Evaluation of Depletion Uncertainty for Spent Fuel Storage Pool using Monte Carlo Random Sampling by Considering Boron Concentration in Depletion Calculation

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1. Introduction

Criticality safety evaluation of spent nuclear fuels (SNFs) employing burnup credit (BUC) requires an uncertainty evaluation of the composition of actinides and fission products from irradiation in a reactor. Since 2011, the U.S. NRC has required the application of the revised guidelines for criticality analysis and operation of spent fuel storage. In addition, the domestic regulatory authorities are demanding the application of the revised guidance. The guidelines recommend the application of boron credit and BUC, including consideration of axial burnup profile, criticality code validation, and uncertainty evaluation by depletion calculations [1]. In order to evaluate the uncertainty by depletion, it is important to consider the inventory of radionuclides that reflects the fuel assembly designs and operation history of the reactor. Operation history parameters include nuclear fuel temperature, moderator temperature, boron concentration in moderator, depletion period, decay period, and discharge burnup.

This paper aims (1) to assess the effect of the change in the boron concentration in moderator on the composition of nuclides of pressurized water reactor (PWR) SNFs and then (2) to analyze the isotopic uncertainties in criticality analysis using the Monte Carlo random sampling method for the spent fuel storage pool.

2. Methods and results

2.1. Computer code system

The TRITON sequence is a multipurpose SCALE control module for transport, depletion, sensitivity, and uncertainty analysis for reactor physics applications [2]. In this study, using SCALE 6.2/TRITON with ENDF/B-VII.1-based 252-group cross-section libraries, the inventories of actinides and fission products were calculated through two-dimensional (2-D) modeling of the nuclear fuel assemblies. The nuclides inventories of the SNFs were estimated by applying the axial burnup profile for eighteen axial nodes.

KENO-VI is a Monte Carlo criticality transport program used to calculate k_{eff}, flux, reaction rates, and other data for three-dimensional (3-D) systems [2]. With KENO-VI, we modeled the region II spent fuel storage rack (SFSR) of domestic WH-F type reactors assuming an infinite array. The nuclides applied for BUC were nine major actinides (i.e., ^{234}U, ^{235}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, and ^{241}Am) and nineteen fission products (i.e., ^{95}Mo, ^{99}Tc, ^{101}Ru, ^{103}Rh, ^{109}Ag, ^{133}Cs, ^{143}Nd, ^{145}Nd, ^{147}Sm, ^{149}Sm, ^{150}Sm, ^{151}Sm, ^{152}Sm, ^{151}Eu, ^{153}Eu, ^{155}Gd, ^{236}U, ^{237}Np, and ^{243}Am) [3]. The workflow of the estimate for SNFs isotopes inventory and random sampling criticality calculation for SFSR is presented in Fig. 1.

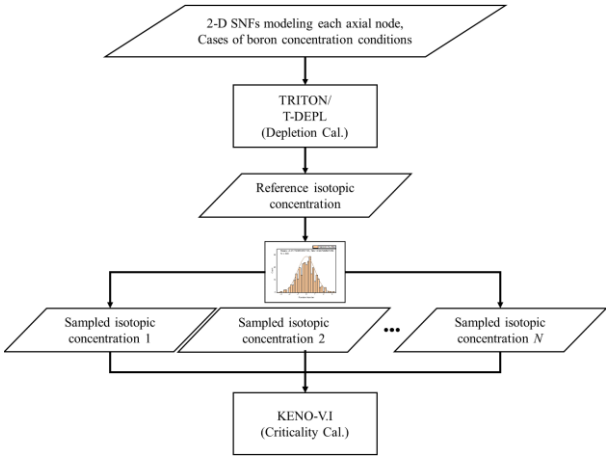

Fig. 1. Workflow of the evaluation of depletion uncertainty by considering boron consideration.

