Demand for TRU nuclide cross-sections from the view point of TRU production and radiotoxicity

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Abstract

The environmental load reduction of nuclear energy is required in Japan, from the view point of public acceptance. Here, the long-term radiotoxicity of radioactive wastes is dominated by trans-uranium (TRU) nuclides. We evaluated the effects of differences between the nuclear data libraries of heavy-metal-nuclide cross-section on the radiotoxicity of LWR spent fuels. In this study, the MVP-BURN code and the JENDL-4.0u nuclear data library were used as a burn-up calculation code and a reference nuclear data library, moreover, only a heavy metal cross section of interest was replaced to JEFF-3.2 or ENDF/B-VII.1 to evaluate the effect of difference between libraries for each nuclides. The calculation results revealed that the productions of Pu-238, Am-241 and Cm-244 with JEFF-3.2 were 8% larger than those with JENDL-4.0u and ENDF/B-VII.1. The thermal energy capture reaction of Pu-238 and 1.356eV resonance capture reaction of Am-243 have a large impact on the radiotoxicity of Pu-238 and Cm-244, consequently, these cross sections should be improved.

1 Introduction

The environmental load reduction of nuclear energy is required in Japan, from the view point of public acceptance due to the increase of safety demand to the nuclear energy utilization. This environmental load is mainly caused by the mass and radiotoxicity of radioactive wastes. Especially, long-term radiotoxicity (>100 years) of the radioactive waste is dominated by trans-uranium (TRU) nuclides [1]. Additionally, most of the TRU nuclides, which are large part of environmental loads, are generated from light water reactors. Therefore, the evaluation of TRU nuclide production in the light water reactors is important to estimate the environmental load of nuclear energy [2-4].

As well known, the amount of TRU nuclide is evaluated through burn-up calculations. Here, the burn-up chain of actinides is shown in Fig. 1. This figure also shows high-radiotoxicity nuclides and major ancestor nuclides among these TRU nuclides, and besides, the radiotoxicity of the TRU nuclides is deduced by radioactivity, type of decay, decay energy and biological-effect of radiation. As shown in this figure, high-radiotoxicity nuclides have some of major ancestor nuclides.

For these reason, the evaluation of high-radiotoxicity nuclide production require many actinide cross-section data from a nuclear data library. However, also as well known, these cross section data have a different value between libraries due to its uncertainty of experimental data.

Fig. 2 shows the difference of a neutron capture cross section for ^{238}Pu between ENDF/B-VII.1, JENDL-4.0u and JEFF-3.2 [5-7]. As shown in this figure,the neutron cross section is different in a range from thermal to epi-thermal energy region. From this result, it would be considered that other nuclide cross section data also have a difference between libraries. Ultimately, these differences should be improved in the future. However, measurements for all nuclides in same time are not realistic option. Therefore, the priority of measurement should be given by the effect of nuclide cross section on TRU nuclide production in burn-up chain from the view point of environmental load reduction.

For these backgrounds, we investigated cross section difference effects between libraries on high-radiotoxicity nuclides; furthermore, we made the requirement for cross section to improve the precision of burn-up calculation on high-radiotoxicity nuclides production.

