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PHOTOFISSION CROSS SECTIONS OF U-233 AND Pu-239 NEAR THRESHOLD INDUCED BY GAMMA RAYS FROM THERMAL NEUTRON CAPTURE *

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ABSTRACT

The photofission cross sections of U-233 and Pu-239 have been studied using monochromatic photons produced by thermal neutron capture in several materials placed in a radial beam hole of the IEA-R1, 2 MW pool type research reactor, in the energy interval from 5.43 MeV to 9.72 MeV. The gamma flux incident on the samples were measured using a (3x3) inch. NaI(Tl) crystal. The photofission fragments were detected in MAKROFOL-KG (solid state nuclear track detector) etched during 30 min. in a KOH (35%wt) solution at 60°C. The efficiency of the detector was obtained using a Californium-252 calibrated source and its value was (0.4323+3%). The tracks were counted by means of an automatic spark counting. Analyzing the photofission data we have observed similarities between the cross sections obtained for the two samples in comparison with other authors. A structure was also observed in the U-233 cross section near the energy of 7.23 MeV. According to the liquid drop model the height of the simple fission barrier were determined: (5.6 ± 0.2) MeV and (5.7 ± 0.2) MeV for U-233 and Pu-239 respectively. The relative fissionability of the samples to U-238 were also determined in each excitation energy and showed to be energy independent: (2.12 ± 0.25) for U-233, and (3.32 ± 0.41) for Pu-239.

SECÇÕES DE CHOQUE DE FOTOFISSÃO DO U-233 E Pu-239 JUNTO AO LIMIAR, INDUZIDAS POR RADIAÇÃO GAMA DE CAPTURA DE NÊUTRONS

RESUMO

As secções de choque de fotofissão do U-233 e do Pu-239 foram obtidas no intervalo de energia entre 5,42 MeV e 9,72 MeV usando fótons monocromáticos produzidos pela captura de nêutrons térmicos em diversos materiais colocados no canal radial do reator IEA-R1. Os fluxos gama incidentes nas amostras foram medidos usando um cristal de NaI(Tl) com dimensões de (3x3) polegadas. Os fragmentos de físsão foram detectados em Makrofol KG, revelados durante 30 minutos numa solução de KOH (35%/massa) à 60°C. A eficiência total de detecção foi obtida usando uma fonte calibrada de Cf-252: E_f = (0,43 + 3,1%). Os traços foram contados eletricamente usando uma câmara de descarga automática. Analizando os dados de

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fotofissão, observou-se muitas semelhanças entre as secções de choque obtidas para as duas amostras em comparação com outros autores. Uma estrutura foi também observada na secção de choque do U-233 próximo à energia de 7.23 MeV. Usando o modelo da gota líquida a altura da barreira simples de fissão foi determinada H = (5,6 + 0,2) MeV e H = (5,7 + 0,2) MeV para os núcleos de U-233 e Pu-239 respectivamente. A fissionabilidade relativa das amostras em relação ao U-238 foi também determinada em cada energia de excitação e mostrou ser independente da energia: (2,21 $\pm$ 0,25) para o U-233 e (3,32 $\pm$ 0,41) para o Pu-239.

INTRODUCTION

Photonuclear studies for nuclei in the actinides region are being still performed by several laboratories due the little data available for those nuclear parameters in literature. The main objective of these studies is to obtain informations about the region of the giant dipole ressonance (GDR - between 10 and 20 MeV) and about the region of low energy, near the photofission and photoneutron thresholds (5 - 10 MeV).

Fission barriers characteristics may be obtained from the photofission cross sections because these reactions make use of the relative simplicity and directeness of the eletromagnetic interaction to explore the process of nuclear fission(1). Photofission experiments may be performed using various kinds of gamma rays sources, with resolutions ranging from a few eletrons-volts to several kilo-eletron-volts(2). These markedly different energy resolutions, permit us only a qualitative comparison between the results obtained by different gamma rays sources. This comparison continues to be useful because of the significance of such measurements and the little data available in the literature, but are always questionable and of limited value.

