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The Application of the Zirconium to the Thermal Neutron Fluence Monitoring at the Irradiation Experiments of the Research Reactor

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A determination procedure for the thermal neutron flux using zirconium is proposed. The zirconium foils and cobalt wires are irradiated simultaneously at the irradiation hole of HANARO in order to measure the thermal neutron flux. It is confirmed that the handling of a zirconium sample is much more convenient than that of the cobalt wire in the processes of sample preparation, irradiation and sample fixing for gamma-ray detection. The activation characteristics of the cobalt and zirconium are investigated, and the gamma-ray spectrum from the irradiated zirconium is analyzed. It is confirmed that the two gamma-ray peaks at 724 and 757 keV from Zr-95 are useful for the flux measurements in the case of an irradiation for several hours at the neutron flux of ~3×10^{13} n/cm^{2}s. The thermal neutron flux measured within 8 days from the zirconium irradiation are slightly smaller than that after 8 days because of the effects of a high count rate and the interference of the gamma-rays from Nb-97. The neutron fluxes measured by zirconium are 15.2~16.7% larger than those by cobalt. Anyhow, the ratio of the flux measured by the cobalt to that by the zirconium can be determined. Therefore, the equivalent thermal neutron fluence for cobalt can be obtained by using the neutron flux determined by zirconium and the flux ratio. The uncertainty in the neutron flux determination is within 1.8%, and it is a little larger than that determined by cobalt only. However, if this uncertainty level is acceptable, it is expected that the above procedure for the thermal neutron flux measurement is very useful when a number of monitors are needed in the irradiation experiments of the research reactors.

KEYWORDS: thermal neutron flux, zirconium, cobalt, HANARO, activation characteristics, neutron fluence, irradiation experiments, research reactors

1. Introduction

An accurate thermal neutron fluence is required for the neutron irradiation applications of research reactors, and it is deduced from the thermal neutron flux and irradiation time. In general, the thermal neutron fluence can be measured by detecting the gamma-rays from the activated thermal neutron flux monitors. In choosing the material for an activation detector, there are several general considerations concerning the nuclear properties of a material such as, the shape and magnitude of the activation cross-section, characteristics of the induced activities, interfering activities and the physical properties[1,2]. In addition, the procedures

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related to the preparation and handling of the sample must be simple and convenient. Gold, cobalt and their alloys with aluminum are widely used in monitoring the thermal neutron fluence because their cross-sections, gamma-ray emission probabilities and half-lives of activities are precisely determined[3,4]. But, when they are used in the fluence range of ~10^{17} n/cm^2, they have several problems. Gold is not proper in this range because of a relatively large activation cross-section and the very short half-life of its daughter nuclei. Cobalt is useful, but it must have a very small size and mass. So, when a number of samples are required in the irradiation experiments, the treatment processes, such as weighing, are inconvenient and painstaking. Its alloy with aluminum can be used instead of pure cobalt, but the uncertainty for the composition is unavoidable.

Zirconium is the stable material which is used in various reactor structures. It has a relatively small radiative capture cross-section. So, the zirconium sample can be large, and it is expected that the manipulation of the zirconium sample is relatively easy.

The analysis tool for the two gamma-rays from Co-60, which is the induced activity of the cobalt, is set up well using the HPGe detector[5]. But, it can be supposed that the uncertainty in the analysis of the induced activity from the zirconium is much larger than that from the cobalt sample.

Therefore, in order to obtain the neutron fluence efficiently in the irradiation experiments of the research reactors, a determination procedure of the neutron flux using zirconium is proposed.

2. Experiments

In order to measure the neutron flux by the activation method with a small uncertainty, it is necessary to determine accurately the peak area of the gamma-ray of interest, the detection efficiency of the gamma-ray detector and the sample weight. And also, the uncertainties of the abundance of each isotope, the neutron cross-sections, the gamma-ray emission probabilities and the half-lives of the decay activities are as small as possible.

From the result of the micro-balance test, it is confirmed that the weighing uncertainty is very small in the range of several hundred μg. The HPGe detector used in the activity measurement of the irradiated sample is calibrated with the standard sources such as Co-60, Eu-152 and Cs-137, etc. The uncertainty of the efficiency calibration is within 1.2%. In the analysis of the gamma-ray spectrum obtained with the HPGe detector, the uncertainty in the determination of the gamma-ray peak area is smaller than 0.5%[6].

