SLOW-NEUTRON CROSS-SECTION FOR URANIUM OXIDE

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ABSTRACT

The total cross section of a polycrystalline sample of uranium oxide has been measured in the energy range from 0.08 ev to 0.0013 ev, using the slow-neutron chopper and time-of-flight spectrometer at the IEAR-1 reactor.

The absorption cross-section was determined subtracting the calculated scattering cross-section affected by the experimental resolution from the measured total cross-section.

The obtained absorption cross-section values are compared with the calculated curve on basis of the single level Breit-Wigner formula. It is observed a reasonable agreement between the experimental points and the theoretical curve.

I. INTRODUCTION

The neutron cross-section of materials that contain fissionable nuclides are rather interesting for reactor technology, because of their effect on the neutron economy and their use in determining the fuel burnup.

The purpose of the present work is to determine the absorption and scattering cross-section, for slow neutrons (neutrons with energy below 0.1 ev), of a polycrystalline sample of natural uranium oxide.

The cross-section study of crystalline materials in the energy range in which the Bragg scattering makes an appreciable contribution is of particular interest because the value of the total cross-section depends mainly on its normal crystalline structure.

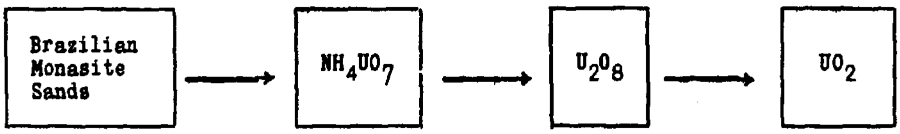
In the thermal energy the absorption cross-section, σ_{ab}, for natural uranium is 7.68 barns and for oxigen it is < 0.004 barns[(1)]. As the σ_{ab} for uranium has not a 1/v dependence[(1)] its value, in the energy range of interest to the present work, could not be determined by a simple extrapolation.

In this work, the absorption cross section of uranium oxide was determined from the measured total cross-section. The result was compared with the calculated curve on basis of Breit-Wigner formalism(2).

II. EXPERIMENTAL

Neutron transmission measurements of UO_2 have been performed in the energy range from 0.08 to 0.0013 ev (1.0 to 8.0 Å) using the slow neutron chopper and time-of-flight spectrometer(3) at IEAR-1 reactor. All measurements were carried out at room temperature.

The UO_2 sample used was cylindrical with 3.2 cm diameter and 1.0 cm thick; the density was of 10.45 g/cm^3. The sample has been sintered at 1400°C. Its procedence is shown in the graph below(4).



The oxide obtained by this process was analysed in order to determine the grade of purity of the sample. The analyses did not present significant quantities of impurities.

III. RESULTS AND DISCUSSION

a) Experimental total cross-section

The total cross section of UO_2, determined through transmission measurements in shown in figure 1. The errors indicated

