Measurement of keV-neutron capture cross sections and capture gamma-ray spectra of Cs-133 and I-127

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Abstract. The neutron capture cross sections and the capture gamma-ray spectra of ^{127}I and ^{133}Cs at incident neutron energies from 15 to 100 keV have been measured by the time-of-flight method. Capture gamma-rays were detected with an anti-Compton NaI(Tl) spectrometer, and the pulse-height weighting technique was applied to derive capture yields. The capture cross sections of ^{127}I and ^{133}Cs were determined using the standard capture cross section of ^{197}Au. The total errors of the cross sections were 3.8-5.1%. The obtained cross sections were compared with evaluated values in JENDL-4.0 and ENDF/B-VII.1. For ^{127}I, the energy dependence is different between the present results and the evaluations. For ^{133}Cs, the evaluated values in JENDL-4.0 agree with the present results but the evaluated values in ENDF/B-VII.1 are smaller than the present results by 14%–18%. The capture gamma-ray spectra of ^{133}Cs and ^{127}I were derived by unfolding the pulse height spectra with detector response functions.

1. Introduction

A key issue of the current nuclear energy production is disposal of high-level radioactive waste from nuclear power plants. Currently-planned geological disposal is controversial because long-lived nuclides existing in spent nuclear fuel require very long-term management. In order to solve the issue, researchers have suggested nuclear transmutation systems that transmute long-lived nuclides to shorter-lived or stable species via neutron-induced reactions. The systems can reduce the impact of nuclear waste disposal on the environment [1].

^{129}I and ^{135}Cs are long-lived fission products (LLFPs), which are desired to be transmuted. The cumulative fission yields of I isotopes and Cs isotopes for tharmal neutron fission of ^{235}U and ^{239}Pu are shown in Tables 1 and 2. As seen in these tables, long-lived fission products exist together with their stable isotopes in spent nuclear fuel. To design the nuclear transmutation systems of LLFPs ^{129}I ($T_{1/2}$ = 15.7 M year) and ^{135}Cs ($T_{1/2}$ = 2.3 M year), accurate neutron capture cross section data are needed not only of the LLFPs but also of the stable isotopes ^{127}I and ^{133}Cs unless isotope separation is carried out.

The neutron capture cross sections of ^{127}I and ^{133}Cs have been evaluated in nuclear data libraries based on experimental data [2–10] but the evaluated cross sections in JENDL-4.0 [11,12] and ENDF/B-VII.1 [13,14] differ by 20% at a maximum in the keV energy region. This motivated us to carry out the present measurements. We measured the neutron capture cross sections of ^{127}I and ^{133}Cs in the energy region from 15 to 100 keV. In addion, we measured the neutron capture gamma-ray spectra of ^{127}I and ^{133}Cs, which give more information on reation mechanism of the neutron capture reaction.

2. Experimental procedure

Details of the experimental procedure were reported in [15]. Only a brief description is given in this report. Experiments were performed using a 3 MV Pelletron accelerator at the Laboratory for Advanced Nuclear Energy at the Tokyo institute of Technology. A pulsed neutron beam was produced thorough the ^{7}Li(p,n)^{7}Be reaction induced by a pulsed proton beam from the Pelletron accelerator. The pulse width of the proton beam was 1.5 ns and the repetition rate was 4 MHz. The proton energy was set at 1.902 MeV, 22 keV above the reaction threshold of ^{7}Li(p,n) reaction. The time-of-flight (TOF) method was employed to deduce the incident neutron energy. Neutrons were detected with a ^{6}Li glass scintillator (scintillator size: 5 mm diam. × 5 mm thick) placed at a flight distance of 30 cm. The detection angle of the ^{6}Li glass scintillator with respect to the proton beam direction was 8.7°, the average angle of neutrons incident on a 55 mm-diamter sample placed at a distance 12 cm from the neutron source. The energy of the incident neutrons distributed from a few keV to 100 keV. The flight path length from the neutron source to the sample was 12 cm.

Neutron capture gamma-rays emitted from the samples were detected with an anti-Compton NaI(Tl) spectrometer [16]. The spectrometer consists of a main NaI(Tl) detector (152 mm diam. × 305 mm length) and an annular anti-Compton NaI(Tl) detector (330 mm outer diam. × 172 mm inner diam. × 356 mm length) surrounding the main NaI(Tl) detector. The spectrometer was shielded with borated polyethylene, borated paraffin, potassium-free lead, cadmium, and ^{6}LiH. We recorded the pulse-height (PH) and the TOF of NaI(Tl) signals in list-data format sequentially.

Samples in chemical forms of CsI and PbI$_2$ were used because cesium and iodine in metal form are chemically unstable in the air. In addition, the natural

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Table 1. Cumulative fission yields (%) of I isotopes for tharmal neutron fission of ^{235}U and ^{239}Pu.

