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ABSTRACT

The total neutron cross section of iron has been measured by transmission within the neutron energy range 0.2879-0.0016 eV, using a slow chopper time-of-flight spectrometer and a crystal spectrometer at the IPEN research reactor. The data were analysed to determine the total inelastic scattering cross section. The results obtained were compared with calculated values based on the Marshall-Stuart theoretical model and the agreement was quite good.

I – INTRODUCTION

The steel components, iron, nickel and chromium, as well as cobalt and zirconium, are important structural materials in the design of nuclear reactors and the precise determination of their total thermal cross sections is desirable.

The development of accurate computational methods for thermal reactor lattices has reached the point at which neutron cross section uncertainties are a major limitation to the confidence with which theory can be applied to practical situations.

In simple reactor systems the most significant uncertainties are largelly of two types: those which arise from approximations to the transport equation employed to simplify the analysis and those which result from uncertainties in the physical data, mainly the neutron cross sections used in the calculations. The mathematical approximations have been received most attention and from such studies the understanding of the behavior of neutron on a reactor lattice was improved. So the uncertainties in the basic cross sections even in the well-known thermal neutron range are still the major source of remaining error in the calculation of reactor characteristics.

In the thermal neutron energy range the ^{235}U and hydrogen cross sections are so well known that they contribute little to the total uncertainty in the multiplications and buckling. The principal uncertainty comes from the steel components Fe, Ni and Cr. The absolute errors in thermal cross section of these components should produce systematic differences between measurements and calculations of a reactor[8].

Except for iron, the measurements and data evaluation about thermal cross sections of the other structural materials for reactors mentioned here, were subject of previous papers[8,18].

In the present work the total cross section of iron has been measured in the energy interval 0.2879-0.0016 eV (or in wavelenght 0.533-7.091Å) by neutron transmission method.

The total inelastic scattering (coherent + incoherent) cross sections has been determined by subtracting the total elastic scattering and capture cross sections from the measured total cross section.

The inelastic scattering cross section is compared with calculations based on the theoretical model formulated by Marshall and Stuart[13,14] and the experimental points are fitted to a polynomial sum in order to be represented by an analytical expression.

II – EXPERIMENTAL

II.1 – Transmission Measurements

The total neutron cross section, σ_T, at each energy is obtained by measuring the transmission of the specimen with monochromatic neutrons. When a collimated neutron beam is perpendicularly incident to the plane surface of a sample, the measured transmission, T, or the ratio between the transmited I and the incident I_o intensities is given by:

$$T = I / I_o = \exp(-N x \sigma_T) \qquad (1)$$

where N is the number of atoms per cm^3 of the sample, x is the sample thickness and $\sigma_T = \sigma_c + \sigma_s$ is the microscopic total cross section of the atom, composed by the capture cross section σ_c and by the scattering cross section σ_s. N is given by:

$$N = \frac{\rho N_o}{A} \qquad (2)$$

where N_o is Avogadro's number, A is the atomic weight and ρ is the density of the sample. For samples in powder form, which is the case for the iron sample used, the density to be used in the calculation depends on the powder compactness, and is computed from mass and volume measurements.

The neutron source for this work was the IPEN swimming pool research reactor operated at 2 MW. Neutron transmissions through the iron samples were measured using the IPEN single crystal spectrometer in two different energy ranges: 0.2879-0.0356 eV (or 0.533-1.516Å) and 0.0051-0.0016 eV (or 4.008-7.091Å). For neutrons with energies in the intermediate interval 0.1000-0.0027 eV (or 0.9-5.5Å) a curved slit slow neutron chopper and time-of-flight spectrometer was used. All the experiments have been carried out at room temperature (295-297K).

II.2 – The Sample

The iron sample was in powder form (from Carlo Erba Co.) with purity higher than 99%. A spectrographic analysis showed the presence of some impurities of low total cross sections, such as: Mn(100ppm), Mg(500ppm), Ni(150ppm), Cu(100ppm) and P($<$ 10ppm).

The iron powder samples was placed in an aluminum holder; and for the I_o incident intensity measurements, a similar but empty aluminum holder has been used in order to eliminate the effect of the lateral plates of the holder.