In the present work measurements of the photofission cross sections of the U-233 and Pu-239 nuclei were made in the energy region near threshold (5.43 to 9.72 MeV), using monochromatic photons produced by neutron capture in several targets in the IEA-R1 research reactor, a high resolution gamma ray source. For this low energy region the data published previously about these nuclei are: Huizenga et al (3) for U-233; Ostapensko(4) for U-233 and Pu-239; Shapiro et al (5) for Pu-239 and Dragnev(6) for Pu-239. Including the GDR region there are the old paper of Katz(7) for U-233 and Pu-239; Gurevich(8) for Pu-239, and the recent paper of Berman et al [9] for U-233 and Pu-239. Only Dragnev[6] has used a gamma rays source of high resolution like ours. So, there are little data available on these parameters at the present time and there is a marked discrepancy among these reported values and a lack of experimental data at several energies near threshold.

For a complete study of the photonuclear reactions in these nuclei, the photoneutron and photoabsorption cross sections are being performed in this energy range, using this neutron capture gamma ray source.

EXPERIMENTAL METHODS

The experimental methods are based on those used in other experiments whose detailed description appears in references[10,11].

Briefly the method consists of a gamma radiation beam produced by thermal neutron capture in several chosen targets placed near the core of IEA-R1 research reactor. This collimated gamma radiation 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 figure 1.

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

$$I(\gamma) = \frac{\text{photopeak area}}{p(E)\ \{1 - \exp\left(\frac{-u(E)}{L}\right)\}} \qquad (1)$$

Where

$p(E)$ is the photofraction

$\frac{u(E)}{L}$ is the efficiency of the scintilattion counter for gamma rays at energy E

The photopeak area was evaluated assuming a gaussian peak shape and making a weighted least square fit to the experimental data.

The denominator of equation 1 that represents the photopeak efficiency was taken from reference[12] due the similarities of the two arrangements. The incertainties of these values are ranging from 5 to 10%.

The gamma ray flux of the principal line for each target used in this experiment are shown in the table I.

Table I - The targets, their principal gamma ray energy and fluxes used in this work.

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

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

The photofission fragments were detected by the fission track registration technique in Makrofol KG (8 μm). The Makrofol foils and the U-233 and Pu-239 samples were irradiated in the form of sandwiches separately. The total efficiency of this technique was obtained in the same way described previously by using a Cf-252 calibrated source. 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 minutes. Finally the total number of tracks were counted by an automatic discharge chamber[13].

The total efficiency of the fission detection was (0.4223 ± 3.1%).

In order to subtract the background counts 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 U-233 and Pu-239 samples were supplied by the International Atomic Energy Agency (IAEA) and contained 52.4 mg and 51.4 mg respectively, deposited in the form of U_3O_8 and PuO_2 on four titanium disks, each one, with an active diameter of 40 mm.

The calibration and isotopic analisys of these samples were made by AERE-HARWELL Chemistry Division(14) using a gravimetry method and the results are described in the table II. These results were confirmed experimentally by us using the gamma spectrometry method for these masses measurements(15).

Table II - Masses of the samples, including the isotopic percentage.

NUCLEI	MASSES (mg)	ISOTOPIC PERCENTAGE ATOMS/%
U-233	1- 13.9	U-233 - 99.702
	2- 13.0	U-234 - 0.236
	3- 12.8	U-235 - 0.012
	4- 12.7	U-238 - 0.050
	Total = (52.4 ± 2%)	
Pu-239	1- 12.8	Pu-238 - 0.01
	2- 12.6	Pu-239 - 99.01
	3- 13.2	Pu-240 - 0.98
	4- 12.8	
	Total = (51.4 ± 2%)	

Systematic uncertainties arising from the photon flux calibration are, ~ 11-13%, from the fission detector efficiency 3.1%; from the masses determinations 2% and from the reproducibility of the photofission counts 2-5%; resulting in a overall systematics uncertainties in the final photofission cross sections about 15%.

