ISOTOPIC CONCENTRATION AND CRITICALITY ANALYSES OF BWR SPENT NUCLEAR FUEL USING CASMO

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ABSTRACT

The two-dimensional fuel assembly burnup code CASMO-4 validation was done for the application of burnup credit through the comparison with experimental values. The experimental data used for the validation were isotopic concentrations and criticality properties measured for the spent nuclear fuel (SNF) discharged from Fukushima-Daini Unit 2 the boiling-water reactor (BWR). The isotopic concentrations and the criticality properties were measured for several axial elevations by the destructive chemical assay and the exponential experiment, respectively.
The measured isotopic concentrations for actinides up to ^{241}Am were evaluated by CASMO-4 with the library based on ENDF/B-IV. The axial operating profiles used for CASMO-4 analyses such as assembly exposure and history-averaged void ratio were calculated by the core burnup code SIMULATE-3. The criticality properties were evaluated by CITATION fixed source calculation using the reactivity preserved macro cross-sections calculated by CASMO-4.
As a result, the relative errors (C/E) of isotopic concentrations were between 0.93 and 1.00 for the major actinides. The relative errors for the criticality properties at six detector heights were between 0.99 and 1.05. In both results, there was no obvious dependency on axial elevation of the fuel assembly, i.e., on void fraction and assembly exposure. From these analyses, it was concluded that the isotropic concentration and the criticality properties predictions of CASMO-4 code system have good accuracies for BWR SNF.

1.INTRODUCTION

Historically, the safety analyses of criticality control systems for spent nuclear fuel (SNF) are based on the assumption that SNF is fresh or in the highest critical state of the fuel life. This conservative assumption results in excessive and costly design requirements for neutron absorbers or spacing of SNF's in the storage casks and disposal facilities. The increasing number of SNF is one of the important issues for utilizing nuclear energy. So the application of burn-up credit, which takes into consideration of the reactivity reduction due to burnup, becomes beneficial by allowing storage casks and disposal facilities to optimize their capacity and improve its cost-effectiveness.
For the application of burn-up credit to criticality control systems, the validation of assembly burn-up codes for accurate prediction of the burn-up properties, such as the isotopic concentrations and the criticality, is important. The isotopic concentrations are utilized for establishing isotopic-inventory biases that have large influence on the

criticality safety analysis. In the same way, the criticality prediction using the predicted isotopic concentration affects directly the establishment of a criticality safety margin.

The BWR SNF experimental data for the validation of assembly burn-up codes are not widely available. Recently, to obtain the reference data for the burn-up credit analyses, the isotopic concentrations and the criticality properties of BWR SNF were examined by the destructive chemical assay and the exponential experiment, respectively. These experiments were done by Japan Atomic Energy Research Institute (JAERI), and they were summarized in the report [1,2].
The exponential experiment measures the neutron flux axial decay constants, which is utilized for the determination of the subcriticality. The exponential experiment has been applied for the subcriticality measurement of PWR SNF, and it is applied for that of BWR SNF in the present experiments. The pulsed neutron method, which measures the neutron flux time decay constants, is generally utilized for the measurement of subcriticality. However, the time decay is significantly affected by the reflector and its sensitivity to the subcriticality becomes less for deeper subcritical state [3]. The exponential experiment is superior to the pulsed neutron method for the deep subcritical measurements.
The analysis of the exponential experiment is essentially performed by the two-dimensional buckling search calculation. According to the JAERI report, however, the criticality properties were obtained under the flux affected by higher mode of the neutron source, and it is impossible to evaluate its effect by the two-dimensional buckling search calculation. To evaluate the actual measured criticality properties, the exponential experiment analysis has to be performed by three-dimensional fixed source calculation.

