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Original Article

Neutron yield and energy spectrum of ^{13}C(alpha,n)^{16}O reaction in liquid scintillator of KamLAND: A Nedis-2m simulation

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ABSTRACT

The ^{13}C (α,n)^{16}O reaction cross-section is important data for nuclear physics, astrophysical, and neutrino physics experiments, however, they exhibit uncertainties due to the discrepancies in the experimental data. In this study, using the Nedis-2m program code, the energy spectrum of α-induced neutrons in a thin carbon target was calculated and the corresponding reaction cross-section was refined in the alpha particle energy range of 5–8 MeV. The results were used to calculate the intensity and energy spectrum of background neutrons produced in the liquid scintillator of KamLAND. The results will be useful in a variety of astrophysical and neutrino experiments especially those based on LS or Gd-LS detectors.

1. Introduction

The ^{13}C (αn)^{16}O reaction has found less attention compared to ^{9}Be (α,n)^{12}C in radioisotopic neutron source production, because the ^{13}C content in carbon is ~1%, and the reaction cross-section is also about 5 times less than that of ^{9}Be (α,n)^{12}C at 5 MeV alpha particle energy. Nevertheless, ^{13}C (α,n)^{16}O is very often used as a source of hard gamma-rays (^{13}C (α,n$_2\gamma$)^{16}O, $E_\gamma = 6.129$ MeV) for spectroscopy calibrations, through the de-excitation of the second excited level of ^{16}O [1–3], and also as a source of tagged neutrons in background measurements using scintillation detectors [5,6].

Recently, the need for a more accurate estimation of the background neutrons produced in the liquid scintillator of KamLAND facility has increased the interest in ^{13}C (α,n)^{16}O [4,5,7–9].

The cross-section library for the ^{13}C (α,n) reactions available in EXFOR (Experimental Nuclear Reaction Data) [10] confirms that the latest experimental data dates back to 2005 through the experimental studies of Harissopulos et al. [7] with an accuracy of ~4% (1σ) for alpha energies ranging from 0.8 to 8 MeV with 10 keV steps. The measurements were carried out with an alpha particle accelerator at the Ruhr University (Germany) using a 22 μg/cm^2 thick carbon target, 99% enriched in ^{13}C. The neutron yield was measured using a 4π detection setup in which 16 helium proportional counters were responsible for thermal neutron detections. The detectors were placed in a 1-m long, 35-cm diameter cylindrical polyethylene moderator with a 7-cm diameter central hole to accommodate the target.

In this study, the neutron detection efficiency was calculated using the Monte Carlo code MCNP for different neutron energies (E_n) before approximated as $\eta(E_n)$ [7] which is further used to convert the measured neutron yield $Y(E_n)$ into ^{13}C (α,n)^{16}O reaction cross-section. A comparison was carried out on the neutron detection efficiency calculated for a standard calibration ^{252}Cf source (±1%) with average neutron energy of 2.3 MeV, showing a promising agreement between the calculation (31.6%) and the measurement (32.1 ± 0.5%). Following the approach used in Harissopulos et al. studies, the neutron energy for $\eta(E_n)$ calculations was determined for the ground level of the residual nucleus ^{16}O (0^+, $j = 0$), which means the contribution of neutrons originating from the excited states of ^{16}O was neglected, and as a result, the total absolute cross-section data of ^{13}C (α,n)^{16}O reaction for the alpha particle energies near the threshold of 5.0136 MeV and above were overestimated by a factor of ~1.4. Peters [9] and Mohr [8] discussed different aspects of these results and emphasized that the contribution of neutrons from the excited states of the residual

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^{16}O nucleus must be taken into account for precise cross-section data.

As far as the literature review shows the measurements carried out by Harissopulos et al. despite some discrepancies are the only publicly available and reliable experimental database for analyzing the ^{13}C (α,n)^{16}O reaction cross-section. The measurement results satisfactorily agree with the experimental data set obtained by West and Sherwood [11] when they are correctly normalized. Therefore, one may obtain invaluable and accurate data for solving problems of modern nuclear physics and astrophysics by correcting the computational part of the methodology used by Harissopulos et al.