RESULTS AND DISCUSSION

Because U-233 and Pu-239 are fissile, special care had to the taken to avoid overestimating the photofission cross sections caused by fission events initiated by moderated neutrons from the beam-hole. By using the adequate filters[11,16] shown in fig. 1 the neutron flux was minimized to ~ zero. The experimental verification of this fact was made by getting some data both with and without cadmium foil wrapped around the samples: the cadmium foil virtualy eliminated the slow neutron fission. The results showed, at least within experimental errors, no difference between the measurements with and without cadmium.

The targets used to produce gamma radiation were chosen in a such way that the secondary gamma rays intensities are < 10% of the main gamma rays in the majority of targets[17].

In order to calculate the photofission cross sections the contributions of the secondary gamma rays with energies above the fission threshold must be taken into account. The relation between the experimental data and the photofission cross section may be seen in the following formula:

$$\sum_i \sigma(\gamma,f)i \; r_i = \frac{F}{\varepsilon . N . \phi}$$

Where:

$\sigma(\gamma,f)i$ = photofission cross section at the energy of the i'th line in the gamma ray spectrum emitted by the target element;

r_i = gamma ray flux of the i'th 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;

ε = total fission detection efficiency;

N = number of uranium or plutonium atoms in the samples;

ϕ = flux of the main gamma ray.

By using the ten targets, the set of linear equations obtained was solved with the same approximations used in references(10-11). Briefly, the solution of the set of equations is found firstly to the quadratic system, formed by using only the main gamma line. Second a linear interpolation is made between the first set of solutions by taking into account the secondary gamma lines. This procedure is repeated until the differences between the evaluated $\Sigma\sigma(\gamma,f)i\ r_i$ and the experimental value $\frac{F}{\epsilon.N.\phi}$ are < 0.1%.

The experimental errors E of the data may be written according to propagation of errors, as:

$$\sum_i \Delta^2\sigma(\gamma,f)i\ r_i^2 = \Delta^2 E$$

and the set of linear equations formed using each target can be solved in the same way as before.

The photofission cross sections obtained for the U-233 and Pu-239 nuclei are given in table III and are compared with the data of other authors in fig. 2 and fig.3.

A comparison between the present results with cross sections data obtained by unfolding bremmstrahlung spectra yields satisfatory agreement. The present data is in a reasonable agreement with the data of Katz(7), Ostapensko(4) and Shapiro(5) for U-233 and Pu-239.

The results of high resolution monoenergetic photon measurements performed with nuclear gamma rays agree with the present data for some cases but do not for others(3,6,9). The data reported by Huizenga(3) for U-233 agree with the present data. A possible structure was observed near the energy of 7.23 MeV for U-233 which is also indicate by the data 7 MeV obtained by Huizenga(3),but the lack of data in this region do not permit a definite conclusion about it.This structure was not observed by other authors. The cross sections reported by Bermann(9) tend to lie higher than ours about 30% in the region until 8.00 MeV, at higher energies the data points agree well for the U-233 nucleus. For Pu-239 nucleus the data points agree very well. There are serious discrepancies between our results and the data reported by Dragnev[6] for Pu-239 who has used a high resolution gamma radiation too.

The most remarkable feature of the present measurements was the peak found at 7.23 MeV for U-233 that may be supported by Huizengas' 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 point measured could easily coincide with peaks or valleys in the underlying fine structure of the cross section.

Several authors[10,11,18,19] have observed a structure in the photofission cross near photoneutron threshold, but unfortunately we do not have data in this region.

RELATIVE FISSIONABILITY OF U-233 AND Pu-239

The parameter relative fissionability was defined by Huizenga et al.[20] as the ratio between the fission yield obtained for a given nuclide relative to the yield obtained for the U-238 at the same excitation energy.

