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¹² C(α,γ)¹⁶ O Capture Cross Section Below 3.2 MeV[†]

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The ${}^{12}C(\alpha, \gamma){}^{16}O$ capture cross section was measured for α -particle energies between 1.86 MeV ($\sigma = 1.1 \pm 0.4$ nb) and 3.11 MeV ($\sigma = 29 \pm 4$ nb) using a pulsed ⁴He⁺ beam from the ORNL 6-MV Van de Graaff accelerator and a 23- by 30-cm NaI(Tl) crystal viewed by six matched, bialkali photomultiplier tubes. An upper limit was obtained for the capture cross section at E_{α} =1.6 MeV. The over-all time resolution (full width at half maximum) of the system for 8-10-MeV pulses due to γ rays is 2.7 nsec. Enriched (99.94%) ¹²C targets ranging in thickness from 98 to 178 μ g/cm² were used. Pulses resulting from fast neutrons [from the ¹³C $(\alpha, n)^{16}$ or reaction] interacting with the NaI(Tl) crystal were further separated from true γ pulses through the use of a new technique based on rise-time distribution differences of the respective neutron- and γ -ray-produced pulses. The face of the crystal (shielded with a 10.2 cm thickness of ⁶LiH) was 12.2 cm from the target. The astrophysical significance of this reaction in the helium-burning sequence of stellar nucleosynthesis is also discussed.

INTRODUCTION

The ${}^{12}C(\alpha, \gamma){}^{16}O$ reaction is a major link in the "helium-burning" sequence of events believed to occur during stellar evolution.¹ The formation of ¹⁶O in stellar interiors by radiative α capture is thought to be strongly influenced by the 7.12-MeV (1⁻) level in 16 O, which is 40 keV below the 12 C + ⁴He threshold.² While the γ -ray width of this state has been measured,³ the dimensionless reduced α width θ_{α}^{2} , has only been indirectly inferred by Loebenstein *et al.*⁴ through the ${}^{6}\text{Li}({}^{12}\text{C}, d){}^{16}\text{O}*$ reaction. They give the limits 0.06-0.14 for the extracted θ_{α}^{2} for the 7.12-MeV state in ¹⁶O. Tombrello⁵ has calculated the interference effects between the 1⁻ level at 7.12 and the 1⁻ state at 9.59-MeV excitation in ¹⁶O, and found that the ¹²C(α, γ) ¹⁶O capture cross section in the region between these "resonances" will be affected. It is proposed that with a sufficiently accurate measurement of the cross section it would be possible to determine θ_{α}^{2} for the 7.12-MeV state. Larson and Spear⁶ found a value of 36 nb for the capture cross section at E_{α} = 3.2 MeV using enriched ¹²C targets, while Adams *et al.*⁷ using time-of-flight methods and natural-carbon targets presented a preliminary value of 10 nb at about $E_{\alpha} = 2.7$ MeV. One of us (RJJ)⁸ measured the cross section using enriched ¹²C targets and a dc He⁺ beam between $E_{\alpha} = 2.54$ and 3.6 MeV, with a recent least-squares analysis of the data yielding the value 4 ± 2.5 nb at the lower point. These measurements lacked the sensitivity required for a reliable determination of the reduced α -particle width of the 7.12-MeV level.

In the present paper, we present the results of a study of the radiative α capture in ¹²C for α -particle energies between 1.6 and 3.2 MeV. A major problem in low-yield α -capture experiments, where the excitation energy lies between 8 and 10 MeV, is the prolific fast-neutron background resulting from the ${}^{13}C(\alpha, n){}^{16}O$ reaction, which has a cross section considerably larger than capture cross sections. Beam interaction with naturalcarbon buildup on targets, collimators, and other material exposed to the beam generates these fast neutrons. Also, certain contaminants or isotopic impurities in the target itself may yield fast neutrons. In all cases, the result (when using a NaI (T1) γ -ray spectrometer) is an approximately exponentially decreasing background in the energy (pulse-height) spectrum in the 8- to 10-MeV region. Various techniques were used in the present experiment to minimize the contribution to the background resulting from these fast neutrons.