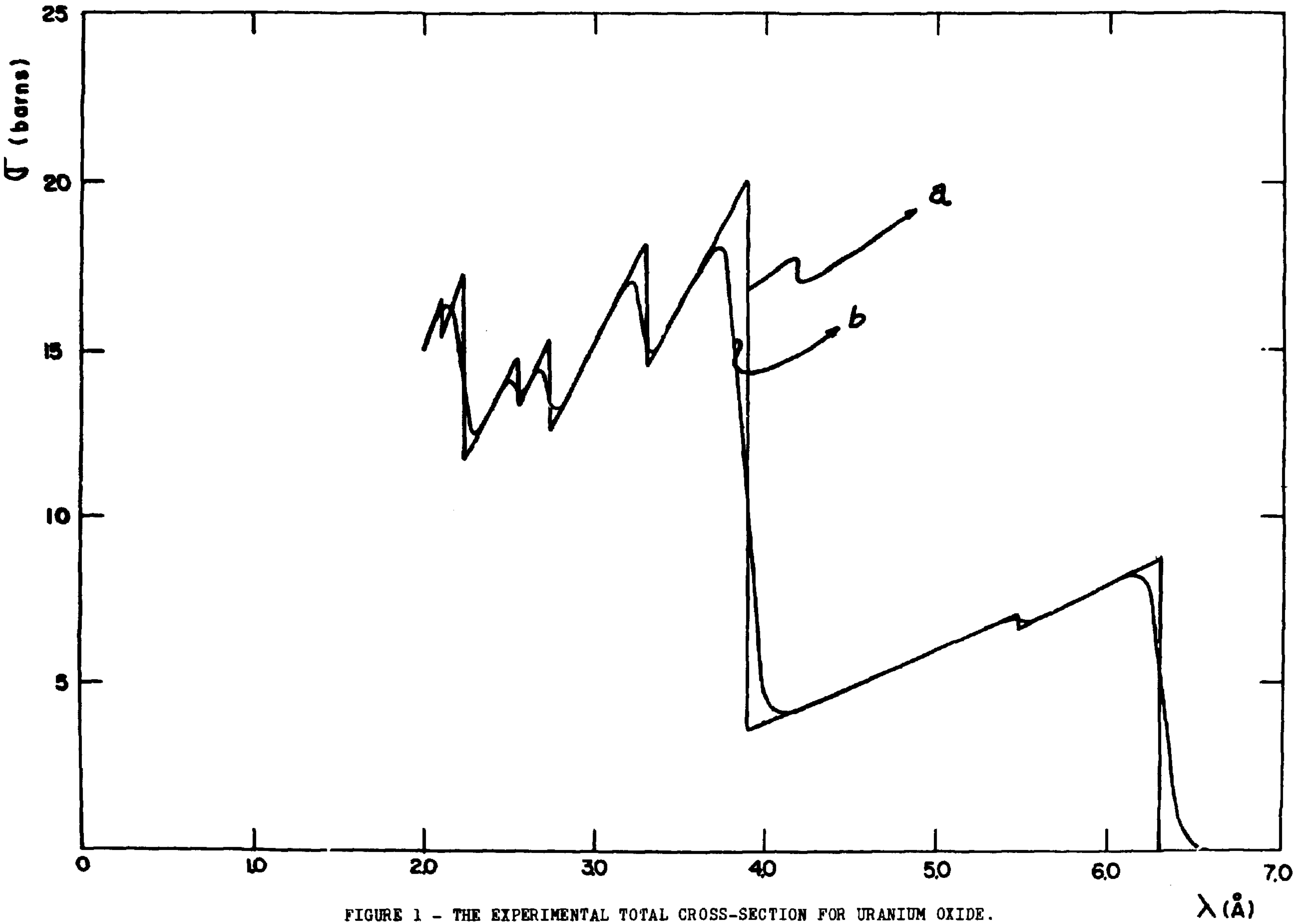


FIGURE 1 - THE EXPERIMENTAL TOTAL CROSS-SECTION FOR URANIUM OXIDE.

are statistical only. The data for $\lambda > 3.6$ Å represent an average over three channels of 8 μs in the time-of-flight analyser and for $\lambda > 5.2$ Å they are an average over results obtained by three independent measurements. The experimental resolution in the edges is indicated in the figure.

b) Theoretical scattering cross-section

The total scattering cross section of a polycrystalline sample is the sum of four terms, incoherent elastic scattering cross-section, coherent elastic scattering cross-section,coherent inelastic scattering cross-section and incoherent inelastic scattering cross-section.

For UO_2, as the nuclear incoherent scattering cross-section of Uranium and Oxigen atoms are very small the incoherent contributions for total scattering cross-section will be neglected. The coherent elastic scattering cross-section (Bragg reflexions) has been calculated assuming for UO_2, at room temperature, a crystalline structure of CaF_2 type, with lattice constant ... $a_o = 5.47$ Å.

The structure factor for a crystal of CaF_2 type depends on the sum of the Miller indices hkl of the crystalline planes.

