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Precision Neutron Total Cross Section Measurements for Natural Carbon at Reactor Neutron Filtered Beams

ABSTRACT: Experimental investigation of the total neutron cross section for natural carbon was fulfilled at Kyiv Research Reactor using neutron filtered beams with energies 24, 59 and 148 keV. The intense neutron beams formed by composite neutron filters at reactor horizontal channels had the fluxes of about 10^6 – 10^7 neutron/cm^2·s at the fixed neutron energies which enabled to measure the neutron cross sections with accuracy better than 1%. Transmission method was used in these measurements. The results of the measurements are presented together with the analysis of the known previous experimental data and the evaluated nuclear data from ENDF libraries. Sample thickness dependence of the observed neutron cross section, measured at the 148 keV filter, has been detected which hypothetically may be connected with existence of a very strong resonance in the ^{13}C neutron cross section in the energy range 119 - 157 keV.

KEYWORDS: carbon, research reactor, neutron filtered beam, total neutron cross section, transmission method.

Introduction

This investigation is devoted to the precise measurements of total neutron cross section for natural carbon. This element is well known as reactor structure material and at the same time as one of the most important scattering standards, especially at energies of less than 2 MeV, where the neutron total and elastic scattering cross sections are essentially identical. The best experimental data in the area 1-500 keV have the uncertainty 1-4% [1, 2]. However, the difference between these data and those founded within R-matrix analysis and unified for all ENDF libraries is evident (Fig.1) especially in the energy range 1-60 keV. The use of the technique of neutron filtered beam developed at the Kyiv Research Reactor makes possible to reduce the uncertainty of the experimental data to 1% and less [3, 4]. These high precision data on natural carbon could stimulate the new run of the R-matrix analysis for carbon.

Experimental Set-up and Measurements

Experimental investigation of the total neutron cross section for natural carbon was fulfilled on the eighth and ninth horizontal channels at the Kyiv Research Reactor (KRR). Experimental installations on horizontal reactor channels (Fig.2) include the systems of filtered neutron beam forming, neutron detector and registration systems, sample management systems and systems of radiation shielding.

The forming system includes the elements of beam collimation and neutron filtration on the way from reactor core to detector. The preliminary forming of necessary beam geometry is realized with two iron and boron carbide collimators. Further beam forming

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takes place in the first three discs of shutter and in outer collimator. By turns lead, textolite and mixture of paraffin with H_3BO_3 are used as material for these collimators. The collimation system provided beam narrowing to 12 mm/m, what corresponded to beam diameter at the sample in 10 mm.

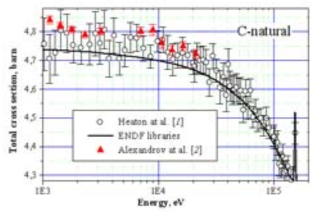

FIG. 1— *Natural carbon neutron total cross sections.*

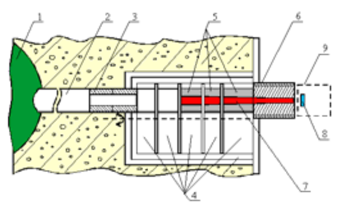

FIG. 2— *Construction of neutron filtered beam to receive the quasi-mono-energy neutron flux. 1 – Beryllium reflector; 2 – horizontal channel tube; 3 – preliminary collimator; 4 – beam shutter disks; 5 – filter-collimator assembles; 6 – outside collimator; 7 – filter components; 8 – sample for activation (optionally); 9 – device for samples removing.*

The elements of neutron filtration system take place in the first three disks of shutter and in outer collimator. To receive the quasi-mono-energy beams with the average energies 24, 59 and 148 keV we used the composite neutron filters consisted of Fe, Al, S, ^{10}B for the first, of ^{58}Ni, S, V, Pb, Al, ^{10}B for the second and of Si, Ti, ^{10}B for the third one. The filter component optimization, to receive the most possible intensity of the main energy line at the most optimal impurity of the parasitic energy lines in neutron spectrum, was carried out by means of calculation using our code FILTER_L [5].

The detection and neutron registration systems included: the proportional hydrogen recoil counters CHM-38 (Gas Filling: 90% H_2 + 9.56% CH_4 + 0.44% $^3He_2$, gas pressure 2280 Torr) or LND 281 (Gas Filling: H + CH^4 + N_2, gas pressure 3240 Torr), electronic blocks, personal computer IBM 286/287 and communication lines.

The management system for experimental samples provides the establishment of the samples on neutron beam with definite alternation. Simultaneously three samples can be loaded into the system and placed in the beam at any sequence and combination.

