20.80 ± 2.12
Mn	7.23	29.79 ± 3.84	26.83 ± 6.20
Pb	7.38	20.88 ± 2.42	34.52 ± 3.99
Fe	7.64	21.82 ± 3.20	37.56 ± 5.27
Al	7.72	26.77 ± 3.73	44.22 ± 6.19
Zn	7.88	26.14 ± 3.70	42.42 ± 7.48
Cu	7.91	29.10 ± 4.32	36.20 ± 4.85
Ni	9.00	72.84 ± 11.57	92.67 ± 14.87
Cr	9.72	98.48 ± 17.28	131.85 ± 25.02

The photofission cross sections obtained in this work for the ^{233}U and ^{239}Pu nuclei are given in the table 3 and are compared with the data of other authors in figs. 2 and 3.

A comparison between the present results with cross section data obtained by unfolding bremsstrahlung spectra yields satisfactory agreement. The present data are in reasonable agreement with the data of Katz [7], Ostapenko [4] and Shapiro [5] for ^{233}U and ^{239}Pu. The results of high resolution monoenergetic photon measurements performed with nuclear gamma rays agree with the present data for some cases but not for others [3,6,9]. The data reported by Huizenga [3] for ^{233}U agree with the present data. A possible structure was observed near the energy of 7.23 MeV for ^{233}U that may be endorsed by the data at 7 MeV obtained by Huizenga [3], but the lack of data in this region does not permit a conclusion about it. This structure was not observed by

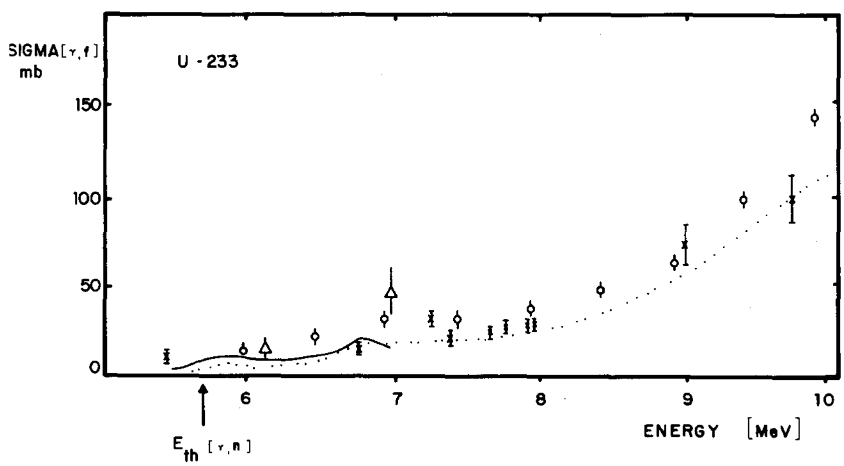

Fig. 2. ^{233}U photofission cross sections, $\sigma(\gamma, f)$ in millibarn. The symbols mean: dotted curve – ref. [7] (1958), open triangles – ref. [3] (1962), open circles – ref. [9] (1986), solid curve – ref. [4] (1981), solid data points – this work (1987). The photoneutron threshold energy reaction (γ, n) is also included, and it was taken from ref. [26].