2.2. Isotopic inventory Estimation of SNFs

The isotopic inventories were estimated for the 17V5H type nuclear fuel assembly. The assembly was assumed 5.0 wt% initial ^{235}U enrichment and 40 MWd/kg discharge burnup. The bounding axial burnup profile is shown in Fig. 2 [4]. In order to calculate the difference in the isotopes inventory of SNFs by the boron concentration in the moderator during reactor operation, four cases of boron concentration conditions were considered during the depletion calculation: three cases using constant boron concentrations of 0 ppm, 600 ppm, 1,200 ppm and a letdown model from 1,200 ppm at the BOC to 10 ppm at the EOC similar to the actual operating conditions. The nuclear fuel assembly was depleted for three cycles; each cycle was eighteen months, and the preventive maintenance period or refueling interval was assumed to be one month between cycles. Fig. 3 presents the boron concentration over three cycles in each condition.

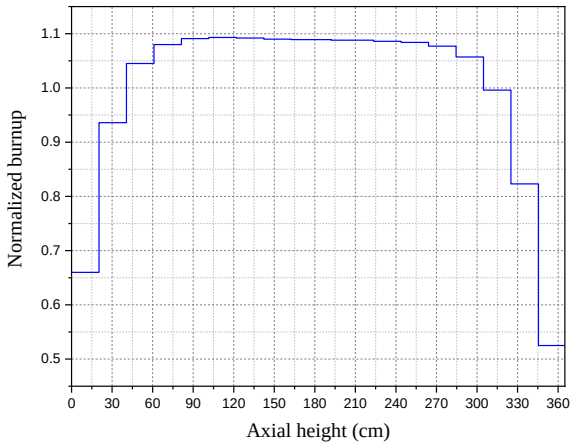

Fig. 2. Bounding axial burnup profile at 40 MWd/kg.

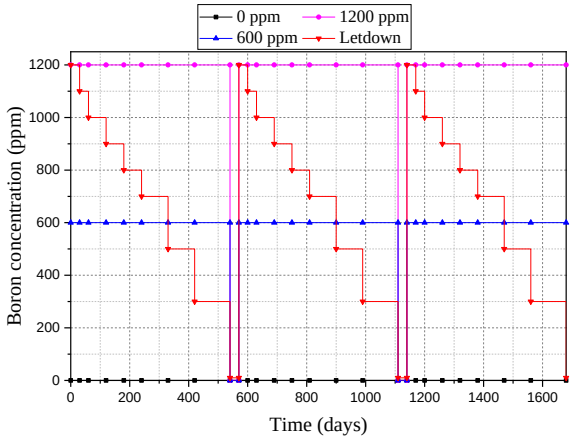

Fig. 3. Boron concentration conditions.

The neutron spectrum is hardened when the nuclear fuel assembly depletion calculation was conducted with a high boron concentration. ^{239}Pu is a very important nuclide because it has a larger thermal fission cross-section. This nuclide is a product of the transmutation and subsequent nuclear decay of fertile isotope ^{238}U. The transmutation and decay chain is shown in Eq. (1) [5].

$$^{238}_{92}\mathrm{U} + {}^{1}_{0}\mathrm{n} \rightarrow {}^{239}_{92}\mathrm{U} \xrightarrow{\beta^-} {}^{239}_{93}\mathrm{Np} \xrightarrow{\beta^-} {}^{239}_{94}\mathrm{Pu} \quad (1)$$

The capture of a neutron in the ^{238}U nucleus yields ^{239}U. The half-life of ^{239}U is approximately 23.5 minutes. ^{239}U decays to ^{239}Np, whose half-life is 2.36 days. ^{239}Np decays to ^{239}Pu. Higher mass number isotopes of plutonium (e.g., ^{240}Pu, ^{241}Pu, and ^{242}Pu) are also created by neutron radiative capture. Fig. 4 shows the transmutation and decay chains from ^{238}Pu to ^{244}Cm. The harder spectrum when boron is considered in moderator leads to higher fissile plutonium contents. Another important nuclide is ^{241}Am due to its high thermal fission cross-section, which is mainly formed by the beta-decay of ^{241}Pu. Therefore, the SNFs depleted under high boron concentration conditions have a high fissile content. When the boron concentration decreases as actual reactor operation, the isotopes inventory of the SNFs is similar to the case where the average value of the boron concentration at the BOC and the EOC is kept constant.