The measured isotopic concentration and criticality properties of BWR SNF are evaluated using CASMO-4 with the axial operating profiles of the assembly. In the case of BWR SNF analyses, the selection of the axial operating profiles for the validation is important because there exist significantly varying moderator density and non-uniform axial nuclear design of burnable poison and fuel. In this study, the isotopic concentration prediction is performed for the validation of CASMO-4 depletion calculation using the history-averaged axial operating profiles calculated by SIMULATE-3 core-following analyses. In addition, the comprehensive accuracy of the criticality property prediction, which includes the overall uncertainty induced by the axial operating profiles and the depletion calculation, are confirmed for the validation of the CASMO-4 code system. In this paper, the experiment and the analysis method are described, and the validity of the CASMO-4 code system is confirmed through the comparison with experimental results.

2. EXPERIMENTAL

The experiments were performed with the BWR SNF discharged from Fukushima-Daini Unit-2 Core. The SNF was the three-cycle burned 8x8 BWR fuel assembly (GE9) with 3.01wt% ^{235}U initial enrichment and an average burnup of 33.3GWd/t. The radial and axial configurations of ^{235}U and Gd initial enrichment in the assembly are shown in Figure 1. During the operation, there was no influence of the control rods or the periphery reflector region to this assembly. The cooling time between the removal from the reactor and the experiments is six years. In this section, the measurement method and the experimental results of the isotopic concentrations and the criticality properties are described.

2.1 MEASUREMENT OF ISOTOPIC CONCENTRATIONS

The isotopic concentrations were measured by destructive radiochemical assay for actinides up to ^{247}Cm and fission products up to ^{154}Sm [4]. The assays were performed on the round sliced samples taken from several axial levels of UO_2 pin (SF98) and Gd_2O_3-UO_2 pin (SF99) to evaluate the effects of the void ratio, exposure and burnable gadolinium poison. The fuel pin positions and the sampling positions in the axial direction are shown in Figure 1. After dissolving the samples in nitric acid solution, the isotopic compositions were measured by a combination of isotopic dilution mass spectrometry, alpha-ray spectrometry and γ-ray spectrometry. The uncertainties of measured data, which depends on the isotopes and its measurement method, are summarized in

Table I. All the concentrations were normalized to the date of the discharge from the reactor except samarium isotopes. The results of ^{239}Pu include the amounts of ^{239}Np (2.4d half-life).

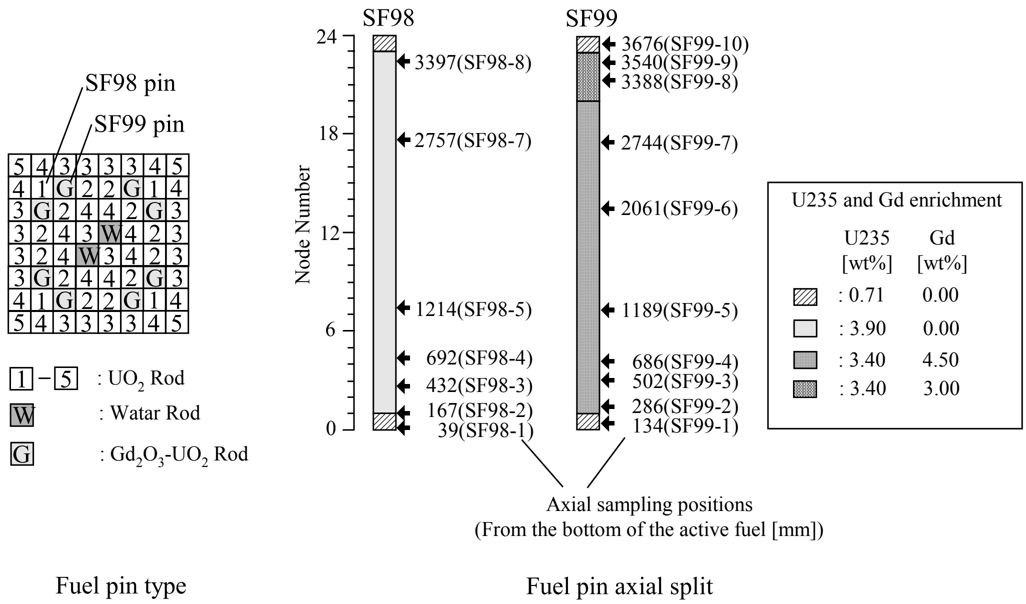

Figure 1 Configurations of spent nuclear fuel

Table I. Measurement uncertainties of isotopic concentrations

Isotopes	Uncertainties [%]
^{235}U	0.1
^{236}U	2.0
^{238}U	0.1
^{237}Np	10.0
^{238}Pu	0.5
^{239}Pu	0.3
^{240}Pu	0.3
^{241}Pu	0.3
^{242}Pu	0.3
^{241}Am	2.0

