AN EXPERIMENTAL FACILITY FOR THE PRODUCTION OF REACTOR NEUTRON CAPTURE GAMMA-RAYS

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AN EXPERIMENTAL FACILITY FOR THE PRODUCTION OF REACTOR NEUTRON CAPTURE GAMMA-RAYS

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RESUMO

Num dos canais transversais do IEAR-1, foi construido um sistema colimador para a radiação gama emitida por núcleos atômicos, após captura de neutrons térmicos[1].

As energias disponíveis variam de 4 a 10 MeV discretamente.

Obtemos com êsse arranjo, acoplados: alta intensidade, boa colimação e energias monocromáticas.

RÉSUMÉ

Un système pour la collimation des rayons gamma émis par des noyaux atomiques après la capture de neutrons thermiques, a été construit dans un des tubes du reacteur à piscine IEAR-1 de cet Institut[1].

Les énergies des rayons gamma disponibles varient entre 4 et 10 million de électron-volts, discrétement.

Le système construit permet l'obtention des lignes gamma monoenergétiques associés à des conditions d'une haute intensité et bonne collimation.

ABSTRACT

In order to obtain a collimated gamma-ray beam from excited nuclei formed by neutron radioactive capture, an experimental device was installed in one of the beam through tubes at the IEAR-1 swimming pool reactor in this Institute.

in order obtain monochromatic gamma rays of high intensity and well collimated, several targets are used in order to cover the range from 4 to 10 MeV.

I - INTRODUCTION

Photonuclear reactions have been studied in recent years through the use of gamma radiation obtained from various sources. Amongst those sources, special reference is made to the use of monochromatic gamma-rays obtained from either natural or artificial radioactive sources, from charged particles capture radiation, annihilation of positrons in flight or from continuous Bremsstrahlung radiation produced in betatrons and synchrotrons; the obtained radiation, in each case, presents some advantages or disadvantages, when compared with each other, in important characteristics such as intensity, energy, width of the line and collimation, as will be inferred from the following considerations.

The gamma radiation emitted by radioactive sources are monochromatic and their intensity depends only on the strength of the source; although the line width is small, the available energies are small and their use for photodesintegration studies is restricted to those nuclei whose neutron binding energy is smaller than the gamma-ray energies available.

The capture gamma radiation from charged particles reactions, such as $F^{19}(p,\alpha,\gamma)O^{16}$ and $Li^{7}(p,\gamma)Be^{8}$ is monochromatic and energies up to about 20 MeV are available; since however their intensity is low, it becomes very difficult to obtain well collimated beams of adequate intensity - even when the distances between the source and the target are of the order of 30 cm.

Most of the studies on the nuclear photoeffect have been made with the continuous Bremsstrahlung spectrum from betatrons. Although the energy of the electrons accelerated in those machines can be known with a high degree of precision, the photon energy distribution in the continuous spectrum cannot be easily decomposed in energy bands of a sufficiently narrow width in order to allow a precise knowledge of the number of photons in those energy intervals. To this difficulty, which arises from the finite width of the targets used for the Bremsstrahlung production another factor, due to the shape of the spectrum which shows a relatively small number of photons near to the tip of the spectrum as compared with the large number of quanta in the low energy regions: consequently, the analysis of the observed experimental data is tedious and rather complex. The procedures used to sort out the relevant cross-sections from the observed excitation functions involve small differences between large numbers - a procedure which gives rise to rather large uncertainties and to errors which are often difficult to be estimated.

In the late years the use of gamma-rays from the annihilation of positrons in flight become widely used and the available energies reach values of several tens of MeV. Positrons are obtained through the pair production in a heavy target bombarded by Bremsstrahlung radiation at the center of a two-stage linear accelerator.

The obtained positrons are further accelerated and selected by a magnetic analyzer. The positron beam thus obtained, impinges on a thin target of a low atomic number material and are annihilated in flight with the target electrons. A beam of monochromatic gamma-rays is thus obtained with a continuously variable energy, which can be obtained through a variation of the positron energies.

The main disadvantages of such a system are its low intensity and the existence of a Bremsstrahlung background.

The last method referred to above requires a strong thermal neutron flux, such as produced by a reactor, and was installed in one of the IEAR-1 swimming pool reactor beam through tubes.

Monochromatic gamma-rays are produced by thermal neutron capture in several substances used as targets and located near to the center of the beam tube, where they face the reactor core and are submitted to a strong neutron flux. In this way it is possible to obtain a high intensity gamma-radiation under low background and excellent collimation.

The gamma-radiation emitted by nuclei which have captured neutrons is limited to a narrow energy range (between 4 and 11 MeV): it is fortunate that the thresholds for several important reactions lay in this energy interval so that several phenomena of interest can be studied.

II - EXPERIMENTAL ARRANGEMENT

The collimator was installed in the upper through tube of the IEAR-1 reactor, as shown in figure 1. A through tube (transversal irradiation channel) was chosen in order to avoid the high gamma-ray and fast neutron background from the radial tubes which "see" the core; besides, the operations of loading and unloading targets are considerably simplified since they can be carried out without disturbing the collimators and detecting equipment.

