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RESUMO

Os fluxos de neutrons rápidos e a distribuição de energias foram medidos em alguns dispositivos de irradiação no reator de 5 Mw, tipo piscina do IEA. O espectro diferencial de neutrons rápidos no intervalo de energias de 0,5 a 10 Mev foi determinado no "beam hole" nº 7, utilizando um "telescópio" simplificado de protons de recuo e técnicas de emulsões nucleares. Técnicas de detetores limiares foram usadas para especificar os espectros de neutrons rápidos dentro do reator. É sugerido emprêgo do sulfato de magnésio como um duplo detetor limiar, através das reações $S^{32}(n,p)\ P^{32}$ (E_{ef}= 2,9 Mev) e $Mg^{24}(n,p)\ Na^{24}$ (E_{ef}= 6,3 Mev), consistindo num modo simples e econômico de medir fluxos rápidos e de estimar a forma do espectro integral de neutrons rápidos. Alguns resultados preliminares são apresentados.

RÉSUMÉ

Les flux de neutrons rapides et la distribution d'énergie ont été mésurés dans quelques dispositifs d'irradiation au réacteur

piscine de 5 Mw de l'IEA. Le spectre différentiel de neutrons rapides dans l'intervalle d'énergie 0,5 à 10 Mev, a été determiné au "beam hole" nº 7, en utilisant un "télescope" simplifié à protons de recul et des techniques d'émulsions nucléaires. Des techniques de détecteurs à seuil ont été utilisées pour spécifier les spectres de neutrons rapides à l'intérieur du réacteur. Le sulfat de magnésium est suggéré comme un double détecteur à seuil par les réactions $S^{32}(n,p)$ P^{32} (E_{ef}= 2,9 Mev) et $Mg^{24}(n,p)$ Na^{24} (E_{ef}= 6,3 Mev), qui constitue une façon simple d'estimer la forme du spectre intégral des neutrons rapides. Quelques résultats préliminaires sont présentés.

ABSTRACT

Fast neutron fluxes and energy distribution were measured in some irradiation facilities at the IEA 5 Mw swimming pool reactor. The differential fast neutron spectrum in the energy interval 0,5 to 10 Mev was determined in beam hole number 7, utilizing a simplified proton recoil telescope and nuclear emulsion techniques. Threshold detector techniques are being used to specify fast neutron spectra inside the reactor. Magnesium Sulfate is suggested as a "double" threshold detector, through the reactions $S^{32}(n,p)$ P^{32} (E_{eff}= 2,9 Mev) and $Mg^{24}(n,p)$ Na^{24} (E_{eff}= 6,3 Mev), providing a simple and low cost way to measure fast fluxes and estimate the shape of the integral fast neutron spectrum. Some preliminary results are presented.

INTRODUCTION

The knowledge of fast neutron fluxes and energy distributions is important in several fields of research reactor utilizations as , for example, radiation damage studies (1), shielding design (2)

and activation analysis (3).

A program was established to measure fast neutron fluxes and spectra in several irradiation facilities at the IEA 5 Mw swimming pool reactor. Beam measurements were started using a simplified proton recoil telescope (4) and nuclear emulsion techniques (5) (6) (7). Threshold detectors (8) (9) (10) and nuclear emulsions (5) (6) (7) may be used for "in-pile" measurements. Thus far, six threshold reactions have been studied covering the range from 0.85 to 8.1 Mev. Magnesium sulfate is suggested as a "double" threshold detector, providing information about fast neutron fluxes above 2.9 Mev. through the reaction S^{32} (n,p) P^{32} and above 6.3 Mev through the reaction Mg^{24}(n,p) Na^{24}. Absolute activities of the resulting nuclides of these reactions are determined simultaneously in a 4 Pi proportional beta counter. Some preliminary results are given below.

BEAM MEASUREMENTS

a) Simplified proton recoil telescope (4)

A simplified proton recoil "telescope" was developed which presents relatively good resolution with an efficiency high enough for measurements in reactor beams whose fluxes are of the order of 10^6 n/cm^2/sec. Good resolution is achieved by the use of thin polyethylene radiators and a small detection solid angle. Background discrimination, usually provided by coincidence measurements, is here attained by a careful design of the recoil chamber. The main features of the chamber are: (a) its large size, in order to avoid wall scattering; (b) proton detector located out of the reactor beam; (c) operation in vacuum, providing a free path for the recoil protons from radiator to detector; and (d) detector collimator and shield made with high Z material.

