Letter

## Measurements of the $^{13}$ C( $\alpha$ , n) $^{16}$ O cross section up to $E_{\alpha} = 8$ MeV

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We present results from direct measurements of the total  $^{13}\text{C}(\alpha,n)$   $^{16}\text{O}$  cross section over laboratory energies  $E_{\alpha}=2.9$ –8.0 MeV, performed with the  $^{3}\text{HeBF}_{3}$  giant barrel neutron detector at the Edwards Accelerator Laboratory. The cross sections reported in this work are considerably lower than prior direct measurements for  $E_{\alpha}>5$  MeV, in agreement with prior corrections based on Hauser-Feshbach estimates. However, applying branching ratios based on Hauser-Feshbach estimates to our data would not reproduce existing direct measurements for partial decay channels. This indicates both the promise and limitations of Hauser-Feshbach for branching ratio estimates of  $(\alpha,n)$  reactions on light nuclides. The  $^{13}\text{C}(\alpha,n)$  thick-target yields inferred from this work for  $E_{\alpha}>6$  MeV are significantly larger than those currently employed in estimates of dark matter and neutrino detector backgrounds.

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The reaction  $^{13}C(\alpha, n)$   $^{16}O$  plays a prominent role in nuclear astrophysics and nuclear applications. It is a nearly ubiquitous background in low-energy  $(\alpha, n)$  and  $(\alpha, \gamma)$  cross section measurements [1], a background for geoneutrino [2] and dark matter [3] detection experiments, a neutron source for the astrophysical *s*-process [4], and a possible  $\alpha$ -fluence monitor reaction for  $\alpha$ -induced reaction measurements [5]. Meanwhile, the reverse reaction  $^{16}O(n,\alpha)$   $^{13}C$  is of interest for nuclear applications [6], making the Nuclear Energy Agency list of high-priority requests [7]. Recently, significant progress has been made in pushing direct measurements of the  $^{13}C(\alpha, n)$  cross section into the energy window of astrophysical interest [8,9]. However, at energies above  $\approx$ 5 MeV, that are of interest for the other applications stated above, the cross section is much more uncertain.

To date, above laboratory  $\alpha$  energies  $E_{\alpha} > 2$  MeV, three angle-integrated direct measurements exist of the  $^{13}\text{C}(\alpha,n)$  total cross section [10–12]. Although there is some level of agreement between the data sets, those of Refs. [10,12] tend to lay roughly 30% below the data of Ref. [11], and resonance-like structures in the data of Ref. [12] tend to be located roughly 40 keV below similar features in the data of Refs. [10,11]. The agreement is improved by applying a  $\times 0.8$  scaling to Ref. [11] as suggested by their note added in proof. Only the data of Ref. [12] cover  $E_{\alpha} > 5.6$  MeV. However, these data are known to be problematic in this energy regime [13–16]. While Ref. [12] assumed a neutron detection

efficiency corresponding to neutron energies for the  $(\alpha, n_0)$  branch of the reaction, at  $E_{\alpha} \approx 5$  MeV, decay branches to excited states in  $^{16}$ O open and have significant cross sections. The neutron energies for these branches are far lower than the ground-state branch and, for the neutron detector used by Ref. [12], the associated neutron detection efficiency was far higher. Therefore, it can be concluded that Ref. [12] has overestimated the  $^{13}$ C( $\alpha$ , n)  $^{16}$ O cross section for  $E_{\alpha} \gtrsim 5$  MeV and therefore has overestimated the associated dark matter and neutrino detector background [2].

