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Cross-section-induced uncertainty evaluation of MA sample irradiation test calculations with consideration of dosimeter data

Kazuteru Sugino [a,*], Kazuyuki Numata [b], Makoto Ishikawa [c], Toshikazu Takeda [d]

[a] Fast Reactor Cycle System Research and Development Center, Japan Atomic Energy Agency, 4002, Narita-cho, Oarai-machi, Ibaraki 319-1393, Japan
[b] NESI Inc., 4002, Narita-cho, Oarai-machi, Ibaraki 319-1313, Japan
[c] Nuclear Science and Engineering Center, Japan Atomic Energy Agency, 2-4 Shirakata, Tokai-mura, Ibaraki 319-1195, Japan
[d] Research Institute of Nuclear Engineering, University of Fukui, 1-2-4 Kanawa-cho, Tsuruga-shi, Fukui 914-0055, Japan

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ABSTRACT

In MA sample irradiation test data calculations, the neutron fluence during irradiation period is generally scaled by using dosimetry data in order to improve calculation accuracy. In such a case, appropriate correction is required to burnup sensitivity coefficients obtained by the conventional generalized perturbation theory because some cancellations occur in the burnup sensitivity coefficients. Therefore, a new formula for the burnup sensitivity coefficient has been derived with the consideration of the neutron fluence scaling (NFS) effect. In addition, the cross-section-induced uncertainty is evaluated by using the obtained burnup sensitivity coefficients and the covariance data based on the JENDL-4.0.

1. Introduction

For the efficient reduction of radioactive waste and toxicity, minor actinide (MA) transmutation with the utilization of fast reactors is one of the most effective prospectives. To attain the effective MA transmutation, the validation and the improvement of MA cross-section data are indispensable. For this purpose, MA sample irradiation tests have been conducted and calculation results of the post irradiation test data are reported in Kotchetkov et al. (2002) at BN-350 and BOR-60, in Tsujimoto et al. (2003) at PFR, in Tommasi et al. (2006) at Phenix and in Sugino et al. (2012) at Joyo. In calculations of above mentioned MA sample irradiation tests, the neutron fluence is scaled by using the dosimeter data for accurate estimation of the compositions in the MA sample. In addition, burnup sensitivity coefficients of the post irradiation test data are utilized in order to evaluate the calculation results and associated uncertainties (Tommasi et al, 2006; Sugino et al, 2012). However, the previous studies treated only the burnup sensitivity coefficients and associated uncertainties related to direct burnup effects, therefore they ignored those related to a neutron field as undermentioned in detail.

The burnup sensitivity coefficient related to the neutron field can be calculated by the generalized perturbation theory accompanying burnup calculation in addition to those related to the direct burnup effects. The current burnup sensitivity theory has been derived by Williams (1979) and Takeda and Umano (1985), where the flux normalization treatment is formulated by the reactor thermal power.

In producing the neutron flux or fluence with core transport calculations, it is generally normalized by the reactor thermal power. However, the neutron fluence should be scaled by the dosimeter data in the MA sample irradiation test calculations in order to improve its accuracy. Therefore, the normal burnup sensitivity coefficient which is obtained by the conventional formula should be appropriately corrected. This is because the biases, which are induced by the neutron field, e.g., the neutron flux levels and the neutron spectra during the irradiation period, should be cancelled from the MA sample calculation data due to the utilization of the dosimeter data.

In the present paper, a new formula is derived for the burnup sensitivity coefficients taking into account the neutron fluence scaling (NFS). The new formula is applied to the burnup sensitivity calculations of the MA sample irradiation test data at Joyo (Sugino et al., 2012).

By using the new formula, the burnup sensitivity coefficients are calculated. Obtained burnup sensitivity coefficients are utilized

* Corresponding author.
E-mail address: sugino.kazuteru@jaea.go.jp (K. Sugino).

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for evaluation of the cross-section-induced uncertainty is calculated with the nuclear data library JENDL-4.0 (Shibata et al., 2011; Chiba et al., 2011; Iwamoto et al., 2011).