For $h + k + 1 = 4n$, where n is an integer, the uranium and oxigen atoms scatter in phase and the reflexions are strong; for $h + k + 1 = 4n \pm 2$ they scatter out of phase and the reflexions are weak; for $h + k + 1 = 4n \pm 1$ only uranium atoms contribue to the scattering and the reflexions has a medium intensity.

The calculated structure factors are

$$F_{hkl}^{4n} = 4b_n \exp\left(\frac{-B_u}{4d^2}\right) + 8\, b_o \exp\left(\frac{-B_o}{4d^2}\right)$$

$$F_{hkl}^{4n\pm2} = 4b_u \exp\left(\frac{-B_u}{4d^2}\right) - 8\, b_o \exp\left(\frac{-B_o}{4d^2}\right)$$

$$F_{hkl}^{4n\pm1} = 4b_u \exp\left(\frac{-B_u}{4d^2}\right)$$

where b_u and b_o are the nuclear scattering amplitude of the uranium and oxigen atoms, B_u and B_o their temperature factors and d is the lattice spacing of the hkl planes.

For a crystalline sample, the coherent elastic scattering cross-section is given by

$$\sigma_{coh}^{elast.} = \frac{N\lambda^2}{2} \sum_{d > \frac{\lambda}{2}}^{hkl} MF^2 d$$

where N is the number of units cell/cm^3, λ is the neutron wavelenght and M is the multiplicity of hkl planes.

The calculation of $\sigma_{coh}^{elast.}$ for UO_2 was carried out assuming $b_u = 0.85 \times 10^{-12}$cm, $b_o = 0.577 \times 10^{-12}$, $B_u = 0.31$ and ... $B_o = 0.49$[5].

The calculated curve for $\sigma_{coh}^{elast.}$ is shown in figure 2.

The other component of total scattering, the coherent inelastic scattering, was calculated assuming the Marshall-Stuart model making use of the incoherent approximation[6].

In the energy region of interest to this work, the calculated coherent inelastic scattering cross section resulted less than the experimental errors in the total cross-section measurement. Because of this reason it will be only considered, in the total scattering, the contribution of the coherent elastic scattering.