2. Materials and methods

The calculated values of the neutron yield and energy spectrum for the ground and all known excited states of the residual nucleus ^{16}O of ^{13}C (α,n) reaction are presented in this section. The results were obtained using the Nedis-2m code based on the cross-section estimates made by Vlaskin et al. [12] and other researchers in this field. The calculation results show that the relative discrepancies among different data remain less than ~5% (2σ).

The above data were used to calculate the intensity and energy spectrum of background neutrons produced in the liquid scintillator of KamLAND. The calculated energy spectrum in which the anisotropy of neutron emission is also incorporated shows a finer structure in the energy spectrum with a better agreement with measurement.

2.1. NEDIS-2m program code

Nedis-2m [12] is capable to calculate the production rate and continuous energy spectra of neutrons generated via (α,n) reaction on Li, Be, B, C, O, F, Ne, Na Mg, Al, Si, P, S, Cl, Ar, and K. It takes into consideration the anisotropic angular distribution of neutrons of (α,n) reaction in the center-of-mass system within the alpha-emitting source material. It also calculates the spontaneous fission spectra with an evaluated half-life, spontaneous fission branching, ν-averaged per fission, and Watt spectrum parameters. The Nedis-2m results can be used as input to other Monte Carlo codes for full-simulation purposes. Nedis-2m is very successful in reproducing the spectrum structures mainly because of its evaluated cross-sections for various excited states of the residual nuclei, as well as the anisotropic angular emission in the center-of-mass system, in particular for ^{7}Li, ^{9}Be, and ^{13}C. The most successful application of the Nedis-2m code has been presented by different authors in the literature [13–16].

The Nedis-2m code uses the following equation for the calculation of the energy spectrum of neutrons produced for every energy level of the residual ^{16}O nucleus of ^{13}C (α,n) reaction:

$$\frac{dN}{dE}(E_n)=\int_{X(E_n,\cos(\vartheta_1))}^{Y(E_n,\cos(\vartheta_2))}\frac{\sigma(E_\alpha,\cos(\vartheta^*))}{\varepsilon(E_\alpha)}\left(\frac{\partial\Omega^*}{\partial E_n}\right)dE_\alpha, \tag{1}$$

where $\varepsilon(E_\alpha)$ is the atomic stopping power of the medium for alpha particles; E_α is the alpha particle energy in the laboratory coordinate system; E_n is the neutron energy in the laboratory coordinate system; $\sigma(E_\alpha \cos(\vartheta^*))$ is the differential cross-section of (α,n) reaction; ϑ* is the neutron emission angle in the center-of-mass system; the limits of integration, $X(E_n,\cos(\vartheta_1))$ and $Y(E_n,\cos(\vartheta_2))$ normally determined by the target thickness and the neutron emission angle in the laboratory coordinate system, are derived from the kinematics consideration of the (α,n) reaction of interest.

In Nedis-2m, the reaction cross-section, $\sigma(E_\alpha,\cos(\vartheta^*))$, in the center-of-mass system for a separate ^{16}O level as an expansion in Legendre polynomials is calculated through the following relation:

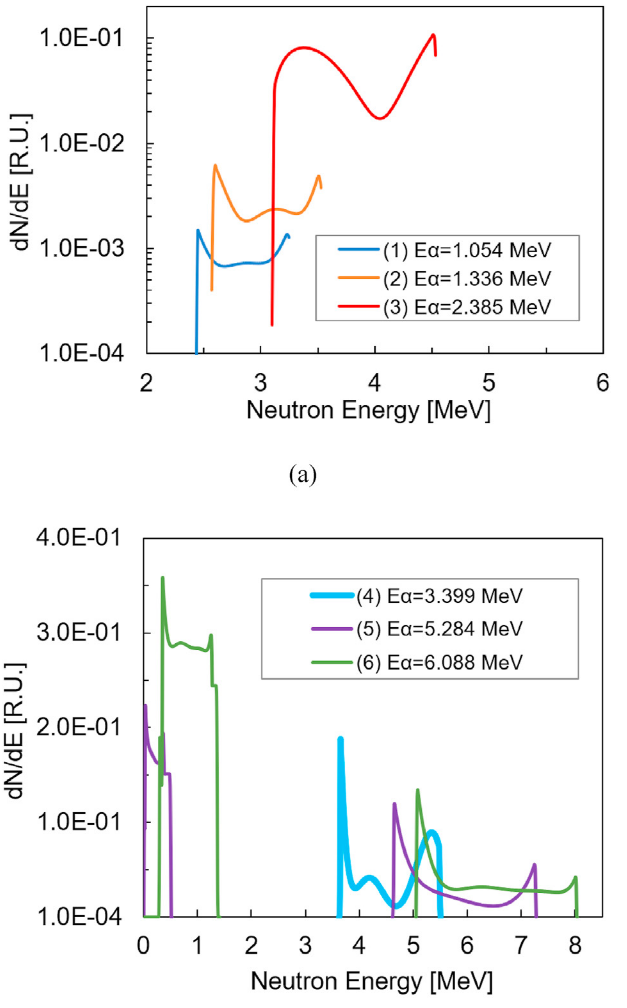
$$\sigma(E_\alpha,\cos(\vartheta^*))=\frac{\sigma_{tot}(E_\alpha)}{4\pi}\left[1+\sum_{\ell}a_\ell(E_\alpha)P_\ell(\cos(\vartheta^*))\right], \tag{2}$$

where a_ℓ coefficients are found through linear interpolations. The values of $\cos(\vartheta^*)$ are uniquely determined by the energies E_n and E_α.

The transition from the solid angle in the center-of-mass system to the neutron energy in the laboratory coordinate system is performed via the following equation:

$$I(E_\alpha)=\frac{\partial\Omega^*}{\partial E_n}=2\pi\frac{d(\cos(\vartheta^*))}{dE_n}=\frac{4\pi}{R(E_\alpha)}=\frac{4\pi}{E_n(0^\circ,E_\alpha)-E_n(180^\circ,E_\alpha)}, \tag{3}$$

where function $R(E_\alpha)=\frac{4m_n m_\alpha E_\alpha}{(m_n+m_r)^2\gamma(E_\alpha)}$ is the energy interval $(E_{n,max}-E_{n,min})$, in which the neutrons are produced for a given value of the alpha particle energy, and function $\gamma(E_\alpha)$, for the



(a)

(b)

Fig. 1. Energy spectrum of neutrons produced in ^{13}C (α,n)^{16}O reaction on a 22 μg/cm^2 target.

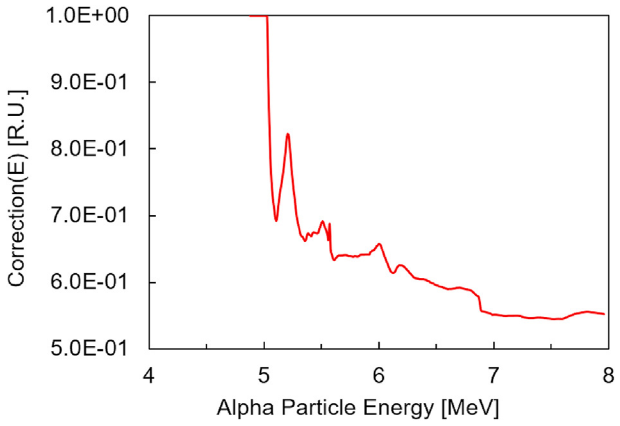


Fig. 2. Detection efficiency ratio $(\eta_{90}/\dot{\eta}(E_{\alpha}))$.