In the present paper, the derivation of the new formula and its application are described in Chap. 2. The MA sample irradiation test calculations for Joyo are briefly introduced in Chap. 3. Subsequently, the evaluation of the cross-section-induced uncertainties by the new formula is reported in Chap. 4. Finally, some concluding remarks complete the present paper.

2. Derivation of a new formula

Evaluated number densities in the irradiated MA sample or dosimeter are dominantly affected by the biases of neutron fluence and cross-sections. Further, applied calculation modelling or approximation also affects the evaluation of the number densities. The present paper assumes that the bias of the calculation modelling or approximation is negligible for simplicity.

Under these assumptions, the bias of the number density ratio in the irradiated MA sample can be presented by the following equation:

$$\Delta C_{MA} = \sum_g \sum_i S^g_{MA,i} \Delta\sigma^g_i + S_{MA,\Phi}\Delta\Phi \tag{1}$$

Here,

ΔX: Bias of any parameter X,
C_{MA}: Number density ratio of the irradiated MA sample,
σ^g_i: Cross-section specified by i of energy group g,
i: Index for nuclide and type of reaction,
$S^g_{MA,i}$: Burnup sensitivity coefficient of $C_{MA,j,k}$ with respect to σ^g_i before NFS,
Φ: Neutron fluence,
$S_{MA,\Phi}$: Burnup sensitivity coefficient of C_{MA} with respect to Φ.

As aforementioned, it is extremely difficult to estimate the uncertainty of neutron fluence. Hence, the dosimeter data should be utilized in the MA sample irradiation test data calculation.

The present study assumes that the U-235 dosimeter is loaded at the same position as the MA sample. Under this assumption, the bias of the number density of the specific FP, e.g. Cs-137 in the U-235 dosimeter can be treated in a similar way to the number density ratio in the MA sample and presented by the following equation:

$$\Delta C_{Dos} = \sum_g \sum_i S^g_{Dos,i} \Delta\sigma^g_i + S_{Dos,\Phi}\Delta\Phi \tag{2}$$

Here,

C_{Dos}: Number density of Cs-137 in the irradiated U-235 dosimeter,
$S^g_{Dos,i}$: Burnup sensitivity coefficient of C_{Dos} with respect to σ^g_i,
$S_{Dos,\Phi}$: Burnup sensitivity coefficient of C_{Dos} with respect to Φ.

Obviously, the burnup sensitivity coefficient with respect to the cross-sections can be calculated by the generalized sensitivity theory accompanying the burnup calculation. The burnup sensitivity coefficients of the Cs-137 number density in the U-235 dosimeter with respect to the cross-sections and neutron fluence can be calculated in the same way as those of the number density ratio in the MA sample, respectively.

The burnup sensitivity coefficient with respect to the neutron fluence can be calculated by taking the difference of number density ratio between the reference state and the fluence increased state in the normal burnup calculations. For the MA sample, the burnup sensitivity coefficients can be obtained by the following formula:

$$S_{MA,\Phi} = \frac{C_{MA}(\Phi + \delta\Phi) - C_{MA}(\Phi)}{\delta\Phi} \tag{3}$$

Here,

δX: Difference or variation of any parameter X.

The burnup sensitivity coefficients for the U-235 dosimeter can be obtained in the same manner.

When utilizing the dosimeter data, the neutron fluence is scaled so as to set the calculation value $C_{Dos,Cal}$ of the U-235 dosimeter data to the experimental value $C_{Dos,Exp}$. Under the assumption that the bias of experimental value is negligible for simplicity in addition to the calculation value, the bias of the U-235 dosimeter data is supposed to be zero as:

$$\Delta C_{Dos} = C_{Dos,Cal} - C_{Dos,Exp} = 0 \tag{4}$$

Under this situation, Eq. (2) can be transformed into the following equation:

$$\Delta\Phi = -\frac{1}{S_{Dos,\Phi}} \sum_g \sum_i S^g_{Dos,i} \Delta\sigma^g_i \tag{5}$$

By substituting Eq. (5) into Eq. (1), the following equation is obtained.

$$\Delta C_{MA} = \sum_g \sum_i \left(S^g_{MA,i} - \alpha \cdot S^g_{Dos,i} \right) \Delta\sigma^g_i \tag{6}$$

Here,

$$\alpha = \frac{S_{MA,\Phi}}{S_{Dos,\Phi}}$$

The burnup sensitivity coefficient after NFS is defined as followings:

$$\tilde{S}^g_{MA,i} = S^g_{MA,i} - \alpha \cdot S^g_{Dos,i} \tag{7}$$

Eq. (7) is the new formula and it shows that the number density ratio in the MA sample has the sensitivity with respect to cross sections due to the NFS.

