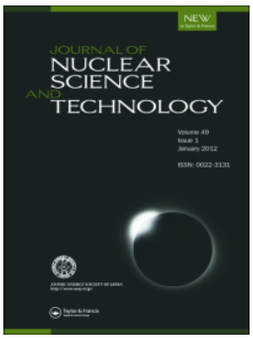

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Measurement of Thermal Neutron Capture Cross Section and Resonance Integral of the Reaction $^{133}Cs(n, \gamma)^{134m,134g}Cs$

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The measurements of the thermal neutron (2,200 m/s neutron) cross section (σ_0) and the resonance integral (I_0) of the $^{133}Cs(n, \gamma)$ reaction were performed by an activation method to obtain fundamental data for research on the transmutation of nuclear wastes. The cross sections for the formations of the isomeric state ^{134m}Cs and the ground state ^{134g}Cs were measured respectively by following the behavior of the γ-ray counting rate after the irradiation.

The thermal neutron capture cross sections and the resonance integrals of the $^{133}Cs(n, \gamma)$ reaction were determined to be 2.70±0.13 b and 23.2±1.8 b for the formation of the isomeric state ^{134m}Cs, and 26.3±1.0 b and 275±16 b for the formation of the ground state of ^{134g}Cs. The results for the reaction $^{133}Cs(n, \gamma)^{134m+g}Cs$ were 29.0±1.0 b and 298±16 b, respectively. As for the thermal neutron capture cross section for the formation of $^{134m+g}Cs$, the evaluated value (29.00 b) of JENDL-3.2 agreed with the present result. The reported value by Baerg *et al.* was in good agreement with the present result within the limits of error on the thermal neutron capture cross section for ^{134m}Cs. On the other hand, the resonance integral for $^{134m+g}Cs$ was 32% smaller than the experimental value by Steinnes *et al.*

KEYWORDS: ***nuclear transmutation, radioactive wastes, fission products, cesium 133, cesium 134, activation method, thermal neutrons, neutron capture, cross sections, resonance integral, isomer ratio, half-life, gamma-ray spectroscopy, experimental data, evaluations***

I. Introduction

The accurate neutron cross section data of long-lived fission product nuclides are of first importance for transmutation study. In case of the transmutation study using reactor neutrons, the thermal neutron capture cross section (σ_0) and the resonance integral (I_0) are needed. In this view point, the present authors have ever measured the neutron cross sections of important nuclides, such as, ^{137}Cs[1][2], ^{90}Sr[3], ^{99}Tc[4], ^{129}I[5] and ^{135}Cs[6].

In the process of the transmutation of ^{135}Cs, the amount of ^{135}Cs increases by the long neutron irradiation through the multi-neutron capture reactions, $^{133}Cs(n, \gamma)^{134}Cs(n, \gamma)^{135}Cs$, when the amount of ^{133}Cs contained in the target sample are large (in practice, the fission yield of ^{133}Cs is relatively large as 6.7%[7]). The thermal neutron capture cross section of the ^{133}Cs is large enough as 29.0 b[8]. Therefore, the ^{134}Cs is produced by the $^{133}Cs(n, \gamma)$ reaction, and possesses the high activity due to its rather short half-life (2.062 yr[9][10]). The ^{135}Cs is produced by the $^{134}Cs(n, \gamma)$ reaction because of the large cross section of ^{134}Cs as reported to be 140 b[8]. The resultant ^{135}Cs is the important nuclide for the transmutation because of its huge long half-life of 2.3×10^6 yr[10]. The first step reaction $^{133}Cs(n, \gamma)^{134}Cs$, therefore, is the noteworthy reaction from the point of primary purpose of the transmutation.

Many authors[11]-[14] have ever measured the neutron capture cross sections of the $^{133}Cs(n, \gamma)^{134}Cs$ reaction. Pomerance ('51) reported the cross section as 29.0±1.5 b[11] which was obtained by a pile-oscillator method. Baerg *et al.* ('60) measured the cross section for the formation of the isomeric state ^{134m}Cs as 2.82±0.07 b[12] by an activation method and Keisch ('61) as 2.44±0.15 b[13]. Takiue *et al.* ('78) measured the cross section for the formation of ^{134}Cs (that is, the sum of the cross sections for the isomeric state ^{134m}Cs and for the ground state ^{134g}Cs) as 28.7±0.7 b[14] by an activation method with a liquid scintillation counting technique.

Many of the older data[11]-[14] were obtained without benefit of the present-day refinements in counting equipment and without adequate knowledge of nuclear data, for example, the accurate neutron cross section data used for the neutron flux monitor, the decay schemes of the products and so on. The previous cross section data are in poor agreement with each other[11]-[14]. And also, there is only one experiment[12] which separately measured the cross sections for the formation of the isomeric state ^{134m}Cs and the ground state ^{134g}Cs. The cross section measurements for the formation of the isomeric and ground states are important since the accurate data of the isomer ratio is useful for the check of nuclear model calculations[15].