	^{127}I	^{129}I	^{131}I
^{235}U	0.1522	0.5388	2.895
^{239}Pu	5.0665	1.3214	3.8562

Table 2. Cumulative fission yields (%) of Cs isotopes for tharmal neutron fission of ^{235}U and ^{239}Pu.

	^{133}Cs	^{135}Cs	^{137}Cs
^{235}U	6.7000	6.5342	6.1636
^{239}Pu	7.0154	7.6171	6.6038

isotope abundance of both ^{133}Cs and ^{127}I is 100%. To estimate contribution from lead in the PbI$_2$ measurement, we also used a metal lead sample. ^{197}Au was also used as a standard sample for neutron capture cross section. The physical form of the CsI and PbI$_2$ samples was powder. The samples were sealed in graphite containers. The diameters of the samples were 55 mm. The weights of the samples were 10.04 g and 9.94 g for CsI and PbI$_2$.

Measurements were repeated cyclically for the CsI sample (CsI run), PbI$_2$ sample (PbI$_2$ run), Pb sample (Pb run), ^{197}Au sample (^{197}Au run) and no sample (blank run).

3. Data analysis

Recorded TOF-PH event data were analyzed. Four foreground and one background TOF gates were set for analysis. The corresponding energies to the four foreground gates were 15–25 keV (Gate 1), 25–35 keV (Gate 2), 35–55 keV (Gate 3), and 55–100 keV (Gate 4). The foreground and the background pulse-height spectra were obtained by sorting the TOF-PH event data with the TOF gates. The net pulse-height spectra were obtained by subtracting the background pulse-height spectra from the foreground pulse-height spectra.

A pulse-height weighting technique [2] was applied to derive the capture yields. The absolute cross sections were determined from standard measurements of a gold sample, based on the well-known cross section of ^{197}Au(n,γ)^{198}Au [17] of ENDF/B-VII.1. We also made corrections for neutron self-shielding and multiple scattering in the samples.

The neutron capture cross section of ^{127}I was obtained by subtracting the yield contribution of Pb from the obtained capture yield of the PbI$_2$ sample. Similarly, the neutron capture cross section of the ^{133}Cs was obtained by subtracting the yield contribution of ^{127}I from the obtained capture yield of CsI.

In addition to statistical errors, systematic uncertainties of the ^{197}Au(n,γ)^{198}Au cross section, weighting function, correction of discriminated gamma-ray contribution below 600 keV, correction of neutron self-shielding and multiple scattering, and impurity correction were included in the data analysis. The major components of the errors were statistics ($\sim$1%), the gold capture cross sections (3%), discriminated gamma-ray corrections ($\sim$1.5%), and self-shielding and multiple scattering (1–2%).

The capture gamma-ray spectrum was derived by unfolding the net pulse-height spectrum with detector response functions.
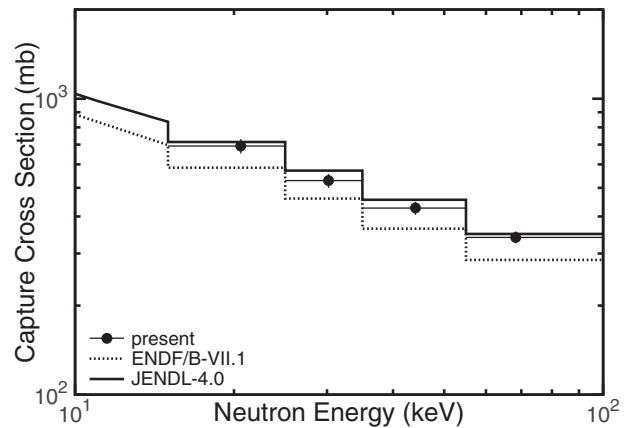

Figure 1. Neutron capture cross sections of ^{127}I. The present results are shown as solid circles. Evaluated data of JENDL-4.0 [11] and ENDF/B-VII.1 [13] are compared with the present results. The evaluations were averaged over the same energy bins as the present experimental data.
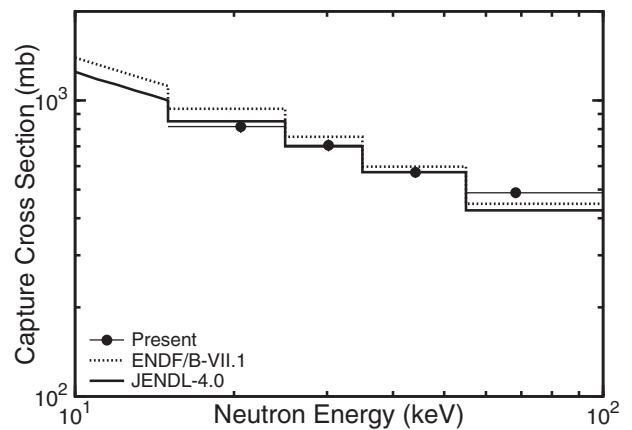

Figure 2. Neutron capture cross sections of ^{133}Cs. The present results are shown as solid circles. Evaluated data of JENDL-4.0 [12] and ENDF/B-VII.1 [14] are compared with the present results. The evaluations were averaged in the same energy bins as the present experimental data.