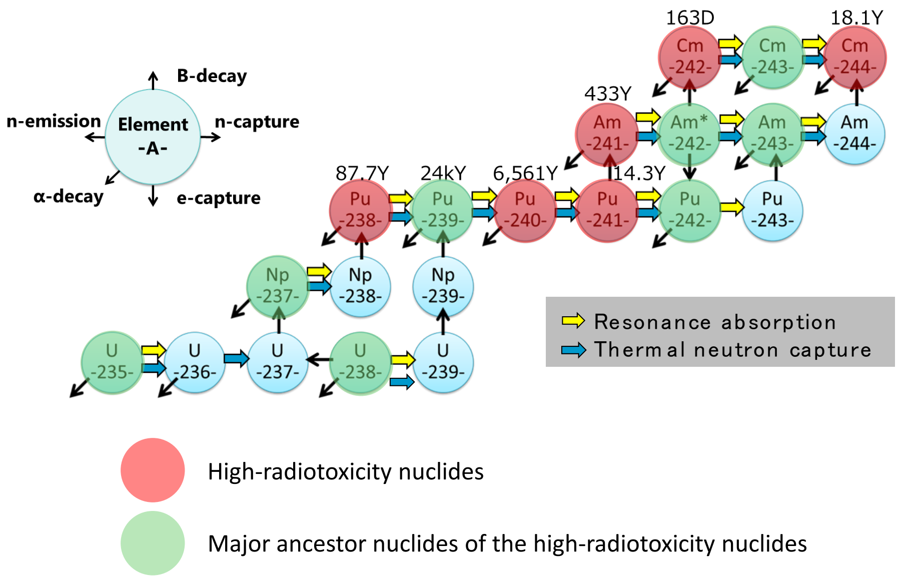


Fig. 1 Burn-up chain of the Actinides

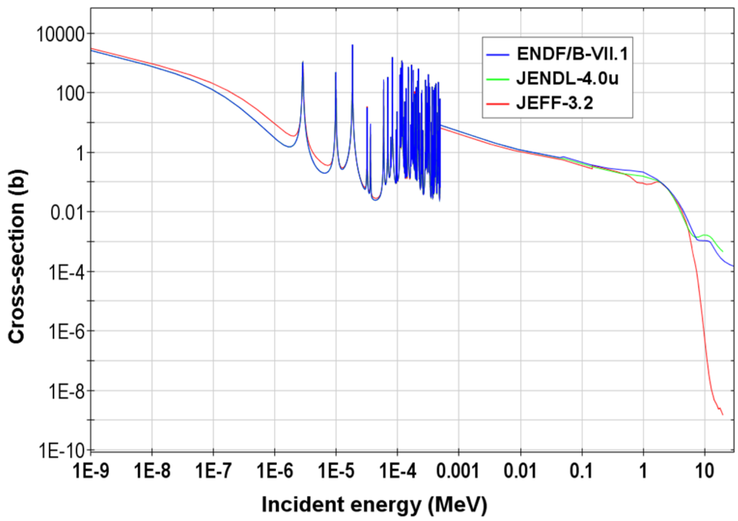


Fig. 2 Neutron capture cross section difference of ^{238}Pu between ENDF/B-VII.1, JENDL-4.0u and JEFF-3.2

2 Calculation method of the effect of nuclear data libraries

The diagram of nuclide cross section data replacement method is shown in Fig. 3. In this study, JENDL-4.0u was used as a reference nuclear data library; furthermore, the cross section data of nuclides were replaced by ENDF/B-VII.1 or JEFF-3.2 one by one to evaluate the effect of the cross section on TRU nuclide production.

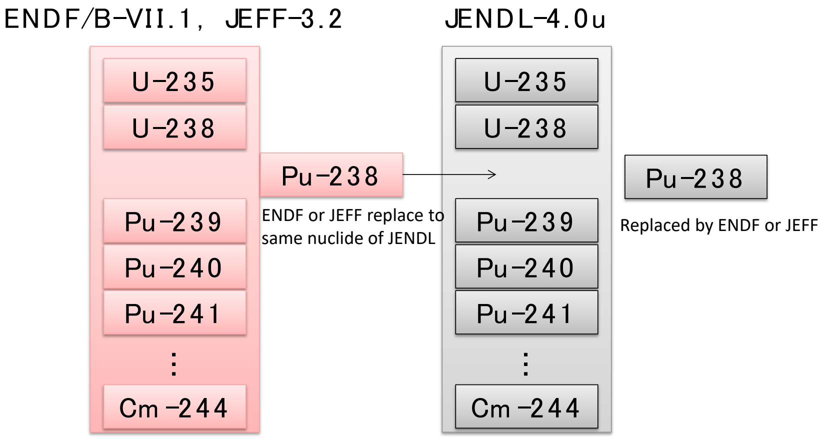


Fig.:3 Replacement met~~t~~hod of nuclide cross section data

The 9x9A type BWR fuel assembly was utilized in the present study. The cross section of this fuel assembly is shown in Fig. 4. Additionally, calculation condition is shown in Table 1. The MVP-BURN code was used as a Monte-Carlo burn-up calculation code [8], additionally; the burnup calculation of the 9x9A type BWR fuel assembly was conducted by MVP-BURN with a typical burn-up condition as shown in Table 1, the number density of each nuclides were evaluated at the burnup of 45GWd/t.

Table 1: Calculation Condition

Item	Condition
CODE	MVP-BURN
Nuclear Data Libraries	JENDL-4.0u(Ref) ENDF/B-VII.1 JEFF-3.2
Power Density	50 kW/L
Fuel pellet diameter	0.956 cm
Pin pitch	1.438 cm
Cladding thickness	0.071 cm
U-235 Enrichment	3.84 wt%
Burn-up	45 GWd/t

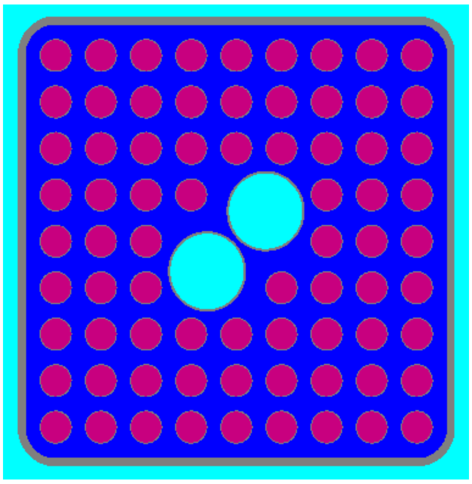


Fig. 4: Cross section image of the 9x9A type BWR fuel assembly

In the present study, number density ratio at the discharge burnup NR_{ij} was defined as equation (1)

$$NR_{ij} = \frac{N_{ij,ENDF}}{N_{i,JENDL}} \quad or \quad \frac{N_{ij,JEFF}}{N_{i,JENDL}} \tag{1}$$

Here, $N_{ij,JENDL}$ is the number density of a nuclide i at the discharge burnup calculated withJENDL-4.0u, $N_{ij,ENDF}$ and $N_{ij,JEFF}$ are discharged number density of nuclide i calculated with the cross section of nuclide j replaced by ENDF or JEFF. We evaluated this NR_{ij} for all nuclides in Table 2.