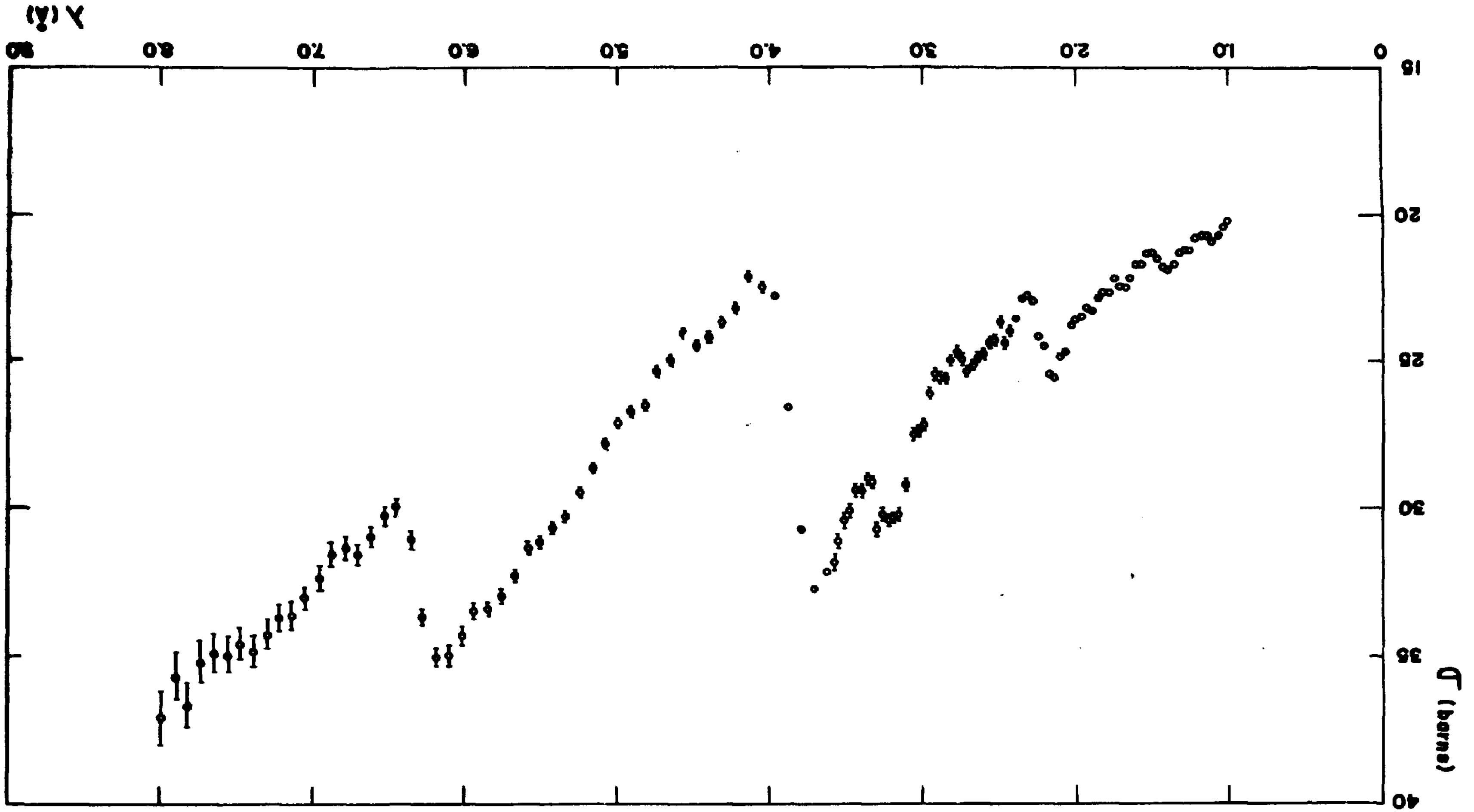


FIGURE 2 - (a) - THE CALCULATED COHERENT ELASTIC CROSS-SECTION FOR URANIUM OXIDE AND
(b) - THE CALCULATED CURVE AFFECT BY EXPERIMENTAL RESOLUTION.

c) Experimental absorption cross-section

For the determination of the absorption cross-section we must consider the contribution of the absorption cross-sections of the nuclides included in the UO_2 molecule; oxigen, uranium 235 and uranium 238. For this, the values of the absorption cross-section, at therma' energy, tabulated in the litterature[1] was used. As the absorption cross-section of the ^{235}U has not an 1/v dependence[1] it is not possible to estimate correctly the absorption cross-section in the total energy range of our measurements. For this reason, in this work, the absorption cross-section was calculated subtracting the theoretical scattering cross-section affected by experimental resolution, from the measured total cross--section. The coherent elastic scattering affected by experimental resolution is shown in figure 2. The absorption cross-section for UO_2 determined by this method is shown in figure 3.

d) Theoretical Absorption cross-section- Breit-Wigner formula

In order to compare with the experimental absorption cross-section, the theoretical curve was calculated from the single level Breit-Wigner formula[2].

For this calculation it is necessary to know the parameters of the near resonances of ^{238}U and ^{235}U. The effect of the oxigen atom resonances was not considered.

