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Measurements of the ^{209}Bi(n, g)^{210g}Bi Reaction Cross Sections at 30 and 520 keV

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Abstract

The cross sections of the ^{209}Bi(n, γ)^{210g}Bi reaction which leads to the ^{210}Po production in the lead-bismuth coolant were measured at the incident neutron energies of 30 and 520 keV. An activation method was adopted with the ^{7}Li(p, n)^{7}Be reaction neutron source. The incident neutron flux on a bismuth sample was obtained from the activities of standard gold samples. The α rays from the ^{210}Po nuclei produced in the bismuth sample were measured with a Si surface barrier detector. The derived cross sections were 0.77±0.20 mb at 30 keV and 0.46 ±0.11mb at 520 keV, which were one tenth and one third of the evaluated values in JENDL Activation Cross Section File, respectively.

1. Introductio n

Recently, the Lead-Bismuth Coolant (LBC) attracts a great deal of attention(1) in the research fields of fast reactors and accelerator-driven systems because it has advantages such as chemical inertness, high boiling point, and low neutron moderation. However, the α-emitter of ^{210}Po with the half-life of 138.376 d(2) is produced in LBC through the ^{209}Bi(n, γ)^{210g}Bi reaction and the β decay of ^{210g}Bi with the half-life of 5.013 d(2). The activity of ^{210}Po in LBC causes a problem when maintaining a system with LBC, and the ^{210}Po production is one of the most important disadvantages of LBC. Therefore, the accurate evaluation of ^{210}Po activity is important in the design of a system with LBC.

From the viewpoint mentioned above, the cross section of the ^{209}Bi(n, γ)^{210g}Bi reaction is very important, but the experimental data are very scarce. In particular, the experimental result of Booth *et al.* at 25 keV(3) is the only one in the keV region. Therefore, the uncertainty of the cross section data contained in evaluat ed nuclear data libraries such as JENDL seems to be large in the keV region.

This paper presents the measurements of the ^{209}Bi(n, γ)^{210g}Bi reaction cross sections at 30 and 520 keV. The experimental procedure and data processing are described in Sections 2 and 3, respectively. The results and discussion are given in Section 4. Finally, the conclusion is given in Section 5.

2. Experimental Procedure

The measurements were performed at the incident neutron energies of 30 and 520 keV with an activation method. The arrangement for neutron irradiation is shown in Fig. 1.

Pulsed neutrons were generated using the ^{7}Li(p, n)^{7}Be reaction by a 1.5-ns wide pulsed-proton beam from the 3-MV Pelletron accelerator of the Research Laboratory for Nuclear Reactors at the Tokyo Institute of Technology. The proton-pulse repetition rate was 4 MHz and the average proton-beam current was about 12 μA in both measurements at 30 and 520 keV. The energy spectrum of neutrons incident on a bismuth sample was measured by means of a time-of-flight (TOF) method with a ^{6}Li-glass detector.

Each bismuth sample was a metallic disk with the diameter of 30 mm and the thickness of 3.0 mm, and sandwiched between two gold disks with the thickness of 0.5 mm. Each sandwiched sample was placed at 20 mm from the neutron source, as shown in Fig. 1. The neutron irradiation time was 20 or 50 h in the measurement at 30 or 520 keV, respectively. The neutron generation rate was monitored with a ^{6}Li-glass detector.

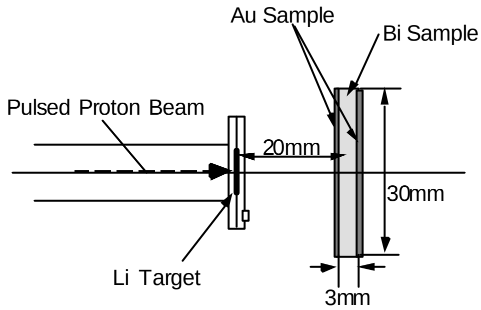

Fig. 1 Arrangement for neutron irradiation

After the irradiation, the 412-keV γ rays from the ^{198}Au nuclei produced in the standard gold samples were measured with a high-purity Ge detector which has the energy resolution of 1.9 keV (FWHM at 1,333 keV) and the relative efficiency of 100 %. The Ge detector was placed in a heavy shield, and the γ-ray measurement was performed for about 11 d to confirm the half-life (2.69517 d[(2)]) of ^{198}Au.

The α rays from the ^{210}Po nuclei produced in the bismuth sample were measured with a Si surface barrier detector which has the energy resolution of 20 keV at 5,486 keV and the sensitive area of 450 mm^2. The Si detector and the bismuth sample were placed in a vacuum chamber (~100 Pa), and the measurement was performed for about 170 d to confirm the half-life of ^{210}Po.

3. Data Processing

The energy spectrum of incident neutrons in each measurement was obtained from the neutron TOF spectrum measured with the ^{6}Li-glass detector, and used in the calculations of the average neutron energy, the average capture cross section of ^{197}Au, etc.