Table 2: Evaluated nuclides

Element	Nuclide
U	^{235}U, ^{238}U
Np	^{237}Np
Pu	^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu
Am	^{241}Am, ^{242g}Am, ^{242m}Am, ^{243}Am
Cm	^{242}Cm, ^{243}Cm, ^{244}Cm

3 Results and discussion

3.1 Results of the number density ratio and cause of the difference

The NR_{ij} results of ENDF/B-VII.1 and JEFF-3.2 are shown in Fig. 5 and Fig. 6. NR_{ij} results replacing JENDL-4.0u with ENDF/B-VII.1 shows the maximum difference of 2 %. In case that Minor Actinide (MA) cross sections were replaced, the maximum difference of NR_{ij} was smaller than 0.5 %. Because, ENDF/B-VII.1 have used the same cross sections data as JENDL-4.0u. While, the differences of some NR_{ij} values from 1.0 in JEFF-3.2 were larger than 8 %. Especially, effect of the ^{238}Pu, ^{241}Am and ^{243}Am cross sections had a large impact on the NR_{ij} values.

NR_{ij} results of each nuclide ($N_{ij,ENDF}/N_{ij,JENDL}$)

	ENDF-U235	ENDF-U238	ENDF-Np237	ENDF-Pu238	ENDF-Pu239	ENDF-Pu240	ENDF-Pu241	ENDF-Pu242	ENDF-Am241	ENDF-Am242g	ENDF-Am242m	ENDF-Am243	ENDF-Cm242	ENDF-Cm243	ENDF-Cm244
U-235	0.998	1.001	1.000	1.001	0.998	1.001	1.000	1.001	1.001	1.000	1.001	1.001	1.000	1.000	1.000
U-238	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000
Np-237	1.004	1.019	1.009	1.000	1.001	1.000	0.999	0.999	0.999	0.999	1.000	1.000	1.000	1.000	1.000
Pu-238	1.004	1.020	0.984	1.001	1.001	1.001	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000
Pu-239	0.999	1.002	0.999	0.999	1.000	1.000	1.001	1.000	1.001	1.001	1.001	1.000	1.000	1.000	1.000
Pu-240	0.998	1.001	1.002	1.000	1.010	0.998	1.001	1.000	1.001	1.000	1.000	1.001	1.000	1.000	1.000
Pu-241	1.002	1.002	1.000	1.000	1.007	1.003	1.000	1.000	1.002	1.002	1.001	1.001	1.000	1.000	1.000
Pu-242	1.001	0.999	0.999	1.000	1.006	1.002	0.999	1.000	0.999	1.000	0.999	1.000	1.000	1.000	1.000
Am-241	1.001	1.001	1.000	1.001	1.006	1.002	1.001	1.001	1.001	1.001	1.001	1.000	1.000	1.000	1.000
Am-242g	1.002	1.001	1.002	1.002	1.008	1.003	1.001	1.002	1.001	1.003	1.001	1.002	1.000	1.000	1.000
Am-242m	1.001	1.002	1.001	1.002	1.006	1.004	1.001	1.002	1.003	1.001	0.998	1.003	1.000	1.000	1.000
Am-243	1.001	0.998	0.999	1.002	1.010	0.997	0.999	1.003	1.002	0.999	1.003	0.996	1.000	1.000	1.000
Cm-242	1.001	1.000	1.000	1.002	1.007	1.002	1.001	1.001	1.001	1.002	1.001	1.001	1.000	1.000	1.000
Cm-243	1.001	1.000	0.998	1.001	1.009	1.005	1.000	1.001	1.000	1.002	1.003	1.001	1.000	1.000	1.000
Cm-244	1.000	0.997	0.999	1.000	1.007	1.002	0.999	0.998	1.000	0.998	1.001	1.002	1.000	1.000	1.000

Changed nuclide (JENDF-4.0u to ENDF/B-VII.1)

Fig. 5: NR_{ij} results of each nuclide based on the ENDF/B-VII.1

NR_{ij} results of each nuclide ($N_{ij,JEFF}/N_{ij,JENDL}$)

