Technical Information

Neutron Flux Gradient of the VT-2 Hole of the JRR-2 Reactor[†]

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1. Introduction

It is known that the neutron flux in a reactor is not uniform throughout the reactor but decreases with the distance from the center. The neutron flux gradient is much higher near the reflector than that in the core of the reactor. Generally, the flux gradient near the D₂O reflector for a large core is known to be no more than 5% across a 2.5 cm irradiation can.

It is desired for the neutron activation analysis to use the irradiation hole with the uniform neutron distribution. The selection of the irradiation hole is also important in other aspects, such as neutron flux, neutron spectra and the temperature elevation during irradiation.

We originally planned to perform the activation analysis of mercury in silicate rocks via the step by step heating method¹⁾. Because of the extremely low abundance of Hg in rocks and the high volatility of the element in various forms, the irradiation at a high neutron flux and at low temperature is highly desired. The neutron flux at the irradiation holes VT-2 and VT-9 of the JRR-2 reactor of the JAERI (Japan Atomic Energy Research Institute) is high enough for the analysis. Moreover, the holes have an equipment of water cooling reservoir during sample irradiation.

In our experiment samples were irradiated in the hole VT-2 of the JRR-2 under the condition of about 50 to 60°C for the period of 180 to 290-hrs at a neutron flux of $3.5 \times 10^{13} \text{ n/cm}^2 \cdot \text{sec}$.

It is noted that the specific activities of 2 or 3 Hg standards varied by about 20% in almost every irradiation. First we attributed this to the problems caused by the standard itself, i.e., by the instability of dilute Hg standard solution, by losses from the evaporation of standard solution, from heat seal and losses during irradiation²⁾.

After elimination of possible causes, the same degree of the neutron flux gradient was observed. This effect may be due to a characteristics of perturbed flux distribution of this hole. Though the whole understanding of the effect remains unsolved, we report the observed neutron flux gradient at the VT-2 hole as well as that at other hole in the reactor. The variation of neutron flux at the hole in other reactors was also investigated.

2. Experimental

The neutron flux variation in a 2.8 cm dia. aluminum irradiation can was measured by comparing the specific activities of the Hg standards (¹⁹⁷Hg and ²⁰³Hg) and/or iron flux monitors (⁵⁹Fe).

The Hg standards were prepared as follows. Ten grams of metallic Hg was dissolved in 30 ml of 2:1 nitric acid solution under heat. The solution was diluted to

[†] JRR-2 原子炉の VT-2 照射孔における中性子束の勾配

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1000 ml with 0.5 N nitric acid solution. The standard solution was prepared by dilution of 10 ml of the above solution to 1000 ml with 0.5 N nitric acid solution. Ten milligrams of the standard solution contains 1 μg of Hg. A series of known amounts of standard solutions, ranging 50 to 90 mg, were weighed and sealed in a 0.4 cm I.D. $\times$ 5.5 cm quartz vial with 0.05 cm of wall thickness.

At first the standard solutions in vials were carefully evaporated at 50°C. Later the standard solutions were directly irradiated without evaporation to avoid possible losses during the process.

About 3.2 mg each of iron wire was weighed with a microbalance and was sealed in a quartz capillary tube with 0.1 cm diameter. Each iron flux monitor was attached to the outside of standard vials. The Hg standard vials with iron flux monitor, wrapped with aluminum foils, were placed in the fixed position in a 2.8 cm dia. aluminum can and were irradiated in the VT-2 hole of the JRR-2 reactor for 180 to 290 hrs at a neutron flux of $3.5 \times 10^{13} \text{ n/cm}^2\text{-sec}$. This reactor is the same type as the CP-5 reactor in Argonne, U.S.A., which is of an enriched-fuel and heavy water type.

As seen in Fig. 1, the VT-2 hole, as well as other nine holes, is situated in heavy water reflector and has a Cd ratio of 21 to 25.

Fig. 1 Position of the irradiation holes of VT-2 and VT-4 of the JRR-2 reactor.