The thermal neutron distribution along the length of the tube was measured with gold foils in order to find out the position of maximum flux to locate the target. The thermal flux at the best position is $(3.63 \pm 0.07) \times 10^{11}$ neutrons/cm^2/sec. at a nominal power of 2 MW. All the main details of the flux measurements are presented in Appendix I.

There is a definite advantage of placing the target at the center of the tube and getting the gamma-ray beam at its end, instead of collimating a neutron beam through the irradiation tube and letting it impinge on the target: in the first case, besides the elimination of the gamma-rays from the core and of the background due to their scattering, there is a net advantage in collimating the beam through 4 m of distance.

The distance between the target and the first collimator results from a compromise of several factors such as the maximum weight which can be placed at the center of the through tube, the problems of gamma-ray heating of the target and the background produced by neutron irradiation of the collimators. The determination of the distance between the target and collimator, length of the collimator, diameter of the source and of the collimator were calculated taking into account the collimator transmission. Such calculations were made for several parameters of interest.

The collimator finally used is 4 m long and has a diameter of 5 cm. Filters of boric acid mixed with paraffin and plastic and boric acid remove both the fast and the thermal neutrons making their background low enough in order not to disturb the experiments.

The length and the location of the filters of paraffin mixed with boric acid and plastic mixed with boric acid were determined experimentally. The total background of both thermal and fast neutrons outside the channel was measured through a physical integration in a manganese sulphate solution and its absolute activity was obtained by intercalibration. This background is smaller than 600 $n/cm^2/sec$ which is the sensitivity of measurement (Appendix II).

In order to maintain the reproductibility of the position of the collimators, wood spacers were used. Outside the reactor biological shield, the collimator shield shows an increase in diameter in order to avoid radiation channeling between the through tube and the collimator plug.

About 10 different targets can be used (Table I) if the choice criterion used is that the targets should show one gamma line of a higher intensity than the other (lines) and that the separation between them is at least 0.5 MeV; besides, the targets should present a low scattering cross section.

It is quite obvious from the presenting remarks that the spectra used are not rigorously monoenergetic. The gamma rays of smaller energy than the main line contribute as a background but when their energy is well below the main line their effect can be either discriminated experimentally or through the threshold of the process under study.

The reactor background, that is, the gamma rays due to the fission products, to the fission process itself, to the capture and activation of structural materials or even of the target activation, become important for energies below about 3 MeV as can be seen from figure 2.

In order to avoid the contribution of gamma radiation from the (neutron, gamma) process in the aluminium tube which surrounds the collimator (through tube) the collimator angle was chosen in such a way that only points from this tube situated far away from the collimator port are "seen".

A special care was also taken to avoid the presence of any material around the target that might give rise to undesirable neutron or gamma background. In this way it was possible to obtain a high ratio between the intensity of the gamma lines of interest and the background.

The targets are placed at a suitable position, without disturbing the collimator arrangement, by means of a simple device which is introduced in the through tube through its opening opposite to the collimator port. The samples used weight between 1 and 2 kilograms; powder or small pieces of target material are packed inside a magnesium container, whenever a solid sample is not available. The advantage of using a magnesium cladding is that its main gamma contribution consists on a low energy line which lies in the background region.

The targets used are of cylindrical shape and, in order to avoid flux depression, their length is chosen in such a way that it is always smaller than the neutron mean free path in the material.

The intensity of the gamma-ray lines outside the channel were calculated by taking into account the collimator transmission and the absorption of the intensity due to the presence of paraffin and boric acid.

Under these assumptions, one can write:

$$\phi_\gamma S = \phi_n \frac{N_0}{A} \sigma_Q \frac{M}{\rho} G f \quad \text{photons/sec}$$

where $S = 19.63\ cm^2$, ϕ_n is the neutron flux, N_0 is the Avogadro's number, A is the mass number of the target, ρ is the density of the target, σ_a is the absorption cross section, G is the collimator transmission and f is the factor of absorption in boric acid and paraffin (80% absorption).

$$\phi_\gamma S = 2.11 \times 10^{11} \left(\frac{\Omega \sigma}{A}\right) M G f$$

$$\phi_\gamma = 1.07 \times 10^{11} \left(\frac{\Omega \sigma}{A}\right) M G f$$

The obtained results are presented in Table II.

The energies available from the targets used are known with precision; tabulated data was used[2].

The alignement of the collimators was checked by mean of gamma radiography from the beam, by inserting at the collimator port end, a small collimator, made of concentric cylinders of lead and aluminium, with a total length of 15 cm. The diameter of the outermost cylinder was 5 cm; since aluminium has a smaller absorption coefficient than lead, concentric rings, alternatively black and white, are obtained under gamma-rays; a measurement of the optical density provides the required information for a perfect alignement (figure 3).

The shape of the beam can be crudely estimated by using a NaI(Tl) 2" x 2" scintillator provided with a 1/8" diameter collimator and displacing it transversally to the beam. The obtained curve is represented in figure 4.

The spectra and the intensities of the lines were determined with a NaI(Tl) 3" x 3" gamma-ray scintillation crystal coupled to a TMC 1024 channel analyser. The crystal coupled to the photomultiplier tube is installed inside a lead shield in order to eliminate the contribution of the room background and the gamma-rays from the target which could be scattered. The details of the arrangement are represented in figure 5.