II.3 – Crystal Spectrometer

The IPEN crystal spectrometer has been described thoroughly in previous paper[6]. Its operation is based on the selective diffraction from a single crystal, governed by the Bragg equation for coherent elastic scattering, given by,

$$\lambda = (2d \sin\theta) / n \qquad \text{with } n = 1,2,3,\ldots \qquad (3)$$

where: n is the order of reflection, λ is the neutron wavelength, d is spacing of the appropriate crystal planes and θ is the glancing angle of the neutron beam with respect to these planes.

From the wave properties of neutrons, one can convert wavelength to energy, using the relationship $\lambda = 0.286/\sqrt{E}$, with λ in Angstrons and E in electronvolts. The several neutron wavelengths are selected changing the Bragg angle θ.

Since any neutron which satisfies equation (3) can be diffracted, higher order contamination is always present in the reflected beam, which has neutrons with the desired wavelength λ and also wavelengths λ/2, λ/3, etc. Polycrystalline filters, which transmit only neutrons with wavelength greater than the cut-off given by twice the larger interplanar spacing, are commonly used to eliminate or attenuate higher order contamination. A beryllium polycrystalline filter, with a cut-off at 3.96 Å, was used in the present experiment.

The measurements of the total cross section of iron with the crystal spectrometer were made under the following experimental conditions: for the wavelength interval from 4.008 to 7.091 Å, a mica crystal monochromator and a beryllium filter placed inside the beam hole were used; in the interval from 0.533 to 1.516 Å a copper(111) crystal monochromator was used and the beryllium filter was removed because at this wavelength interval the higher order contamination can be neglected[6]. The crystal monochromator has been displaced from its maximum Bragg reflection position for the background measurements.

II.4 – Time-of-Flight Spectrometer

The IPEN curved slit slow-neutron chopper and time-of-flight spectrometer has been described elsewhere[1,9]. This experimental apparatus was used only for the intermediate wavelength interval since at high and slow wavelengths the crystal spectrometer is more suitable.

The chopper was placed in front of a tangential beam hole or through tube, so that the reactor core was not "seen" directly, the neutron source being a volume of moderator (H_2O) contained in the portion of the through tube located in front of the core. The distance from the center of the neutron source to the center of the chopper was 3.30 m. A shielded boron trifluoride proportional counter 12" x 1" diameter, filled with BF_3 gas at a pressure of 60 cm of Hg, located at a known distance from the chopper, was utilized as neutron detector. Cadmium slits were used to define the neutron beam between chopper and detector. A small low efficiency BF_3 detector (99% transmission for 2200 m/s neutrons) 1" long x 1/4" diameter was located between the reactor beam hole and the chopper, for monitoring the neutron flux.

Experiments have been carried out with good wavelength resolution because the polycrystalline iron total cross section curve presents discontinuities due to crystalline effects[2]. So, high chopper speeds, from 10000 to 13000 RPM, were used[19]. An 8 μsec channel length in a TMC 1024 channel time-of-flight analyser and flight paths of 3.0m for wavelength from 0.9 to 3.0 Å and 1.6 m for wavelength from 3.0 to 5.5 Å, were adopted. The wavelength resolution varied from 0.04 Å at λ = 1. Å to 0.1 Å at λ = 5.5 Å.

Transmission measurements were made in the usual way, with the sample in and out the pulsed beam; background measurements were made with a 0.7 mm thick cadmium plate located between the sample position and the detector.

The following calculations were carried out for each analyser channel: correction for counting losses; normalization of the counting data with respect to monitor reading, subtraction of the measured background; total neutron cross section computation from transmission measurements, with statistical errors; calculation of the wavelength for each channel number, according to the overall calibration.

III – RESULTS AND DISCUSSION

III.1 – Experimental Total Cross Section

A series of σ_T measured values for polycrystalline iron at room temperature are shown in Figure 1. These data, with only the statistical errors indicated, represent the result of an average made with at least two independently measured values. For almost all the experimental values the errors are smaller than 2% and in the wavelength interval 1.0-3.4 Å error bars are smaller than the dimensions of the points.

With the aim of making a comparison, all the experimental results previously published[7,10,11,12] and compiled in CINDA 76/77[4] and CINDA 79[5], are also shown in Figure 1. The main discrepancies near the Bragg edge regions can be attributed to the poor resolution of these measurements.