After the NFS, the uncertainty of the number density ratio in the MA sample is expressed in the matrix form by the next formula:

$$V(C_{MA}) = \tilde{\mathbf{G}}_{\mathbf{MA}} \mathbf{M} \tilde{\mathbf{G}}^{\mathbf{t}}_{\mathbf{MA}} \tag{8}$$

Here,

$V(X)$: Variance of any parameter X,
$\tilde{\mathbf{G}}_{\mathbf{MA}}$: Vector with components of $\tilde{S}^g_{MA,i}$,
$\mathbf{M}$: Covariance matrix of cross-section data.
$\mathbf{t}$: Transpose of vector,

If the dosimeter is not applied, the uncertainty of the number density ratio in the MA sample is expressed by the next conventional formula:

$$V(C_{MA}) = \mathbf{G}_{\mathbf{MA}} \mathbf{M} \mathbf{G}^{\mathbf{t}}_{\mathbf{MA}} + S^2_{MA,\Phi} \cdot V(\Phi) \tag{9}$$

Here,
$\mathbf{G}_{\mathbf{MA}}$: Vector with components of $S^g_{MA,i}$.

Apparently, the conventional evaluation includes the uncertainty due to the neutron fluence though the new evaluation can rationalize it as shown in Eqs. (8) and (9).

3. MA sample irradiation test data at Joyo

The present study treats the calculation data of the MA sample irradiation tests B9 and SMIR-26 conducted at Joyo (Sugino et al., 2012).

The MA samples irradiation tests B9 and SMIR-26 had been conducted at the Joyo MK-II core in 1994 through 1999. In the tests, MA samples had been irradiated for 251 or 276 effective full power days. The present paper treats the MA samples loaded at three positions in the Joyo reactor core as shown in Fig. 1; in the core midplane of the specified irradiation test fuel subassembly (SA) and the core midplane and the upper part of the material irradiation test reflector SA. Neutron field of these three positions can be characterized by the energy dependence of Am-241 capture reaction rate as shown in Fig. 2. "Position F" indicates the energy dependence at the core midplane of the irradiation test fuel SA or position F, which is similar to that of normal driver fuels. "Position R1" indicates the energy dependence at the core midplane of the material irradiation test reflector SA or position R1, which is much softer than that at position F. "Position R2" indicates the energy dependence at the upper part of the material irradiation test reflector SA or position R2, which is further softer than that at position R1.

In each position of the MA samples loaded, several types of dosimeters were also loaded. Without these dosimeter data, it is almost impossible to assure sufficient reliability in calculation results because they would involve unquantified uncertainties due to calculation modelling in the irradiation history, e.g., time dependence of the reactor thermal power, and treatment of neutron transport or estimation of the neutron fluence at the local point such as a MA loaded position. Therefore, the neutron fluence at the MA loaded positions should be estimated with sufficient accuracy or scaled by utilizing the dosimeter data, which causes insurance of reliability in the calculation results. For above mentioned reason, enriched U-235 dosimeter data were utilized for the NFS in the MA sample irradiation test data.

