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A Comparative Study on Effective One-Group Cross-Sections of ORIGEN and FISPACT to Calculate Nuclide Inventory for Decommissioning Nuclear Power Plant

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ABSTRACT

Background: The radionuclide inventory calculation codes such as ORIGEN and FISPACT collapse neutron reaction libraries with energy spectra and generate an effective one-group cross-section. Since the nuclear cross-section data, energy group (g) structure, and other input details used by the two codes are different, there may be differences in each code's activation inventory calculation results. In this study, the calculation results of neutron-induced activation inventory using ORIGEN and FISPACT were compared and analyzed regarding radioactive waste classification and worker exposure during nuclear decommissioning.

Materials and Methods: Two neutron spectra were used to obtain the comparison results: Watt fission spectrum and thermalized energy spectrum. The effective one-group cross-sections were generated for each type of energy group structure provided in ORIGEN and FISPACT. Then, the effective one-group cross-sections were analyzed by focusing on ^{59}Ni, ^{63}Ni, ^{94}Nb, ^{60}Co, ^{152}Eu, and ^{154}Eu, which are the main radionuclides of stainless steel, carbon steel, zircalloy, and concrete for decommissioning nuclear power plant (NPP).

Results and Discussion: As a result of the analysis, ^{154}Eu and ^{59}Ni may be overestimated or underestimated depending on the code selection by up to 30%, because the cross-section library used for each code is different. When ORIGEN-44g, -49g, and -238g structures are selected, the differences of the calculation results of effective one-group cross-section according to group structure selection were less than 1% for the six nuclides applied in this study, and when FISPACT-69g, -172g, and -315g were applied, the difference was less than 1%, too.

Conclusion: ORIGEN and FISPACT codes can be applied to activation calculations with their own built-in energy group structures for decommissioning NPP. Since the differences in calculation results may occur depending on the selection of codes and energy group structures, it is appropriate to properly select the energy group structure according to the accuracy required in the calculation and the characteristics of the problem.

Keywords: Effective One-Group Cross-Section, Energy Group Structure, ORIGEN, FISPACT, Decommissioning NPP, Activation Inventory Calculation

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Introduction

A decommissioning plan for nuclear power plants (NPPs) should be established by considering the residual radioactivity. Due to the radioactivity during decommissioning NPP, the decommissioning plan should be well evaluated to consider waste management, radiation protection, and environmental impacts. For an efficient decommissioning plan, it is essential to predict the radioactivity of NPPs accurately because it can induce cost reduction, efficiency improvement, flexible plans, radioactive waste minimization, and worker safety in decommissioning NPP [1].

In the radioactive wastes of decommissioning NPPs, the principal component of the radioactive inventory is the activation of the construction materials. Contamination of the decommissioning NPPs results from radioactive releases from the fuel, together with the activated products of corrosion and erosion which occurred during normal operation or unplanned events. For neutron-activated wastes, the radioactivity concentration is the highest near the reactor core. As the distance from the core increases, the concentration of the radionuclide inventory generally decreases. With respect to the commercial NPPs that have been operated for several decades, more than 99% of the total radioactivity are neutron-activated wastes at decommissioning [2].

Neutron-activated wastes in or near the core emit a lot of radiation, making them difficult to access during the decommissioning planning stage. For an efficient decommissioning plan, the radioactivity should be well-evaluated through computational simulation and verified with partial samplings and laboratory analysis. In this study, the activation results using Oak Ridge Isotope GENeration (ORIGEN) and FISPACT codes were compared and analyzed mainly according to the energy group structures.

Materials and Methods

For the inventory calculation, the nuclear cross-sections should be considered for several 10 thousand reactions to many radionuclides. However, it takes huge computational time to treat all the reactions using continuous or multi-group cross-sections. Thus, the effective one-group cross-section is generally calculated by collapsing the continuous or multi-group cross-sections with neutron spectrum using the following equation:

$$\bar{\sigma} = \frac{\int \sigma(E)\phi(E)dE}{\int \phi(E)dE}. \tag{1}$$

where $\bar{\sigma}$ is the effective one-group cross-section, $\sigma(E)$ is the energy-dependent cross-section, and $\phi(E)$ is the energy-dependent neutron flux.