Tree type of carbon samples were used in these measurements: 1) solid samples from reactor graphite (C 99.9%); 2) powder samples, loaded into aluminum container (C 99.9%); 3) carbon discs, each of them has thickness 1mm and diameter 30.4 mm (C 99.997 %).

For determination of background counting rate the polyethylene samples with thickness 4.730 – 0.550 g/cm^2 were used. For high statistics accuracy, the measurements were carried out during 30-40 hours for each sample. To remove the influence of instability factors, the samples at neutron beam were replaced every minute.

Sample transmission was calculated for each carbon sample measurement series (from 2 to 25) with formulae:

$$T = \frac{N_{SMP} - N_{SNMP+PE}}{N_{DB} - N_{DB+PE}} \quad , \tag{1}$$

where

N_{SMP} – beam after sample,

N_{SMP+PE} – beam after sample + polyethylene,

N_{DB} – direct beam,

N_{DB+PE} – direct beam + polyethylene.

Then the transmission was averaged over 50 - 500 channels of 1024 channels of proton recoil counter. These averaged values then were averaged over all series of measurements. The total cross section for sample was determined as

$$\sigma_x = -\frac{1}{n_x} \ln < T > , \tag{2}$$

The total uncertainty included the statistical inaccuracy of measurements, sample weight and dimensions inaccuracies:

$$\Delta\sigma_x = -\frac{1}{n_x} \sqrt{\left(\frac{d < T >}{< T >}\right)^2 + (\sigma_x dn_x)^2} \ , \tag{3}$$

Filter 24 keV

We used here the composite neutron filter consisted of Fe, Al, S, B-10 to receive the quasi-mono-energy beam with the average energy 24.01±1.74 keV and neutron flux about $4.4.10^5$ n/cm^{2}s. The purity of beam was about 99.8%, other additions to the main spectra were negligible: 73 keV – 0.14%, 351keV – 0.02%.

The filter components (g/cm^2), used in these experiments for filtered neutron beam forming at the energy 24 keV, are presented in the Table 1.

TABLE 1—24 keV filter components.

^{10}B (85%)	Al	S	Fe
1.0	112.81	62.1	236.22

Fig. 3 show the calculated neutron spectra with peak energy 24.01 keV for filter used in experiment. The limits of 95% response function for 24 keV filter spectrum were defined as 19.3-25.8 keV.

Three samples of reactor carbon (99.9%) with thickness 0.1275925, 0.1751940 and 0.3027865 atoms/barn were used in these measurements. The final value of total neutron cross section, averaged over all samples was 4.687± 0.006 barn (relative accuracy 0.13%).

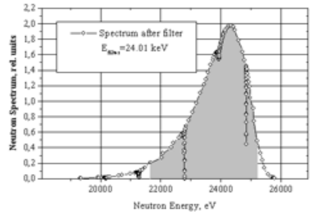

FIG. 3— *Calculated neutron spectrum after filter with energy 24.01 keV. The filled area corresponds to 95% response function.*

Filter 59 keV

We used here the composite neutron filter consisted of ^{58}Ni, S, V, Pb, Al and ^{10}B to receive the quasi-mono-energy beam with the average energy 58.98±2.60 keV. The purity of beam was about 98.4%, other additions to the main spectra were negligible: 83 keV – 0.02%, 366 keV – 1.6%.

The filter components (g/cm^2), used in these experiments for filtered neutron beam forming at the energy 59 keV, are presented in the Table 2.

TABLE 2—59 keV filter components.

^{10}B (85%)	^{58}Ni (99.3%)	S	V	Al	Pb
0.2	83.3	133.0	18.33	5.4	28.34

Fig. 4 show the calculated neutron spectra with peak energy 58.98 keV for filter used in experiment. The limits of 95% response function for 59 keV filter spectrum were defined as 51.92-60.22 keV.

Three samples of reactor carbon (99.9%) with thickness 0.0437451, 0.0870349 and 0.2100811 atoms/barn were used in these measurements. The final value of total neutron cross section, averaged over all samples was 4.480± 0.039 barn (relative accuracy 0.87%).

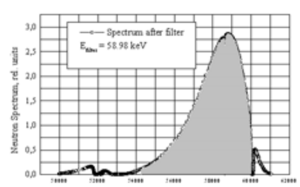

FIG. 4— *Calculated neutron spectrum after filter with energy 58.98 keV. The filled area corresponds to 95% response function*

Filter 148 keV

We used here the composite neutron filter consisted of Si, Ti and ^{10}B to receive the quasi-mono-energy beam with the average energy 148.09±17.60 keV. The purity of beam was about 95.4%, contribution of higher energy lines is negligible – 0.6 %. The low-energy lines in the region of (0.1– 2) keV and 54 keV make more considerable contributions – about 0.18% and 4.2%, but in process of treatment they may be separated due to the proton recoil counter spectroscopy abilities. The intensity of this filter was very large and we used additional Pb to reduce the counting rate to the reasonable value to avoid the large corrections.

