Measurement of Cross Sections of the ^{210}Po Production Reaction by keV-Neutron Capture of ^{209}Bi

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The cross sections of the ^{209}Bi(n,γ)^{210g}Bi reaction were measured in the keV-neutron region, using an activation method with the detection of α rays from ^{210}Po. Pulsed keV neutrons were generated from the ^{7}Li(p,n)^{7}Be reaction by a 1.5-ns bunched proton beam from the 3-MV Pelletron accelerator of the Research Laboratory for Nuclear Reactors at the Tokyo Institute of Technology. The ^{209}Bi samples were metallic bismuth evaporated on a gold backing. After sample irradiation, the 412-keV γ rays from the gold backing were measured with an HPGe detector, and then the α rays from ^{210}Po nuclei in the bismuth sample were measured with a Si surface barrier detector. The derived cross sections were 1.9±0.4 mb and 0.62±0.14 mb at the average neutron energies of 30 keV and 534 keV, respectively. The present value at 30 keV is in agreement with the previous measurement at 24 keV of Booth *et al.*, but about a quarter of the evaluation of JENDL Activation cross section File. The present result at 534 keV is the first experimental one around 500 keV, and is about a half of the evaluation of JENDL Activation cross section File.

1. Introduction

The Lead-Bismuth Eutectic Coolant (LBEC) attracts a great deal of attention in the research fields of fast breeder reactors and accelerator driven systems because of its advantages such as chemical inertness, high boiling point, low neutron moderation, etc. However, the α-emitter of ^{210}Po with the half-life of 138 d is produced in LBEC through the ^{209}Bi(n,γ)^{210g}Bi

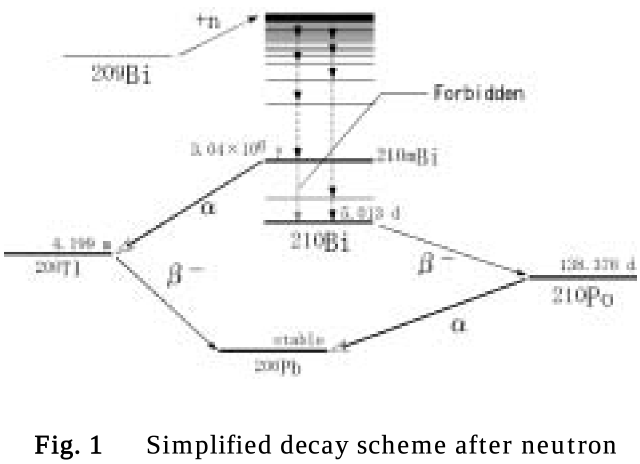

Fig. 1 Simplified decay scheme after neutron capture of ^{209}Bi

reaction and the following β^- decay of ^{210g}Bi with the half-life of 5.01 d. **Figure 1** shows a simplified decay scheme after the neutron capture of ^{209}Bi. The radioactivity of ^{210}Po causes a problem when maintaining a system with LBEC. Moreover, as seen from Fig. 1, ^{210m}Bi with the half-life of 3.04×10^6 y is produced through the competitive neutron capture reaction of ^{209}Bi(n,γ)^{210m}Bi. Such a long-live nuclide continuously accumulates in proportion to neutron irradiation time, which will bring about an important problem on nuclear wastes. Therefore, the ^{210}Po and ^{210m}Bi production cross sections, *i.e.* the ^{209}Bi(n,γ)^{210g}Bi and ^{209}Bi(n,γ)^{210m}Bi reaction cross sections, are very important to evaluate the radioactivity of ^{210}Po and ^{210m}Bi in a system with LBEC.

In astrophysics, ^{209}Bi is the end point of S-process nucleosynthesis in stars and its keV neutron capture cross sections are important for the study on the nucleosynthesis. In nuclear physics, the cross sections are important for understanding the keV-neutron capture reaction mechanism.