2.2 EXPONENTIAL EXPERIMENT

The criticality properties were measured by the source-moving exponential experiment [1,2]. The SNF without the channel box was set in the fuel receiving pool with the ^{252}Cf-neutron source on one side and the ^{235}U neutron detector on the opposite side of the assembly as illustrated in Figure 2. Three fuel rods were already taken out for the isotopic concentration measurement.

Because of its deep subcritical state, the neutron flux in the experimental system multiplies but rapidly decays away. At certain point where the source is sufficiently apart, higher modes of neutron flux decay away. In other words, the axial flux distribution has asymptotic exponential shape which is dominated by the fundamental mode. Under the fundamental mode, the decay constant, i.e., the slope of the axial flux distribution, represents the degree of the subcriticality. Therefore the decay constant is regarded as an index of the eigenvalue.

In the experiment, the criticality properties of decay constant called γ-value was measured by source-moving exponential experiment in the axial direction as a criticality property. The variation of neutron count rate at a fixed detector elevation of SNF was measured by moving the neutron source in the axial direction about the detector. After subtracting the background neutron count rate, the γ-value at the elevation was obtained by fitting the measured neutron count rate variations with the following single exponential function.

$$\varphi(t) = \alpha \exp(-\gamma t) \tag{1}$$

where α, γ and t are constant, the decay constant (γ-value), and the neutron source level from the bottom of the active fuel.
The measurements of the γ-values were done at six axial detector positions to evaluate the effects of void ratio and exposure. Two γ-values at each detector positions were obtained by fitting both the descent and ascent slope of the neutron count rate variations. The measured γ-values are summarized in Table IV. The statistical errors of the fitting were reported to be smaller than 3%.