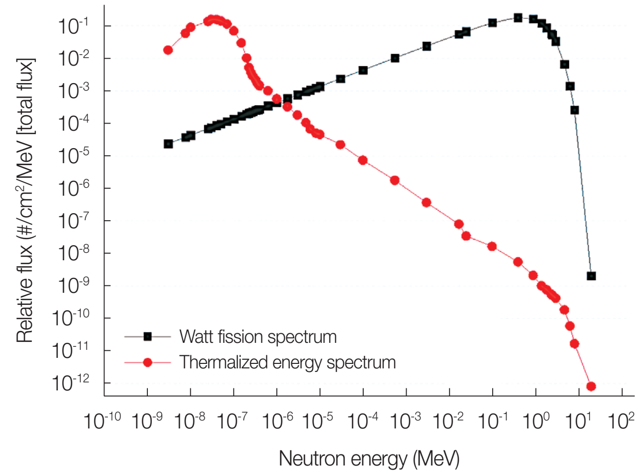

Fig. 1. Watt fission spectrum and thermalized energy spectrum in this study.

1. Neutron Energy Spectrum

The neutron-induced activation rate such as (n,γ), (n,p), (n,xn) depends on the neutron energy. For example, the (n,γ) reaction is mainly caused by the thermal and resonance neutrons, and the (n,p) reaction dominantly by fast neutrons. Therefore, the following neutron spectra were used to account for the differences in the dominant neutron energy range depending on the reaction: Watt fission spectrum and thermalized energy spectrum. The Watt fission spectrum employed in this work is the neutron energy distribution of fission for uranium whose average neutron energy is about 1 MeV. Hence, it was selected to consider the reactions for the high-energy range. Also, the thermalized energy spectrum was, in this work, generated to consider the low neutron energy range at the calandria of the Canada Deuterium Uranium (CANDU)-6 reactor, in which low energy neutrons are dominant. Fig. 1 shows those spectra used in this study. The thermalized neutron spectrum in the pressure tube of CANDU-6 reactor was, in this work, calculated by using Monte Carlo N-Particle (MCNP) [3] to see the effect on the activated nuclide inventory at the energy region in which thermal neutrons are dominant. The effective one-group cross-section using the thermalized spectrum was employed for the ORIGEN and FISPACT calculation.

2. Computational Codes

ORIGEN in the SCALE code package [4], used as a code for licensing of the United States Nuclear Regulatory Commission (NRC), is one of the most widely used computational codes for evaluating a radionuclide inventory. The effective one-group cross-section library can be produced using the

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Table 1. Energy Group Structure of ORIGEN and FISPACT Computational Code

Energy	ORIGEN					FISPACT				
	238g	200g	49g	47g	44g	315g	175g	172g	100g	69g
< 1 eV	50.0 (20.9)	27.0 (13.4)	21.0 (42.0)	4.5 (9.4)	21.0 (46.7)	43.5 (13.8)	6.5 (3.7)	49.5 (28.6)	5.5 (5.4)	31.5 (45.0)
1 eV–1 keV	119.5 (50.0)	31.5 (15.7)	14.5 (29.0)	7.0 (14.6)	9.5 (21.1)	88.0 (27.8)	28.0 (15.9)	67.0 (38.7)	28.0 (27.7)	20.0 (28.6)
1 keV–1 MeV	47.0 (19.7)	79.5 (39.6)	6.0 (12.0)	18.5 (38.5)	6.0 (13.3)	123.5 (39.1)	79.5 (45.2)	36.5 (21.1)	40.5 (40.1)	14.0 (20.0)
> 1 MeV	22.5 (9.4)	63 (31.3)	8.5 (17.0)	18 (37.5)	8.5 (18.9)	61.0 (19.3)	62.0 (35.2)	20.0 (11.6)	27.0 (26.7)	4.5 (6.4)

Values are presented as number of region (%).
"g" stands for the energy group library.