	JEFF-U235	JEFF-U238	JEFF-Np237	JEFF-Pu238	JEFF-Pu239	JEFF-Pu240	JEFF-Pu241	JEFF-Pu242	JEFF-Am241	JEFF-Am242g	JEFF-Am242m	JEFF-Am243	JEFF-Cm242	JEFF-Cm243	JEFF-Cm244
U-235	0.998	1.001	1.001	1.002	0.998	0.999	1.000	1.001	1.000	1.001	1.001	1.001	1.000	1.000	1.000
U-238	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000
Np-237	1.002	1.009	1.006	0.999	1.000	0.997	1.000	0.999	1.002	0.998	0.999	1.001	0.999	0.999	1.000
Pu-238	1.003	1.010	0.995	0.908	1.003	0.998	1.001	1.000	1.009	1.000	1.000	1.002	1.000	1.000	1.001
Pu-239	1.000	0.998	1.000	1.003	1.002	0.999	1.000	1.001	0.999	1.000	1.000	1.000	1.000	1.000	0.999
Pu-240	1.000	0.998	1.000	0.998	1.010	1.009	1.000	1.000	1.001	1.001	1.001	0.999	0.999	0.999	1.000
Pu-241	1.000	0.998	1.003	1.004	1.009	0.996	0.999	1.000	1.000	1.000	1.000	1.002	1.000	1.000	1.000
Pu-242	1.000	0.998	1.001	1.001	1.009	0.995	1.001	1.001	1.000	0.999	1.001	1.000	1.001	1.001	0.999
Am-241	1.000	0.998	1.002	1.004	1.007	0.996	1.000	1.000	0.916	1.001	1.001	1.001	1.000	1.000	1.000
Am-242g	1.000	0.998	1.003	1.003	1.009	0.998	1.001	1.001	1.023	1.004	1.001	1.002	1.001	1.001	1.000
Am-242m	1.000	0.998	1.004	1.005	1.007	0.996	1.001	1.002	1.024	1.003	0.997	1.003	1.001	1.001	1.000
Am-243	0.999	0.995	0.999	1.000	1.007	0.997	0.999	0.999	0.999	1.003	0.997	1.040	0.996	0.996	1.001
Cm-242	1.000	0.998	1.002	1.002	1.008	0.996	1.001	1.001	1.030	1.003	1.001	1.002	1.001	1.001	1.000
Cm-243	1.002	1.001	1.001	1.001	1.009	0.996	1.002	1.003	1.041	1.004	1.001	1.000	0.994	0.994	1.000
Cm-244	1.002	0.992	0.997	1.000	1.010	0.994	1.000	1.000	1.002	1.000	0.998	0.917	0.999	0.999	1.000

Changed nuclide (JENDF-4.0u to JEFF-3.2)

Fig. 6: NR_{ij} results of each nuclide based on the JEFF-3.2

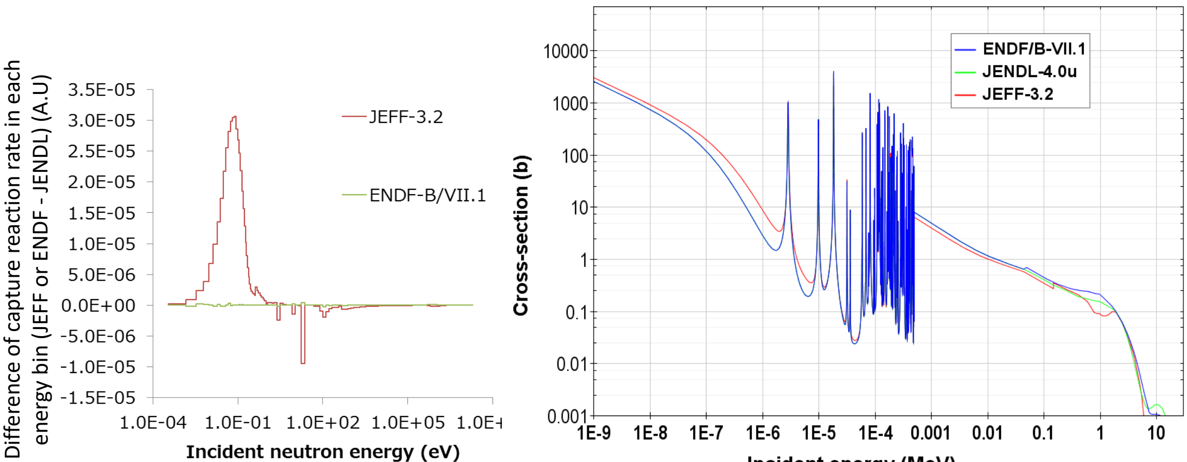
Accordingly, the different of reaction rate and major cross sections are shown in Fig. 7 to Fig. 9 to realize important energy region in each nuclides. Here, the different of the reaction rate Dif_R is defined as equation (2).

$$Dif_R = (\textit{Reaction rate by JEFF-3.2 or ENDF-B/VII,1}) - (\textit{Reaction rate by JENDL-4.0u}) \quad (2)$$

Firstly, Fig. 7 (a) shows the Dif_R of ^{238}Pu. As shown in this figure, the difference of capture reaction rate replacing JENDL-4.0u with JEFF-3.2 was dominated by thermal region. This difference came from capture cross section difference in the thermal energy region between these nuclear data libraries as shown in Fig. 7 (b).

