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A PRECISE DETERMINATION OF THE TERMAL NEUTRON ABSORPTION CROSS SECTION OF B^{10} AND NATURAL BORON BY TIME OF FLIGHT

by

A. J. DERUYTTER (C.E.N.-S.C.K.) A. PROSDOCIMI (EURATOM)

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0·019) barns m/μs. The deduced values for the boron absorption cross section at the reference neutron velocity of 2200 m/s are for natural boron $\sigma_a = 760{\cdot}8 \pm 1{\cdot}9$ barns and for the enriched boron $\sigma_a = 3703 \pm 8{\cdot}9$ barns. These values yield for the absorption cross section of the B^{10} isotope at the reference velocity 3840 ± 11 barns as deduced from the natural boron results, and 3837 ± 9 barns from the enriched sample.

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A PRECISE DETERMINATION OF THE THERMAL NEUTRON ABSORPTION CROSS SECTION OF B^{10} AND NATURAL BORON BY TIME OF FLIGHT*

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(*Received* 7 *July* 1962)

Abstract—The total cross section of B_2O_3 has been measured from 0·006 eV to 0·082 eV with the BR-1 slow-chopper. Samples of natural boron with an isotopic abundance of (19·81 ± 0·02) per cent B^{10} atoms, and boron enriched up to (96·515 ± 0·013) per cent were used for these measurements. From the accurate total cross-section values obtained, the ratio of the boron absorption cross section to the reciprocal of neutron velocity was calculated. This quantity remains a constant over the entire energy range covered and its mean value for natural boron is $\bar{K} = (1{\cdot}674 \pm 0{\cdot}0041)$ barns m/μs and for the enriched boron $\bar{K} = (8{\cdot}15 \pm 0{\cdot}019)$ barns m/μs. The deduced values for the boron absorption cross section at the reference neutron velocity of 2200 m/s are for natural boron $\sigma_a = 760{\cdot}8 \pm 1{\cdot}9$ barns and for the enriched boron $\sigma_a = 3703 \pm 8{\cdot}9$ barns. These values yield for the absorption cross section of the B^{10} isotope at the reference velocity 3840 ± 11 barns as deduced from the natural boron results, and 3837 ± 9 barns from the enriched sample.

1. INTRODUCTION

THE need for reducing the uncertainties in the boron cross section arises from its widespread use as a reference absorber for measurements of other cross sections, and from its use in boron loading specifications for reactors and in reactor physics calculations.

This widespread use is due to the high absorption cross section of the B^{10}-isotope, almost entirely due to the $B^{10}(n, \alpha)Li^7$ reaction, which shows a rigorous $1/\sqrt{E}$-behaviour in an extensive energy range.

In order to remove some discrepancies shown by previous determinations (DEBUS *et al.*, 1962) neutron transmission measurements of natural and enriched boron samples have been made with a high degree of precision in an energy range around 0·0253 eV. Special care was used in the mass-spectrometric determinations of the isotopic abundances of B^{10} in the samples, as this was the main reason of deviations in the absorption cross section of B^{10}, deduced from natural boron cross section measurements in the past. All other associate problems such as the determination of the number of B^{10} atoms/cm^2 in the cells, the exclusion of any trace of light hydrogen from the cells and chemical purity of the solutions were extensively investigated.

The use of highly enriched boron samples minimizes the influence of other cross sections used in the calculation of $\sigma_a(B^{10})$ from the measured total cross section of B_2O_3 and this reduces the error.

The use of 'natural boron' is not adequate to define a standard of neutron absorption with precision. As mass-spectrometric measurements of isotopic abundances seem now more reliable, we have considered as the fundamental value the absorption cross section of the B^{10}-isotope at 2200 m/s. From this value one easily calculates the absorption cross section of any boron standard, from a precise mass-spectrometric determination of the isotopic ratio.

2. SAMPLES

Two solutions of deuterated boric acid in heavy water have been prepared, one containing natural, the other highly enriched boron. The use of quartz containers eliminated any exchange of boron atoms with other materials. A preliminary analysis checked the chemical purity of the boron. No traces of other elements with high neutron cross sections were detected, which could necessitate a correction to the measured total cross section of B_2O_3.

Two identical planeparallel cells (Fig. 1) made of optically polished quartz were filled with these solutions. The use of D_3BO_3 and handling of the samples in a dry atmosphere avoided any trace of light hydrogen in the cells. It is estimated that the hydrogen contamination was less than 0·1 per cent.

Two identical cells were filled respectively with pure heavy water (99·9 per cent) and helium to serve as blanks.

