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Luis Paulo Geraldo, Laercio Antônio Vinhas* and Marília Teresa Freitas Cesar

DEPARTAMENTO DE FÍSICA E QUÍMICA NUCLEARES
* DEPARTAMENTO DE PROTEÇÃO RADIOLÓGICA

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ABSTRACT

The photodisintegration of ^{237}Np has been studied using monochromatic photons produced by thermal neutron capture in several materials. The partial cross sections $\sigma_{\gamma,f}$ and $\sigma_{\gamma,n}$ were measured in the energy interval from 5.43 MeV to 10.83 MeV. Analysing the photofission data according to the liquid drop model, the height (E_f) and the curvature ($\hbar\omega$) of the simple fission barrier were determined: E_f = (5.9 ± 0.2) MeV and $\hbar\omega$ = (0.8 ± 0.4) MeV. For the competition between photoneutron emission and fission (Γ_n/Γ_f) a constant value was found (1.28 ± 0.15) in the energy range 6.73 – 10.83 MeV. From this result the following nuclear temperatures for ^{237}Np were extracted on bases of some models of levels density: T = 0.84 ± 0.06 MeV (Fujimoto-Yamaguchi model) and T = 0.60 ± 0.04 MeV (Constant Nuclear temperature model).

ESTUDO DE REAÇÕES FOTONUCLEARES JUNTO AO LIMIAR PARA O ^{237}Np, COM RADIAÇÃO GAMA DE CAPTURA DE NÊUTRONS TÉRMICOS

RESUMO

A fotodesintegração do ^{237}Np foi estudada com fotons monocromáticos produzidos pela captura de nêutrons térmicos em vários materiais. As secções de choque parciais $\sigma_{\gamma,f}$ e $\sigma_{\gamma,n}$ foram medidas no intervalo de energia entre 5,43 MeV e 10,83 MeV. Analisando os dados da fotofissão com o auxílio do modelo da gota líquida, determinaram-se a altura (E_f) e a curvatura ($\hbar\omega$) da barreira de fissão simples: E_f = (5,9 ± 0,2) MeV e $\hbar\omega$ = (0,8 ± 0,4) MeV. A competição entre emissão de fotonêutrons e fissão (Γ_n/Γ_f) mostrou-se constante no intervalo de 6,73 MeV à 10,83 MeV e igual a (1,28 ± 0,15). Este resultado, analisado em termos de alguns modelos teóricos sobre densidade de níveis, fornece as seguintes temperaturas nucleares: T = 0,84 ± 0,06 MeV (modelo de Fujimoto-Yamaguchi) e T = 0,60 ± 0,04 MeV (modelo da Temperatura Nuclear Constante).

INTRODUCTION

The great number of photonuclear studies performed up to now has employed a continuous bremsstrahlung spectrum, produced by the impact of an accelerated electrons beam against a target. In these sources the gamma ray energy can be continuously changed, however, the spectrum for each energy is also continuous and it extends to the kinetic energy of the incident electron. The lack of a precise knowledge of the bremsstrahlung spectrum makes the experimental data analysis a hard and a complex task. Furthermore, the poor resolution of the gamma lines, even in the case of the bremsstrahlung monochromator (100 KeV), is not enough to resolve possible fine structures in the cross sections.

The feasibility of using reactors as an intense and discrete gamma radiation alternative source, was pointed out by Jarczyk el al(15) in 1961. Posteriorly, this technique was implanted in several laboratories where a research reactor was available. Since then, various applications and studies have been developed and a review on this subject can be found in Moreh's paper(20). In this gamma source type, used in this work, the lines widths are extremelly narrow (few eV). However the energies choice is restricted and it is not possible to measure the cross section at continuously varying gamma ray energies.

Measurements on photofission and photoneutron cross sections of ^{237}Np, employing capture gamma ray sources, had not been made so far. The few data available in the literature at the present time for these parameters, were obtained with bremsstrahlung radiation. The results have shown a marked discrepancy among the reported values and a lack of experimental information at several energies near threshold. For this reason it is important that further results should be obtained, even with the same techniques and at the same energies, in order to improve the nuclear fission data analysis near threshold for this nuclide. On the other hand, in working with capture gamma ray source, other contributions can be added such as: fine structures in these cross sections and experimental information at energies not yet explored.