Table 2. Major Radionuclides by Neutron Activation in Commercial Reactors

Item	Stainless steel	Carbon steel	Concrete	Zircalloy
Radio-waste level classification				
ILW-LLW	^{59}Ni, ^{63}Ni, ^{94}Nb	^{94}Nb	-	^{59}Ni, ^{63}Ni, ^{94}Nb
LLW-VLLW-CW	^{60}Co	^{60}Co	^{152}Eu, ^{60}Co, ^{154}Eu	-
Radiation protection	^{60}Co	^{60}Co	^{152}Eu, ^{60}Co, ^{154}Eu	^{60}Co

ILW, intermediate-level waste; LLW, low-level waste; VLLW, very low-level waste; CW, clearance waste.

COUPLE module with the JEFF multi-group nuclear cross-section [5].

The European Atomic Energy Community (EURATOM) and United Kingdom Atomic Energy Authority (UKAEA) have supported the development of FISPACT [6]. FISPACT was developed based on the FISPIN1 inventory code designed for fission reactor calculations, with the addition of the calculation ability for more nuclides and more reactions compared with FISPIN1. The effective one-group cross-section for FISPACT is based upon the European Activation File (EAF) library [7].

The ORIGEN code converts 44-, 47-, 49-, 200-, and 238-group (g) structures into one valid group section, while the FISPACT code converts 69g, 100g, 172g, 175g, and 315g structures. The FISPACT-315g has "flt," "fis," and "fus" options as a micro-flux weighting, FISPACT-175g "flt" and "fus," FISPACT-172g "fis" and "flt," FISPACT-100g "fus," and FISPACT-69g "fis." The "flt" option applies the arithmetic mean of continuous energy in the interval to the representative nuclear cross-section of the energy bin. In contrast, the "fis" and "fus" options determine the representative cross-section considering the energy distribution for fission and fusion. The characteristics of the energy group structure of ORIGEN and FISPACT are summarized in Table 1.

The effective one-group cross-sections for the nuclides of interest were also generated using MCNP to set as a reference value. MCNP code employs the ENDF/B-VII continuous energy nuclear cross-section library [8]. In this work, the comparison results of effective one-group cross-sections were expressed as a ratio of two codes, ORIGEN/MCNP or FISPACT/MCNP. In the ORIGEN and FISPACT calculations, 238g and 315g were used among several group structures, respectively. The 238g and 315g structures were divided in more detail than other structures and evenly divided into the low and high energy (all energy) sections. Therefore, the uncertainties of the collapsing process applying 238g and 315g structures are the lowest among that of the group structures.

3. Neutron Activated Nuclides of Interest

In Korea, the first pressurized water reactor (PWR), Kori Unit 1, and the first pressurized heavy-water reactor (PHWR), Wolsong Unit 1, have been permanently shut down recently and are preparing to be decommissioned. The neutron-activated nuclides of interest in PWR are nearly the same as those in PHWR since the core structures of both reactors are almost similar.

Stainless steel and carbon steel are are mainly used for reactor core structures of both Kori Unit 1 and Wolsong Unit 1. Zircalloy is one of the key materials in Wolsong Unit 1 for guide tubes, etc., with a low thermal neutron absorption cross-section and strong corrosion resistance from high-temperature cooling water. Reinforced concrete is used as a bio-shield in both NPPs.

Table 2 shows the significant nuclides in radioactive waste classification and worker exposure for neutron-activated stainless steel, carbon steel, zircalloy, and concrete. The elements

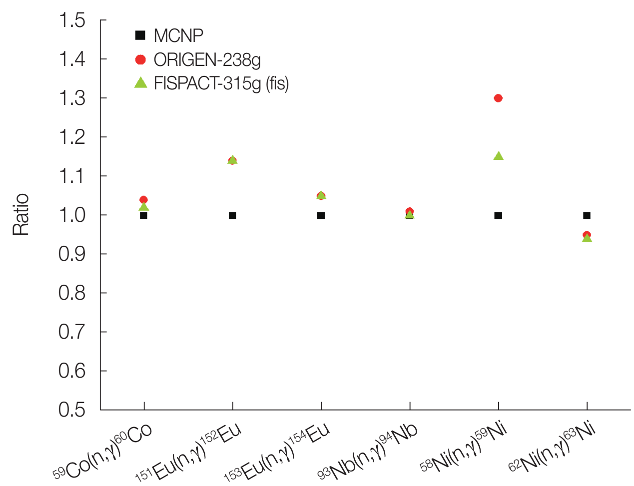

Fig. 2. Collapsed one-group cross-section ratio using Watt fission spectrum.