The neutron transmission of the empty quartz cells, measured for each of them at 0·1 per cent had a mean

* Work done by a joint Euratom-CEN group.

TABLE 1.—SAMPLE SPECIFICATIONS

Sample specifications	Natural boron	Enriched boron	Heavy water
Isotopic concentration B^{10}/B	0·1981 ± 0·0002	0·96515 ± 0·00013	—
Concentration of B_2O_3 (% by weight)	1·67712 ± 0·00018	0·38591 ± 0·00011	—
Solution density at 25°C (g/cm³)	1·1162 ± 0·0001	1·1072 ± 0·0001	1·104485 ± 0·000001
Solution thickness (cm)	2·003 ± 0·001	2·005 ± 0·001	2·004 ± 0·001

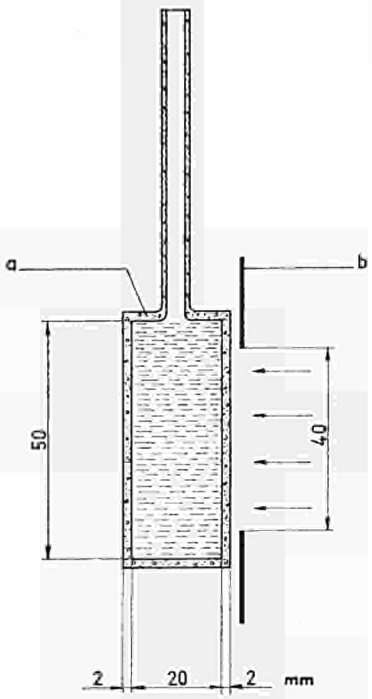

FIG. 1

value of 92·5 per cent. Relative differences of 0·2 per cent were minimized by a suitable choice of the cells to be filled with the four samples, moreover their influence on the final error is still less.

The neutron transmission of the boron samples varied from 30 to 70 per cent, i.e. suitable values to reduce both background and statistical effects.

The specifications of the samples are shown in Table 1. The quoted errors are standard deviations. Details about the measurements are given by DEBUS *et al.* (1962) for isotopic concentration, by LAUER *et al.* (1961) for chemical analysis, by MORET (1962) for thickness determination.

3. EXPERIMENTAL PROCEDURE AND APPARATUS

The total cross section of B_2O_3 has been calculated from the transmission of the samples measured by neutron time-of-flight spectrometry in the energy range 0·006 eV to 0·082 eV. (In this calculation the transmission of the D_2O cell was used to account for the density variation of the boron solutions.) The He cell simulated the empty cell.

The neutron beam of the thermal column of the BR-1 reactor at Mol, pulsed by a slow chopper (CEULEMANS *et al.*, 1962), was transmitted through the samples toward a bank of BF_3-detectors, with their axis perpendicular to the beam direction. Then the BF_3-pulses were fed into a multichannel time-of-flight analyser. The 1/v-behaviour of the measured cross section allowed the use of low resolution: 42 to 56 μsec/m, yielding higher intensity and statistical accuracy per channel.

The four samples were placed in the neutron beam by means of a precise positioning device. Cadmium diaphragms of 4 cm dia. and relative transmissions identical within 0·1 per cent, collimated the beam. Any long-term drifts of the experimental conditions were minimized by means of the successive exposure of the four samples in the beam during a time cycle of 10^3 sec.

Both the multichannel time-of-flight analyser and the sample holder were driven automatically according to a pre-set programme.

Suitable monitoring of the neutron beam eliminated the influence of power-level fluctuations of the reactor. For this purpose a parallel-plate fission flow counter with a low efficiency ^{235}U layer (DERUYTTER, 1960), was placed in the direct beam before the rotor. The diameter of the sensitive volume of the counter was made larger than the diameter of the neutron beam and the axis of the counter coincided with the axis of the beam. Statistical errors on the monitor counts were kept low by making its counting rate at least two orders of magnitude higher than that of each individual channel.

In this way all the partial countings could be normalized to their respective monitor counting, printed out by the multichannel analyser at the end of each cycle.

Similar measurements were made with Cd filters to correct for the epicadmium and background neutrons.