The activities of the gold samples after the neutron irradiation were obtained from the 412-keV γ-ray peak-area counts, the emission probability of the 412-keV γ ray (0.99812[(2)]), and the detection efficiency of the Ge detector. The obtained activities are shown in Fig. 2, where the solid circles show the activities of the front gold sample in the measurement at 520 keV. On the other hand, the activity Au(t) of a gold sample at the time t after the end of neutron irradiation is expressed as follows:

$$\mathrm{Au}(t) = N\sigma\phi(1-e^{-\lambda T})e^{-\lambda t}, \qquad (1)$$

where N is the number of ^{197}Au nuclei in the gold sample, λ the decay constant of ^{198}Au, and T the irradiation time. The average capture cross section σ of ^{197}Au was calculated from the neutron energy spectrum described above and the standard capture cross sections[(4)] of ^{197}Au in ENDF/B-VI. The neutron flux ϕ was obtained by fitting Eq. (1) to the measured activities of the gold samples, as shown by the solid line in Fig. 2.

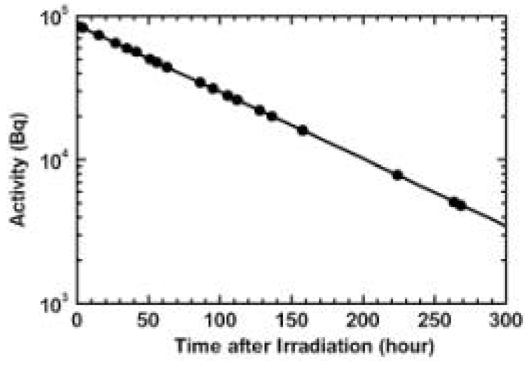

Fig. 2 Measured and calculated ^{198}Au activities in the front gold sample in the measurement at 520 keV

Two of α-ray spectra observed in the measurement at 520 keV are shown in Fig. 3 together with the background spectrum. The thick solid line shows the foreground spectrum measured after 3 days from the end of irradiation, and the dotted line shows that after 25 days. Since the bismuth sample was very thick in comparison with the range (about 15 μm) of the 5.304-MeV α ray from ^{210}Po, the α-ray spectra were continuous ones with the end point of 5.3 MeV. The β rays from ^{210g}Bi are observed below about 1 MeV in the spectrum after 3 days but disappear in the spectrum after 25 days because of the short half-life (5.013 d) of ^{210g}Bi. The thin solid line shows the background spectrum, where the intensity below about 2 MeV is dominant.

The energy spectrum of α rays incident on the Si detector was calculated with a Monte-Carlo method in order to derive the detection efficiency for the α rays isotropically emitted in the bismuth sample. As for the density distribution of ^{210}Po in the sample, the $1/r^2$ dependence from the neutron source was taken into account in the calculation. The dashed line in Fig. 3 shows the calculated spectrum whose intensity is normalized to that of the observed spectrum after 3 days.

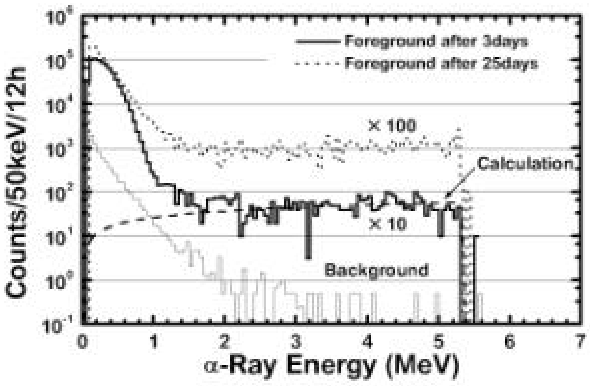

Fig. 3 Measured and calculated α-ray spectra in the measurement at 520keV

From the analysis of measured α-ray spectra, the activities of ^{210}Po in the sample after the irradiation were obtained in each measurement, as shown in Fig. 4, where the open circles with the error bars show the obtained activities in the measurement at 520 keV. On the other hand, the polonium activity Po(t) of a bismuth sample is expressed as follows:

$$\mathrm{Po}(t) = \frac{n \sigma_g \phi}{\lambda_b - \lambda_a}\{\lambda_b (1 - e^{-\lambda_a T}) e^{-\lambda_a t} - \lambda_a (1 - e^{-\lambda_b T}) e^{-\lambda_b t}\}, \qquad (2)$$

where λ_b and λ_a are the decay constants of ^{210m}Bi and ^{210}Po, respectively, n the number of ^{209}Bi nuclei in the bismuth sample, and ϕ the neutron flux obtained above. The ^{209}Bi(n, γ)^{210g}Bi reaction cross section σ_g for the incident neutron energy spectrum was derived by fitting Eq. (2) to the measured activities. The fitted curve is shown by the solid line in Fig. 4.

In addition to the statistical error, the following errors were taken into account for the error of the final results: the errors due to the standard capture cross sections of ^{197}Au (3 %), the 412-keV γ-ray detection efficiency (1 %), and the α-ray detection efficiency (20 %) including the effect of neutron self-shielding and multiple-scattering in the bismuth sample.