EXPERIMENTAL TECHNIQUE

The discrete and monochromatic γ-lines where obtained with an experimental apparatus mounted inside a radial beam hole of the IEA-R1, 2 Mw(9) pool type research reactor. In this arrangement the γ-radiation is obtained from neutron capture in several elements used as target and placed near the reactor core, in front of the beam hole. The collimation assembly for the gamma radiation as well as the special neutron filters are similar to those employed by Young and Donahue(29) and described in detail elsewhere(9).

The gamma flux incident on the sample was measured with a 3" x 3" NaI (T1) crystal. The targets employed, the main line energy, and the γ-flux incident on the ^{237}Np sample, are listed in Table I.

Table I

Targets Used, Principal γ-Line Energies and Flux incident on the Sample

TARGETS	E (MeV)	ϕ(γ/cm^2 . s)
^{32}S	5.43	(5.1 ± 0.5) x 10^5
^{162}Dy	5.58	(2.4 ± 0.2) x 10^5
^{48}Ti	6.73	(6.1 ± 0.6) x 10^5
^{55}Mn	7.23	(5.7 ± 0.6) x 10^5
^{207}Pb	7.38	(1.9 ± 0.2) x 10^5
^{56}Fe	7.64	(6.3 ± 0.6) x 10^5
^{27}Al	7.72	(6.8 ± 0.6) x 10^5
^{64}Zn	7.88	(2.4 ± 0.2) x 10^5
^{63}Cu	7.91	(4.6 ± 0.4) x 10^5
^{85}Ni	9.00	(3.3 ± 0.3) x 10^5
^{52}Cr	9.72	(2.7 ± 0.3) x 10^5
^{14}N	10.83	(2.8 ± 0.3) x 10^4

The photofission fragments were detected by the fission track registration technique in Makrofol KG(8μ). The procedure with specific details on this technique were described in another paper(10). Using a ^{252}Cf calibrated source a total efficiency of 0.371 ± 0.011 was obtained for this technique.

Neutrons from the (γ,f) and (γ,n) reactions were detected by a 4π long counter, as by Caldwell and Dowdy(5). This long counter has 60 ^{3}He detectors, distributed in four concentric rings inside a polythene cube where the neutrons are slowed down. The detection efficiency was measured using the same ^{252}Cf source employed in the track detectors calibration. The results showed that within 15 cm around the detector central position the efficiency is practically constant and equal to 0.4331 ± 0.0014.

RESULTS AND DISCUSSION

Photofission Cross Section

The cross sections obtained for the ^{237}Np (γ,f) reaction are given in Table II, corrected already for secondary γ-lines contribution in a way similar to the one described by Mafra et al(18). The results are compared with data of other authors in Figure 1.

Table II

Photofission Cross Sections of ^{237}Np

E (MeV)	$\sigma_{\gamma,f}$ (mb)
5.43	(5.6 ± 1.0)
5.58	(8.1 ± 1.2)
6.73	(32.3 ± 5.1)
7.23	(10.4 ± 2.7)
7.38	(22.5 ± 2.6)
7.64	(19.8 ± 3.0)
7.72	(27.8 ± 3.5)
7.88	(36.4 ± 6.3)
7.91	(43.0 ± 5.1)
9.00	(41.0 ± 7.7)
9.72	(62.8 ± 16.4)
10.83	(205.0 ± 33.1)

In Figure 1 two possible structures can be distinguished: one near the photoneutron threshold (6.62 MeV) which was never observed before and another, poorly resolved, around 8 MeV. The latter shows the same trend as the structure observed by Caldwell and co-workers(6) at 8.5 MeV but, unfortunatelly, it cannot be resolved due to lack of data between 8 and 9 MeV.

