International Atomic Energy Agency

INDC(EGY)-003/L

INDC INTERNATIONAL NUCLEAR DATA COMMITTEE

MEASUREMENTS OF THE TOTAL NEUTRON CROSS-SECTIONS OF U AND UO_2 BELOW 2 eV AT DIFFERENT TEMPERATURES

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The data is available as EXFOR 30636

November 1982

IAEA NUCLEAR DATA SECTION, WAGRAMERSTRASSE 5, A-1400 VIENNA

Reproduced by the IAEA in Austria
November 1982

82-7150

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ABSTRACT

The total neutron cross-sections of natural uranium and its oxide are measured using two time of flight spectrometers, installed in front of two of the ET-RR-1 reactor horizontal channels, and also by a neutron diffraction spectrometer. The measurements were carried out at room temperature in the energy range from 2 eV-0.002 eV and at 210°C, for neutron energies below 0.005 eV.

The coherent scattering cross-section of U was deduced both from the Bragg cut-offs observed in the behaviour of the total neutron cross-section of both U and UO_2 at cold neutron energies and the neutron diffraction pattern obtained at room temperature.

1. Introduction

Work has been started at the ET-RR-1 reactor on measurements of the total cross-sections of U and UO_2 at neutron energies below 1 eV. These measurements are carried out with time-of-flight technique, for micro-crystalline samples, at temperatures starting from the liquid nitrogen temperature and up to 1000°K. Such work should provide some new values for the incoherent and thermal diffuse scattering cross-sections. These are required both for the build up of the fuel element spectra, and the study of the effects of the phase transformation in the uranium crystal structure.

The total neutron cross-section measurements of U and UO_2 were carried out by Beshai [1] in the neutron energy range from 4.5 meV to 25 meV at 20°C. This work also contains some calculations, carried out both for U and UO_2, concerned with the energy positions of the Bragg edges. However Beshai reported that the measurements at 750°C were not

successful due to the changes which take place in the sample's density[1].
The present work deals with total neutron cross-section measurements carried out both for U and UO_2 in the energy range from 2.5 meV-1.0 eV at room temperature and at 483°K.

2. EXPERIMENTAL DETAILS

2.1. The Natural Uranium Sample

The natural uranium sample used is from U-0.05 at.% Ru alloy. The alloy was prepared under vacuum, in a Cu crucible, using an electron-beam melting furnace. The Ru powder was compressed into small pellets which were placed underneath the U-button before starting the melting operation. To obtain well homogenized alloy, the specimens were melted four times and were inverted after each melt. Chemical analysis of 3 specimens obtained from each button showed that the alloy was perfectly homogeneous. Gamma graphic examination of every button showed that the specimens were perfectly sound and contained neither defects nor cavities. The chemical analysis of the uranium used in these experiments is given as : (in ppm weight average)

Cd	Cr	Cu	Fe	Mn	Ni	Si	Na	C	O_2	N_2
<0.5	6	5	50	5	21	27	5	295	53	44

Al	As	Co	Li	Mg	Pb	K	Ag	Be	H_2
<20	<5	<2	<2	10	<3	<20	3	<0.2	5

From the chemical analysis one can see that the contribution due to impurities, as well as the inserted Ru, to the total cross-section of U sample does not exceed 0.5 barns at the thermal point. One U sample of thickness 15.8 gm/cm^2 was used during the present cross - section measurements.

The lattice parameters a,b and c of the orthorombic alpha uranium lattice having addition of 0.05 at. % Ru were calculated from the x-ray diffraction charts. These charts were obtained from the U specimens using a Siemens x-ray diffractometer. Only the reflections of x-rays from the surface layers of a polycrystalline polished surface were recorded. The radiation employed was that from a cobalt target with an iron filter to eleminate the K -radiation. The diffraction lines, using a scintillation counter, were obtained over the range 2θ = 11-145°. The x-ray diffraction pattern of the U-0.05 at.% Ru alloy showed that the (111), (002), (021), and (110) peaks were shifted from those of α-uranium to higher 2θ values, thus indicating contraction of the a,b and c parameters. These peaks were used for calculation of the lattice parameters by a trial and error method. The results are listed in Table I. It was pointed out that Ru is an effective grain refining element when added to uranium even in very small amounts.