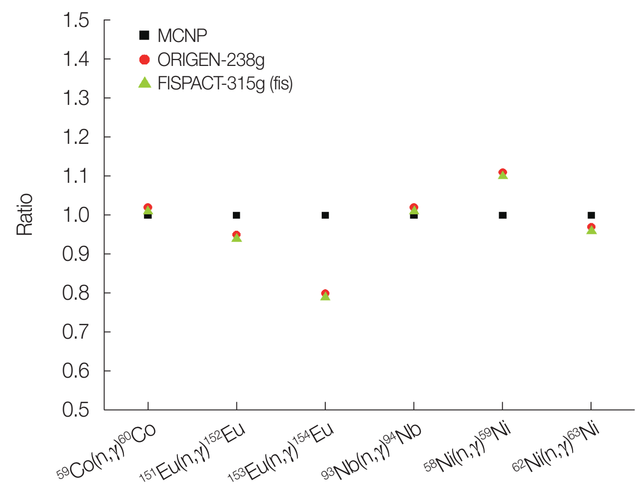

Fig. 3. Collapsed one-group cross-section ratio with thermalized energy spectrum.

of Table 2 (^{59}Ni, ^{63}Ni, ^{94}Nb, ^{60}Co, ^{152}Eu, and ^{154}Eu) are dominant radioactive isotopes made from neutron-induced activation of the materials of the core structures.

Results and Discussion

1. Comparison of Results from Each Computational Code

In order to analyze the difference due to code selection, the results calculated by applying the structure with the largest number of groups embedded in each computer code were compared with the results using MCNP. The effective one-group cross-sections were calculated using MCNP continuous energy, ORIGEN-238g, and FISPACT-315g "fis" multi-group energy structures for the two types of energy spectra. Figs. 2 and 3 show the effective one-group cross-sections as a ratio of those using two codes, ORIGEN/MCNP or FISPACT/MCNP.

The ratios distributed from 0.93 to 1.30 as shown in Fig. 2 if the Watt fission spectrum was used, and from 0.80 to 1.11 for the thermalized energy spectrum as shown in Fig. 3. These comparative results explained that the ratios of the results of two codes to MCNP were caused by their own cross-section libraries for both neutron spectrums. In the case of the watt-fission spectrum being applied (Fig. 2), the largest difference to the MCNP was 1.3 of ORIGEN/MCNP for ^{59}Ni, and in the thermalized energy spectrum (Fig. 3), the largest difference to the MCNP was 0.80 of FISPACT/MCNP for ^{154}Eu.

This difference is due to the nuclear cross-section library used in each code, as shown in Fig. 4. For example, in the $^{153}Eu(n,\gamma)^{154}Eu$ reaction, the effective one-group cross-sections have a difference between applying the Watt fission spectrum and the thermalized energy spectrum. For the Watt fission spectrum, all the ratios for ORIGEN/MCNP and FISPACT/MCNP were close to 1 despite the cross-section difference. On the other hand, for the thermalized energy spectrum, the results were close to 0.8, which is mainly caused by the cross-section differences near thermal energy, as shown in Fig. 5.

As shown in Table 2, the key radionuclides in activated concrete are ^{152}Eu, ^{60}Co, and ^{154}Eu from the viewpoints of the radiowaste level classification and worker exposure. The nuclides are produced by the (n,γ) reaction of europium and cobalt impurities in concrete. The cross-section of ^{152}Eu is tens of times higher than that of ^{154}Eu in the thermal energy region, as shown in Fig. 6. The immediate decommissioning strategy was selected in Korea, and it is expected that decommissioning will be carried out from 5 to 20 years after permanent shutdown. During this period, since the radioactivity of ^{154}Eu is comparable to only 2%–3% of the total radioactivity in the concrete, worker dose induced by ^{154}Eu and the degree of contribution to waste classification will be negligible compared with the ^{60}Co and ^{152}Eu, which contribute more than 90% of the total radioactivity. Therefore, the difference in the nuclear cross-section of ^{154}Eu for each code has little effect on the radiowaste level classification and worker exposure.

