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Section A

A new experimental apparatus for production and utilization of capture gamma rays

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This paper describes an experimental apparatus mounted at a tangential beam tube of the IPEN IEA-R1 research reactor, for production and utilization of capture gamma-rays. By changing the target material, it was possible to produce up to 30 gamma-ray lines in the 5 to 11 MeV energy range providing at least 18 new energies to be explored in photonuclear reactions studies, besides those usually obtained with similar gamma ray sources. The gamma-ray flux density produced by each target was measured with a Ge(Li) spectrometer. The efficiency calibration for this detector was performed, in the 5 to 11 MeV energy interval, using samples of nickel and nitrogen as reference standards. Least square fitting methods and covariance matrix have been applied in the experimental data analysis.

1. Introduction

Photonuclear reactions near threshold (5–11 MeV) have been pointed out [1,2] as an excellent means of studying physics of low energy fission, especially because of the restricted angular momenta in the entrance channel (mainly produced by dipole (E1) and quadrupole (E2) photoabsorption) and the few decay modes of the compound nucleus basically (γ, γ'), (γ, f) and (γ, n).

During the last 40 years, a number of photonuclear experiments, in the actinide region, have been performed using bremsstrahlung beams (resolution higher than 10%; bremsstrahlung monochromator, annihilation in flight of positrons, Compton scattered neutron capture gamma-rays (1 to 5% of resolution); and monoenergetic gamma radiation from neutron capture reactions (resolution of few eV). However, there are still few data available in the literature on photonuclear cross sections for several actinide nuclei and large discrepancies are apparent among the reported values, mainly in the energy region near threshold (5–11 MeV) (see for example refs. [3,5,7]. Consequently, it is expected that new experimental information on these parameters be still relevant not only in eliminating the discrepancies mentioned above but also to better understand the fission phenomenon at low excitation energies.

A systematic investigation of photonuclear reactions for some actinide nuclei has been undertaken [4–8] in this laboratory using monochromatic photons from neutron capture. However, the small number of experimental data (lower than 12) obtained in all experiments has not been appropriated neither to extract safe information on fission barrier parameters nor to resolve possible structures in the photonuclear cross sections.

The main purpose of the present work was the construction and calibration of an experimental arrangement at IEA-R1 reactor, in order to be able to utilize up to 30 capture gamma-rays lines, with discrete energies ranging from 5 to 11 MeV, for photonuclear reactions studies.

2. Experimental arrangement

The experimental apparatus used to produce the discrete and monochromatic gamma-rays has been mounted at the BH4-12 tangential beam hole of the IPEN IEA-R1 research reactor (2 MW) and is described in detail elsewhere [9]. In summary, gamma radiation with energy in the 5.265 to 10.829 MeV interval is produced from thermal neutron capture in different target materials, when they are placed near the reactor core, in the center of the tangential channel. At a maximum power of 2 MW, the flux density of thermal neutron at the target irradiation position amounted to 6.2×10^{11} n/cm^2 s. The changes of the thermal neutron flux in this position has been monitored by means of a self-powered neutron detector (SPND) with silver converter, which was installed in-

side the tangential beam tube, very close to the capture target. In order to obtain proper collimation of the gamma beam, two lead collimators were used. The first one, the main collimator with 250 cm length, was placed in the tangential tube and yielded a parallel beam of 3 cm diameter at the outside of the reactor wall, approximately 1 m before the sample (actinide nuclei) irradiation position. The second collimator, 50 cm length, which was installed 473 cm away from the reactor wall yielded a parallel beam having a diameter of 6 mm and was employed for gamma flux density measurements with a Ge(Li) spectrometer.

The background of neutrons at the sample irradiation position was carefully studied by optimizing the signal to background ratio. The best neutron filters combination found was: 22 cm polythene, 34 cm polypropilene and 9.5 cm borated paraffin. With this neutron filters combination in the beam path, the neutron flux measured with a 4π long counter detector [5] was about 30 n/s.

The target materials chosen for this work are listed in table 1 along with the mass employed and the measured gamma-ray lines with respective intensities (I_γ). These gamma radiations are not necessarily the main gamma ray lines emitted by the neutron capture targets but the ones representing the most convenient for gamma ray spectroscopy and/or for photonuclear studies. The targets were constructed with the same geometry by packing the materials inside aluminum containers having the following dimensions: 18 cm length, 7.6 cm diameter and 1.27 mm thickness.

Due to the relatively large number of targets employed in this experiment, it was necessary to construct a lead chamber for safe storage of these materials after irradiations. This chamber has been mounted near the beam tube entrance, where the targets are placed (or taken off) for irradiation.