For a well isolated resonance, the capture and fission cross-sections calculated by Breit-Wigner are given by

$$\sigma_{cap} = \pi\lambda^2 \; g \; \sqrt{E} \; \frac{\Gamma_n^o \; \Gamma_\gamma}{(E-E_o)^2 + (\frac{\Gamma}{2})^2}$$

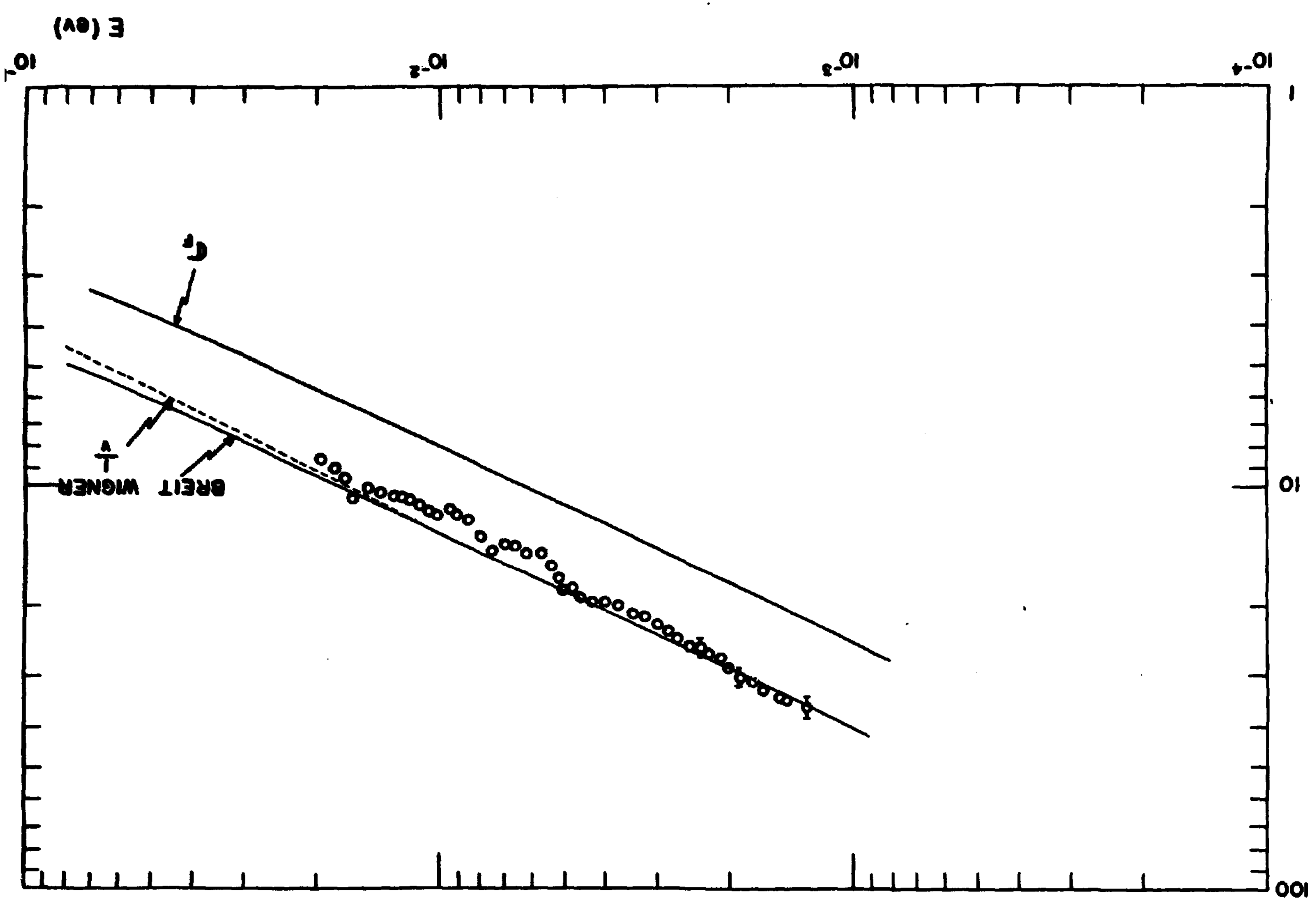


FIGURE 3 - THE EXPERIMENTAL ABSORPTION CROSS-SECTION FOR URANIUM OXIDE.

$$\sigma_{fiss} = \pi\lambda^2 g \sqrt{E} \frac{\Gamma_n^o \Gamma_f}{(E-E_o)^2 + (\frac{\Gamma}{2})^2}$$

with

$$g = \frac{1}{2} \left[1 \pm \frac{1}{2 I+1} \right]$$

In the equations, $2\pi\lambda$ is the wavelenth of a neutron with energy E, E_o is the energy of the neutron at resonance, I is the target nucleus spin and Γ_n^o, Γ_f, Γ_γ are the widths of levels for emission of a neutron, for fission and for emission of gamma radiation, Γ is the total width of the levels.

