Precise Measurements of Neutron Capture Cross Sections for FP

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The thermal neutron capture cross sections (σ_0) and the resonance integrals (I_0) of some fission products (FP), such as ^{137}Cs, ^{90}Sr, ^{99}Tc, ^{129}I and ^{135}Cs, were measured by the activation and γ-ray spectroscopic methods. Moreover, the cross section measurements were done for other FP elements, such as ^{127}I, ^{133}Cs and ^{134}Cs. This paper provides the summary of the FP cross section measurements, which have been performed by authors.

1. Introduction

In nuclear waste management, the major 29 fission product (FP) nuclides shown in **Table 1** are important nuclides as the objective nuclides for transmutation. For the study of transmutation by using reactor neutrons, the accurate data are needed on the neutron capture cross sections (σ_0) and the resonance integrals (I_0) in order to estimate the accurate reaction rates of those FP nuclides. However, there are few cross section data on those FP nuclides. If any, most of the data have large errors. By using the recently developed measuring equipments and the accurate γ-ray emission probability (I_γ) data, one could obtain more accurate cross section data than that measured previously. Accordingly, we had started to measure the cross sections of FP nuclides to obtain the accurate ones. In the beginning five nuclides, ^{137}Cs[1], ^{90}Sr[2], ^{99}Tc[3], ^{129}I[4] and ^{135}Cs[5] , were chosen from the nuclides listed in Table 1 because of their large fission yields and long half-lives, and then the cross sections of these nuclides were measured by the activation and γ-ray spectroscopic methods.

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 affect the neutron economics; the reaction rate of the target nuclei is reduced. Moreover, some of these stable nuclei breed more radioactive nuclei by the neutron capture process. It is also necessary for the transmutation study to accurately estimate these influences caused by stable nuclei involved in the FP targets. Consequently, the cross sections of the stable nuclei, such as ^{127}I[6], ^{133}Cs[7], were measured.

2. Brief Outline of Analysis

Since the details of Westcott's convention[8] that we used to determine the cross sections and neutron fluxes were described elsewhere[1], here we present only a brief outline of the analysis. Equations based on Westcott's convention can be rewritten by using simplified flux notation[1] as follows:

$$R/\sigma_0 = \phi_1+\phi_2 s_0 , \quad (1)$$

for irradiation without a Cd shield capsule,

$$R'/\sigma_0 = \phi_1' + \phi_2' s_0 , \quad (2)$$

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 s_0 is the parameter defined by

$$s_0 = \frac{2}{\sqrt{\pi}} \frac{I_0'}{\sigma_0}, \quad (3)$$

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:

$$I_0 = I_0' + 0.45\sigma_0 , \quad (4)$$

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,

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

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).

3. Experiment

The cross section measurements were performed by the activation and γ-ray spectroscopic methods. The information of each experiment is summarized in **Table 2**.

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 (two foil method[9]) at the irradiation position. The method of measuring the neutron flux was the same as that for the cross section measurements. Using the well-known data of both the cross sections σ_0 and the parameter s_0 for cobalt and gold, the values of the flux terms, i.e. $\phi_{1,2}$ and $\phi'_{1,2}$, were determined by solving the simultaneous equations for cobalt and gold from Eqs. (1) and (2) in **Sec.2**. For example, **Table 3** summarizes the experimental results of the neutron fluxes in the case of Rikkyo Reactor together with the R and R' values of the flux monitors.

The yields of γ-rays emitted from the irradiated targets were measured by a high purity Ge detector with a 90% relative efficiency to a 7.6cm×7.6cmϕ(NaI) detector and an energy resolution of 2.1keV FWHM at 1.33MeV of ^{60}Co. The details of the data taking system were described elsewhere[4]. An example of the gamma-ray spectrum is shown in **Figure 1**. As can be seen in Figure 1, the two γ-rays originated from ^{100}Tc, 540 and 591keV γ-rays, were clearly measured. The cross sections of ^{99}Tc were deduced from the measured γ-ray intensities according to the Westcott's convention.

4. Results and Discussion

The results of the cross sections obtained in this work are summarized in **Tables 4**[10-18] together with the previously reported data. The brief discussion for each target is made as follows:

Cesium-137 Our result of σ_0 was about twice larger than the previous one[10] and the I_0 was obtained for the first time.

