Study of Burnup Reactivity and Isotopic Inventories in REBUS Program

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Abstract

The REBUS program provides the measurement data of the critical water levels and water level reactivity coefficients of critical cores which contain the fresh and irradiated fuel bundles of PWR UO2 and BWR MOX fuel. The average burnups of irradiated fuels are 54.5GWd/t for PWR UO2 and 60.6GWd/t for BWR MOX. Using these data, the burnup reactivity was determined to be -2.40%dk/kk' for the PWR UO2 fuel bundle and -2.48%dk/kk' for the BWR MOX fuel bundle. Theoretical analysis was performed to reproduce the burnup reactivity, which comprises burnup calculation to obtain the isotopic inventories of the irradiated fuel and core calculation to obtain neutron multiplication factors. The deviations (biases) of the calculated burnup reactivity from the measurement are -0.12 for the PWR UO2 fuel bundle and +0.04 %dk/kk' for the BWR MOX fuel bundle. Study for splitting the calculation bias of the burnup reactivity into those of inventory and reactivity calculations was also performed using measured isotopic inventories of the irradiated fuel. The study shows that the biases for the PWR UO2 fuel bundle are +0.13 %dk/kk' for the inventory calculation and -0.25 %dk/kk' for the reactivity calculation; those for the BWR MOX fuel bundle are +0.17%dk/kk' for the inventory calculation and -0.13 %dk/kk' for the reactivity calculation, which indicates that the biases of inventory calculation and reactivity calculation partly compensate each other to make the total biases smaller in the adopted analysis tools and models in this study.

1. Introduction

For precise estimation of the burnup reactivity of nuclear systems including irradiated fuel, analysis tools are required to accurately calculate the number densities and effective microscopic cross sections of the nuclides in the irradiated fuel. In order to validate and improve analysis tools, it is effective to compare the analysis results with the measurement of burnup reactivity. The REBUS program has a unique feature, that is, direct determination of the burnup reactivity of UO2 and MOX fuel bundles, which were fabricated from irradiated fuel assemblies discharged from commercial LWRs.[1-11)] The program includes core physics experiment in the LWR critical test facility VENUS, nondestructive measurement of the burnups of irradiated fuel rods by gamma-ray spectroscopy, and radiochemical isotopic analysis of pellet samples. In the critical experiment, a core tank was loaded with a 27x27 square lattice core that consists of a 7x7 cell test bundle in the core center, and 3.3 and 4.0 wt% enriched UO2 fuel rods surrounding the test bundle as a driver region. Five types of bundles were tested in the experiment: (1) fresh BR3 (PWR) MOX fuel, (2) irradiated BR3 MOX fuel, (3) irradiated BWR MOX fuel, (4) fresh PWR UO2 fuel, and (5) irradiated PWR UO2 fuel. **Figure 1** and **2** show the core radial configurations for the PWR UO2[3)] and BWR MOX fuel test bundles.[10)] The average burnups of the fuel bundles of the PWR UO2 and BWR MOX are 54.5 and 60.6 GWd/t, respectively. The analysis of the experimental data have been performed by the participants of the program.[1-11)] One of findings in the analysis is that, while measured burnup reactivity values were well reproduced by core calculations, marked discrepancies between the calculated and measured inventories were observed for both irradiated UO2 and MOX fuel. The present authors have indicated that the biases in inventory and reactivity calculations compensate each other, which makes the total biases of the burnup reactivity small for the BR3 MOX fuel bundle.[11)]

