



## The production of nitrogen-13 by neutron capture in boron compounds

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### Abstract

The  $^{10}\text{B}(\alpha, n)^{13}\text{N}$  reaction is studied as an activation process in a variety of solid boron-containing neutron shielding materials. The source of  $\alpha$ -particles is the neutron capture reaction  $^{10}\text{B}(n, \alpha)^7\text{Li}$ . Samples of boron carbide, boron oxide, and boron nitride are irradiated with thermal neutrons and the rate of  $^{13}\text{N}$  production is determined.  $^{13}\text{N}$  promptly decays, emitting a positron. This positron efficiently annihilates with electrons in the material and the resultant 511 keV gamma ray is detected. For each of the above-mentioned materials, the rate of  $^{13}\text{N}$  production is  $(1-2) \times 10^{-10}$  per captured neutron.

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### 1. Introduction

In boron compounds,  $^{13}\text{N}$  may be produced through the  $^{10}\text{B}(\alpha, n)^{13}\text{N}$  reaction. This reaction has been studied previously by bombarding boron targets with alpha particles emitted by radioactive sources [1] or produced by ionization and acceleration of helium [2,3]. The same reaction can also

be induced by irradiating boron compounds with neutrons; the neutron capture reaction  $^{10}\text{B}(n, \alpha)^7\text{Li}$  yields 1.47 MeV  $\alpha$ -particles<sup>1</sup> that can react with neighboring  $^{10}\text{B}$  atoms. Because boron compounds are commonly used in neutron shielding, the resulting  $^{13}\text{N}$  activation and following  $\beta^+$  decay can constitute an unwanted source of background radiation.

The probability of producing  $^{13}\text{N}$  through neutron irradiation is of particular interest to

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<sup>1</sup> This reaction also yields 1.78 MeV  $\alpha$ -particles with a branching ratio of 6.1%.

experiments for measuring the neutron lifetime by detecting the decay of magnetically trapped ultracold neutrons (UCN). In one such experiment [4], the vast majority of approximately  $10^{12}$  cold neutrons entering the trapping apparatus are absorbed in boron-containing shields surrounding the trapping region (only  $10^4$  of the neutrons are trapped). Because the detection system used in this neutron lifetime experiment is sensitive to gamma rays, and because the  $862.6 \pm 0.3$  s decay lifetime of  $^{13}\text{N}$  [5] is comparable to that of the free neutron,  $885.7 \pm 0.8$  s [6],  $^{13}\text{N}$  decays in the shielding materials might lead to a potential systematic effect.

Perry and Wilson [7] estimated the probability of producing neutrons by secondary activation of the  $^{10,11}\text{B}(\alpha, n)^{13,14}\text{N}$  reaction following neutron capture [7]. They were interested in this reaction's potential to reduce the effectiveness of boron as a reactor poison. Their calculations predict yields of  $6 \times 10^{-7}$  and  $5 \times 10^{-7}$  neutrons per 2 MeV  $\alpha$ -particle in natural isotopic abundance boron and boron carbide, respectively; if a reasonable fraction of this neutron yield were due to the reaction in  $^{10}\text{B}$ , the  $^{13}\text{N}$  produced could be sufficient to create a background in the neutron lifetime experiment.

In this study, boron-containing targets are irradiated with thermal neutrons. The resulting  $\alpha$ -particles initiate the  $^{10}\text{B}(\alpha, n)^{13}\text{N}$  reaction. The  $^{13}\text{N}$  atoms produced in this manner are detected by counting the 511 keV annihilation gamma rays induced by the positrons emitted in the  $^{13}\text{N}$  decays. The results of the decay rate measurements, combined with a measurement of the neutron flux used in their radiation, yield the number of  $^{13}\text{N}$  atoms produced per neutron.

