Measurements of $^{89}$Y(n,2n)$^{88}$Y and $^{89}$Y(n,3n)$^{87}$Y, $^{87m}$Y cross sections for fast neutrons at KIRAMS

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Abstract. A proton cyclotron MC-50 in Korea Institute of Radiological & Medical Science (KIRAMS) is used to carry out neutron activation experiments with $\text{Y}_2\text{O}_3$ targets irradiated with neutron beams of a continuous spectrum produced by proton beams on a thick beryllium target. Neutrons are generated by $^7\text{Be}$ (p, n) reaction with an incident proton intensity of 20 $\mu$A. The neutron spectra generated by proton beams of 30, 35, and 40 MeV are calculated by GEANT4 simulations. Nb powders are used for neutron flux monitoring by measuring the activities of $^{92m}$Nb through the reaction $^{93}$Nb (n, 2n). By using a subtraction method, the average cross section of $^{89}$Y(n,2n) and $^{89}$Y(n,3n) reactions at the neutron energies of 29.8 ± 1.8 MeV and 34.8 ± 1.8 MeV are extracted and are found to be close to the existing cross sections from the EXFOR data and the evaluated nuclear data libraries such as TENDL-2015 or EAF-2010.

1. Introduction

In the development of fast neutron systems, such as an Accelerator Driven System [1], nuclear data for neutron induced reactions at energies above 20 MeV are required, but they are not well known. For example, it is known that the content of Yttrium can improve the corrosion resistance of Zirconium-base alloy, but the data on $^{89}$Y(n, 2n) reactions are also used for the measurement of cross sections of neutron induced reactions in the high energy region [8,9]. Measurements at neutron energies above 20 MeV are only scarcely measured with large uncertainties [5]. In this work, we thus consider the neutron induced reactions of Yttrium for fast neutrons.

However, in the high energy region above 20 MeV, the measurement of cross sections poses difficulties in the measurement and analysis because mono-energetic neutron beam is not easy to get. To obtain an accurate (n, xn) cross section, a mono-energetic neutron facility is needed. Neutron time-of-flight (n-TOF) facilities are developed and available worldwide [6,7]. Quasi-monoenergetic neutrons produced by $^7\text{Li}$t(p,n) and $^9\text{Be}$t(p,n) reactions are also used for the measurement of cross sections of neutron induced reactions in the high energy region [8,9]. In this work, we consider the neutron induced reactions of Yttrium for fast neutrons.

2. Neutron spectra

Neutrons were produced by proton beams of 30, 35, and 40 MeV on a Be target of thickness of 10.5 mm, and the neutron spectra were obtained by using the GEANT4 code [10] with a data-based hadronic model discussed in Ref. [11]. Since the Be target is thick, the resulting neutron spectra were continuous with respect to neutron energy. Figure 4 of Ref. [12] shows that the magnitude of the neutron flux simulated by GEANT4 increases with the energy of the incident proton beam. However, it is remarkable that the neutron spectra are rather flat at neutron energies above 5 MeV and then drop sharply at the neutron energies close to the incident proton energies.

Let us denote by $\Phi_{E_p}(E_n)$ the neutron spectrum generated by the incident proton energy $E_p$. If we subtract $\Phi_{E_3}(E_n)$ from $\Phi_{E_0}(E_n)$, the difference $\Delta\Phi_1 = \Phi_{E_0}(E_n) - \Phi_{E_3}(E_n)$ can be obtained. Likewise, the difference $\Delta\Phi_2 = \Phi_{E_2}(E_n) - \Phi_{E_3}(E_n)$ can be obtained. These two distributions are plotted in Fig. 1, and the two deduced peaks can be fitted by Gaussian functions.