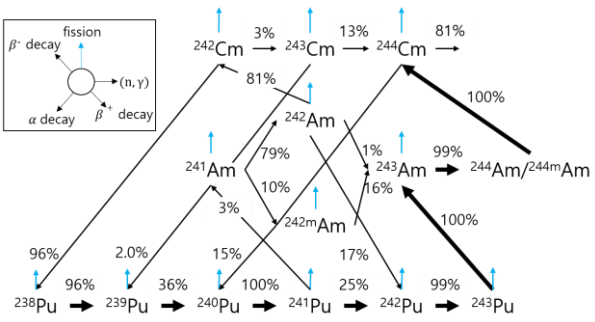

Fig. 4. Predominant path up to ^{244}Cm and transmutation ratio [5].

Fig. 5, 6, and 7 compare the major actinides' inventories of SNFs to be stored in the SFSR after 2-D depletion calculations for each of the eighteen axial nodes for the considered four different cases of the boron concentration. The nuclide inventories unit was normalized to the atom density when the boron concentration was 0 ppm for each nuclide.

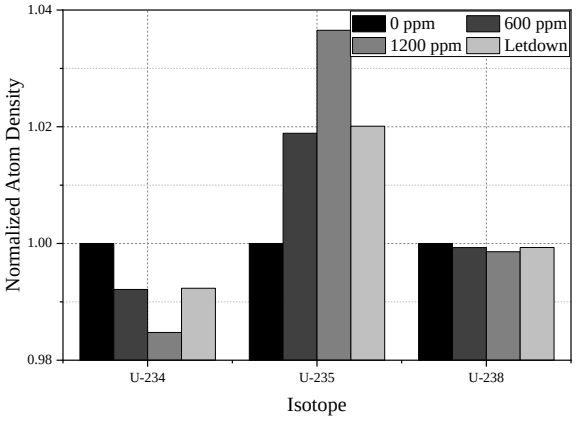

Fig. 5. Uranium nuclide inventories for different boron concentration conditions.

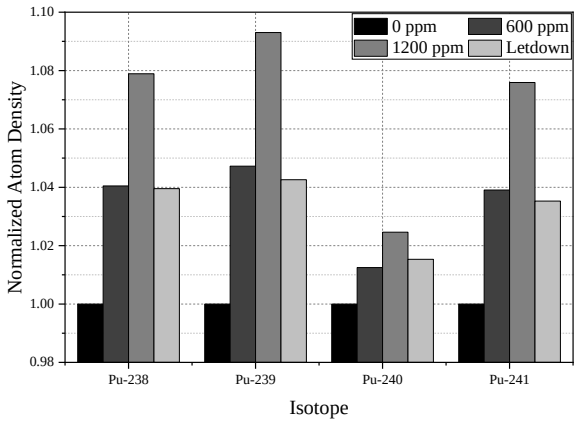

Fig. 6. Plutonium nuclide inventories for different boron concentration conditions.

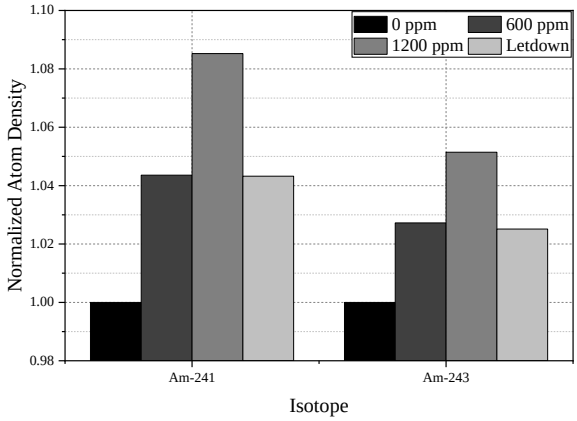

Fig. 7. Americium nuclide inventories for different boron concentration conditions.