## 2. Experimental method

### 2.1. $^{13}\text{N}$ production and detection

Samples of boron carbide ( $\text{B}_4\text{C}$ ), boron nitride (BN) and boron oxide ( $\text{B}_2\text{O}_3$ ) are used. The  $\text{B}_4\text{C}$  sample was obtained in the form of a hot-pressed 3.1 cm diameter, 4.2 mm thick disk, with a  $^{10}\text{B}$  isotopic mass fraction of  $\approx 9\%$  [8]. The BN sample (with natural  $^{10}\text{B}$  isotopic abundance of 19.9%) was machined into a 3.0 cm diameter and 4.1 mm

thick disk [9]. The  $\text{B}_2\text{O}_3$  ( $^{10}\text{B}$  isotopic mass fraction of  $6.0 \pm 0.5\%$ ) was prepared by melting powder into a glass ingot and grinding into a 3.0 cm diameter and 9.6 mm thick disk [8,10]. All three target samples are “thick” relative to the path lengths of both the neutrons and the  $\alpha$ -particles, such that essentially all of these particles are absorbed in the target.

Each sample is irradiated with neutrons from the thermal column facility at the National Institute of Standards and Technology (NIST) Center for Neutron Research (NCNR). This facility consists of a 30 cm diameter spherical cavity located in an approximately 1.4 m  $\times$  1.3 m  $\times$  0.9 m graphite moderator. The graphite is positioned inside the biological shield of the reactor and has a well-characterized thermal spectrum ( $T = 293$  K) [11]. The graphite assembly and shielding are shown in Fig. 1. Access to the thermal column cavity is through a 1.2 m long, 5.1 cm diameter cylindrical hole that penetrates the reactor biological shield. Each sample is mounted to the end face of a 3.5 cm diameter, 3.5 cm long cylindrical piece of acrylic. This assembly is placed into one end of a 3.8 cm diameter aluminum tube that is inserted into the thermal column cavity (see Fig. 1). Once the sample is positioned, the boral curtain separating the thermal column from the reactor vessel is raised to allow neutrons to enter the cavity. A neutron fluence of  $\approx 10^{11}$   $\text{cm}^{-2}\text{s}^{-1}$  strikes the sample (see Section 2.2). The samples are each irradiated for 30 min. (The time required to raise and lower the boral curtain – about 30 s – introduces an insignificant uncertainty into the measurement of the total irradiation time.) The sample is then removed from the graphite cavity and quickly transferred (with a delay of about 5 min) to a lead-encased high purity germanium detector.

For each irradiated boron compound, the energy spectra of the emitted gamma radiation are collected in 150 s time intervals for several hours. An example of such an energy spectrum is shown in Fig. 2. For each 150 s spectrum, the area of each peak of interest is integrated after background subtraction and recorded as a function of time. These data are used to determine the rate of  $^{13}\text{N}$  production in the boron samples.

