Measurement of Resonance Integral of the $^{90}Sr(n, \gamma)^{91}Sr$ Reaction

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To obtain fundamental data for research on nuclear transmutation method of radioactive wastes, the resonance integral (I_0) of the $^{90}Sr(n, \gamma)^{91}Sr$ reaction was measured with an activation method.

1. INTRODUCTION

Accurate data of the thermal neutron capture cross section (σ_0) and the resonance integral (I_0) are required for the research on nuclear transmutation of fission product (FP) nuclides with large fission yields and long half-lives, so that the present authors have measured the σ_0 and the I_0 of FP. ^{90}Sr is one of the most important nuclides in the low-level radioactive nuclear wastes, and remains for long period of time because of its long half-life (28.8 yr). However, the cross section data of ^{90}Sr have not been prepared sufficiently. There was only a value of upper limit on I_0 which was measured by Harada *et al.* in 1994 [1]. They measured σ_0 and I_0 of the $^{90}Sr(n, \gamma)^{91}Sr$ reaction by an activation method in the research reactor JRR-4 at JAERI. The upper limit of I_0, however, was only obtained in that experiment The reason for this is as follows: the ratio of the epithermal flux component to total neutron flux was so small (e.g. 0.033 ± 0.007 [1]) at the irradiation position in JRR-4 that the sufficient yields of ^{91}Sr were not obtained in case of the irradiation within a Cd capsule. It was found that it was possible to precisely measure I_0 of ^{90}Sr by using more harder neutron filed in Kyoto University Reactor (KUR) than that in JRR-4, and then the measurement of I_0 for the $^{90}Sr(n, \gamma)^{91}Sr$ reaction was planed once more.

2. EXPERIMENTS

$SrCl_2$ solution containing 3.7×10^5 (Bq) of ^{90}Sr was poured into a high purity quartz tube, which was 8mm in diameter and 100mm in length. $SrCl_2$ solution containing 900 (Bq) of ^{85}Sr was also added into the tube. Strontium-85 was used as a tracer for ^{90}Sr because ^{85}Sr was a γ-ray emitter. After drying of the solution, the tube was shaped into an

ampoule that was 25mm in length.

A target was constructed from the Sr ampoule and flux monitors, i.e. Co/Al and Au/Al alloy wires. Wires of Co/Al alloy (Co: 0.46 wt%, 0.381 mm in diameter) and Au/Al alloy (Au: 0.112 wt%, 0.510 mm in diameter) were used to monitor the neutron flux at the target position. Because ^{59}Co and ^{197}Au have different sensitivities to thermal and epithermal neutrons, these wires are adequate to determine the thermal and the epithermal fractions of neutron flux The amount of Co and Au contained in wires were determined by weight measurement in a microbalance.

The target was put into an Al capsule, and then the Al capsule was housed in an irradiation capsule. In the case of irradiation to measure I_0, the target was housed in a Cd capsule, which was 10mm in diameter, 26mm in length and 0.5mm in thickness. During the neutron irradiation, the Al capsule have two roles: **(a)** Target confinement and **(b)** heat removal of the Cd capsule. This was confirmed by a test irradiation with no target for 1 hour in hydraulic transfer tube of KUR. Irradiations in hydraulic transfer tube of KUR were performed for 10 hours without and within the Cd capsule, respectively. After the cooling for more than 12 hours, targets were pulled out from the irradiation capsules.

The chemical separations were accordingly performed to eliminate ^{24}Na nuclide from the Sr samples. The irradiated Sr targets was washed with 2.5 mol/l $(NH_4)_2SO_4$ solution, and then Sr nuclides were precipitated as a form of Sr_2SO_4. The Na nuclides were retained in the solvent solution, so that they were removed from the Sr precipitates with a filter, which was 25mm in diameter and 0.1μm in thickness. The filter was wrapped with a vinyl tape, and then used as a measurement sample.