In the present study, from the above viewpoints, the ^{210}Po production cross sections in the keV-neutron region were measured with an activation method, and then the ^{210m}Bi production cross sections were determined from the differences between the total capture cross sections of ^{209}Bi and the ^{210}Po production cross sections.

2. Experimental Procedure

The experiments were performed, using the 3-MV Pelletron accelerator of the Research Laboratory for Nuclear Reactors at the Tokyo Institute of Technology. Pulsed keV neutrons were generated from the ^{7}Li(p,n)^{7}Be reaction by a 1.5-ns bunched proton beam from the Pelletron accelerator. The incident neutron spectrum on a ^{209}Bi sample was measured by means of a Time-Of-Flight (TOF) method with a ^{6}Li-glass scintillation detector. Each ^{209}Bi sample was metallic bismuth evaporated on a gold backing whose diameter and thickness were 15 mm and 0.1 mm, respectively. The thickness of the bismuth layer is about 5 μm. The sample was located at a distance of 2 or 5 cm from the neutron source and at an angle of 0 ° with respect to the proton beam axis. The irradiation time was 40 to 60 h. After each irradiation, the 412-keV γ rays from the gold backing were measured for 3 d with an HPGe detector with the relative efficiency of 100 %, and then the α rays from ^{210}Po nuclei in the bismuth sample were measured with a Si surface barrier detector for several months.

3. Data Processing

Figure 2 shows a normalized incident neutron energy spectrum, $\eta(E_n)$, which was derived from a TOF spectrum observed with the ^{6}Li-glass detector. An average capture cross section of ^{197}Au, $<\sigma_{Au}>$, was defined as follows, using $\eta(E_n)$:

$$<\sigma_{Au}>=\int \sigma_{Au}(E_n)\eta(E_n)dE_n \,, \qquad (1)$$

where $\sigma_{Au}(E_n)$ is the standard capture cross section of ^{197}Au taken from ENDF/B-VI[1] and is shown in Fig. 2. The decay curve of radioactivity of ^{198}Au was obtained from the measurement of 412-keV γ rays, and the radioactivity, A, at the end (t' = 0) of neutron irradiation was determined from the decay curve. Then, the neutron flux, ϕ, was obtained from the following relation:

$$A = N <\sigma_{Au}> \phi(1-e^{-\lambda T_{irr}}) \,, \quad (2)$$

where λ is the decay constant of ^{198}Au, N the number of ^{197}Au nuclei, and T_{irr} the irradiation time.

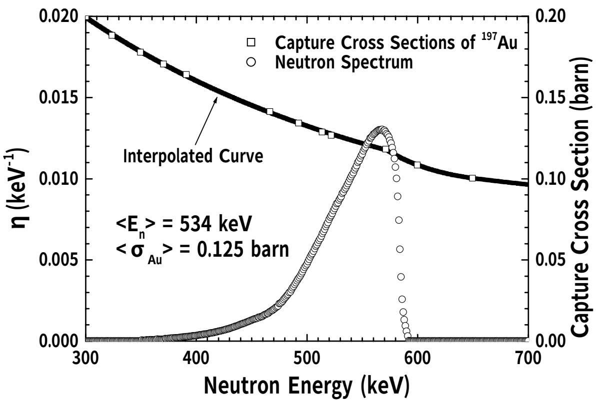

Fig. 2 Normalized incident neutron spectrum at the 534 keV experiment and the standard capture cross sections of ^{197}Au