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The present experiment is intended for the accurate measurement of the thermal neutron cross sections and the resonance integrals for the formation of the isomeric state ^{134m}Cs and the ground state ^{134g}Cs which are produced by the ^{133}Cs(n, γ) reaction.

II. Experiment

1. Target Preparation

High purity (99.99%) cesium chloride powder was used as targets. Six targets were prepared as the following manners. First, six cylindrical quartz cases (7 mm in outer diameter, 1 mm in thickness and 8 mm in outer length) were weighed on a microbalance (Mettler model-UM3). About 12 mg of the CsCl powder was poured into each quartz case. All cases containing the powder were housed in an oven and then heated at 120°C for about 100 min to dry the CsCl powder. After drying the powder, the quartz cases with the CsCl powder were weighed on the microbalance. The amount of CsCl target was estimated from the increase in weight of each quartz case. Finally, each quartz case was covered tightly with a quartz plug, and was sealed with ceramic bond made from aluminum oxide.

Three of these six targets were irradiated without a Cd capsule and the others within a Cd capsule, which was 1 mm in thickness, 22 mm in outer diameter and 63 mm in outer length. The Cd capsule was used to absorb the thermal neutron flux at the irradiation position, and to reduce the sensitivity to thermal neutrons.

The Au/Al alloy wire of 0.112 wt% of Au (0.510 mm in diameter) and the Co/Al alloy wire of 0.46 wt% of Co (0.381 mm in diameter) were used as activation detectors to monitor the neutron flux. Cobalt has a small resonance integral, while gold has a large resonance integral compared with its thermal neutron cross section. Thus cobalt and gold differ from each other in sensitivities to thermal and epithermal neutrons, therefore, these monitors are useful for the determination of the thermal and the epithermal neutron fluxes[16]. **Table 1** shows the nuclear data[8][10] used to determine the neutron flux. The Au/Al and Co/Al wires were weighed on the microbalance, and were 16.87 mg and 16.56 mg, respectively. The weighed wires were sealed with plastic bags.

2. Neutron Irradiation

The irradiation of the targets was performed with the Research Reactor TRIGA MK-II at Rikkyo University. Each target was housed in a polyethylene irradiation capsule, and placed in a rotary specimen rack (RSR), which was located in the graphite reflector surrounding the reactor core.

The irradiation was performed as the following procedure: A set of two ^{133}Cs targets and a Au/Al flux monitor wire was irradiated for 10 min without the Cd capsule, and another set for 25 min within the Cd capsule. And then, a Co/Al wire was irradiated at the same irradiation position for 1 h without the Cd capsule, and one for 2 h within the Cd capsule. After this, two sets of the irradiations of two Au/Al wires without and within the Cd capsule were performed to check the deviation of the neutron flux. These irradiations were repeated in another day where a set of a Cs target and a Au/Al flux monitor wire was irradiated within and without the Cd capsule, and a Co/Al wire within and without the Cd capsule.

3. Activity Measurement

A high purity Ge detector was used to measure the γ-rays from the irradiated targets. Its performance was characterized as 90% relative efficiency to a 7.6 cm×7.6 cmϕ NaI(Tl) detector and 2.1 keV full width at half maximum (FWHM) at the 1.33 MeV peak of ^{60}Co. The full energy peak efficiencies of the detector were calibrated with standard γ-ray sources of ^{152}Eu and ^{133}Ba. The signals from the detector were fed to a fast data acquisition system, and the γ-ray spectra measured were recorded on the memory of a personal computer. Details of the data taking system were described elsewhere[4]. The correction for the pulse pile-up loss of the full energy peak intensities was considered as the following manner[4]. Signals generated by an external pulse generator were fed into the test input of detector. The correction factor was estimated from the loss of the intensity of peak made by the input pulses.

III. Analysis

Figures 1(a) and (b) show the γ-ray spectra of the Cs targets irradiated without and within the Cd capsule, respectively. The counting period was 2 h for both spectra. The γ-rays, originating in the isomeric state ^{134m}Cs and the ground state ^{134g}Cs produced by the ^{133}Cs(n, γ) reaction, are observed at energies of 127, 563, 569, 605, 796 and 802 keV.

After the irradiation, the activities of ^{134m}Cs and

Table 1 Nuclear characteristics[8][10] of the neutron flux monitors

Material (Al alloy wire)		Diameter (mm)	Half-life	σ_0 (b)	s_0	Detected γ-rays		G_γ
						Energy (MeV)	Intensity (%)	
Au	0.112 wt%	0.510	2.696 d	98.65±0.09	17.22±0.32	0.412	95.5	0.995
Co	0.46 wt%	0.381	5.271 yr	37.18±0.06	1.738±0.061	1.173	99.90	0.998
						1.332	99.98	0.998