Fast neutrons undergo n-p scatterings in a thin poly - ethylene foil and the emerging recoil protons that form an angle $\theta = 20°$ with the direction of the incident neutron beam, are detected by a CsI (Tl) scintillator. Protons lose almost all their energies in the CsI (Tl) crystal and the energy distribution is determined by pulse-high analysis in a multi - channel analyser. Neutron spectra can be calculated from proton energy distributions by means of the relation $E_n = E_p \sec^2 \theta$. At neutron energies ranging from 2 to 12 Mev the detection efficiency varies from 10^{-5} to 10^{-7} and the chamber intrinsic resolution is 8%. The overall resolution is somewhat poorer due to the scintillation detector contribution. With well polished crystals and good photomultipliers total resolutions of 10% (f.w.h.m.) can be obtained.

The spectrum of fast neutrons was measured at the exit of Beam hole nº 7 (BH-7) in the IEA swimming pool reactor operating at 2 Mw. Figure 1 gives the experimental set up and Figure 2 the resulting curve with the core arrangement nº 59, without a graphite reflector. The fission spectrum shape (11) (12) is modified by the hardening effect of the pool water and bismuth filter located inside the collimator. The valley observed at about 3.7 Mev is due to the broad peaks presented by the oxigen and bismuth total cross sections for neutrons at this energy. The hydrogen cross section tends to modify the shape of the fission spectrum at low neutron energies. The mean energy in the interval 2.5 - 7.5 Mev of this emerging spectrum is 4.3 Mev.

NUCLEAR EMULSION TECHNIQUES

The method utilized is described in detail by L. Rosen in reference 5. For the measurement of fast neutron spectra Ilford

K-2 nuclear emulsions were used as both radiator and detector. This method allows the detection of neutrons with an energy as low as 0.5 Mev. In this case the basic reaction is the elastic collisions of neutrons with the hydrogen nuclei contained in the gelatin of the emulsion. The recoil protons leave latent images of their trajectories in the emulsion which, after developing, come out as characteristics tracks. These tracks can then be identified and measured with a microscope. The track lenght is a function of the proton energy which is related to the corresponding neutron energy by the expression:

$$E_p = E_n \cos^2 \theta$$

where

E_p = proton energy
E_n = neutron energy
θ = angle subtended by the trajectories of the incident neutrons and recoil protons tracks

The number of incident neutrons per unit area, per unit time and per unit energy interval is determined considering: the number of hydrogen atoms per cm^3 in the emulsion, the neutron-proton scattering cross-section, the probability for the tracks being entirely contained in the emulsion, the solid angle for acceptance of tracks, and the scanned volume.

Ilford type K-2 plates, 3" x1", with a 200 micron thick emulsion were irradiated during 30 seconds, at the exit of BH-7 parallel to the reactor beam. The reactor power was 2 Mw and the fission neutrons were collimated by a 3.30 m tube, and traversed the following materials : 11 cm graphite, 1 cm H_2O, 1 cm Al, 7.5 cm Pb and 0.255 cm Cd.

The emulsions were developed by a combination of the two

solution method and the temperature method (5). This technique proved to be the most convenient for this particular application.

Plate analysis were carried out by using a MEOPTA Type B-36 Binocular Microscope, adapted in our laboratory for emulsion work. A 100x oil immersion objetive and a 10x eyepiece with a calibrated graticle were used.

The relation between the track length of the recoil proton and its energy may be obtained by the formula

$$E = kL^n$$

where the constants, according to Barkas (15), are:

$1 \text{ Mev} < E_p < 10 \text{ Mev}$ $\quad n = 0.604 \pm 0.008$

$\quad k = 0.221 \pm 0.005$

The neutron energy E_n is given by the relation

$$E_n = \frac{E_p}{\cos^2 \theta}$$

where : E_p = recoil proton energy

$\cos \theta = \cos \alpha \cos \ell$

α and ℓ = angles defining the position of the tracks as measured at the microscope.

The error in the energy was less than 8%.

Data for the energy calculation has been processed by a IBM-1620 computer.

The absolute neutron flux is given by the expression

$$F(E_n)\, dE_n = \frac{\int N(E_p)\, d E_p}{n\, P(L)\, \sigma_{n,p}(E_n)\, \overline{\cos\theta}\, V\, \Omega}$$

where:

$N(E_p)$ = number or protons per energy interval at energy E_p

n = number of hydrogen atoms per cm^3 of emulsion

$P(L)$ = probability for a given track to be contained entirely in the emulsion

V = scanned volume

Ω = solid angle of acceptance

$\overline{\cos\theta}$ = mean value of the angle subtended by the trajectories of incident neutrons and recoil proton tracks.