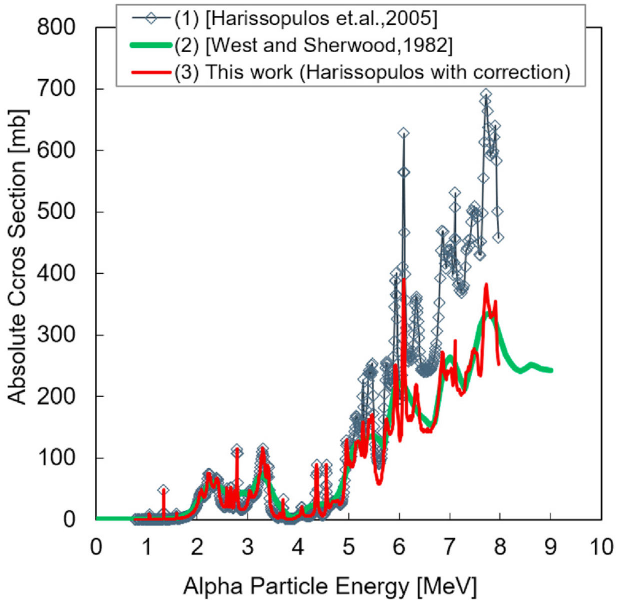


Fig. 3. Absolute total cross-section for $^{13}C\ (\alpha,n)^{16}O$ reaction.

reaction energy Q, is determined by the expression $\gamma(E_{\alpha}) = \left[\frac{m_n m_{\alpha} E_{\alpha}}{m_r(m_r+m_n)Q+m_r(m_r+m_n-m_{\alpha})E_{\alpha}}\right]^{1/2}$; m_r is the mass of the residual nucleus; m_n is the mass of the alpha particle; m_n is the neutron mass.

The values of the detection efficiency $\dot{\eta}(E_{\alpha i})$ of the energy spectrum for the neutrons emitted when a thin carbon target is irradiated with alpha particles of energy $E_{\alpha i}$ were found by the following equation:

$$\dot{\eta}(E_{\alpha i}) = \frac{\int_{E_{min}}^{E_{max}} \eta(E_n)\frac{dN}{dE}(E_n)dE}{\int_{E_{min}}^{E_{max}} \frac{dN}{dE}(E_n)dE}, \tag{4}$$

where $\eta(E_n)$ is the calculated neutron detection efficiency taken from Ref. [7] for the monoenergetic neutrons with energy E_n; E_{min} and E_{max} are the lower and upper limits of the neutron spectrum, respectively.

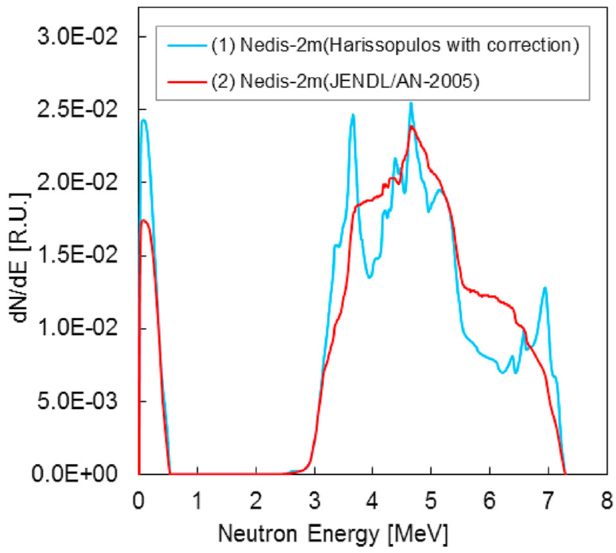


Fig. 4. Energy spectrum of neutrons produced in $^{13}C\ (\alpha,n)\ ^{16}O$ reactions in the liquid scintillator of KamLAND: (1) calculation using the Nedis-2m program based on the estimates of the cross-sections made by authors in the present work and (2) the cross-sections estimated in Ref. [17].

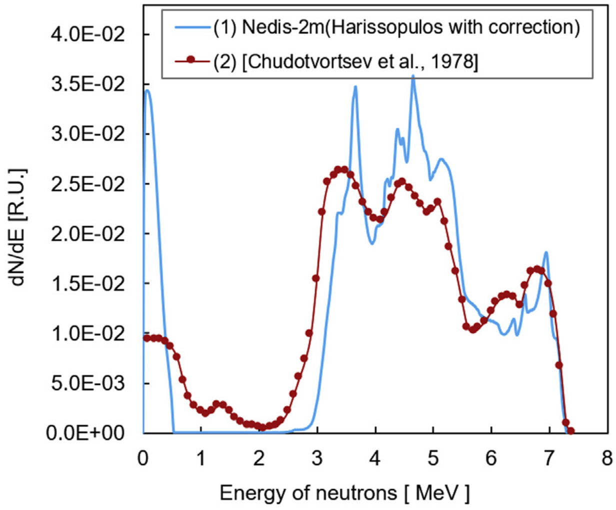


Fig. 5. Energy spectrum of neutrons of ^{210}Po-$^{13}C\ (\alpha,n)$-source.