Fig. 5 shows that a higher boron concentration leads to a higher ^{235}U inventory, while the boron concentration gives only a small effect on ^{238}U concentration. Fig.6 and 7 show the inventories of two fissile plutonium isotopes (i.e., $^{239, 241}$Pu), and two fissile americium isotopes (i.e., $^{241, 243}$Am) are higher for higher boron concentration than for the lower boron concentration.

2.3. Monte Carlo uncertainty sampling method

The evaluation of the propagation of the isotopic uncertainties to criticality calculation is usually conducted in two steps. In the first step, isotopic biases and bias uncertainties are determined by comparing the measured compositions of the radiochemical assay data and those calculated using the depletion calculation code. The ratios of the measured isotopic concentration to the calculated isotopic concentration for a sample j are X_n^j as Eq. (2).

$$X_n^j = M_n^j / C_n^j , \tag{2}$$

where M_n^j and C_n^j represent the measured and calculated concentrations, respectively, for a nuclide n of a sample j. Then, the sample mean of the ratios and standard deviation for the nuclide n are calculated using Eq. (3) and Eq. (4).

$$\overline{X_n} = \sum_{j=1}^{N_n} X_n^j / N_n , \tag{3}$$

$$S_n = \sqrt{\sum_{j=1}^{N_n} (X_n^j - \overline{X_n})^2 / (N_n - 1)} , \tag{4}$$

where N_n is the total number of samples for nuclide n. However, it should be noted that they are not true values of the mean and standard deviation because they are evaluated using only a limited number of experimental samples. Tolerance intervals have been introduced as a means to account for uncertainty due to sample size. A statistical tolerance interval defines the limits within which a stated proportion of a population is expected to lie, based on a sample measured from this population [6, 7]. Previous work [6] evaluated the isotopic biases and bias uncertainties using the SCALE 6.1/TRITON-2D code with the 238-group ENDF/B-VII.0 cross-section libraries for various PWR SNFs. In this work, the sample means and standard deviations evaluated by the previous work [6] were utilized to sample the isotopic concentrations. The isotopic bias is Eq. (3), and bias uncertainty (σ_n) is written by Eq. (5).

$$\sigma_n = S_n \times tf_2^{N_n} \tag{5}$$

where $tf_2^{N_n}$ represents the two-sided tolerance limit factor with N_n samples for the nuclide n. Fig. 8 presents isotopic bias and bias uncertainty values for PWR SNFs compositions.

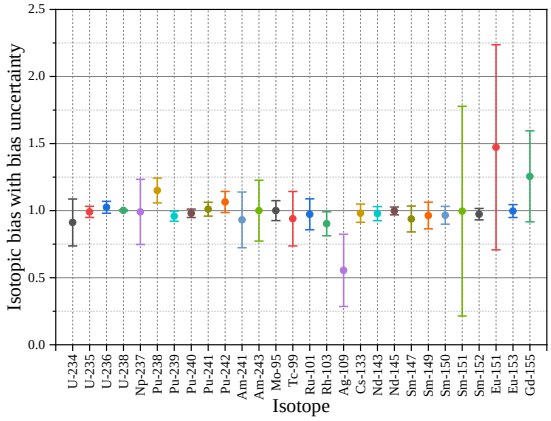

Fig. 8. Isotopic biases and bias uncertainties of major actinides and fission products.