4. TREATMENT OF THE DATA

The data from the different measuring cycles were submitted to standard statistical tests, corrected for

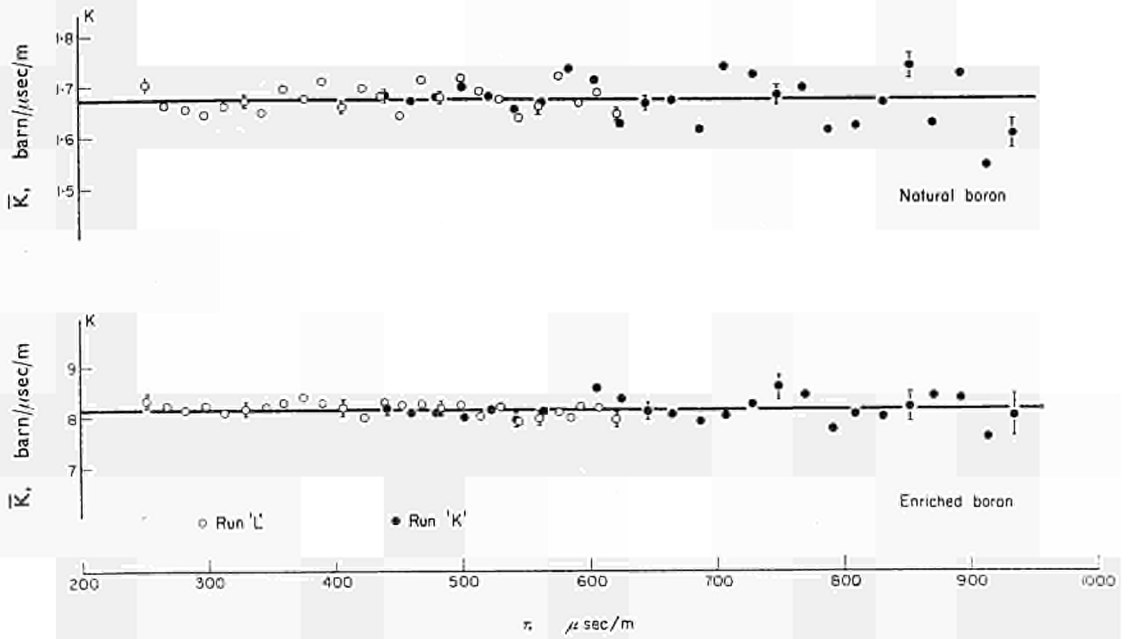

FIG. 2

the background and epicadmium neutrons, and normalized to the monitor counting. The data thus selected were then totalized to deduce the transmission T of the B_2O_3 solutions, both natural and enriched, and the transmission T_0 of the heavy water.

The total cross section of the B_2O_3 molecule is given by:

$$\sigma_t(B_2O_3) = \frac{M}{N_A}\left[\frac{1}{dc\rho}\ln(1/T) - \frac{1}{d_0\rho_0}\left(\frac{1}{c}-1\right).\ln(1/T_0)\right]. \quad (1)$$

c is the concentration of B_2O_3 in the solution; ρ and ρ_0 are respectively the densities of the solution and the heavy water; d and d_0 are respectively the solution and the D_2O thickness; M is the molecular mass of B_2O_3 and N_A is Avogadro's number.

The contribution to the total cross section of B_2O_3 by other cross sections than the boron absorption cross section is assumed 21 barns (HUGHES *et al.*, 1958; EGELSTAFF, 1957; TITMAN *et al.*, 1951). The uncertainty of this value, about 5 per cent, does not affect the results appreciably.

Indeed its contribution to $\sigma_t(B_2O_3)$ at 0·0253 eV is 1·3 per cent for natural boron and only 0·3 per cent for the enriched sample; and it remains less than 2·5 per cent for the highest energy covered in this experiment. The quantity:

$$K = \frac{\sigma_a(B)}{\tau} = \frac{\sigma_t(B_2O_3) - 21}{2\tau} \quad (2)$$

should be a constant, independent of τ, if $\sigma_a(B)$ is proportional to 1/v. τ represents the flight time reduced to μs/m.

In Fig. 2, K is plotted as a function of τ for natural and enriched boron, and the 1/v-behaviour is seen to be valid in the energy range covered. This verification allowed a calculation of the weighted mean value by least squares treatment of the 50 experimental K-values by the time-of-flight analysis.

From (2) it is seen directly that

$$\sigma_a(B^{10})_{2200} = \frac{\bar{K}}{v_0 r} \quad (3)$$

where v_0 is the velocity in m/μs of 0·0253 eV neutrons and r is the isotopic ratio $B^{10}/(B^{10}+B^{11})$ of the sample, neglecting $\sigma_a(B^{11}) = 0{\cdot}005$ barn (HUGHES *et al.*, 1958).