2.2. Nedis-2m simulation of neutron energy spectrum and detection efficiency

To demonstrate the contribution of (α,n) neutrons originating from the excited levels and to show the generation of non-monoenergetic neutrons in a 22 $\mu g/cm^2$-thick target, a Nedis-2m calculation example is presented for individual alpha particle energies, $E_{\alpha i}$, on targets made of carbon, 99% enriched in ^{13}C.

In these energy spectrum calculations of the main energy levels for the residual ^{16}O nucleus at alpha particle energies above the threshold of 5.0136 MeV, σ_{tot} cross-section was taken from Ref. [17] obtained using the detailed equilibrium method from the cross-section of the reverse reaction $^{16}O\ (n,\alpha)^{13}C$. The cross-sections for the first and second excited ^{16}O levels were estimated by Vlaskin based on integral energy spectrum measurements of ^{227}Ac-^{13}C, ^{244}Cm-^{13}C, and ^{210}Po-^{13}C neutron sources and the photon yields

Table 1
Characteristics of (α,n) reaction on a thick CH1.87 target of KamLAND scintillator ($E_\alpha = 5.304$ MeV).

Energy levels of the residual nucleus ^{16}O [MeV]	Decay mode [4]	$E_{threshold}$ [MeV]	$E_{n,min}$ [MeV]	$E_{n,max}$ [MeV]	Neutron yield per 10^6 alpha particles with $E_\alpha = 5.304$ MeV	
					Nedis-2m (Harissopulos with correction)	Nedis-2m (JENDL/AN-2005)
0	–	0	2.0437	7.295	5.753E-2	6.040E-2
6.049	$e^- + e^+$	5.0136	0	0.5317	6.970E-3	5.223E-3
6.130	γ	5.1195	0	0.4050	5.445E-4	7.734E-4
6.919	–	6.1514	–	–		–
7.117	–	6.4104	–	–		–

with an energy of 6.13 MeV [1–3,18]. The cross-sections for the third and fourth excited levels were obtained from Ref. [17] based on the breakup probability calculation of the compound nucleus into the corresponding levels with a neutron emission.

The data on the angular distributions of (α,n) neutrons in the center-of-mass system were taken from Refs. [19,20], whilst the atomic stopping power data of alpha particles, in units of 10^{-15} eV $cm^2 \cdot atom^{-1}$, were obtained using the equations taken from Ref. [21].

The selected values of $E_{\alpha i}$ (i.e., the energy spectrum of the produced α-n neutrons) shown in Fig. 1 were calculated by taking into account the contribution of all known excited states of ^{16}O and associated anisotropy. These spectra contain the peaks of different neutron energies whose characterization, following the work of Harissopulos et al. is impossible by the same average energy value when the neutrons escape from the target at an angle of 90° with respect to the beam of incident alpha particles. This approach requires preliminary verification in some cases. Therefore, in a further study, the neutron energy spectra and the detection efficiency for each $E_{\alpha i}$ value taken from Ref. [7] were calculated whose simulation results are shown in Figs. 2 and 3.

Fig. 2 shows the variation of the neutron detection efficiency obtained for neutrons emitted at an angle of 90° (η_{90}) to the detection efficiency obtained in this work ($\acute{\eta}(E_\alpha)$) as a function of E_α. The ratio, $correction(E) = \eta_{90}/\acute{\eta}(E_\alpha) = \sigma_{tot,\ this\ work}(E_\alpha)/\sigma_{tot,[Harissopulos\ etal.,2005]}(E_\alpha)$, was used to correct the values of the total absolute cross-section $\sigma_{(tot,\ this\ work)}$ of ^{13}C (α, n) ^{16}O reaction. The results of this adjustment are shown in Fig. 3 (see curve (3)).