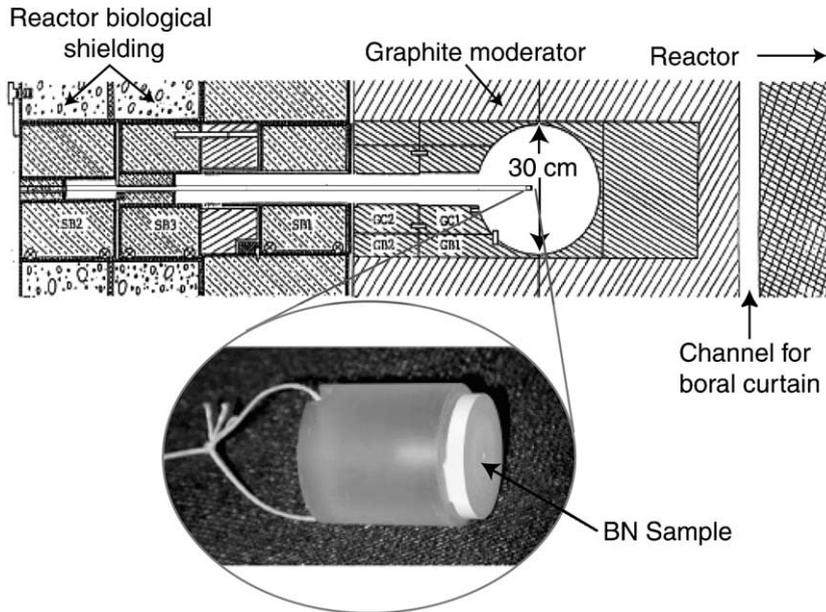


Fig. 1. Schematic view of the thermal column. Inset: a picture of the boron nitride sample mounted on the acrylic cylinder.

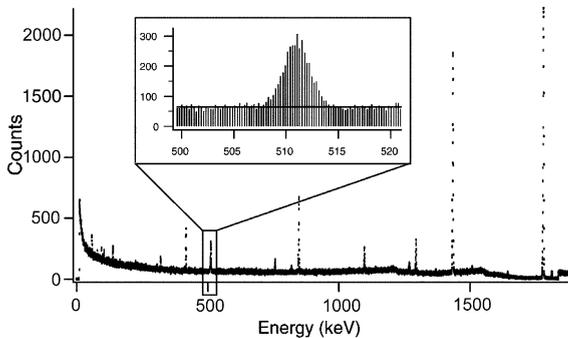


Fig. 2. A gamma ray spectrum obtained from the irradiated boron carbide sample using a high-purity germanium detector. Inset: the peak at 511 keV.

## 2.2. Neutron fluence measurement

The fluence of neutrons incident on the samples is determined by irradiation of 1.27 cm diameter, 23  $\mu\text{m}$  thick copper foils. In order to ensure that the fluence is measured in the same environment as that of the boron samples, the copper foil in each irradiation is placed on a face of the boron carbide sample, with the boron carbide sample mounted in the same manner as described above. Two irradiations are performed, each for 10 min. In the first,

the copper foil is positioned such that it faces the reactor core; in the second, the copper foil is between the boron carbide disk and the acrylic cylinder. Following irradiation, each copper foil is transferred within minutes to a high purity germanium detector to count the gamma rays emitted in the decays of the neutron capture products,  $^{64}\text{Cu}$  and  $^{66}\text{Cu}$ .

The energy spectra of the emitted gamma radiation are recorded in 60 s intervals for 4 h. The resulting spectra contain peaks from  $^{64}\text{Cu}$  ( $\tau_{^{64}\text{Cu}} = 18.322 \pm 0.003$  h [5]) at 511 keV (from positron annihilation) and 1.346 MeV and peaks from  $^{66}\text{Cu}$  decay ( $\tau_{^{66}\text{Cu}} = 7.39 \pm 0.02$  min [5]) at 834 keV and 1.039 MeV. For each of these energies, the time-dependent countrates of the gamma rays are measured as described above. The data are fitted to  $Y = Ae^{-t/\tau}$ , with  $\tau$  fixed to the known average decay lifetime and  $A$  extracted from the fit. The fluence of neutrons incident on the copper foil during the irradiation,  $\phi$ , is given by

$$\phi = \frac{Ae^{t_w/\tau}}{\sigma N_E \epsilon_E (1 - e^{-t_{\text{irr}}/\tau})}, \quad (1)$$

where  $t_w$  is the time between the end of the irradiation and the start of data collection,  $t_{\text{irr}}$  is the

Table 1

Measurements of the neutron fluence incident on the front face of the sample, oriented toward the reactor core, and on the opposite face of the sample, as described in the text