5. ERROR ANALYSIS

Several directly measured quantities are involved in the calculation of $\sigma_t(B_2O_3)$ according to (1) of Section 4. Their partial errors contribute with different weights to build up the total error of each single value deduced from the different energy channels.

Nevertheless it could be realized that the highest contribution to the error of each σ_t is due to the random distribution of the detector counts in such a way that the errors of other quantities are scarcely appreciable in comparison.

In the next step, when the values of K are computed from σ_t, the error on τ must be taken into account.

TABLE 2

Sample	Mole per cent B^{10}	$\bar{K}$, barn m/μsec	$\sigma_a(B)_{2200}$ barns	$\sigma_a(B^{10})_{2200}$ barns
Natural boron	19·81 ± 0·02	1·674 ± 0·0041	760·8 ± 1·9	3840 ± 11
Enriched boron	96·515 ± 0·013	8·15 ± 0·019	3703 ± 8·9	3837 ± 9

TABLE 3

Reference	Experimenter	Year	Energy (eV)	Apparatus	$\sigma_a(B^{10})_{2200}$ barns
SCHMITT *et al.* (1960)	ORNL	1960	0·018–0·4	Fast chopper	3848 ± 38
SCHMITT *et al.* (1960)	ORNL	1960	0·022–0·042	Fast chopper	3803 ± 48
SAFFORD *et al.* (1960)	Columbia Univ.	1960	0·003–0·1	Crystal spectr.	3838 ± 11
SAFFORD *et al.* (1960)	Columbia Univ.	1960		Crystal spectr.	3858 ± 25
DEBUS *et al.* (1962)	Euratom-CEN	1961	0·006–0·082	Slow chopper	3840 ± 11
DEBUS *et al.* (1962)	Euratom-CEN	1961	0·006–0·082	Slow chopper	3837 ± 9

This error originates from uncertainties on the flight-path length and on the flight time. The former arises essentially from the finite thickness of the sensitive volume of the neutron detectors, the latter from the definition of the zero of the flight time.

The flight path, defined as the distance between the reaction centre of the BF_3-counters and the chopper is measurable to 0·01 per cent. The paths of the detected neutrons, however, have a spread of 0·3 per cent around this nominal value.

The calibration of the time-scale was achieved throughout the determination of the Bragg cut-off of the total cross section for a polycrystalline sample of α-iron. Its error affects the single K-values with uncertainties ranging from 0·2 to 0·6 per cent; in Fig. 2 the total errors of K are displayed for some typical points.

Furthermore the correction for multiple scattering was estimated to be much less than 0·1 per cent and has therefore been neglected.

Finally, upon calculation of the mean $\bar{K}$ of the series of the K-values, its standard error, as quoted in Table 2, is evaluated according to the weighted least-squares method. At this point the interpretation of the errors displayed for $\sigma_a(B)_{2200}$ is straightforward.

Systematic errors on τ have not been accounted for, but an analysis on possible sources has been carried out (DERUYTTER *et al.*, 1961) showing that the arithmetic sum could yield no more than 0·13 per cent on the single τ-values. The error given in Table 2 for the isotopic concentration of B^{10} represents only the statistical fluctuations in the mass-spectrometric determinations. Systematic errors (mass discrimination) are under investigation. However, there are reasons to believe that mass discrimination is small. There is, in fact, good agreement between measurements of the same sample on mass spectrometers of different design. The value of 19·81 per cent B^{10} is in good agreement with results of other laboratories (DEBUS, 1962; private discussion at KAPL and ORNL), where mass discrimination has been measured. Furthermore, mass discrimination would have a very different effect on natural and enriched boron; the good agreement between the cross-section values for B^{10} obtained on both samples indicates that mass discrimination is practically non-existing. In any case the results of the investigation on mass discrimination will be published as soon as possible.

6. RESULTS AND CONCLUSIONS

The results for $\bar{K}$, $\sigma_a(B)$ and $\sigma_a(B^{10})$ are summarized in Table 2.

The errors quoted are standard deviations. The agreement between the values of the absorption cross section of the B^{10}-isotope calculated from the natural and the enriched sample is exceedingly good.

Table 3 shows a comparison of the most recent results for $\sigma_a(B^{10})_{2200}$. The table is limited to recent measurements either taken with enriched boron, or those giving a defined isotopic ratio when natural boron was used.

The agreement is satisfactory within the quoted errors.

The mean value for the two determinations of the present report is $\sigma_a(B^{10}) = 3838 \pm 7$ barns. Moreover a weighted mean deduced from all the data mentioned in Table 3 yields $\sigma_a(B^{10}) = 3839 \pm 9$ barns.

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