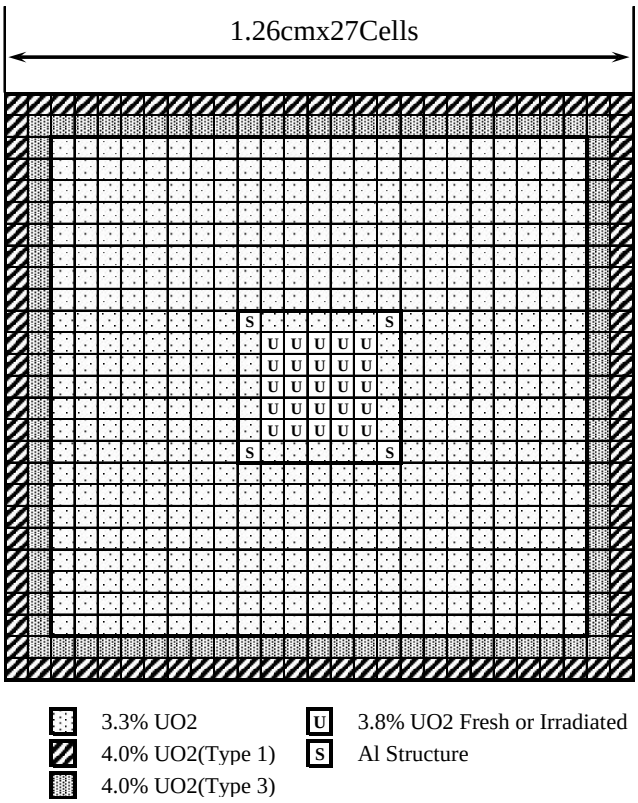


Fig. 1 Core radial configuration with fresh or irradiated PWR UO2 fuel test bundles[3)]

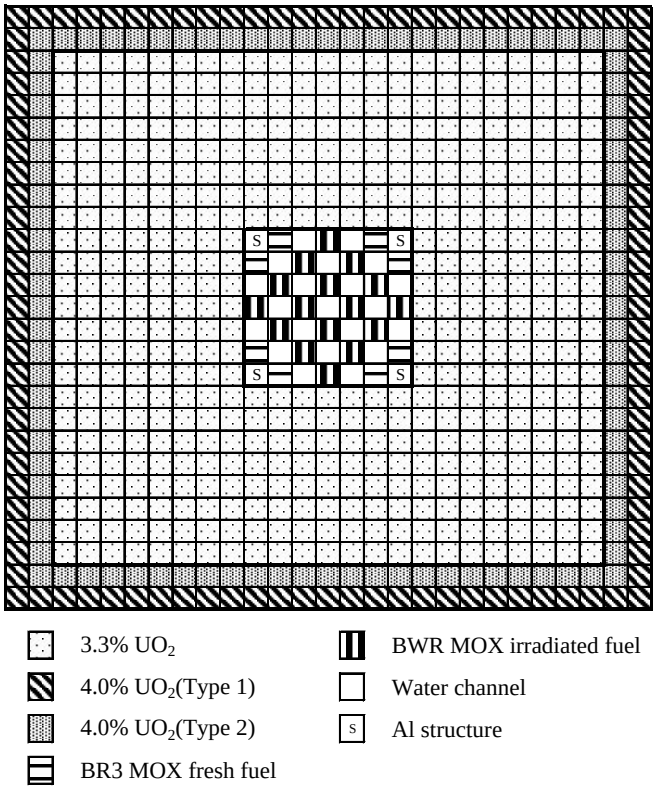


Fig. 2 Core radial configuration with irradiated BWR MOX fuel test bundle[10)]

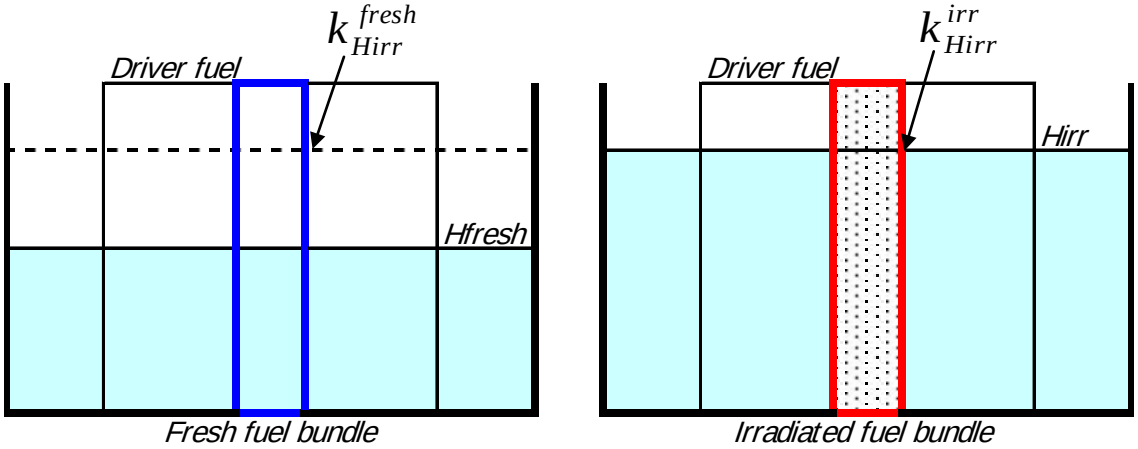
In the present study, the ratios of calculated to measured inventories (C/Es) were utilized to estimate probable inventories for the PWR UO2 and BWR MOX fuel bundles, and burnup reactivity was re-analyzed with the probable inventories. Based on the results, the biases in inventory and reactivity calculations for burnup reactivity were discussed.

