Experimental Studies on Particle and Radionuclide Production Cross Sections for Tens of MeV Neutrons and Protons

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Abstract. Experimental studies are described for (1) differential thick-target neutron yields and double-differential neutron emission cross sections for (p,n) and (d,n) reactions, (2) fragment production reactions, and (3) radionuclide production for neutron- and proton-induced reactions in the tens of MeV region and upgrading of the 7Li(p,n) neutron source.

INTRODUCTION

Recent progress in accelerator utilization requires various nuclear data in intermediate energy region for the design of the accelerator system and radiation safety [1]. In particular, intense proton accelerators under construction and planned are very strong neutron sources and require detailed information on neutron production reactions of accelerated beam and on the effects of secondary neutrons such as radiation effects and activation of materials. The nuclear data in this energy region play a crucial role also in the space technology, i.e., shielding of space crafts and analysis of radiation effects to semiconductors. For the analysis of radiation effects, information on energy-angular distribution data are needed not only for light charged particles such as p, d, t, α but also for heavy charged particles, Li, Be, C, O and so on (fragments) that will have strong radiation effects due to their large LET (linear energy transfer). Experimental data for fragment production are very few because of experimental difficulty.

We have been conducting experiments on the following items in tens of MeV region using a K=110 MeV AVF cyclotron of Tohoku University and others, and a quasi mono-energetic neutron source with the 7Li(p,n) reaction:

1) differential thick target yields and double-differential neutron production cross sections (DDX) of (p,n) and (d,n) reactions for Ep=35, 50, 70 MeV and Ed=25 and 40 MeV using a beam swinger TOF spectrometer,

2) production cross sections of fragments for neutron- and proton-induced reactions using a Bragg curve spectrometer and the energy-TOF (E-TOF) technique for particle identification. We are developing an experimental technique for the purpose.

3) radio- nuclides production using the activation technique, and

4) upgrading of the 7Li(p,n) neutron source to increase the neutron production rate that is essential for study of neutron induced reactions.

EXPERIMENTAL FACILITY

Figure 1 shows the layout of Tohoku University AVF cyclotron facility [2,3]. The cyclotron accelerates protons up to 90 MeV, deuterons to 65 MeV and other heavy ions to ten’s of MeV/u. The cyclotron is equipped with 1) five experimental rooms and 2) a beam swinger magnet, 3) well-collimated neutron flight path up to 44 m, and 4) a beam chopping system for reduction of the beam frequency. These apparatuses enable neutron angular distribution measurement with high resolution TOF technique. In addition, the cyclotron is also equipped with 5) a quasi mono-energetic 7Li(p,n) neutron source, which used for experiments of neutrons-induced activation and semiconductor soft-error.
DIFFERENTIAL TTY AND DDX FOR (P,N) AND (D,N) REACTION [6-9]

The schematic view of the experimental apparatus for neutron production experiment is shown in Fig. 2. The incident angle of the beam is changed by a beam swinger magnet from -5 to 145 deg. Therefore, in this geometry, neutron angular distribution can be measured with a fixed neutron detector arrangement with tight collimation. In the case of TTY measurement, targets with full-stop thicknesses served as a Faraday. For the purpose, the target is surrounded with a copper mesh biased to -300 VDC for secondary electron suppression. For thin targets, the beam transmitted the target is bent to a beam dump made of full stop carbon acting as a Faraday cup. The beam dump is shielded from detectors with shadow bars made of iron and copper, and concrete walls. The neutron detectors were NE213 scintillator, 14 cm-diam by 10 cm thick or 5 cm-diam by 5 cm thick. Both detectors are equipped with n-γ discriminators. To cover almost full range of neutron spectrum, measurement was done in two different flight paths: 11.5 m and 3.5 m. The measurement with long flight path employed the larger neutron detector and pulse height bias of ~2 MeV proton, and the measurement with shorter path employed the smaller detectors and lower pulse height bias, ~0.6 MeV proton. The detector efficiency was obtained with calculation using the modified version of SCINFUL [4,5] whose efficiency was confirmed [5] to be valid within 5%. The data by high bias and low bias were in general agreement in overlapping region. The final data were derived by combining two data.

The data were corrected for backgrounds, detector efficiency, and attenuation in the target and air in the flight path. The attenuation correction was made using the LA-150 [10] data library or experimental total cross section data [11]. Errors were estimated considering the contributions from counting statistics, detector efficiencies and errors of cross sections data employed in the attenuation correction. Until now, measurements were done for C, Al, Ta, W, and Pb at 50 MeV [7] and 70 MeV, for Fe, Cu at 35, 50, and 70 MeV [8]. The present experiments provided angle-dependent neutron emission spectrum down to 0.8 MeV or so. The results were compared with other experiments if available and LA-150, which is the modern evaluation. The present (p,n) data of C and Cu for 70 MeV show good agreement with those by Meigo for 68 MeV [12].

Here typical data for C and W for 50 and 70 MeV are presented. Figure 3 illustrates the results of C, W(p,n) reactions in comparison with LA-150 library. LA-150 reproduces the experimental data generally well. In detail, however, the C data show marked underestimation of experiments in high energy region because of lacks of neutron emission through 13C(p,n) reaction and too soft neutron spectrum. For W, LA-150 similarly underestimates the high-energy portion of the spectrum. Such trend is commonly observed for other elements in both energies. To trace the reason of the discrepancy, we are promoting DDX measurements using thin targets.

For (d,n) reactions, measurements were done for Li, C, and Al, which are of prime importance for the design and operation of IFMIF (International Fusion Material Irradiation Facility). Past experiments on Li were discrepant markedly [14] and new high quality data were required for practical database and also for model refinement of neutron spectrum.
In the case of Li, measurement was done also for thin targets to clarify the primary process in the neutron emission. In Fig. 4 illustrated are the results of TTY and neutron emission cross section for 40 MeV deuterons.