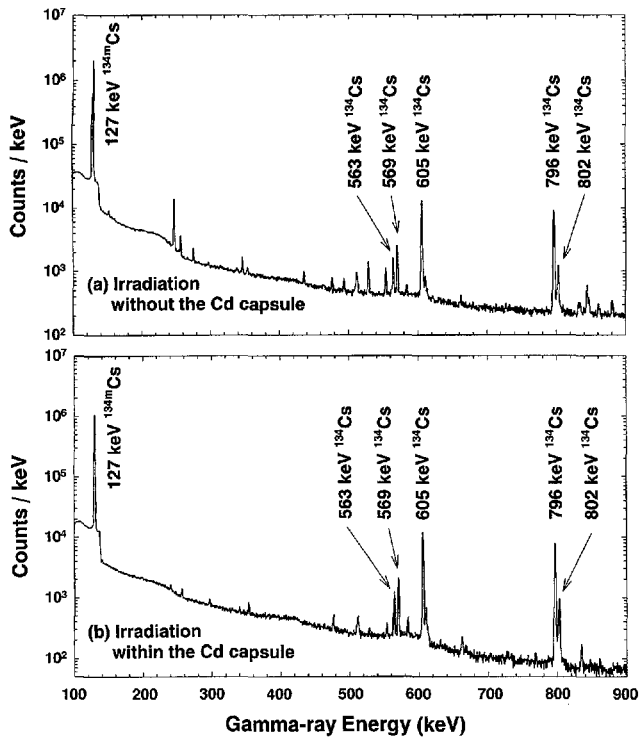

Fig. 1 Gamma-ray spectra of Cs targets irradiated (a) without and (b) within the Cd shield capsule by reactor neutrons

Table 2 Nuclear data[10] used for the analysis of the reaction ^{133}Cs$(n, \gamma)^{134m,134g}$Cs

Nuclide	Half-life	Detected γ-rays	
		Energy (keV)	Intensity (%)
^{134m}Cs	2.913±0.001 h†	127	12.7±0.3
^{134g}Cs	2.062±0.005 yr[9][10]	563	8.38±0.05
		569	15.43±0.11
		605	97.56±0.32
		796	85.44±0.38
		802	8.73±0.04

† This value was obtained in this work.

^{134g}Cs in the Cs targets, and of ^{60}Co and ^{198}Au in the flux monitor wires were determined from the γ-ray peak counts for each isotope. The nuclear data[8][10] used in the present analysis are listed in **Tables 1** and **2**. A partial decay scheme[10] of ^{134m}Cs and ^{134g}Cs is shown in **Fig. 2**.

The intensities of γ-ray peaks in the spectra are given by

$$Y_m = \varepsilon_m b_m N_m(\exp(-\lambda_m t_1) - \exp(-\lambda_m t_2)), \quad (1)$$

$$Y_g = \varepsilon_g b_g \Big[N_g(\exp(-\lambda_g t_1) - \exp(-\lambda_g t_2)) + \frac{\lambda_g}{\lambda_g - \lambda_m} N_m(\exp(-\lambda_m t_1) - \exp(-\lambda_m t_2)) - \frac{\lambda_m}{\lambda_g - \lambda_m} N_m(\exp(-\lambda_g t_1) - \exp(-\lambda_g t_2)) \Big], \quad (2)$$

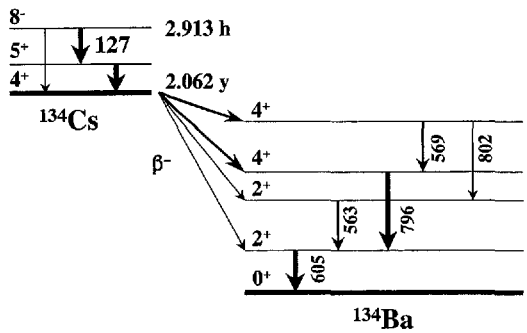

Fig. 2 A partial decay scheme of ^{134}Cs

where Y_j is the γ-ray peak count, ε_j the detection efficiency, b_j the emission probability per decay, λ_j the decay constant, N_j the number of nuclei produced by the reaction, t_1 the start time of measurement, and t_2 the stop time of measurement. The times t_1 and t_2 were measured from the end of the irradiation. The subscript j of Y, ε, b, λ and N denotes the isomeric state ^{134m}Cs (8^-) in case of j=m and the ground state ^{134g}Cs (4^+) in case of j=g. The quantities N_j (j=m, g) are able to be expressed as follows:

$$N_m = \frac{N_0}{\lambda_m} R_m(1 - \exp(-\lambda_m t_{irr})), \quad (3)$$

$$N_g = \frac{N_0}{\lambda_g}(R_m + R_g)(1 - \exp(-\lambda_g t_{irr})) + \frac{N_0}{\lambda_m - \lambda_g} R_m(\exp(-\lambda_m t_{irr}) - \exp(-\lambda_g t_{irr})), \quad (4)$$

where N_0 is the number of ^{133}Cs nuclei contained in the target, R_j the reaction rate for the production of the ^{134j}Cs, and t_{irr} the irradiation period.