Table 1: Lattice Parameters of the U-0.05 at.% Ru Alloy

Ru (at.%)	2θ (deg) for α-Uranium (h,k,l)				Lattice Parameters (Å)			Volume of U.C (Å)³	Grain Size
	(110)	(021)	(002)	(111)	a	b	c	a.b.c.	μ
0	40.80	41.50	42.34	46.24	2.848	5.861	4.944	82.5259	200
0.05	40.82	41.52	42.35	46.25	2.846	5.860	4.942	82.4205	100

2.2. The UO_2 Sample

The sample used was prepared from spec. pure fine UO_2 powder (grain size less than 20 μ) The UO_2 powder is packed in a Cu container with a thin Cu window (0.2 mm thick). One sample of thickness 8.43 gm/cm^2 was used during the measurements.

The neutron diffraction pattern of the UO_2 powder was measured using the crystal diffractometer, installed in front of one of the horizontal channels of the ET-RR-1 reactor. Monochromatic neutrons with wavelength λ =1.02 Å selected out of the reactor spectrum by diffraction from Zn single crystal cut along the (002) plane. The UO_2 sample was enclosed in a vanadium cylindrical container (5 cm height and 1.5 cm in diameter). The neutron diffraction pattern obtained for angles 2θ between 10°-60° at room temperature is represented in Fig. 1. Eight well separated peaks were identified confirming the UO_2 crystal structure.
The integrated intensities for the observed reflections were calculated using the area method and then corrected for background and Lorentz factor. The coherent scattering amplitude of U was deduced by fitting the corrected integrated intensities using the reliability factor method. The minimum reliablity factor R was found to be 3% for a coherent scattering amplitude value b_u(8.5±0.2)fm of uranium, with a value of the coherent scattering amplitude of oxygen b_o=5.8 fm

2.3. Total Neutron Cross-Section Measurements

The measurements were performed using two TOF spectrometers installed in front of two of the ET-RR-1 reactor horizontal channels. The measurements were carried out for neutrons below 5 meV using an automatically controlled heater, where the temperature could be fixed within ±5°C in the interval between 25-500°C. The spectrometer's resolution at different intervals of the whole energy range, under consideration, could be varied from 3 μ sec/m to 20 μ sec/m. The spectrometers are described in Ref[2-4].

3. RESULTS AND DISCUSSION

3.1. Natural Uranium

Fig. 2 shows the dependence of the total neutron cross-section (closed circles), on both wavelength and energy, as measured for natural uranium at room temperature in the energy range from 2.0 eV-2 meV. In the same figure are also represented the total neutron cross-sections (open circles) measured at 483°K for neutron energies below 5 meV. The errors indicated in Fig. 2 are statistical ones. The contribution due to ^{235}U was incoherently subtracted from the measured total cross-section ; the residual cross-section (^{238}U) is also given in Fig. 2 (as closed triangles). From Fig. 2, one can see that the behaviour of the uranium cross-section measured at room temperature is in good agreement with the reported in the BNL-325 [5].

The cut-off values of $\Delta\sigma_{hkl}$ due to reflection of neutrons with wavelengths λ, from planes with Miller indices hkl, the multiplicity factor M_{hkl} and inter planer distances d_{hkl} are given in Table 2. The coherent scattering amplitude of U (last column of Table 2) is calculated taking into account both the spectrometer's resolution and the Debye Waller factor. The average value of b_u was determined and found to be (8.4±0.2) fm Consequently the coherent scattering cross-section of U is σ_s=(8.9±0.4) barns. This value is in good agreement with the values (8.5±0.06) fm and (8.90±0.16) b reported in Ref. [5].

The difference between the U total cross-section measured at 483°K and that measured at room temperature was attributed to the contribution of the one phonon annihilation process. The measured difference was found to be (1.2±0.2) b at neutron energy 3 meV. This difference was calculated, under the same conditions of Ref.[6],

Table 2: The U Bragg cut-offs

h k l	M_{hkl}	d_{hkl} Å	λ_{hkl} Å	$\Delta\sigma_{hkl}$ (barns)	b_{coh} (fm)
110	4	2.568	5.136		
021	4	2.5248	5.0497		
002	2	2.475	4.950	11.0±2.0	8.5±0.8
111	8	2.278	4.557		
112	8	1.782	3.565	8.0±1.0	8.4±0.5
131	8	1.534	3.069	1.6±0.5	8.3±1.3
023	4	1.4384	2.8768		
041	4	1.406	2.813	1.3±0.5	8.4±1.6
110	8	1.387	2.774		
042	4	1.261	2.523		
221	8	1.242	2.484	2.6±0.5	8.4±0.7
133	8	1.1534	2.3069		
114	8	1.1143	2.2286		

considering a U Debye temperature 207°K and found to be 0.8 b. Such slight disagreement may be due to the contribution of the multi-phonon process.