2. Burnup Reactivity

A principle of measurement of burnup reactivity in the REBUS program is schematically shown in **Fig. 3**. Burnup reactivity is expressed by

$$\rho = \{1-(1/k_{Hirr}^{irr})\}-\{1-(1/k_{Hirr}^{fresh})\}, \quad (1)$$

where k_{Hirr}^{irr} is k_{eff} with the critical water level *Hirr* of a core loaded with a irradiated fuel bundle, and k_{Hirr}^{fresh} is that with the water level *Hirr* of a core loaded with a fresh fuel bundle.



Hirr: the critical water level of a core loaded with an irradiated fuel bundle

Hfresh: the critical water level of a core loaded with a fresh fuel bundle

Fig. 3 Principle of measurement of burnup reactivity in the REBUS program

2.1 Determination of Burnup Reactivity with Measurement Data

In the critical measurement, k_{Hirr}^{irr} = 1.0, and therefore

$$\rho = -\{1-(1/k_{Hirr}^{fresh})\}. \quad (2)$$

Applying measured water level reactivity coefficients $(\partial\rho/\partial h)$,

$$\rho = -\int_{Hfresh}^{Hirr}(\partial\rho/\partial h)dh, \quad (3)$$

where *Hfresh* is the critical water level of a core loaded with a fresh bundle. One group diffusions theory gives,

$$\rho = 1-\{1+M^2B_{rad}^2+M^2\pi^2/(h+\delta_z)^2\}/k_\infty,$$

and $(\partial\rho/\partial h)$ is expressed as a function of a water level *h* by

$$(\partial\rho/\partial h) = 2\pi^2M^2/k_\infty(h+\delta_z)^3 = \alpha/(h+\delta_z)^3, \quad (4)$$

where M^2 is a migration area, B_{rad}^2 a radial buckling, δ_z a reflector saving, k_∞ an infinite multiplication factor and α a constant. Typical experimental errors in one σ are 0.05% for *h*, 2.5% for δ_z, 1.3% for $(\partial\rho/\partial h)$ in the fresh fuel bundle core, and 0.05% for *h*, 1.6% for δ_z, 0.7% for $(\partial\rho/\partial h)$ in the irradiated fuel bundle core. Two values of α were obtained from the measured water level reactivity coefficients of the fresh and irradiated fuel bundle cores. Using these values, two burnup reactivity values were derived, as shown in **Table 1**. Since the experiment was not performed on fresh fuel bundle for the BWR MOX fuel bundle core, the fresh BR3 MOX fuel bundle core was used as a substitute. The reactivity difference between the fresh BWR MOX and BR3 MOX fuel bundle cores with *Hfresh* of the fresh BR3 MOX core was calculated with a continuous energy Monte Carlo code MVP.[12] It showed that the

reactivity with the fresh BWR MOX fuel bundle is larger than that with the BR3 MOX fuel bundle by 0.13%dk/kk'. This result was applied to the measurement reactivity of the BWR MOX fuel bundle. The corrected value was shown in Table1, which is to be compared with the calculated value. The obtained burnup reactivity values are systematically larger for those with α values of the irradiated fuel bundle cores than those with α values of the fresh fuel bundle cores. This is probably due to a simple model shown by Eq. (4). Calculated average values and standard deviations of the two values were regarded as measurement values and their errors, respectively.