EXPERIMENTAL METHOD

For this measurement we used the terminally pulsed ⁴He⁺ beam (currents ≤ 5 mA) from the Oak Ridge National Laboratory 6-MV Van de Graaff Accelerator. The He⁺ pulses had a time resolution [Full width at half maximum (FWHM)] of less than 2 nsec. A relatively large (23- by 30-cm) NaI(Tl) scintillator viewed by six matched bialkali photomultiplier tubes was used as the γ -ray detector. The efficiency of the NaI(T1) detector (positioned at an 84.5° angle with respect to the beam axis) was determined by computing the total intrinsic efficiency from tabulated attenuation coefficients.⁹ The "peak" efficiency was then obtained by multiplying this quantity by the peak-to-total ratio which was experimentally determined with 9.17-MeV γ rays from the ${}^{13}C(p,\gamma){}^{14}N$ reaction. This reaction and the ²⁴Mg(α, γ)²⁸Si reaction were used to calibrate the energy-analyzing magnet of the accelerator. The resonance energies used were obtained from published values.¹⁰ Targets (prepared by the Isotopes Division of ORNL) enriched to 99.94% in ¹²C were fabricated by "cracking" enriched acetylene with a high-frequency discharge onto 0.025-cm tantalum backings. Thicknesses determined by weighing (with a microbal-



FIG. 1. Electronic circuit used for the ${}^{12}C(\alpha, \gamma){}^{16}O$ capture cross-section measurement. The leading-edge discriminator could be set to trigger at a point equivalent to 2.5 MeV on the pulse; thus, the fast-neutron-induced events could be further separated from the prompt- γ -induced events.

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ance) the backings before and after deposition ranged between 98 ± 2 and $178 \pm 3 \ \mu g/cm^2$. The angle between the beam axis and the perpendicular to the plane of the target was 45°. At first we used a 4- by 1024-channel two-dimensional analyzing system (Fig. 1) in order to search for cascade radiation from the 9.59-MeV level; however, no definite indication of cascade radiation was observed, and the measurements reported here result from a study of the ground-state transition only. The signal for the α -beam pick off was obtained at the target through the use of a ferritecore transformer suggested by one of the authors (RLM). This permitted the energy of the pulsed beam to be varied without changing the delay times of the system. The NaI(Tl) crystal was completely encased in 10.2 cm of lead except for the front face (12.2 cm from the target), which was shielded with 10.2 cm of ⁶LiH which further attenuated the fast neutrons emitted by the target chamber. An ultraclean target assembly was used consisting of all-metal vacuum seals (mainly indium "O" rings) from the diffusion pump to the target holder. The diffusion pump was trapped with a liquid-nitrogencooled baffle. Also, the beam had to traverse two liquid-nitrogen-cooled in-line traps before striking the target. An Orb-Ion¹¹ pump was also used between the two in-line traps for part of the experiment. Typical operating pressures at the target were $\leq 2 \times 10^{-7}$ Torr. A suppressor ring in front of the target was biased at -300 V to minimize secondary electron emission from the target. Also, there was a magnetically activated holder which enabled a MgO "flag" to be placed into the He⁺ beam. γ radiation resulting from the ²⁴Mg(α, γ)-

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FIG. 2. Schematic representation of 8-10-MeV pulses due to fast neutrons and γ rays in a large NaI(Tl) crystal showing the additional delay of the fast-neutron-induced pulse as a result of inelastic scatterings within the crystal. The time interval (t_4-t_3) will be larger than the time interval (t_2-t_1) .

²⁸Si reaction permitted a simple and quick means for adjusting the time and energy windows, and to ensure proper operation of the complete electronics system before the commencement of long dataacquisition periods.

The 400-channel analyzer (gated for "on-time" pulses) was used to monitor the energy (pulseheight) spectrum. A new method¹² of discriminating against fast neutrons based on rise-time distribution differences was used (Fig. 2). By setting the level of the leading-edge discriminator to the energy equivalence of approximately 2.5 MeV, it is possible to measure the additional delay due to inelastic scattering of the fast neutron, thereby spreading the distribution of pulses due to fast neutrons toward the direction of late-arrival times. A spectrum comparison of this effect will be presented in the following section.

The data were dumped through a CDC-160-A computer on magnetic tape in binary-coded decimal format. This tape was rewritten onto an 18bit binary tape using the ORNL CDC-1604 computer. The format of this binary tape was then compatible with a series of programs developed by Marusak¹³ for the PDP-7 computer that allows one



FIG. 3. Time-of-flight pulse-height spectrum for the ${}^{12}C (\alpha, \gamma){}^{16}O$ reaction obtained at $E_{\alpha} = 3.11$ MeV (at the center of the target) corresponding to an excitation energy E_x in ${}^{16}O$ of 9.5 MeV. The leading-edge discriminator was set at a low level (to eliminate the observation of time delays due to inelastic scattering of fast neutrons) for comparison with the next figure. Note that the time calibration is 0.8 nsec/channel. The line through the data is a guide to the eye.