In the second step, the isotopic compositions are randomly sampled with the probability density distribution for the measured to calculated isotopic ratios, and they are utilized for criticality calculations. Then, to evaluate the propagation of the isotopic uncertainties to k_{eff} for SFSR, a Gaussian distribution with zero mean and the unity standard deviation is used to sample the isotopic concentrations for nuclides with more than ten samples with the Eq. (6).

$$C_{n,b}^{k} = C_{n,b}^{ref} \times (\overline{X_n^b} + \sigma_n^b \times RN_n^k), \quad (6)$$

where $C_{n,b}^{k}$ represents the concentration of nuclide *n* in a fuel mixture of burnup *b* for criticality calculation *k* adjusted for isotopic bias and bias uncertainty. $C_{n,b}^{ref}$ represents the calculated concentration of nuclide *n* in a fuel mixture of burnup *b*. $RN_{n,b}^{k}$ represents the random number sampled with the standard normal distribution. The reference isotopic concentrations were used to calculate the reference k_{eff} value (k_{eff}^{ref}). It is noted that the reference k_{eff} value was calculated with no adjustment of the isotopic concentration. The reference k_{eff} value was used to calculate the depletion bias and bias uncertainty in k_{eff}. For each case of the criticality calculations, each isotopic concentration ($C_{n,b}^{k}$) was independently sampled using Eq. (6) with different random numbers from the other nuclides. As the number of criticality calculations increased, the estimated k_{eff} values approach a normal distribution with the mean ($\bar{k}_{eff}$) and standard deviation (σ_{keff}) given by Eq. (7) and Eq. (8), respectively.

$$\bar{k}_{eff} = \sum_{i=1}^{N_c} k_{eff}^{i} / N_c, \quad (7)$$

$$\sigma_{k_{eff}} = \sqrt{\sum_{i=1}^{N_c} (k_{eff}^{i} - \bar{k}_{eff})^2 / (N_c - 1)}, \quad (8)$$

where k_{eff}^{i} and N_c are the effective multiplication factor for criticality calculation *i* in the series of N_c criticality calculations and the total number of criticality calculations, respectively. The difference between the reference k_{eff} and the average k_{eff} value calculated by Eq. (7) represents the bias in k_{eff} as Eq. (8).

$$\beta_{depl} = \bar{k}_{eff} - k_{eff}^{ref}. \quad (9)$$

Bias uncertainty in k_{eff} at a 95% probability, 95% confidence level is calculated with Eq. (10).

$$\Delta k_{depl} = \sigma_{k_{eff}} \times tf_1^{N_c}, \quad (10)$$

where $\sigma_{k_{eff}}$ is determined with Eq. (8) and $tf_1^{N_c}$ is the one-sided tolerance-limit factor for the normal distribution corresponding to the number of calculated k_{eff} values (N_c), at a 95% probability and(?) 95% confidence level.

Finally, the total depletion uncertainty, $\beta_{depl} + \Delta k_{depl}$, in k_{eff} for the SFSR was evaluated by Eq. (11).

$$\beta_{depl} + \Delta k_{depl} = \begin{cases} (\bar{k}_{eff} - k_{eff}^{ref}) + \sigma_{k_{eff}} \times tf_1^{N_c}, & if\ \bar{k}_{eff} > k_{eff}^{ref}, \\ \sigma_{k_{eff}} \times tf_1^{N_c}, & if\ \bar{k}_{eff} \le k_{eff}^{ref} \end{cases} \quad (11)$$

As shown in Eq. (11), the positive bias was added, but the negative bias was set to zero for the conservative estimation of the total depletion uncertainty [6, 7].

2.4. Result of criticality calculation and uncertainties

In this study, a script code was made to generate random numbers and automatically prepare input files to execute SCALE 6.2/KENO-VI using Python3. Random numbers were generated differently for each of the eighteen axial nodes, the twenty-eight nuclides applied to the BUC, and each criticality calculation. And then, criticality calculations for SFSR were performed 300 times according to each boron concentration condition.