Strontium-90 Our result of σ_0 was in agreement with the value obtained by McVey et al. [11], but not agreement with the value by Zeisel [12]. The I_0 of ^{90}Sr was also measured in this work. However, the upper limit value of the I_0 ($I_0 \leqq 0.16$ b) was obtained. That remains to be re-measured in the future.

Technetium-99 The cross section σ_0 was almost equal to the previous data, but the resonance integral was about twice of that by Lucas et al.[13]. The possible reasons were that Lucas et al. used the huge amount of target, therefore the self-shielding effect influenced their results.

Iodine-129 Our data of σ_0 was 12% larger than the previously reported one[14]. This discrepancy was probably caused by the lack of the contribution of the isomeric state ^{130m}I because the existence of the isomeric state was not known.

Cesium-135 The value of σ_0 obtained in this experiment was almost the same as that reported by Baerg et al.[15]. On the other hand, the value of I_0 was about 2/3 of that by Berg et al. They used only one kind of flux monitor, i.e. Co/Al alloy wire, therefore they did not take the contribution of the epithermal component into consideration sufficiently.

Iodine-127 and Cesium-133 There are some discrepancies between our results and reported data[16,17]. There may be still some problems in cross section data of not only FP nuclides but also stable FP elements. It seems that the cross sections of other stable nuclides have to be re-measured.

5. Conclusions

The thermal neutron capture cross sections and the resonance integrals of some FP nuclides were measured by the activation and γ-ray spectroscopic methods.

Some future plans, which aim for the more precise measurements of the cross sections, are shown below:

(1) the measurement of the isomer production ratio of ^{138m}Cs to ^{138g}Cs in the ^{137}Cs(n, γ) ^{138}Cs reaction;
(2) the measurement of the emission probability (I_γ) of γ-ray from ^{100}Tc by β-γ coincidence technique [19];
(3) the measurement of the resonance integral of the ^{90}Sr(n,γ)^{91}Sr reaction;
(4) the measurements of the cross sections for long-lived FPs, such as ^{79}Se(6.5×10^5 y), ^{126}Sn($\sim1\times10^5$ y), ^{107}Pd(6.5×10^6 y), ^{93}Zr(1.53×10^6 y), ^{166m}Ho(1200 y).

The plans of (1),(2) and (3) are now in progress at the Research Reactor Institute, Kyoto University under join-research.

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Table 1 Major 29 fission product nuclides for the nuclear waste management

Nuclide	Half-Life (year)	Nuclide	Half-Life (year)	Nuclide	Half-Life (year)
^{129}I	1.57×10^{7}	^{108m}Ag	418	^{155}Eu	4.7611
^{107}Pd	6.5×10^{6}	^{151}Sm	90	^{102}Rh	2.9
^{135}Cs	2.3×10^{6}	^{121m}Sn	55	^{125}Sb	2.7582
^{93}Zr	1.53×10^{6}	^{137}Cs	30.07	^{147}Pm	2.6234
^{99}Tc	2.1×10^{5}	^{90}Sr	28.78	^{134}Cs	2.062
^{126}Sn	1×10^{5}	^{113m}Cd	14.1	^{171}Tm	1.92
^{79}Se	6.5×10^{4}	^{152}Eu	13.542	^{109}Cd	1.270
^{94}Nb	2.03×10^{4}	^{93m}Nb	16.13	^{106}Ru	1.007
^{166m}Ho	1200	^{85}Kr	10.756	(^{14}C	5730)
^{158}Tb	180	^{154}Eu	8.593		
		^{146}Pm	5.53		