3.2. The uranium oxide

Fig. 3 shows the dependence of the total neutron cross-section (closed circles) measured for UO_2 at room temperature in the energy range from 2 eV-2 meV. The cross-sections measured for neutron energies below 5 meV at 483°K are also represented in the same figure (open circles). The measured cross-sections are in good agreement with those reported before [1], in the energy range from 0.028 eV-4.5 meV.

The coherent scattering amplitude of U was calculated from the Bragg cut-offs, observed in the total cross-section of UO_2 at room temperature,(see Table 3)

Table 3: The UO_2 Bragg cut-offs

h k l	M_{hkl}	d_{hkl}(Å)	λ_{hkl}(Å)	$\Delta\sigma_{hkl}$(barns)	b_{coh} (fm)
220	12	1.9341	3.8663	14.0±2.0	6.6±0.5
311	24	1.649	3.299	4.4±0.6	9.1±0.5
400	6	1.367	2.735	3.0±0.5	8.4±0.7
331	24	1.2551	2.5102	1.7±0.3	8.6±0.6
422	24	1.1166	2.2333	6.6±0.2	8.5±0.5
440	12	0.9673	1.9346	2.2±0.1	8.7±0.5

considering that the coherent scattering amplitudes of U and oxygen are of the same sign. The value[5],5.8 fm was chosen for the coherent scattering amplitude of oxygen. It was found that the value of the U-coherent scattering amplitude is (8.6±0.5) fm which is in reasonable agreement with the value (8.5±0.2) fm calculated from the neutron diffraction pattern of the same sample.

The contribution due to the one phonon annihilation process in the total neutron cross-section was calculated for 3 meV neutron energy and the difference between the 483°K and the room temperature, and found to be 0.6 b. This value is of the same order of magnitude of the difference between the total neutron cross-sections measured at 483°K and room temperature.

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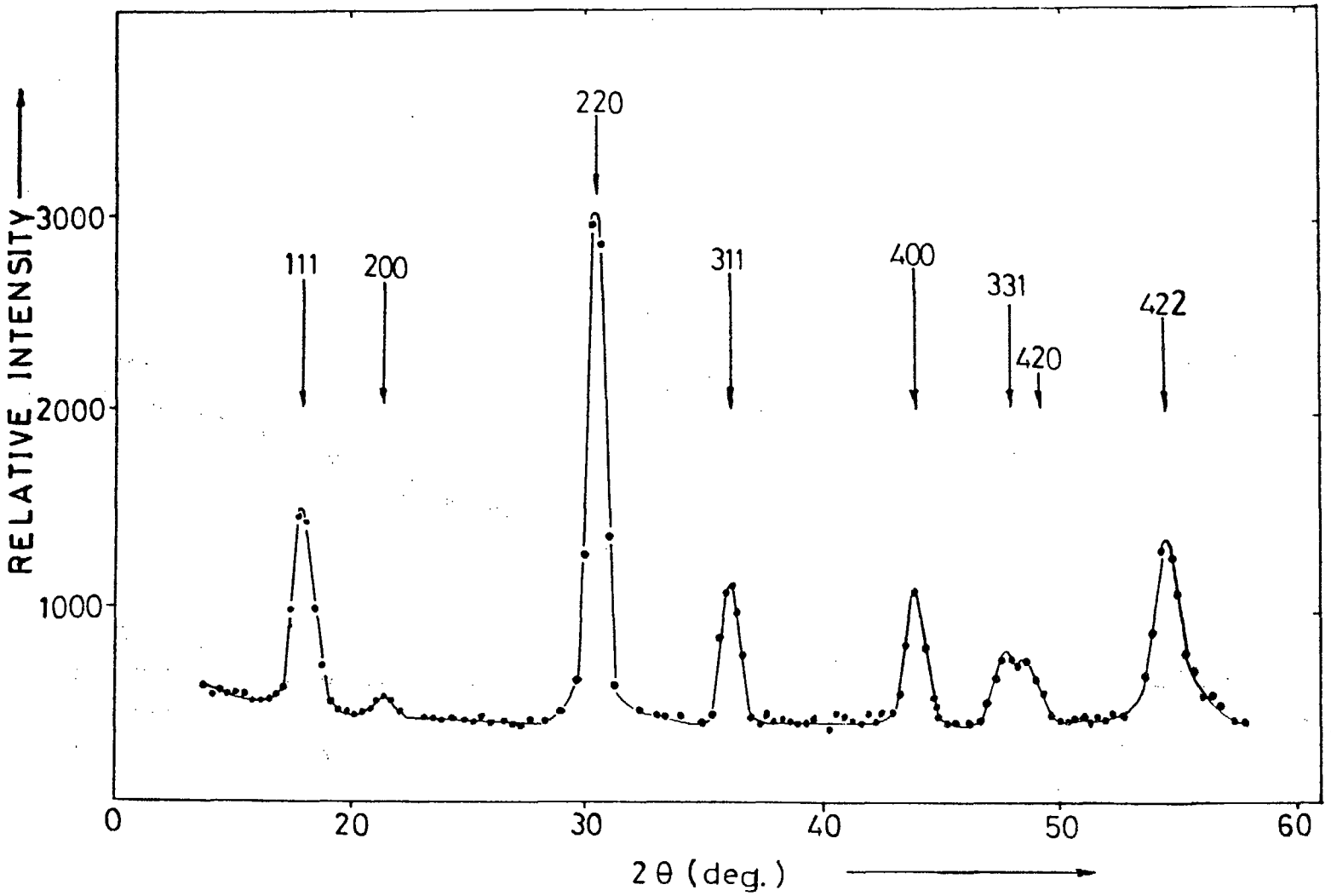
Figure Captions