The number of ^{210}Po nuclei was obtained by solving the following differential equations:

$$\frac{dN_{Bi}(t)}{dt} = n <\sigma> \phi - \lambda_\beta N_{Bi}(t) \,, \qquad (3)$$

$$\frac{dN_{Po}(t)}{dt} = \lambda_\beta N_{Bi}(t) - \lambda_\alpha N_{Po}(t) \,, \qquad (4)$$

in the period of irradiation, and after irradiation,

$$\frac{dN_{Bi}(t')}{dt'} = -\lambda_\beta N_{Bi}(t') \,, \qquad (5)$$

$$\frac{dN_{Po}(t')}{dt'} = \lambda_\beta N_{Bi}(t') - \lambda_\alpha N_{Po}(t') \,, \qquad (6)$$

where N_{Bi} and N_{Po} are the numbers of ^{210g}Bi and ^{210}Po nuclei, respectively, n the number of ^{209}Bi nuclei, λ_β and λ_α the decay constants of ^{210g}Bi and ^{210}Po, respectively, and <σ> the average cross section for the ^{210}Po production. Assuming that ϕ and n were constant, these equations were solved and the α activity of ^{210}Po after irradiation, $A_{Po}(t') = \lambda_\alpha N_{Po}(t')$, was obtained as follows:

$$A_{Po}(t') = n <\sigma> \phi \frac{\lambda_\alpha}{\lambda_\alpha - \lambda_\beta}\left\{(1-e^{-\lambda_\beta T_{irr}})e^{-\lambda_\beta t'} - \frac{\lambda_\beta}{\lambda_\alpha}(1-e^{-\lambda_\alpha T_{irr}})e^{-\lambda_\alpha t'}\right\}. \qquad (7)$$

By comparing Eq.(7) with the growing and decaying curve of the α activity of ^{210}Po obtained from the measurement of α rays, the cross section, <σ>, was derived with the α-ray detection efficiency, ε, described below.

The ^{209}Bi samples had medium thickness (about 5 μm) in comparison with the range (about 15 μm) of 5.3-MeV α rays from ^{210}Po. A thin sample might be better than the present medium sample to make α-ray spectroscopy with good energy resolution, but it must have the disadvantage of poor statistics. In contrast, a thick sample might have the advantage of good statistics, but it must cause continuous α-ray spectra whose shapes depend on the surface condition of thick sample. From the above consideration, the medium thickness was chosen in the present study. Monte-Carlo simulation was performed to check the sample thickness and determine the α-ray detection efficiency. **Figure 3** shows the simulated and measured α-ray spectra at the 534-keV experiment. The simulated spectrum is in good agreement with the measured one.

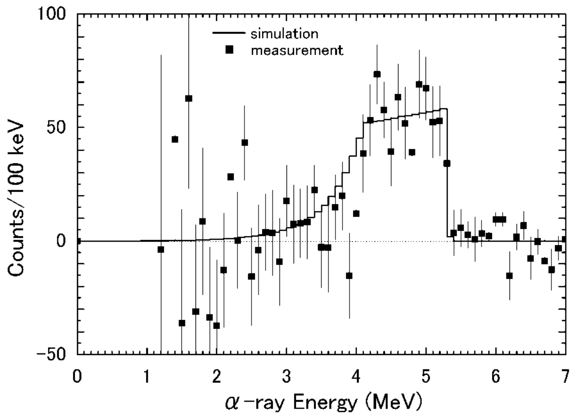

Fig. 3 Comparison between simulated and measured α-ray spectra

The ^{210m}Bi production cross sections were determined as the differences between the ^{210}Po production cross sections and the corresponding total capture cross sections which were previously measured by a prompt γ-ray detection method in our laboratory.

4. Results and Discussion

The derived ^{210}Po and ^{210m}Bi production cross sections are listed in **Table 1**, and are compared with a previous measurement[2] and the evaluation of JENDL Activation cross section File (JENDL/A-96)[3] in **Fig. 4**. The present result of the ^{210}Po production cross section at 30 keV is in agreement with the previous measurement at 24 keV of Booth *et al.*[2], but is about a quarter of the evaluation of JENDL/A-96. Booth *et al.* adopted a double ratio-comparison method with the standard of ^{127}I, using a Sb-Be photoneutron source for the 24-keV neutrons and a reactor neutron source for the thermal neutrons. They detected the β-rays from ^{210g}Bi and ^{128}I, separately. The present result of the ^{210}Po production cross section at 534 keV is the first experimental one around 500 keV, and is about a half of the evaluation