For the purpose of the study of this parameter,the U-238 photofission cross sections published in references[21,22] were taken. These authors were chosen because they used a gamma source type similar to the one used in this work. In reference[30] can be seen a complete comparison between the U-238 photofission cross sections obtained by several authors using several kinds of gamma sources.

The results presented by Mafra[21] and Manfredini[22] for U-238 photofission cross sections are shown in tabel IV and are similar within experimental errors. We have used mainly the data of Mafra[21] with the, exception of 7.23MeV energy,where we have used the result of Manfredini[22] that agree better with on results.

The $\sigma_{\gamma,f}$(U-233) / $\sigma_{\gamma,f}$(U-238) and $\sigma_{\gamma,f}$(Pu-239) / $\sigma_{\gamma,f}$(U-238) ratios at excitations energies coincident with those studied for U-238 are also shown in table IV.

The resulting values appear to be independent of excitation ener gy in the (6.73 - 9.72)MeV range, at least within experimental errors, excluding the result at 7.23 MeV for U-233, where a structure in the photofission cross section was obtained for this nuclei.

The mean values obtained were (2.12 ± 0.25) for U-233 and (3.32 ± 0.39) for Pu-239 meaning that U-233 and Pu-239 are more fissile than U-238 in this energy range.

The values are in excellent agreement with the results of other, workers shown in the table V.

Some authors[20] have correlated the parameter relative fissionability to the liquid droplet model parameter ($\frac{Z^2}{A}$). Our results are in excellent agreement with this correlation as can be seen in figure 4.

STUDY OF THE FISSION BARRIER FOR U-233 AND Pu-239

The experimental information obtained in this work is insufficient to specify all the parameters needed to describe the double humped fission barrier of U-233 and Pu-239. However the barrier height predicted 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 ħω. The transmition T_f of this simple barrier was calculated by Hill et al [26] and is represented by the equation:

$$T_f(E) = \{1 + \exp 2\pi (\frac{H-E}{\hbar\omega}) \}^{-1}$$

(where E is the excitation energy).

The fission barrier transmition can be expressed as:

$$T_f(E) = T_\gamma(E) \frac{\sigma_{\gamma,f}(E)}{\sigma_a(E) - \sigma_{\gamma,f}(E)}$$ for excitation energies below the photoneutron threshold.

The fission barrier transmition was obtained for U-233 and Pu-239 from the measured photofission cross sections at energy 5.43 MeV the unique result lower than the photoneutron threshold.

The total photoabsorption cross section $\sigma_a(E)$ was taken from reference[9], and the gamma ray transmission $T_\gamma(E)$ was taken from reference[26]. The expression for this transmission is semi-empirical and between 4.5 MeV and 6.5 MeV may be approximated by the equation[29].

$$T_\gamma\left(\frac{1}{2}, E\right) = 0.1 \exp \frac{(E_\gamma - 6.02)}{0.41} \quad \text{where } E_\gamma \text{ (MeV)}$$

The curvature of the barrier was taken from reference[26] by using a value for $\hbar\omega$ according to the liquid drop model.

The values of the barrier parameters determined for U-233 and Pu-239 are listed and compared with other data in table VI.

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

_EGEND OF THE FIGURES

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

Fig.2 - U-233 photofission cross sections, $\sigma(\gamma,f)$ (millibarn). The sym bols 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

The photoneutron threshold energy reaction (γ,n) is also included and was taken from ref. (28).

Fig.3 - Pu-239 photofission cross sections, $\sigma(\gamma,f)$ (millibarn). The sym bols 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

The photoneutron threshold energy reaciton (γ,n) is also included and was taken from ref. (28).

Fig.4 - Relative fissionability of several nuclei as a function of $\frac{Z^2}{A}$ liquid drop model parameter.

The data represented by, open circles were taken from ref.(20) and the data represented by a black square and the best fitted curve (dashed curve) were taken from ref.(11).

TABLE III - U-233 and Pu-239 photofission cross section as function of gamma ray energy.