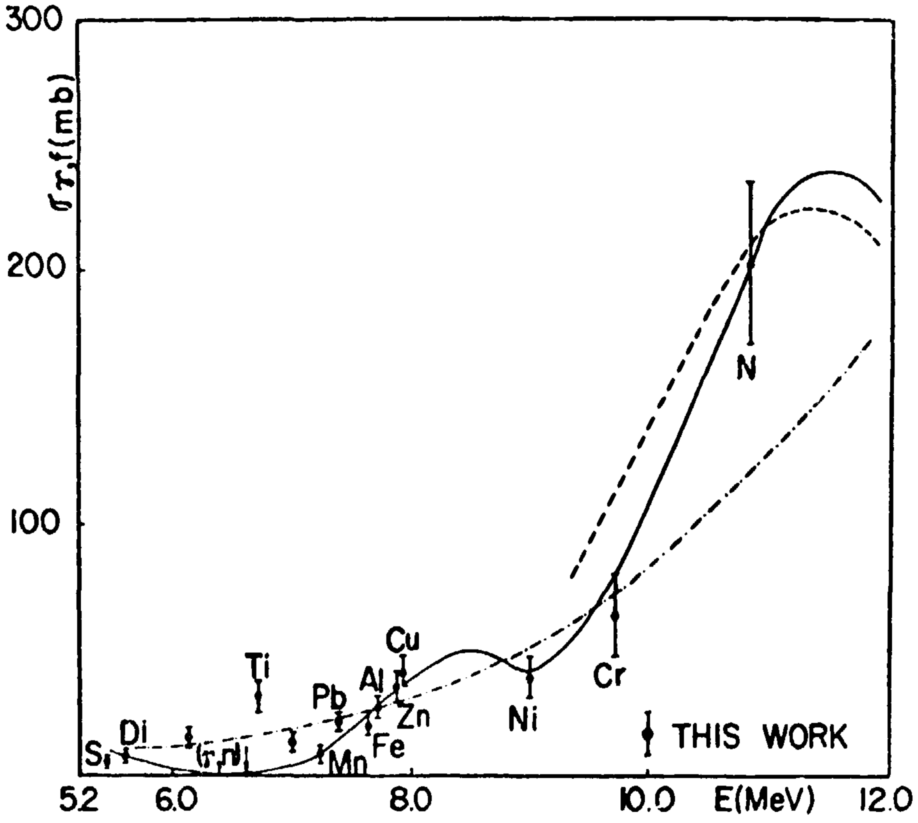

Figure 1 – Photofission Cross Sections of ^{237}Np. Solid Line Represents the Results of Caldwell et al(6), Dashed Curve Represents the Results of Veyssiere et al(25), Dashed Dot Line Represents the Results of Katz et al(16) and the open Circles Represent the Results of Clarke(7).

The peak observed near 6.62 MeV which represents the most remarkable feature of these measurements, is supported by Clarke's(7) results. Using photons produced in the F(p,αγ)O reaction, this author found that the ^{237}Np photofission cross section was lower at 7 MeV than at 6.14 MeV. This indicates the possibility of existence of a local maximum between these two energies, supporting the result of the present work. This same phenomenon had already been observed previously(21, 27) for ^{238}U and ^{232}Th nuclei and at the present moment, it is an important characteristic of the photofission cross sections for these nuclei.

The interpretation of this peak near the photoneutron threshold is still a question with a lot of controversy. The opinion on this question has been divided between those who have used the fission resonance interpretation(17, 18) and those who felt that neutron competition provided a convincing explanation(8, 12).

In the experimental results of Khan and Knowles(17) and Anderl and co-workers(2) for ^{235}U and Yester and co-workers(28) for ^{236}U, one can also observe a hint of structure at the photoneutron threshold in the photofission cross sections, although with intensity much lower than those found for the nuclei mentioned previously. With the result obtained here for ^{237}Np, it is very tempting to consider this phenomenon as being another typical characteristic of the photofission in heavy elements, beyond the giant resonance.

Relative Fissionability of ^{237}Np

The difficulties in obtaining accurate cross sections as a function of monochromatic photon energy, from experiments conducted with bremsstrahlung spectra, have led some authors to compare results between nuclides, in order to express their measurements. The most common of these comparisons which was defined by Huizenga and co-workers(13) as relative fissionability, represents the ratio between the fission yield obtained for a given nuclide relative to the yield obtained for ^{238}U at same excitation energy.

A comparative study has been performed between the results of this work, with data of other authors whom have employed the relative fissionability to express their results. For this purpose the ^{238}U photofission cross sections published by Mafra et al(18) and Manfredini et al(19) were taken. These authors were chosen because they used a gamma source type very similar to the one used in this work.