The decay curves of 127 keV γ-ray from the ^{134m}Cs are shown in **Fig. 3**. In the analysis of the ^{134m}Cs, the parameters λ_m and R_m were determined by fitting Eq. (1) to the decay curves by the least squares method, where λ_m gave the half-life ($T^m_{1/2}$) of the ^{134m}Cs. The resultant half-life of the ^{134m}Cs was determined as 2.913±0.001 h, and in good agreement with the evaluated value of 2.914±0.003 h[10].

Figure 4 shows the growth curve of 605 keV γ-ray from the ^{134g}Cs involved in the Cs target irradiated without the Cd capsule. In the analysis of the ^{134g}Cs, an unknown paramcter R_g was determined by the least squares method with Eq. (2). In Eq. (2), the parameters λ_m ($=\ln 2/T^m_{1/2}$) and R_m were fixed to the values determined in the present analysis of the ^{134m}Cs, and the parameter λ_g was fixed to the value[10] obtained from the half-life data.

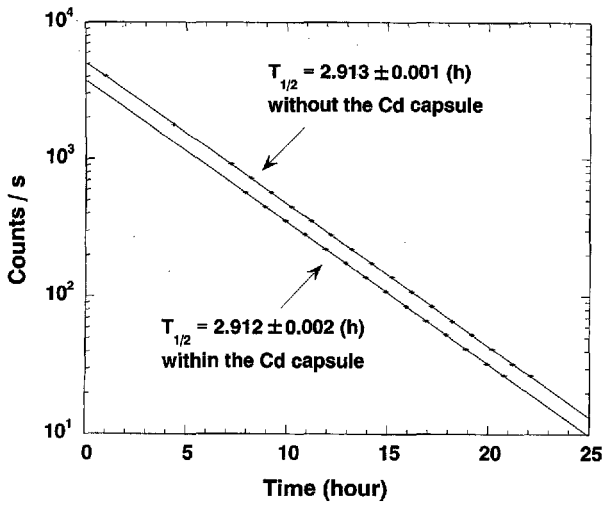

Fig. 3 Decay curves of the 0.127 MeV γ-ray emitted from the isomeric state ^{134m}Cs

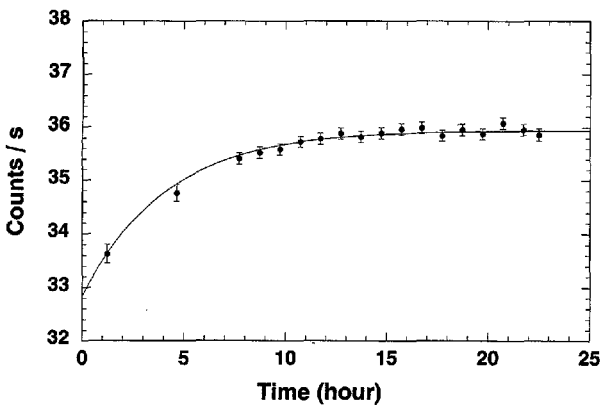

Fig. 4 Growth curve of the 0.605 MeV γ-ray emitted from the ground state ^{134g}Cs

The cross sections were calculated using the convention given by Westcott[16]. A brief outline is only given here to indicate the method used in this analysis. The measured reaction rate, R (corresponding to R_m or R_g mentioned above), is equated to the product of the effective cross section, $\hat{\sigma}$, and the neutron flux, nv_0, where n is the neutron density and v_0 is 2,200 m/s, thus

$$R = nv_0\hat{\sigma}. \tag{5}$$

The effective cross section $\hat{\sigma}$ is defined by

$$\hat{\sigma} = \sigma_0(g + rs), \tag{6}$$

where σ_0 is the 2,200 m/s cross section, r is a measure of the relative density of epithermal neutrons, g and s are factors which depend on the deviation of the cross sections from the $1/v$ law. In this analysis, it is assumed that g=1 and the factor s is written as $\sqrt{T/T_0}s_0$, where s_0 is related to the reduced resonance integral I_0' (with the $1/v$ part subtracted), which is defined by

$$s_0 = \frac{2I_0'}{\sqrt{\pi}\sigma_0}. \tag{7}$$

Thus,

$$\hat{\sigma} = \sigma_0(1 + r\sqrt{T/T_0}s_0), \tag{8}$$

where T is the neutron temperature and T_0 is 293.6 K corresponding to 2,200 m/s neutrons. The term $r\sqrt{T/T_0}$ is a constant (Westcott's index[17]) for the irradiation position in a reactor.