When the peak of the difference $\Delta\Phi_1(E_n) = \Phi_{E_0}(E_n) - \Phi_{E_3}(E_n)$ is fitted by a Gaussian function, the central value of the peak is 34.8 MeV with the width of 3.6 MeV. Similarly, $\Delta\Phi_2(E_n) = \Phi_{E_2}(E_n) - \Phi_{E_3}(E_n)$ is fitted with the peak energy of 29.8 MeV and the width of 3.6 MeV. These central values agree well with the flux-weighted average energies defined by

$$\langle E_n \rangle = \frac{\int_{E_{n_{\min}}}^{E_{n_{\max}}} \Delta\Phi(E_n) dE_n}{\int_{E_{n_{\min}}}^{E_{n_{\max}}} \Delta\Phi(E_n) dE_n}.$$  \(\text{(1)}\)
current of 20 µ proton beam energies of 30, 35, and 40 MeV and the beam current close to 50 µ beams in the range from 20 to 50 MeV at a maximum beam cyclotron at KIRAMS. This cyclotron provides proton The experiment was conducted by using the MC-50 3. Experiments

Figure 1. The difference spectra of the simulated neutron spectra. The red dotted and black solid curves are for the \( \Delta \Phi_1(E_n) = \Phi_{30}(E_n) - \Phi_{35}(E_n) \) and \( \Delta \Phi_2(E_n) = \Phi_{35}(E_n) - \Phi_{30}(E_n) \), respectively.

Figure 2. The geometry of the Be target, the neutron collimators, and the sample target.

In Eq. (1), the cut-value \( E_c \) is chosen as 31 MeV and the maximum energy \( E_{\text{max}} \) is chosen as 39 MeV for the \( \Delta \Phi_1(E_n) \), and \( E_c \) and \( E_{\text{max}} \) for \( \Delta \Phi_2(E_n) \) are 26 and 34 MeV, respectively.

3. Experiments

The experiment was conducted by using the MC-50 cyclotron at KIRAMS. This cyclotron provides proton beams in the range from 20 to 50 MeV at a maximum beam current close to 50 µA. In the present work we used three proton beam energies of 30, 35, and 40 MeV and the beam current of 20 µA.

The samples used for the experiment were of two types; one is yttrium oxide powder (99.99% pure) with an average mass of 3.03 g (for the three Y samples used) and another is a niobium powder (99.95% pure) with an average mass of 3.02 g (for the three Nb samples used). These samples were placed downstream along the neutron beam at a distance of 100 cm from the bottom of the collimator as shown in Fig. 2. All the six samples are placed inside a circle of 6 cm radius, which corresponds to \( \theta = 1.6^\circ \), where \( \theta \) is the angle of the neutron momentum with respect to the incident proton beam direction. The irradiation time was 5400 s.

After irradiation, the gamma-ray activities from the produced isotopes were measured by a shielded ORTEC HPGe detector.

4. Data analysis

Experimentally measured activities of \(^{88}\text{Y}\) or \(^{87}\text{Y}\) are proportional to the integration of cross sections \( \sigma(E_n) \) for \(^{88}\text{Y}\) (n, 2n) or \(^{87}\text{Y}\) (n, 3n) reactions multiplied by a neutron flux \( \Phi(E_n) \).

\[
A = N \int_{E_{\text{th}}}^{E_{\text{max}}} \sigma(E_n) \Phi(E_n) \, dE_n,
\]

(2)

where \( E_{\text{th}} \) is the threshold energy of each reaction, and \( N \) is a constant. By using a subtraction method [13], we can extract the cross sections at the central value of the neutron Gaussian peak. The cross section at the neutron energy of 34.8 MeV can be extracted by the following

\[
\sigma = \frac{\left( \frac{A_{30}}{N_{30}} - C \frac{A_{35}}{N_{35}} \right) - \int_{E_{\text{th}}}^{E_{\text{max}}} \sigma(E_n) \cdot (\Phi_{30}(E_n) - C \Phi_{35}(E_n)) \, dE_n}{\int_{E_{\text{th}}}^{E_{\text{max}}} \Phi_{30}(E_n) - C \Phi_{35}(E_n) \, dE_n},
\]

(3)

where \( C \) is a factor to be explained shortly. We take the cross sections from the TENDL-2015 data library [14] for \( \sigma(E_n) \) in Eq. (3), because the TENDL-2015 seems to reproduce best the EXFOR data for these reactions.