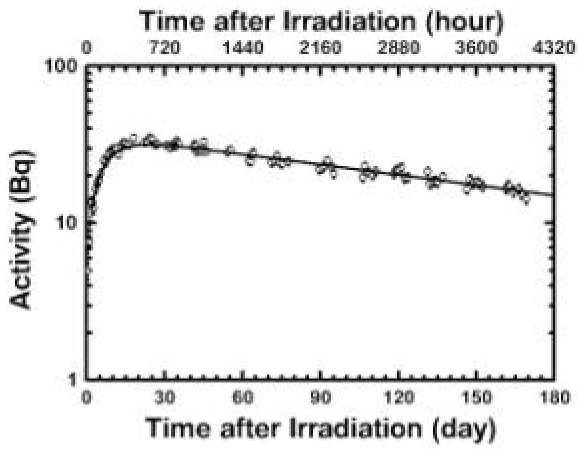

Fig. 4 Measured and calculated ^{210}Po activities in the bismuth sample in the measurement at 520 keV

4. Results and Discussion

The cross sections of the ^{209}Bi(n, γ)^{210g}Bi reaction were derived as 0.77±0.20 mb at 30 keV and 0.46±0.11 mb at 520 keV. In Fig. 5, the present results are compared with that of Booth *et al.*[3] and the evaluated values in JENDL Activation Cross Section File[5]. The result of Booth *et al.* is 1.8±0.7 mb at 25 keV and about two times as large as the present one at 30 keV. The JENDL evaluations are ten times at 30 keV and three times at 520 keV in comparison with the present results.

Booth *et al*. adopted a double-ratio comparison method(3); they used ^{127}I as a standard isotope, and they used an Sb-Be photo neutron source for the 25-keV neutron activation and a water-moderated Po-Be neutron source for the thermal neutron activation. They needed the activation cross sections of ^{127}I and ^{209}Bi at thermal energy and that of ^{127}I at 25 keV to derive their ^{209}Bi result at 25 keV. If the current values(6)-(8) for these reference cross sections are adopted instead of the values given in their paper, the value of 1.9 mb is obtained instead of the reported value of 1.8 mb. Namely, their large value cannot be ascribed to the reference cross sections used in their data analysis.

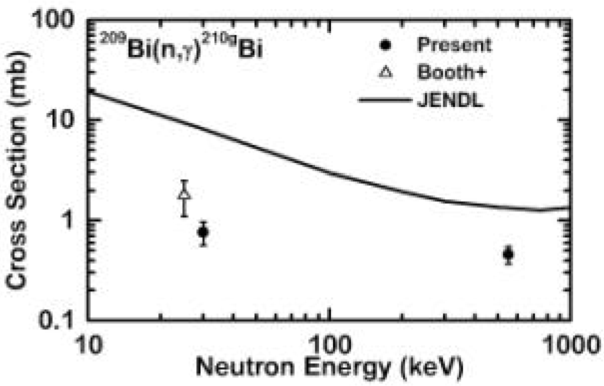

Fig. 5 Cross sections of the ^{209}Bi(n, γ)^{210g}Bi reaction in the keV region

In the JENDL evaluation of the ^{209}Bi(n, γ)^{210g}Bi reaction cross section, the statistical model calculation with a spherical optical potential was mainly adopted above 10 keV. In the evaluation, the calculated cross sections for the ^{209}Bi(n, γ)$^{210g+m}$Bi reaction, *i.e.* total capture reaction, were adjusted so as to reproduce the experimental results of Macklin and Halperin(9) and Voignier *et al*.(10). However, the results of Macklin and Halperin are about three times as large as the recent measurements of Mutti *et al*.(11) and Kawakami *et al*.(12) below 60 keV, and both results of Voignier *et al.* and Macklin and Halperin are about two times as large as the very recent result of Saito *et al*.(13) around 500 keV. The adjustment directly affects the calculated values for the ^{209}Bi(n, γ)^{210g}Bi reaction cross section. Therefore, one of the reasons for the large values in JENDL should be ascribed to the incorrect adjustment.

It is worth noting that the effect of the α rays from the ^{210m}Bi nuclei produced in the bismuth sample on the present results was negligibly small because of the very long half-life (3.04×10^6 y(2)) of ^{210m}Bi.

5. Conclusion

The cross sections of the ^{209}Bi(n, γ)^{210g}Bi reaction were measured at the incident neutron energies of 30 keV and 520 keV using an activation method with the ^{7}Li(p, n)^{7}Be reaction neutron source and standard gold samples. The derived cross sections were 0.77 ± 0.20 mb at 30 keV and 0.46 ± 0.11 mb at 520 keV.

The previous experimental result of Booth *et al.* at 25 keV was about two times as large as the present result at 30 keV. The evaluated values in JENDL Activation Cross Section File were about ten times at 30 keV and about three times at 520 keV in comparison with the present results. One of the reasons for the large values in JENDL was ascribed to the incorrect adjustment of the statistical model calculation.

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