Measurements of Thermal Neutron Capture Cross Sections for some FP Nuclides

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The thermal neutron capture cross sections (σ_0) and the resonance integrals (I_0) of some FP elements, such as ^{80}Se, ^{94}Zr, ^{124}Sn, ^{127}I and ^{133}Cs, were measured by the activation and γ-ray spectroscopic method.

1. Introduction

The thermal neutron capture cross sections (σ_0) and the resonance integrals (I_0) of long-lived fission products (FP) is required for the transmutation study by reactor neutrons in the nuclear waste management. The nuclear waste, sometimes, contains a large amount of stable nuclei having the same atomic number as that of long-lived FP. These stable nuclei absorb thermal neutrons during the neutron irradiation of the nuclear waste and disturb the neutron filed near the objective target nuclei; the reaction rate of the target nuclei will be reduced. Also, more radioactive nuclei may be produced by the neutron capture process of the stable nuclei. Therefore, the cross section data are needed for the stable nuclei, such as ^{80}Se, ^{94}Zr, ^{124}Sn, ^{127}I and ^{133}Cs.

2. Experiment

The cross section measurements for those nuclei were performed by the activation and γ-ray spectroscopic method. The high purity(99.99%) natural SeO_2, ZrO_2, SnO_2 and CsCl powders were used as targets. The ^{127}I elements were also used for the target, which were involved in the ^{129}I standardized solution as the contamination, since the measurements were performed together with those for ^{129}I. The adequate amount of each sample was poured into each quartz case. The wires of 0.112wt% Au/Al alloy (0.510mm in diameter) and 0.46wt% Co/Al alloy (0.381mm in diameter) were used as activation detectors to monitor the neutron flux. The method of measuring the neutron flux was the same for the cross section measurements, and will be explained later in **Sec. 3**.

The irradiations were performed in the rotary specimen rack (RSR) of the Research Reactor TRIGA MK-II at Rikkyo University, whose thermal neutron flux was 5.0×10^{11} n/cm^{2}sec and the epithermal index of Westcott's convention[(1)] 0.039. Each target was irradiated without and within a Cd capsule (1mm in thickness, 22mm in outer diameter and 63mm in outer length). The Cd capsule was used to reduce the flux of thermal neutrons at the target.

The yields of γ-rays emitted from the irradiated targets were measured by a high purity Ge detector with a 90% relative efficiency and an energy resolution of 2.1keV FWHM at 1.33MeV of ^{60}Co. The details of the data taking system were described elsewhere[(2)].

3. Analysis

The following equations obtained by modifying Westcott's convention[(1)] were used for analysis to deduce neutron fluxes at the irradiation position and cross sections. Equations based on Westcott's convention are rewritten[(2)] by using simplified flux notation as follows;

$$R/\sigma_0 = \phi_1 + \phi_2 s_0 \tag{1}$$

for irradiation without the Cd shield capsule,

$$R'/\sigma_0 = \phi_1' + \phi_2' s_0 \tag{2}$$

for irradiation with the Cd shield capsule. Here, the R (or R') is the reaction rate, σ_0 the thermal neutron (2,200m/s neutron) capture cross section, and s_0 is defined by

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

where I_0' is the reduced resonance integral after subtracting the 1/v 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/v contribution given by assuming the Cd cut-off energy to be 0.5eV.

Using the known data of the cross sections σ_0 and the parameter s_0 for cobalt and gold, the values of the flux terms $\phi_{1,2}$ and $\phi'_{1,2}$, were determined by solving the simultaneous equations for cobalt and gold from Eqs. (1) and (2). **Table 1** summarized the experimental results of the R and R' values of the flux monitors, and also the neutron flux.

After the deletion of σ_0, Eqs.(1) and (2) give the relation,

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

so that the s_0 value for each target nuclide is obtained from R/R' value of each irradiated target. The value of σ_0 is obtained 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. Results and Discussion

The results of the cross sections were shown in **Tables 2-6** together with the previously reported data.