III.2 – Theoretical Cross Sections

When a beam of thermal neutrons interacts with a system of nuclei, such as the polycrystalline iron sample, besides neutron capture, four different types of scattering may be distinguished[3]: elastic coherent, elastic incoherent, inelastic coherent and inelastic incoherent. So, the total cross section is a sum of five terms and for iron the magnetic scattering contribution must also be considered.

The capture cross section and the elastic scattering cross section are easily calculated; nevertheless, due to the slow convergence of the sum over all kinds of multiphonon processes and integrals over the phonon spectrum of the material, several difficulties appear in the evaluation of inelastic scattering cross sections. Therefore, in the present work, the total inelastic scattering (coherent plus incoherent) cross section has been determined by subtracting the theoretical capture and elastic scattering cross sections from the measured total cross section.

The capture cross section, σ_c, of iron is calculated using the equation: $\sigma_c(\lambda) = 1.409\,\lambda$ (Å) barns, which is obtained from the value of the capture cross section at thermal energy, σ (1.80 Å) = (2.55 ± 0.05) barns[11] and considering the 1/v energy dependence.

The elastic coherent scattering cross section, with nuclear and magnetic contributions included, is given by[2]

$$\sigma_{coh}^{elast} = \frac{N_c \lambda^2}{2c} \sum_{d_{hkl} > \lambda/2} [\,(F_{Nuc}^2 + q^2\ F_{Mag})\, dj\ e^{-2w}\,]_{hkl} \quad (4)$$

where N_c is the number of unit cells per cm^3, c is the number of nuclei per unit cell, hkl are the Miller indices, d is the interplanar distance, e^{-2w} is the Debye-Waller factor and j is the multiplicity factor for each hkl family of planes.

The unit-cell structure factor, F_{Nuc} is given by

$$F_{Nuc}\ (h,k,l) = b_{coh} \sum_{j=1}^{c} e^{2\pi i\,(hx_j + hy_j + lz_j)} \quad (5)$$

b_{coh} being the coherent scattering amplitude and x, y, z the coordinates of the atoms in the unit cell.

The magnetic structure factor, F_{mag}, obbeys a formula similar to that one given above for F_{Nuc}, but with b_{coh} being substituted by p which is the magnetic scattering amplitude. When h + k + l is an odd number, both F_{Nuc} and F_{mag} are zero; when it is an even number, they are $2b_{coh}$ and $2p$, respectively. The q^2 that multiplies F_{mag} in the equation for σ_{coh}^{elast}, comes from a vector defined by[2] $\vec{q} = \vec{\varepsilon}(\vec{\varepsilon}\cdot\vec{K}) - \vec{K}$ with $\vec{K}$ and $\vec{\varepsilon}$ denoting the magnetization vector and the scattering vector,