Equation (5) with Eq. (8) is rewritten in the form of the simplified neutron flux notation:

$$R/\sigma_0 = \phi_1 + \phi_2 s_0, \tag{9}$$

for irradiation without a Cd capsule, and

$$R'/\sigma_0 = \phi_1' + \phi_2' s_0, \tag{10}$$

for the irradiation within a Cd capsule. Using the known data of the cross sections σ_0 and the parameters s_0 for cobalt and gold in Table 1, 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. (9) and (10):

$$R^{(')}_{\mathrm{Au}}/\sigma_{0,\mathrm{Au}} = \phi_1^{(')} + \phi_2^{(')}s_{0,\mathrm{Au}}, \tag{11a}$$

$$R^{(')}_{\mathrm{Co}}/\sigma_{0,\mathrm{Co}} = \phi_1^{(')} + \phi_2^{(')}s_{0,\mathrm{Co}}. \tag{11b}$$

The experimental results of the R and R' values are listed in **Table 3**. **Figure 5** shows the experimental relations between R/σ_0 (or R'/σ_0) and s_0. The R/σ_0 intercepts and the slopes of the solid lines in Fig. 5 give the $\phi_1^{(')}$ and $\phi_2^{(')}$, respectively. The results of neutron fluxes are also summarized in Table 3. The thermal neutron flux at the irradiation position was $4.4\times10^{11}n/(\mathrm{cm}^2\cdot\mathrm{s})$, and Westcott's epithermal index $r\sqrt{T/T_0}$ was obtained as 0.039.

From Eqs. (9) and (10), following relation can be derived,

Table 3 Results of neutron flux measurements in Rotary Specimen Rack of Rikkyo Reactor

Irradiation type	Irradiation period (min)	Reaction rates of the flux monitors (10^{-11}/s)		Reduced neutron flux ($10^{11}n/\mathrm{cm}^2\cdot\mathrm{s}$)	
		^{60}Co	^{198}Au		
Without Cd	10	1.76±0.04	7.31±0.15	ϕ_1=4.42 ±0.09	ϕ_2=0.173±0.006
Within Cd	25	0.155±0.003	3.20±0.06	ϕ'_1=0.099±0.004	ϕ'_2=0.183±0.006
Ca ratio		11.3±0.3	2.28±0.07		

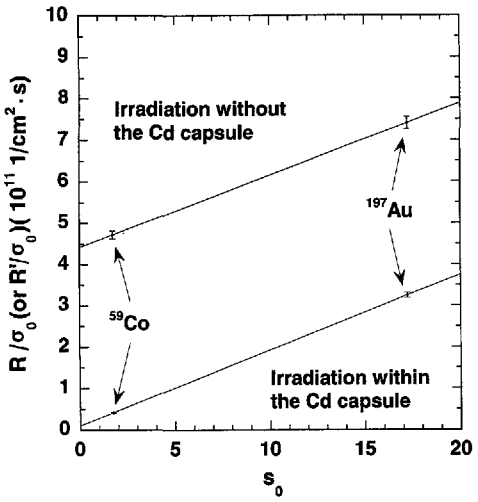

Fig. 5 Experimental relation between R/σ_0 (or R'/σ_0) and s_0 obtained by flux monitor wires irradiated without and within the Cd shield capsule

$$s_0 = -\frac{\phi_1 - \phi_1'(R/R')}{\phi_2 - \phi_2'(R/R')}, \tag{12}$$

so that the s_0 values for the ^{133}Cs$(n, \gamma)^{134m,134g}$Cs reactions are obtained from their R/R' values of the irradiated Cs targets. The value of σ_0 is obtained by substituting the s_0 into Eq. (9), and then the value of I_0' is calculated from Eq. (7).

The Cd cut-off energy (E_c) was estimated as follows: The target in this experiment was set at the center in the cylindrical Cd capsule, which was 22 mm in outer diameter, 63 mm in outer length and 1 mm in thickness. In consideration of the present geometrical configuration of the Cd capsule, the mean path length in the Cd wall was calculated for the neutrons that entered the Cd wall and contributed to the target activation. The mean path was calculated as 1.3 mm for the present Cd capsule. For this mean path, the Cd cut-off energy was estimated to be 0.547 eV according to the method of Westcott[17]. There is no appreciable resonance[18] in the energy region up to 5.9 eV, therefore there is very little variation in the reduced resonance integral for slightly changed values of E_c. The resonance integral I_0 was calculated by assuming the Cd cut-off energy E_c to be 0.5 eV.

The $1/v$ contribution to the resonance integral for the Cd cut-off energy E_c is given by

$$I(1/v) = \int_{E_c}^{\infty} \sigma_0 \sqrt{\frac{E_0}{E}} \cdot \frac{dE}{E} = 2\sigma_0 \sqrt{\frac{E_0}{E}}. \tag{13}$$

For E_0 and E_c taken as 0.0253 eV and 0.5 eV, the $1/v$ contribution to the resonance integral is estimated to be

$$I(1/v) = 0.45\sigma_0. \tag{14}$$

The resonance integral I_0 is given by the sum of the reduced resonance integral I_0' by Eq. (7) and the $1/v$ contribution by Eq. (14), thus, written in the following form:

$$I_0 = I_0' + 0.45\sigma_0. \tag{15}$$

In addition to the statistical errors, the following systematic errors are included in the errors of σ_0 and I_0.