As can be seen in fig. 1 the chamber has a lead wall of 10 cm thickness and an appropriated window of glass–lead alloy that allows the visualization inside the chamber. With a kind of manual elevator it is possible to store the targets in a shelf containning 30 compart-

Table 1
Characteristics of the target materials employed in this work and the results obtained for the γ-ray flux density calculations

Target element	Target compound	Mass [g]	Measured γ-ray energy [keV]	I_γ Ref. [17]	γ-ray flux γ/cm² s
Yb	Yb_2O_3	1410	5265.70	5.78	$(1.76\pm0.13)\times10^4$
K	K_2CO_3	1260	5380.63	8.90	$(1.91\pm0.14)\times10^4$
S	solid (α)	1500	5420.50	59.08	$(7.68\pm0.55)\times10^4$
Dy	Dy_2O_3	200	5607.30	2.93	$(1.08\pm0.08)\times10^4$
Hf	HfO_2	1400	5723.50	2.18	$(2.62\pm0.29)\times10^3$
Cd	metal	620	5823.90	2.13	$(8.07\pm0.80)\times10^3$
In	metal	1580	5891.90	0.60	$(2.51\pm0.17)\times10^3$
Y	Y_2O_3	1500	6079.80	77.49	$(8.25\pm0.52)\times10^4$
Er	Er_2O_3	1200	6228.60	0.89	$(2.60\pm0.16)\times10^3$
Na	Na_2CO_3	1120	6395.40	22.18	$(2.21\pm0.13)\times10^4$
Ca	$Ca(OH)_2$	1500	6419.90	38.89	$(2.35\pm0.15)\times10^4$
Hg	HgO	1700	6457.50	5.20	$(1.77\pm0.11)\times10^4$
Nd	Nd_2O_3	1780	6501.70	8.15	$(4.16\pm0.25)\times10^4$
Sm	Sm_2O_3	610	6538.30	0.25	$(7.85\pm0.81)\times10^2$
Gd	Gd_2O_3	520	6748.70	2.25	$(6.16\pm0.37)\times10^3$
Ti	metal	2280	6750.78	24.17	$(3.32\pm0.19)\times10^5$
Be	BeO	1500	6809.41	63.75	$(3.67\pm0.22)\times10^3$
V	V_2O_5	1440	6873.90	10.73	$(4.73\pm0.28)\times10^4$
As	As_2O_3	1920	7019.45	2.47	$(1.11\pm0.07)\times10^4$
Mn	metal	3000	7057.81	11.06	$(7.61\pm0.44)\times10^4$
Pb	metal	5000	7367.70	94.06	$(1.03\pm0.06)\times10^4$
Cl	$MgCl_2$	1170	7413.80	10.42	$(5.50\pm0.03)\times10^4$
Se	SeO_2	1320	7418.70	3.12	$(1.62\pm0.10)\times10^4$
Fe	metal	2330	7638.29	26.41	$(2.03\pm0.11)\times10^5$
Al	metal	2260	7723.85	27.43	$(4.54\pm0.25)\times10^4$
Zn	metal	3560	7863.30	10.58	$(3.03\pm0.17)\times10^4$
Cu	metal	3060	7914.50	30.82	$(6.65\pm0.37)\times10^4$
Ni	metal	3620	8998.80	37.74	$(1.11\pm0.07)\times10^4$
Cr	metal	2670	9720.30	26.97	$(3.54\pm0.24)\times10^4$
N	$C_3N_3(NH_2)_3$	1500	10829.18	14.12	$(2.42\pm0.19)\times10^3$

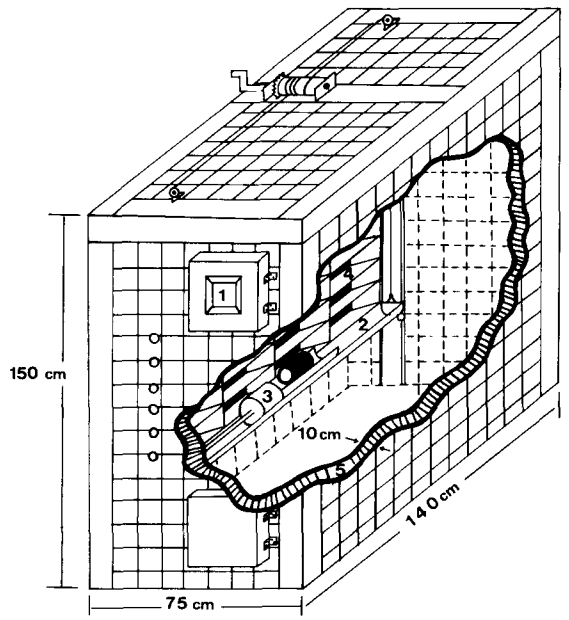

Fig. 1. Lead chamber for target storage. (1) Lead glass alloy window. (2) Manual elevator. (3) Target carier. (4) Target compartments. (5) Lead wall.

ments. The targets are put in (or out) the chamber through a rectangular hole located at the upper part of the left wall.