Secondly, Fig. 8 (a) and (b) show the Dif_R and capture cross section of ^{241}Am. Here, capture cross section was drawn in liner scale to make it easier to understand. As shown in both figures, capture reaction rate difference between JEFF-3.2 and JENDL-4.0u was dominated by resonance region.

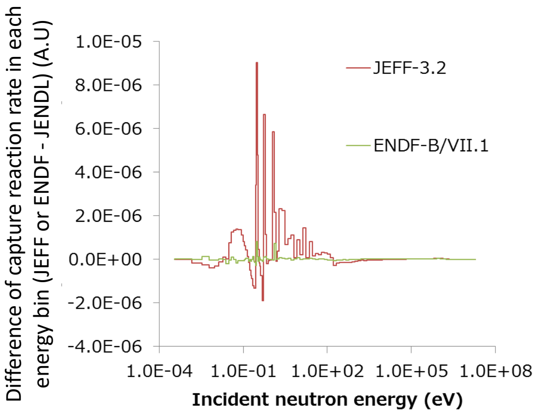
Finally, Fig. 9 (a), (b) and (c) show the Dif_R and capture cross section of ^{243}Am. As shown Fig. 9 (a), reaction rate difference was dominated by almost one resonance cross section. The capture cross sections of each library in 0.1-10 eV are drawn in Fig. 9 (b), additionally, resonance cross sections at 1.356 eV are shown in Fig. 9 (c). It was confirmed that the resonance cross section of ^{243}Am at 1.356 eV had a difference larger than 2000 b among theese libraries. In addition, measurement values between these libraries were compared in Fig. 10. As shown in this figure, the latest evaluated libraries do not support measurement value, but TENDL-2015 supported the measurement value. For these results, resonance cross section at 1.356 eV of ^{243}Am could have a large uncertainty.



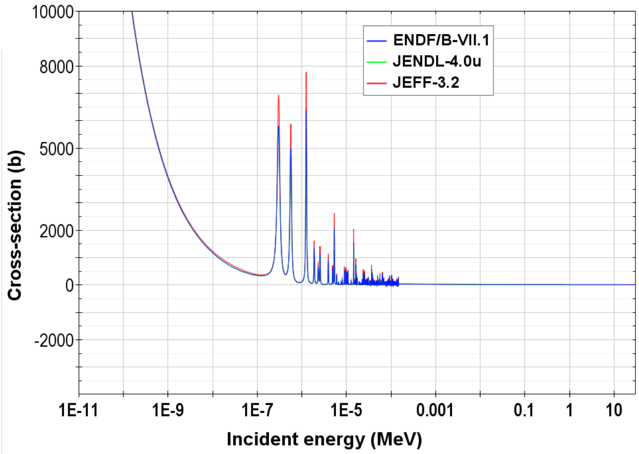
(a) Difference of ^{238}Pu capture reaction rate in each energy bin

(b) Comparison of ^{238}Pu capture cross section

Fig. 7: Reaction rate and capture cross section difference of ^{238}Pu between JENDL-4.0, ENDF/B-VII.1 and JEFF-3.2