For U^{238} it is sufficient to take into account the fourth nearest resonances. The parameters of these resonances are(8)

E_o(ev)	Γ (mev)	Γ_n^o (mev)	Γ_γ (mev)
6;67	27.5	0.56	26
20.8	31.8	2.16	21.9
36.6	63	5.6	29
66	49	2.8	25.6

For U^{235} were considered the following resonances(9)

E_o(ev)	Γ (mev)	$2_g\ \Gamma_n^o$ (mev)	Γ_γ (mev)	Γ_{fiss} (mev)
-1.45	259	3.050	33	233
-0.02	97	0.00072	34	63
0.29	114.7	0.00516	32.3	82.5
1.135	148	0.0143	42	106

In the figure 3 it is shown the absorption cross-section calculated using the Breit-Wigner formula. It is also shown the calculated fission cross-section of UO_2 using the same formula.

For E_n < 5 mev the agreement with the experimental points is very good. In the Bragg edge region the discrepances may be attributed to the calculated coherent elastic scattering cross--section and to the not perfect simulation of the resolution effect on the theoretical scattering curve.

ACKNOWLEDGEMENT

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RESUMO

Foi medida a secção de choque total do óxido de urânio, para nêutrons de energia entre 0,08 ev e 0,0013 ev, usando o espectrômetro de tempo de vôo instalado no reator IEAR-1.

A secção de choque de absorção foi determinada subtraindo-se da secção de choque total medida a secção de choque de espalhamento calculada teòricamente afetada pela resolução experimental.

A secção de choque de absorção assim obtida é comparada com a calculada pela fórmula de Breit-Wigner. Observa-se uma concordância razoável entre os pontos experimentais e a curva teórica.

RÉSUMÉ

On a mesuré la section efficace totale de l'oxide d'uranium pour les neutrons d'energie entre 0.08 ev et 0.0013 ev. On a employé le spectromètre à temps de vol de l'IEA.

La section efficace d'absorption a été determinée en retirant de la section efficace de diffusion calculée theoriquement et affectée par la resolution expérimentale.

La section efficace d'absorption ainsi obtenue est comparée avec alle calculee par la formule de Breit-Wigner. On constate un bon accord entre les points experimentaux et la courbe theorique.

REFERENCES

(1) "Neutron Cross-Sections", vol. III, Second Editions, Supplement nº 2, report BNL 325 (1965).

(2) "Progress in Nuclear Energy", series I, vol. 1, ed. by R.A. Charpe, J. Horowits, D.J. Highes and D.J. Litter (1956).

(3) S.B. Herdade, L.Q. Amaral, C. Rodrigues and L.A. Vinhas, report IEA-136 (1967).

(4) T.D. Santos, H.M. Haydt, C.T. de Freitas - Report IEA nº 92.

(5) B.T. Wills, Proc. Royal Society of London, Series A, 274, 122 (1963).

(6) W. Marshall and R.N. Stuart, "Inelastic Scattering of Neutrons in Solids and Liquids", IAEA, Viena, 75 (1961).

(7) L.Q. Amaral, L.A. Vinhas, C. Rodrigues and S.B. Herdade - Nucl. Instrum, Meth. 63, 13, (1968).

(8) L.M. Bollinger, R.E. Coté, D.A. Dalleberd and G.E. Thomas, Phys. Rev. 105, 661 (1957).

(9) F.J. Shore and V.L. Sailor, Phys. Rev. 112, 191 (1958).