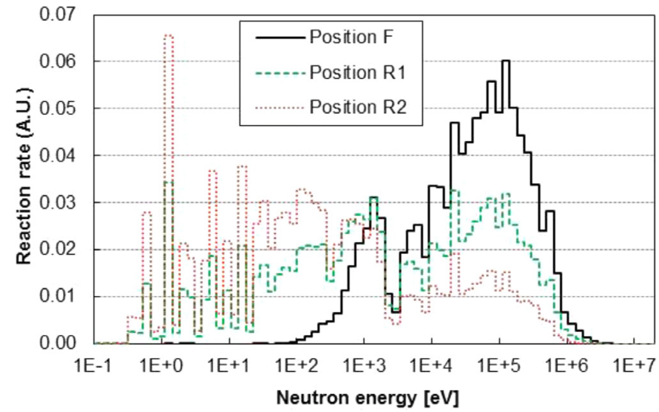

Fig. 2. Comparison in the energy dependence of the Am-241 capture reaction rates among MA loaded positions.

4. Calculations

4.1. Calculation condition

The present paper treats the Am-241 and Am-243 samples for the demonstration of the new formula. As calculation items, specified number density ratios are selected which have major sensitivity coefficients of around unity with respect to the capture cross-sections of the target MA. Concerning the Am-241 sample, the number density ratio of Am-242m to Am-241, where Am-242m is produced by the capture reaction of Am-241 with Am-242g, shows the sensitivity of around unity to the capture cross-section of Am-241. Concerning the Am-243 sample, the number density ratio of Cm-245 to Cm-244, where Cm-244 is produced by the capture reaction of Am-243 and decay of Am-243, has the sensitivity of around unity to the capture cross-section of Cm-244.

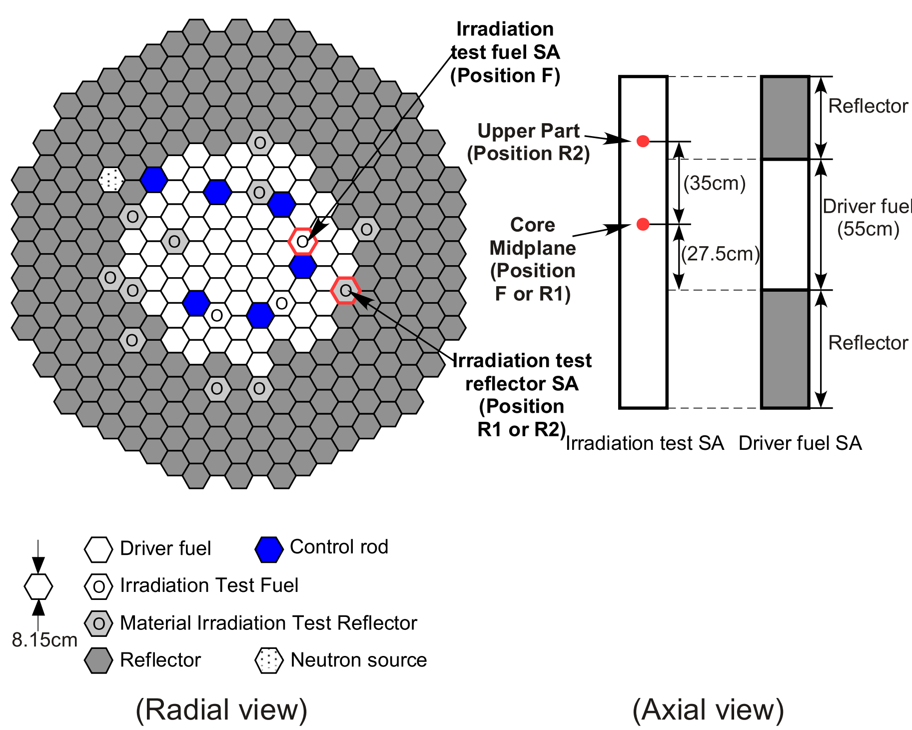

Fig. 1. Core layout of Joyo MK-II and the MA sample loaded positions.

Table 1
Calculation items of the MA sample irradiation tests at Joyo.

Index	Type of MA sample or dosimeter	Type of number density ratio or number density	Cross-section with major sensitivity
A1C	Am-241	Am-242m/Am-241	Am-241 capture
C4C	Am-243	Cm-245/Cm-244	Cm-244 capture
U5F	U-235	Cs-137	U-235 fission

Further, the present paper utilize the U-235 dosimeter. Concretely, measured number density of Cs-137, which was produced mainly by the fission reaction of U-235, are utilized in order to scale the neutron fluence at the MA sample irradiated positions so as to improve the calculation accuracy.