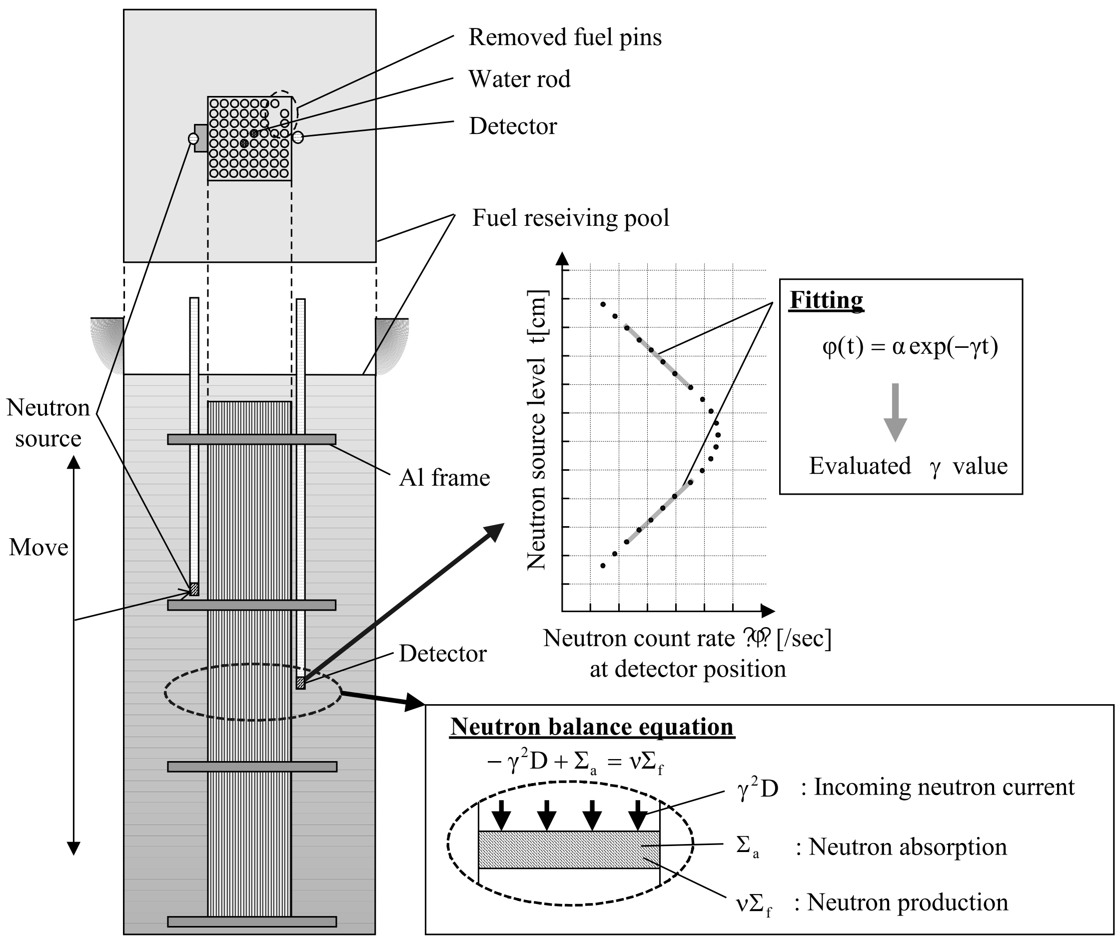

Figure 2 Exponential experiment and γ-value

In the case which the fitting is done under the fundamental mode, the γ-value is expressed by the negative material buckling at the detector elevation as in Equation (2) [3]. In addition, the following two important relationships, Equation (3) and (4), are derived from Equation (2). The Equation (3) is the neutron balance equation at the detector elevation, and the $-\gamma^2 D$ term is the incoming neutron current as illustrated in Figure 2. The Equation (4) is an expression of the two-dimensional multiplication factor with the γ-value which shows the γ-value as the index of the eigenvalue.

$$\gamma^2 = -B_m^{\ 2} \equiv -\frac{\overline{\nu\Sigma_f} - \overline{\Sigma_a}}{\overline{D}} \tag{2}$$

$$-\gamma^2 \overline{D} + \overline{\Sigma_a} = \overline{\nu\Sigma_f} \tag{3}$$

$$k_{\infty(2D)} = \overline{\nu\Sigma_f} \big/ \overline{\Sigma_a} = 1 - \gamma^2 \overline{D} \big/ \overline{\Sigma_a} \tag{4}$$

$B_m^{\ 2}$, D, Σ_a, $\upsilon\Sigma_f$ and $k_{\infty(2D)}$ are material buckling, diffusion coefficient, macroscopic cross sections for absorption, production and two-dimensional multiplication factor, respectively.
According to the Equation (2), the γ-values measured in the fundamental mode can be calculated by direct two-dimensional axial buckling search calculation. However, the γ-values were in fact measured under the flux affected by the higher mode of the neutron source because of its deep subcriticality. Therefore, the flux variation must be calculated first by the three-dimensional fixed source calculation. Then the γ-value can be evaluated by fitting the flux variation. High order mode could have been avoided if the distance between the neutron source and the detector is large enough, but then it is difficult to keep the neutron count rate high without the radiation-shielding problem.

3 ANALYSIS

3.1 ANALYSIS OF ISOTOPIC CONCENTRATIONS

The isotopic concentrations of SNF were evaluated by CASMO two-dimensional fuel assembly burnup calculation with the library based on ENDF/B-IV. The assembly burnup calculations were performed for each axial zone with single-assembly infinite lattice assumption using the axial-burnup profiles, i.e., the assembly exposure and the history-averaged void ratio, calculated by SIMLATE-3 core burnup code. The twenty-four axial zone-averaged profiles were obtained from the core-following calculation by SIMULATE-3. The assembly geometry was exactly modeled in the CASMO-4 calculation except for the spacer grid. Note that this evaluation method might not be assured when the heterogeneity of axial composition or the local distortion of the burnup profile in the axial zone is significant. From these calculations, the isotopic concentrations are evaluated for every pin cells.