The filter components (g/cm^2), used in these experiments for filtered neutron beam forming at the energy 148 keV, are presented in the Table 3.

TABLE 3—148keV filter components.

^{10}B (85%)	Si	Ti
1.0	197.18	12.3

Fig. 5 shows the calculated neutron spectra with peak energy 148.09 keV for filter used in experiment. The limits of 95% response function for 148 keV filter spectrum were defined as 118.71–157.01 keV.

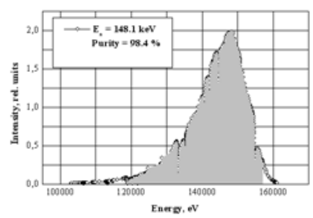

FIG. 5— *Calculated neutron spectrum after filter with energy 148.1 keV. The filled area corresponds to 95% response function.*

Nine samples of carbon with thickness from 0.008888 to 0,24788 atoms/barn were used in these measurements: 6 solid samples from reactor graphite (C 99.9%), 2 powder samples, loaded into aluminum container with internal diameter 31.75 mm (C 99.9%) and 1 sample in the form of carbon disk with thickness 1 mm (C 99.997%).

Our experimental results, obtained for carbon samples with different thickness, are shown in Fig. 6. As is obvious the observed total neutron cross sections depend on the sample thickness. Linear extrapolation to zero thickness, executed by means LSM using experimental results for 5 the most thin carbon samples, given unshielded value of the total neutron cross section for carbon (4,6551 ± 0,0477) barn.

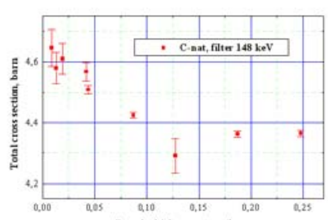

FIG. 6— *The observed experimental total neutron cross sections for natural carbon, obtained at the filter beam with energy 148 keV.*

Discussion of results

Fig. 7 represents our results for C-nat. total neutron cross sections at energies in the intervals 19.3–25.8 keV, 51.9–60.2 keV, and 118.7–157.0 keV together with the known experimental data from database EXFOR/CSISRS [1, 2, 6–10] and ENDF libraries.

For 24 keV our result is in a good consent with result by R. Block [6] - 4.684 ± 0.009 barn, measured with Fe-Al filter and time-of-flight at Linac spectrum.

Previous measurements at our reactor with somewhat other filter on the basis of Si (energy 55 keV) gave the values 4,48±0,039 barn [7] and 4.497±0.089 barn [8], rather close to our result for the 59 keV filter. The value of total cross section, obtained for the 55 keV filter in [9] 4,37±0,15 barn lies significantly below, but its measurement accuracy is low - 3.4%.

The value of the C-nat. total cross section, measured in our experiments with neutron filter at the energy 148 keV and corrected on self-shielding effect, is considerably higher than ones, obtained at the same Si filter by V. Vertebnyj [8] 4.309±0.018 barn and by Pham Zuy Hien [9] 4.280±0.013 barn. They are rather close to our results, obtained for carbon samples with thickness 0,12759– 0,24788 atoms/barn. Unfortunately information about thickness of samples, used in [8] and [9], is not available.

As it follows from the last edition of BNL-325 (Neutron Cross Sections, v.1, Academic Press, 1984), the isotope ^{13}C, part of which in natural carbon takes only 1.1%, has strong p-resonance with energy 152.9 ±1.4 keV (J=2, Γn=3.7 ± 0.7 keV, $\Gamma\gamma$=4.0 ± 1.6 keV).

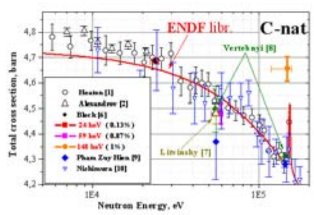

FIG. 7— *Our results for C-nat. total neutron cross sections for the 148 keV filter, experimental data from database EXFOR/CSISRS and ENDF libraries*

So, dependence of total neutron cross section from sample thickness, observed in our experiments, can be explained by self-shielding effect. But unshielded value of C-nat. cross section (4,6551 ± 0,0477) barn, estimated by us as averaged neutron cross section for energy range (118.71-157.01) keV, points out that in this energy range there is very strong resonance. Its neutron width may be much more than 3.7 keV. We consider this our result as preliminary. Investigation will be continued.

Acknowledgments

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