Isotope	Energy (keV)	Fluence (front) ( $10^{11} \text{ cm}^{-2} \text{ s}^{-1}$ )	Fluence (rear) ( $10^{11} \text{ cm}^{-2} \text{ s}^{-1}$ )
$^{64}\text{Cu}$	511	$1.8 \pm 0.2$	$0.52 \pm 0.05$
$^{66}\text{Cu}$	833	$1.8 \pm 0.4$	$0.60 \pm 0.15$
$^{66}\text{Cu}$	1039	$1.9 \pm 0.5$	$0.53 \pm 0.13$
$^{64}\text{Cu}$	1356	$1.8 \pm 0.1$	$0.52 \pm 0.03$
Mean		$1.81 \pm 0.08$	$0.52 \pm 0.02$

time of irradiation,  $\sigma$  is the neutron capture cross-section of the target nuclide,  $N$  the number of atoms of the target nuclide,  $I_E$  is the number of gamma rays emitted at the energy  $E$  per radioactive decay and  $\epsilon_E$  is the absolute efficiency of the detector at energy  $E$ , taking into account the geometry of the copper foil and its placement on the detector; this value is determined independently in separate measurements [12].

The neutron fluence is determined for two placements of the copper foil at each of the four energies and is summarized in Table 1. Taking the weighted mean of the flux calculations from the four peaks yields a neutron fluence of  $\phi_1 = (1.81 \pm 0.08) \times 10^{11} \text{ cm}^{-2} \text{ s}^{-1}$  incident on the face of the sample oriented toward the reactor core and  $\phi_3 = (5.2 \pm 0.2) \times 10^{10} \text{ cm}^{-2} \text{ s}^{-1}$  incident on the opposite face (fewer neutrons strike the face opposite the reactor core because they can only strike this face via reflection off the walls of the thermal column and the acrylic piece). We estimate the neutron fluence  $\phi_2$  incident on the cylindrical side of each sample to be the average of these two.

### 2.3. Calculation of $^{13}\text{N}$ production rate

The 511 keV gamma rays detected in the boron samples after neutron irradiations originate from both  $^{13}\text{N}$  decay positrons and high-energy pair production. An analysis of the complete spectra reveals peaks indicating the presence of  $^{28}\text{Al}$ ,  $^{56}\text{Mn}$  and  $^{24}\text{Na}$  in all three samples. Each of these nuclides emits gamma rays of sufficient energy to undergo pair production, thus producing positrons that annihilate to yield 511 keV gammas as well. To account for these additional contributions,

