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Absolute Calibration of the NBS Standard Thermal Neutron Density

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A moderating geometry providing a constant thermal neutron flux from two radium-beryllium (α,n) sources has been constructed. The flux at the center of the geometry has been calibrated by absolute alpha counting from boron films exposed in pulse ionization chambers. The boron capture cross section has been obtained by comparison of the NBS boron with the Argonne "standard" boron, and a value of $n\hat{v}=3918$ (± 2 percent) n/cm^2 sec has been determined for the flux.

1. Introduction

The NBS standard thermal neutron flux was designed to provide a uniform flux in a region suitable for the absolute calibration of foils, small neutron detectors, and dosimeters. This flux is obtained by the moderation of neutrons in paraffin and carbon from two radium-beryllium (α,n) sources and does not have a precise Maxwellian velocity distribution. If a thin foil having a thermal neutron-capture cross section varying as $1/v$ is placed in the flux, the rate of neutron capture is $C=\Sigma_i n_i v_i \sigma_i N$, where n_i, is the neutron density, σ_i is the capture cross section at velocity v_i and N is the number of nuclei in the foil.

As $v_i\sigma_i$ is a constant independent of velocity,

$$C=nNK=nN(\hat{\sigma}\hat{v}). \quad (1)$$

Thermal-capture cross sections, $\hat{\sigma}$, are usually listed [1][1] at the most probable Maxwellian velocity at room temperature, $\hat{v}=2.2\times10^5$ cm/sec.

Measurement of the absolute capture rate by a thin foil thus yields the thermal neutron density unambiguously. This density is usually multiplied by $\hat{v}$, and $n\hat{v}$ represents a flux density of monoergic neutrons that is equivalent to the original flux for foil activation of $1/v$ absorbers. For a flux with a Maxwellian-velocity distribution, $\Sigma_i n_i v_i = n\bar{v}=1.128\ n\hat{v}$. In this work $n\hat{v}$ has been used to represent the flux, which has been standardized by the absolute counting rates obtained from boron films exposed in pulse ionization chambers.

2. Description of Geometry

The geometry providing the NBS thermal neutron flux (fig. 1) consists of two 1-curie radium-beryllium (α,n) sources imbedded in lead cylinders 8½ inches in diameter. Two inches of paraffin and 4 inches of carbon are between the lead cylinders and the exposure slot. Carbon surrounds the exposure slot so that the depression in neutron density [2] caused by absorbing foils will be small. The effect would be much greater if paraffin alone were used for moderation. At the center of the slot the neutron flux should be nearly isotropic, and from symmetry considerations the gradient of the neutron density should be zero. Therefore, a pure scatterer placed at the center will not alter the flux.

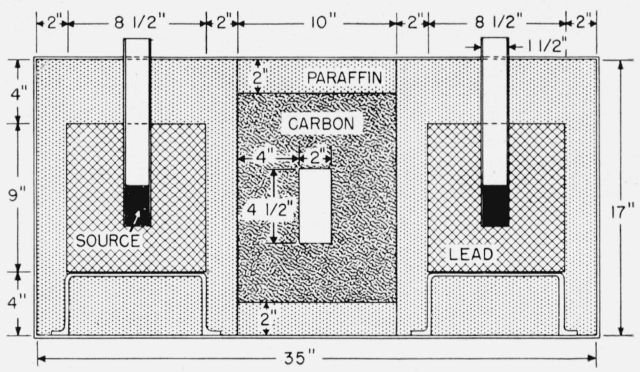

FIGURE 1. *Standard neutron flux geometry.*

3. Description of Pulse Ionization Chambers

An aluminum chamber of pillbox design, 2⅝ in. in internal diameter and 1⅛ in. deep, having a boron film clamped to the cover plate, was used for the measurements [3]. Either an aluminum parallel plate mounted on Teflon insulators 1.4 cm from the film or a 1.14-mm carbon-steel wire (1.7 cm distant) was used for electron collection. A mixture of 5 percent of carbon dioxide and 95 percent of argon was used as the filling gas.

Boron films were deposited by vacuum evaporation of natural boron from carbon crucibles over an area 2.0 by 3.4 cm^2 on Pt-coated 0.015-in. quartz wafers or on Pt foils.

[1] Figures in brackets indicate literature references at the end of this paper.

An aluminum holder that slides into the exposure slot contains the pulse chamber and cathode follower. The holder exterior has been milled for the snug fit of a 0.020-in. cadmium cover.

Pulses from the chamber were amplified by a Bell-Jordan preamplifier and amplifier system and fed into a three (integral) channel discriminator unit. Most of the measurements were made with a 2-μsec differentiating time constant; a few were repeated with a 20-μsec time constant.