The collimator used for that purpose is 20 cm long and presents a diameter of 1/4" in order that the gamma-ray beam will impinge on the central region of the crystal; with this precaution, the resolution is improved since the probability of escape decreases. In order to avoid the scattering of the gamma radiation by the collimator in order to assure that only gamma-rays parallel to the collimator axis will reach the crystal, special care was taken in the alignement of both the outside beam collimator and the crystal collimator. Besides aligning those collimators geometrically, special care was taken to assure that the target was properly aligned in order that the maximum gamma-ray intensity would be obtained.

The intensities of the lines corresponding to the targets used were computed from the areas under the total absorption peaks whose shape was assumed as gaussian.

The photopeak efficiency, that is, the ratio of the area under the photopeak and the area corresponding to the whole response of the system is obtained by a Monte Carlo process[3].

In this process of computation, the history of a photon and of all its secondary radiations is simulated. For each incident gamma-ray, the total quantity of energy which is absorped by the crystal is computed and a pulse is registered in the channel corresponding to that energy.

The details of the intensity determination using the Monte Carlo process will be published shortly.

Figure 6 shows the spectrum obtained with a nickel target, where the principal line and the escape peaks and other additional lines are readily observable. An approximate calculation of the intensity of the nickel gamma-rays gives a value of 6.10^5 photons/cm^2/sec, in good agreement with the calculated values.

In order to avoid the variation of the gamma-ray intensity with any fluctuation of the reactor power, a monitor was used all the time: the monitor used was a BF_3 counter ("microneutron") introduced in a through tube which is quite close to the beam tube which contains the target.

TABLE I

Targets	Energy (MeV)	σ_a Q (barn)	σ_a Q/ σs_γ
Si^{28}	3.54	9.6×10^{-2}	6.9×10^{-3}
	4.93	12	8.4
Mg^{24}	3.92	3.2	2.7
C^{12}	4.95	0.26	0.43
S^{32}	5.43	31	19
Y^{89}	6.07	59	15
Ca^{40}	6.42	17.5	8.0
Ti^{48}	6.75	238	100
Be^{9}	6.82	0.75	1.9
Pb^{207}	7.38	15.8	19
Fe^{56}	7.64	76	29
Al^{27}	7.73	4.8	3.7
Ni^{58}	9.00	119	43
Cr^{54}	9.72	11.5	6.7
N^{14}	10.83	0.88	1.3

Q is the number of photons corresponding to 100 neutrons captured[(2)]

TABLE II

Elements	Q σ/A	φ γ/MGf	φ γ/MG	G	φ γ/M	M (grams)
C^{12}	2.16×10^{-4}	2.41×10^{6}	4.82×10^{5}	8.25×10^{-6}	3.97	1.230×10^{3}
Be^{9}	8.32×10^{-4}	9.28×10^{6}	1.855×10^{6}	8.45×10^{-6}	1.57×10	7.20×10^{2}
S^{32}	0.97×10^{-2}	1.08×10^{8}	2.16×10^{7}	9.75×10^{-6}	2.11×10^{2}	5.00×10^{2}
Pb^{207}	7.62×10^{-4}	8.50×10^{6}	1.699×10^{6}	9.75×10^{-6}	1.66×10	1.000×10^{3}
Fe^{56}	1.36×10^{-2}	1.51×10^{8}	3.02×10^{7}	8.40×10^{-6}	2.54×10^{2}	2.700×10^{3}
Al^{27}	1.78×10^{-3}	1.98×10^{7}	3.96×10^{6}	7.95×10^{-6}	3.15×10	2.720×10^{3}
Ni^{58}	2.05×10^{-2}	2.29×10^{8}	4.57×10^{7}	8.75×10^{-6}	4.00×10^{2}	1.500×10^{3}
Cr^{54}	2.87×10^{-3}	3.20×10^{7}	6.40×10^{6}	9.75×10^{-6}	6.24×10	1.000×10^{3}
N^{14}	6.29×10^{-4}	7.01×10^{6}	1.403×10^{6}	8.25×10^{-6}	1.16×10	8.08×10^{2}
Ti^{48}	4.95×10^{-2}	5.52×10^{8}	1.104×10^{8}	8.8×10^{-6}	9.71×10^{2}	1.000×10^{3}

