Measurements of the neutron activation cross sections for Bi at 287 and 370 MeV

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*Neutron activation cross section /
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Summary. We present cross section measurements for neutron-induced activation of Bi, at 287 and 370 MeV. These values were derived from the activation method using a quasi-monoenergetic neutron field based on the ${}^7\text{Li}(p, n)$ reaction. In separate experiments, samples were irradiated with neutrons derived from ${}^7\text{Li}(p, n)$ reaction at either 0° or 30° for proton beam axis. This approach allows the subtraction of the low energy neutron components. The measured cross sections are compared with the findings of other studies, and evaluated in relation to nuclear data files.

1. Introduction

Activation cross sections for high energy neutrons are important nuclear data, needed for the estimation of residual radioactivities in accelerator facilities. In addition they are fundamental in order to decipher cosmic-ray irradiation histories when evaluating the amounts of cosmogenic nuclides stored in extraterrestrial matter. While there are many measurements of the neutron activation cross sections below 20 MeV, comparable measurements for energies above 20 MeV are scarce [1–5], with no measurements above 200 MeV. Neutron-activation cross sections are evaluated assuming that neutron-induced cross sections above 100 MeV are approximately equal to proton-induced cross sections of the same energy. The cross section data of ${}^{209}\text{Bi}(n, xn)$ reactions were reported up to 150 MeV [1] and are applied for high energy neutron spectrometry. But evaluation of the neutron-induced activation above 150 MeV are not well established due to absence of experimental data.

This study measures the neutron activation cross sections of Bi above 200 MeV by using a quasi-monoenergetic neutron field. The quasi-monoenergetic neutron field was produced from enriched ${}^7\text{Li}$ that is bombarded by 300 and 392 MeV protons. The spectra of these neutron fields are not

purely monoenergetic, with both a high energy peak coming from the ${}^7\text{Li}(p, n)$ reaction, and a low energy tail resulting from the consequent break-up reaction. Thus, two different experiments were performed by using two different neutron beams at 0° and 30° for proton beam axis. Subtracting the low energy neutron contribution produced by the 30° beam angle from the results of the 0° beam resolves the contribution made by low energy neutrons, as described in Ref. [5].

2. Experiments

2.1 Neutron field from the ${}^7\text{Li}(p, n)$ reaction

Irradiation experiments were performed in the neutron field produced by the ${}^7\text{Li}(p, n)$ reaction using the ring-cyclotron of the Research Center for Nuclear Physics (RCNP), Osaka University. The quasi-monoenergetic neutrons were emitted from a 1.1-cm-thick ${}^7\text{Li}$ target, bombarded with 300 and 392 MeV protons. The neutrons emitted in the forward direction were extracted into the time-of-flight (TOF) room through an iron collimator of $12 \times 10 \text{ cm}^2$ aperture, embedded inside a 150-cm-thick concrete wall. Charged particles were stopped in the collimator, hence removed from the neutron flight path, by a bending magnet placed within the collimator. The neutron spectra were measured by using the TOF method. A 12.5 cm diameter, 20 cm long NE213 liquid scintillation detector was placed at the 0° , then later at 30° directions. The flight path of spectrum measurement (distance between Li target and detector) are listed in Table 1.

Table 1. The conditions of the irradiation experiments.

Proton energy (MeV)		Flight path for spectra measurement (m)	Distance between Li target and Bi sample (m)	Irradiation time (h)
300	0°	17.1	8.05	25.7
	30°	16.1	7.05	17.9
392	0°	45.0	8.03	26.7
	30°	44.0	7.03	22.0

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Fig. 1. Schematic view of the experimental set up. (upper panel: 0° irradiation, lower panel: 30° irradiation).

Fig. 2. Neutron energy spectrum generated by the ${}^7\text{Li}(p, n)$ reaction. (upper panel: 392 MeV irradiation, lower panel: 300 MeV irradiation).

A schematic view of the experimental set up is shown in Fig. 1. The neutron flight time was measured as the temporal difference between the timing signal from the accelerator and the neutron detector signal. The neutron spectra were obtained after corrections for detector solid angle, proton beam current and detection efficiency that was calculated using CECIL [6] and SCINFUL-QMD [7, 8] codes. The measured neutron spectra are shown in Fig. 2.