The measurement of a simple, well known fast neutron spectrum has been carried out preliminarily, in order to check our experimental instruments and procedures. Figure 3 shows the spectrum of neutrons from the reaction $H^3(d,n)$, in the direction of the incident deuterons (at 0º), obtained near the tritium target of a High Voltage Engineering Model FN neutron generator, with 100 Kev deuterons. The accepted neutron energy value for this deuteron energy and at an angle 0º with the deuteron beam direction is E_n = 14,75 Mev.

The fast neutron energy distribution as measured at the exit of BH-7, IEA-R1 research reactor, with the technique just described is shown in figure 4. The statistical accuracy of the method is represented by the vertical bars. The fission spectrum (solid line) as well as some measurements carried out with the proton recoil telescope described before (small circles) can also be seen. The fission spectrum was not fitted to any experimental point and is shown here only for comparison. Effects of the

hydrogen, oxigen and lead (or bismuth) cross sections on the shape of the spectrum as was observed in the results obtained with the proton recoil telescope can also be observed in the nuclear emulsions results.

IN-PILE MEASUREMENTS

a) Threshold detectors

Measurement of fast neutron flux by activation techniques are based on the fact that some nuclear reactions induced by neutrons and resulting in radioactive nuclides are endoergic and hence, possible only for neutrons having energies above a certain threshold. Some (n,f), (n,n'), (n,p), (n,α) and (n,2n) reactions have threshold energies that are convenient for the detection of fast neutrons in reactors.

An effective threshold energy, E_{eff}, is defined so that :

$$\sigma_s \int_{E_{eff}}^{\infty} f(E)\, dE = \sigma_s F_{eff} = \int_{E_T}^{\infty} \sigma(E)\, f(E) dE \quad (1)$$

where σ_s is the saturation value of the cross section (maximum mean value); f(E), the differential neutron flux; F_{eff}, the total fast flux above E_{eff}; $\sigma(E)$, the cross section for the particular reaction at energy E; and E_T, the true threshold energy.

Some effective cross sections and threshold energies have been determined experimentally in several swimming pool reactors (8) (9) (Table 1).

An alternative approach, if the fast neutron spectrum is not too different from a fission spectrum, consists of determining a mean cross section $\overline{\sigma}$, through the relation:

$$\overline{\sigma} = \int_0^\infty f(E)\ \sigma(E)\ dE \Big/ \int_0^\infty f(E)\ dE \qquad (2)$$

A method for calculating effective threshold energies for a fission spectrum is mentioned in reference (13).

In both cases, cross section curves as a function of neutron energy are approximated by step functions where:

$$\begin{aligned} \sigma(E) &= 0 && \text{for} \quad E < E_{eff} \\ \sigma(E) &= \sigma_s \quad \text{or } \overline{\sigma} && \text{for} \quad E > E_{eff} \end{aligned} \qquad (3)$$

In this paper effective saturation value cross sections are used rather than mean cross sections.

Let us consider a sample that has been exposed to neutrons and which contains an isotope wherein a threshold reaction can be induced. The total flux above E_{eff} can be evaluated by the expression:

$$F_{eff} = \frac{A_o}{N \sigma_s (1 - e^{-\lambda t})}, \qquad (4)$$

where A_o is the absolute activity of the resulting nuclide in the moment irradiation finishes; N is the number of target nuclei in the sample; σ_s, the experimental effective cross section; λ, the desintegration constant of the resulting radionuclide; and t, the irradiation time.

Table 1 gives some data on the threshold reactions thus far used to measure fast flux in the IEA reactor. All samples are irradiated inside a cadmium shield in order to reduce the influence of competitive (n,γ) reactions with slow neutrons. Indium and aluminium are irradiated and counted in the form of thin metal foils. Other elements or compounds containing target nuclides are irradiated either as powdered samples or pellets.

Absolute activities are measured in a 4 Pi beta proportional counter and in an end-window GM counter with defined geometry (14). Special techniques are used in the preparation of samples for counting in the 4 Pi detector. Small aliquots of solutions prepared with the irradiated samples are deposited on thin aluminized mylar supports (0.8 mg/cm^2), previously treated with insulin, to provide a uniform spread of the sample after drying. With this procedure, self absorption correction becomes negligible for some radionuclides involved.