The zirconium foils with a thickness of 0.127 mm and mass of 10~50 mg are irradiated at the thermalized irradiation hole of the HANARO, 30 MW research reactor. The cobalt wires with diameters of 0.05 and 0.381 mm, and a mass of 0.2~0.8 mg are irradiated at the same time as the zirconium. The irradiation time and sample weight are varied in order to obtain the appropriate activity for the gamma-ray detection. When the gamma-rays from the irradiated sample are detected, the source-to-detector distance is about 25 cm. In the various processes of the sample preparation, irradiation and sample fixing for the gamma-ray detection, it is confirmed that the handling of the zirconium sample is much more convenient than that of the cobalt wire.

3. Characteristics of the Irradiated Zirconium

The activation characteristics of the cobalt and zirconium are represented in Table 1. The

region of interest for the gamma-ray spectrum obtained from the irradiated zirconium sample with the HPGe detector is shown in figure 1. The weight of the sample is 15.522 mg, and the irradiation, cooling and collection(live) time is 18076, 73024, and 4700 seconds, respectively.

Table 1 Activation characteristics of the cobalt and zirconium[7,8].

Isotope	Abun-dance [%]	Radiative capture cross-section			g-factor	Induced activity	Half-life
		at 0.0253 eV	Max. avg. at 0.0253 eV	Resonance integral			
Co-59	100	37.18 b	32.96 b	75.51 b	1.0005	Co-60 (Co-60m)	5.28y (10.4m)
Zr-90	51.45	11.12 mb	9.862 mb	173.6 mb	1.0003	Zr-91	stable
Zr-91	11.22	1.247 b	1.106 b	6.943 b	1.0003	Zr-92	stable
Zr-92	17.15	229.2 mb	203.2 mb	713.9 mb	1.0003	Zr-93	1.53×10^6 y
Zr-94	17.38	49.81 mb	44.16 mb	320.7 mb	1.0003	Zr-95	64.02 d
Zr-96	2.80	22.80 mb	20.22 mb	5.863 b	1.0007	Zr-97	16.90 h

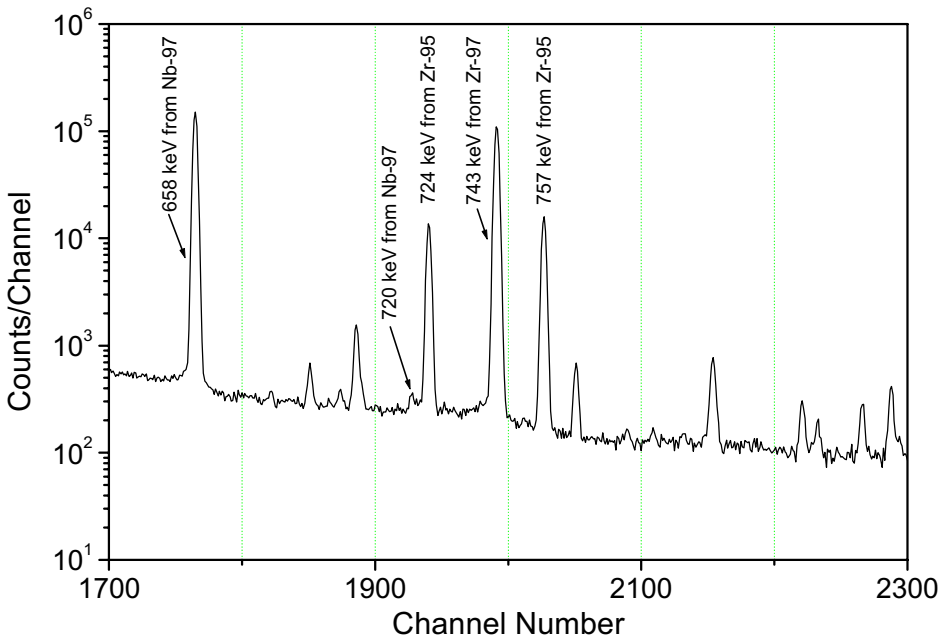

Fig. 1 The region of interest for the gamma-ray spectrum obtained from the irradiated zirconium.