The effective one-group cross-sections for the $^{58}Ni(n,\gamma)^{59}Ni$ reaction were also different by the cross-section difference. If the Watt fission spectrum was applied, the ratio between the ORIGEN and MCNP was shown as 1.30, and if the thermalized energy spectrum was involved, the ratio of 1.11. In the

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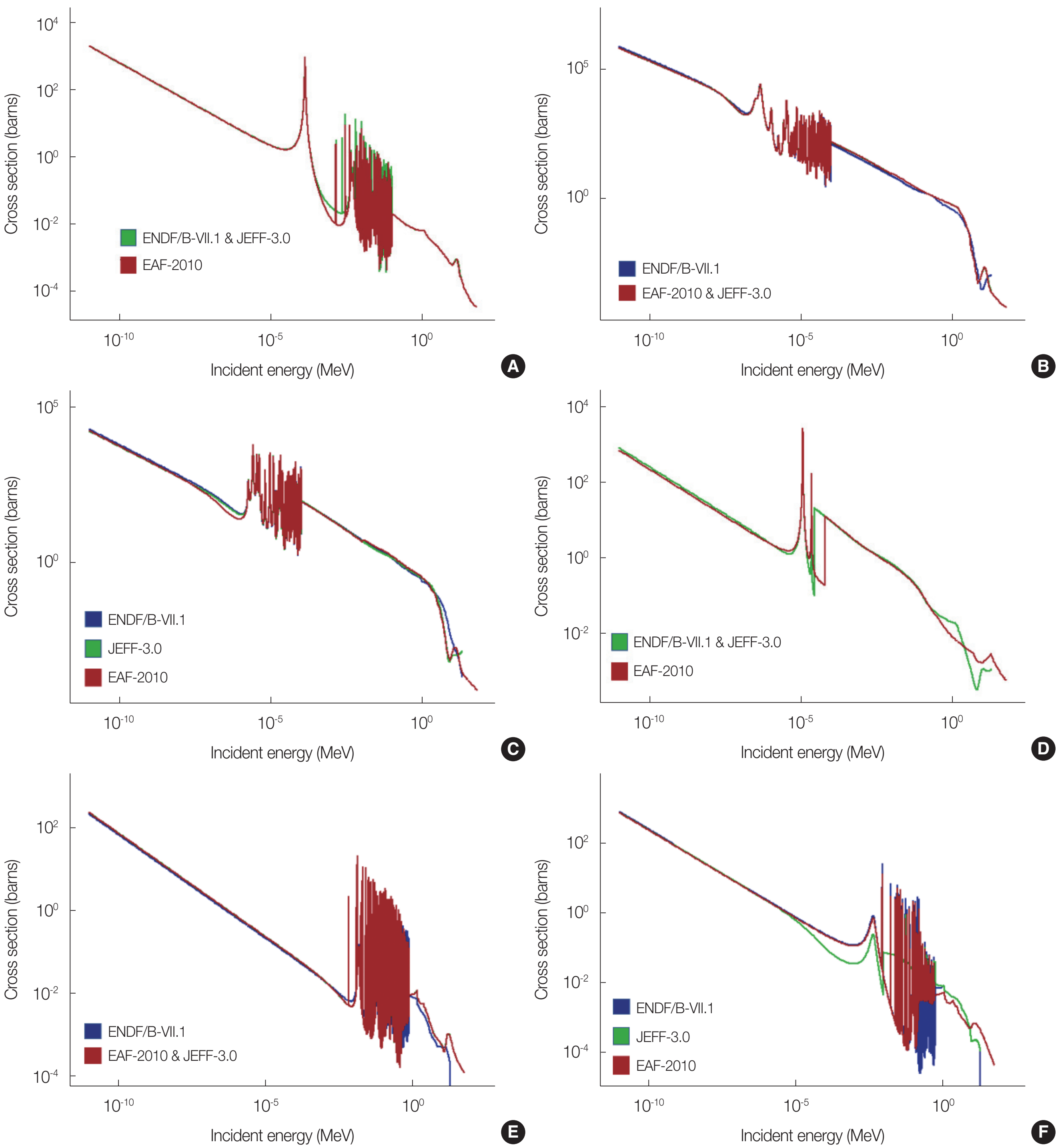

Fig. 4. Neutron-induced activation cross-section for six major nuclides of the three computational codes. (A) $^{59}Co(n,\gamma)^{60}Co$. (B) $^{151}Eu(n,\gamma)^{152}Eu$. (C) $^{153}Eu(n,\gamma)^{154}Eu$. (D) $^{93}Nb(n,\gamma)^{94}Nb$. (E) $^{58}Ni(n,\gamma)^{59}Ni$. (F) $^{62}Ni(n,\gamma)^{63}Ni$.

case of FISPACT calculation, it was found to be 1.15 and 1.10, respectively.