While the  ${}^{13}C(\alpha, n)$  branching has been measured for some decay branches and  $(n, \alpha)$  measurements, e.g., Refs. [2,17– 19], most others rely on estimates. One approach for estimating branchings is to use the Hauser-Feshbach (HF) formalism, as adopted by Ref. [15] for  $^{13}C(\alpha, n)$  and by Ref. [20] for light nuclides more generally. As the HF formalism is based on average nuclear properties, it is expected that decay branchings based on this formalism will be accurate on average when considered as an ensemble, provided that the transmission functions are correct. However, substantial deviations are likely when considering light nuclides, where a small number of intermediate states are involved and the choice of optical potential is less clear, as is the case for  $^{13}C(\alpha, n)$  at the energies of interest for this work. As such, new direct measurements mitigating the issues of Ref. [12] are desirable. Therefore, in this work we report measurements of the  $^{13}$ C( $\alpha$ , n)  $^{16}$ O cross section using the recently developed <sup>3</sup>HeBF<sub>3</sub> giant barrel (HeBGB) neutron counter [21] up to  $E_{\alpha} = 8$  MeV. HeBGB provides a near-constant neutron detection efficiency over the corresponding large range of neutron energies (up to  $E_n = 9.5 \text{ MeV } [21]$ ), mitigating the impact of the  $(\alpha, n)$  decay branchings on the inferred  ${}^{13}C(\alpha, n)$  yield.

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Measurements were performed at the Edwards Accelerator Laboratory at Ohio University [22], where the full setup is described in Ref. [21]. A helium beam produced by an Alphatross ion source was accelerated using a 4.5 MV T-Type tandem Pelletron to an energy between  $E_{\alpha} = 2.9-8.0$  MeV, departing the accelerator as He<sup>2+</sup>. The beam was analyzed with an energy-calibrated 90° dipole magnet and slits with a gap of 0.152 cm, resulting in a beam energy uncertainty of 0.2%. The beam passed through a 0.5-cm-diameter goldplated collimator located approximately 48 cm upstream of the target location and was impinged on a <sup>13</sup>C target located on a gold-plated ladder at the center of the HeBGB moderator. The <sup>13</sup>C target, produced by Arizona Carbon Foil, Inc. via electron-beam evaporation of carbon enriched to ≥99% <sup>13</sup>C onto a copper substrate, was determined to have a 13C thickness (areal density) of  $n_t = (1.12 \pm 0.05) \times 10^{18}$  atoms/cm<sup>2</sup> via  $\alpha$ -elastic scattering,  $\alpha$  energy-loss measurements, and a scan of the 1.053 MeV  $^{13}$ C( $\alpha$ , n) resonance. The incident charge was measured by summing the beam current of the electrically isolated target ladder and target chamber within HeBGB. To ensure the entirety of the beam was on target, at each measurement energy the beam was first tuned through an empty frame in the target ladder and the integrated beam charge  $Q_{\alpha}$  was recorded using a downstream Faraday cup. At periodic energy spacings, measurements were performed on a copper substrate identical to the backing used for the <sup>13</sup>C targets in order to ascertain the neutron background. The contribution of  $^{65}\mathrm{Cu}(\alpha,n)$  to the yield was negligible until  $E_{\alpha} \approx 7$  MeV, reaching nearly 25% at  $E_{\alpha} = 8$  MeV. The yields from the <sup>13</sup>C target were corrected for this background, with an uncertainty assumed to be half of the correction. Neutrons were detected using the <sup>3</sup>He and BF<sub>3</sub> neutron-sensitive proportional counters, configured as described in Ref. [21]. A pulser, adjusted to provide a signal outside of the neutron spectrum, was used to monitor the proportional counter deadtime and therefore the live fraction  $f_{live}$  of the data acquisition.

While HeBGB provides nearly  $4\pi$  coverage, having a nearly constant absolute efficiency of 0.075(12) for isotropic neutrons, our MCNP6 [23] simulation results show a dependence of the efficiency on the neutron angular distribution [21]. We have taken this into account in our efficiency at each beam energy  $\epsilon(E_{\alpha})$  using the current best estimate of the angular distributions. Up to  $E_{\alpha} = 5$  MeV, our angular distributions were determined via R-matrix calculations [24]. The R-matrix calculations were taken from the ENDF/B-VIII.0 evaluation [25], updated for more recent narrow resonance information [26], which reproduced the angular distribution data from Ref. [27]. These relative efficiency corrections were typically  $\approx 10\%$ , but were as large as 27% in some cases. For  $E_{\alpha} > 5$  MeV, where no experimental angular distribution data were available, we estimated the  ${}^{13}C(\alpha, n)$  angular distribution based on HF calculations performed with TALYS [28], which follows the formalism described by Ref. [29]. This approach necessarily relied on adopting fractions for the various decay branches. To estimate the uncertainty due to this assumption, we performed efficiency corrections assuming two extreme cases, that all of the decays for  $E_{\alpha} > 5$  MeV proceed through either the  $(\alpha, n_0)$  or  $(\alpha, n_2)$  branches in order to get upper and lower bounds for the correction. These two

