Thermal-neutron cross section for $^{10}B(n,t)2\alpha$ via ^3He-^4He mass spectrometry

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The thermal cross section for the reaction $^{10}B+n\rightarrow{}^3H+{}^4He+{}^4He$ is determined to be 4.47±0.15 mb by neutron irradiation of H_3BO_3 followed by measurement of ^3He (from decay of ^3H) and ^4He in a static mass spectrometer. Some samples contained boron of normal isotopic composition and some were enriched in ^{10}B, and irradiations were carried out in a highly thermalized neutron flux and in the core of a light-water-moderated reactor.

Kavanagh and Marcley[1] have recently determined a value of 7±2 mb for the $^{10}B(n,t)2\alpha$ reaction cross section at thermal energies. This result is in sharp contrast to previous measurements of 3 b by Lal and Craig[2] and 50±5 mb by Cserpak *et al.*[3] When we became aware of the 3 b value, it was decided to attempt a remeasurement, primarily because if the high value is correct, serious systematic error is present in measurements of ultratrace Li via thermal-neutron activation followed by mass spectrometric assay of ^3He (from decay of ^3H) where we have assumed that in most cases the only reaction which interferes significantly with $^6Li(n,t)\alpha$ is $^2H(n,\gamma)^3H$.[4] We then learned that Li contamination in some of the glass samples was a likely source of systematic error in the 3 b measurement.[5] A reliable value for the $^{10}B(n,t)2\alpha$ thermal reaction cross section is also important because of ^3H production in reactor shielding materials, and some intriguing geophysical speculations.[1]

Solutions containing known concentrations of H_3BO_3 were made by dissolving weighed amounts of National Bureau of Standards (NBS) standard reference materials (SRM's) 951 ($^{10}B/^{11}B=0.2472$) and 952 ($^{10}B/^{11}B=18.80$) (Ref. 6) in distilled water. Aliquots were pipetted into polyethylene tubes, then weighed, freeze-dried, and placed in lead containers which were evacuated and pinch-sealed as previously described.[4] Neutron irradiations were carried out in a position close to the edge of the core of the light-water moderated reactor at McMaster University

TABLE I. ^3H and ^4He from $^{10}B(n,t)2\alpha$.

Series	Sample[a]	Thermal neutron flux (cm^{-2} s^{-1})	Irradiation time (h)	Cooling time (days)	Facility[b]	^3H[c] (10^{14} g^{-1} ^{10}B)	^4He[c] (10^{18} g^{-1} ^{10}B)	^3H R_{Cd}	^4He R_{Cd}	$^{10}B(n,t)2\alpha$ thermal cross section (mb)
I	951 (4)	8.8×10^{12}	3	10–13	McM	0.47 ±0.09	21.4 ±0.9	2.0 ±0.7	119±14	4.3 ±0.5
I	951 Cd (2)	~0	3	10–13	McM	0.23 ±0.06	0.18 ±0.02			
II	951 (2)	3.2×10^{11}	24	212	NBS	0.070±0.006	6.42 ±0.03			4.2 ±0.4
II	951 (3)	3.4×10^{11}	10	178	NBS	0.042±0.010	2.70 ±0.14			6.0 ±1.5
II	952 (3)	3.4×10^{11}	10	177	NBS	0.036±0.005	2.86 ±0.09			4.8 ±0.7
III	951 (2)	8.9×10^{12}	3	285	McM	0.482±0.021	21.6 ±0.4	2.03±0.14	106±3	4.37±0.11
III	951 Cd (3)	~0	3	285	McM	0.238±0.013	0.204±0.005			
III	952 (2)	8.9×10^{12}	3	286	McM	0.518±0.025	23.1 ±0.8	2.13±0.12	108±5	4.62±0.12
III	952 Cd (2)	~0	3	286	McM	0.243±0.006	0.213±0.006			
Weighted mean								2.09±0.18	107±5	4.47±0.15

[a]SRM's 951 and 952 are boron isotope standard reference materials (Ref. 6). Numbers in parentheses are numbers of samples analyzed.
[b]McM denotes McMaster University light-water-moderated reactor. NBS indicates graphite thermal column attached to the National Bureau of Standards heavy-water-moderated reactor.
[c]Errors are estimates of random and systematic components at a level of 1σ. 2σ errors are given for weighted mean values of R_{Cd} and the $^{10}B(n,t)2\alpha$ thermal cross section.

and also in a graphite thermal column attached to the heavy-water-moderated reactor at the National Bureau of Standards. In the McMaster irradiations, some samples were wrapped with cadmium to determine the relative amounts of ^{3}H and ^{4}He from neutrons above and below the cadmium cutoff energy of about 0.5 eV.