Table II and III shows C/E's for the isotopic concentrations of UO_2 pin and UO_2-Gd_2O_3 about ten actinides up to ^{241}Am by CASMO-4. These isotopes are identified to have the most significant effect on the multiplication factor of SNF. There is no obvious trend of C/E's on axial elevation, i.e., on void fraction and pin exposure (excluding the results of SF98-1, SF98-2, SF99-1, SF99-2, SF99-9 and SF99-10 which is located near the edge of the fuel pins). In the edge region corresponding to the natural enrichment uranium zones and its adjacent zones, some results showed large discrepancies. It may be due to the fact that the effect of axial heterogeneity to the burnup spectrum is not considered in the CASMO-4. The pin averaged C/E's were between 0.93 and 1.00 for the major actinide except ones in the edge region. The effect of the burnable poison on the accuracy of the isotopic prediction was not observed.

Table II. Results of isotopic concentrations analyses (UO_2 pin)

Isotopes	SF98-1	SF98-2	SF98-3	SF98-4	SF98-5	SF98-7	SF98-8	Average
^{235}U	0.96	0.82	0.97	0.93	0.88	0.94	0.99	0.94
^{236}U	1.02	1.16	1.01	1.01	1.01	1.00	0.99	1.00
^{238}U	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
^{237}Np	0.91	1.55	1.10	1.05	1.33	1.14	1.13	1.15
^{238}Pu	0.73	1.67	0.96	0.98	1.06	1.01	0.92	0.99
^{239}Pu	0.92	1.06	0.96	0.95	0.93	0.91	0.94	0.94
^{240}Pu	0.97	1.30	0.96	0.93	0.93	0.91	0.93	0.93
^{241}Pu	0.88	1.38	0.94	0.94	0.92	0.92	0.93	0.93
^{242}Pu	0.88	1.83	0.92	0.93	0.97	0.93	0.91	0.93
^{241}Am	0.49	0.78	0.91	1.03	1.11	1.14	0.82	1.00

Table III. Results of isotopic concentrations analyses (UO_2-Gd_2O_3 pin)

Isotopes	SF99-1	SF99-2	SF99-3	SF99-4	SF99-5	SF99-6
^{235}U	0.90	1.01	0.96	1.01	0.91	0.98
^{236}U	1.02	0.99	1.01	1.00	1.01	0.99
^{238}U	1.00	1.00	1.00	1.00	1.00	1.00
^{237}Np	0.81	0.93	0.91	1.03	0.94	1.01
^{238}Pu	0.99	1.05	1.03	1.10	1.31	1.11
^{239}Pu	0.88	0.98	0.96	1.00	0.93	0.95
^{240}Pu	1.00	0.99	0.98	0.98	0.95	0.95
^{241}Pu	0.83	0.93	0.95	0.97	0.92	0.95
^{242}Pu	0.91	0.90	0.96	0.93	0.97	0.97
^{241}Am	0.88	0.91	0.82	1.06	0.98	1.07

Isotopes	SF99-7	SF99-8	SF99-9	SF99-10		Average
^{235}U	0.97	1.01	1.01	1.02		0.97
^{236}U	1.00	0.98	1.00	0.98		1.00
^{238}U	1.00	1.00	1.00	1.00		1.00
^{237}Np	0.97	0.97	0.97	1.09		0.97
^{238}Pu	1.17	1.03	1.07	1.38		1.12
^{239}Pu	0.94	0.98	1.05	1.09		0.96
^{240}Pu	0.95	0.97	1.00	0.97		0.96
^{241}Pu	0.94	0.97	1.07	1.04		0.95
^{242}Pu	0.97	0.95	1.02	0.95		0.96
^{241}Am	1.05	0.71	0.52	1.81		0.95