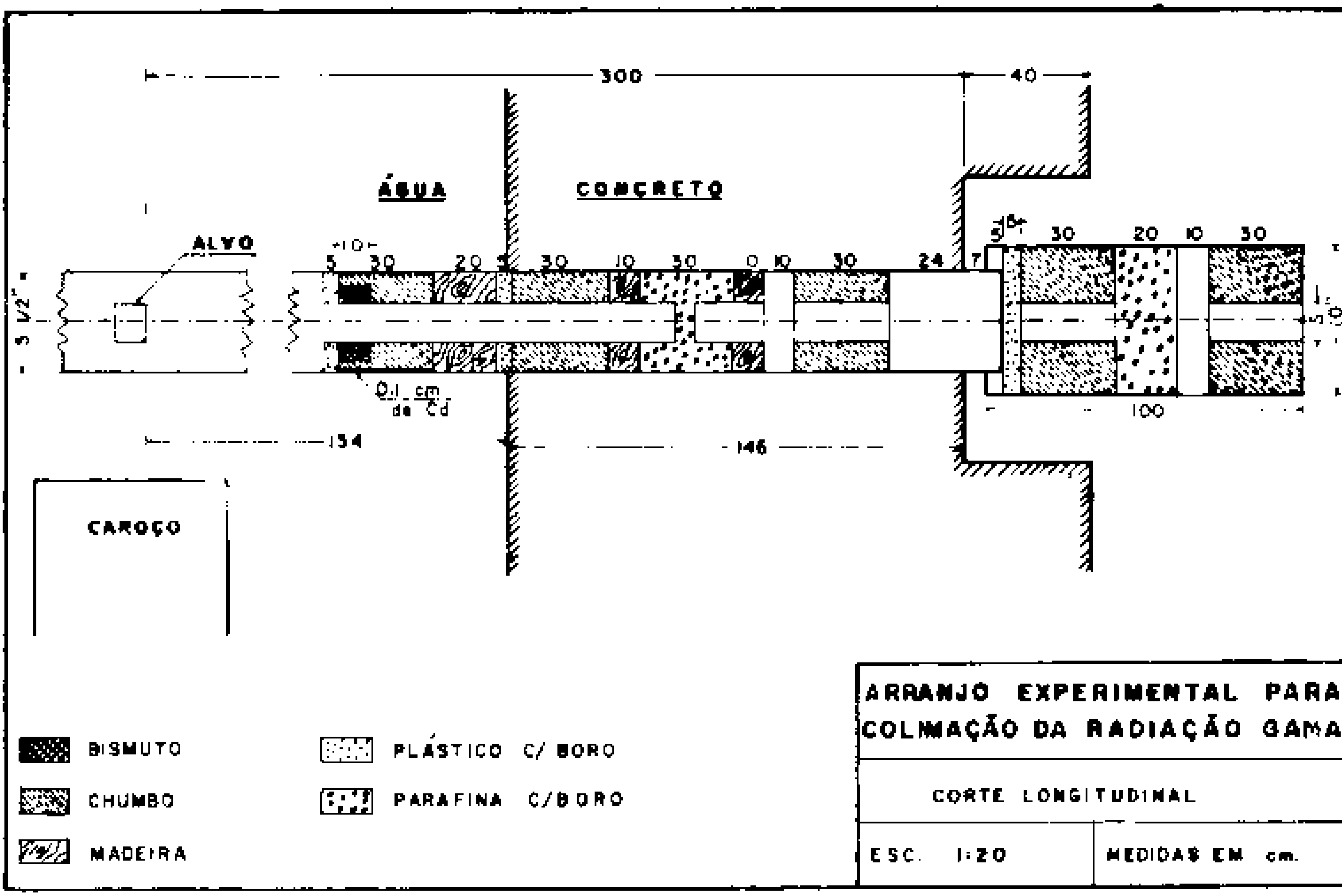

300
40
ÁGUA
CONCRETO
ALVO
5 1/2"
0.1 cm de Cd
154
146
100
CAROÇO
BISMUTO
CHUMBO
MADEIRA
PLÁSTICO C/ BORO
PARAFINA C/BORO
ARRANJO EXPERIMENTAL PARA COLMAÇÃO DA RADIAÇÃO GAMA
CORTE LONGITUDINAL
ESC. 1:20
MEDIDAS EM cm.