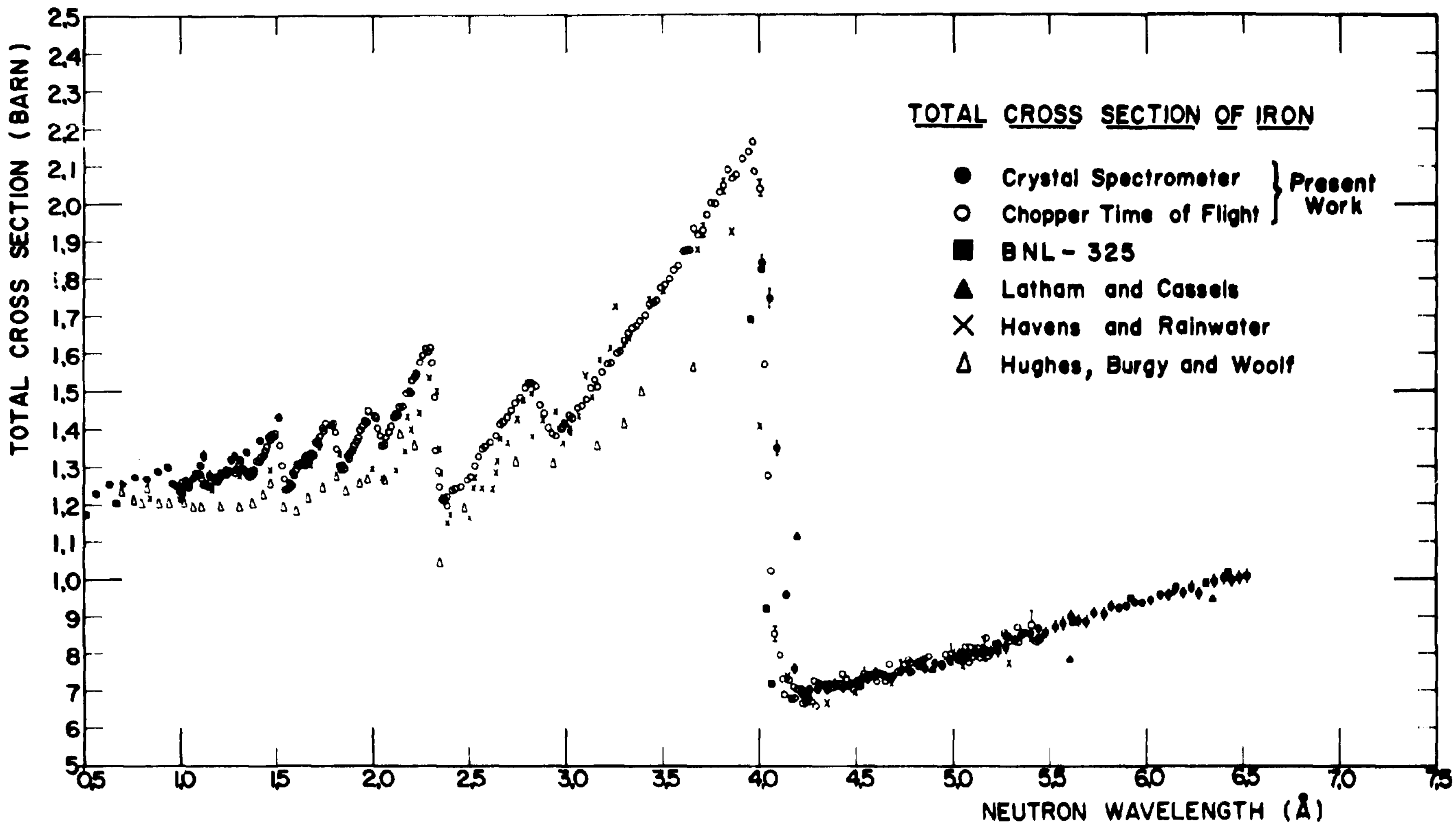

Figure 1 – Total cross section of polycrystalline iron at room temperature

respectivelly.

The elastic incoherent scattering cross section is given by[3]

$$\sigma_{inc}^{elast} = \frac{\sigma_i \lambda^2}{Y} \left[1 - \exp\left(- \frac{Y}{\lambda^2}\right) \right] \qquad (6)$$

with σ_i being singles nucleus incoherent scattering cross section and Y following the mathematical expression:

$$Y = \frac{12h}{Mk\theta} \left[1/4 + \Lambda\left(\frac{\theta}{T}\right) \right] \qquad (7)$$

where h is the Planck constant, k is the Boltzmann constant, M is the atomic mass, $\Lambda\left(\frac{\Theta}{T}\right)$ is the function, T and Θ are the sample and the Debye temperature respectivelly.

All the parameters for iron used in the calculations are listed in Appendix 1.

III.3 – Total Inelastic (coherent plus incoherent) Scattering Cross Section

The determination of the total inelastic scattering cross section was made by subtracting the capture and the total elastic scattering cross sections, calculated following the formalism shown in item III.2, from the total cross section measured. The data for the total inelastic scattering obtained by this procedure are shown in Figure 2. The error attributed to each point is the same as that of the experimental value of total cross section used in its determination.

Theoretical values of σ_{tot}^{inel}, calculated following the model introduced by Marshall and Stuart[13] and also described by Marshall and Lovesey[15], are represented in the figure as the full line. By this model, the difficulties in the calculation of inelastic scattering cross sections caused by the multiphonon processes, are solved by two suggestions introduced by Plackzek[16,17]: incoherent approximation and mass expansion.