We remark that \( \Delta \Phi_i \) (i = 1, 2) in Fig. 1 has non-zero values in the low energy regions while it is desirable that \( \Delta \Phi_i = 0 \) at low energies. Thus, in the calculation of the integrals in Eq. (3), the non-zero values of \( \Delta \Phi_i \) can introduce errors in the analysis. To minimize the effect due to the non-zero values of \( \Delta \Phi_i \) in the low energy region from the threshold energy \( (E_{\text{th}}) \) to a cut-value \( (E_c) \), we introduce a neutron flux cancellation factor by defining \( C \) as a factor which will make the integral

\[
\frac{E_c}{E_{\text{th}}} \int_{(E_{\text{th}})}^{(E_c)} (\Phi_{30}(E_n) - C \Phi_{35}(E_n)) \, dE_n = 0,
\]

(4)

zero. The values of \( C \) is about 1.27 for \( \Delta \Phi_1 \) and is about 1.34 for \( \Delta \Phi_2 \).

Here we remark that the integrated experimental activities of \(^{58}\text{Mn}\) and \(^{24}\text{Na}\) from \(^{56}\text{Fe}\) (n,p)\(^{56}\text{Mn}\) and \(^{27}\text{Al}\) (n, \( \alpha \))\(^{24}\text{Na}\) reactions, respectively, are about 10% larger than those estimated by using the neutron flux shown in Fig. 4 of Ref. [12]. This 10% difference in the experimental and calculated activities may be traced to the uncertainty in the neutron flux. To check this uncertainty independently, we used a Nb powder to monitor the neutron flux. When Nb powders are placed together with Y samples, \(^{92}\text{mNb}\) are excited due to \(^{93}\text{Nb}\) (n, 2n) reaction. We compared the experimentally measured activity with the calculated activity of \(^{92}\text{mNb}\) from \(^{92}\text{Nb}\) (n, 2n) reaction.

5. Results

The reactions observed in this work are listed in Table 1, and the extracted cross sections for \(^{89}\text{Y}\) (n, 3n) \(^{87}\text{mY}, \)
Table 1. The reactions observed and their characteristics.

| Nuclear reaction     | $T_{1/2}$ | $E_n$ [MeV] | $E_{\gamma}$ [keV] | $I_{\gamma}$ [%] |
|----------------------|-----------|-------------|-------------------|-----------------|
| $^{89}\text{Y}(n,3n)^{87m}\text{Y}$ | 13.37 h   | 21.06       | 380.79            | 78.0            |
| $^{89}\text{Y}(n,3n)^{87}\text{Y}$  | 79.8 h    | 21.06       | 388.53            | 82.0            |
| $^{89}\text{Y}(n,2n)^{88}\text{Y}$  | 106.65 d  | 11.61       | 898.04            | 93.7            |

Table 2. The cross sections extracted for $^{89}\text{Y}(n,3n)^{87m}\text{Y}$, $^{89}\text{Y}(n,3n)^{87}\text{Y}$ and $^{89}\text{Y}(n,2n)^{88}\text{Y}$ reactions.

| $E_n$ [MeV] | $\sigma_{n,3n}$ [mb] | $\sigma_{n,3n}$ [mb] | $\sigma_{n,2n}$ [mb] |
|-------------|-----------------------|-----------------------|-----------------------|
| 29.8 ± 1.8  | 352 ± 58              | 143 ± 16              | 312 ± 332             |
| 34.8 ± 1.8  | 498 ± 80              | 205 ± 34              | 817 ± 362             |

Figure 3. The cross sections extracted for $^{89}\text{Y}(n,3n)^{87m}\text{Y}$ are plotted by the squares at two neutron energies and are compared with the EXFOR data, TENDL and EAF cross sections.