Figure 1

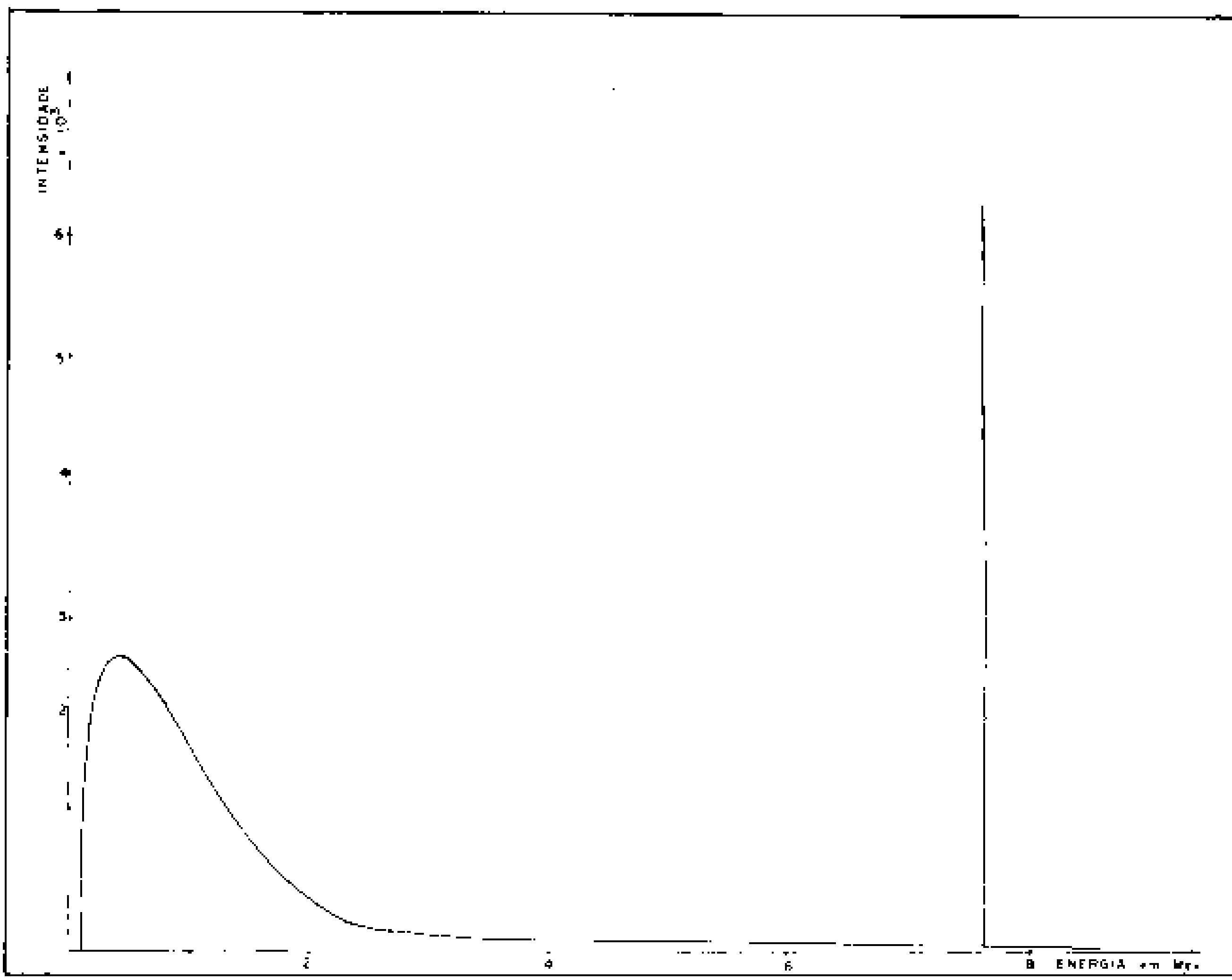


Figure 2

Figure 3

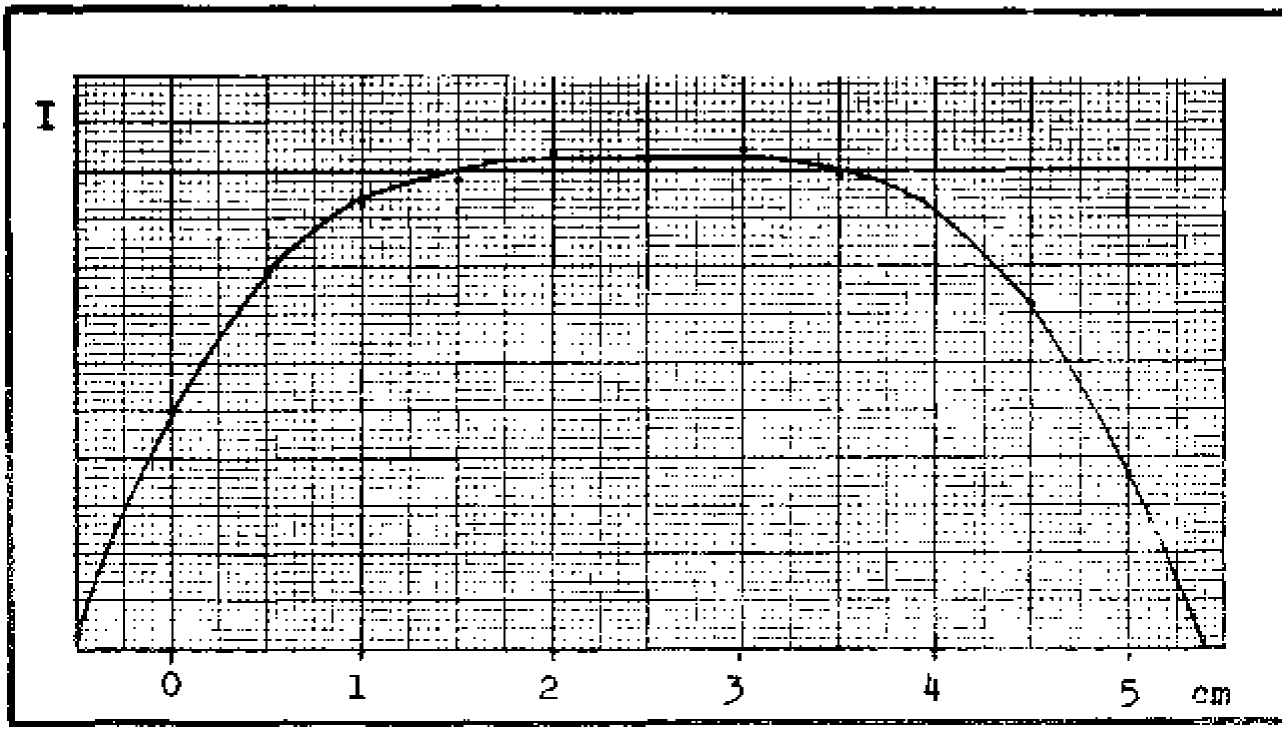
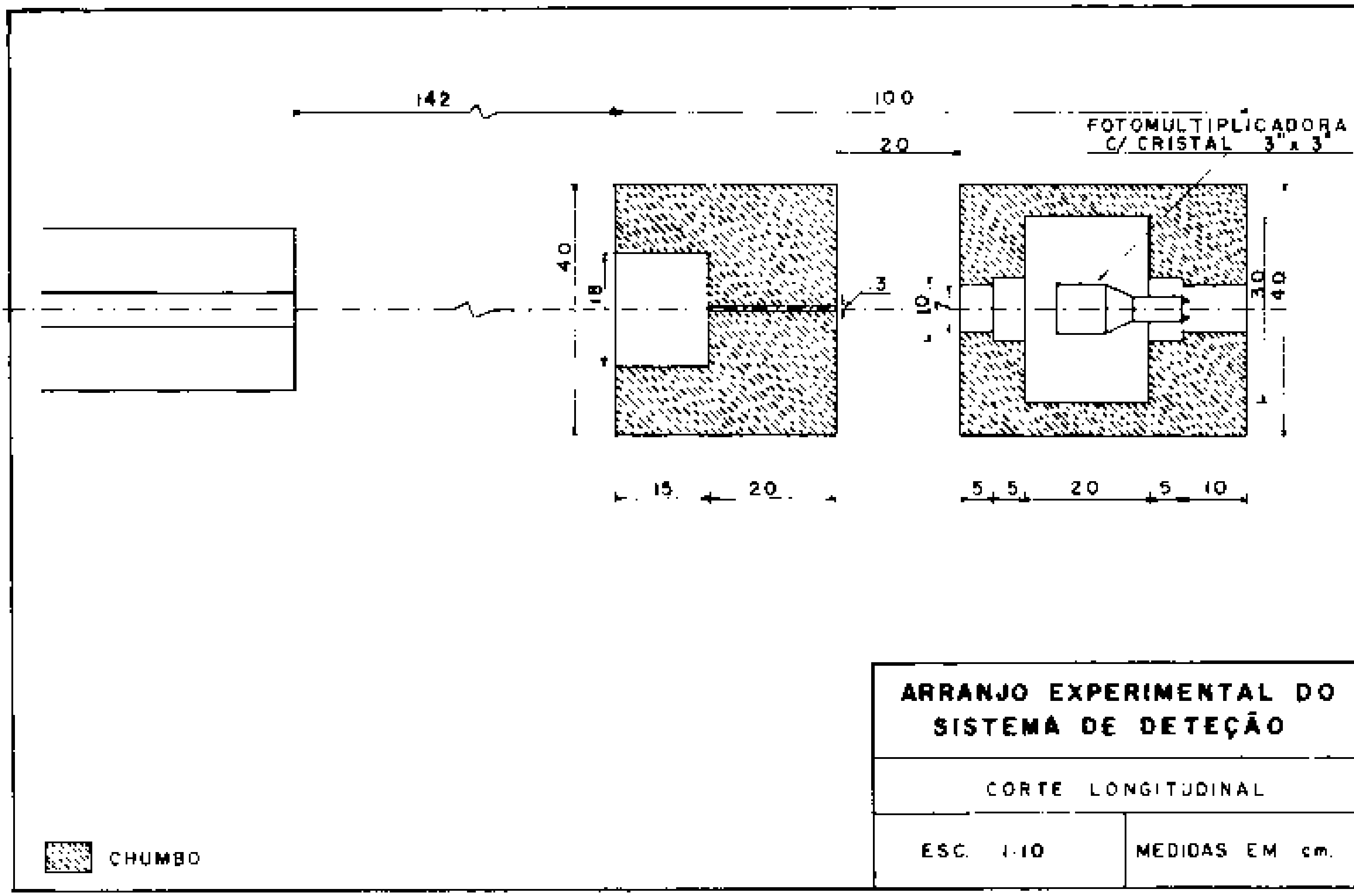


Figure 4 - Beam Profile



142
100
20
FOTOMULTIPLICADORA
C/ CRISTAL 3" x 3"
CHUMBO
ARRANJO EXPERIMENTAL DO
SISTEMA DE DETEÇÃO
CORTE LONGITUDINAL
ESC. 1:10
MEDIDAS EM cm.