As shown in figure 1, the induced activities of the zirconium by neutron irradiation are Zr-95, Zr-97 and their daughters such as Nb-95 and Nb-97. In the case of an irradiation for several hours at the neutron flux of ~3×10^{13} n/cm^{2}s, the main peaks of the measured gamma-ray spectrum from the irradiated zirconium are located at the energies of 658, 724, 743 and 757 keV. The 743 keV peak is not proper for the flux determinations due to the overlap of the gamma-rays from Zr-97 and Nb-97m. Also, the 658 keV peak from Nb-97 is not suitable because of the short half-life of Nb-97. Therefore, the two peaks at 724 and 757 keV from Zr-95 are applicable for the flux measurements. When the 724 keV peak from Zr-95 is analyzed, the interference of the 720 keV gamma-rays from Nb-97 which is the

daughter of Zr-97 must be considered. Since the half-life of Zr-97 is 16.9 hours, and it is much shorter than that of Zr-95, it is negligible if the cooling time is long.

Figure 2 shows the dead time of the gamma-ray detection system measured as a function of the cooling time of the zirconium sample. The irradiation time of the zirconium sample is settled so that the peak area of 724 keV gamma-rays from Zr-95 may become 54000 during the detection time of 2400 seconds. Also, in the figure, the dead time anticipated from the decay curves of Zr-95 and Zr-97 is represented. As shown in the figure, in an early detection within 3 days, the gamma-rays which mainly contribute to the dead time originate from Zr-97, and after 5 days from the irradiation, the gamma-rays from Zr-95 are the main contributions to the total count rate. If an early detection for the gamma-ray is required, the detection after 48 hours from the irradiation is possible without a large uncertainty.

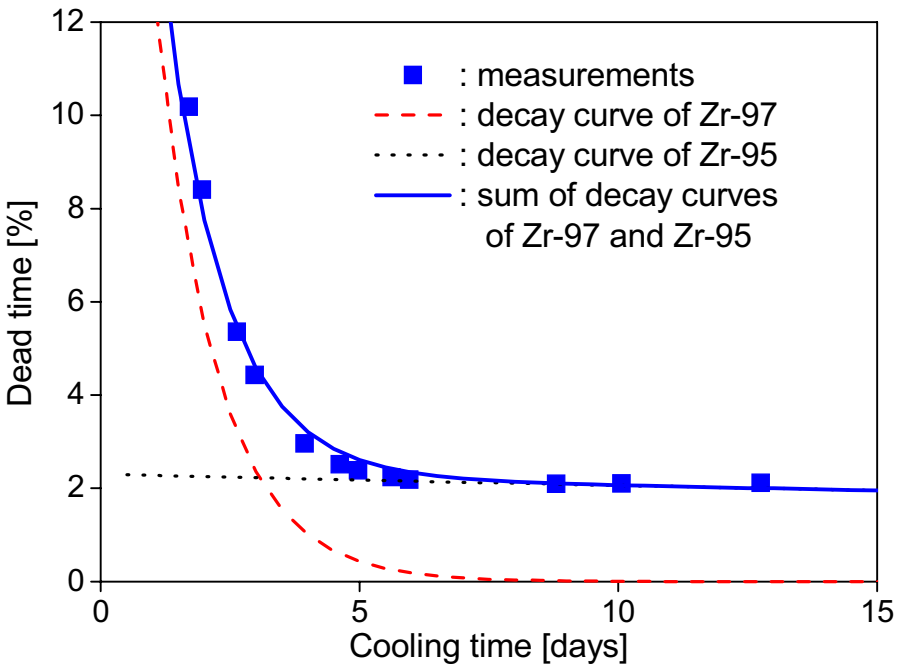

Fig. 2 Measured and anticipated dead times of the gamma-ray detection system as a function of the cooling time of the zirconium sample.

4. Determination of the Thermal Neutron Flux Using Zirconium

Table 2 represents the thermal neutron fluxes measured by the cobalt and zirconium samples irradiated at the same conditions. The neutron fluxes measured by zirconium are 18.4~20.2% larger than those by the cobalt with a diameter of 0.381 mm, and they are 15.2~16.7% larger than those by the cobalt with a diameter of 0.05 mm. The difference related to the diameter of the cobalt wire is caused by the self shielding effect. From the experiments to investigate the self shielding effect of the cobalt wire, it is confirmed that the neutron flux determined by the cobalt wire with a diameter of 0.381 mm is underestimated by 4.94%. The self shielding effects of the 0.05 mm cobalt wire and zirconium foil with a thickness of 0.127 mm are negligible.