FIG. 4. Time-of-flight pulse-height spectrum at $E_{\alpha} = 2.70$ MeV. The leading-edge discriminator adjusted to trigger at an energy equivalent to 2.5 MeV. The fast-neutron-induced peak has been broadened toward late-arrival times. Note that the time calibration is 0.22 nsec/channel.

FIG. 5. Pulse-height spectrum for time-offlight data corresponding to α -particle energy of 2.47 MeV showing a considerably decreased ratio of γ -ray-induced peak area to fast-neutron-induced peak area as compared to the one observed in Fig. 4.

FIG. 6. Pulse-height spectrum for time-offlight data at $E_{\alpha} = 2.28$ MeV.

to use the associated light pen in subtracting backgrounds and extracting peak areas. In analyzing the data, corrections (up to 12%) have been made for the attenuation of the γ radiation in traversing the ⁶LiH shield. Also, corrections were made for the observed angular distribution (assuming a $0^{+\frac{p}{2}} 1^{-\frac{p}{2}} 0^{+}$ radiative transition) using computed correction factors in the sum over Legendre polynomials.¹⁴

TIME-OF-FLIGHT SPECTRA

For the following figures, the α -particle energies given correspond to the energies half way through the target. The curves connecting the data points are guides to the eye. The time-of-flight pulse-height spectrum at E_{α} = 3.11 MeV shown in Fig. 3 was obtained with an enriched ¹²C target having a thickness of 113 μ g/cm². It should be noted that in this figure the new "inelastic-delay" technique for discrimination was intentionally rendered inactive (by setting the leading-edge fast-discriminator level very low). For comparison with the following figures where this technique was activated, note that the peak due to fast neutrons (the large one) has a FWHM of about 12 nsec, whereas, for the γ -ray-produced peak, the FWHM is approximately 2.5 nsec. The pulse-height spectrum was obtained near the peak in the ${}^{12}C(\alpha,\gamma){}^{16}O$ resonance at E_{α} = 3.24 MeV, corresponding to the broad ($\Gamma_{\rm cm}$ ~600 keV) 1⁻ level at 9.59-MeV excitation in ¹⁶O. The measured cross section is 29 ± 4 nb. For the time-of-flight spectrum obtained at $E_{\alpha} = 2.70 \text{ MeV}$ (Fig. 4) where the "inelastic-delay"

technique was used (note that the time calibration is now 0.22 nsec/channel), the peak due to the fast neutrons has a FWHM of about 30 nsec where the additional spread is toward late-arrival times. The γ -ray-produced peak still has a time resolution (FWHM) of about 2.5 nsec. The measured cross section is 14 ± 2 nb. The time-of-flight spectrum obtained at $E_{\alpha} = 2.47$ MeV (Fig. 5) yields a value of 5.0 ± 0.7 nb for the cross section. From Fig. 6 it can be seen that the ${}^{12}C(\alpha, \gamma){}^{16}O$ capture cross section $(3.5 \pm 0.5 \text{ nb})$ is decreasing more rapidly in this energy region than the fast-neutron background due to the ${}^{13}C(\alpha, n){}^{16}O$ reaction. The data shown in Fig. 7 for $E_{\alpha} = 1.86$ MeV was smoothed for channels having more than 15 counts, using the formula $S_j = 0.33(S_{j-1} + S_j + S_{j+1})$, where S_{i} = the number of counts in the *j*th channel. The measured capture cross section was found to be 1.1 ± 0.3 nb. Time-of-flight spectra taken with a cleaned out-gassed Ta blank at a few α -particle energies between 2 and 3 MeV yielded no observable γ peaks and very little indication of pulses due to fast neutrons.