2.2 Bismuth sample irradiation

Two types of irradiation were performed in the present study. The Bi samples were placed at an angle of 0° to the proton beam to measure activities induced by both high energy peak neutrons and low energy neutrons, while to measure activities induced by low energy neutrons only, Bi samples were placed at an angle offset 30° from the direction of the proton beam. The conditions of the irradiation experiments (distance between Li target and sample) are listed in Table 1. Average proton beam intensity was about 1 μA . During the irradiation time, the proton beam current at the beam dump was recorded with a digital current integrator, connected to a multi-channel scaler (MCS) to monitor the fluctuations of the proton beam currents. After irradiation, the gamma rays emitted from the irradiated samples were measured with a high-purity germanium (HPGe) detector to be connected to 8192 multichannel analyzers. The samples were measured several times in order to identify newly created radio-active nuclides by their half-lives.

3. Data analysis

3.1 Reaction rates

Reaction rates of radioactive nuclides produced in the samples were determined from the gamma-ray spectra and decay curves, after first accounting for any beam current fluctuations during the irradiation and corrections in the peak efficiency of the HPGe detector using the EGS4 code [9]. The reaction rates per proton, corrected for the above mentioned effects, are given as follows:

$$R = \frac{\lambda C}{\gamma \varepsilon N (1 - e^{-\lambda t_m}) e^{-\lambda t_c} \sum_{i=1}^n I_i (1 - e^{-\lambda \Delta t}) e^{-\lambda (n-i) \times \Delta t}} \quad (1)$$

where λ is the decay constant (s^{-1}), C is the total counts of gamma-ray peak area, γ is the branching ratio of gamma-rays, ε is the peak efficiency, N is the number of atoms in the sample, t_m is measurement time(s), t_c is the cooling time(s), I_i is the beam current for the irradiation time interval Δt (proton). The physical properties of the radioactive nuclides measured in this study are listed in Table 2 [10].

3.2 Cross section estimation

The measured neutron spectra at 0° show a high energy peak and a low energy tail (Fig. 2), while the high energy peak that is prominent at 0° being absent from the 30°. To remove the component of nuclides produced by the low energy tail, the 30° spectrum is normalized to the one measured at

Table 2. The physical properties of radioactive nuclides.

Reaction	Half-life	Gamma-ray energy (keV)	Branching ratio (%)
$^{209}\text{Bi}(n, 9n)^{201}\text{Bi}$	103 min	629.1	26
$^{209}\text{Bi}(n, 7n)^{203}\text{Bi}$	11.76 h	820.23	29.6
$^{209}\text{Bi}(n, 6n)^{204}\text{Bi}$	11.22 h	374.76	81.8
$^{209}\text{Bi}(n, 5n)^{205}\text{Bi}$	15.31 d	703.45	31.1
$^{209}\text{Bi}(n, 4n)^{206}\text{Bi}$	6.243 d	803.1	98.9
$^{209}\text{Bi}(n, x)^{183g}\text{Os}$	13.0 h	381.763	89.6
$^{209}\text{Bi}(n, x)^{199}\text{Tl}$	7.42 h	455.46	12.4
$^{209}\text{Bi}(n, x)^{200}\text{Pb}$	21.5 h	147.7	38.2
$^{209}\text{Bi}(n, x)^{201}\text{Pb}$	9.33 h	331.17	76.9

0° by equalizing the neutron fluence in low energy region, then the corrected result is then obtained by subtracting the normalized 30° spectrum from the 0° spectrum, as shown in Fig. 3. Thus, the subtraction of the nuclide component produced by a beam angled at 30° from one angled at 0° gives a yield produced only by high energy peak neutrons. The cross section for peak neutron energy, σ can be obtained as follows:

$$\sigma = \frac{R_0 - R_{30} f_1 f_2}{\phi_{\text{peak}}} \quad (2)$$

Fig. 3. Corrected spectrum obtained by subtracting the normalized 30° spectrum from the 0° spectrum. (Upper panel: 392 MeV irradiation, lower panel: 300 MeV irradiation).