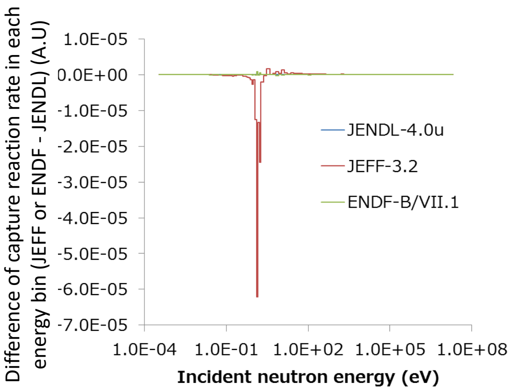


(a) Difference of ^{241}Am capture reaction rate in each energy bin

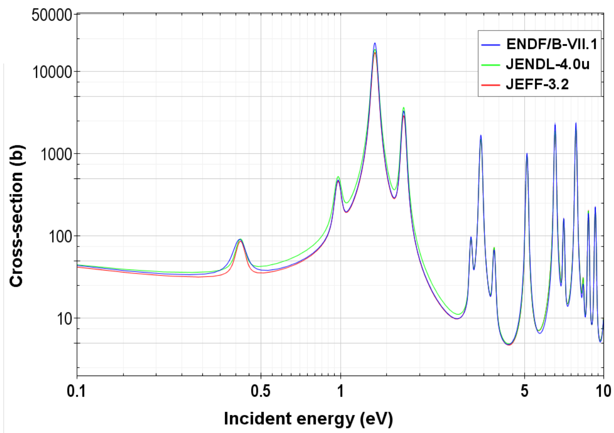


(b) Comparison of ^{241}Am capture cross section

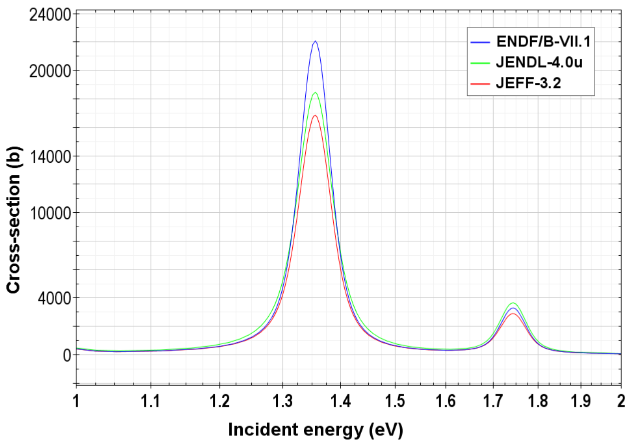
Fig. 8: Reaction rate and capture cross section difference of ^{241}Am between JENDL-4.0, ENDF/B-VII.1 and JEFF-3.2



(a) Difference of ^{243}Am capture reaction rate in each energy bin



(b) Comparison of ^{243}Am capture cross section



(c) Comparison of ^{243}Am resonance capture cross section at 1.356eV

Fig.9: Reaction rate and capture cross section difference of ^{243}Am between JENDL-4.0, ENDF/B-VII.1 and JEFF-3.2

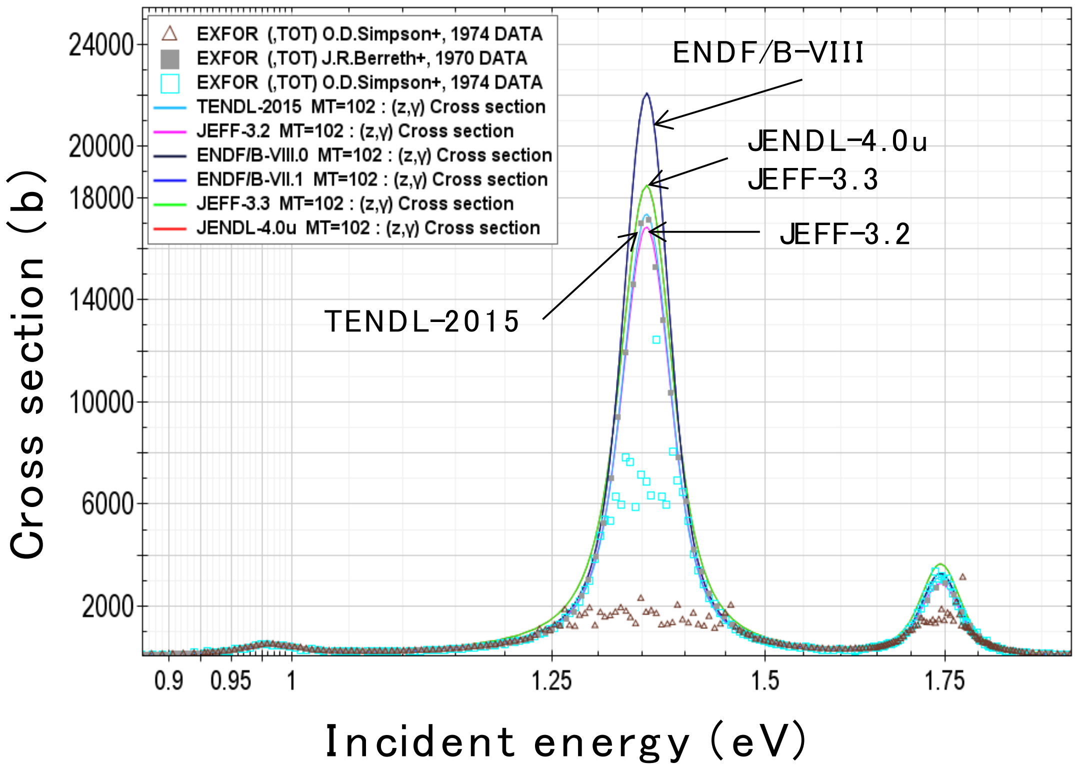


Fig.10: Measured value and libraries data of ^{243}Am total cross section [9-13]

3.2 Prioritization of requirement for nuclear data improvement

The important nuclide cross sections to estimate actinides discharged number density were realized in section 3.1. However, importance classifying of these cross sections were required for efficient nuclear data improvement. Hence, requirements for nuclear data improvement were prioritized in this section.

The relative composition of nuclides causing radiotoxicity in the UO_2 and MOX fuel are shown in Fig. 11. As shown in this figure, ^{238}Pu, ^{241}Pu and ^{244}Cm had a large composition at a discharged spent fuel. On the other hand, ^{238}Pu, ^{241}Am and ^{243}Am had a large sensitivity for actinides discharged number density as mentioned in section 3.1. Here, all actinide cross sections is insensitive to ^{241}Pu number density as shown in Fig. 5 and Fig. 6. Additionally, post irradiation experiment (PIE) data of ^{241}Pu number density shows good agreement with calculated result [14]. Therefore, ^{241}Pu was excluded from prioritization target.