The $\sigma_{\gamma f}(^{237}\text{Np})/\sigma_{\gamma f}(^{238}\text{U})$ ratios at excitation energies coincident with those studied for ^{238}U are shown in Table III. The resulting values appear to be independent of excitation energy in the range 5.58 – 7.91 MeV, at least within the experimental error. The low value (1.1) found at 9 MeV is possibly a consequence of the structure observed near 8 MeV for ^{237}Np and which was not present in the correspondent case for ^{238}U. With this restriction, one can say that, in the 5.58 – 9 MeV range, the ^{237}Np *relative fissionability is constant* and that on the average this nuclide is 2.5 times more fissile than ^{238}U This is in excellent agreement with the results of other authors (Table IV), showing a consistency between the cross sections obtained in the present experiment for ^{237}Np and those found in refs. 18 and 19 for ^{238}U.

Table III

Relative Fissionability of ^{237}Np

E(MeV)	$\sigma_{\gamma f}$(U–238) (mb)*	$\dfrac{\sigma_{\gamma f}(\text{Np–237})}{\sigma_{\gamma f}(\text{U–238})}$
5.58	3.73 ± 0.70**	2.2 ± 0.5
6.73	10.4 ± 1.7	3.1 ± 0.7
7.23	3.7 ± 2.4	2.8 ± 1.9
7.38	10.2 ± 1.1	2.2 ± 0.3
7.64	10.0 ± 4.3	2.0 ± 0.9
7.72	9.2 ± 2.6	3.0 ± 0.9
7.88	11.1 ± 3.4	3.3 ± 1.1
7.91	14.3 ± 1.5	3.0 ± 0.5
9.00	37 ± 11	1.1 ± 0.4

* – results presented by Mafra and co-workers(18).

** – results presented by Manfredini and co-workers(19).

Table IV

Relative Fissionability of ^{237}Np Compared with Results of Other Authors

$\dfrac{\sigma_{\gamma f}(\text{Np-237})}{\sigma_{\gamma f}(\text{U-238})}$	ENERGY INTERVAL (MeV)	REFERENCE
(2.52 ± 0.31)	5.58 – 9.00	This work
(2.50 ±)	5.00 – 12.00	(14)
(2.44 ± 0.07)	12.00 – 20.00	(13)
(2.16 ±)	5.00 – 20.00	(16)

Recently, Aleksandrov and co-workers(1) observed that the independence of the ^{237}Np relative fissionability on excitation energy is not maintained at energies greater than 100 MeV. According to these results the value of $\sigma_{\gamma f}(^{237}\text{Np})/\sigma_{\gamma f}(^{238}\text{U})$ falls down to 1.9 between 100 – 240 MeV and decreases down to 1.64 between 400 – 1200 MeV. This shows a somewhat increase in the ^{238}U fission probability relative to the ^{237}Np at those excitation energies.

Study of Fission Barrier for ^{237}Np

For excitation energies below the photoneutron threshold, the neutron width Γ_n is zero and one can express the fission barrier transmission (penetrability) T_f as:

$$T_f(E) = T_\gamma(E)\,\frac{\sigma_{\gamma f}(E)}{\sigma_a(E) - \sigma_{\gamma f}(E)} \qquad (1)$$

The fission transmission $T_f(E)$ was extracted from the measured fission cross sections, using the giant resonance extrapolation from ref. 25 for the photon absorption cross section $\sigma_a(E)$, as a way to take into account the cross section $\sigma_{\gamma,\gamma'}$. The gamma ray transmission $T_\gamma(E)$ was taken from Vandenbosch and Huizenga(23) and it is based on a semi empirical expression for the dependence of the transmission on energy, on spin, and on the Fermi gas level parameter "a", normalized to experimental values obtained from resonance capture on several even-A nuclei. Between 4.5 and 6.5 MeV the transmission was approximated by the following empirical equation (based on Vandenbosch-Huizenga's study):

$$T_\gamma(1/2, E) = 0.1 \exp\left[\frac{E_\gamma - 6.02}{0.41}\right] \qquad (2)$$

where E_γ is in MeV.

With the results of this calculation for $T_f(E)$, one can determine the fission barrier parameters for ^{237}Np, using the transmission(11) of a single inverted parabolic barrier of height E_f and curvature $\hbar\omega$, such as predicted by the liquid drop model,

$$T_f(E) = \left[1 + \exp\left[2\pi\,\frac{(E_f - E)}{\hbar\omega}\right]\right]^{-1} \qquad (3)$$

The values of the barrier parameters determined for ^{237}Np are listed and compared with data of other authors in Table V.