3.2 EXPONENTIAL EXPERIMENT ANALYSIS

As described previously, the three-dimensional fixed source calculation was necessary to evaluate the present experimental γ-value which was affected by the higher mode of the neutron source.
The three-dimensional fixed source calculations were performed by CITATION three-dimensional diffusion code to evaluate the flux variations at the detector positions. The statistically high accurate evaluation of the flux variation by Monte-Carlo code is considered to be impractical since the effective multiplication factor of the system is too small to obtain sufficient neutron count rate for the fitting region.
In the analyses, the combination of heterogeneous single-assembly calculation by CASMO-4 and pin and water cell homogeneous calculation by CITATION was utilized as illustrated in Figure 3. From the CASMO-4 heterogeneous unit assembly burnup calculations, the cell homogeneous macro cross-sections were calculated for every pin cell and water reflector. Using these macro cross-sections, cell homogeneous calculations were performed by CITATION. The thermal neutron flux variations at detector positions were calculated by moving the neutron source in the axial direction about the fixed detector level. The γ-value was then obtained by fitting with the single exponential function in the same way as the experiment.

In the present analyses, the neutron current leakage to the surrounding reflector for BWR SNF is significantly larger than PWR SNF due to its small assembly size and consequently results in large transport effect. Under this condition, the multiplication factor calculated at each axial level by CASMO-4 two-dimensional transport calculation was not preserved by CITATION pin cell homogeneous diffusion calculation. This problem dose not arise when the radial geometry is homogeneously treated. However, the radial heterogeneities are indispensable to evaluate the effect of higher mode to the radial flux distribution properly. This transport effect influences axial leakage directly and results in an inaccurate γ-value evaluation. To remedy this effect, the nine-group macro cross-sections (absorption and ν-fission) for CITATION were corrected to preserve the CASMO-4 reactivity for each cell. With these corrected macro cross-sections, the γ-values were evaluated excluding the transport effect.

To determine the calculational parameters such as energy group structure, mesh size, and neutron spectrum utilized in the CITATION calculation, three kinds of sensitivity studies were performed. First, the energy group structure of the macro cross-sections calculated by CASMO-4 was varied from two to nine, and the effect on γ-value was evaluated. As shown in Figure 4, the γ-values have converged at about the seven to nine energy groups. Second, the axial and radial mesh divisions of the cell in CITATION was varied as illustrated in Figure 4. The γ-values have converged at about 2cm for axial mesh and about 0.5cm for radial mesh. Third, the effects of fission and neutron-source spectrum on γ-value were evaluated using the ^{235}U fission spectrum and the weighted average fission spectrum of ^{235}U and ^{239}Pu based on the SNF composition calculated by CASMO-4. The γ-values were found to have same value, and it is confirmed that the γ-value is not sensitive to the fission and neutron-source spectrum. Therefore, from the sensitivity studies, the following parameters are used for the CITATION calculation: the nine-group macro cross-sections; about 2cm and 0.5cm for axial and radial mesh size, respectively; and the weighted average fission spectrum.

Figure 5 shows C/E's for the γ-values by CITATION with the macro cross-section calculated by CASMO-4. The analyses were done at six detector heights of eleven γ-values. The C/E's were between 0.99 and 1.05 and average C/E were 1.01 except for the result of 369cm and 304cm detector position. The γ-value prediction show slightly large discrepancy in the natural enriched uranium zone (369cm position) and the spacer grid position (304cm position) because the accuracy of isotopic concentration prediction is low in these positions and the macro cross-sections are not generated in the spectrum reflecting such axial heterogeneity. The dependency of C/E's on axial elevation, i.e., on void fraction and assembly exposure was not observed.