Nickel-59, caused by $^{58}Ni(n,\gamma)^{59}Ni$, is generated from nickel, which comprises about 10% of the stainless-steel structure in the core. In terms of the radiowaste level classification, ^{59}Ni is one of the essential nuclides for classifying stainless steel as an intermediate-level waste (ILW) according to the domestic regulations. If some structures made of stainless steel are located inside the core (baffle, shroud, barrel, top/bottom plate, etc., of PWR), the amount of ILW may change depend-

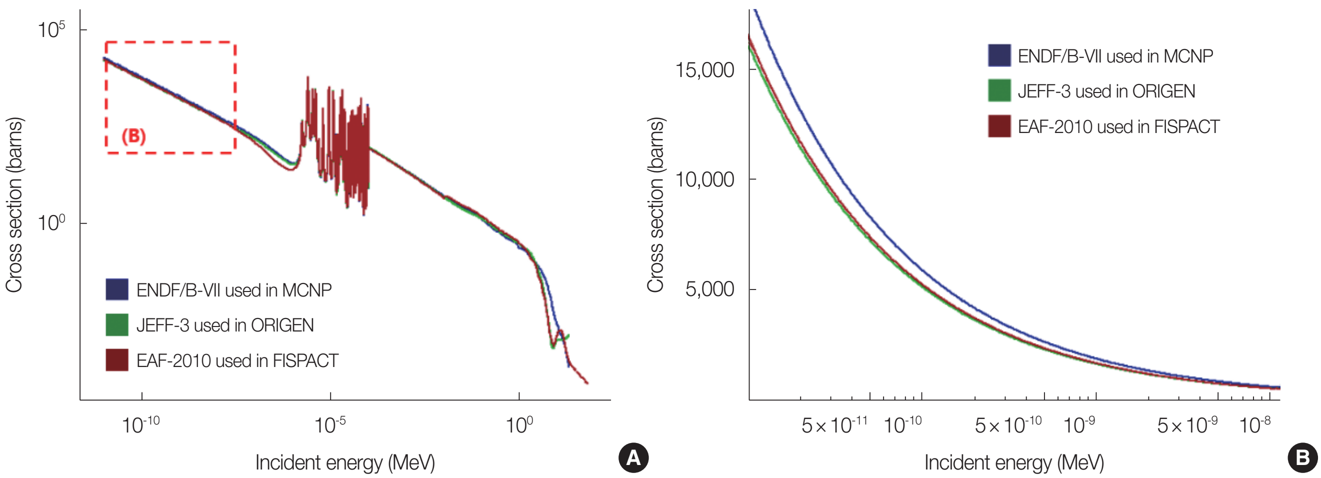

Fig. 5. (A) Neutron-induced activation cross-section of the three computational codes for ^{153}Eu(n,γ)^{154}Eu reaction (log-log scale) and (B) cross-sections of the reaction in the thermal neutron energy region shown in the red dotted box in (A) (log-linear scale).

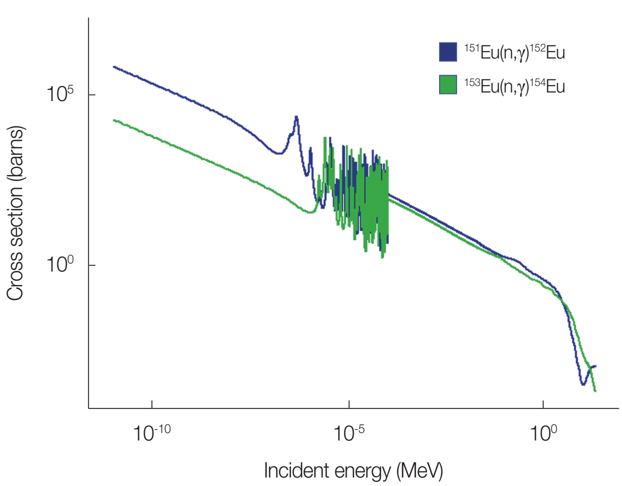

Fig. 6. Neutron-induced activation cross-section of ^{151}Eu(n,γ)^{152}Eu and ^{153}Eu(n,γ)^{154}Eu reaction (log-log scale).

ing on the application of the nuclear cross-section.