Selenium-80

The cross sections for the formations of the isomeric state ^{81m}Se and the ground state ^{81g}Se were measured respectively by following the γ-ray counting rate after the irradiation. The present results are listed in Table 2 together with the values reported previously[(3)-(6)]. The present data of

$\sigma_{0,m+g}$ and $I_{0,m+g}$ in the reaction $^{80}Se(n,\gamma)^{81m,g}Se$ support the evaluated ones. The $\sigma_{0,m}$ for the formation of the ^{81m}Se was 43% smaller than the value (0.08 b) reported by Holden et al(4). This reason is owed to the revision of the γ-ray emission probability data from 9.7%(7) to 12.7%(8).

Zirconium-94

The present results are listed in Table 3 together with the data reported previously(9),(10),(11). The σ_0 value for ^{94}Zr supports the evaluated one. The present result of I_0 was 14% smaller than the evaluated one, but close to the value given by Santry et al(10). The ratio of I_0/σ_0 was found to be 5.78 which agreed with the measured value by Moens et al(9). The present values for ^{94}Zr have the consistency in point of the results of the σ_0 value and the I_0/σ_0 ratio. This fact supports the reliability of the present values.

Tin-124

The present results for ^{124}Sn are listed in Table 4. There have been no previously reported values for the reaction $^{124}Sn(n,\gamma)^{125g}Sn$. The measurements of the $\sigma_{0,g}$ and $I_{0,g}$ for the formation of ^{125g}Sn were performed in the present work. However, the γ-ray emission probability data for ^{125g}Sn had 30% error. After now, the accurate measurement of the γ-ray emission probability should be performed by means of β-γ coincidence counting technique(12).

Iodine-127

The present results for ^{127}I are listed in Table 5 with the data reported(13),(14). The value of the thermal neutron capture cross section was 6.40±0.29 b and in agreement with the evaluated and previously reported data within the limit of the error, but not in agreement with the data, 4.7±0.2 b by Friedmann et al(14). The difference between the present result and the result by Friedmann et al. may be caused by the difference of the γ-ray emission probability used for the deduction of reaction rate from the γ-ray peak counts. Friedmann et al. used a value of 0.16 for the emission probability of the 443keV γ-ray from the reaction product ^{128}I. The emission probability of this γ-ray was measured recently by Miyahara et al.(15) by using a two dimensional 4πβ–γ coincidence method, and was determined as 0.1261±0.0008. If the new value of 0.1261 for the emission probability of the 443keV γ-ray is used in the analysis of the data by Friedmann et al., their result of the σ_0 will change to 5.96 b and becomes close to the value obtained in the present analysis.

Cesium-133

The cross sections for the formations of the isomeric state ^{134m}Cs and the ground state ^{134g}Cs were measured respectively by following the γ-ray counting rate after the irradiation. The present results for ^{133}Cs are listed in Table 6 together with the reported data(16)-(20). As shown in Table 6, many authors have ever measured the thermal neutron capture cross sections and the resonance integrals for the formations of the ^{134m}Cs and $^{134m+g}Cs$. With respect to the thermal cross section, the present result agrees with data in references within the limits of the error. The thermal cross section leading to the $^{134m+g}Cs$ agreed with the evaluated value in JENDL-3.2 (29.00 b(3)), and one leading to ^{134m}Cs was in good agreement with the data reported previously, e.g. 2.82 ±0.07 b by Baerg et al(19).

On the other hand, the resonance integral for $^{134m+g}Cs$ was 25% smaller than the evaluated value in JENDL-3.2 (396.2 b(3)).

5. Conclusion

①For the $^{80}Se(n,\gamma)$, $^{94}Zr(n,\gamma)$, $^{124}Sn(n,\gamma)$, $^{127}I(n,\gamma)$ and $^{133}Cs(n,\gamma)$ reactions, the thermal neutron capture cross sections and the resonance integrals were measured to obtain fundamental data for research on the transmutation of nuclear wastes.