Table 1 presents the summary of aforementioned process, which shows that the compiled data covers the two MAs and U-235 by means of sensitivity to capture and fission cross-sections relating to Am-241, Cm-244 and U-235. Hereafter, indices A1C, C4C and U5F are used for the convenience as described in Table 1.

4.1.1. Burnup sensitivity coefficient with respect to the neutron fluence

As aforementioned, the burnup sensitivity coefficient with respect to the neutron fluence can be calculated by taking the difference of number density ratio between the reference state and the fluence increased state in the normal burnup calculations as indicated by Eq. (3) for the MA sample. Assuming that all over the neutron fluence is proportional to the reactor thermal power, the burnup sensitivity coefficients with respect to the neutron fluence can be rewritten as:

$$S_{MA,\Phi} \approx \frac{C_{MA}(P+\delta P) - C_{MA}(P)}{\delta P} \tag{10}$$

Here,

P: Base reactor thermal power.

The value $C_{MA}(P+\delta P)$ is obtained by increasing only the reactor thermal power in some extent with other parameters fixed in the calculation of $C_{MA}(P)$.

The burnup sensitivity coefficients for the U-235 dosimeter can be obtained in the same manner.

In the present study, the burnup sensitivity coefficients with respect to the neutron fluence is obtained by increasing the reactor thermal power by 10 percent from the base state.

4.1.2. Burnup sensitivity coefficient with respect to the cross-section

The energy dependence of burnup sensitivity coefficients is treated with the 70-group structure. The present study applies the 2-D diffusion theory for the calculation of the burnup sensitivity coefficients, which is performed by the burnup sensitivity calculation code PSAGEP (Tatsumi and Hyoudou, 2004).

Concerning the burnup treatment, a simple straight burnup calculation, which starts with the core of overall fresh fuels and does not treat the fuel exchange during irradiation and cooling periods, is carried out for the burnup sensitivity coefficients.

The burnup sensitivity coefficients consist of five terms, which are direct, number density, neutron flux, adjoint neutron flux and power normalization terms (Takeda and Umano, 1985). In treating the burnup sensitivity coefficients of the number density, the direct and the adjoint neutron flux terms become zero. Therefore, the present paper treats the number density, the neutron flux and the power normalization terms.

Further, the burnup sensitivity coefficient of the number density ratio is given by the difference of those of the number densities. For example, the burnup sensitivity coefficient of the number density ratio of Am-242m to Am-241 is calculated by taking difference of those of the number densities between Am-242m and Am-241.

4.1.3. Cross-section-induced uncertainty

The burnup sensitivity coefficients are utilized for the evaluation of the cross-section-induced uncertainty. In the evaluation, the covariance data of the JENDL-4.0 (Iwamoto et al., 2011) are applied as well as the MA sample irradiation test data calculations for Joyo. In the present paper, the uncertainty is defined by one standard deviation, represented by a 68 percent confidence level.

4.2. Calculation result

4.2.1. Burnup sensitivity coefficient with respect to the neutron fluence

Table 2 presents the calculation results of the burnup sensitivity coefficients of the number densities in the MA samples and the number density ratio in the U-235 dosimeter with respect to the neutron fluence. It is found that the burnup sensitivity coefficients are around unity. This means that the relative changes of the number densities in the irradiated MA sample and dosimeter are almost proportional to the neutron fluence, which are reasonable.

4.2.2. Burnup sensitivity coefficient with respect to the cross-section

As representative examples of the burnup sensitivity coefficients related to the neutron field, three types of the burnup sensitivity coefficients of Index A1C are illustrated in Figs. 3 through 5.

Fig. 3 presents the power normalization term of the burnup sensitivity coefficients with respect to the Pu-239 fission cross-section at position F. The burnup sensitivity coefficients of the dosimeter or Index U5F are similar to those of the MA sample or Index A1C before NFS. By the NFS, significant cancellation occurs in burnup sensitivity coefficients between the MA sample and the dosimeter as intended, which results in almost zeros as shown in the burnup sensitivity coefficients of Index A1C after NFS.