2. Comparison of Results using Various Energy Group Structures in Each Code

In this section, differences due to the selection of nuclear cross-sections are excluded, and only the effect of cross-section collapsing was considered. Therefore, ORIGEN-238g and FISPACT-315g (fis) were referred, respectively, and the calculation results for other group structures embedded in each code were compared and analyzed. The calculation results are shown in Tables 3 and 4.

If the Watt fission spectrum was applied to the ORIGEN calculations, the ratios for each energy group structure over ORIGEN-238g were calculated to be 0.95–1.05. In particular, if the thermalized energy spectrum was applied, the ratios for 44g/238g and 49g/238g were calculated as 1.00–1.01.

Since ORIGEN was developed originally to calculate the inventory of nuclear fuel, it has built-in multi-group nuclear cross-sections based on the spectrum of the core region similar to the thermalized energy spectrum of this study. It is, therefore, noted that the effective one-group cross-section was relatively well predicted compared with that using FISPACT, even though only five groups are employed below 1 eV in the case of 47g. Nevertheless, using 47g and 200g is likely to over/underestimate the results compared with using 44g and 49g.

For the case of the FISPACT code, the trend of the ratios using the Watt fission spectrum and the thermalized energy spectrum is different according to the energy group structure. If the Watt fission spectrum was applied, all the ratios were calculated to be 0.90–1.03 even if the group structures were different, which are deemed to be relatively small. If the thermalized energy spectrum was involved, the ratios for 100g/315g and 175g/315g were calculated up to 1.71 and 1.73. Since FISPACT is a tool that can evaluate all neutron environments, such as nuclear fission and fusion, 175g and 100g are not recommended in the systems where the reaction in the low energy region is dominant, such as a fission reactor.

For the FISPACT-315g structure in which the energy bins are finely divided, no effect is expected for each energy region due to the micro-flux weighting options. Also, a similar trend was observed in the 172g structure.

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Table 3. Collapsed One-Group Cross-Section Ratio Using Watt Fission Spectrum

Neutron-induced reaction	ORIGEN					FISPACT								
	238g	200g	49g	47g	44g	315g			175g		172g		100g	69g
						fis	flt	fus	flt	fus	fis	flt	fus	fis
$^{59}Co(n,\gamma)^{60}Co$	1.00	0.97	1.04	0.99	1.04	1.00	1.00	1.00	0.95	0.98	0.93	1.03	0.99	0.90
$^{151}Eu(n,\gamma)^{152}Eu$	1.00	1.00	1.04	1.03	1.04	1.00	1.00	1.00	1.00	0.99	1.00	1.00	1.01	1.00
$^{153}Eu(n,\gamma)^{154}Eu$	1.00	1.00	1.03	1.02	1.03	1.00	1.00	1.00	0.99	1.00	1.00	0.99	1.00	1.00
$^{93}Nb(n,\gamma)^{94}Nb$	1.00	1.00	1.03	0.99	1.03	1.00	1.00	1.00	1.01	1.01	1.01	1.01	1.01	1.03
$^{58}Ni(n,\gamma)^{59}Ni$	1.00	0.98	1.05	0.98	1.05	1.00	1.00	1.00	1.01	1.01	1.02	1.02	1.02	1.03
$^{62}Ni(n,\gamma)^{63}Ni$	1.00	1.05	1.01	1.05	0.99	1.00	1.00	1.00	1.00	1.00	1.01	1.01	1.01	1.01

"g" stands for the energy group library. "flt," "fis," and "fus" indicate an option as a micro-flux weighting.