Targets (Energy MeV)	U-233 $\sigma_{\gamma,f}$(mb)	Pu-239 $\sigma_{\gamma,f}$ (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	58.48 ± 17.28	131.85 ± 25.02

TABLE IV - Relative fissionability of U-233 and Pu-239. The U-238 photofission cross sections are also included.

Targets - Energy (MeV)	U-238 (21) $\sigma(\gamma,f)$mb	U-238 (22) $\sigma(\gamma,f)$mb	$\frac{\sigma_{\gamma,f}(U\text{-}233)}{\sigma_{\gamma,f}(U\text{-}238)}$	$\frac{\sigma_{\gamma,f}(Pu\text{-}239)}{\sigma_{\gamma,f}(U\text{-}238)}$
Ti - 6.73	10.40 ± 1.70+	12.50 ± 1.10	1.35 ± 0.30	2.00 ± 0.38
Mn - 7.23	3.70 ± 2.40	7.17 ± 1.50+	4.15 ± 1.02	3.74 ± 1.16
Pb - 7.38	10.20 ± 1.10+	12.60 ± 1.60	2.05 ± 0.32	3.38 ± 0.53
Fe - 7.64	10.00 ± 4.30+	12.10 ± 3.10	2.18 ± 0.99	3.76 ± 1.70
Al - 7.72	9.20 ± 2.60+	7.15 ± 0.56	2.91 ± 0.90	4.81 ± 1.51
Zn - 7.88	11.10 ± 3.40+		2.35 ± 0.79	3.82 ± 1.35
Cu - 7.91	14.30 ± 1.50+	18.9 ± 6.70	2.03 ± 0.37	2.53 ± 0.42
Ni - 9.00	37.00 ±11.00+	29.0 ± 1.80	1.97 ± 0.66	2.50 ± 0.85
		MEAN VALUES	*2.12 ± 0.25	3.32 ± 0.39

* mean value calculed without 7.23 MeV energy data.

\+ these values were used as the U-238 and Pu-239 relative fissionability calculation.

TABLE V - Comparison of relative fissionability of U-233 and Pu-239 measured by several authors

$\frac{\sigma_{\gamma,f} - \text{U-233}}{\sigma_{\gamma,f} - \text{U-238}}$	$\frac{\sigma_{\gamma,f} - \text{Pu-239}}{\sigma_{\gamma,f} - \text{U-238}}$	Energy interval MeV	Reference
2.12 ± 0.25	(3.32 ± 0.41)	6.73 -9.00	This work
2.49	2.51	12 - 20	Mc. Elhinney(24)
(2.36-2.62±4-20%)	(3.10-3.51±4-20%)	12 - 20	Huizenga (20)
(2.30)	(3.55)	5 - 20	Katz (7)
	3.00	5 - 12	Ivanof (23)

TABLE VI - Comparison of the single and double fission barriers parameters of U-233 and Pu-239 measured by several authors.

U-233		Pu-239		Reference
Ef(MeV)	hω(MeV)	Ef(MeV)	hω(MeV)	
5.6 ± 0.2	0.56	5.7 ± 0.2	0.57	This work
5.7 ± 0.3		5.8 ± 0.3		Vandenbosch (20)
		6.43 ± 0.20	1.00 ± 0.10	Back (27)
		5.50	0.55	