When a scattering process involves two or more phonons, the conservation of energy and momentum is not restrictive. Therefore the multiphonon scattering tends to be a smoothly varying function of scattering angle and incident energy. In these circunstances the interference of the scattering from two different nuclei can be neglected. Thus the cross section can be evaluated as though all the scattering was incoherent. This is known as Plackzek's incoherent approximation.

Plackzek also noticed that if, in place of a multiphonon process expansion, the cross section is rearranged as an expansion in the ratio of neutron mass(m) to nuclear mass(M), then a rapidly convergent series is obtained and, generally, the sum of the first three terms is sufficient to obtain good results. This series is known as Plackzek mass expansion.

According to the Marshall model[13], the total inelastic scattering cross section, $\sigma_{tot}^{inel} = \sigma_{inc}^{inel} + \sigma_{coh}^{inel}$, is obtained as following:

Using the mass expansion, the total incoherent cross section is given by

$$\sigma_{inc}^{tot} = \sigma_i \left\{ 1 + \left(\frac{m}{M}\right) A_1(x,t) + \left(\frac{m}{M}\right)^2 A_2(x,t) + \left(\frac{m}{M}\right)^3 A_3(x,t) \right\} \qquad (8)$$

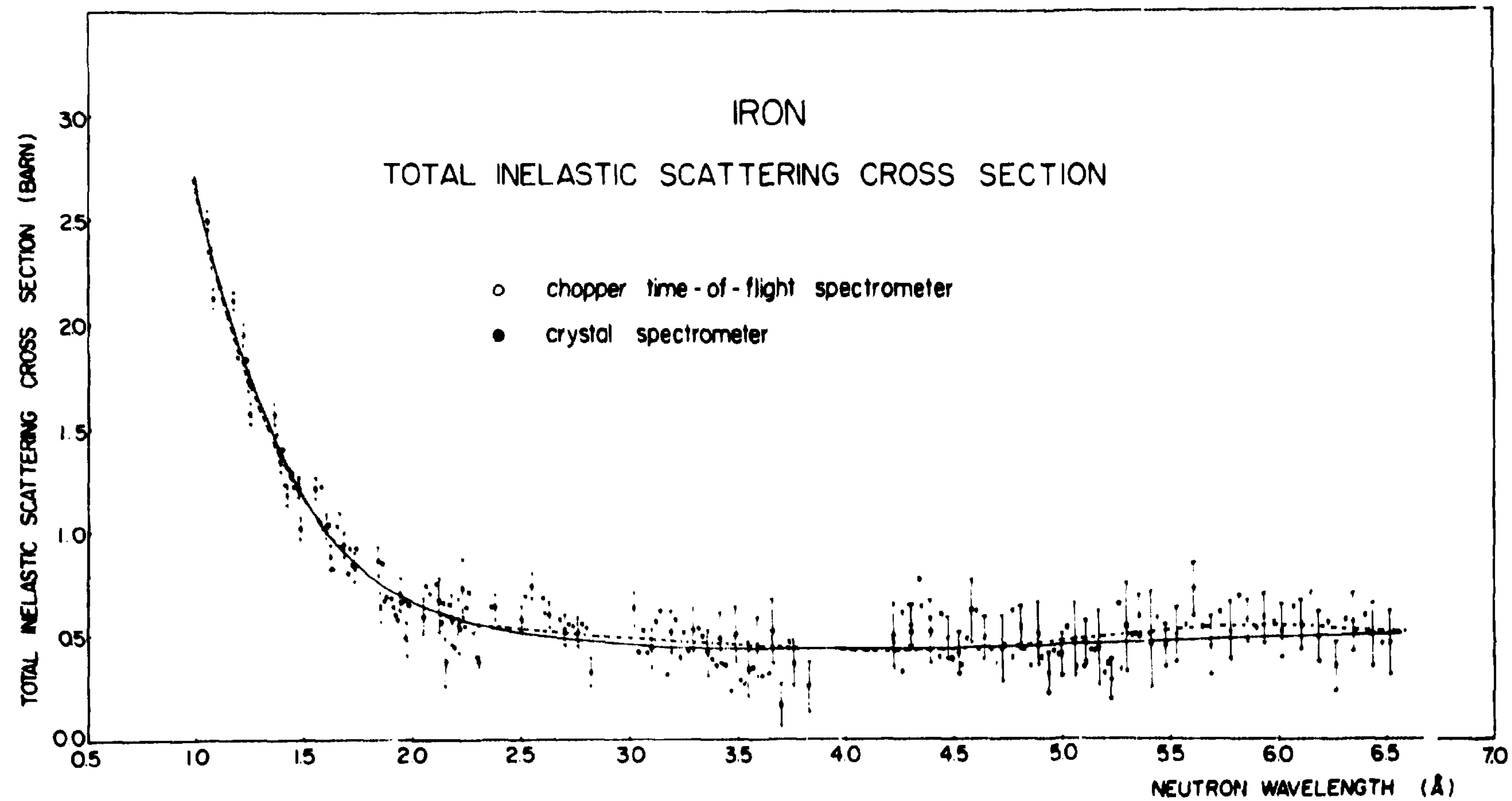

Figure 2 – Total inelastic scattering cross section of iron. The full line represents the theoretical values calculated according to Marshall and Stuart model. The dashed line represents a polynomial sum adjusted by last square fit

where $x = \sqrt{\frac{E}{k\Theta}}$ and $t = \frac{T}{\Theta}$; coefficients A_1, A_2 and A_3 have been calculated by Marshall and Stuart and are tabulated in references[13]. The σ_{inc}^{inel} was obtained by subtracting from σ_{inc}^{tot} the elastic incoherent cross section given by expression (6).

Finally, considering the incoherent approximation, the inelastic coherent scattering is given by

$$\sigma_{coh}^{inel} = \frac{4\pi b_{coh}^2}{\sigma_i}\,\sigma_{inc}^{inel} \qquad (9)$$

In Figure 2, the satisfactory agreement between the calculated curve and the experimental points can be seen, indicating the properness of the theoretical model employed.