Table 1 Determination of burnup reactivity with measurement data

Fuel bundle	PWR UO2		BWR MOX	
	Fresh	Irradiated	Fresh	Irradiated
Critical water level (cm)	59.05	81.38	61.10	84.84
Water level reactivity coefficient ($/cm)	0.186	0.0924	0.191	0.0887
Reflector saving (cm)	12.60	13.23	12.81	14.91
Averaged reflector saving (cm)	12.92		13.86	
α (pcm cm^2)	5.65x10^7	6.26x10^7	6.08x10^7	6.42x10^7
Burnup reactivity (%dk/kk')	-2.28	-2.52	-2.29	-2.42
Averaged burnup reactivity and error (%dk/kk') (relative error)	-2.40 ± 0.17 (7%)		-2.36 ± 0.09 (4%) -2.48 ± 0.10 (4%)*	

* Corrected value (see text)

2.2 Calculation of Burnup Reactivity

Burnup reactivity values were obtained by Eq. (1). Effective multiplication factors k_{Hirr}^{irr} and k_{Hirr}^{fresh} were analyzed by transport core calculations using a lattice calculation code SRAC[13] with the nuclear data library JENDL-3.3[14] and a transport calculation code THREEDANT.[15]

2.2.1 Isotopic Inventory Calculations

Axial burnup distributions in a 1-cm pitch were determined by gamma-ray spectroscopy of Cs-137 for the test fuel rods, which compose the irradiated fuel test bundles. Depending on the axial variation of burnup, three and six axial divisions were adopted for the irradiated PWR UO2 and BWR MOX fuel rods, respectively. For each axial region, isotopic inventories were obtained by burnup calculations in infinite assembly models with the SRAC code coupled with the nuclear data library JENDL-3.2.[16] The burnup calculations took into account the irradiation history of the fuel assemblies from which the fuel rod were selected. The detail descriptions are given in Refs. 3 and 10. The obtained isotopic inventories for the irradiated fuel were used to obtain the values of k_{Hirr}^{irr} and k_{Hirr}^{fresh} in the core calculation.

2.2.2 Calculated Burnup Reactivity

The calculated values of burnup reactivity are shown in **Table 2**. The biases of burnup reactivity, which are defined as the calculated reactivity minus the measured, are from -0.12 to 0.04%dk/kk' and are comparable to measurement errors.

3. Biases due to Inventory and Reactivity Calculations

3.1 Comparison between Calculated and Measured Inventories

One fuel pellet sample was taken for each of the PWR UO2 fuel and BWR MOX fuel, and the inventories of major Actinide and FP nuclides were measured by radiochemical analysis. The burnup value measured by gamma-ray spectroscopy was also given for the samples. Burnup calculations were performed for the fuel pellet samples in the same way mentioned in **Section 2.2.1** with the target burnups obtained by gamma-ray spectroscopy. **Figures 4** and **5** show the ratios of the calculated to measured inventory (C/Es) for actinide and FP nuclides. They also show uncertainties ($2\ \sigma$) of C/Es caused by the measurement errors. While the calculations generally well reproduce the measurements for the PWR UO2 fuel, marked overestimation is observed for Np-237, Am-241, Am-243 and metallic FP nuclides (Ru-101, Rh-103, Pd-105, -108 and Ag-109) and slight overestimation is observed for Pu-239 and 241. For the BWR MOX fuel, marked overestimation appears in Np-237, Pu-239, -241, Am-241, -242m, -243, Cm-245 and metallic FP nuclides.

Table 2 Calculated burnup reactivity

Fuel bundle	PWR UO2		BWR MOX	
k-effective	k_{Hirr}^{irr}	k_{Hirr}^{fresh}	k_{Hirr}^{irr}	k_{Hirr}^{fresh}
	0.99445	1.02004	0.99721	1.02205
Burnup reactivity (%dk/kk')	-2.522		-2.437	
Bias of burnup reactivity (Calculated - measured) (%dk/kk')	-0.1227		+0.0438	
C/E	1.051		0.982	