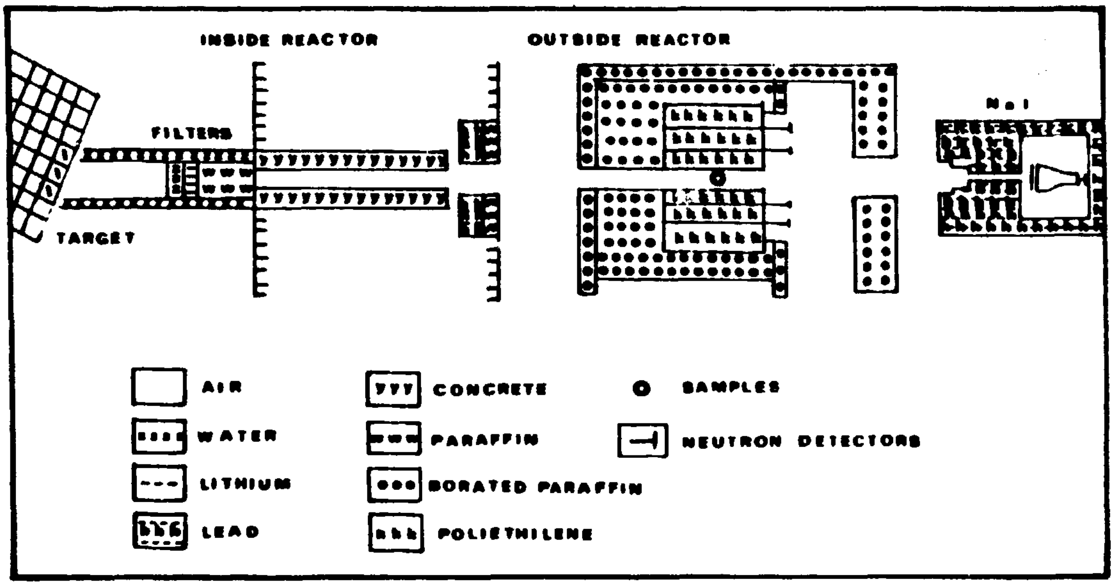

FIGURA 1

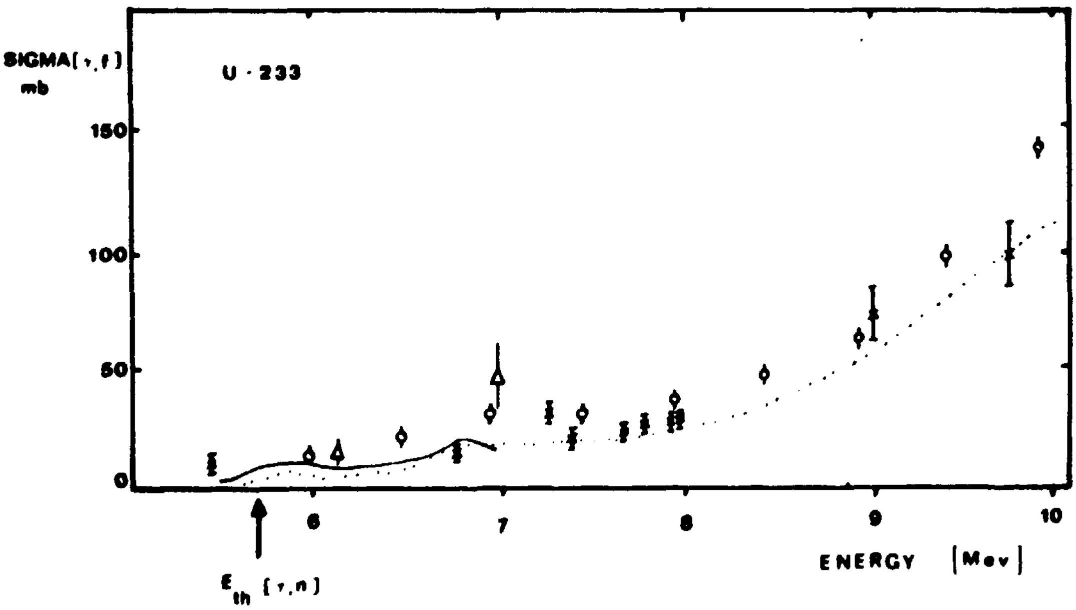

FIGURA 2

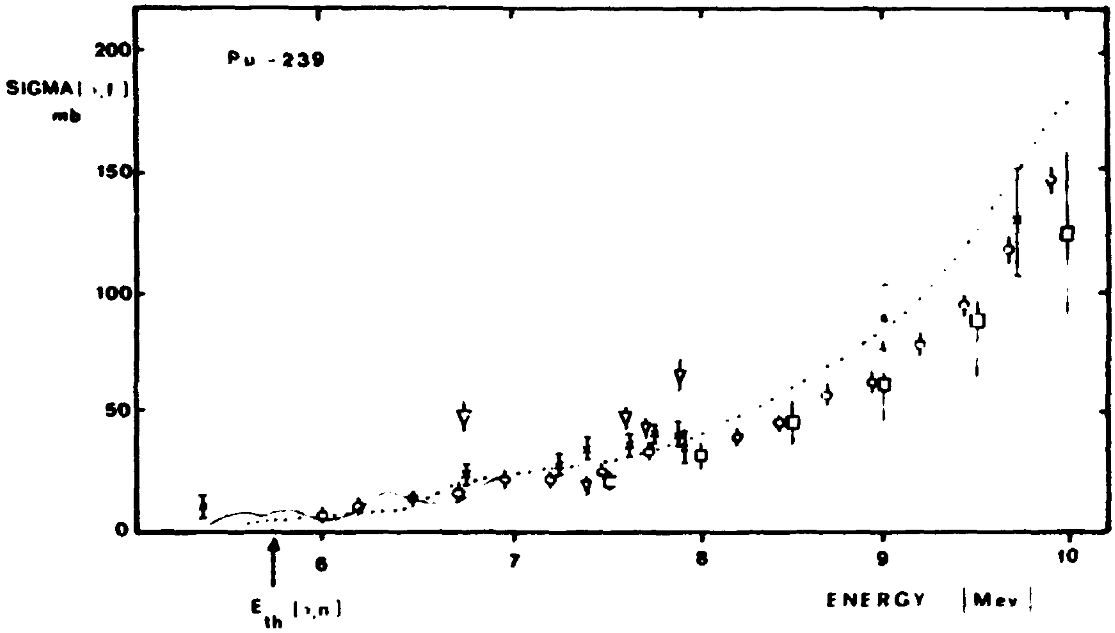