For nuclear data purpose it is convenient to describe the total inelastic scattering cross section by a simple analytical expression, independently of the tabulated coefficients. With this aim, the determined values of total inelastic cross section were adjusted by a polynomial sum by least squares fit. The best fit was obtained using the following mathematical expression.

$$\sigma_{tot}^{inel} = 13.369994 - 19.850450\,\lambda + 12.568190\,\lambda^2 - $$
$$- 4.147218\,\lambda^3 + 0.747187\,\lambda^4 - 0.069507\,\lambda^5 + $$
$$+ 0.0026095\,\lambda^6 \qquad (10)$$

The dashed line in Figure 2 represents the total inelastic scattering calculated by the above expression.

IV – CONCLUSION

By measuring a large number of points with good precision and resolution and by the calculations performed, the present work intends, to give an improvement to the knowledgement of the total neutron cross section of iron at thermal an subthermal energy regions.

From the total inelastic scattering cross section determination, an analysis of the validity of the Marshall and Stuart model was made and also a simple analytical expression was obtained describing this partial cross section, which allows its use as nuclear data.

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APPENDIX I

Parameters and constants for iron used in the theoretical calculations of partial cross section:

- Capture cross section (thermal energy): σ_{cap} = 2.66 barn
- Coherent scattering cross section: σ_{coh} = 11.37 barn

– Incoherent scattering cross section: $\sigma_{inc} = 0.43$ barn

– Coherent scattering amplitude: $b_{coh} = 0.951 \times 10^{-12}$ cm

– Lattice parameter: $a_o = 2.86106$ Å

– Atomic mass: 55.85 amu

– Debye Temperature: 453K

– Parameter $q^2 = 2/3$

– Magnetic scattering amplitude $p = \left(\frac{e^2}{mc^2}\right) S_{eff} f . \mu_n$

where

$\left(\frac{e^2}{mc^2}\right)$ – classical radius of electron

S_{eff} – effective spin quantum number = 1.11

μ_n – magnetic moment of neutron in nuclear magnetons = 1.913

f – amplitude form factor given by Steinberg and Wick (Phys. Rev. 76:994(1949)).