In the DDX data, neutrons feeding to low-lying states of \(^6\)Be are apparent as an origin of so called “high energy tail” in TTY.
FIGURE 4. Differential TTY (top) and neutron emission cross sections (bottom) of Li (d,n) reactions at $E_d=40$ MeV.

FRAGMENT MEASUREMENT

Generally, fragments are difficult to detect because of large LET and most of past data on fragment production were obtained by activation or radiochemical techniques. Therefore, the information on energy-angular distribution, which is indispensable for the dosimetry and single event effect analysis, are very few. Following the experiments for right charged particle production [15], we are developing a experimental technique for direct measurement of energy-angular distribution of fragments.

One is the Bragg curve spectrometer (BCS) [16], which is a gridded-ionization chmaber shown in Fig. 5 and provides information on energy, atomic number $Z$, stopping power from a single counter. Thus BCS is widely used for spectroscopy of heavy ion reactions. We have developed BCS that is appropriate for fragment detection in neutron- and proton-induced reaction. To apply to neutron-induced reaction, BCS was designed with special feature of internal target, high-$Z$ elements for structural elements, and neutron collimation. By using the BCS, we could observe fragment spectra for 65 MeV neutrons [17], and continued improvement [18].

FIGURE 5. Schematic view of the BCS developed by our group to apply to neutron-induced reactions [17,18].

The BCS technique has been applied also to the proton-induced reactions to obtain complementary information to neutron-induced reaction, and also on the relation between neutron-induced reaction and proton-induced reaction.

Figure 6 shows particle separation by pulse-height vs Bragg peak for proton-induced reaction on a thin polyethylene target. The Bragg peak information is derived by differentiating anode signal with short time constant (~0.25µs) as usual, and the target is placed outside of BCS. The separation is fairly good above threshold.

FIGURE 6. Two-dimensional plot for pulse-height (X) vs Bragg peak (Y) of the present BCS for 70 MeV protons on polyethylene target.
Experimental data that can be compared with the present data are very few, but the present data proved to be qualitatively consistent with proton-induced data at 45 MeV and 100 MeV by Roche et al. [19]. In Fig. 7, the spectrum of Li deduced from the data in Fig. 6 is shown (dot) in comparison with the PHITS code employing QMD model for cascade process, the LA-150 library, and experimental data by Roche et al. [19].

There are large differences among calculations. In order to extend the dynamic range of the measurement to both high-energy side and low-energy side, we are developing data analysis method employing digital signal processing and data correction method for particles that are not stopped in BCS.

In the case of BCS, inherent limitation in dynamic range exists. On the other hand, the E-TOF method can cover almost whole range of energy spectrum if detectors are thick enough and timing resolution is good enough, although this method cannot be applied to neutron-induced reaction because of very small solid angle. For the reason, we are now developing E-TOF method for fragment measurement using a set of micro-channel plate for timing pickup and Si detector backed with a CsI(Tl) detector [18]. In Fig. 8, an example of a two-dimensional spectrum for timing (Y) vs energy (X) is shown. The result is promising. It will be possible obtain systematic data for fragment production by neutron- and proton-induced reactions by combining the BCS and E-TOF techniques.

FIGURE 7. Energy spectrum of Li deduced from the data in Fig. 6 (dot) in comparison with theoretical calculations, PHITS, LA-150, see text for detail.

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FIGURE 8. Energy vs. TOF scatter plot for fragments from proton-induced reaction on thin polyethylene film

RADIO-NUCLIDE PRODUCTION

We are conducting measurement of activation or radionuclide production cross sections for neutrons and protons, deuterons using a beam from the cyclotron [20-22].

Neutron activation data are obtained by using the $^7\text{Li}(p,n)$ quasi-monoenergetic neutron source shown in Fig. 2. Figure 9 illustrates neutron activation cross sections of $^{14}\text{N}(n,2n)$ reactions obtained with activation technique [21]. These data are indispensable for radiation safety analysis around accelerators. Experimental data for neutron-induced reaction are very few because of lacks of neutron sources with sufficient intensity and difficulty for data correction for continuous neutrons existing in the $^7\text{Li}(p,n)$ neutron source. Reflecting the situation, there are marked disagreement among calculations. The present experiments provided first new data above 30 MeV.

Data were also obtained for $^7\text{Be}$ production yield and cross section of $\text{Li}(d,x)^7\text{Be}$ reaction. The experiments were conducted concurrently with neutron spectrum measurement described in section 2. In the experiment, a stack of Li metal targets was prepared under Ar atmosphere and bombarded by deuteron beam. The activity of each Li plate was measured after TOF measurement with HP Ge detector. The total yield of $^7\text{Be}$ provides total production yields, and the activity for each plate gives cross section for energies of deuterons for each plate. In reduction of cross section, attenuation of deuteron flux in Li stack was taken into account. The results of $^7\text{Be}$ yields and $^7\text{Be}$ production cross sections are shown in Fig. 10, together with model calculations. There are large differences among experiments and theoretical calculation.
FIGURE 9. $^{14}$N(n,2n) cross section [21].

FIGURE 10. $^7$Be production yield (top) and excitation of $^7$Be production cross section (bottom).

For these experiments, the present $^7$Li(p,n) neutron source is not strong enough in neutron flux. Then, now we are installing a new source employing geometry in which target-to-sample distance can be shortened much compared with the present one.

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REFERENCES

1. e.g., http://jkj.tokai.jaeri.go.jp/index_j.html.