Figure 5

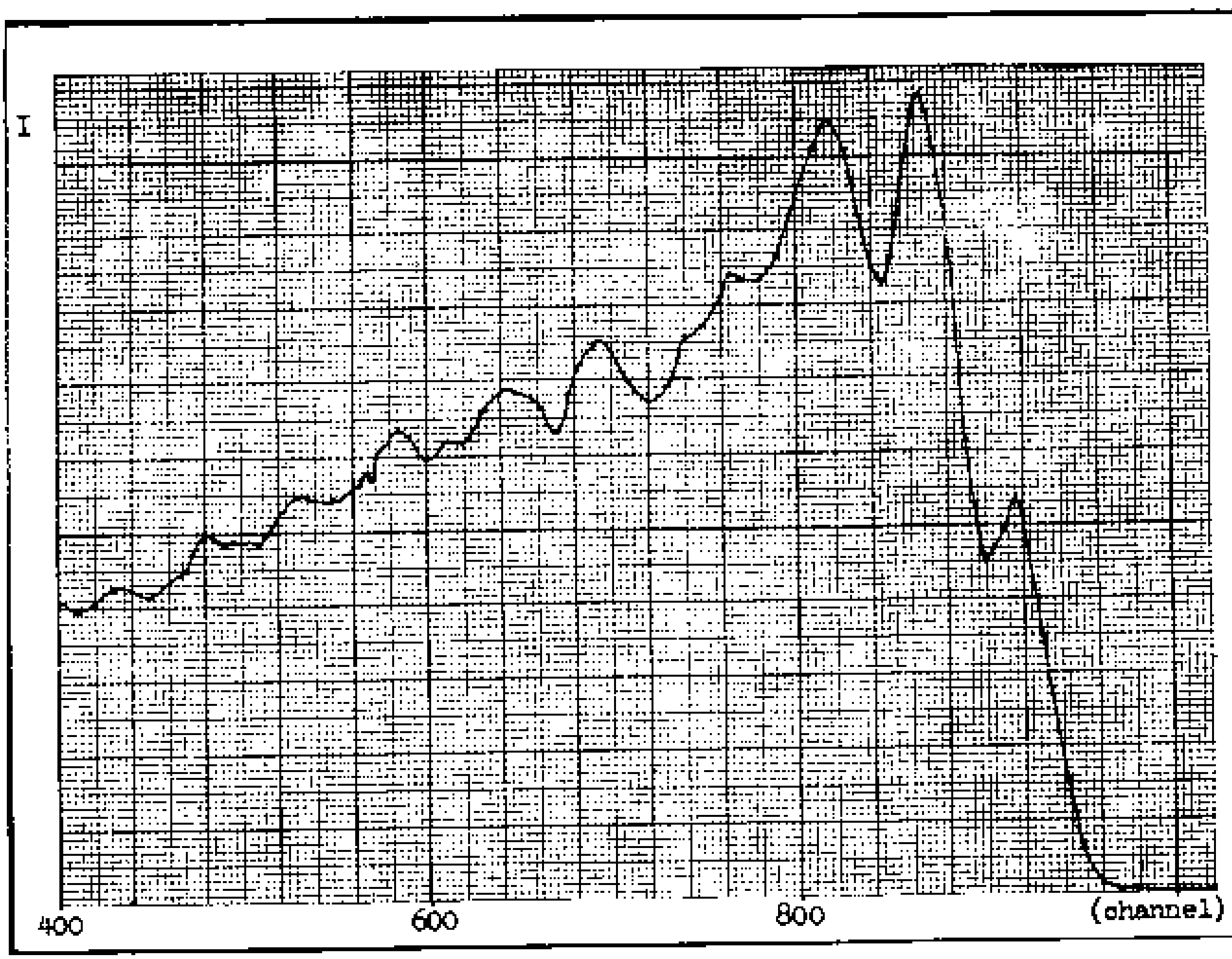
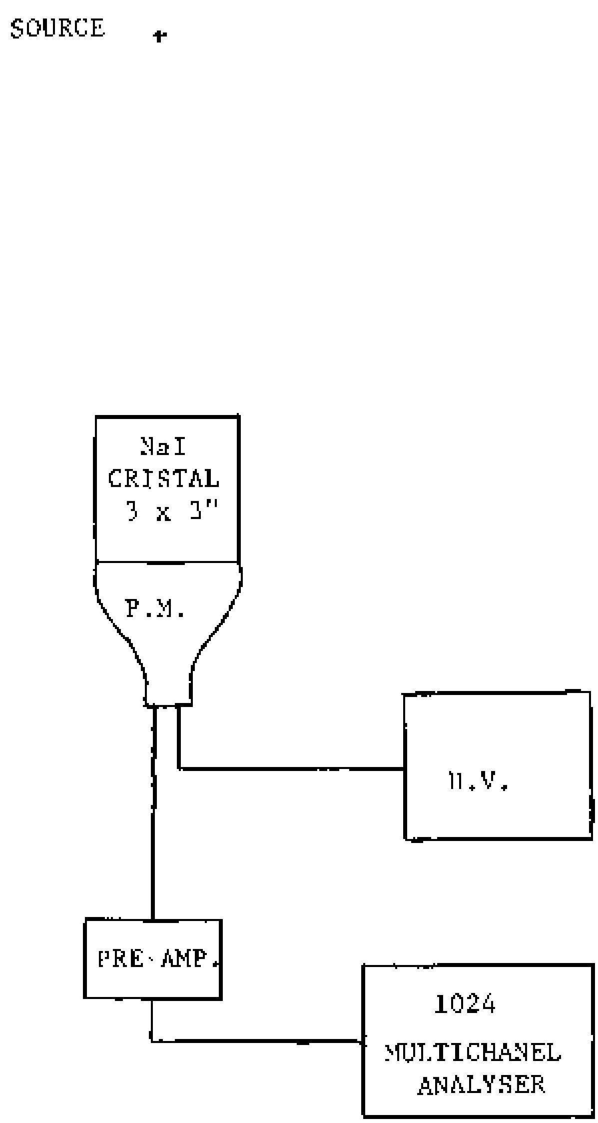


Figure 6 - Spectrum of a nickel target with a NaI 3"x3" crystal

APPENDIX I

In order to determine the thermal neutron flux in different positions inside the through tube, thin gold foils were irradiated; those measurements were made both with naked and with cadmium covered foils.



The thickness of the gold foils used was of 0.1 mm with a mass of 5 mg considered to be sufficiently small to avoid any distortion of the flux distribution in the region of the measurement.