Criticality calculations were performed for SFSR using the Monte Carlo random sampling method for the concentration of nuclide in SNFs according to the boron concentration condition in depletion calculation. The reference k_{eff} value, k_{eff} values, average k_{eff} values, and standard deviations of k_{eff} values for four different cases of boron concentration conditions are shown in Fig. 9, 10, 11, and 12, respectively.

Under the high boron concentration condition in depletion calculation, the neutron spectrum is relatively hardened compared to the low boron concentration condition, and the neutron absorption reaction rate of ^{238}U increases. Consequently, much more of the inventories of fissile nuclides (e.g., ^{235}U, ^{239}Pu, ^{241}Pu, and ^{241}Am) remain in the SNFs as discussed in Sec. 2.2, which leads to the reactivity increase. For the reference criticality calculation without the Monte Carlo random sampling, we analyzed the fission-to-absorption ratio for the considered boron concentration conditions to quantitatively understand the effect of boron concentration on the reactivity. The fission-to-capture ratios for 0 ppm, 600 ppm, 1,200 ppm, and letdown conditions were estimated to be 0.83144, 0.83596, 0.84187, and 0.83589, respectively, which explains a higher boron concentration leads to a higher reactivity.

Table I summarizes the evaluations of the propagation of the isotopic uncertainties to criticality uncertainties for each boron concentration condition. The bias, bias uncertainty of k_{eff}, and total depletion uncertainty in k_{eff} were calculated in the given Eq. (9), (10), and (11), respectively. The reference k_{eff} values for the 0 ppm, 600

ppm, 1,200 ppm, and letdown conditions were estimated to be 0.8362, 0.84063, 0.84721, and 0.84054, respectively. Assuming that the change in reactivity according to boron concentration is linear, it can be calculated as 10 pcm per 1 ppm.

It is noted that the results for the letdown condition were similar to those of the 600 ppm condition because 600 ppm boron concentration is roughly the average concentration for the letdown condition case. However, it is very difficult to consider the true history of the boron concentration change in the propagation of depletion isotopic uncertainty to $k_{eff.}$ So it is necessary to assess the appropriate boron concentration such that the uncertainty does not lead to the violation of conservatism. As shown in Table I, the bias uncertainties of k_{eff} range from 0.0183 to 0.0196 Δk.

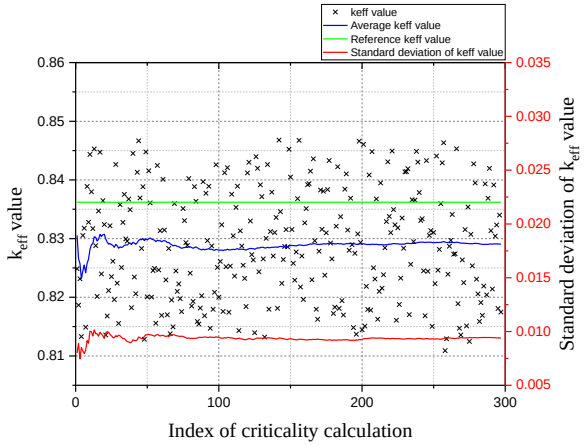

Fig. 9. Distribution of k_{eff} values, their averages, and standard deviations for SFSR during no boron reactor operating condition.

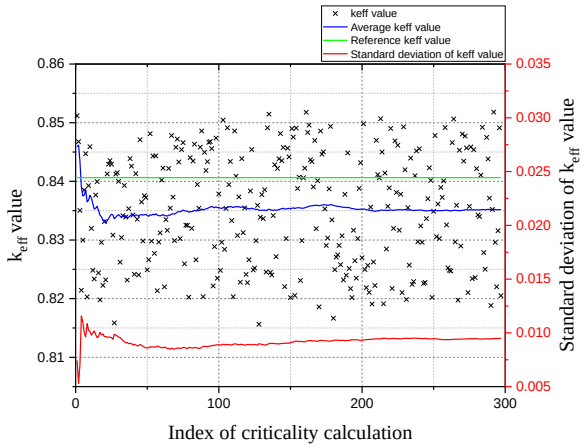

Fig. 10. Distribution of k_{eff} values, their averages, and standard deviations for SFSR during 600 ppm reactor operating condition.