The water tank of 132 cm $\times$ 10 cm dia. was fitted to the hole of VT-2. The irradiation can was placed in the cage of 6 cm dia. and soaked in the water tank. The whole samples in the can were submerged into water through three small holes on both top and bottom of the aluminum can. The irradiation was performed in water at low temperature.

After cooling for five days from the end of irradiation, the outside of standard vials and iron flux monitor tubes were washed with a hot nitric acid solution. The standard vials were weighed. No significant change in weight were observed after irradiation.

A 60 cc Ge(Li) detector coupled with a Northern Scientific 4000 channel pulse-height analyzer was used for the radioactivity

Fig. 2 The neutron flux variation across 2.8 cm dia. aluminum can at the hole VT-2 of the JRR-2 reactor; measured from (a) the relative specific activities of Hg standards (run #12), (b) The relative specific activities of four Hg standards (vial Nos. 6, 7, 8 and 9) and nine iron flux monitors (run #14). Silicate samples in vial Nos. 1, 2, 3, 4 and 5.

measurement. The γ -ray photopeaks at 77 keV of ^{197}Hg , 279 keV of ^{203}Hg for mercury standard and 1,095 keV of ^{59}Fe for iron flux monitors were measured.

The neutron flux variations were also measured for the VT-4 hole of the same reactor, the VG-7 hole of the JRR-3 and the F-21 hole of the TRIGA reactor at Rikkyo University.

3. Results and Discussion

3.1 Neutron flux variation at the VT-2 hole

A typical example (run #12) of the neutron flux variation observed in the VT-2 irradiation hole is shown in Fig. 2a.

A great discrepancy was observed in the specific activities of one standard to another. The specific activity is the highest at position #1 and gradually goes down to the minimum at position #6. Though the exact positions for #12 and #13 are not certain, the values of 0.84 and 0.91 are just between the maximum and the minimum value. The standard #5 and #14 were used for different experiment. The specific activity variation does not correlate with the weights of

standard solutions and quartz vials and thickness of quartz wall.

Prior to the above irradiation, the same fourteen standards were first irradiated at the F-21 hole of the TRIGA II reactor at Rikkyo University for 15 hrs at a neutron flux of $1.5 \times 10^{12} \text{ n/cm}^2 \cdot \text{sec}$.

The neutron flux variation at the F-21 hole is listed in Table 1 along with that of the VT-2 hole.

As seen in Table 1, the specific activities of the fourteen standards for the irradiation at the TRIGA reactor, were almost identical with the deviation of about 2%. This precludes possible errors in weighing the standard solution and losses during the short irradiation. The flux depressions caused by both silica wall and different amount of Hg were of very little effect. Then these standards were irradiated at the VT-2 hole for 183 hrs at a higher neutron flux.

A great discrepancy was observed in the specific activities of the standards.

The result for another irradiation (run #14) is shown in Fig. 2b. Four mercury standards and five silicate rock samples

Table 1 Specific activity variation of Hg standard solutions irradiated at the F-21 and VT-2 holes

Vial No.	Weight of std. soln. (mg)*	Weight of Quartz vial (g)	Specific activities of Hg standards		
			F-21 (TRIGA II) (count/ μg)	VT-2 (JRR-2)*** (count/ng)	Relative value
1	43.6	1.04	787 $\pm$ 14	2,083 $\pm$ 23	1.00
2	62.3	1.00	780 $\pm$ 11	2,028 $\pm$ 19	0.97
3	60.5	0.99	802 $\pm$ 12	1,899 $\pm$ 19	0.91
4	36.8	1.02	760 $\pm$ 15	1,777 $\pm$ 24	0.85
5	69.1	1.02	772 $\pm$ 11	...	...
6	55.6	1.03	779 $\pm$ 12	1,664 $\pm$ 18	0.80
7	79.7	1.07	794 $\pm$ 10	1,712 $\pm$ 15	0.82
8	67.9	1.04	771 $\pm$ 11	1,749 $\pm$ 17	0.84
9	65.7	1.10	786 $\pm$ 18	1,878 $\pm$ 18	0.90
10	60.7	1.10	769 $\pm$ 12	1,997 $\pm$ 19	0.96
11	63.2	1.61**	773 $\pm$ 11	2,016 $\pm$ 19	0.97
12	71.2	1.63**	743 $\pm$ 10	1,740 $\pm$ 16	0.84
13	71.5	1.55**	761 $\pm$ 11	1,890 $\pm$ 17	0.91
14	57.5	1.30**	762 $\pm$ 12	...	...
			av. 774 $\pm$ 15	av. 1,869 $\pm$ 140	

* Ten milligrams of the standard solution contains 1 μg of Hg.