3. Results and discussion

Gamma flux density measurements for all targets listed in table 1 were carried out by counting the emitted prompt gamma-rays with a Ge(Li) detector (5%, 25 cm^3, ORTEC). Determination of an appropriate counting efficiency for the configuration of the present work, was an essential part of the experiment. Details concerning the method employed are well documented elsewhere [9,10] and it will not be described here in any depth. In summary, the relative efficiency calibration for the Ge(Li) detector has been performed, in the energy interval from 4857.33 to 10829.18 keV, using samples of nickel (50.5 g) and nitrogen (melamine 114.2 g) as reference standards in a similar way as employed in refs. [11–14]. These samples produce several capture gamma rays lines, in all energy range of interest, with reasonably well known (< 6.8%) emission probabilities.

The efficiency for each selected energy $\varepsilon_f(E_\gamma)$ has been determined by the following equation:

$$\varepsilon_f(E_\gamma) = \frac{\text{AREA}}{N\sigma\phi I_\gamma \Omega t}, \qquad (1)$$

where, AREA is the photopeak counting, N the number of nuclei for a particular isotope, σ the capture cross section, ϕ the thermal neutron flux density, I_γ the gamma-ray emission probability, Ω is the solid angle, and t the counting time.

The set of gamma-ray lines selected for the efficiency calibration are listed in table 2 along with the relevant nuclear data.

The thermal neutron flux impinging on the nickel or nitrogen sample has been measured by the gold foil activation technique [15]. In order to better handle the systematic uncertainties in neutron flux calculation, two gold foils placed on opposite sides of each sample were irratiated.

From the measured spectra in a multichannel analyser system (PCA-8000 from Nucleus Inc.) the photopeak areas of the selected gamma-rays lines were determined by using the proper software from PCA-8000 for isolated peaks or a Fortran program named ANALYSIS [9] for peaks which were not well resolved. This program has been written to analyse peaks using a Gaussian shape with exponential tail in the low and

Table 2
Gamma-ray lines selected from the standard samples

Isotope	Cross section [b] Ref. [18]	Isotopic fraction Ref. [19]	Energy [keV] Refs. [14,20]	I_γ Refs. [12,14,20]
^{58}Ni	4.6 ±0.3	68.27	4858.85	1.53 (04)
^{14}N	0.075 ± 0.0075	99.634	5533.38	19.75 (21)
^{14}N	0.075 ± 0.0075	99.634	6322.34	18 67 (14)
^{58}Ni	4.6 ±0.3	68.27	6583.65	2.63 (06)
^{62}Ni	14.5 ±0.3	3.59	6838.16	85.9 (59)
^{60}Ni	2.9 ±0.2	26.10	7820.25	54.8 (25)
^{58}Ni	4.6 ±0.3	68.27	8121.09	4.56 (10)
^{14}N	0.075 ± 0.0075	99.634	8310.14	4.22 (05)
^{58}Ni	4.6 ±0.3	68.27	8534.21	25.14 (56)
^{58}Ni	4.6 ±0.3	68.27	8999.18	52.71 (116)
^{14}N	0.075 ± 0.0075	99.634	10829.09	13.65 (21)

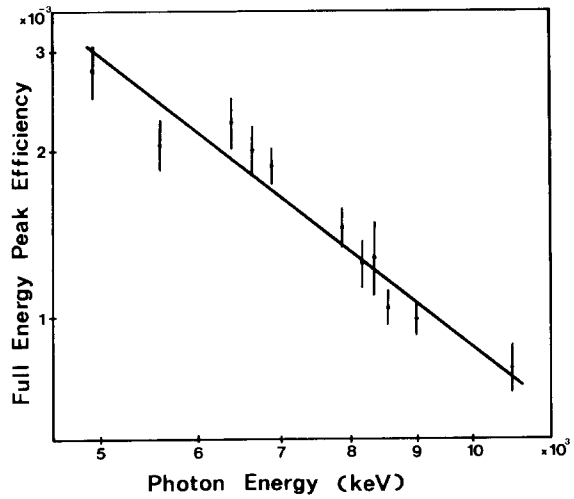

Fig. 2. The full energy peak efficiency for the Ge(Li) detector.

high energy side and an optional function (step, linear or second degree) for the background. The final counting for each analysed photopeak has been obtained after correction for pileup effects, self-absorption, counting dead time and attenuation by the neutron filters.