Table II and figure 5 present some results obtained with threshold detectors in the pneumatic rabbit station number 3. Figure 3 gives an idea of the relative position of this irradiation facility and the reactor core. Its closest distance to the core is about 8 inches. The solid curve in figure 5 is the U^{235} integral fission spectrum normalized to the Al^{27} (n,α) Na^{24} point. The experimental curve shows a hardening of the fission spectrum due to interactions of the virgin fission neutrons with the pool water and graphite reflector. All experimental points are averages taken from different measurements. The overall error in each point is very difficult to evaluate in determinations of this kind, as the extent of uncertainties in cross section and effective threshold energy data are unknown. Statistical errors were kept below 3% in all absolute activity measurements.

Some compounds have been considered for utilization as "double" threshold detectors. So far, $MgSO_4.1H_2O$ has been successfully utilized. A known amount of the salt is irradiated in a polyethylene vial. After irradiation this sample is dissolved in a known volume of distilled water. An Aliquot of this solution is transferred by means of a micropipet to an aluminized mylar support (0.8 mg/cm^2 thick) for a 4 Pi beta counting. The decays of Na^{24} and P^{32} formed in the reactions $Mg^{24}(n,p)\ Na^{24}$ and $S^{32}\ (n,p)\ P^{32}$ are followed simultaneously and the respective absolute activities are resolved graphically. This double threshold detector technique provides a simple and low-cost way to measure fast neutron fluxes and to estimate the shape of the integral energy spectrum. The compound used is very common and gas flow counters with 2 Pi or 4 Pi geometries are very easy to construct.

b) Nuclear emulsions

Nuclear emulsions are also programmed for in-pile measurements. For extended anisotropic sources only integral measurements are possible. A method devised by J.E. Evans and mentioned in reference (5) may be applied.

CONCLUSIONS

The data presented in this paper must be considered solely as preliminary results obtained in the evaluation of several experimental techniques for the specification and measurement of reactor fast neutron spectra. Most of the apparatus utilized must be improved and some completely redesigned. Some modern techniques using exothermic reactions and solid state detectors such as the He^3 coincidence spectrometer may be

considered.

With the experience accumulated up to date in threshold detector techniques, a routine program for the measurement of fast neutron fluxes and spectra, related to the utilization of the IEA Swimming Pool Reactor in radiation damage studies, can be established.

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TABLE 1

Threshold Detector Reaction Data for Swimming Pool Reactors

Reaction	Half-Life	Effective Threshold Energy (Mev)		Experimental Effective Cross Section (mb)	
		(1)	(2)	(1)	(2)
In^{115} (n,n') In^{115m}	4.5 h	-	~0.85	-	230
P^{31} (n,p) Si^{31}	2.62 h	2.4	3.1	75	142
S^{32} (n,p) P^{32}	14.3 d	2.9	2.9	300	300
Cl^{35} (n, α) P^{32}	14.3 d	4.1	-	178	-
Mg^{24} (n,p) Na^{24}	15.1 h	6.3	7.5	48	190
Al^{27} (n, α) Na^{24}	15.1 h	8.1	8.1	111	116

(1) - BMI - 1486 (1960), reference (8)

(2) - J. Romanko and W.E. Dungan, General Dynamics NARF - GTR reactor, reference (7).

Instituto de Energia Atômica 5 Mw swimming pool reactor

Pneumatic Rabbit Station N°3 - Integral Fast Neutron Spectrum

Effective Threshold Energy (Mev)	Neutron Flux above Threshold -n/cm^2 sec (2Mw)	Neutron Flux per Watt Above Threshold-n/cm^2. sec. watt	Detector Used
~ 0.85	1.75×10^{11}	8.74×10^{4}	In foil
2.4	1.18×10^{11}	5.90×10^{4}	$(NH_4)_2 HPO_4$ pellet
2.9	7.60×10^{10}	3.80×10^{4}	$MgSO_4$ powder and Sulfur pellet
4.1	5.30×10^{10}	2.65×10^{4}	NH_4Cl pellet
6.3	1.11×10^{10}	5.54×10^{3}	$MgSO_4$ powder
8.1	3.90×10^{9}	1.95×10^{3}	Al foil