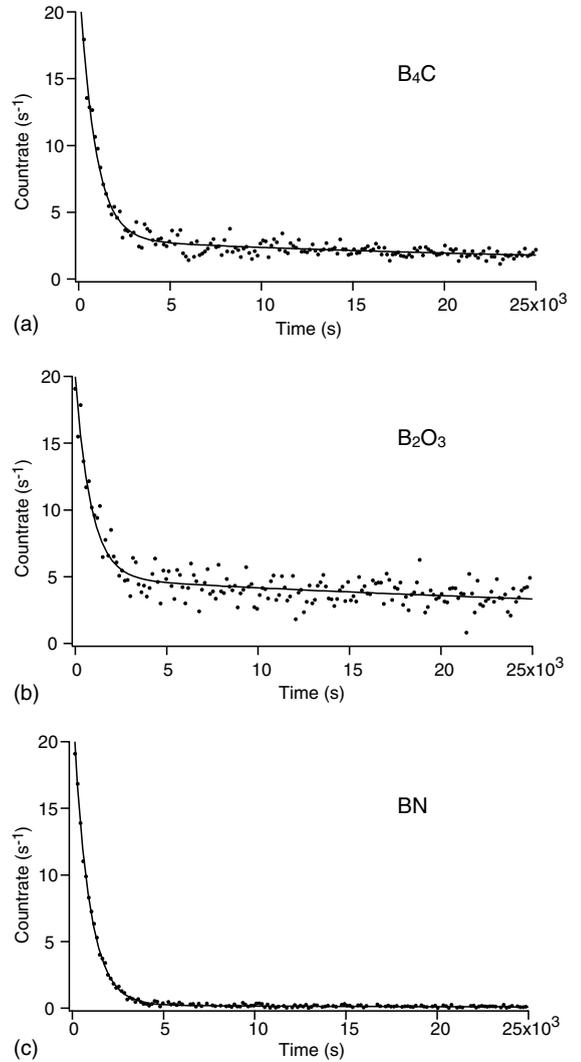


Fig. 3. The 511 keV gamma ray decay count rates are fitted to combinations of  $^{28}\text{Al}$ ,  $^{13}\text{N}$ ,  $^{56}\text{Mn}$  and  $^{24}\text{Na}$  decays. Details of the fits are described in the text and Table 2.

the 511 keV decay data are fit to the functional form

$$Y = Ae^{-t/\tau_{\text{Al}}} + Be^{-t/\tau_{\text{N}}} + Ce^{-t/\tau_{\text{Mn}}} + De^{-t/\tau_{\text{Na}}}, \quad (2)$$

where  $\tau_{\text{Al}} = 194.0 \pm 0.1 \text{ s}$ ,  $\tau_{\text{N}} = 862.6 \pm 0.3 \text{ s}$ ,  $\tau_{\text{Mn}} = 3.721 \pm 0.001 \text{ h}$  and  $\tau_{\text{Na}} = 21.581 \pm 0.002 \text{ h}$  are the lifetimes of  $^{28}\text{Al}$ ,  $^{13}\text{N}$ ,  $^{56}\text{Mn}$  and  $^{24}\text{Na}$ , respectively [5]. The 511 keV decay data, along with the associated fits, are shown in Fig. 3. Results from these fits are summarized in Table 2.

Table 2  
Fit coefficients of Eq. (2) for different samples

Sample	A (Al)	B (N)	C (Mn)	D (Na)
B <sub>4</sub> C	0.0 ± 0.7	20.3 ± 0.5	0.7 ± 0.2	2.3 ± 0.1
B <sub>2</sub> O <sub>3</sub>	0.0 ± 1.3	15.0 ± 0.9	0.4 ± 0.2	4.5 ± 0.1
BN	0.6 ± 0.2	22.8 ± 0.1	0.18 ± 0.03	0.07 ± 0.01

Table 3  
The number of <sup>13</sup>N atoms produced per neutron, extracted from Eq. (3)

	<sup>13</sup> N atoms produced per neutron
B <sub>4</sub> C	(1.7 ± 0.5) × 10 <sup>-10</sup>
B <sub>2</sub> O <sub>3</sub>	(1.1 ± 0.3) × 10 <sup>-10</sup>
BN	(2.0 ± 0.5) × 10 <sup>-10</sup>

The coefficient  $B$  in Eq. (2) gives the number of 511 keV gamma rays from <sup>13</sup>N decay detected at the start of the detection time. In terms of the fitting coefficient  $B$ , the number of <sup>13</sup>N atoms formed per neutron striking the sample is given by

$$N = \frac{4\pi B e^{t_{\text{irr}}/\tau_{\text{N}}}}{(1 - e^{-t_{\text{irr}}/\tau_{\text{N}}})\epsilon \int_S I \Omega \phi \, dA}, \quad (3)$$

where  $I$  is the number of 511 keV gamma rays resulting from positron annihilation in the sample and in the aluminum casing of the detector,  $\epsilon$  is the intrinsic efficiency of the detector,  $S$  represents the surface of the sample;  $\phi$  is the fluence of neutrons incident on the region  $dA$  of the surface and  $\Omega$  is the solid angle subtended by the detector at a source point in the region  $dA$ . It was found that in each of these compounds,  $\approx(1 \text{ to } 2) \times 10^{-10}$  <sup>13</sup>N atoms are formed per neutron capture (see Table 3).