The absolute activity of those foils was determined by means of the procedure which makes use of a NaI(Tl) scintillation crystal whose pulses were analysed with a 1024 channel analyser (TMC).

In order to have a crystal response which could be compared with its theoretical response, the source was situated at a sufficiently large distance from the crystal in order that it could be considered as a point source and to avoid gamma-ray scattering on the scintillation crystal shield.

The absolute activity is obtained through integration of the counts under the 411 keV photopeak, which was estimated as a gaussian[4] curve, and by taking into account the total efficiency K(E).

$$K(E) = p(E)\ G\ (1 - e^{-\mu(E)\ L})$$

where

$(1 - e^{-\mu(E)\ L})$ is the crystal intrinsic efficiency

G, the geometric efficiency

p(E), the photofraction, that is, the ratio between the number of pulses due to the total absorption of the photons of energy E and the total number of pulses due to the photons of energy E. The photopeak can be represented by the Gaussian,

$$y = y_o\ e^{-\frac{(x - x_o)^2}{b}} \qquad (1)$$

$$\log y = \log y_o - \frac{(x - x_o)^2}{b}$$

Putting $\log y_o = A$ and $-1/b = B$, we have

$$\log y = A + B\,(x - x_o)^2 \qquad (2)$$

which is a straigh line of log y against $(x - x_o)^2$.

The parameters A and B from the stright line (2) can be determined through a least squares method

$$A = \frac{\Sigma\, n(x)\ (x - \bar{x})^2 \quad \Sigma\, n(x)\ \ln n(x)}{\Sigma\, n(x)\ \Sigma\, n(x)\ (x - \bar{x})^4 - \left[\Sigma\, n(x)\ (x - \bar{x})^2\right]^2} -$$

$$- \frac{\Sigma\, n(x)\ (x - \bar{x})^2\ \Sigma\, n(x)\ (x - \bar{x})^2\ \ln n(x)}{\Sigma\, n(x)\ \Sigma\, n(x)\ (x - \bar{x})^4 - \left[\Sigma\, n(x)\ (x - \bar{x})^2\right]^2}$$

$$B = \frac{\Sigma\, n(x) \quad \Sigma\, n(x)\ \ \ln n(x)\ (x - \bar{x})^2}{\Sigma\, n(x)\ \Sigma\, n(x)\ (x - \bar{x})^4 - \left[\Sigma\, n(x)\ (x - \bar{x})^2\right]^2} -$$

$$- \frac{\Sigma\, n(x)\ (x - \bar{x})^2\ \Sigma\, n(x)\ \ln n(x)}{\Sigma\, n(x)\ \Sigma\, n(x)\ (x - \bar{x})^4 - \left[\Sigma\, n(x)\ (x - \bar{x})^2\right]^2}$$

where x represents the channel order and n(x) the total counts corresponding to the x^{th} channel.

By integrating the equation (1) from $-\infty$ to $+\infty$, one obtains

$$Y = y_o \sqrt{b\,\pi} \qquad \therefore \qquad \frac{Y}{\sqrt{b\,\pi}} = y_o$$

$$\log y_o = A = \log \frac{Y}{\sqrt{b\,\pi}}$$

$$\frac{Y}{\sqrt{b\,\pi}} = e^{A} \qquad\qquad Y = e^{A}\sqrt{b\,\pi}$$

where $Y = N_p$ is the number of integrated counts

$$N_p = e^{A}\sqrt{-\frac{\pi}{B}}\text{ , where B is negative} \qquad (3)$$

Since N_p is given by the equation (3), knowing the cadmium ratio and the efficiency, one can write:

$$A_{\text{without Cd}} = A_s = \frac{N_p}{K(E)} \quad \text{and} \quad A_{\text{with Cd}} = A_c = A_s/f$$

$$\therefore \qquad \boxed{\phi = \frac{A_s - A_c}{N\sigma}}$$

In order to check up this neutron flux determination, the absolute activity of the gold foils was also determined by the defined solid angle method with Geiger Muller counter and, independently, by beta-gamma coincidences[5].

The neutron flux so obtained was 3.73×10^{11} n/cm^2/sec being in well agreement with the other value.

APPENDIX II

In order to determine the number of fast and slow neutrons, an intercalibration, as described below, was made.

About 50 mg of manganese sulphate was irradiated in a neutron beam of known flux; since its mass is small, it could be considered as a point source Its absolute activity due to Mn^{56} (2,58 hour half-life) was measured considering the 850 KeV photopeak as a Gaussian and using the photopeak efficiency and the total efficiency from the tables[4].

This manganese sulphate sample was dissolved in one liter of H_2SO_4 and the scintillation crystal was immersed in the solution under a reproductible geometrical arrangement; the crystal efficiency was determined in these conditions.

To determine the neutron background contamination of the gamma-ray beam, 60 g of manganese sulphate, dissolved in one liter of H_2SO_4, were irradiated in the gamma-ray beam.

The solution was kept in a plastic container inside a paraffin layer with the purpose of increasing the detection efficiency. The whole system was shielded against neutrons from the counting room by means of suitable borated paraffin and cadmium.

The activity of this manganese sulphate solution was measured in the same conditions as stated above; since the efficiency of the system was known, the activity due to the incident neutrons was measured and the corresponding total neutron flux evaluated.

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