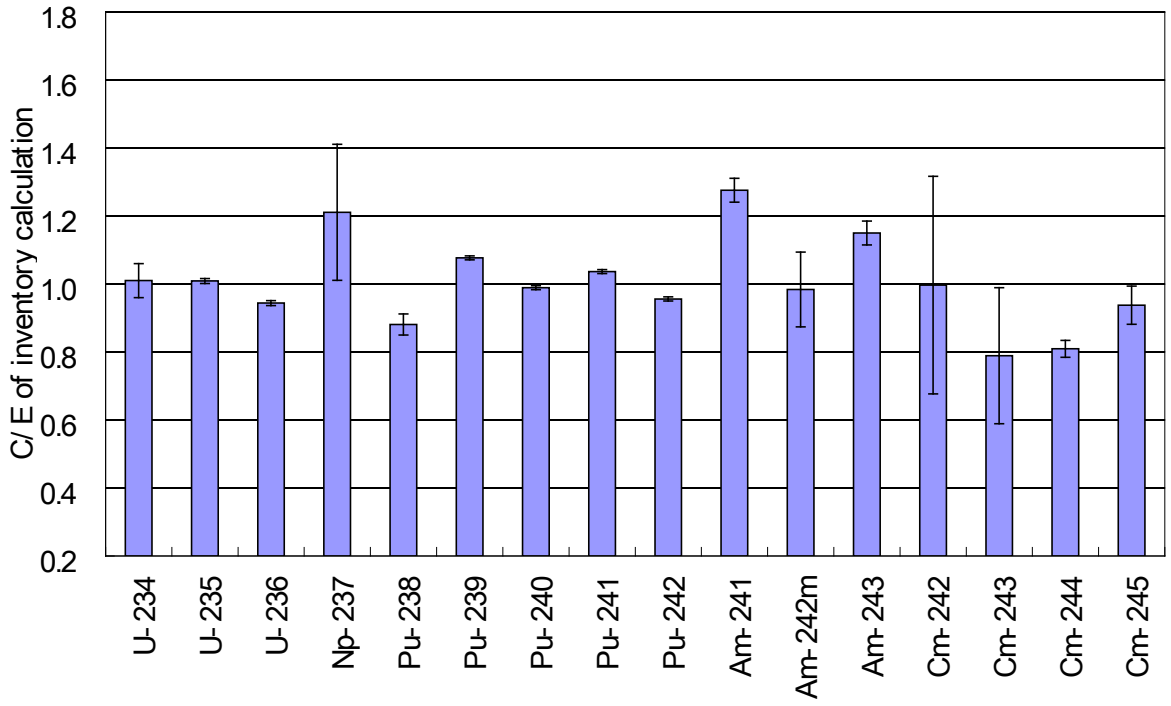
3.2 Correction of Calculated Isotopic Inventories

Probable isotopic inventories were estimated by using above-mentioned C/Es for the fuel pellet samples. It was assumed that (1) the C/Es in the comparison can be applied to all fuel rods of the test bundle, and (2) C/Es are unity at the burnup = 0 and proportional to the burnup. With the assumption (2), corrected isotopic inventory for a nuclide *i* in a segment *j* in the fuel bundle is given by

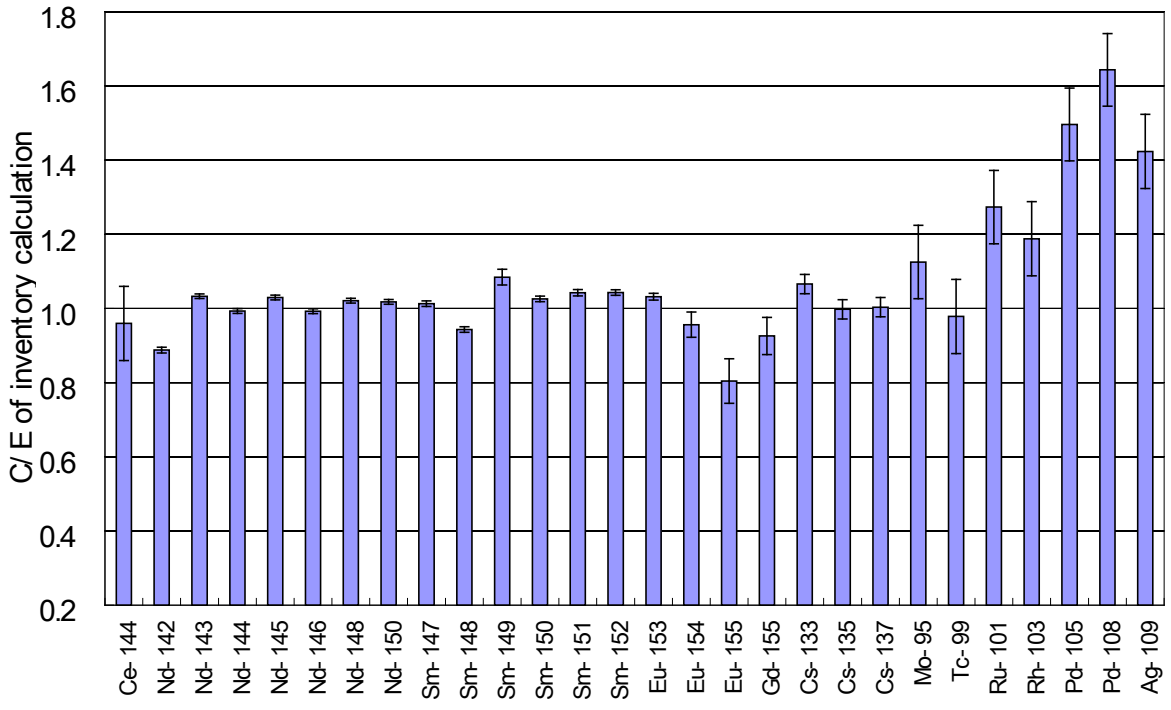
$$E_i^j = C_i^j \left[\{(E_i/C_i) - 1\} \left(B^j/B\right) + 1 \right], \quad (1)$$