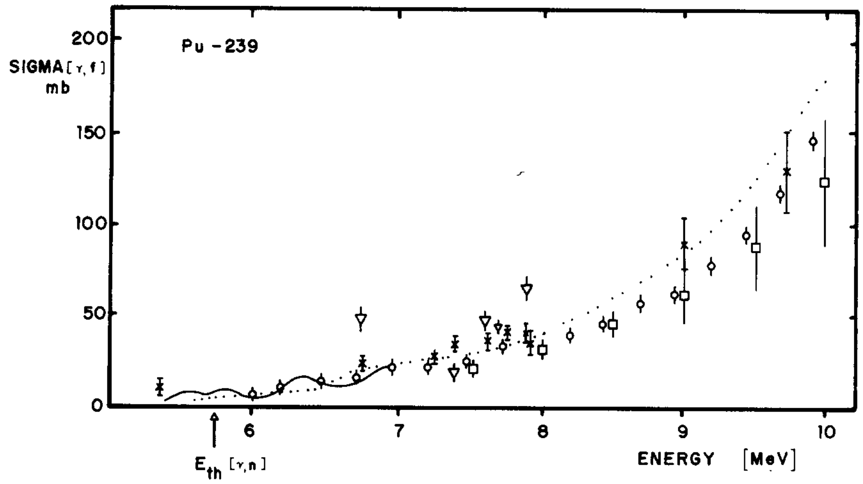

Fig. 3. ^{239}Pu photofission cross sections, $\sigma(\gamma, f)$ in millibarn. The symbols mean: dotted curve – ref. [7] (1958), open triangles – ref. [6] (1973), open squares – ref. [5] (1971), open circles – ref. [9] (1986), solid curve – ref. [4] (1981), solid data point – this work (1987). The photoneutron threshold energy reaction (γ, n) is also included, and it was taken from ref. [26].

other authors. The cross sections reported by Bermann [9] have a tendency to lie higher than our values by 30% in the region up to 8.0 MeV. At greater energies the data points agree very well. There are serious discrepancies between our results and the data reported by Dragnev [6] for ^{239}Pu who has used also a high resolution gamma radiation.

The most remarkable feature of the present measurements was the peak found at 7.23 MeV for ^{233}U that may be supported by Huizenga's results [3]. However, our measurements were of such high resolution (comparable to or smaller than the spacing between levels in the compound nucleus) that the data points measured could easily coincidence with peaks or valleys in the underlying fine structure of the cross section.

Table 4
Relative fissionability of ^{233}U and ^{239}Pu

Targets and energy [MeV]		Relative fissionability of ^{233}U	Relative fissionability of ^{239}Pu
Ti	6.73	1.35 ± 0.30	2.00 ± 0.38
Mn	7.23	4.15 ± 1.02	3.74 ± 1.16
Pb	7.38	2.05 ± 0.32	3.38 ± 0.53
Fe	7.64	2.18 ± 0.99	3.76 ± 1.70
Al	7.72	2.91 ± 0.90	4.81 ± 1.51
Zn	7.88	2.35 ± 0.79	3.82 ± 1.35
Cu	7.91	2.03 ± 0.37	2.53 ± 0.42
Ni	9.00	1.97 ± 0.66	2.50 ± 0.85
Mean value		2.12 ± 0.25 [a]	3.32 ± 0.41

[a] Mean value calculated without the 7.23 MeV energy datum.

3.1. Relative fissionability of ^{233}U and ^{239}Pu

The relative fissionability parameter as defined by Huizenga et al. [18] represents the ratio between the fission yield obtained for a given nuclide relative to the yield obtained for the ^{238}U nucleus at the same excitation energy. For the purpose of studying this parameter, the photofission cross sections of ^{238}U were taken from refs. [19,20], because these authors have used a gamma source type similar to the present one.

The relative fissionability of ^{233}U and ^{239}Pu at excitations energies coincident with those studied for ^{238}U are shown in table 4.

The resulting values appear to be independent of excitation energy in the 6.73–9.72 MeV range, at least within experimental errors, excluding the result at 7.23 MeV for ^{233}U where a structure in the photofission cross section was obtained for this nucleus which is not present in the ^{238}U photofission data.

Table 5
Comparison between the relative fissionability of ^{233}U and ^{239}Pu measured by several authors

Relative fissionability of ^{233}U	Relative fissionability of ^{239}Pu	Energy [MeV] interval	Ref.
2.12 ± 0.25	3.32 ± 0.41	6.73– 9.00	This work
2.49 ± –	2.51 ± –	12.00–20.00	McElhinney [22]
2.36–2.62	3.1–3.51		
± 4–20%	± 4–20%	12.00–20.00	Huizenga [18]
2.30 ± –	3.55 ± –	5.00–20.00	Katz [7]
–	3.00 ± –	5.00–12.00	Ivanof [21]

The mean values obtained were 2.12 ± 0.25 for ^{233}U and 3.32 ± 0.41 for ^{239}Pu indicating that ^{233}U and ^{239}Pu are more fissile than ^{238}U in this energy range.

These values are in excellent agreement with the results of other authors, as shown in table 5.

A correlation between the relative fissionability and the parameter Z^2/A of the liquid drop model was performed. It is in good agreement with the trend, as can be seen in fig. 4.