The neutron flux measured by using the zirconium foil is higher than that by the cobalt wire. It is supposed that the difference comes from the contributions of the epithermal and fast

neutrons to the reaction rate or the uncertainties of the nuclear and atomic data of the zirconium. The origin of the difference between the neutron flux determined by the cobalt and that by the zirconium is currently being analyzed in detail. Anyhow, we can determine the ratio of the flux measured by the cobalt to that by the zirconium. Therefore, we can obtain the equivalent thermal neutron fluence for the cobalt using the neutron flux determined by the zirconium and the ratio.

Table 2 Measured thermal neutron fluxes by the cobalt and zirconium samples irradiated in the same conditions.

Sequential number	Neutron flux determined by Co [a)] [n/cm^{2}s]		Neutron flux determined by Zr[b)] [n/cm^{2}s]	b)/a)
	Co wire with ϕ=0.381 mm	Co wire with ϕ=0.05 mm		
1	3.223×10^{13}		3.815×10^{13}	1.184
2	3.217×10^{13}		3.847×10^{13}	1.196
3	3.164×10^{13}		3.803×10^{13}	1.202
4		3.105×10^{13}	3.577×10^{13}	1.152
5		3.118×10^{13}	3.638×10^{13}	1.167
6		3.142×10^{13}	3.654×10^{13}	1.163

Figure 3 represents the thermal neutron flux determined by the zirconium sample as a function of the cooling time.

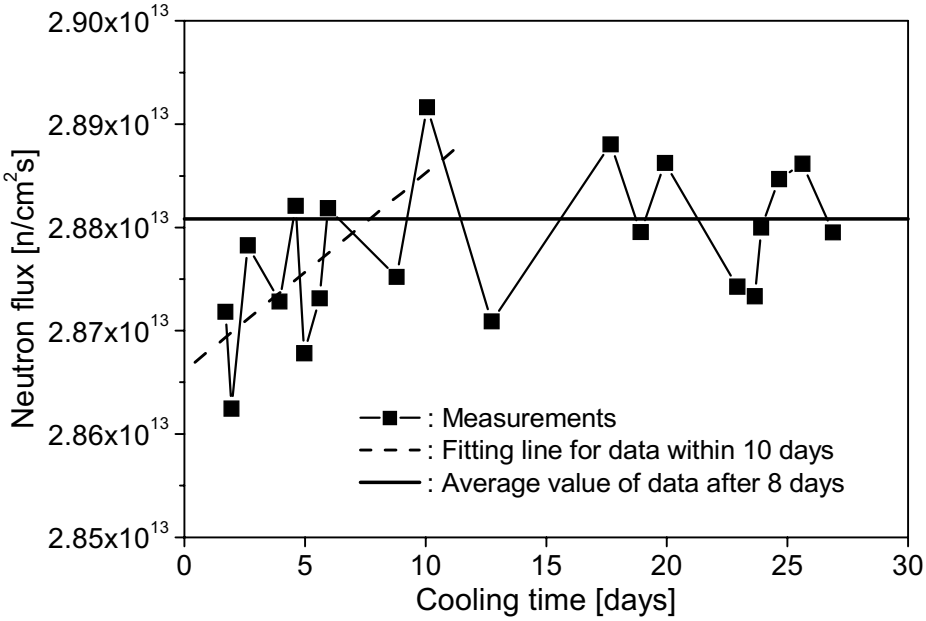

Fig. 3 Thermal neutron flux determined by the zirconium sample as a function of the cooling time.

The measurements within 8 days are slightly smaller than those after 8 days because of the effects of a high count rate and the interference of the gamma-rays from Nb-97. So, it is preferable to have a cooling time of more than 8 days.

The uncertainty in the neutron flux determination is within 1.8%, and it is a little larger

than that determined by the cobalt only. However, if this uncertainty level is acceptable, it is expected that the above procedure for the thermal neutron flux measurement is very useful when a number of flux monitors are needed in the irradiation experiments of the research reactors.

5. Conclusion

A determination procedure for the thermal neutron flux using the activation method with zirconium foil is proposed. Although the uncertainty in the neutron flux determination using zirconium and cobalt is a little larger than that determined by cobalt only, it is expected that the above procedure for the thermal neutron flux measurement is very useful when a number of monitors are needed in the irradiation experiments of the research reactors.

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