Table V

Comparison of Single barrier Parameters of ^{237}Np

E_f(MeV)	$\hbar\omega$(MeV)	REFERENCE
5.9 ± 0.2	0.8 ± 0.4	This work
5.7 ± 0.3	0.8	(3)
5.6 ± 0.3	–	(23)

This barrier height (5.9 MeV) represents, in principle, the energy for which the penetration is equal to 1/2 for the lowest transition state ($J = 3/2$, $K = 1/2$).

In this evaluation, a single barrier penetrability was used because of the insufficient number of experimental information to specify all the parameters needed to describe a double humped barrier. The value found for E_f, however, represents the energy of the highest barrier to a good approximation.

Photoneutron Cross Section

As mentioned earlier, the neutron counting obtained with the long counter corresponds to the total neutron emission in the photonuclear processes. To this counting is associated a total neutron production cross section σ_N, such as:

$$\sigma_N (E) = \sigma_{\gamma,n} (E) + \bar{\nu}\sigma_{\gamma,f} (E) \tag{4}$$

The values determined for σ_N are listed in Table VI also corrected for secondary γ-lines contribution.

Table VI

Total Neutron Production Cross Sections of ^{237}Np

E(MeV)	σ_N(mb)
5.43	(3.0 ± 1.7)
5.58	(12.9 ± 2.2)
6.73	(100.2 ± 16.6)
7.23	(39.5 ± 10.4)
7.38	(78.5 ± 9.4)
7.64	(70.4 ± 11.1)
7.72	(97.1 ± 12.2)
7.88	(133.4 ± 25.4)
7.91	(156.5 ± 20.0)
9.00	(166.2. ± 28.9)
9.72	(261.5 ± 68.8)
10.83	(910.6 ± 212.2)

According to Eq. 4, the photoneutron cross section $\sigma_{\gamma,n}$ can be obtained by using the experimental results for $\sigma_{\gamma,f}$ and σ_N as long as the average number of prompt neutron emited per fission, $\bar{\nu}$, is nown.

Experimental data of $\bar{\nu}$ using bremsstrahlung photons with energies ranging from 8 to 13 MeV, have been published by Caldwell and Dowdy(5). The variation of $\bar{\nu}$ on excitation energy determined by these authors and employed in this work, is given by the following relationship:

$$\bar{\nu}(E) = 0.4027 + 0.2505\ E_\gamma \tag{5}$$

With the cross sections values presented in Table II and Table VI together with $\bar{\nu}(E)$ given according to Eq. 5, the photoneutron cross section were determined and the results are listed in Table VII. These results are compared with data of other authors in Figure 2.

Table VII

Photoneutron Cross Sections of ^{237}Np

E (MeV)	$\sigma_{\gamma,n}$(mb)
6.73	(32.7 ± 7.2)
7.23	(16.4 ± 4.0)
7.38	(27.8 ± 6.4)
7.64	(23.9 ± 4.5)
7.72	(32.1 ± 6.1)
7.88	(46.9 ± 10.8)
7.91	(53.9 ± 10.9)
9.00	(57.3 ± 9.8)
9.72	(88.1 ± 16.2)
10.83	(272.2 ± 171.9)