### 3. Discussion

The comparison of these results with theoretical expectations is complicated due to the energy-dependent range of the  $\alpha$ -particle in the samples, coupled with the lack of information on the energy-dependent cross-sections. In general, the samples with the lower <sup>10</sup>B concentrations yield slightly lower <sup>13</sup>N production rates as expected.

If the total  $\alpha$ -particle production from neutron capture and the following  $(\alpha, n)$  reaction is estimated using the results in [7,13], one obtains the

yield of neutrons (from both <sup>10</sup>B and <sup>11</sup>B) of  $1.9 \times 10^{-7}$  in natural isotopic abundance boron carbide. It is also estimated that the neutron yield is about two times larger in the <sup>11</sup>B( $\alpha, n$ ) reaction than the <sup>10</sup>B( $\alpha, n$ ) reaction [14,15], implying a production of  $\approx 1 \times 10^{-8}$  <sup>13</sup>N atoms for every incident neutron striking a sample of our 9% <sup>10</sup>B mass fraction boron carbide sample. The rate of <sup>13</sup>N production observed, however, is about a factor of 50 smaller.

Both Perry's [7] and Liskien's [13] calculation used the  $(\alpha, n)$  cross-section of natural boron measured by Walker [1] in 1949. Walker's measurement has a low energy resolution (0.3 MeV) and did not resolve resonance peaks at low energies. We performed a similar calculation using the recent <sup>11</sup>B( $\alpha, n$ ) cross-section data of [16] and the data of [17] for the energy loss of  $\alpha$ -particles in boron. We estimate only a  $1.7 \times 10^{-8}$  probability of an exiting  $\alpha$ -particle participating in the <sup>11</sup>B( $\alpha, n$ )<sup>14</sup>N reaction in natural isotopic abundance B<sub>4</sub>C, about a factor of 10 smaller than the previous estimates. Unfortunately, <sup>10</sup>B( $\alpha, n$ ) cross-section data for low energy  $\alpha$ -particles are not currently available. Shire et al. [2,3] did measure the neutron yields resulting from  $\alpha$  bombardment of <sup>10</sup>B and <sup>11</sup>B close to the resonant energy 1.51 MeV. The yield curves suggest that the  $(\alpha, n)$  cross-section of <sup>10</sup>B approaches zero rapidly below the resonant energy, whereas that of <sup>11</sup>B remains nonzero (in agreement with the data of [16]). A small <sup>10</sup>B( $\alpha, n$ )<sup>13</sup>N cross-section for  $\alpha$ -particle energy at 1.47 MeV could explain the low probability of the  $(\alpha, n)$  reaction in <sup>10</sup>B relative to the probability of the reaction in <sup>11</sup>B.<sup>2</sup>

<sup>2</sup> Even though the <sup>10</sup>B( $\alpha, n$ )<sup>13</sup>N cross-section for 1.78 MeV  $\alpha$ -particles is larger, the production of <sup>13</sup>N resulting from them is still on the order of  $10^{-10}$  per neutron due to the small branching ratio.

The measurements from the three different samples agree well with each other. The discrepancy with theory can be resolved if the neutron yield of 1.47 MeV alpha bombardment on  $^{10}\text{B}$  is about 10 times smaller than on  $^{11}\text{B}$ . Future measurements of the  $^{10}\text{B}(\alpha, n)$  cross-section will help to clarify this.

Our observation that the  $^{10}\text{B}(\alpha, n)^{13}\text{N}$  reaction is less probable than anticipated has positive implications for the neutron lifetime experiment. With a production rate of  $\approx 10^{-10}$   $^{13}\text{N}$  atoms per incident neutron, the number of  $^{13}\text{N}$  atoms formed in the neutron shielding materials is two orders of magnitude less than the number of neutrons trapped. Our simulation shows that even if the background subtraction technique [4] used in the neutron lifetime experiment is only 90% effective, the systematic error introduced by the background from the  $^{13}\text{N}$  decay will be less than  $10^{-6}$ .

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