Fig. (1) : The UO_2 neutron diffraction pattern obtained at room temperature.

Fig. (2) : The total neutron cross-section of U

Fig. (3) : The total neutron cross-section of UO_2



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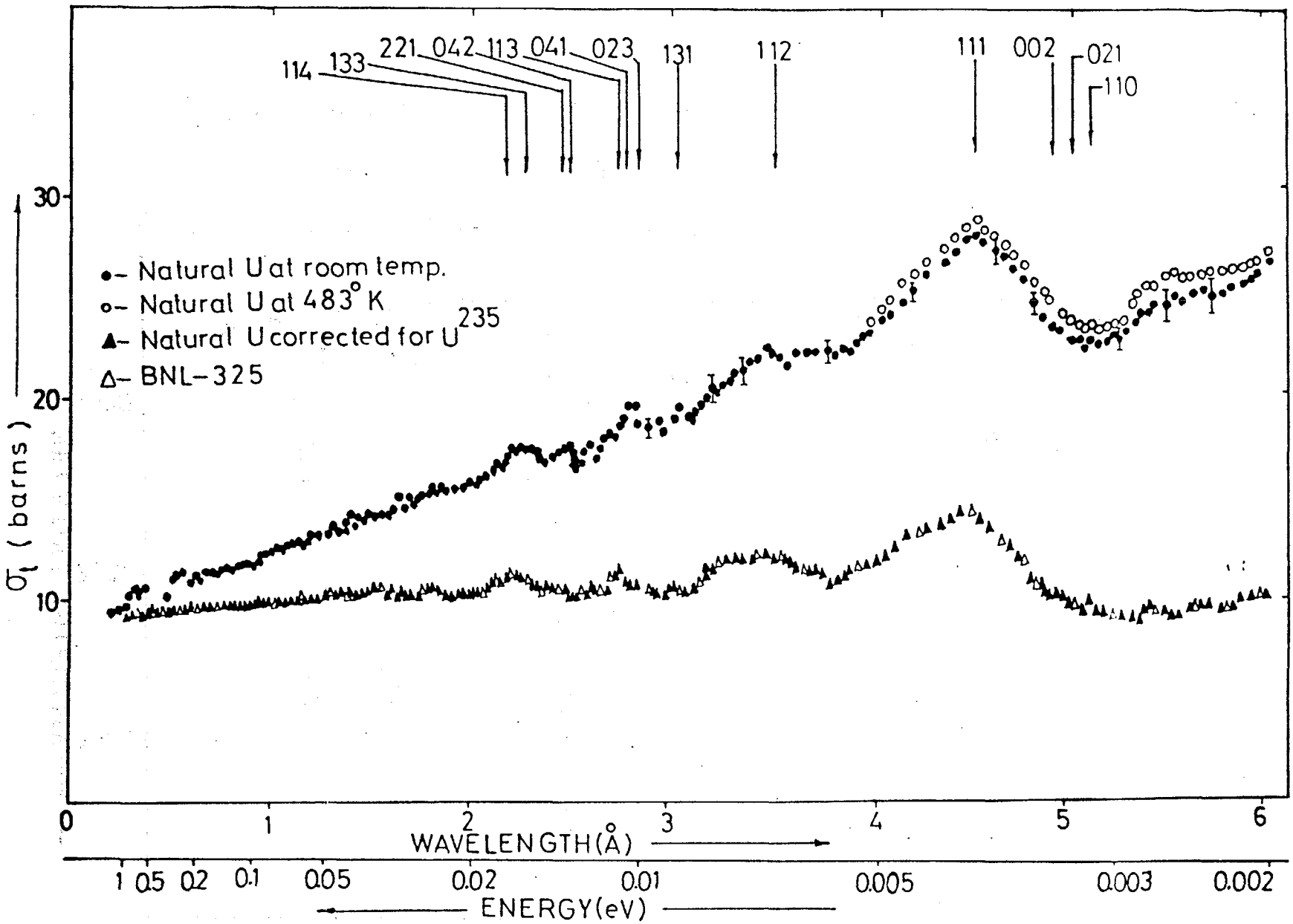