(1) Error in the γ-ray full energy peak efficiency resulting from the uncertainty of the source intensity used for the calibration ·····2.0%
(2) Error in the measurement of the neutron flux resulting from the uncertainty of the nuclear data used for gold and cobalt ·····2.5%
(3) Error in the measurement of weight of target ·····1.0%
(4) Error in the emission probability per decay of the 127 keV γ-ray ·····2.4%
〃 563 keV γ-ray ·····0.6%
〃 569 keV γ-ray ·····0.7%
〃 605 keV γ-ray ·····0.3%
〃 796 keV γ-ray ·····0.4%
〃 802 keV γ-ray ·····0.5%

IV. Results

The thermal neutron capture cross sections were obtained from the above analysis of each γ-ray (127, 563, 569, 605, 796 and 802 keV γ-ray) emitted from the irradiated Cs target. **Table 4** shows the thermal neutron capture cross section ($\sigma_{0,m}$), the values of $s_{0,m}$, $I_{0,m}'$ and $I_{0,m}$ for the formation of the isomeric state ^{134m}Cs and the weighted averages of these values for Run #1~#3. The thermal neutron capture cross section ($\sigma_{0,g}$), the values of $s_{0,g}$, $I_{0,g}'$ and $I_{0,g}$ for the formation of the ground state ^{134g}Cs are shown in **Table 5**. The weighted average of these results are also shown in Table 5. The weighted average was taken excluding the result from the 802 keV γ-ray, because some kind of γ-ray peak could be included in the 802 keV γ-ray peak. The reason was that (a) the difference between the result from 802 keV γ-ray and the weighted average of all results from the 5 γ-rays (563, 569, 605, 796 and 802 keV γ-rays) went beyond the limits of the error, (b) the 802 keV γ-ray peak intensity was 19% larger than that deduced from data of the γ-ray emission probability, and also (c) the value of FWHM was appreciably large, 2.2 keV, for the 802 keV γ-ray compared with that of 1.9 keV for the 796 keV γ-ray.

The thermal neutron capture cross sections and the

Table 4 Experimental results of the cross section $\sigma_{0,m}$ and the resonance integral $I_{0,m}$ for the ^{133}Cs$(n, \gamma)^{134m}$Cs reaction obtained from the analysis of 127 keV γ-ray

	Run #1	Run #2	Run #3	**Weighted average**
$\sigma_{0,m}$ (b)	2.77±0.13	2.64±0.12	2.69±0.13	**2.70±0.13**
$s_{0,m}$	8.10±0.52	10.4±0.7	9.91±0.63	**9.19±0.58**
$I_{0,m}'$ (b)	19.9±1.6	24.3±1.9	23.7±1.9	**22.0±1.7**
$I_{0,m}$ (b)	21.1±1.6	25.5±2.0	24.9±1.9	**23.2±1.8**

Table 5 Experimental results of the cross section $\sigma_{0,g}$ and the resonance integral $I_{0,g}$ for the $^{133}Cs(n, \gamma)^{134g}Cs$ reaction obtained from the analysis of each γ-ray

	0.563 MeV	0.569 MeV	0.605 MeV	0.796 MeV	0.802 MeV	**Weighted average**[†]
$\sigma_{0,g}$ (b)	24.8±0.9	26.0±1.0	26.4±0.9	26.9±1.0	32.4±1.2	**26.3±1.0**
$s_{0,g}$	12.3±0.6	10.5±0.5	11.3±0.5	11.3±0.5	11.5±0.5	**11.3±0.5**
$I'_{0,g}$ (b)	270±16	243±14	265±15	269±15	330±19	**263±15**
$I_{0,g}$ (b)	281±16	255±15	277±15	281±16	345±19	**275±16**

[†] The weighted average excluding the result from the 0.802 MeV γ-ray

Table 6 Comparison of the present results with the previously reported data

Author (Year)	$\sigma_{0,m}$	$\sigma_{0,g}$	$\sigma_{0,m+g}$	$I_{0,m}$	$I_{0,g}$	$I_{0,m+g}$	Ref.
Present results	**2.70±0.13**	**26.3±1.0**	**29.0±1.0**	**23.2±1.8**	**275±16**	**298±16**	
Pomerance ('51)			29.0±1.5				(11)
Bayly *et al.* ('58)			30.4±1.7				(19)
Bidinosti *et al.* ('58)			39.9[†]			320	(20)
Baerg *et al.* ('60)	2.82±0.07		30.4±0.8	34.4±1.9		461±25	(12)
Keisch *et al.* ('61)	2.44±0.15						(13)
Brown *et al.* ('61)			33.4±2.6			370±50(S)	(21)
			33.4±3.2			375±50(J)	
			32.9±4.7			350±80(X)	
Sims *et al.* ('68)			29.2±2.3			495±17	(22)
Steinnes ('72)						437±26	(23)
Takiue *et al.* ('78)			28.7±0.7				(14)
JENDL-3.2 ('97)			29.00			396.2	(24)

