α-particle Induced Yield of 6.13 MeV γ-rays in Carbon

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ABSTRACT

Results from a preliminary accelerator-based experiment conducted at the Harwell Tandem Van de Graaff generator to estimate the thick target integrated over angle yield of 6.129 MeV γ-rays from α-particles in the MeV range stopping in natural carbon are briefly outlined. The results should be useful in planning future detailed experiments to generate data for applications and also to benchmark thin target cross section evaluations. The estimated yields referred to a natural ^{13}C abundance of 1.09 atom % at 5.597, 5.801, and 5.999 MeV in units of 6.129 MeV γ-rays per 10^8 α-particles are: 1.04, 1.62 and 3.31, respectively. An indicative overall relative uncertainty at the 68 % confidence level is estimated to be about 6%, although we caution that, being undertaken only as a feasibility study, the checks and balances we would usually conduct were not performed.

Key words: α-induced reaction, thick target yield in carbon, nuclear data

INTRODUCTION

The $^{13}C(\alpha,n)^{16}O$ reaction is exothermic with a Q-value of approximately 2.2156 MeV. As the α-particle energy increases the accessible levels of the compound nucleus ^{17}O are probed but below about 5.0135 MeV the only energetically allowed neutron emission possibility leaves ^{16}O in the 0^+ ground state (gs). This is a consequence of ^{16}O being doubly magic ("closed shell") which results in a relatively high energy first excited state, which is at 6.0491 MeV above the gs and is also 0^+. A 6.049 MeV γ-ray is never seen, however, as the state decays via internal pair conversion. Above 5.1192 MeV the second excited state in ^{16}O is accessible, this is the 6.1299 MeV 3^- state, which decays promptly to the gs with the emission of a sharp characteristic γ-ray of approximately 6.129 MeV. There is then a gap until 6.1487 MeV α-energy when the 6.9171 MeV 2^+ state opens up. In practice the observation of 6.129 MeV γ-radiation provides an indication of carbon present as an impurity on special nuclear materials. For the reasons outlined between 5.12 MeV and 6.13 MeV α-energy the 6.129 MeV γ-production cross section maps out the (α, n_2) reaction uniquely and is also valuable for determining partial neutron-production cross section [1]

The measurements reported here were made as part of a feasibility study for a later program of measurements which was never done. However, we have not seen a similar study, and so even though preliminary in nature we feel there is still value in outlining our findings. γ-ray yield measurements, Y_γ, were made at 5.6, 5.8 and 6.0 MeV.

THE EXPERIMENTAL SETUP

The experiment was performed at the UKAEA Harwell Tandem Van de Graaff generator. The ion-source at the top injects singly charged negative ions into the accelerator tube. They get accelerated in the upper section of the machine towards the positively charged terminal (adjustable up to a maximum of about 6 MV) where they undergo charge exchange and become positive ions and are then accelerated a second time, but by repulsion away from the terminal, in the lower section of the machine. The beam is extracted via an analyzing magnet, slit and lens system towards the target.

The target consisted of a 3 mm thick Cu disc with a central recess 10 mm in diameter and 2 mm deep packed with carbon powder. The disc was mounted at the end of a 500 mm long Ta Faraday cup attached to the Harwell Tandem accelerator. The target was bombarded by α-particles and γ-rays were detected at 55-degrees with respect to the incident beam in a 101 cm^3 Princeton Gamma-Technology's (PGT) n-type (i.e. donor impurity doped) coaxial high-purity Ge detector with a relative efficiency of about 25%. Forced air cooling was applied to the back of the disc throughout the irradiations. Beam currents were limited to the nano-Ampere (nA) range to preserve the target. Short exposures were used as a precaution to minimize neutron damage to the Ge detector and to preserve the fragile target. Approximately 1200 net counts were accumulated in the full energy peak.

The target was prepared by the Actinide Chemistry Group at UKAEA, Harwell. The amorphous carbon target material was enriched in ^{13}C to enhance the signal and was manufactured by blending a remnant of (20.1±0.1) mg of ^{13}C powder (99 at% with an assumed uncertainty of ±0.3 at%) with (176.2±0.1) mg of ^{nat}C powder (enrichment not checked) in a small hard-steel ball-mill. A portion (about 160 mg) of the powder recovered was pressed into the recess at 155 MPa to create a pellet thick compared to the range α-particles used. After pressing the disc was fired under dry nitrogen in a furnace we believe in order to stabilize the product. The target is thicker than the range of the incident α-particles and to the exact density and thickness of the target do not need to be known in the yield analysis.