where E_i^j is a corrected inventory, C_i^j is a calculated inventory, E_i/C_i is the inverse of the C/E for the nuclide *i*, B^j is the burnup of the segment *j*, *B* is the burnup value measured by gamma-ray spectroscopy for the pellet sample of the radiochemical analysis.

In order to study which nuclides cause the dominant effect of the inventory correction on the reactivity of the irradiated fuel bundle cores, the sensitivity of inventory change to reactivity of a typical infinite fuel cell in the irradiated fuel bundle in the experimental core was also calculated, and the reactivity change of the infinite cell caused by the inventory correction was estimated. **Figures 6** and **7** show the results. Prominent nuclides in the reactivity change are Np-237, Pu-239, -241, Am-241, metallic FP nuclides for the PWR UO2 fuel, and Pu-239, -241, Am-241 and metallic FP nuclides for the BWR MOX fuel. The total reactivity changes of the infinite fuel cells are -0.70 and -1.49%dk for the PWR UO2 and BWR MOX fuels, respectively.

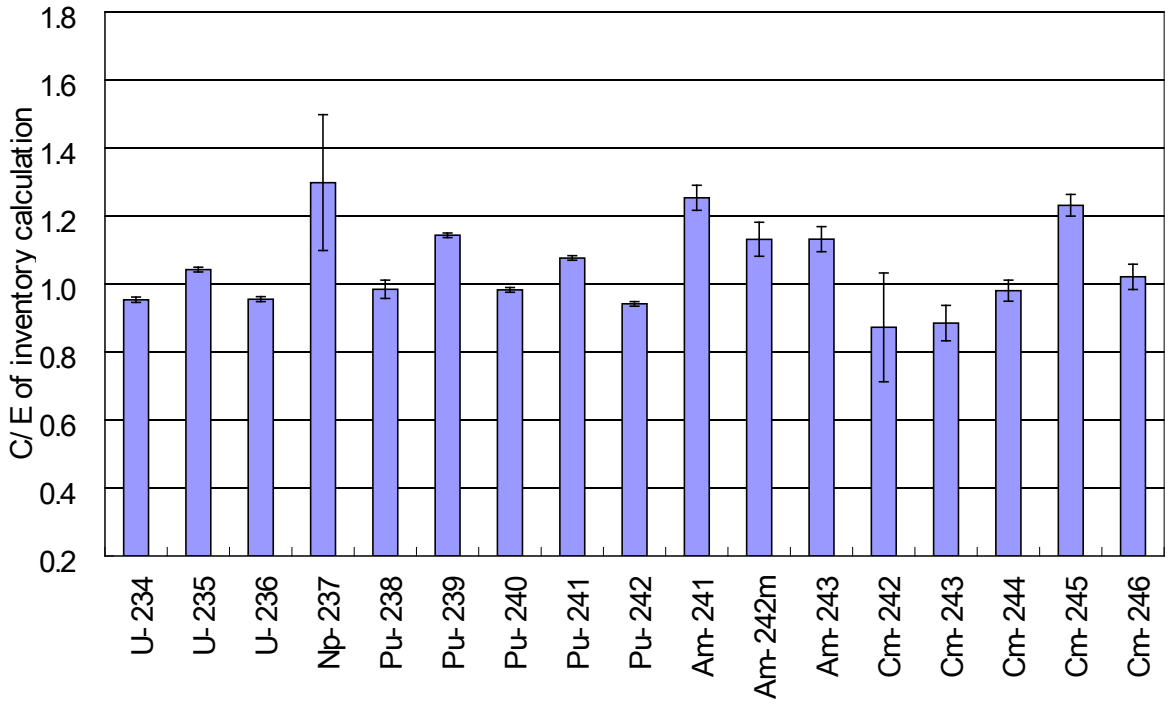


(a) Actinide

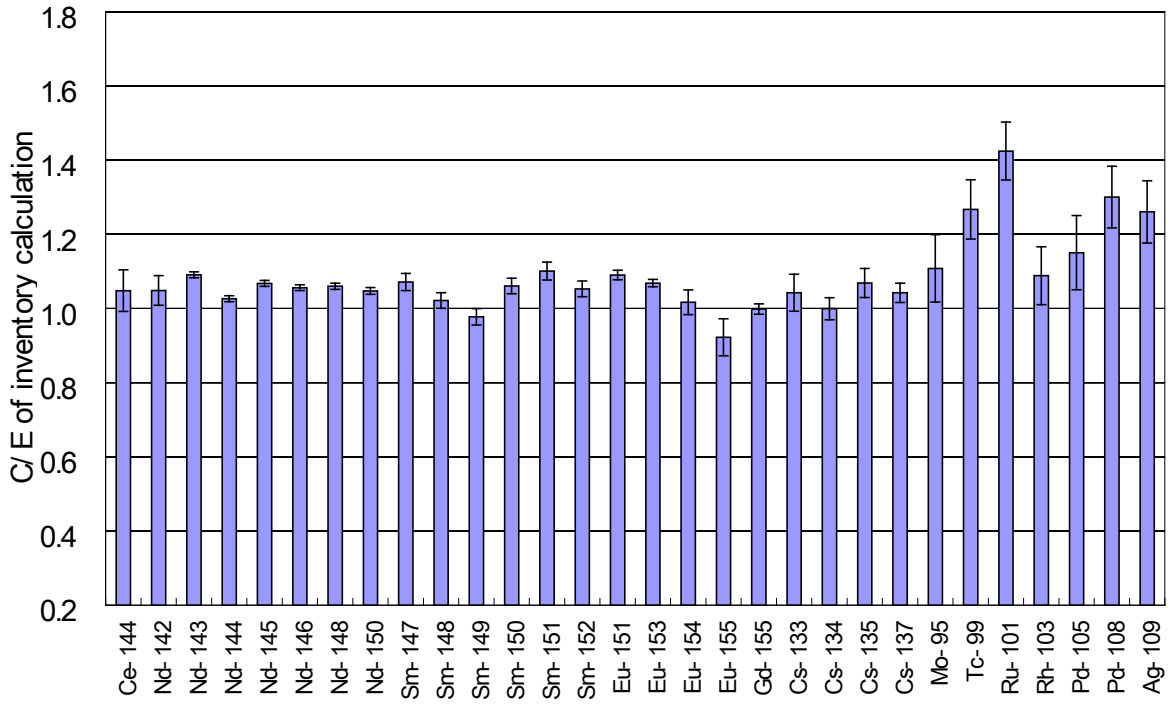


(b) FP

Fig. 4 Comparison between calculated and measured isotopic inventories for the PWR UO2 fuel (uncertainties (2 σ) caused by measurement errors)