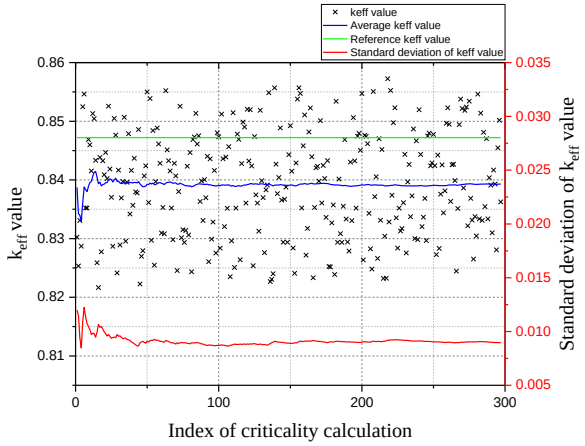

Fig. 11. Distribution of k_{eff} values, their averages, and standard deviations for SFSR during 1,200 ppm reactor operating condition.

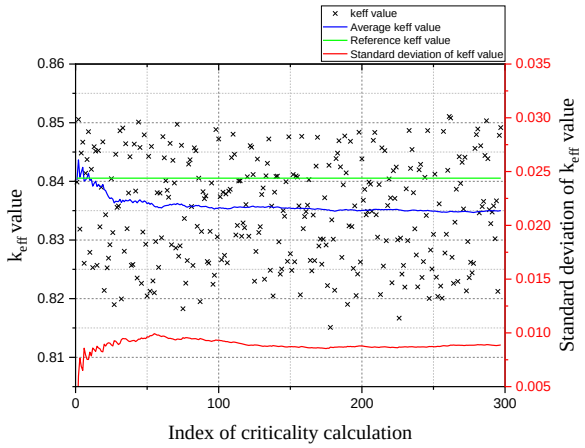

Fig. 12. Distribution of k_{eff} values, their averages, and standard deviations for during letdown reactor operating condition.

Table I. Comparison of the criticality uncertainties results for boron concentration condition.

	Reference k_{eff}	Average k_{eff}	Standard deviation of k_{eff}	Bias of k_{eff}	†Bias uncertainty of k_{eff}	††Total depletion uncertainty
0 ppm	0.83620	0.82905	0.00940	-0.00715	0.01941	0.01941
600 ppm	0.84063	0.83515	0.00951	-0.00548	0.01964	0.01964
1200 ppm	0.84721	0.83929	0.00896	-0.00792	0.01850	0.01850
Letdown	0.84054	0.83502	0.00888	-0.00552	0.01834	0.01834

† One-sided tolerance factor ($tf_1^{N_c}$) is 2.065 [8].

†† Negative bias was set to zero.

3. Summary and conclusion

This study analyzed the build-up of nuclides constituting PWR SNFs according to the history of boron concentration during depletion calculation. Moreover, we analyzed the depletion uncertainties in criticality analysis using the Monte Carlo random sampling method for the spent fuel storage pool. As a result, the build-up of the fissile isotopes in the SNF increases because the neutron spectrum was relatively hardened under the high boron concentration condition compared to the low boron concentration condition during depletion calculation. Due to this, the reactivity increases when it is stored in the SFSR. Therefore, it is necessary to consider the effect of boron concentration during depletion calculation for the conservative criticality safety analysis of the SFSR. Finally, the propagation of the isotopic uncertainty consideration of the effect of boron concentration during depletion calculation to the multiplication factor uncertainty was estimated from 0.0183 to 0.0196 Δk.

ACKNOWLEDGEMENTS

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