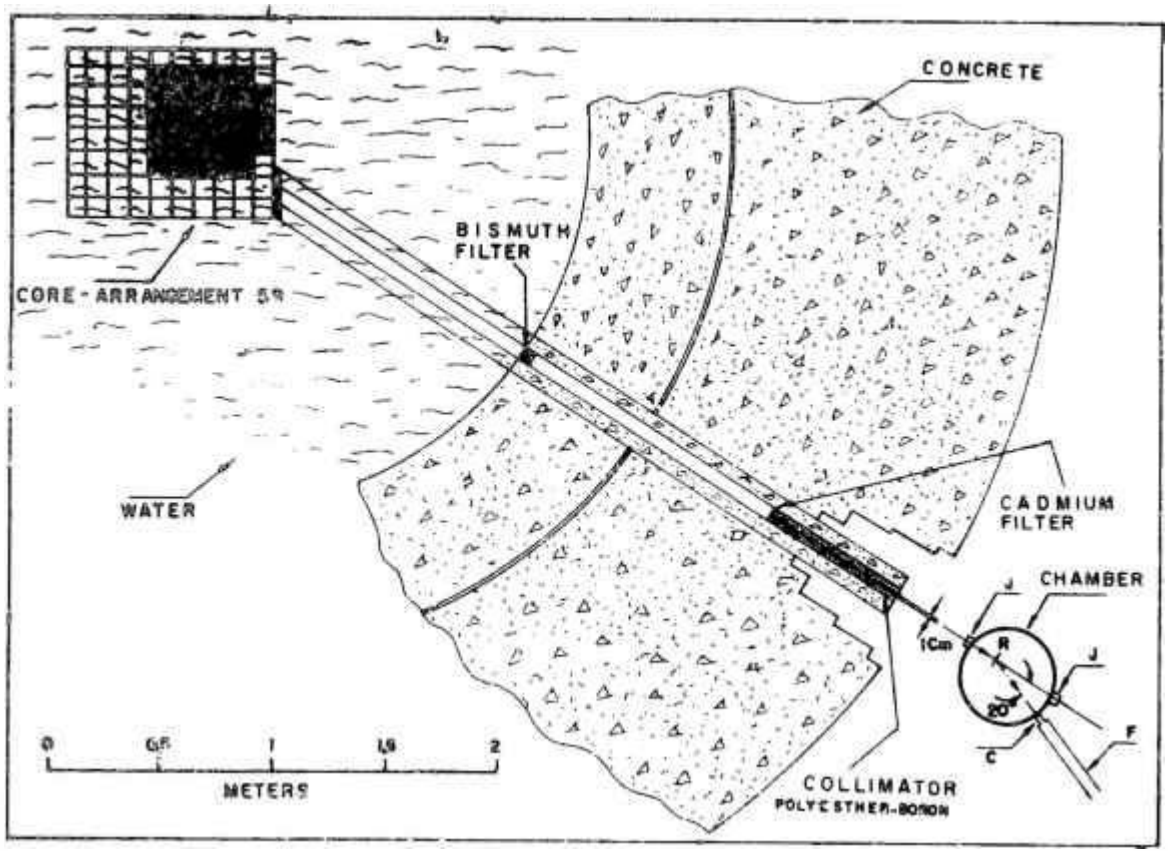


Fig.1

Experimental arrangement at beam-hole no.7 for the measurements of fast-neutron spectra.
J = thin aluminum windows, R= Polyethylene radiator, C = CsI (Tl) crystal, F = photomultiplier and preamplifier.