where R_0 and R_{30} are the reaction rate (1/atom/ μC) for 0° and 30° irradiation, respectively, f_1 is the normalization factor to equalize the neutron fluence in the low energy range, 0.77 and 1.11 for 300 and 392 MeV irradiation respectively, f_2 is the correction factor for the detector solid angle, 0.77. ϕ_{peak} is the high energy peak neutron fluence of corrected spectrum, 3.23×10^4 and 2.47×10^4 n/cm²/ μC for 300 and 392 MeV irradiations, respectively. The energy ranges of the peak neutron fluences are 240–320 and 300–400 MeV for 300 and 392 MeV irradiations, respectively. The energy ranges of the low energy neutron are 0–240 and 0–300 MeV for 300 and 392 MeV irradiations, respectively. Averaged neutron energies for the high energy peaks of the corrected spectra are 287 and 370 MeV, for 300 and 392 MeV primary proton energy, respectively. The uncertainties in the high energy peak of the corrected spectra, evaluated from FWHM of high energy peak, amount to 10 and 13 MeV for the 300 and 392 MeV irradiations, respectively. Contributing to the estimate of uncertainty in the cross section measurements are the counting statistics (< 15%), detector efficiency (Ge detector (10%), NE213 detector (15%)), beam current monitoring (5%) and correction for contribution of low energy tail (10%).

4. Results and discussions

The cross sections obtained for $^{209}\text{Bi}(n, xn)^{201,203,204,205,206}\text{Bi}$, $^{209}\text{Bi}(n, x)^{183g}\text{Os}$, $^{209}\text{Bi}(n, x)^{199}\text{Tl}$, $^{209}\text{Bi}(n, x)^{200,201}\text{Pb}$ are shown in Fig. 4a–i, respectively, and tabulated in Table 3. The shaded symbols are results from this work, and the open symbols are as cited from other experimental data by Kim *et al.* [1] and Michel *et al.* [11]. Here, our results are compared with the proton-induced reaction results of Michel *et al.* [11] due to the absence of neutron activation data above 200 MeV, as mentioned above. The curves shown in Fig. 4 are derived from the ENDFB-VI high energy file [12] and JENDL-HE [13]. The ENDFB-VI high energy file [12] and JENDL-HE [13] agree well with some the findings of this study, as is apparent in Fig. 4a and f, however, other results presented here are greater than in these other studies, such as Fig. 4b–e and g–i. From Fig. 4b–d and h, it can be seen that our results agree well with those of Michel [11] at 370 MeV, but that they are lower at 287 MeV. Fig. 4e, shows a good agreement between this study and that of Michel [11].

Table 3. The physical properties of radioactive nuclides.

Reaction	300 MeV irradiation		392 MeV irradiation	
	Cross section (mb)	Error (mb)	Cross section (mb)	Error (mb)
$^{209}\text{Bi}(n, 9n)^{201}\text{Bi}$	28	10	24	7
$^{209}\text{Bi}(n, 7n)^{203}\text{Bi}$	46	17	35	10
$^{209}\text{Bi}(n, 6n)^{204}\text{Bi}$	46	18	36	11
$^{209}\text{Bi}(n, 5n)^{205}\text{Bi}$	56	25	40	14
$^{209}\text{Bi}(n, 4n)^{206}\text{Bi}$	53	26	44	14
$^{209}\text{Bi}(n, x)^{183g}\text{Os}$			8.1	1.9
$^{209}\text{Bi}(n, x)^{199}\text{Tl}$	75	19	93	28
$^{209}\text{Bi}(n, x)^{200}\text{Pb}$	55	14	81	26
$^{209}\text{Bi}(n, x)^{201}\text{Pb}$	65	17	81	25

Fig. 4. Excitation function for nuclides produced from Bi.

Based on Fig. 4, the following can be stated: (1) nuclear data file results are less than our measurements, except for nuclides of higher threshold energy. (2) at 370 MeV, our results agree with proton-induced reaction data, but at 287 MeV, proton-induced data differ from our results for some reactions.

Further systematic experimental studies are required for evaluation of neutron-induced activation cross sections.

5. Conclusion

Neutron activation cross sections induced in Bi at 287 and 370 MeV were measured by the activation method using quasi-monoenergetic neutron fields from the ${}^7\text{Li}(p, n)$ reaction. These experimental results will be useful as benchmark values to evaluate nuclear data and investigate the accuracy of calculation codes.

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