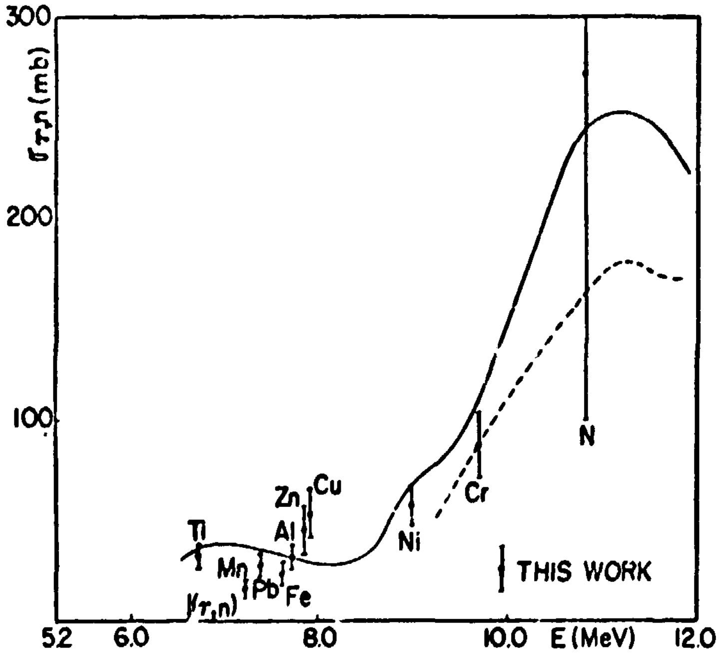

Figure 2 – Photoneutron Cross Sections of ^{237}Np. Solid Line Represents the Results of Caldwell et al(6) and the Dashed Curve Represents the Results of Veyssiere et al(25).

The photoabsorption cross section $\sigma_a(E)$, represented as the sum of the two processes, are shown in Figure 3. Some authors[6, 25] have studied the behaviour of the total photon interaction cross section for ^{237}Np in the giant resonance region (9 – 18 MeV). In order to compare the results, Figure 3 shows the extrapolations to lower energies of the measurements fit of these authors. Figure 3 also shows the curve obtained by Zhuchko and co-workers[30] representing the best fit for the heavy nuclei experimental data obtained by several authors, in the energy inteval of 6 – 10 MeV. The photoabsorption cross sections for heavy nuclei, in this interval energy, were considered as being equal.

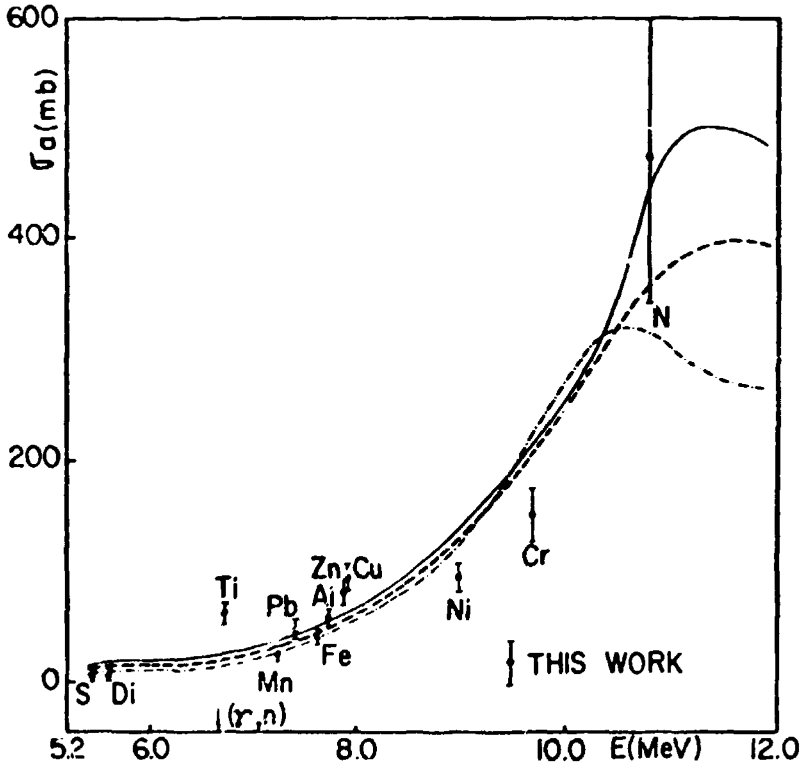

Figure 3 – Photoabsorption Cross Sections of ^{237}Np. Solid Line Represents the Results of Caldwell et al[6], Dashed Curve Represents the Results of Veyssiere et al[25] and the Dashed Dot Line Represents the Results of Zhuchko et al[30].

Despite the reasonable approximation between our results and the three curves, there is a better agreement with the fit of Zhuchko and co-workers, at least below 10 MeV. The peak obseved by us near 6.62 MeV, however, was not identified showing a possible danger in taking these extrapolations too literally.

Competition Between Neutron Emission and Fission

The competition between neutron emission and fission at energies above the photoneutron threshold may be described as:

$$\frac{\Gamma_n}{\Gamma_f} = \frac{\sigma_{\gamma,n}}{\sigma_{\gamma,f}} \tag{6}$$

The values determined for Γ_n/Γ_f ratios are listed in Table VIII. As it can be seen in this table, between 6.7 and 11 MeV the competition Γ_n/Γ_f is constant, at least within experimental error. Therefore, we adopted an average value of 1.28 ± 0.15 for the competition between the two processes. Table IX shows a good agreement between this result and those reported by other authors.