The effective average enrichment relative to ^{nat}C is calculated as follows:

$$F = \left(\frac{e}{e_0}\right) = \frac{\frac{m_0}{e_0 A_{13} + (1-e_0)A_{12}} + \frac{m_1}{e_1 A_{13} + (1-e_1)A_{12}} \cdot \frac{e_1}{e_0}}{\frac{m_0}{e_0 A_{13} + (1-e_0)A_{12}} + \frac{m_1}{e_1 A_{13} + (1-e_1)A_{12}}}$$

where

$F = \left(\frac{e}{e_0}\right)$ is the enhancement factor in the signal achieved by using the carbon blend rather than carbon of natural isotopic abundance (the letter 'e' denoting 'enrichment' as will be made clear below)

m_0 is the mass of ^{nat}C added to the ball-mill

m_1 is the mass of enriched C feed added to the ball-mill

e_0 and e_1 are the ^{13}C atomic fractions in the natural and enriched carbon feed stocks, respectively,

and,

A_{12} and A_{13} are the molar masses of ^{12}C and ^{13}C, respectively.

The isotopic abundances of ^{13}C in natural terrestrial materials is known to exhibit significant variation [2]. Taking $e_0 = (1.09 \pm 0.03)\ at\%$ for carbon we obtain $F = \left(\frac{e}{e_0}\right) = (10.430 \pm 0.057)$.

The order of the experiments was 5.8, 6.0 and 5.6 MeV. No repetitions, repeats or other verification measurements which would ordinarily be performed were performed in this scoping study which had a modest goal of 50 % accuracy. However, visual inspection of the target after the experiment revealed nothing noteworthy.

The Brookhaven Instruments Corporation [bicorp.com] current integrator was nominally accurate to about 0.5 % but leakage currents limited the present results to about 2% rsd (relative standard deviation).

The α-particle beam spot was about 6 mm across and was centered in the target by viewing a quartz disc which replaced the target. Beam energy measurements were accurate to 0.003 MeV (3 keV) and were made by Rutherford backscattering off a thin Au foil relative to ^{244}Cm (5.8048 MeV). Although routine [3], set-up and calibration was tedious, and this work benefited greatly by piggy-backing on an independent experiment.

The full energy peak (FEP) efficiency of the detector was established at 250 mm from the end cap using a range of point-line sources (^{56}Co, ^{152}Eu, ^{137}Cs and ^{60}Co). The overall uncertainty for the 1.3325 MeV line from ^{60}Co was about 0.8%. The relative efficiency was extended above 10 MeV using a variety of thermal neutron capture γ-spectra (principally Cl, N, Ti) generated at the Badger facility off the UKAEA Harwell DIDO reactor [4, 5]. Polynomial fitting weighted by the inverse of the independent variances was used to estimate the 6129 MeV to 1.3325 MeV efficiency ratio with an uncertainty of about 4.5 %. The absolute value was 5.8826x10^{-5} count/γ. The single and double escape peak (SEP & DEP) efficiencies are of similar magnitude measured to be 6.2488x10^{-5} count/γ and 5.1940x10^{-5} count/γ, respectively. In practice the FEP, SEP, and DEP offers three statistically independent estimates of the γ-ray yield. Net areas were determined using a simple three region of interest algorithm which also generated defensible counting uncertainties. We took the statistically weighted mean of the yield estimates based on the FEP, SEP and DEP and assigned the Poisson internal rsd as the statistical uncertainty. So, for

example, at 5.8 MeV we have net counts of (1178±36), (1267±39), and (1226±38) in the FEP, SEP and DEP, respectively, which converted to yield and combined as described results in a precision of about 1.86 %.

The efficiency at 6.129 MeV as a function of source to end cap separation was determined experimentally using a physically small sealed ^{238}Pu/^{13}C source. The behavior over the interval 50 to 400 mm was well represented by the functional form:

$$\varepsilon(r) = \frac{\varepsilon_o}{(r+d)^2}$$

where d represents the effective depth of an equivalent point detector behind the end cap and was estimated to have a value of (33.90±0.88) mm. This was the way the absolute efficiency measured at 250 mm was scaled to the current location of (196±2) mm.