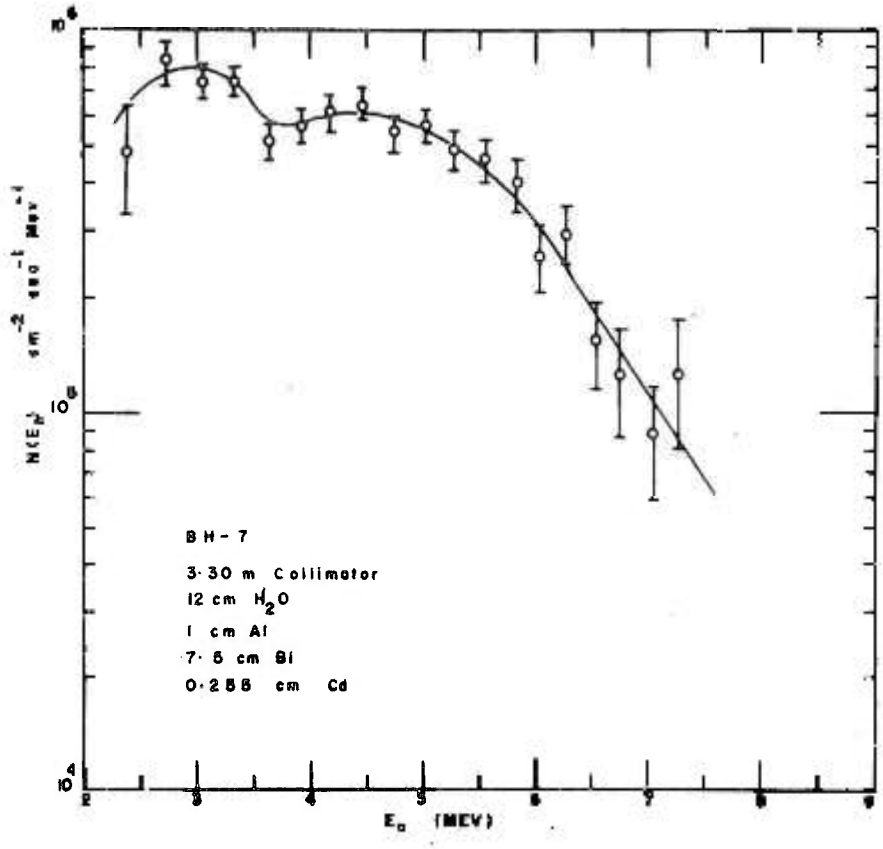


Fig.2

Fast neutron spectrum measured at the exit of beam hole Nº 7. Reactor core without a graphite reflector.