After a waiting time to allow growth of ^{3}He from ^{3}H, mass spectrometric measurements of ^{3}He and ^{4}He ion currents were made relative to ion currents from aliquots of air containing known amounts of helium. The atmospheric ^{3}He-^{4}He ratio[7] of $1.384\pm0.006\times10^{-6}$ and a tritium half-life of 12.38 ± 0.03 y (Ref. 8) were used to calculate amounts of ^{3}H. Samples contained 1–3 μg ^{10}B in H_3BO_3 deposited inside polyethylene tubes which act as catcher foils for tritons and alpha particles produced during neutron irradiation. Several blank polyethylene tubes were irradiated with samples to determine the amounts of ^{3}H and ^{4}He produced in the polyethylene by neutrons, and the amounts of Li and B added during sample handling. The correction to observed ^{3}He for ^{3}H produced via ^{2}H$(n,\gamma)^3$H in cadmium-wrapped samples was about 0.3%, and varied from 15% to 29% in unwrapped samples. The correction to observed ^{4}He for the threshold reaction ^{12}C$(n,\alpha)^9$Be, pertinent only for McMaster irradiations, was less than 0.1%. The effect of addition of Li or B during sample handling was found to be negligible. For one series, blank tubes and aliquots of SRM's 951 and 952 were prepared in the same way as for samples, but were not irradiated. Subsequent mass spectrometric analysis showed that ^{3}H contamination in the polyethylene or the H_3BO_3 was negligible compared to that produced by ^{10}B$(n,t)2\alpha$. It will be noted that initial Li contamination of H_3BO_3 can lead to systematic error. If we suppose that *all* the ^{3}H attributed to thermal neutrons on ^{10}B is due instead to the thermal-neutron reaction ^{6}Li$(n,\alpha)^3$H, then SRM 951 contains 1.5 ppm Li and SRM 952 contains 6.8 ppm. Although such initial contamination is extremely unlikely in view of the purification procedures used for these materials[6] it was decided to measure the Li content of SRM 951 by atomic absorption spectrometry. A solution in distilled water containing 0.0404 g H_3BO_3 g^{-1} was analyzed and shown to contain <0.4 ppb Li which is equivalent to <10 ppb Li in the H_3BO_3, which means that $<0.7\%$ of the tritium attributed to the thermal-neutron reaction ^{10}B$(n,t)2\alpha$ is due to ^{6}Li$(n,\alpha)^3$H.

Results of the mass spectrometric measurements are given in Table I. Thermal-neutron cross sections for ^{10}B$(n,t)2\alpha$ are expressed relative to the accepted value of 3837 ± 9 b at 0.0253 eV for ^{10}B$(n,\alpha)^7$Li.[9] For NBS thermal-column irradiations, the ^{10}B$(n,t)2\alpha$ cross section was determined directly relative to the ^{10}B$(n,\alpha)^7$Li cross section. For irradiations in the McMaster reactor the effect of high-energy neutrons was determined by measurements on samples wrapped with 1 mm cadmium and irradiated in the same core position. The cadmium ratio R_{Cd} is defined in the usual way as the ratio of ^{3}H atoms g^{-1} ^{10}B in bare samples to that in cadmium-wrapped samples. The low R_{Cd} value of 2.09 ± 0.18 (weighted mean $\pm2\sigma$) for ^{3}H from ^{10}B is due to the relatively large cross sections for the threshold reactions ^{10}B$(n,\alpha)^7$Li* then ^{7}Li*$\rightarrow{}^3$H$+{}^4$He, and ^{10}B$(n,t)^8$Be* then ^{8}Be*$\rightarrow{}^4$He$+{}^4$He. Figure 1(a) shows the differential neutron flux spectrum measured by McCormack[10] near the edge of the McMaster reactor core. Although the flux measurements were made when the reactor was operating at 5 MW vs 2 MW for our irradiations and in a different core location, we believe that McCormack's neutron flux spectrum is accurate in a relative sense over the neutron energy range to about $\pm5\%$ for our purpose. Figure 1(b) shows the excitation function for ^{10}B(n,t) assuming that the cross section closely follows the $1/v$ Fermi law from 10^{-10} to 10^{-3} MeV, and then increases rapidly to 39 mb at 2.6 MeV. From 2.6 to 15 MeV we use cross sections measured by Frye and Gammel,[11] Wyman *et al.*,[12] and Suhaimi *et al.*[13] From 10^{-3} to 0.6 MeV, cross sections