branches were chosen as they have quite different predicted angular distributions, they produce quite different energies for the outgoing neutrons, and they were estimated to be the two dominant decay branches [15]. The typical total correction for the efficiency at  $E_{\alpha} > 5$  MeV was 14% (relative), where on average a 5% relative correction stemmed from decay branching assumptions.

In the Supplemental Material [31], we provide a table of  $E_{\alpha}$ , the measured cross section and uncertainty, the dead-time and background-corrected measured yields and uncertainties, the branching fraction to each decay branch  $b_i$ , and, for each decay branch, the Legendre polynomial decomposition of the efficiency  $\epsilon_{\ell}$ , and the adopted Legendre polynomial coefficients  $a_{\ell}$ . An additional Supplemental Material [31] file explains how to employ these data, while a full description of the method will be contained in Ref. [24]. This enables a straightforward reanalysis of our results when improved angular distribution and/or branching data become available.

Over the entire energy range of our measurements, the bulk of the efficiency correction arises from the  $a_2$  Legendre coefficient. This coefficient is usually positive in the R-matrix calculations and is always positive for the TALYS calculations. The generally positive  $a_2$  coefficients result in the efficiency almost always being reduced relative to the isotropic value. This finding is easily understood from Fig. 7 of Brandenburg  $et\ al.\ [21]$ , where an efficiency reduction in the forward and backward directions is shown. This angular dependence of efficiency is expected for any detector of this type which has the neutron detector tubes located parallel to the beam axis with approximate axial symmetry.

In order to calculate the energy at the center of the target, for which we report a cross section, we employ a stopping power based on the world data [32] and commonly used theoretical stopping powers [33] at these energies, adopting the difference between the largest and smallest stopping powers for the stopping power uncertainty and the median of these extremes as the stopping power. For the energy range relevant for this work, the experimental stopping powers [34–36] agree to within 7% and are in agreement with predictions from SRIM2013 [33].

The cross section at each energy is calculated according to the thin-target approximation, where the reported energy is the center-of-mass energy at the center of the target. In the present case, this approximation introduces negligible error, except where there are resonance structures with widths comparable to or smaller than the energy loss in the target, which is approximately 20 keV. The cross section is described by  $\sigma(E_{\alpha}) = Y(E_{\alpha})/[n_{\rm t}\epsilon(E_{\alpha})]$ , where  $Y(E_{\alpha})$  is the number of detected neutrons per incident beam particle and  $\epsilon(E_{\alpha})$  is the total detection efficiency. Our  $^{13}C(\alpha,n)$  cross section results are shown in Figs. 1 and 2. Our systematic cross section uncertainty budget is summarized in Table I.

For  $E_{\alpha} \lesssim 5$  MeV, our results are generally in agreement with prior works, as shown in the lower panels of Fig. 1. We see the same cross section features, but the energy of these features tends to be in between the energies reported by Refs. [11,12]. For  $E_{\alpha} \approx 3$ –4 MeV, our reported cross sections are generally in between the cross sections reported by Refs. [11,12], but agree with both data sets given our neutron