[†] Effective cross section

resonance integrals for the formation of the isomeric and ground states obtained in the present work are listed in **Table 6** together with the data reported in references[11)-14)19)-24)]. 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 neutron capture cross sections, the reported values range from 28.7 to 33.4 b. The present result agrees with many reported data within the limits of the error. With respect to the resonance integrals, on the other hand, the discrepancies among the reported values were large and the reported ones range from 320 to 495 b. Although the values of the resonance integrals quoted from Refs. (12), (20), (21) and (22) were obtained by the irradiation experiments in the identical NRX reactor at Chalk River, the disagreement among these values was evidently remarkable. If the error is taken into consideration, the present value of the resonance integral agrees with some of the reported data, *e.g.* Refs. (20) and (21).

The results of the isomer ratios are summarized in **Table 7** together with the previous data[12)25)26)]. The definition of the isomer ratio was the same as that in Ref. (26). The isomer ratio for the thermal cross section is given by $\sigma_H/(\sigma_H+\sigma_L)$, where the σ with the subscript H or L denotes the formation cross section leading to "High-spin" or "Low-spin" state, respectively. In case of ^{134}Cs, σ_H and σ_L are expressed as σ_m and $(\sigma_m+\sigma_g)$, respectively, since the decay probability from the isomeric state ^{134m}Cs to the ground state ^{134g}Cs is 100%. The present result of the isomer ratio is compared with the previous data in **Fig. 6**. The present data resulted in supporting the previous ones.

Table 7 Results of isomer ratios for the $^{133}Cs(n, \gamma)^{134}Cs$ reaction

	Isomer ratio	
	Thermal cross section	Resonance integral
Present results	**0.085±0.005**	**0.072±0.007**
Ref. (12)	0.085±0.003	0.070±0.006
Ref. (25)	0.079±0.008	——
Ref. (26)	0.086±0.013	0.073±0.011

V. Discussion

With the pile oscillator method, Pomerance obtained a value of 29.0±1.5 b[11)] for the $\sigma_{0,m+g}$. He used the thermal neutron capture cross section of ^{197}Au as the reference cross section. The value he used for ^{197}Au cross section was 95 b, which is 3.7% smaller than the recent reference value, 98.65 b. His result becomes 30.1 b when the value of 98.65 b is used.

Bayly *et al.*, using the activation method with the

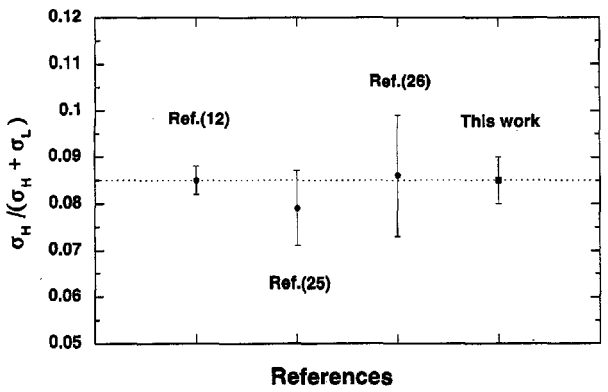

Fig. 6 Experimental isomer ratio for thermal neutron capture together with the other reported values

NRX reactor and the mass spectrometer analysis, found a value of 30.4±1.7 b[19] for the $\sigma_{0,m+g}$. A value of 2.19 yr for the half-life of ^{134}Cs was employed in their analysis. Their value becomes 28.6 b when the half-life of 2.062 yr[10] is used.

Using the NRX reactor, Bidinosti *et al.* measured the effective cross section for the formation of the $^{134m+g}$Cs as 39.9 b[20] by the activation method and the mass spectrometer analysis. Their result is about 11 b larger than the present result for the thermal neutron capture cross section $\sigma_{0,m+g}$. The reason is that the ^{133}Cs has a large resonance at the energy of 5.9 eV. Assuming 29 b for the thermal neutron capture cross section of ^{133}Cs, they estimated the resonance integral to be 320 b. This value is close to the present value.

Baerg *et al.*[12] measured the cross section and the resonance integral for the formation of ^{134m}Cs and those for the formation of $^{134m+g}$Cs simultaneously. The irradiation was also performed in the NRX reactor. Their values of the $\sigma_{0,m}$ and $\sigma_{0,m+g}$ are in agreement with those obtained in the present work. They assumed σ_0=36.68*b* and I_0=75 b for ^{59}Co. These values of ^{59}Co have been corrected to σ_0=37.18 b and I_0=74 b[8], which are used in the present analysis. With this revision, the values of the $\sigma_{0,m}$, $\sigma_{0,m+g}$, $I_{0,m}$ and $I_{0,m+g}$ are found to be 2.86 b, 30.7 b, 34.4 b and 455 b, respectively. The influence of the revision on the results is only 1.4%. They also used 2.19 yr for the half-life of ^{134}Cs. If the reanalysis is performed with the evaluated value of 2.062 yr[10] instead of 2.19 yr, the value of the $\sigma_{0,m+g}$ becomes 28.9 b, which is close to the present value. Baerg *et al.* performed the cross section analysis with Co data only. If our data are analyzed with the same manner used by them, the values of the $\sigma_{0,m}$, $\sigma_{0,m+g}$, $I_{0,m}$ and $I_{0,m+g}$ will be derived to be 2.59 b, 28.0 b, 24.2 b and 321 b, respectively. Though the thermal neutron capture cross section $\sigma_{0,m+g}$ obtained by Baerg *et al.* is close to the present result, the resonance integral is 55% lager than the present value. The present value 321 b of the resonance integral $I_{0,m+g}$ is in good agreement with that reported by Bidinosti *et al.*