4. Analysis of Data

Measurements were made with and without cadmium, and the cadmium-difference counting rate was determined as a function of discriminator setting for three different boron films. This procedure almost entirely eliminates spurious counts from the data and the cadmium-difference counting rate is confined to captures of neutrons of energy less than 0.3 ev. To avoid multiple- and back-scattering corrections the alpha-particle counting rate was determined at a discriminator level two-thirds or three-fourths that of the maximum pulse height of the 1.473-Mev alpha-particle ionization from the main branch of the reaction:

$$n+B^{10}\begin{cases}\nearrow Li^{*7}+He^4 \text{ (93.5 percent)}\\ \searrow Li^7+He^4 \text{ (6.5 percent)}.\end{cases}$$

Li^{*7} refers to the 0.48-Mev excited state. The particles from the reaction proceeding directly to the ground state have 20.7 percent more kinetic energy than those from the main branch.

A correction must be made for the alpha particles that are degraded below the bias level by ionization losses in the film. For 1.5-Mev alphas it is safe to assume that the energy of a particle emerging from a plane film is given by $E=E_0(1-x/R\cos\theta)$ where x is the perpendicular distance from the origin of the particle to the surface of the film, θ is the angle of the particle direction with respect to the normal, and R is the particle range in the film material. If $2N_0$ alphas are produced per second in the film of thickness T, the energy distribution of the emerging particles becomes

$$\frac{dN}{dE}=\frac{N_0R}{2E_0T} \quad \text{for} \quad E_0>E>E_0\left(1-\frac{T}{R}\right) \tag{2}$$

$$=\left(\frac{dN}{dE}\right)_{\max} \text{ a constant independent of } E, \text{ and}$$

$$\frac{dN}{dE}=\frac{N_0T}{2E_0R\left(1-\frac{E}{E_0}\right)^2} \quad \text{for} \quad E<E_0\left(1-\frac{T}{R}\right). \tag{3}$$

The number of particles with energy greater than E is given by

$$N_E=N_0\left[1-\frac{T}{2R\left(1-\frac{E}{E_0}\right)}\right] \quad \text{for} \quad E<E_0\left(1-\frac{T}{R}\right). \tag{4}$$

Combining (2) and (4)

$$\frac{T}{R}=\frac{N_E}{1-\frac{T}{2R\left(1-\frac{E}{E_0}\right)}}\frac{1}{2E_0\left(\frac{dN}{dE}\right)_{\max}}.$$

At $E=\frac{3}{4}E_0$,

$$\frac{T}{R}\left(1-\frac{2T}{R}\right)=\frac{N_{3/4}E_0}{2E_0\left(\frac{dN}{dE}\right)_{\max}}. \tag{5}$$

For thick films eq (5) was used to determine T/R, but for thin films eq (3) must be used because the amplifier noise causes a deviation in dN/dE near the maximum energy, E_0.

The path of the ionizing particle in the gas distorts the energy spectrum because the pulse height at the collecting electrode is proportional to the mean voltage of the ionization electrons divided by the total voltage drop between electrodes. With wire collection this effect is minimized, and T/R may be extracted without too much difficulty.

All three films were counted with parallel-plate collection in the aluminum chamber. Film 2 was counted with a collecting wire in the aluminum chamber, and T/R was determined for film 3 in a similar brass chamber (fig. 2). Another film, which had a smaller thickness correction than the others, was measured with wire collection in the aluminum chamber, but the film was unfortunately spoiled before analysis. All parallel-plate measurements were made with a filling of 95 percent of argon and 5 percent of carbon dioxide at 4 atmos-

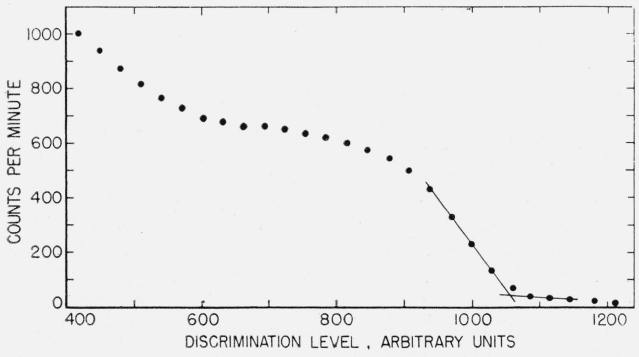

FIGURE 2. *Film 3 pulse-height distribution.*

Brass chamber; 1— atm A-CO_2; 1,500 volts; wire collection.

pheres, and measurements of T/R with wire collection were made at a variety of pressures. As the alphas are counted with 2 π geometry, the neutron capture rate is twice the corrected alpha counting rate.

Carbon was also deposited during the boron evaporation, obviating a boron mass determination by weighing. A technique for determination of the boron content of the films was developed by microchemical modification of the fixed pH titration method of Foote [4]. After analysis each film backing was replaced in the chamber, which was reinserted in the flux as a check of the efficiency of the boron removal. The counting rates at the alpha-plateau discrimination level were 2 to 3 counts per minute, the normal background for the boron-free chamber.