A high purity Ge detector was employed to measure the γ rays from the irradiated targets and monitor wires. Its performance was characterized by a relative efficiency of 90 % to a 7.6 cm×7.6 cmϕ NaI(Tl) detector and an energy resolution of 2.1 keV full width at half-maximum(FWHM) at the 1.33 MeV peak of ^{60}Co. The peak detection efficiencies were calibrated with γ-ray sources, i.e. ^{152}Eu, ^{137}Cs and ^{22}Na, whose activities were well determined. Signals from the detector were fed to a fast data acquisition system, and γ-ray spectra data were recorded on a hard disk of a personal computer. Details of the data taking system were described elsewhere **[2].** The measurement samples were mounted on the surface of the Ge detector. An example of γ-ray spectrum after the chemical processing was shown in **Fig. 1**. As seen in Fig.1, two γ rays revealed themselves around the energy points of 750 keV and 1024 keV. The γ-ray measurements were performed for about 12hours in the case of the irradiated Sr sample within the Cd capsule and for about 15hours in the case of that without the Cd capsule. Spectra data were saved every 3 hours to confirm whether observed γ rays were originated in ^{91}Sr or not. **Fig. 2** shows the decay curves for the counts from the 1024 keV γ-ray peak. The half-life was found to be 9±1(h), which was in agreement with the evaluated half-life value of ^{91}Sr (9.63 h **[3]**) within the limit of errors. Therefore, it was confirmed that the 1024 keV γ ray has originated from ^{91}Sr.

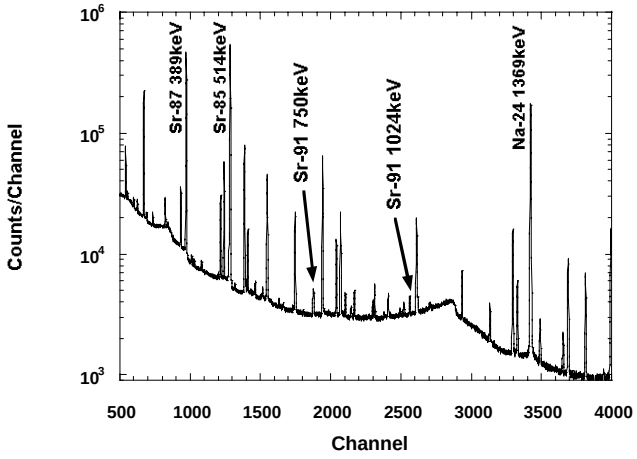

Fig.1 Gamma-ray spectrum of Sr after chemical procedure

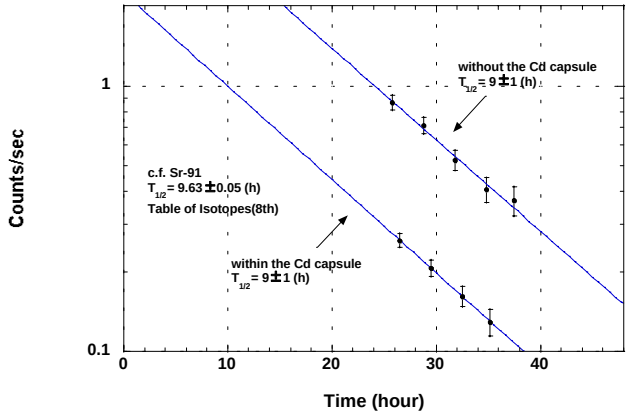

Fig. 2 Decay curves of 1024keV γ-ray emitted form ^{91}Sr

3. ANALYSIS

Since the details of the method that we used to determine the thermal cross section and resonance integral were described elsewhere, here a brief outline of the analysis is only presented.

Westcott's convention **[4]** can be rewritten by using simplified flux notation as follows:

$$R/\sigma_0 = \phi_1 G_{th} + \phi_2 s_0 G_{epi}, \quad (1)$$

for irradiation without a Cd shield capsule,

$$R'/\sigma_0 = \phi_1' G_{th} + \phi_2' s_0 G_{epi}, \quad (2)$$

for irradiation with a Cd shield capsule. Here, the R (or R') is the reaction rate and σ_0 the thermal neutron (2,200m/s neutron) capture cross section; ϕ_1 and ϕ_1' are neutron flux components in the thermal energy region, and ϕ_2 and ϕ_2' are those in the epithermal energy region. The neutron flux components were obtained with flux monitors. The results of the neutron fluxes were listed in **Table 1**.