Keisch *et al.* measured the cross section for the formation of ^{134m}Cs and obtained 2.44±0.15 b[13]. Their result is 10% smaller than the present value. They measured the neutron activation cross section as 2.85 b for the reactor spectrum. The effect of epithermal neutrons was examined by the irradiation with a Cd cover, and found to be 0.41 b. The thermal cross section was estimated as 2.44 b after subtraction of the contribution of epithermal neutrons. The contribution from the thermal component in the case of the irradiation with the Cd cover was not considered sufficiently, therefore the thermal neutron capture cross section may be underestimated.

As shown in Table 6, Brown *et al.*[21] measured the $\sigma_{0,m+g}$ and the $I_{0,m+g}$ at three irradiation positions (S, J, X) in the NRX reactor, whose values of the index $r\sqrt{T/T_0}$ were different. The parameters $s_{0,m+g}$ are estimated from their data as 12.0 for position S, 12.2 for J and 11.5 for X. In comparison with the present value ($s_{0,g}$=11.3), these values are large. So this point could be one reason for the discrepancy of the resonance integrals.

Sims *et al.* found the cross section, $\sigma_{0,m+g}$, to be 29.2±2.3 b[22] by the activation method in the NRX reactor. Their result is close to the present value. The parameter $s_{0,m+g}$, however, is large as 18.6, therefore the resonance integral becomes to be 495 b and 67% larger than the present value. It should be noted that the parameter $s_{0,m+g}$ obtained by Sims is remarkably different from that by Brown in spite of the same use of the NRX reactor.

Steinnes *et al.* used the resonance integral of ^{197}Au (1,550 b) as a comparator and found the resonance integral of ^{133}Cs to be 437±26 b[23]. With the same manner of analysis mentioned by Steinnes *et al.*, the resonance integral is derived to be 327 b from the present data.

Takiue *et al.* performed the absolute measurements of the induced ^{134}Cs activity with a liquid scintillation counter and obtained the cross section as 28.7±0.7 b[14]. They used the value of 2.046 yr for the ^{134}Cs half-life. With the revised value of 2.062 yr[10], the value of the cross section becomes 28.9 b, which is close to the present value. The present and their values are in good agreement with the evaluated data 29.00 b[24].

VI. Conclusion

The thermal neutron capture cross sections and the resonance integrals for the formations of the isomeric state ^{134m}Cs and the ground state ^{134g}Cs from the ^{133}Cs(n, γ) reaction were measured by using an activation method to obtain fundamental data for research on the transmutation of nuclear wastes.

The analyses of the reaction rates of the ^{133}Cs targets and the flux monitor wires irradiated without and within the Cd capsule gave the thermal neutron cross sections and the resonance integrals as follows:

$$\sigma_{0,m} = 2.70 \pm 0.13\text{b}, \quad I_{0,m} = 23.2 \pm 1.8\text{b}$$

for the formation of the isomeric state ^{134m}Cs,

$$\sigma_{0,g} = 26.3 \pm 1.0\text{b}, \quad I_{0,g} = 275 \pm 16\text{b}$$

for the formation of the ground state ^{134g}Cs, and also

$$\sigma_{0,m+g} = 29.0 \pm 1.0\text{b}, \quad I_{0,m+g} = 298 \pm 16\text{b}$$

for the formation of $^{134m+g}$Cs.

The thermal neutron capture cross section leading to the $^{134m+g}$Cs agreed with the data reported by Takiue *et al.* (28.7±0.7 b[14]), Pomerance (29.0±1.5 b[11]), Bayly *et al.* (30.4±1.7 b[19]), Baerg *et al.* (30.4±0.8 b[12]) and Sims *et al.* (29.2±2.3 b[22]), and with the evaluated value in JENDL-3.2 (29.00 b[24]). The thermal neutron capture cross section leading to ^{134m}Cs was in good agreement with the data reported previously, *e.g.* 2.82±0.07 b[12] by Baerg *et al.* On the other hand, most of the previous resonance integrals range from 350 b to 495 b and these values are higher by 17–66% than the present measurement. The value of the isomer ratio calculated by the present cross sections was in good agreement with the previous ones by Baerg *et al.*[12] and Bishop *et al.*[26]

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