There is considerable evidence indicating a slight variation of the boron isotopic ratio with geographic origin [5] (or processing). The boron evaporated for use in the chambers was obtained by the thermal decomposition of diborane supplied by the Naval Research Laboratory. From the diborane a sample of boric-acid solution was prepared for comparison with a solution of the standard boric oxide of the Argonne National Laboratory by the danger-coefficient method in the Argonne low-energy pile. The boron cross section of this standard has been accurately measured as 755 ± 3 barns [6] and verified by the Brookhaven National Laboratory's result of 753 ± 3 [7]. The Argonne comparison gave a value of 746 ± 7 barns for the NBS boron. Dibeler and Mohler [8] have measured the B^{10}/B^{11} isotopic ratio for the NRL diborane and obtained a value of 0.251 as compared with 0.253 for a sample from the University of Southern California (both the USC and Argonne boron are presumably of California origin), which is in line with the Argonne comparison.

Table 1 summarizes the boron-film data. I_b has a weighted average of 1.761×10^6 disintegrations per mole of boron in the NBS flux.

TABLE 1. *Boron-film data*

Film	Boron content	T/R	N_0, corrected counting rate	I_b, Twice counting rate per mole of boron
	Moles		*Counts/sec*	*Disintegration /sec mole*
1	1.416×10^{-5}	0.050	12.57	1.775×10^6
2	1.012	.0505	8.84 (parallel plate)	1.747
			8.93 (wire)	1.764
3	1.751	.102	15.29	1.753

$I_b=n\hat{v}\hat{\sigma}L=1.761\times10^6$ where $\hat{\sigma}=746$ barns, and $L=6.025\times10^{23}$ atoms/mole. Finally, $n\hat{v}=3{,}918$ neutrons/cm^2 sec.

5. Sources of Error

It is somewhat difficult to estimate the errors precisely in this experiment. For each film the alpha plateau counting rate was determined with a statistical accuracy of at least 0.7 percent. For the thickest film, the thickness correction was 18 percent at $E/E_0=2/3$, but since T/R is probably good to 5 percent this introduces an error of only 0.9 percent in the corrected counting rate. The boron cross section should be accurate within 1 percent. Repeated analyses of standard solutions of boric acid containing 100 to 300 micrograms of boron had a standard error within 1.5 percent. Effects such as the neutron density depression and shielding by the platinum backing (including the boron) are each less than 0.15 percent.

When parallel plate collection is used a slight correction must be made to formula (4) because of the path of the particles in the gas. For the thickest film this correction is less than 0.4 percent at $E/E_0=3/4$.

Film 1 did not have a precise measurement of T/R and was analyzed, using a 10-ml buret for the NaOH titrant before the technique was refined with the use of a Gilmont ultramicroburet-pipet of 1-ml capacity subdivided into 1,000 divisions. This film has been assigned an error of 3.3 percent. Films 2 and 3 yielded measurements of $n\hat{v}$ good to 2.2 percent. The weighted average should be accurate to 1.5 percent (standard error).

Systematic errors may be present, e. g., isotopic enrichment during evaporation or loss of boric acid by volatilization during solution of the boron film in warm nitric acid. Assuming the systematic errors to be of the same order of magnitude as the statistical errors, then $n\hat{v}$ equals 3,918 neutrons/cm^2 sec ($\pm$2-percent standard error).

6. Comparison with Oak Ridge National Laboratory

Ritchie and Klema of the Oak Ridge National Laboratory have measured the thermal neutron flux in slot 11 of their standard graphite pile by beta-gamma coincidence measurements of activated gold foils. They obtained a value for $n\hat{v}\hat{\sigma}_{Au}$ of 5.905×10^4, and assuming $\hat{\sigma}_{Au}$ is 95 barns, $n\hat{v}$ equals 622 (±5 percent) neutrons/cm^2 sec. Identical gold foils were simultaneously exposed to the ORNL and NBS fluxes. The Oak Ridge foil was flown to the Bureau and alternately counted with the NBS foil under the same end-window beta counter. Exposures were made with and without cadmium, and the ratio of the cadmium difference counting rate is

$$\frac{A_{NBS}}{A_{ORNL}}=\frac{(n\hat{v})_{NBS}}{(n\hat{v})_{ORNL}}=6.42\ (\pm2.5\ \text{percent}).$$

The ratio of the independently calibrated fluxes is

$$\frac{(n\hat{v})_{NBS}}{(n\hat{v})_{ORNL}}=\frac{3918}{623}=6.29,$$

which agrees with the gold-foil ratio within the experimental error.

7. References

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