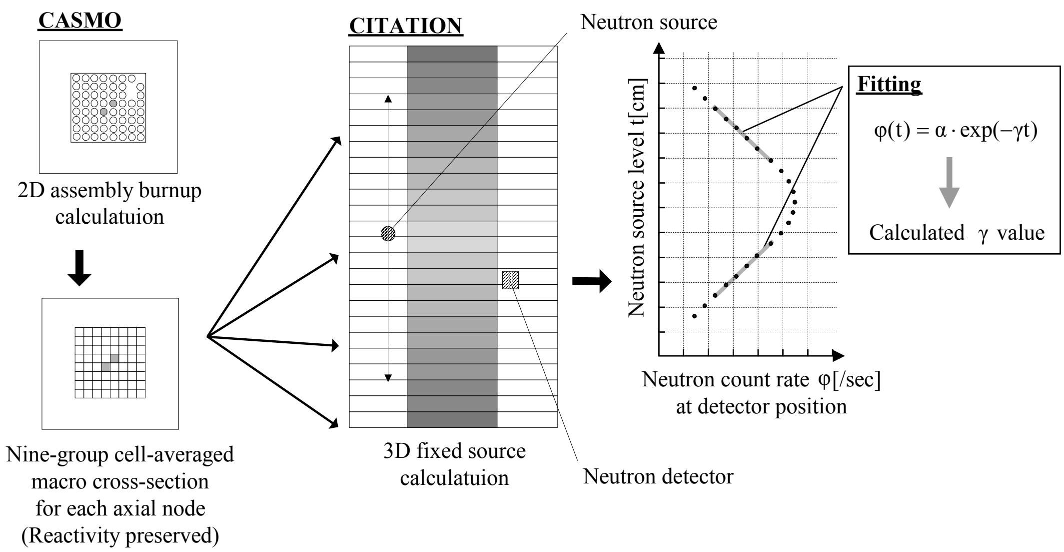

Figure 3 Analysis of γ-value

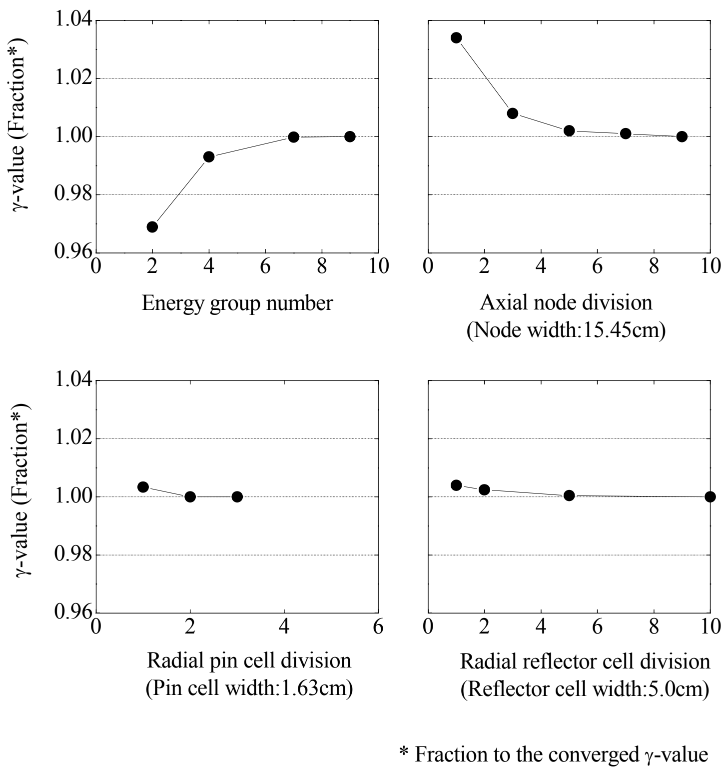

Figure 4 Results of sensitivity studies

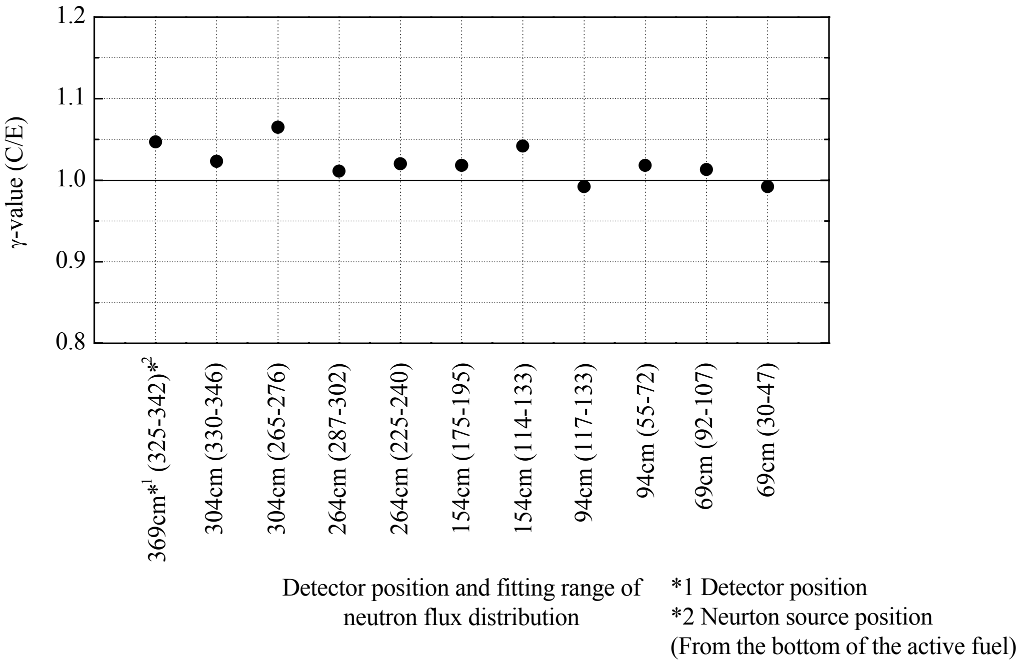

Figure 5 Results of γ-value analyses

Table IV. Results of γ-value analyses

Detector Position*[1] [cm]	Fitting Range*[1] [cm]	Measured γ [/cm]	Calculated γ [/cm]	C/E
369	325 – 342	0.133 ± 0.001	0.139	(1.047)*[2]
304	330 – 346	0.135 ± 0.002	0.138	(1.023)*[2]
	265 – 276	0.135 ± 0.003	0.144	(1.065)*[2]
264	287 – 302	0.139 ± 0.002	0.140	1.011
	225 – 240	0.140 ± 0.001	0.143	1.020
154	175 – 195	0.140 ± 0.003	0.142	1.018
	114 – 133	0.139 ± 0.002	0.145	1.042
94	117 – 133	0.146 ± 0.004	0.145	0.992
	55 – 72	0.143 ± 0.002	0.146	1.018
69	92 – 107	0.144 ± 0.004	0.146	1.013
	30 - 47	0.140 ± 0.002	0.139	0.992

*1 From bottom of active fuel (full active fuel length: 370.8cm)
*2 Excluding from the average C/E calculation

4 CONCLUSIONS

The measured isotopic concentration and the criticality properties (γ-value) of BWR SNF were benchmarked for the validation of CASMO-4 code. The CASMO-4 burnup calculations were performed with the library based on ENDF/B-IV using the axial operating profiles calculated by SIMULATE-3. The γ-values were evaluated by CITATION three-dimensional fixed-source calculation using the reactivity preserved macro cross-sections calculated by CASMO-4 to consider the effect of the higher mode of the neutron source.
As a result, the pin averaged C/E's for the isotopic concentrations were between 0.93 and 1.00 for the major actinides except ones in the fuel edge region. The C/E's for the criticality properties at six detector heights were between 0.99 and 1.05, and the average C/E was 1.01. Therefore, the depletion calculation by CASMO-4 using the axial operating profiles calculated by SIMULATE-3 was proven to be capable of accurately predicting the isotropic concentrations and criticality properties of BWR SNF. This result implies the alternative usage of history-averaged axial operating profiles provided by core-following calculations instead of actual fuel burnup history data.

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