Table 1 Neutron flux measured in Hydraulic Transfer tube at KUR

	ϕ_1 or ϕ_1' (10^{14} n/cm^2 · sec)	ϕ_2 or ϕ_2' (10^{13} n/cm^2 · sec)
without the Cd	ϕ_1=1.063 ±0.007	ϕ_2 =0.243±0.004
within the Cd	ϕ_1' =0.033±0.001	ϕ_2' =0.404±0.001

The G_{th} and G_{epi} are self-shielding factors to thermal and epi-thermal neutrons, respectively. The G_{th} and G_{epi} are unity in the following analysis in current target conditions. The s_0 is the parameter defined by

$$s_0 = \frac{2}{\sqrt{\pi}} \frac{I_0'}{\sigma_0}, \tag{3}$$

where I_0' is the reduced resonance integral, i.e. the resonance integral after subtracting the 1/υ components. The resonance integral I_0 is calculated as follows:

$$I_0 = I_0' + 0.45\sigma_0, \tag{4}$$

where $0.45\sigma_0$ is the 1/υ contribution given by assuming the Cd cut-off energy to be 0.5eV.

Eqs.(1) and (2) give the relation,

$$s_0 = -\frac{\phi_1 - \phi_1'(R/R')}{\phi_2 - \phi_2'(R/R')}, \tag{5}$$

so that the value of s_0 is obtained from R/R' value of each irradiated target. The σ_0 is derived by substituting the s_0 into Eq.(1), and then the values of I_0' and I_0 are calculated from Eqs.(3) and (4).

4. PRELIMINARY RESULTS AND DISCUSSION

Reaction rates were obtained from the 1024 keV γ-ray count data, and listed in **Table 2**.

Table 2 Preliminary results of reaction rates and cross sections of ^{90}Sr(n,γ)^{91}Sr reaction

Irradiation Type	Chemical yield	^{90}Sr(n,γ)^{91}Sr reaction			
		Reaction rates (1/s)	σ_0 (mb)	s_0	I_0 (mb)
Without the Cd capsule	0.836±0.110	1.559±0.215 ×10^{-12}	**11.7±1.7**	**11.2±2.3**	**121±29**
Within the capsule Cd	0.770±0.025	5.678±0.310 ×10^{-13}			
		References:			
		[1] Harada *et al.*	15.3± $^{1.3}_{4.2}$	≦11	≦160
		[5]Zeisel	800±50		
		[6]McVey *et al.*	14.0±2.4		
		[7]Lone et al.	9.7±0.7		

With the experimental results of the reaction rates and neutron flux, quantity s_0 was derived from Eq.(5), and then the resonance integral was obtained by s_0, and Eq.(3) and (4). The present result for the resonance integral of ^{90}Sr was

found to be 121±29 (mb). By substituting s_0 into Eq.(1), the thermal neutron capture cross section was obtained as 11.7±1.7 (mb). The present results were summarized in Table 2 together with the evaluated data .

The present result for I_0 was found to be 121±29 (mb), and it was within the upper limit reported in Ref.**[3]**. The neutron capture cross section was also reported in Ref.**[3]** as 15.3±1.3 (mb) assuming the quantity s_0 was zero. The σ_0 can be estimated from this value and the present result for s_0, the σ_0 was obtained as 11.2±1.3 (mb). This value was in good agreement with the present result, σ_0 = 11.7±1.7 (mb), within the limit of errors. It was found that the present results were consistent with those in Ref. **[3]**.

5. CONCLUSION

To obtain fundamental data for research on nuclear transmutation method of radioactive wastes, the resonance integral of the $^{90}Sr(n, \gamma)^{91}Sr$ reaction was measured with an activation method. However, there is still a problem which is to perform the chemical procedure more precisely, therefore it should be note that these results were only preliminary ones.

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