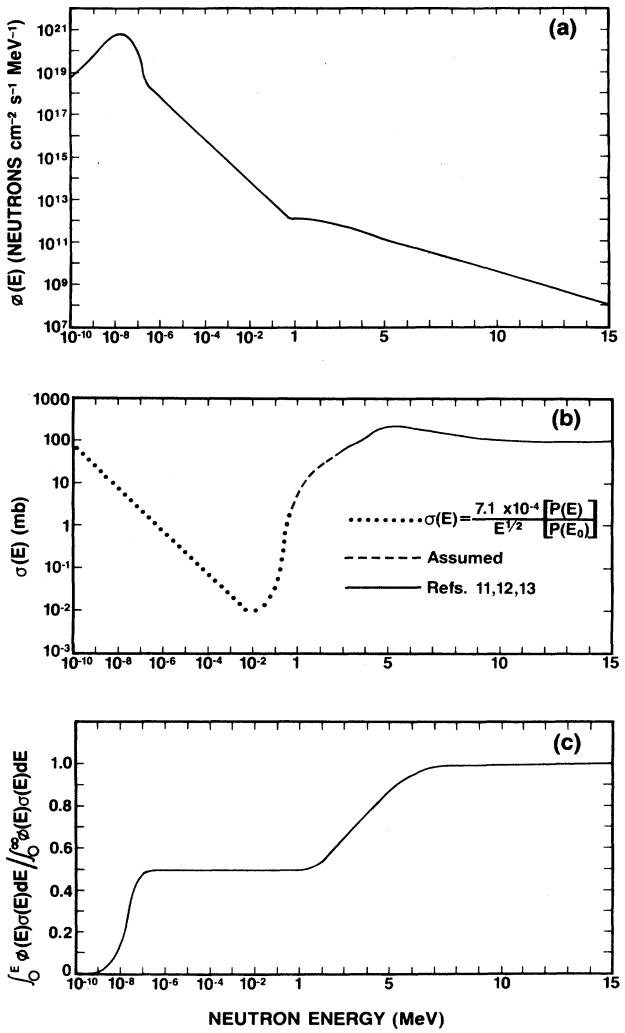

FIG. 1. (a) The measured differential neutron flux spectrum in the McMaster light-water-moderated reactor core from Ref. 10. (b) The ^{10}B(n,t) excitation function used to calculate cumulative fractional ^{3}H production illustrated in (c). $P(E)$ and $P(E_0)$ are ^{3}H barrier-penetration factors (Ref. 14) at E and thermal energies, respectively. The neutron energy scale is logarithmic below 1 MeV, and linear above 1 MeV.

(relative to the measured cross section at thermal energies) were estimated using barrier-penetration factors which were calculated using the Wentzel-Kramers-Brillouin (WKB) approximation given by Humblet *et al.*,[14] assuming a channel radius of 4.1 fm[1]. Above 0.6 MeV, the calculations give cross sections which seem far too large, so from 0.6 to 2.6 MeV estimates were made by drawing a smooth curve. Under these assumptions, R_{Cd} is calculated to be 2.02 with an estimated error of $\pm 7\%$. Although the assumption that the cross sections closely follow the $1/v$ law to 10^{-3} MeV is probably correct, the calculated values from 10^{-3} to 0.6 MeV could be in error because structure effects are ignored. Be that as it may, it should be noted that the calculated value of R_{Cd} is not very sensitive to the assumed cross sections between 10^{-3} and 2.6 MeV because this energy range accounts for only 10.7% of total ^{3}H. The good agreement between calculated R_{Cd} and the measured value indicates that if resonances exist in the $^{10}B(n,t)2\alpha$ excitation function below 1.5 MeV, they are of minor importance for ^{3}H production by neutrons in a typical light-water-moderated reactor core.

In summary, our value of 4.47 ± 0.15 mb is in accord with the recent value of Kavanagh and Marcley[1] but not with other measurements, and thus it seems certain that production of tritium by the thermal-neutron reaction $^{10}B(n,t)2\alpha$ is negligible in most instances using the method for trace lithium assay based on the thermal-neutron reaction $^6Li(n,t)\alpha$.[4]

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