For a comparison, $\sigma_{tot,[West\ and\ Sherwood,1982]}(E_\alpha)$ was also added as shown in Fig. 3, represented by curve (2), which was obtained by differentiating the data of the total neutron yield on a thick carbon target of natural composition from Ref. [11]. The comparison results of curves (2) and (3) show a promising agreement.

In neutron spectrum calculations for the liquid scintillator of KamLAND facility, the corrected total absolute cross-section is illustrated in Fig. 3, where the data of curve (3) was normalized (multiplied by 1.13) by the experimental value of the neutron yield at E_α equal to 5.304 MeV from Ref. [11]. All partial cross-sections were normalized accordingly.

Here it is assumed that the liquid scintillator of the setup is a thick $CH_{1.87}$ hydrocarbon target with a density of ~0.78 g/cm^3 [22], and the energy spectrum of neutrons was obtained for ^{210}Po alpha particles with an energy of 5.304 MeV.

3. Results and discussion

The calculation results of spectral and integral characteristics of ^{13}C $(\alpha,n)^{16}O$ reaction in the liquid scintillator of KamLAND are shown in Figs. 4 and 5, as well as in Table 1, whilst Fig. 4 compares the calculated spectrum based on the cross-sections estimated by Murata et al. [16], assuming an isotropic neutron emission.

The calculation results (see Figs. 4 and 5, curve (1)) showed that the neutron spectrum corresponding to the ground level of ^{16}O extends from 2.044 MeV to 7.3 MeV and has three characteristic peaks of ^{210}Po-^{13}C (α,n) source (the characteristic peaks are shown in Fig. 5, see curve (2) [23]): i.e., 3.66, 4.66 and 6.94 MeV. For the isotropic case of neutron emission (see Line 2 in Fig. 4), only one peak is observed at $E_n = 4.66$ MeV. The neutrons associated with the excited states of ^{16}O lie in the energy range of 0–0.532 MeV.

Note that the calculated energy spectrum of neutrons generated in ^{13}C $(\alpha,n)^{16}O$ reaction, taking into account the anisotropy of neutron emission and the contribution of excited ^{16}O states, shows a finer structure of the neutron energy distribution which is generally in agreement with measurement (see Fig. 5). The experiment gives the idea behind the energy spectrum of neutrons leaving the ^{210}Po-^{13}C (α,n)-source containing ^{210}Po and ^{13}C, whilst the calculation is responsible for the energy spectrum of neutrons produced in the reaction ^{13}C $(\alpha,n)^{16}O$.

Table 1 summarizes the calculated values of neutron yield for the ground and excited levels of the residual nucleus ^{16}O obtained using the Nedis-2m code based on the cross-section estimates made by Vlaskin in this and other works and also the cross-sections estimated in Ref. [17]. The results of the present calculations agree with each other with errors not exceeding ~5% (2σ) and are recommended by authors as an update of the old reference tables used in nuclear physics experiments.

4. Conclusions

In this work, using the Nedis-2m program code, the energy spectrum of neutrons produced within a carbon target with a thickness of 22 μg/cm^2 (see Figs. 2 and 3), taking into account the contribution of all known excited states of the residual nucleus, and corrected ^{13}C $(\alpha,n)^{16}O$ reactions in the energy range of alpha particles ranging from 5.0136 to 8.00 MeV.

The data obtained in this study were used to calculate the intensity and energy spectrum of background neutrons produced in the liquid scintillator of KamLAND (see Fig. 4). The calculated spectrum shows a finer structure of the neutron distribution and, in general, is in good agreement with the measurement (see Fig. 5). As shown in Fig. 5, the discrepancy between the measurement and calculation is since the measurement is responsible for the energy spectrum of neutrons leaving the ^{210}Po-^{13}C (α,n)-source containing ^{210}Po and carbon, and the calculation gives the energy spectrum of neutrons produced in the reaction ^{13}C $(\alpha,n)^{16}O$.

The results of the present study are important in astrophysical and neutrino experiments based on LS or Gd-LS detectors and are recommended by authors as an update of the old reference tables used in nuclear physics experiments.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

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