** Quartz vial wall is 0.1cm thick.

*** Run #12.

together with iron flux monitors were irradiated for 237 hrs at the VT-2 hole. The observed specific activity variations for both mercury (^{197}Hg and ^{203}Hg) and iron (^{59}Fe) are quite similar as the result of the previous irradiation (run #12).

After careful examination of the data for the previous irradiations at the hole, the same tendency was observed in almost every irradiation. The observed variation is systematic and appears as if the irradiation were performed at an anisotropic neutron flux.

Interpretation of the above experimental results are summarized as follows; 1) by anisotropic thermal neutron flux, 2) by epithermal or fast neutron, 3) by self-shielding, 4) by interfering nuclear reactions, 5) by the thermal neutron enhancement and 6) by unsymmetrical shielding from surrounding materials⁹⁾.

None of the above flux perturbation effect can reasonably explain the observed great flux gradient up to 20% across a 2.8 cm can.

3.2 Neutron flux variation at the VT-4 hole

Result obtained from the VT-4 hole irradiation at the JRR-2 for 265 hrs is shown in Fig. 3. The hole has a neutron flux of $2 \times 10^{13} \text{ n/cm}^2 \cdot \text{sec}$ with the Cd ratio of 20 to 25.

Fig. 3 The neutron flux variation at the hole VT-4 of the JRR-2 reactor measured from the specific activities of eight iron fluxmonitors (run #13).

Table 2 Neutron flux variation at the VG-7 hole of the JRR-3, estimated from the measurements of the specific activities of Hg-203 and Fe-59

Vial No.	Content of vial	Relative specific activities*	
		Hg-203 (standard)	Fe-59 (flux monitor)
1	at the bottom of the can	...	0.97
6	" "	...	0.96
7	" "	...	0.99
8	" "	...	1.01
9	" "	...	1.00
10	" "	...	1.05
11	" "	...	1.01
12	aqueous standard	0.99	0.99
13	" "	1.00	1.00
14	" "	1.01	1.02
			av. 1.00 ± 0.03
2	6.5cm upper position	...	0.92
3	" "	...	0.93
4	" "	...	0.92
5	" "	...	0.92
15	aqueous standard	0.94	0.95
16	" "	0.94	0.93
			av. 0.93 ± 0.01

* Specific activities (count/g) of Hg-203 and Fe-59 are normalized to the vial No. 13.

Counting errors were less than 1%.

For the lack of cooling system, this hole precludes the irradiation of liquid standards.

The specific activity variation for eight iron flux monitors attached to the outside of quartz vial was fairly uniform but a little gradient was observed. The value of the flux gradient, up to 5% in 2.8 cm dia. can, may be real and quite reasonable for this type of reactor.

3-3 Neutron flux variation at the VG-7 hole

The JRR-3 is a natural uranium heavy water type reactor. The VG-7 hole at the RI train in the graphite reflector has a neutron flux of 2×10^{12} n/cm²·sec and the Cd ratio is approximately 150. The liquid standards of Hg and iron flux monitors were irradiated at the hole for 280 and 285 hrs.

Dimension of the can was 2.5 cm I.D. $\times$ 13 cm in length. For the irradiation, samples can be placed both at the bottom and the upper part.

The neutron flux gradient at this hole was insignificant (Table 2). The vertical variation showed about 7% difference in 6.5 cm apart. Result of another separated run also supported the above observation.

4. Conclusion

The conclusion of this study is summarized as follows:

- 1) The neutron flux gradient at the VT-2 hole of the JRR-2 reactor was significantly high. This hole is not suitable for the neutron activation analysis, though the cause of the effect is not understood.
- 2) The normal and unperturbed neutron flux was observed at the VG-7 hole of the JRR-3 reactor.

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