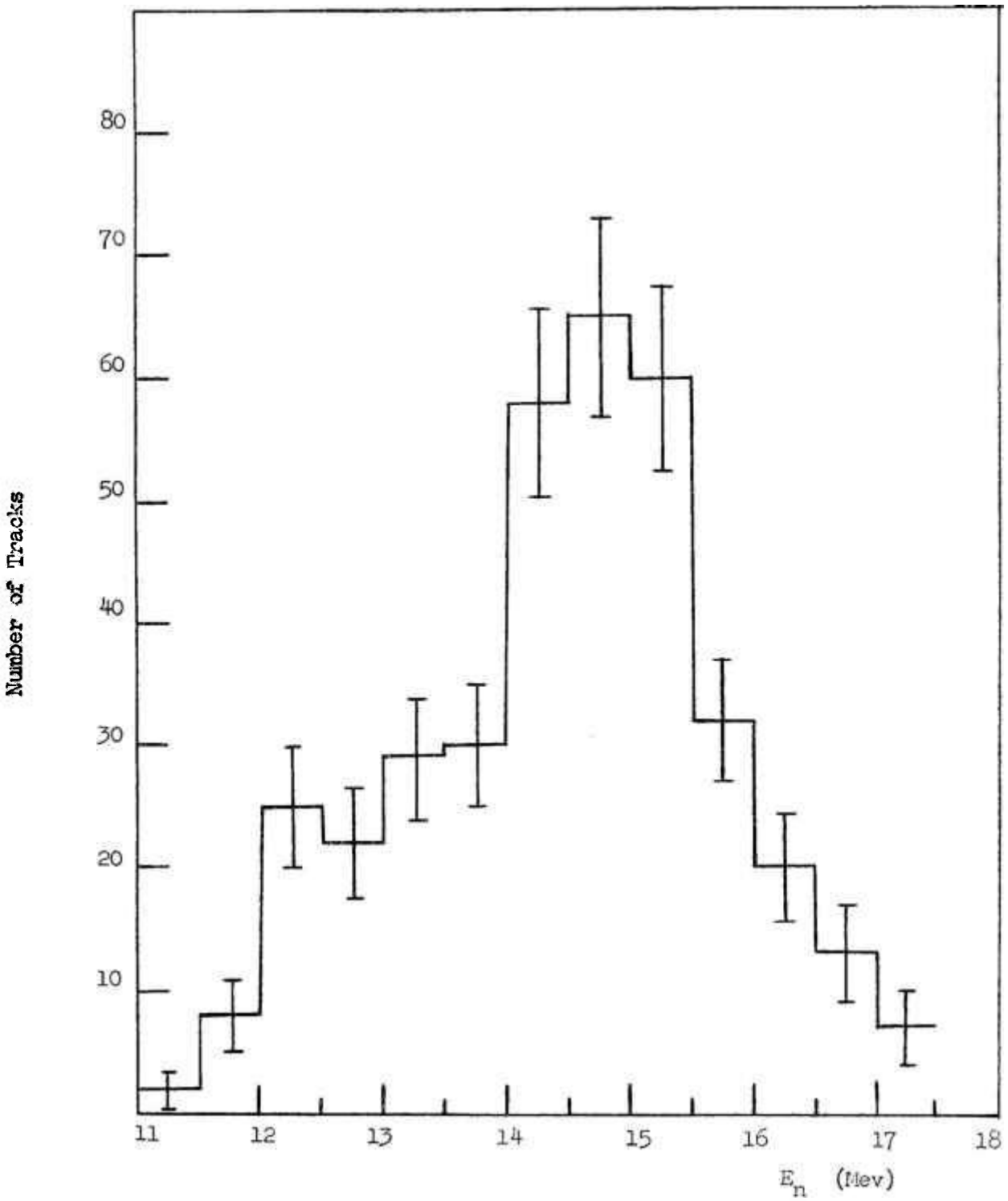


Figure 3

Neutron spectrum from the reaction $H^3(d,n)\,He^4$, measured at 0° with Ilford Type K-2 Nuclear Plates. High Voltage Engineering Model PN 400 Pulsed Neutron Source, Reactor Physics Division (IEA)

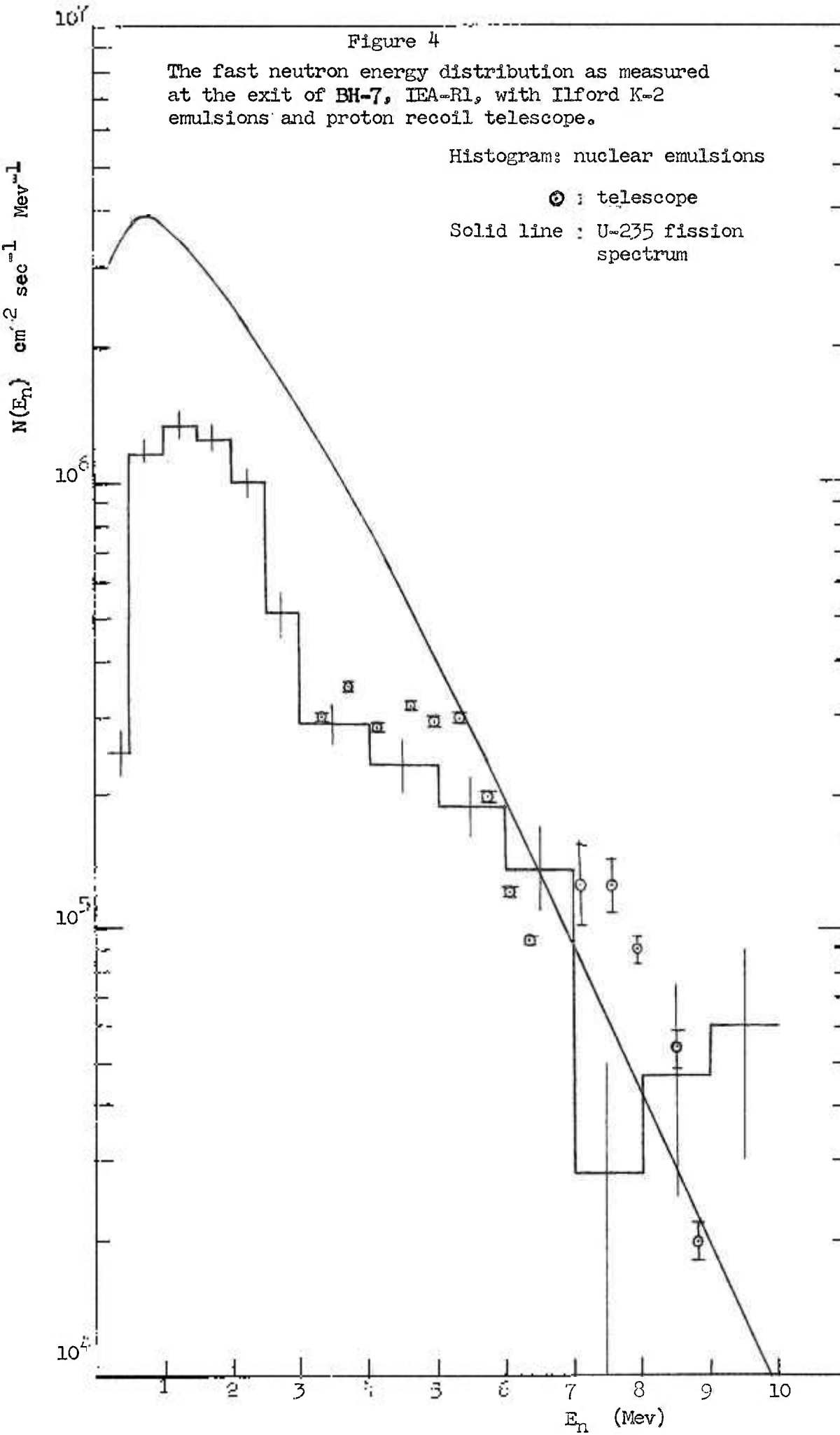
Figure 4

The fast neutron energy distribution as measured at the exit of BH-7, IEA-R1, with Ilford K-2 emulsions and proton recoil telescope.