RESUMO

Foi medida a secção de choque total do ferro para nêutrons, por transmissão, no intervalo de energias entre 0.2879 a 0.0016 eV, utilizando um espectrômetro de tempo de vôo e um espectrômetro de cristal instalados no reator de pesquisa do IPEN. Os dados foram analisados de modo a determinar a secção de choque total de espalhamento inelástico. Os resultados obtidos foram comparados com valores calculados com base no modelo teórico de Marshall-Stuart.

REFERENCES*

1. AMARAL, L. Q.; VINHAS, L. A.; RODRIGUES, C.; HERDADE, S. B. Certain aspects of the calibration and resolution of slow neutron spectrometers. *Nucl. Instr. Methods*, 63:13-22, 1968.

2. BACON, G. E. *Neutron diffraction*. Oxford, Clarendon, 1962.

3. CASSELS, J. M. The scattering of neutrons by crystals. *Prog. Nucl. Phys.*, 1:215, 1950.

4. CINDA 76/77. Vienna, International Atomic Energy Agency, 1977.

5. CINDA 79. Vienna, International Atomic Energy Agency, 1979.

6. FULFARO, R. *Estudo das reflexões de ordem superior em cristais monocromadores de neutrons*. Campinas, 1970. (Tese de doutoramento, Universidade Estadual de Campinas).

7. HAVENS JR., W. W. & RAINWATER, L. J. Slow neutron velocity spectrometer studies. IV. Au, Ag, Br, Fe, Co, Ni, Zn. *Phys. Rev.*, 83(6):1123-36, 1951.

(*) Bibliographic references related to documents belonging to IPEN Library were revised according with NB-66 of Associação Brasileira de Normas Técnicas.

8. HELLENS, R. L. *The sensitivity of the reactor characteristics to cross section incertainties below 100 eV.* Upton, N. Y., Brookhaven National Laboratory, Mar. 1966. (BNL-10058)

9. HERDADE, S. B.; AMARAL, L. Q.; RODRIGUES, R.; VINHAS, L. A. *Experimental study of a curved slit slow-neutron chopper and time-of-flight spectrometer.* São Paulo, Instituto de Energia Atômica, Feb. 1967. (IEA-Pub. 136).

10. HUGHES, D. J.; BURGY, M. T.; WOOLF, W. E. Scattering and polarization of neutrons in an iron single crystal. *Phys. Rev.*, 80:481-2, 1950.

11. HUGHES, D. J. & SCHWARTZ, R. B. *Neutron cross sections.* Upton, N. Y., Brookhaven National Laboratory, 1958. (BNL-325).

12. LATHAM, R. & CASSELS, J. M. The inelastic scattering of very slow neutrons by iron. *Proc. Phys. Soc. (London)*, 65A:241-5, 1952.

13. MARSHALL, W. & STUART, R. N. *The scattering of neutrons from polycrystalline materials.* Livermore, Calif., Lawrence Radiation Laboratory, Apr. 1959. (UCRL-5568).

14. MARSHALL, W. & STUART, R. The scattering of neutrons from polycrystalline materials. In: INTERNATIONAL ATOMIC ENERGY AGENCY. *Inelastic scattering of neutrons in solids and liquids: proceedings of a symposium on..., held in Vienna, 11-14 Oct., 1960. Vienna, 1961.* p.75-85 (proceedings series).

15. MARSHALL, W. & LOVESEY, S. W. *Theory of thermal neutron scattering: the use of neutrons for investigation of condensed matter.*

16. PLACZEK, G.; NIJBOER, B. R. A.; VAN HOVE, L. Effect of short wavelength interference on neutron scattering by dense systems of heavy nuclei. *Phys. Rev.*, 82(3):392-403, 1951.

17. PLACZEK, G. Incoherent neutron scattering by polycrystals. *Phys. Rev.*, 93:895-6, 1954

18. SALAMA M.; ADIB, M.; ABDEL-KAWI, A.; EL-MINIAWY, S.; EL-KHOSHT, M.; HAMOUDA, I. Measurements of the total neutron cross section of chromium, cobalt and zirconium in the energy range 3 MeV to 1 eV. *Atomkernenergie*, 22887-89, 1973.

19. VINHAS, L. A. *Estudo de interação de nêutrons lentos com o ferro policristalino.* Campinas, 1970. (Tese de Doutoramento, Universidade Estadual de Campinas).

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