A trace of deuterium in the ion sources used has been suggested as a possible source of background in a ${}^{12}C + \alpha$ experiment, since the D_2^+ molecular ion would follow the main ${}^{4}\text{He}^+$ beam through the accelerator quite closely. The bombardment of ${}^{12}C$ by 1.7-MeV deuterons results in 9.4-MeV cascade radiation to the 2.31-MeV first excited state in ${}^{14}N$. Of the several reasons to consider this background negligible, perhaps the strongest is based on travel time. The 2-MeV He⁺ ions take



FIG. 7. Time-of-flight pulse-height spectrum at $E_{\alpha} = 1.86$ MeV corresponding to an excitation energy in ¹⁶O of 8.6 MeV. The data for this spectrum has been smoothed by averaging in groups of three channels, for all channels containing more than 15 counts.

over 1.9 μ sec to travel from the pulser in the Van de Graaff terminal through buncher, accelerator, analyzing magnet, and beam piping to the target. The heavier D_2^+ ions arrive more than 6.2 nsec later. Since the cross sections we measured are based on the area under the He⁺-induced γ -ray peaks with 2.7-nsec resolution (FWHM), any deuteron reaction products should have been well resolved. In fact, if present, they are obscured by the prominent ${}^{13}C(\alpha, n){}^{16}O$ background (see Figs. 3-8).

An essentially null measurement for the capture cross section was obtained at $E_{\alpha} = 1.6$ MeV, and the data are presented in Fig. 8. After a bombardment corresponding to 0.133 C of He⁺ on a $174-\mu g/cm^2$ enriched ¹²C target, there was no significant peak in the time-of-flight spectrum to indicate γ -ray yield resulting from the ¹²C(α, γ)¹⁶O reaction. Based on the background present in the region of interest, an upper limit of 0.3 nb has been assigned to the capture cross section at $E_{\alpha} = 1.6$ MeV.

The ${}^{12}C(\alpha, \gamma){}^{16}O$ capture cross sections measured in this experiment are summarized in Fig. 9. Although other factors (such as detector efficiency, target thickness, and statistics) were noted, the major contribution to the errors in the low-energy cross-section measurements was the subtraction of background. The value of 29 ± 4 nb at $E_{\alpha} = 3.11$ MeV is in reasonable agreement with the value of 36 nb reported by Larson and Spear⁶ at $E_{\alpha} = 3.2$ MeV. The value of 14 ± 2 nb at $E_{\alpha} = 2.7$ MeV is somewhat higher than the 10 nb value measured by Adams *et al.*,⁷ but the difference is not felt to be significant. The measured capture cross sections presented here are also consistent with



FIG. 9. The ${}^{12}C(\alpha,\gamma){}^{16}O$ capture cross section measured in the present experiment. The errors include the effects due to target-thickness uncertainty, detector-efficiency uncertainty, current integration, statistics, and background subtraction. Refer to text for comparison of data with previous results.

values (for example, 32 ± 6 and 4 ± 2.5 nb at $E_{\alpha} = 3.4$ and 2.5 MeV, respectively) derived from a recent least-squares analysis of the data obtained earlier by one of us (RJJ).⁸



FIG. 8. Time-of-flight pulse-height spectrum at $E_{\alpha} = 1.6$ MeV. Note that in this spectrum there is essentially no evidence of a γ -ray-induced peak resulting from the ¹²C (α , γ) ¹⁶O reaction.

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ASTROPHYSICAL INTEREST

The significant parameter required for a reasonable determination of the stellar reaction rate for the ${}^{12}C(\alpha,\gamma){}^{16}O$ reaction is the reduced α -particle width θ_{α}^2 , since the two 1⁻ states in ${}^{16}O$ involved at 7.12 and 9.59 MeV interfere and cause an (α,γ) cross section that is directly related to the unknown θ_{α}^2 of the lower state. The calculation of this effect is being investigated by Tombrello.⁵

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Radiative np Capture and Meson Exchange Currents*

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The experimental cross section for radiative np capture is 334.2 ± 0.5 mb. Theoretical calculations to date yield a result about 10% lower. In this work we calculate a number of mesonic exchange currents, using a simple phenomenological method and nonrelativistic wave functions. We obtain an increase of about 2-3%, almost entirely due to π exchange. Since this is numerically too small to resolve the discrepancy, we discuss other possible contributions to the cross sections. A review section is included in which we discuss previous work on the impulse approximation and the meson exchange effect; no discrepancies remain between the numerically accurate theoretical treatments, and the errors in the remaining treatments are discussed.

1. INTRODUCTION

It has been emphasized,¹ notably by Noyes,² that the cross section for thermal-neutron capture by hydrogen is about 10% higher than most theoretical estimates. The work of Cox, Wynchank, and Collie (CWC),³ for example, indicates that the cross section is 334.2 ± 0.5 mb. The extraordinary accuracy and consistency of this measurement with other measurements taken with various target