Fig. 4 presents the neutron flux of the burnup sensitivity coefficients with respect to the U-238 inelastic scattering cross-section at position F. The burnup sensitivity coefficients of Index U5F resemble those of Index A1C before NFS with around one third in amplitude. After the NFS, the burnup sensitivity coefficients of Index A1C are not significantly reduced as shown in the burnup sensitivity coefficients of Index A1C after NFS. This is because the energy dependence of the Am-241 capture cross-section is rather different from that of the U-235 fission cross-section in the energy range of above 50 keV and the inelastic scattering by U-238 exclusively affects the neutron spectrum in this energy range.

Fig. 5 presents neutron flux term of the burnup sensitivity coefficients with respect to the Fe-56 elastic scattering cross-section at position R2. The burnup sensitivity coefficients of Index U5F are similar to those of Index A1C before NFS above 100 eV. After the NFS, the burnup sensitivity coefficients of Index A1C are significantly reduced in the energy range of above 100 eV as shown in the burnup sensitivity coefficients of Index A1C after NFS. Slight cancellation in the sensitivity coefficients below 100 eV is due to the large difference in the energy dependence between the Am-

Table 2
Burnup sensitivity coefficients of the number density ratio in the MA samples and the Cs-137 number density in the U-235 dosimeter with respect to the neutron fluence.

Index	Position		
	F	R1	R2
A1C	0.99	1.00	1.00
C4C	1.01	1.02	1.01
U5F	0.95	0.96	0.97

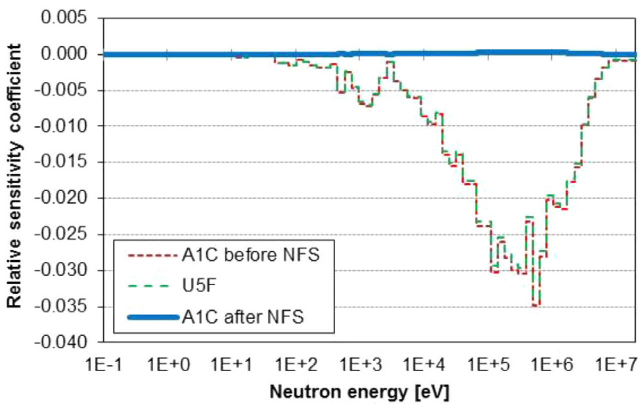

Fig. 3. Power normalization term of the burnup sensitivity coefficients of Index A1C with respect to the Pu-239 fission cross-section at Position F.

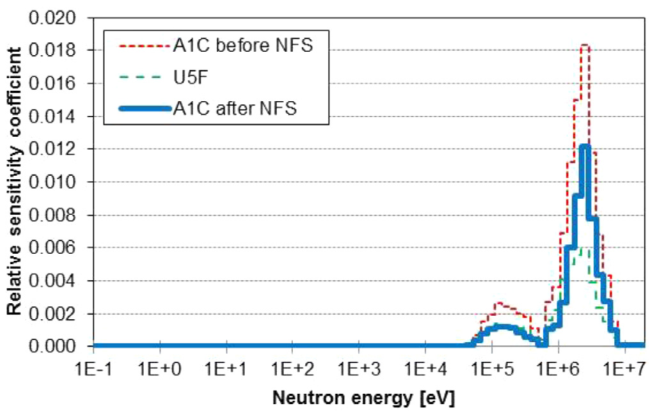

Fig. 4. Neutron flux term of the burnup sensitivity coefficients of Index A1C with respect to the U-238 inelastic scattering cross-section at Position F.