Fig. (2): The total neutron cross-section of U

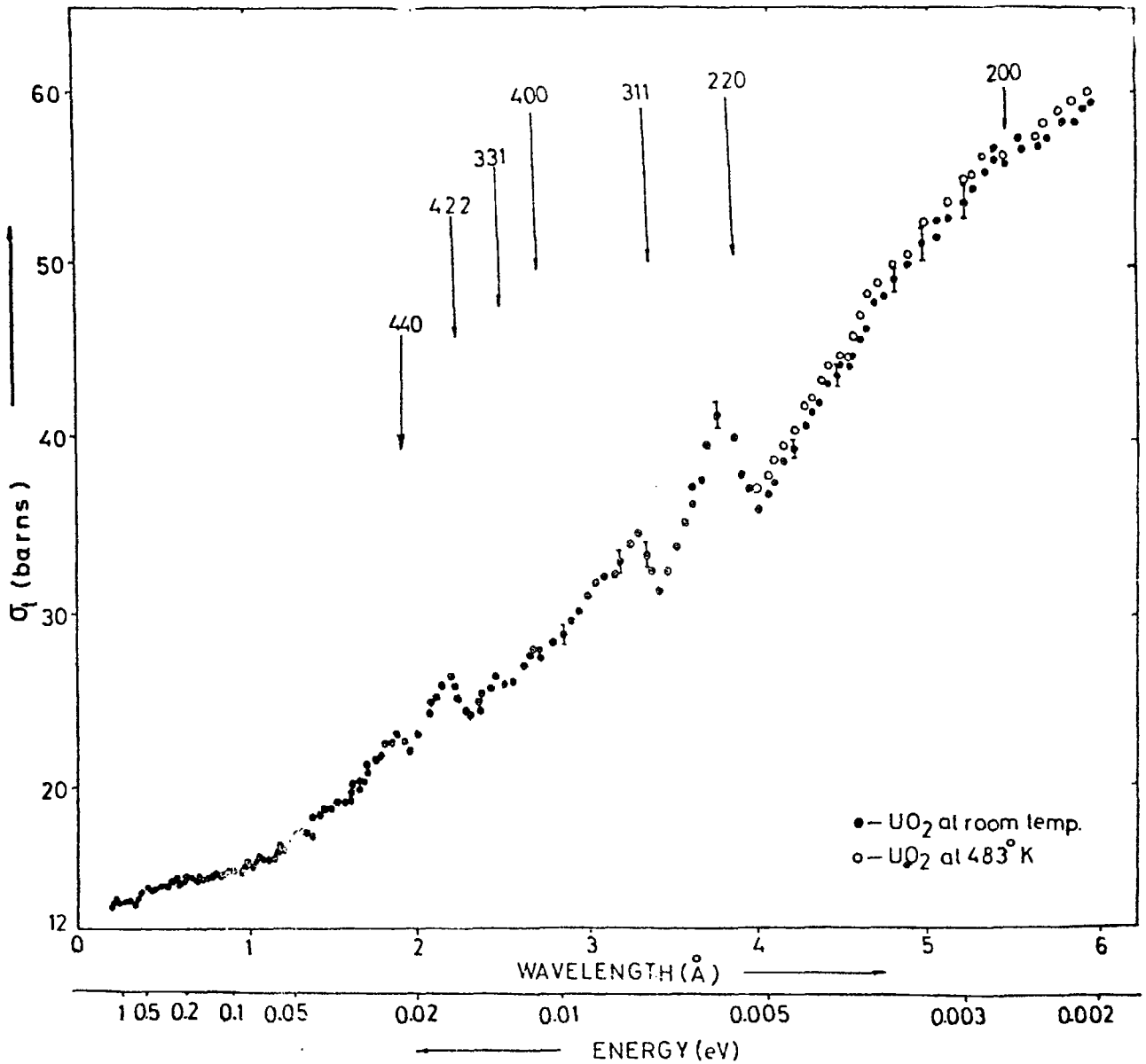

Fig. (3): The total neutron cross-section of UO_2