Histogram: nuclear emulsions

⊙ : telescope

Solid line : U-235 fission spectrum



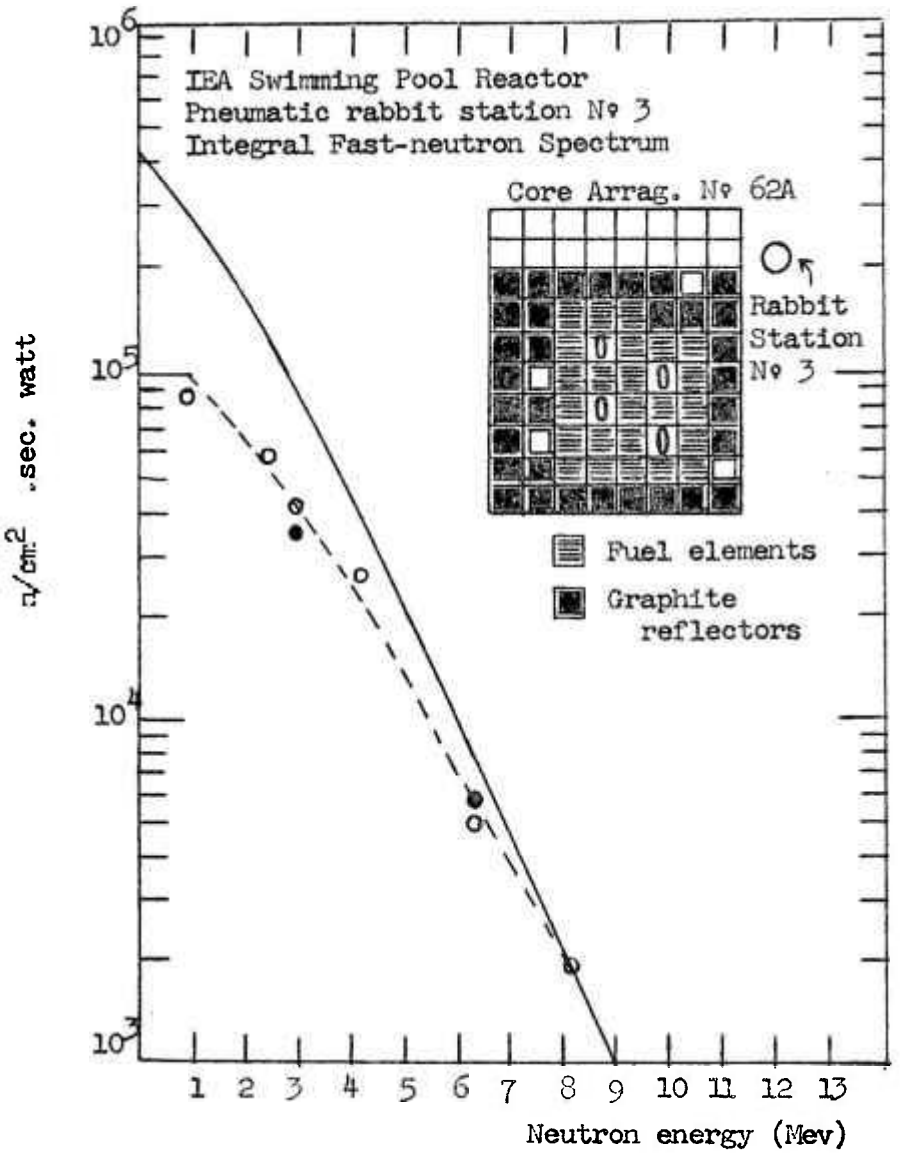


Fig. 5

IEA Swimming Pool Reactor. Integral fast-neutron spectrum in pneumatic rabbit station № 3. Solid curve is U^{235} integral fission spectrum normalized to the Al^{27} (n,α) Na^{24} point. Black circles are measurements made with $MgSO_4$ double threshold detector.