Table VIII

Values Obtained for ^{237}Np Γ_n/Γ_f Ratio

E (MeV)	Γ_n/Γ_f
6.73	1.01 ± 0.27
7.23	1.57 ± 0.56
7.38	1.23 ± 0.31
7.64	1.21 ± 0.29
7.72	1.15 ± 0.26
7.88	1.29 ± 0.37
7.91	1.25 ± 0.29
9.00	1.40 ± 0.36
9.72	1.40 ± 0.45
10.83	1.33 ± 0.86

Table IX

Comparison Between our Result for ^{237}Np Γ_n/Γ_f Competition and Those Reported by Other Authors

Γ_n/Γ_f	ENERGY INTERVAL(MeV)	GAMMA SOURCE	REFERENCE
1.28 ± 0.15	6.7 —— 10.8	neutron capture	this work
0.90 ± 0.20	9.6 —— 11.6	positron annihilation	(25)
1.27 ± 0.35	7.6 —— 11.0	positron annihilation	(6)
1.00	8.0 —— 12.2	Bremsstrahlung	(24)

It would be interesting, now, to analyze the implications of our result for Γ_n/Γ_f according to theoretical models for levels density. The theoretical expressions for Γ_n/Γ_f, which better explain this Γ_n/Γ_f behaviour, were derived from the Fujimoto-Yamaguchi and Constant Nuclear Temperature Models and are represented respectively by:

$$\frac{\Gamma_n}{\Gamma_f} = \frac{A^{2/3}T}{10} \exp\left[\frac{(E'_f - B'_n)}{T}\right] \tag{7}$$

and

$$\frac{\Gamma_n}{\Gamma_f} = \frac{2A^{2/3}T}{10} \exp\left[\frac{(E'_f - B'_n)}{T}\right] \tag{8}$$

where E'_f and B'_n are the effective threshold for the respective processes.

For ^{237}Np, which is an odd-even nucleus, $E' = E_f + \Delta_f$ and $B'_n = B_n$. In this study, a correction for E_f of $\Delta_f = 0.7$ MeV(4, 23) was used.

The fission threshold E_f for ^{237}Np was derived from the equation of Swiatecki(22). According to some authors(18, 24), this formula probably gives the best systematic threshold fission values in the actinides mass region. In this manner a value of $E_f = 5.15$ MeV was obtained for the ^{237}Np so that $E'_f = 5.85$ MeV.

The threshold for photoneutron reaction according to Wapstra and Gove(26) is 6.62 MeV and thus $B'_n = 6.62$ MeV. Using these effective thresholds in Eqs. 7 and 8, the following nuclear temperatures were necessary, in order to reproduce the competition $\frac{\Gamma_n}{\Gamma_f} = 1.28 \pm 0.15$ found in this work:

T = (0.84 ± 0.06) MeV – Fujimoto-Yamaguchi

and

T = (0.60 ± 0.04) MeV – Constant Nuclear Temperature

In these evaluations, only the Γ_n/Γ_f experimental error was considered.

Table X shows a summary of nuclear temperatures evaluated for ^{237}Np. The values are compared with those obtained by other authors in measurements performed for several heavy nuclei. An examination of this Table reveals a poor agreement among the authors. This can be explained by the different effective thresholds employed and by the fac that nuclear temperature obtained from different types of information often disagree.

Table X

Summary of Nuclear Temperatures Evaluated for Several Heavy Nuclei

T(MeV)	MODEL	NUCLEI	ENERGY (MeV)	REFERENCE
0.60 ± 0.10	Statistic	^{237}U	9.5 —— 11.5	(25)
0.90	Constant temperature	^{238}U	6.0 —— 9.0	(18)
1.40	Constant temperature	^{232}Th	6.0 —— 9.0	(18)
0.40	Constant temperature	several	several	(23)
0.60 ± 0.04	Constant temperature	^{237}Np	6.7 —— 10.8	this work
0.60	Fujimoto-Yamaguchi	several	several	(24)
0.84 ± 0.06	Fujimoto-Yamaguchi	^{237}Np	6.7 —— 10.8	this work
1.35	Neutron evaporation	several	several	(24)

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