The known sources of experimental error considered in the efficiency calibration and the range of the magnitudes involved are: peak area 0.35 to 11.6%, capture cross sections 2.1 to 10%, thermal neutron flux 2.0% and gamma ray emission probability 0.75 to 6.9%. These error components were employed to generate the covariance matrix for the gamma-ray detector calibration data [16]. As is apparent the uncertainties in the capture cross sections have represented one of the most important contribution for the total error.

Fig. 2 shows a plot of the full energy peak efficiency (ε_f) as a function of the photon energy (E_γ). The following empirical expression [10]

$$\ln(\varepsilon_f) = \sum_{i=1}^{n} p_i (\ln E_\gamma)^{i-1}, \qquad (2)$$

has been fitted to calibration data using least squares fitting and the best curve obtained was for $n = 2$ (χ^2 normalized $\simeq 1$). The results of the two parameters fit, represented by the solid line in fig. 2, are as follow: $p_1 = 8.9436$ ($\pm 14.1\%$), $p_2 = -1.7337$ ($\pm 8.1\%$) and the parameter error correlation $= -0.99$.

The gamma-ray flux density calculations obtained for each target are listed in table 1 together with the resulting total errors. The partial sources of experimental error which were considered in these calculations are the following: gamma-ray counting statistics 0.3 to 9.0%, collimator area determination 0.7% and efficiency calibration for Ge(Li) detector 5.4 to 7.5%.

4. Conclusions

This work demonstrates that the present experimental apparatus for capture gamma-ray production may be an excellent tool for studying the physics of low energy fission when compared with similar gamma-ray sources.

The utilization of a high energy resolution technique for gamma-ray spectra analysis allowed the inclusion of other target materials and at least 18 new gamma-ray lines may be now explored for photonuclear reactions studies at threshold energies.

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References

[1] B.S. Bhandari and I.C. Nascimento. Nucl. Sci. Eng. 60 (1976) 19.
[2] S. Bjornholm and J.E. Lynn. Rev. Mod. Phys. 52 (1980) 725.
[3] V.V. Varlamov, B.S. Ishkhanov, M.E. Stepanov, V.V. Surgutanov, A.A. Khoronenko and A.P. Chernjaer, Photofission of Nuclei (Institute of Nuclear Physics of MSV, 1989).
[4] O.Y. Mafra, S. Kuniyoshi and J. Goldemberg. Nucl. Phys. A 186 (1972) 110.
[5] L.P. Geraldo, L.A. Vinhas and M.T.F. Cesar. Nucl. Sci. Eng. 89 (1985) 150.
[6] L.P. Geraldo, J. Phys. 12 (1986) 423.
[7] M.A.P.V. Moraes and M.T.F. Cesar, Nucl. Instr. and Meth. A 277 (1989) 467.
[8] L.P. Geraldo, M.T.F. Cesar and M.A.P.V. Moraes, Nucl. Sci. Eng. 110 (1992) 128.
[9] R. Semmler, M. Sc. Thesis University of São Paulo (1993).
[10] L.P. Geraldo and D.L. Smith, Nucl. Instr. Meth. A 290 (1990) 499.
[11] H. Seyfarth, A.M. Hassan, B. Hrastnik, P. Gottel and W. Delang, Nucl. Instr. Meth. 105 (1972) 301.
[12] A.W. Carbonari and B.R.S. Pecequilo, Proc. Int. Conf. Nucl. Data for Science and Technology, ed. S. Igarasi (1988) p. 133.
[13] International Atomic Energy Agency, IEA-TECDOC-619 (Vienna 1991)p. 24.
[14] A.F.M. Ishaq, A. Robertson, W.V. Prestwich and T.J. Kenett, Z. Phys. A 281 (1977) 365.

[15] L.P. Geraldo, M.S. Dias and M.F. Koskinas, Radiochim. Acta 57 (1992) 63.
[16] D.L. Smith, Report ANL/NDM-62 (Argonne National Laboratory, 1981).
[17] M.A. Lone, R.A. Leavitt, D.A. Harrison, At. Data and Nucl. Data Tables 26 (1981) 511.
[18] S.F. Mughabghab, M. Divadeenam and N.E. Holden, Neutron Cross Sections, vol. 1 (Academic Press, New York, 1981).
[19] R.C. West, CRC Handbook of Chemistry and Physics (Boca Raton, CRC, 1990).
[20] T.J. Kennett, W.V. Prestwich and J.S. Tsai, Nucl. Instr. Meth. A 249 (1986) 366.