Table 2 Target preparation and neutron irradiation for each experiment

Nuclide	Irradiation	Method to determine the number of target nuclei	Target amount
^{137}Cs	JRR-4 at JAERI (4×10^{13}n/cm^{2}s)	Comparison of γ-ray intensities of the target (^{137}Cs) and the product nuclei (^{138}Cs) (an isotope ratio method; **IRM**)	CsCl containing about 0.4MBq of ^{137}Cs
^{90}Sr	JRR-4 at JAERI (4×10^{13}n/cm^{2}s)	Same method as ^{137}Cs by using ^{85}Sr as a tracer of strontium. (**IRM**)	$SrCl_2$ containing about 2MBq of ^{90}Sr
^{99}Tc 370kBq	Rikkyo Reactor (5×10^{11}n/cm^{2}s)	Liquid scintillation counting of β-rays (an efficiency tracing method; **ETM**)	The standardized solution containing about of ^{99}Tc
^{129}I	Rikkyo Reactor (5×10^{11}n/cm^{2}s)	Liquid scintillation counting of β-rays (**ETM** + coin.-anticoin. Counting)	About 60ml solution containing about 2600Bq of ^{129}I for the irradiation within a Cd capsule, about 260Bq of ^{129}I for the irradiation without it.
^{135}Cs ^{135}Cs	JRR-3 at JAERI (1×10^{14}n/cm^{2}s)	Mass analysis with a quadrupole mass spectrometer and IRM (**IRM** + mass analysis)	About 0.37MBq of ^{137}Cs which contains
^{133}Cs	Rikkyo Reactor (5×10^{11}n/cm^{2}s)	weight measurement	About 12mg of the high purity(99.99%) ^{nat}CsCl
^{127}I	Rikkyo Reactor (5×10^{11}n/cm^{2}s)	The amount of ^{127}I in the KI target was used as the reference to determine the amount of ^{127}I in the ^{127}I-^{129}I target	^{127}I as a contamination contained in the ^{129}I solution

Table 3 Results 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 4. Summary of thermal neutron capture cross sections (σ_0) and resonance integrals (I_0) of important fission product nuclides for transmutation studies

Nuclide	Half-Life	Previous Data (Author and reported year)	Data of JNC
^{137}Cs	30 years	σ_{eff}=0.11±0.03 b (Stupegia '60[10])	σ_0= 0.25±0.02 b I_0= 0.36±0.07 b ('93)
^{90}Sr	29 years	σ_0=0.0140±0.0024 b (McVey '83[11]) σ_{eff}=0.8±0.5 b (Zeisel '66[12])	σ_0= $15.3\pm^{1.3}_{4.2}$ mb $I_0 \leqq 0.16$ b ('94)
^{99}Tc	2.1×10^5 years	σ_0= 20±2 b I_0'= 186±16 b (Lucas '77[13])	σ_0= 22.9±1.3 b I_0= 398±38 b ('95)
^{129}I	1.6×10^7 years	σ_0= 27±2 b I_0= 36±4 b (Eastwood '58[14])	σ_0= 30.3±1.2 b I_0= 33.8±1.4 b ('96)
^{127}I	(stable)	σ_0= 4.7±0.2 b I_0= 109±5 b (Friedmann '83[16])	σ_0= 6.40±0.29 b I_0= 162±8 b ('99)
^{135}Cs	3×10^6 years	σ_0= 8.7±0.5 b I_0= 61.7±2.3 b (Baerg '58[15])	σ_0= 8.3±0.3 b I_0= 38.1±2.6 b ('97)
^{134}Cs	2 years	σ_{eff}=134±12 b (Bayly '58[18])	σ_{eff}=141±9 b ('99)
^{133}Cs	(stable)	σ_0= 30.4±0.8 b I_0= 461±25 b (Baerg '60[17])	σ_0= 29.0±1.0 b I_0= 298±16 b ('99)

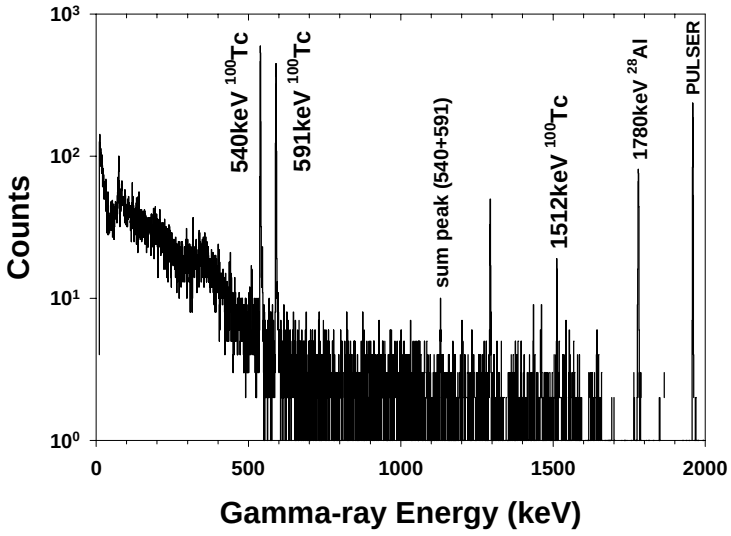

Figure 1 Gamma-ray spectrum of an irradiated ^{99}Tc sample target