Table 3 shows the prioritization results. In the results, first priority was ^{238}Pu capture cross section in thermal energy region. Because, the radiotoxicity of ^{238}Pu had a large weight in UO_2 and MOX spent fuel. Additionally, the latest libraries (JEFF-3.3, it use the same data as JENDL-4.0u (July, 2013)) do not consider the latest experiment data as shown in Fig. 12. Consequently, the cross section of ^{238}Pu would be better to be improved as first priority from view point of radiotoxicity evaluation.

The second priority was ^{243}Am capture cross section. The capture reaction of ^{243}Am produces ^{244}Cm which had a large radiotoxicity as shown in Fig. 11. Especially, the radiotoxicity of ^{244}Cm is important during 100 years after discharge due to half-life of ^{244}Cm (18.1 y). Additionally, ^{243}Am cross section had the same problem of ^{238}Pu cross section; the latest library data (JENDL-4.0u) do not consider latest experiment data as shown in Fig. 14 [19] (2014).

Finally, third priority was ^{241}Am. The radiotoxicity of ^{241}Am was not so high. However, radiotoxicity of ^{241}Am was important to design of radioactive waste disposal site. Because, the half-life of ^{241}Am (432.6 y) is longer than ^{238}Pu (87.7 y) and ^{244}Cm (18.1 y), therefore, ^{241}Am had long-term environmental load.

Table 3: Prioritization results of improve requirement for the nuclear data

Priority	**nuclides**	**Energy range**	**Reason and *comment***
1	**Pu-238**	**1meV~1.0eV**	1.**Large weight of radiotoxicity in UO2 and MOX spent fuel** 2.***Latest libraries not considered latest experiment results***
2	**Am-243**	**Resonance of 1.356eV and 1.744 eV**	1.**radiotoxicity of Cm-244 from Am-243 have large impact during 100y from discharge** 2.***Latest libraries not considered latest experiment results***
3	**Am-241**	**0.1~100eV**	1.**Large impact for long term (Am-241) and short term (Cm-242) radiotoxicity.** 2.***Large differences were exist between libraries and experiment results***

