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Experimental studies on the neutron emission spectrum and induced radioactivity of the ⁷Li(d,n) reaction in the 20–40 MeV region

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Abstract

To improve the data accuracy of the neutron emission spectrum of the ⁷Li(d,n) reaction and the radioactivity (⁷Be, ³H, etc.) accumulated in the ⁷Li target in IFMIF, we have measured the neutron emission spectrum and the radioactivity of ⁷Be induced in the lithium target for 25 MeV deuterons at the Tohoku University AVF cyclotron (K = 110) facility. Neutron spectra were measured with the time-of-flight (TOF) method at four laboratory angles by using a beam swinger system and a well collimated TOF channel. Induced radioactivity was measured by detecting the gamma-rays from ⁷Be with a pure Ge detector. Experimental results are compared with other experimental data. The present result of neutron emission spectra are in qualitative agreement with other experimental data but that of ⁷Be production was much larger than expected by the recent codes. Measurement will be extended to several incident energies up to 40 MeV. © 2002 Elsevier Science B.V. All rights reserved.

1. Introduction

For the design and operation of the neutron production target of IFMIF, detailed knowledge is required on the energy-angular neutron emission spectrum of the ⁷Li(d,n) reaction, and the radioactivity (⁷Be, ³H, etc.) accumulated in the target [1]. The neutron flux and spectral data are indispensable for precise estimation of the neutron irradiation effects, and the radioactivity accumulation is of great concern for the management of the target. Several works were undertaken on these subjects in the course of the FMIT project, but the data status is not good enough as shown by marked differences among experimental data [2–6].

To improve the data accuracy of the neutron energyangular distribution and the predictability of the radioactivity accumulation in the IFMIF ⁷Li target, we have

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started experiments on the neutron emission spectrum of the ⁷Li(d,n) reaction and the radioactivity induced in the target using the AVF cyclotron (K = 110) at the Tohoku University Cyclotron and Radioisotope Center (CY-RIC) [7].

This paper presents the experiments on the (1) neutron emission spectra and the (2) production of radioactive nuclide, ⁷Be, of the Li(d,n) reaction for 25 MeV deuterons. The neutron spectra were measured with the time-of-flight (TOF) method at four laboratory angles between 4° and 40° using a beam swinger system and a well collimated TOF channel. The amount of ⁷Be accumulated in the lithium target was measured by counting the gamma-rays from ⁷Be using a pure Ge detector.

Experimental results are compared with other experimental data.

2. Experimental apparatus

The layout of the target room in CYRIC is presented in Fig. 1. A deuteron beam accelerated by the AVF cyclotron was transported to the target room no. 5 which is equipped with a beam-swinger system and a neutron flight channel as shown in Fig. 1. The beam swinger system changes the incident angle of the beam onto the target from -5° to 145° and enables angular distribution measurement without modifying the detector setup. The neutron flight channel is 44 m-long.

The lithium target was a metallic plate of elemental lithium. Care was taken on the chemical purity of the lithium target: the target was prepared by mechanical pressing of a lithium ingot under argon atmosphere to avoid oxygen contaminant and the lithium ingot was cleaned up thoroughly prior to the pressing to avoid carbon contamination. The lithium target was 7.5 mm thick to stop the incident beam. The size of the target was about $2 \text{ cm} \times 4 \text{ cm}$ which is larger sufficiently than the deuteron beam spot, about 5 mm in diameter. It was set on a remotely-controllable target changer without a backing together with a beam viewer of aluminum oxide. The support frame was isolated from the ground to read a beam charge induced on the target.

The target chamber was shielded with 1 m thick concrete walls having a beam channel for collimators to reduce neutron and gamma-ray backgrounds. Iron collimators, 10 cm-diameter, were inserted into the beam channel to collimate neutrons from the target.

Emitted neutrons were detected by a NE213 scintillation detector, 14 cm-diameter \times 10 cm-thick or 2 in.-diameter \times 2 in.-thick equipped with pulse-shapediscrimination (PSD). The detector was placed around 11 and 3.5 m from the target, respectively, (Fig. 1). The shorter flight path was adopted to measure the low energy part of the neutron spectrum because the RF frequency was as high as about 15 MHz, and the beamchopper for frequency reduction was not available at the time of the experiment. However, this high repetition rate of the beam caused frame-overlap and limited the spectrum measurements above around 9 MeV or so.



Fig. 1. The layout of TR 5 at CYRIC.

The TOF, PSD and pulse-height data were collected event by event as three parameter list data for off-line analysis [8].

3. Experimental procedure

Typical pulse width was 1-1.5 ns in FWHM, and the beam current on the target was around 5 nA. The beam current was digitized and recorded by a multi-channel scaler for normalization of the neutron TOF spectrum and the ⁷Be production measurement. The TOF data were obtained at 4°, 20°, 30° and 40°.

The activity of ⁷Be accumulated in the lithium target was measured by detecting 477 keV gamma-rays due to the decay of ⁷Be. The lithium target bombarded by a deuteron beam during the neutron spectrum measurement was measured with a pure Ge detector (Euricis Mesurese GPC50 -195-R) and a multi-channel analyzer.