RESULTS

We assume that the γ-ray yield, Yγ, observed at 55 degrees in the laboratory frame is representative of the yield into 4π. Correction for attenuation in the Cu backing was allowed for based on a central ray approximation ('narrow beam' using NIST XCOM mass attenuation coefficients excluding elastic scattering which redirects rather than removes photons), and the observed result was corrected to ^{nat}C by dividing by the F-factor calculated for the C-blend. The uncertainty in the deadtime correction was taken to be about 10% of the correction factor estimated as the ratio of the real time to live time ratio reported by the multichannel analyzer. The γ-ray yields measured are reported in Table 1 as function of the kinetic energy of the α-particles. The total measurement uncertainty (TMU) of each of the three yields was evaluated to be about 6%.

E_α (MeV)	**Y_γ (6.129 MeV γ / 10^8 α)**
5.597	1.0437
5.801	1.6207
5.999	3.3108

Table 1. Results of the present experiment. γ-ray yield (Yγ) as a function of the kinetic energy of α-particles stopping in ^{nat}C. The nominal overall relative standard deviation is about 6%.

The TMU was estimated by propagation of variance which rests on linearizing the measurement equation about the expectation value of each of the predictor variables. The measurement equation is simply the ratio of γ-rays emitted divided by the number of α-particles incident and adjusted to natural atomic abundance. Algebraically this can be expressed as follows:

$$Y_\gamma = \frac{1}{F} \cdot \frac{\langle C_{net}/\varepsilon \rangle / f_{atten}}{N_\alpha}$$

where $\langle C_{net}/\varepsilon \rangle$ denotes the weighted average over the FEP, SEP and DEP results, as discussed in the text, for the number of γ-rays emitted from a transparent source based on the ratio of the number of net peak counts observed divided by the corresponding free-air efficiency; f_{atten} is the attenuation factor through the back of the target at 55 degree slant angle; N_α is the number of α-particles on target; and F is the isotopic abundance factor discussed in the text.

For illustration the TMU at 5.8 MeV is estimated to be about 5.7 % based on the quadrature sum of contributions listed in Table 2.

Contributor	**Relative Standard Deviation (%)**
Counting precision	1.86
Current integration	2
Absolute efficiency normalization at 1.3325 MeV	0.8
Extrapolation to 6.129 MeV	4.5
Adjustment to (196±2) mm	1.74
Uncertainty in the effective depth	0.15
Dead time correction	0.5
Attenuation correction (0.9527±0.0014)	0.15
Enrichment correction factor, F	0.55

Table 2. Uncertainty budget for the thick target $^{13}C(\alpha,n_2)^{16}O$+6.129 MeV γ-yield in ^{nat}C at 5.8 MeV.

DISCUSSION and CONCLUSIONS

Thick target γ-yield measurements from C are complementary to high-resolution thin-target energy-dependent cross section measurements. By offering a stronger signal and lower susceptibility to certain uncertainties they can help normalize, or constrain, thin-target shape data during cross-section evaluation. We report results from a feasibility experiment performed in 1990 and provide an indicative uncertainty analysis. These can be used in planning future experiments. The potential exists for several

improvements, for example the uncertainties in both the current integration and counting precision could both readily be reduced by a factor of four. Such improvements are advisable to reduce random point to point variability. However, the overall uncertainty is dominated by the uncertainty in the absolute γ-ray detection efficiency. There is some scope for improvement and best modern practices and reference data should be used in future work to ensure that future experiments generate results with the greatest long-lasting impact. An array of detectors should be used to confirm the angular distribution expeditiously. Attention to the preparation of well characterized and stable targets is a pre-requisite to success and monitor reactions should be added to check all aspects of the data collection and analysis.

The present work provides data at just three, widely spaced energies that are well above threshold. We offer no guidance on how to interpolate between points because we expect there to be structure dependent on the underlying resonant nature of the underlying thin-target reaction cross section. It is for future measurements to map out the fine structure.

The data will be provided to the EXFOR data base.

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