Figure 4. The cross sections extracted for $^{89}\text{Y}(n,3n)^{87}\text{Y}$ are plotted by the squares at two neutron energies and are compared with the EXFOR data, TENDL and EAF cross sections.

Figure 5. The cross sections extracted for $^{89}\text{Y}(n,2n)^{88}\text{Y}$ are plotted by the squares at two neutron energies and are compared with the EXFOR data, TENDL and EAF cross sections.

6. Summary

A proton cyclotron MC-50 in KIRAMS is used to carry out neutron activation experiments with Y$_2$O$_3$ targets irradiated with neutron beams of a continuous spectrum produced by proton beams on a thick beryllium target. By using a subtraction method, the cross section of $^{89}\text{Y}(n,2n)$ and $^{89}\text{Y}(n,3n)$ reactions at the neutron energies of 29.8 ± 1.8 MeV and 34.8 ± 1.8 MeV are extracted and are found to be close to the existing cross sections from the EXFOR data and the evaluated nuclear data libraries such as TENDL-2015 or EAF-2010. We show continuous energy neutron spectra can be used to extract the (n, xn) cross sections.

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References

[1] Y. Kadi, J.P. Revol, Design of an accelerator-driven system for the destruction of nuclear waste, Lectures given at the Workshop on Hybrid Nuclear Systems for Energy Production, Utilisation of Actinides & Transmutation of Long-Lived Radioactive Waste. Trieste, 3–7 September 2001
[2] D.R. Nethaway, Nucl. Phys. A 190, 635 (1972)
[3] L.R. Veeser, E.D. Arthur, P.G. Young, Phys. Rev. C 16, 1792 (1977)
[4] J. Vrzalová, O. Svoboda, A. Kráša, A. Kugler, M. Majerle, M. Suchopár, V. Wagner, Nucl. Instrum. Methods Phys. Res. Sect. A 726, 84 (2013)
[5] V. Wagner, M. Suchopár, J. Vrzalová, P. Chudoba, T. Herman, O. Svoboda, B. Geier, A. Kráša, M. Majerle, A. Kugler, J. Phys. Conf. Ser. 533, 012052 (2014)
[6] C. Guerrero, et al., Eur. Phys. J. A 49, 27 (2013)
[7] W. Mondeleers, P. Schillebeeckx, GELINA, a neutron time-of-flight facility for high-resolution neutron data measurements, Notiziario neutroni e luce di sincrotrone, 11, 19 (2006)
[8] H. Harano, N. Ralf, Metrologia 48, S292 (2011)
[9] M. Ibaraki, M. Baba, T. Miura, T. Aqki, T. Hiroishi, H. Nakashima, S.I. Meigo, S. Tanaka, J. Nucl. Sci. Technol. 39 405 (2002)

[10] S. Agostinelli, et al., Nucl. Instrum. Methods Phys. Res. Sect. A 506, 250 (2003)

[11] J.W. Shin, T.-S. Park, Nucl. Instrum. Methods Phys. Res. Sect. B 342 194 (2015)

[12] J.W. Shin, S.-I. Bak, C.M. Ham, E.J. In, K.J. Min, Y. Zhou, T.-S. Park, S.-W. Hong, V.N. Bhoraskar, Nucl. Instrum. Methods Phys. Res. Sect. A 797, 304 (2015)

[13] J.K. Park, S. Kwon, S.W. Lee, J.T. Kim, J.-S. Chai, J.W. Shin, S.W. Hong, J. Korean Phys. Soc. 58 1511 (2011)

[14] https://tendl.web.psi.ch/tendl_2015/neutron.html

[15] http://www-nds.iaea.org/exfor