4. Results and discussion

4.1. Capture cross sections

The capture cross sections of ^{127}I and ^{133}Cs were derived with errors of about 5% in the neutron energy region from 15 to 100 keV. The present neutron capture cross sections are shown together in Figs. 1 and 2. The evaluated cross sections of JENDL-4.0 [11,12] and ENDF/B-VII.1 [13, 14] are also shown in the figures. The cross section values are averaged over the same energy bins as the present experimental data for comparison. For ^{127}I, the evaluated cross sections are higher than the present results at the lowest energy (15–25 keV) and lower at the highest energy (55–100 keV). For ^{133}Cs, the evaluated cross section of JENDL-4.0 is in good agreement with the present results. On the other hand, the cross section of ENDF/B-VII.1 is lower than the present results by 14–18%.

4.2. Capture gamma-ray spectra

The neutron capture gamma-ray spectra of ^{127}I and ^{133}Cs were derived by unfolding the net pulse height spectra with
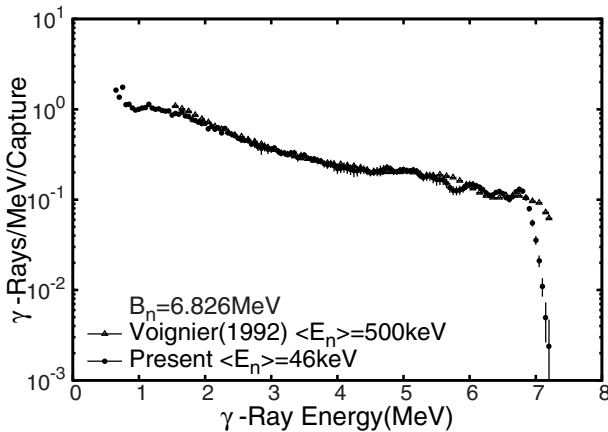
Figure 3. Capture gamma-ray spectrum of ^{127}I in the neutron energy region from 15 to 100 keV. Previous measurement [18] is compared with the present results.
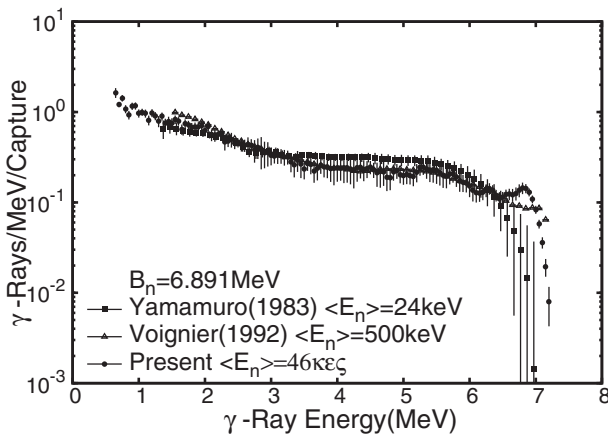

Figure 4. Capture gamma-ray spectrum of ^{133}Cs in the neutron energy region from 15 to 100 keV. Previous measurements [9], [18] are compared with the present results.

Table 3. Multiplicities of observed capture gamma-rays.

Nuclide	$B_n + <E_n>$ [MeV]	Multiplicties
^{127}I	6.872	2.67±0.02
^{133}Cs	6.937	2.56±0.04

detector response functions, as shown in Figs. 3 and 4. The previous measurements are also shown for comparison.

Voignier et al. [18] measured the capture gamma-ray spectra of ^{127}I and ^{133}Cs at a neutron energy of 500 keV. Yamamuro et al. [9] measured the capture gamma-ray spectra of ^{133}Cs in the neutron energy 24 keV. Except for difference of high energy endpoint due to the neutron energy difference, overall shape of the spectra agrees between the previous and measurements.

The gamma-ray multiplicities ($E_\gamma \geq 0.65$ MeV) calculated from the gamma-ray spectra are shown in Table 3.

5. Conclusions

The neutron capture cross sections of ^{127}I and ^{133}Cs were measured with the errors of 3.8–5.1% at incident neutron energies from 15 to 100 keV.

Comparison with evaluated cross sections of JENDL-4.0 and ENDF/B-VII.1 shows that the present results of

^{127}I and the evaluations are in almost good agreement but have slightly different energy dependence.

In comparison of the capture cross section of ^{133}Cs, the JENDL-4.0 evaluation agrees with the present results but the ENDF/ B-VII.1 evaluation is by 14-18% lower than the present results.

The capture gamma-ray spectra of ^{133}Cs and ^{127}I were derived by unfolding the net pulse-height spectra. The gamma-ray multiplicities ($E_\gamma \geq 0.65$ MeV) were obtained from the capture gamma-ray spectra. Capture gamma-ray spectra give more information to determine parameters of nuclear reaction model calculations. The present gamma-ray spectra will be used in future theoretical work.

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