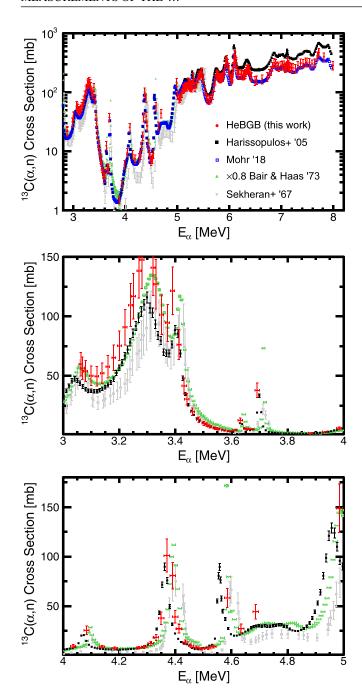


FIG. 1.  $^{13}$ C( $\alpha$ , n) total cross section results from this work compared to direct measurements of Refs. [10–12]. The results of Ref. [11] have been multiplied by  $\times 0.8$  as suggested by their note added in the proof. The data of Ref. [12] have also been adjusted as suggested by Ref. [15]. The upper panel shows the entire energy range measured in this work, while the lower panels show selected energy ranges. Uncertainties for this work are dominated by a correlated systematic uncertainty of 16%.

detection efficiency uncertainty. For  $E_{\alpha} \approx 4-5$  MeV, our cross sections are closer to the results of Ref. [12]. The discrepancies in  $E_{\alpha}$  are likely due to underestimation of uncertainties in beam energy calibration and energy loss corrections. Discrepancies in reported cross section magnitudes are likely due

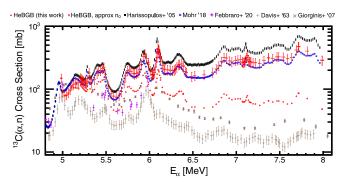


FIG. 2.  $^{13}C(\alpha, n)$  total cross section results from this work compared to direct measurements of Ref. [12], the adjusted data of Ref. [12] suggested by Ref. [15], the  $(\alpha, n_0)$  results from Ref. [2], and the  $(\alpha, n_0)$  results obtained by applying the  $n_0$  branching calculated in Ref. [15] to the total cross section reported in this work ("approx  $n_0$ "). Results are also shown for  $(\alpha, n_0)$  when applying reciprocity (using EXFOR [30]) to the  $^{16}O(n, \alpha)$  data of Refs. [17,18]. Uncertainties for this work are dominated by a correlated systematic uncertainty of 16%.

to inadequate accounting of target thickness and/or neutron detection efficiency uncertainties. In the latter case, we have found the impact of the  $^{13}$ C( $\alpha$ , n) angular distributions and branchings to be important.

Figure 2 shows our results in the energy range where neutron-emission channels beyond the ground-state branch open. Our total cross section results are remarkably close to the results of Ref. [15], which provided a HF based correction to the measurements of Ref. [12]. For  $E_{\alpha} > 5.5$  MeV, our results are on average 4% higher than those of Ref. [15], who indicated that their total cross section results are likely accurate to within 15%. However, when applying the  $n_0$  decay branching calculated in Ref. [15] to the present work, there is poor agreement with the  $(\alpha, n_0)$  measurements of Ref. [2] and those inferred by applying reciprocity to the  ${}^{16}\mathrm{O}(n,\alpha)$  data of Refs. [17,18]. For  $E_{\alpha} > 5.5$  MeV, our average deviations from Refs. [2,18] are about 50%. Deviations from Ref. [17] are much larger, but we note that  $(\alpha, n_0)$  cross sections inferred from that work are not in agreement with Refs. [2,18], which roughly agree with each other. As such, it appears that the decay branchings of Ref. [15] are correct on average, but are inadequate for predicting the values for specific decay branches. This is consistent with the HF formalism, where predictions are expected to be correct, on average, provided that the transmission functions are correct. The HF predictions are expected to be most accurate when averaged over a range

TABLE I. Summary of systematic uncertainty estimates for the  $^{13}\text{C}(\alpha, n)$   $^{16}\text{O}$  cross section measurements.