4. Data analysis

4.1. Neutron spectrum

Neutron TOF spectra gated by a PSD signal and lower pulse-height bias were converted into energy spectra on the basis of the following equations:

$$E_n(i) = mc^2 - m_0 c^2 = m_0 c^2 \left(\frac{1}{\sqrt{1-\beta^2}} - 1\right),$$

$$\beta = \frac{v_n(x)}{c} = \frac{L}{cT_n(i)},$$

where m_0 is a rest mass of a neutron, *i* is the channel number of the event, *L* is a flight path and *c* is a light velocity.

As noted above, the high repetition rate of beam caused frame-overlap of the neutron TOF spectrum in which the low energy parts of neutron spectra were masked with high energy neutrons in the succeeding pulse. We attempted to eliminate the effect by setting multistage biases to the two-dimensional data for TOF vs pulse-height. This procedure was effective but not sufficient to deduce the spectrum down to several MeV, and the low limit of the spectrum was around 9 MeV.

Then, the spectra were divided by the detector efficiency and the solid angle to convert into differential yields. The efficiency vs energy curves of detectors were calculated by a revised version of the Monte Carlo code SCINFUL [9] that was verified to be accurate with in $\pm 5\%$ up to 80 MeV by Meigo [10]. The spectra were normalized by the integrated beam current.

4.2. ⁷Be activity

The induced ⁷Be activity was determined from the gamma-ray counts by the pure Ge detector, and the decay constant. The efficiency of the Ge detector was determined by the calculation using the Monte Carlo code EGS 4 [11]. The calculated results were confirmed at several energy points with standard gamma-ray sources.

5. Results and discussion

5.1. Li(d,n) neutron spectra

The present results for the Li(d,n) neutron spectrum at four laboratory angles are shown in Fig. 2. The data are given as the neutron spectrum per unit solid angle and Coulomb. In the present data, the low limit is as high as around 9 MeV because of the frame-overlap effects. The error bars of the spectra represent the statistical errors alone. It should be noted that the absolute



Fig. 2. The neutron yields of the Li(d,n) reaction at 4° , 10° , 20° and 40° for incident deuteron energy of 25 MeV.



Fig. 3. Comparison of the present Li(d,n) neutron spectrum with the data by Lone et al. [3].

value includes uncertainty due to secondary electron emission in the target.

In Fig. 3, the present data at 4° are compared with the experimental data by Lone et al. at 23 MeV [3]. The data of Lone et al. are reported only for 0° .

The experimental spectra are divided into two parts; high energy tail region probably due to direct stripping reactions and a main peak region centered around 10 MeV which is expected as a main neutron source in IFMIF. The present data are in good agreement with those by Lone in the 10–23 MeV energy region while they are systematically higher than the latter in the high energy tail region. In this region, the data by Sugimoto [12] at 32 MeV are closer to the present one rather than those by Lone et al. It will be necessary to clarify the spectrum in this energy region because neutrons in the high energy tail will cause much more damage due to large helium production cross-sections and the higher energy of primary-knock-on atoms.

The intensity of the high-energy tail is maximum around 20° while total neutron yields are maximum at 4°. The former conclusion is in agreement with Sugimoto [12] and can be interpreted by the angular momentum effects in the ${}^{7}L(d,n_{0,1})^{8}Be$ reaction. Therefore, to reduce the influence of high-energy neutrons and obtain higher intensity irradiation field, neutrons to angles close to 0° will be preferable. It is important to measure the lower energy part of the neutron spectrum for the ${}^{7}Li(d,n)$ reaction to assess the applicability of calculation codes and models for the neutron emission spectrum of the reaction [13].

5.2. ⁷Be activity

The measured ⁷Be activity was compared with other experimental data and calculation by the code IRAC [14] used for activity assessment of accelerator components. Fig. 4 shows the comparison of the present results, other



Fig. 4. ⁷Be production rate via the Li(d,n) reaction.



Fig. 5. ⁷Li(d,2n)⁷ Be reaction cross-section.

experimental data [15,16] and the calculation using the IRAC code. These values are for thick targets. Above 15 MeV, experimental data are very few but the present results at 25 MeV look consistent with the data around 22 MeV [15] and at 40 MeV [16], but are substantially higher than the IRAC calculation. The production of ⁷Be via the (d,n) reaction on elemental lithium will be due to the ⁷Li(d,2n) and the ⁶Li(d,n) reactions mainly. Fig. 5 shows the comparison of the experimental data and IRAC calculation for the 7Li(d,2n) reaction cross-section. It is apparent that the IRAC code underestimates the 7 Li(d,2n) cross-sections and this maybe a principal reason of the underestimation of the IRAC code for the ⁷Be production rate through the (d,n) reaction of elemental lithium. Similar underestimation is reported even for the new code [16]. The experiment will be extended to higher deuteron energy to obtain benchmark data for activity accumulation and to ³H production using the liquid scintillation technique or equivalent.

6. Summary

The present paper described the measurement of (1) neutron energy-angular distributions and (2) the ⁷Be production rate via the (d,n) reaction on thick elemental lithium at 25 MeV deuteron energy performed using the K = 110 AVF cyclotron at Tohoku University CYRIC. In the spectrum measurement, the spectrum of high energy tail could be observed clearly although the low limit of spectrum was rather high because of a lack of a beam chopping system. The measurements will be extended to lower neutron energy and higher incident energies.

The experimental values of the ⁷Be production rate were much smaller than expected by the IRAC code probably due to underestimation of the ⁷Li(d,2n) reaction. This measurement will also be extended to higher incident energies.

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