②The σ_0 of ^{80}Se, ^{127}I and ^{133}Cs agreed with JENDL-3.2 and previous data. However, the I_0 of ^{94}Zr, ^{127}I and ^{133}Cs were different from JENDL-3.2 and any other data.

③The cross sections for the formations of the isomeric and ground states, such as $^{81m,g}Se$ and $^{134m,g}Cs$, were measured separately.

④The σ_0 and I_0 for the formation of ^{125g}Sn was obtained for the first time.

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Table 1 Summary of the neutron flux measurements in RSR of Rikkyo Reactor

Irradiation Type	Irradiation period	Reaction rates of the flux monitors ^{60}Co	^{198}Au (10^{-11} /s)	ϕ_1 or ϕ_1' (10^{11} n/cm^{2}sec)	ϕ_2 or ϕ_2'
bare	10min	1.76 ±0.04	7.31±0.15	4.42 ±0.09	0.173±0.004
with Cd	25min	0.155±0.003	3.20±0.06	0.099±0.004	0.183±0.004
Cadmium ratio		11.3 ±0.3	2.28±0.07		

Table 2 Results of Cross Sections for the $^{80}Se(n,\gamma)^{81m,\ 81g}Se$ Reactions

Author (Year)	$\sigma_{0,m}$	$\sigma_{0,g}$	$\sigma_{0,m+g}$	$I_{0,m}$	$I_{0,g}$	$I_{0,m+g}$
Present results	0.046±0.002	0.542±0.042	0.588±0.042	0.147±0.005	0.847±0.071	0.994±0.071
JENDL-3.2[3]			0.610			0.9739
Holden[6]('68)	0.08 ±0.01[4]	0.53 ±0.04	0.61 ±0.05[5]			2.0 ±0.6

Table 3 Results of Cross Section for the $^{94}Zr(n,\gamma)^{96}Zr$ reaction

	σ_0 (b)	I_0 (b)	$Q=I_0/\sigma_0$
Present Result	0.0479±0.0010	0.277±0.007	5.78±0.19
JENDL-3.2[3]	0.04981	0.3207	
Moens[9] ('79)	----------	----------	5.78±0.12
Santry[10] ('73)	0.0475±0.0024	0.218±0.010	4.6
Fulmer[11] ('71)	0.052 ±0.003	0.30 ±0.03	5.77±0.67

Table 4 Results of Cross Section for the $^{124}Sn(n,\gamma)^{125g}Sn$ Reaction

	σ_0 (b)	I_0 (b)
Present Results	0.0042±0.0013	0.083±0.025
Reference	(no data)	

Table 5 Results of Cross Section for the $^{127}I(n,\gamma)^{128}I$ Reaction

	σ_0 (b)	I_0 (b)	$Q=I_0/\sigma_0$
Present Results	6.40±0.29	162±8	25.3±1.7
JENDL-3.2[3]	6.200	148.2	
Friedman[14]('83)	4.7 ±0.2	109±5	
Moens[9]('79)			24.6
Ryves [13]('70)	6.12±0.12	145±9	

Table 6 Results of Cross Sections for the $^{133}Cs(n,\gamma)^{134m,\ 134g}Cs$ Reactions

Author (Year)	$\sigma_{0,m}$	$\sigma_{0,g}$	$\sigma_{0,m+g}$	$I_{0,m}$	$I_{0,g}$	$I_{0,m+g}$
Present results	2.70±0.14	26.3±1.1	29.0±1.1	23.2±1.8	275±16	298±16
JENDL-3.2[3]('97)			29.00			396.2
Takiue et al.[16]('78)			28.7±0.7			
Steines [17]('72)						437±26
Sims et al.[18]('68)			29.2±2.3			495±17
Keish et al.[19]('61)	2.44±0.15					
Baerg et al.[20]('60)	2.82±0.07		30.4±0.8	34.4±1.9		461±25