3.2. *Study of the fission barrier for ^{233}U and ^{239}Pu*

The experimental results obtained in this work are insufficient to specify all the parameters needed to describe the double-humped fission barrier of ^{233}U and ^{239}Pu. However, the barrier height predicts by the liquid drop model represents the energy of the higher of the two barriers to a good approximation and may be visualized as a single inverted parabolic barrier of height H and curvature $\hbar w$. The transmission T_f of this simple barrier was calculated by Hill et al. [23] and is represented by the equation:

$$T_f = \{1 + \exp[2\pi(H - E)/\hbar w]\}^{-1}, \qquad (4)$$

where E is the excitation energy.

The fission barrier transmission can be expressed as:

$$T_f(E) = T(E)\frac{\sigma(\gamma, f)(E)}{\sigma(a)(E) - \sigma(\gamma, f)(E)}, \qquad (5)$$

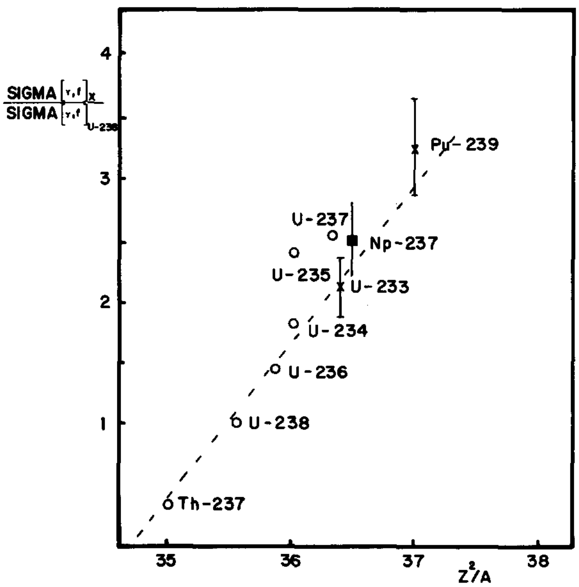

Fig. 4. Relative fissionability of several nuclei as a function of Z^2/A, the liquid drop parameter. The data represented by open circles were taken from ref. [18] and the data represented by a black square and the best fitted curve (dashed curve) were taken from ref. [11].

Table 6
Comparison between the single and double fission barrier parameters of ^{233}U and ^{239}Pu measured by several authors

^{233}U

E_f [MeV]	$\hbar w$ [MeV]	Ref.
5.6 ± 0.2	$0.56 \pm -$	This work
5.7 ± 0.3	–	Vandenbosch [24]

^{239}Pu

E_f [MeV]	$\hbar w$ [MeV]	Ref.
5.7 ± 0.2	$0.57 \pm -$	This work
5.8 ± 0.3	–	Vandenbosch [24]
6.43 ± 0.20 [a]	1.00 ± 0.1	Back [25]
$5.50 \pm -$ [a]	$0.55 \pm -$	

[a] Double fission barrier parameters.

for excitation energies below the photoneutron threshold.

The fission barrier transmission was obtained for ^{233}U and ^{239}Pu from the measured photofission cross sections at energy 5.43 MeV, the unique datum lower than the photoneutron threshold.

The total photoabsorption cross section $\sigma(a)(E)$ was taken from ref. [9], and the gamma ray transmission was taken from ref. [24]. The expression for this transmission is semiempirical and, between 4.5 and 6.5 MeV, may be approximated by the equation [27]:

$$T(0.5, E) = 0.1 \exp\left(\frac{(E - 6.02)}{0.41}\right), \qquad (6)$$

where E is in MeV.

The curvature of the barrier was taken from ref. [24] by using a value for $\hbar w$ according to the liquid drop model.

The values of the barrier parameters determined for ^{233}U and ^{239}Pu are listed and compared with other data in table 6.

The barrier heights. (5.6 ± 0.2) MeV and (5.7 ± 0.2) MeV for ^{233}U and ^{239}Pu respectively represents in principle the energy where the penetration is equal to 0.5 for the lowest transition state, $(J = 3/2;\ K = 1/2)$ for ^{233}U and $(J = 1/2;\ K = 1/2)$ for ^{239}Pu.

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