FIGURA 3

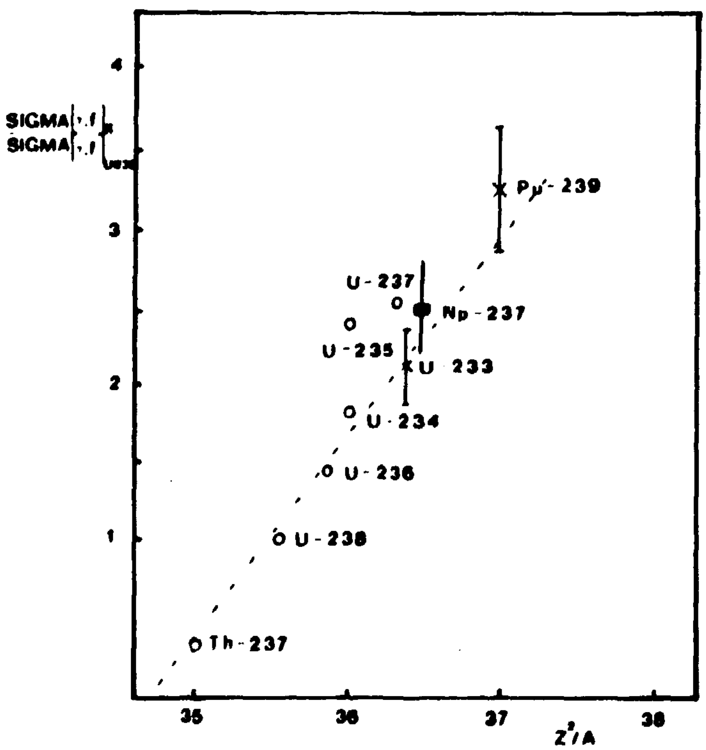

FIGURA 4

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