Table 4. Collapsed One-Group Cross-Section Ratio Using Thermalized Neutron Energy Spectrum

Neutron-induced reaction	ORIGEN					FISPACT								
	238g	200g	49g	47g	44g	315g			175g		172g		100g	69g
						fis	flt	fus	flt	fus	fis	flt	fus	fis
$^{59}Co(n,\gamma)^{60}Co$	1.00	0.99	1.00	0.96	1.00	1.00	1.00	1.00	1.24	1.22	0.99	0.99	1.35	1.00
$^{151}Eu(n,\gamma)^{152}Eu$	1.00	0.99	1.01	1.03	1.01	1.00	1.00	1.00	1.73	1.46	0.99	0.99	1.71	0.99
$^{153}Eu(n,\gamma)^{154}Eu$	1.00	0.99	1.00	0.95	1.00	1.00	1.00	1.00	1.30	1.28	0.99	0.99	1.46	1.00
$^{93}Nb(n,\gamma)^{94}Nb$	1.00	0.99	1.00	0.96	1.00	1.00	1.00	1.00	1.23	1.21	0.99	0.99	1.34	1.00
$^{58}Ni(n,\gamma)^{59}Ni$	1.00	0.99	1.00	0.96	1.00	1.00	1.00	1.00	1.25	1.22	1.00	1.00	1.35	1.00
$^{62}Ni(n,\gamma)^{63}Ni$	1.00	0.99	1.00	0.96	1.00	1.00	1.00	1.00	1.24	1.22	1.00	1.00	1.34	1.00

"g" stands for the energy group library. "flt," "fis," and "fus" indicate an option as a micro-flux weighting.

Conclusion

This study compared effective one-group cross-sections calculated by ORIGEN and FISPACT for the major radioactive nuclides in decommissioning NPP. The factors causing the difference in the results were analyzed: nuclear cross-section library and interval of energy group structure. ORIGEN and FISPACT code generate the effective one-group cross-section from multi-group energy spectra. Because each code uses different cross-section libraries, meaningful differences may exist in inventory calculations for specific nuclides such as ^{154}Eu and ^{59}Ni. Regarding the radiowaste level classification and worker exposure, an effect from ^{154}Eu has less than 10% of an impact of ^{152}Eu produced from the same europium impurity. For ^{59}Ni isotope, depending on the nuclear cross-section employed in the codes, the ^{59}Ni activation inventory will vary up to 30%, which may over/underestimate the amount of ILW for decommissioning NPP. Additionally, for ^{60}Co nuclides known to be the most important for worker exposure, a slight difference existed in the cross-section libraries used in the two codes. Judgment of the validity of the nuclear cross-section data used for each code is beyond the scope of this study and is therefore not further considered here. Except for some nuclides that differ depending on the nuclear cross-section library, the difference in the calculation results by applying each code seems to be tiny.

ORIGEN-47g, ORIGEN-200g, FISPACT-100g, and FISPACT-175g are mainly used for the application to fusion or neutron shielding problems. The four types of group structure are not recommended for fission furnace activation inventory calculations, even though the results will be similar to ORIGEN-238g and FISPACT-315g in some cases. On the other hand, ORIGEN-44g, 49g, 238g, and FISPACT-69g, 172g, 315g are recommended for activation calculations in fission reactors. Calculation results according to the energy group structure of the above six types with ORIGEN and FISPACT show a difference of less than 1% in the thermalized energy spectrum, which is similar to the environment of a nuclear fission reactor. In other words, all of the key nuclides affecting the radioactive waste classification and worker exposure due to neutron-induced activation in NPPs are produced by (n,γ) reactions with a high nuclear cross-section in low energy. Therefore, the results using ORIGEN-44g and -49g having well-subdivided group structure in low energy region agree well with those using ORIGEN-238g, and, in a similar way, the results using FISPACT-69g and -172g are also consistent with those using FISPACT-315g. It is, therefore, appropriate to properly select the energy group structure according to the accuracy required in the calculation and the characteristics of the problem.

Conflict of Interest

No potential conflict of interest relevant to this article was reported.

Author Contribution

Conceptualization: Cha G. Data curation: Cha G. Funding acquisition: Kim M, Kim H. Methodology: Cha G, Lee M. Visualization: Cha G, Lee M. Writing - original draft: Cha G. Writing - review & editing: Kim S. Investigation: Cha G, Lee M. Supervision: Kim S. Validation: Kim S.

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