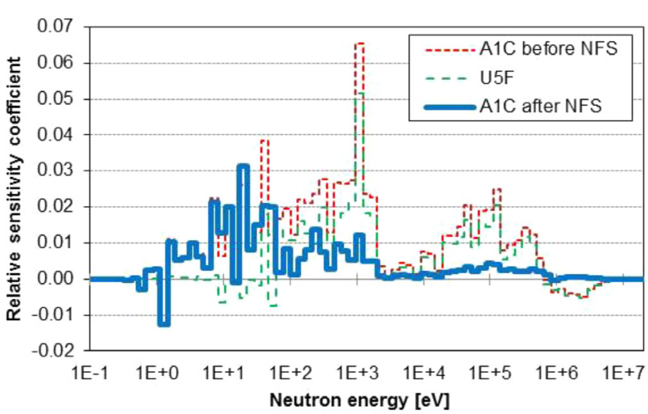

Fig. 5. Neutron flux term of the burnup sensitivity coefficients of Index A1C with respect to the Fe-56 elastic scattering cross-section at Position R2.

241 capture cross-section and the U-235 fission cross-section in this energy range.

Thus, the power normalization terms, which are related to the fuel nuclides only, are mostly canceled by the NFS. The neutron flux terms, which are related to both the fuel and non-fuel nuclides, are canceled to some extent according to the similarity of the energy dependence of the MA capture cross-section to that of the U-235 fission cross-section.

4.2.3. Cross-section-induced uncertainty

Table 3 presents the comparison in uncertainties of each term by the treatment of the NFS. The uncertainties originated by the number density term is slightly increased by the NFS due to the addition of the U-235 fission cross-section contribution. The uncertainties originated by the neutron flux term are reduced to some extent by the NFS as imagined from Figs. 4 and 5. In addition, the uncertainties originated by the power normalization term are mostly eliminated by the NFS as imagined from Fig. 3. Thus, it is confirmed that the NFS effectively reduces the neutron field uncertainty. Furthermore, it is found that the uncertainties originated by the neutron flux term in the reflector region as indexed by R1 and R2 is larger than those in the fuel region as indexed by F. This is because contributions by steel nuclides are larger in the reflector region than those in the fuel region.

The total cross-section-induced uncertainties without NFS are larger than those with NFS due to the difference in those originated by the neutron field.

Concerning the uncertainties applied in the previous study (Sugino et al., 2012), which correspond to those of the number density term with NFS in Table 3. It is remarked that the uncertainties in the previous study are systematically underestimated and should be replaced with those of the total ones with NFS in Table 3. Nevertheless, the experimental and the modeling uncertainties (Sugino et al., 2012) are almost smaller than the cross-section-induced uncertainty, which leads to the re-confirmation of the effectiveness of the present test data with higher reliability.

Thus, the new formula enables us to quantify the uncertainties of the MA sample irradiation test calculation due to the cross-section with NFS procedure. Further, the new formula has rationalized the uncertainty due to the cross-section in comparison with the conventional formula in the burnup sensitivity coefficient calculations.

5. Conclusion

A new formula was derived to correct the burnup sensitivity coefficients taking into account the neutron fluence scaling due to the dosimetry data utilization in the MA sample irradiation test data calculations. Subsequently, the uncertainty induced by the cross-section was evaluated by using the obtained burnup sensitivity coefficients and the nuclear data library JENDL-4.0.

It was found that the new formula can rationalize the uncertainty due to the cross-section originated by the neutron

Table 3
Results of the evaluation in the cross-section-induced uncertainty (%).

Index	Position	Number density		Neutron flux		Power normalization		Total	
		Without NFS	With NFS	Without NFS	With NFS	Without NFS	With NFS	Without NFS	With NFS
A1C	F	6.5_8	6.6_2	1.6	1.0	0.3	0.0	6.8	6.7
	R1	6.6_9	6.7_4	2.6	1.4	0.3	0.0	7.2	6.9
	R2	7.2_5	7.2_9	4.9	2.0	0.4	0.0	8.7	7.6
C4C	F	25.0_7	25.0_8	1.6	0.9	0.4	0.0	25.1	25.1
	R1	11.8_1	11.8_4	6.5	5.4	0.4	0.0	13.5	13.0
	R2	9.8_9	9.9_3	9.6	6.8	0.4	0.0	13.8	12.0

irradiation field in comparison with the conventional formula where the neutron fluence scaling is not taken into account.

Thus, the present study clarified the effectiveness of the new formula by utilizing the neutron fluence scaling with the dosimeter data.

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