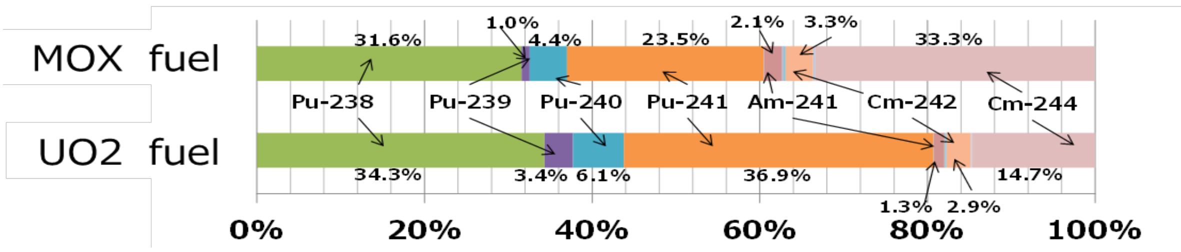


Fig. 11: Relative radiotoxicity composition in the discharged spent fuel

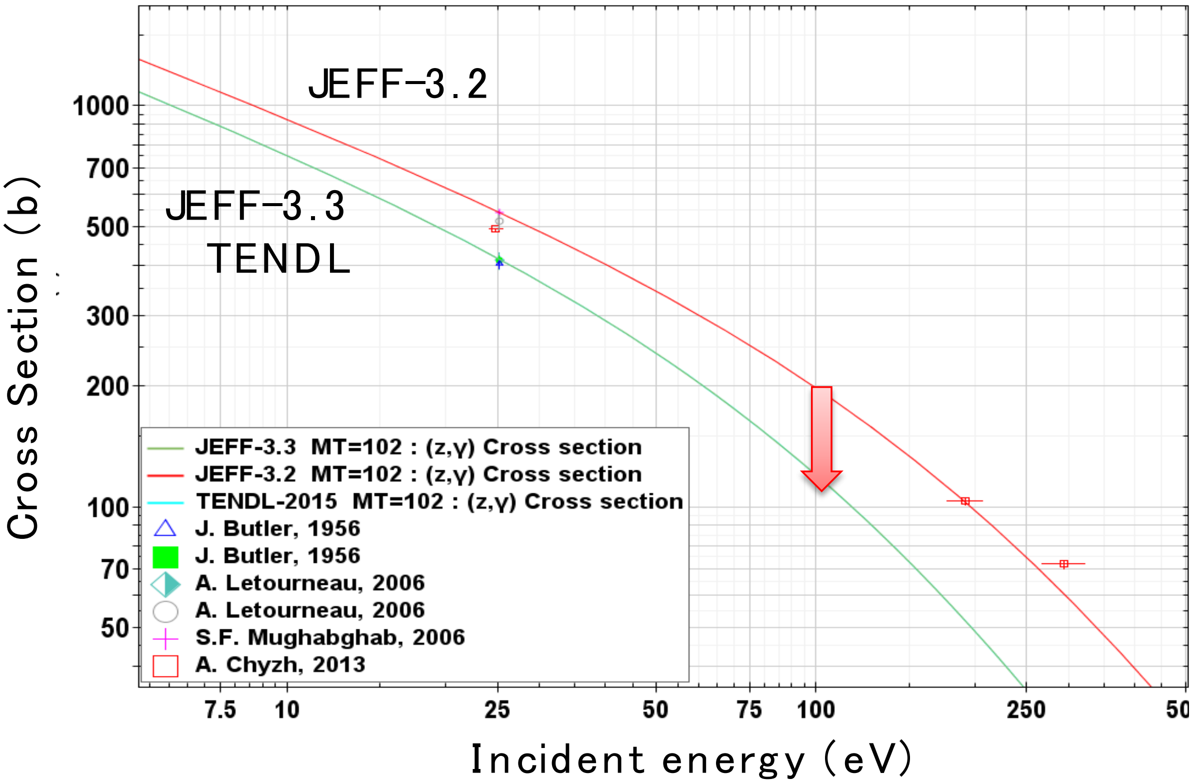


Fig. 12: Comparison of ^{238}Pu neutron capture cross section data of libraries and experiment data [15-18]

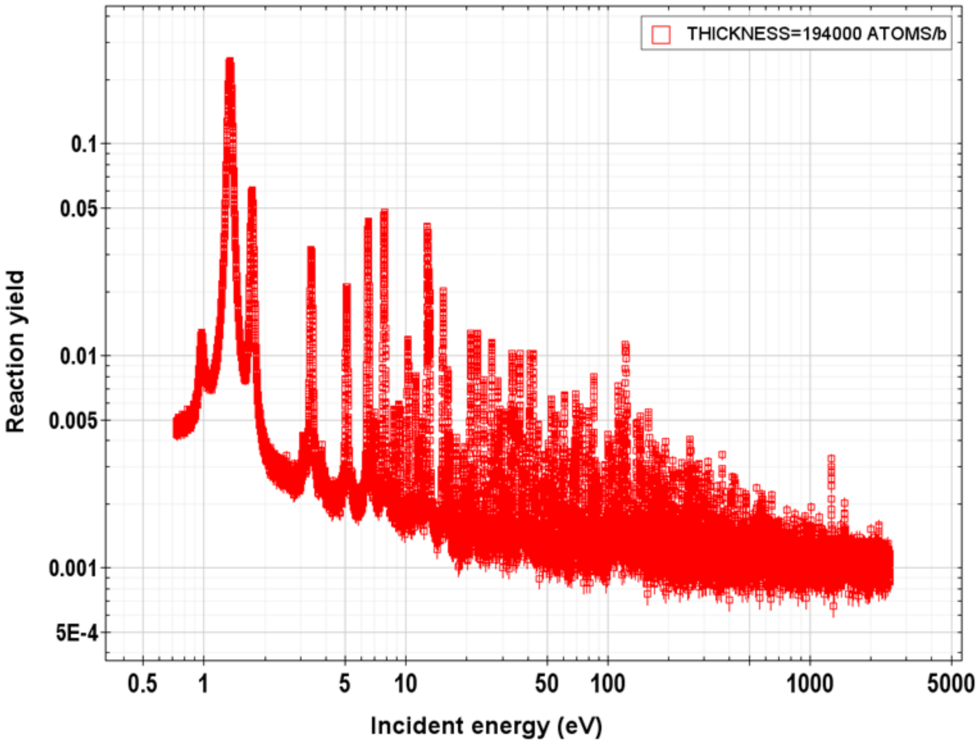


Fig. 14: Experiment data of ^{243}Am capture reaction measured by E. Mendoza et. Al. [19]

4 Conclusion

The cross section sensitivity for the discharged number density of actinide nuclides were demonstrated, furthermore, requirements for the nuclear data were prioritized. For the present study, the priorities of the cross section improvement of ^{238}Pu, ^{243}Am and ^{241}Am were summarized as shown in Table 3.

Moreover, the latest libraries of these nuclides uses some common data, namely, important nuclear data for environmental load estimation were shared between libraries. Therefore, if these nuclear data have a serious error, it would lead systematic error in different libraries. Additionally, the latest nuclear data libraries of ^{238}Pu, ^{243}Am and ^{241}Am have large difference from the latest experiment data. Hence, these nuclides still have room to improve theoretically and experimentally.

Especially, ^{238}Pu, ^{243}Am and ^{241}Am have a large impact to estimate radiotoxicity, decay heat, the volume of radioactive waste and the area of final disposal site. Therefore, improvement in the accuracy of these cross section are important for the utilization of nuclear energy, additionally, these nuclides cross section improvement are clearly important for the future of nuclear industry.

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