Table 1 ^{210}Po and ^{210m}Bi production cross sections

Neutron Energy (keV)	Cross Sectrions (mb) ^{210}Po	Cross Sectrions (mb) ^{210m}Bi
30	1.94±0.39 (20%)	0.65±0.40 (61%)
534	0.62±0.14 (22%)	0.76±0.16 (21%)

of JENDL/A-96. As for the ^{210m}Bi production cross section, the present result at 30 keV is about one third of the evaluation of JENDL/A-96. The present result at 534 keV is in agreement with the evaluation of JENDL/A-96 within the error, but this agreement seems to be accidental. **Figure 5** shows the branching ratios derived from the present results and the evaluations of JENDL/A-96. The branching ratios derived from the present results are smaller than those from the evaluations by about 30 to 50 %, but both the neutron-energy dependences seem to agree with each other.

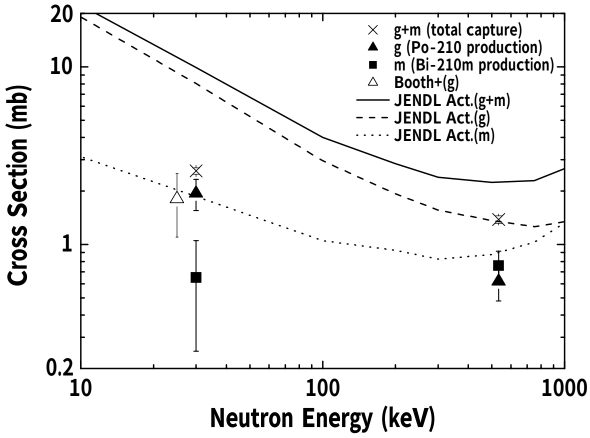

Fig. 4 ^{210}Po and ^{210m}Bi production and total capture cross sections in the keV region

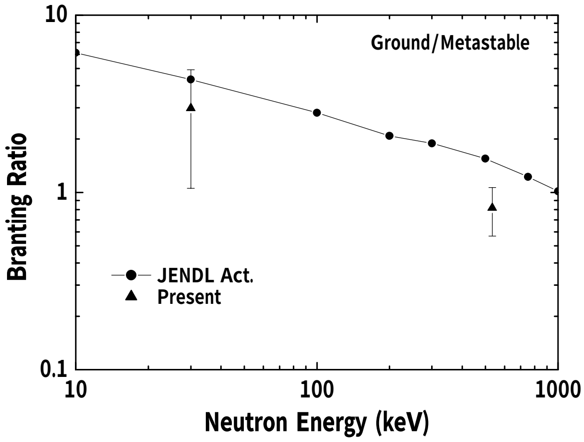

Fig.5 Branching ratios derived from the present results and the JENDL/A-96

5. Conclusion

The ^{210}Po and ^{210m}Bi production cross sections were derived at the average incident neutron energies of 30 and 534 keV. The derived cross sections were a half to a quarter of the evaluated values of JENDL/A-96 except for the ^{210m}Bi production cross section at 534 keV. The branching ratios derived from the present cross sections were smaller than those from the JENDL/A-96 by about 30 to 50 %, but both the neutron-energy dependences were similar.

The present results are important for the evaluation of radioactivity of ^{210}Po and ^{210m}Bi produced in a system with LBEC. The present small values for the ^{210}Po production cross sections might relieve the ^{210}Po problem of LBEC.

References

[1] "ENDF/B-VI data file for ^{197}Au(MAT=7925)," evaluated by Young, P.G. (1984).

[2] Booth, R., Ball, W.P., MacGregor, M.H.: *Phys. Rev.*, **112**, 226 (1958).

[3] JENDL Activation cross section File 96(JENDL/A-96) for ^{209}Bi, evaluated by Yamamuro, N. (1989).