Systematic uncertainty contribution	%
Neutron detection efficiency	16
Target thickness	4
Integrated beam current	1
Total	16

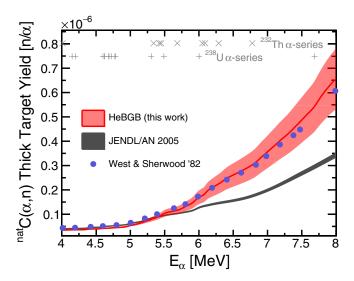


FIG. 3.  $^{\mathrm{nat}}\mathrm{C}(\alpha,n)$  thick-target yields, assuming a carbon target with natural abundances [this energy range is below the  $^{12}\mathrm{C}(\alpha,n)$  threshold.], calculated using our cross section data for  $E_{\alpha} > 5$  MeV and the data of Ref. [12] below (HeBGB), where the bands of each represent their total uncertainties, compared to yields resulting from the evaluated cross section of Ref. [20] (JENDL/AN 2005), as well as measured  $^{\mathrm{nat}}\mathrm{C}(\alpha,n)$  thick-target yields from Ref. [38]. The  $^{232}\mathrm{Th}$  and  $^{238}\mathrm{U}$  decay series  $E_{\alpha}$  with decay branches greater than 1% are also shown by the  $\times$  and +, respectively.

of energies and a number of final states, but can produce significant deviations from experiment over smaller energy regions and for specific decay channels. Since it is the overall cross section that is important for  $(\alpha, n)$  background contributions to neutrino and dark matter detectors, our results show that HF based estimates of decay branchings are adequate to within tens-of-percent precision for  $^{13}C(\alpha, n)$ . Whether or not this accuracy extends to other light nuclei is an open question. These findings also support the  $n + ^{16}O$  optical potential that was utilized in Ref. [15].

We calculated infinitely thick target yields to assess the potential impact of our results on neutrino and dark matter detector backgrounds contributed by the  $^{13}C(\alpha, n)$  reaction. Reference [37] employed the evaluated cross section of Ref. [20] for  $E_{\alpha}$  above the  $(\alpha, n_1)$  threshold, thus we compare to yields calculated with that data set. In Fig. 3, we compare thick-target yields calculated based on our cross section measurements to yields estimated using the evaluated cross sections of Ref. [20], along with  $^{\rm nat}C(\alpha, n)$  thick-target

yields measured by Ref. [38], where we also highlight  $E_{\alpha}$  of typical actinide contaminants [37] for context. Where our data are sparse at  $E_{\alpha} < 5$  MeV we use the cross section data of Ref. [12]. We use the same stopping powers as discussed previously for our energy loss uncertainty.

Thick-target yields calculated using our cross section data are in excellent agreement with the direct measurements of Ref. [38]. However, when compared to yields calculated with the commonly adopted data set of Ref. [20], it is apparent that yields are significantly larger for  $E_{\alpha} > 6$  MeV. For example, at  $E_{\alpha} = 6$  MeV, our upper estimate for the yield is nearly 50% larger. At the  $\alpha$ -decay energy of  $^{214}$ Po (7.687 MeV) from the  $^{238}$ U decay series, our best estimate for the yield is 56% larger, while our lower and upper limits are 27% and 87% larger, respectively. This suggests that the  $^{13}$ C( $\alpha$ , n) background estimates for neutrino and dark matter detector backgrounds, e.g., as in Ref. [37], are likely underestimates.

In conclusion, we report  $^{13}C(\alpha, n)$  cross sections measured with the HeBGB detector at the Edwards Accelerator Laboratory at Ohio University. We find general agreement with prior results up to  $E_{\alpha} \approx 5$  MeV. Above this energy, our results are well below the only prior direct measurement of the total cross section over this energy region. Our total cross sections are in agreement with HF based corrections applied to prior direct measurement data, but the  $n_0$  partial cross sections inferred from applying this HF correction to our results do not agree with directly measured  $(\alpha, n_0)$  cross sections. This highlights both the promise and limitations of applying HF calculations of  $(\alpha, n)$  reactions for low-mass nuclides. The <sup>nat</sup>C $(\alpha, n)$ thick-target yields reported in this work are significantly larger than commonly used evaluated cross sections at  $E_{\alpha} > 6$  MeV. This suggests that contributions from the  ${}^{13}C(\alpha, n)$  reaction to the background of neutrino and dark matter detectors are larger than current estimates.

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