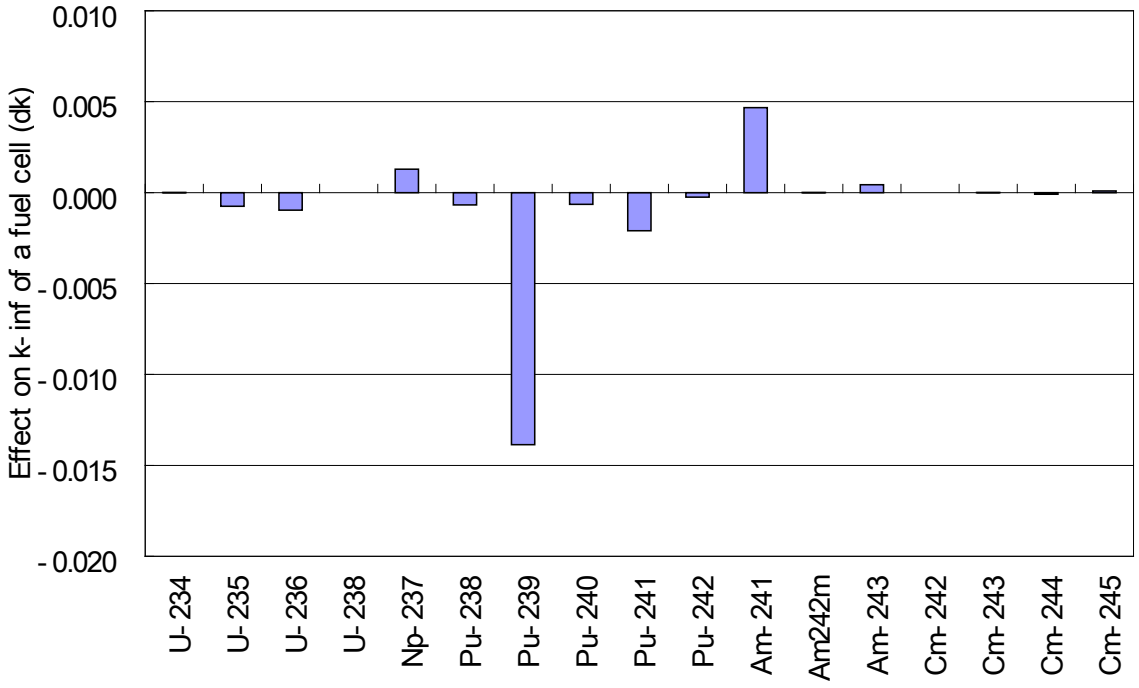


(a) Actinide

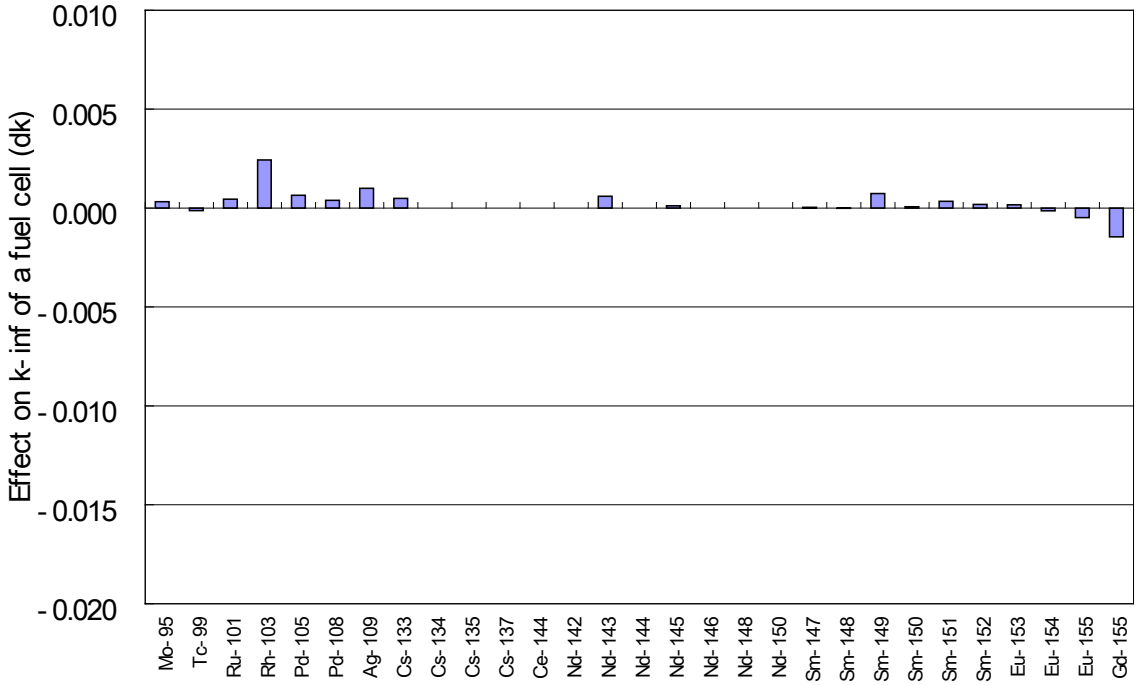


(b) FP

Fig. 5 Comparison between calculated and measured isotopic inventories for the BWR MOX fuel (uncertainties (2σ) caused by measurement errors)

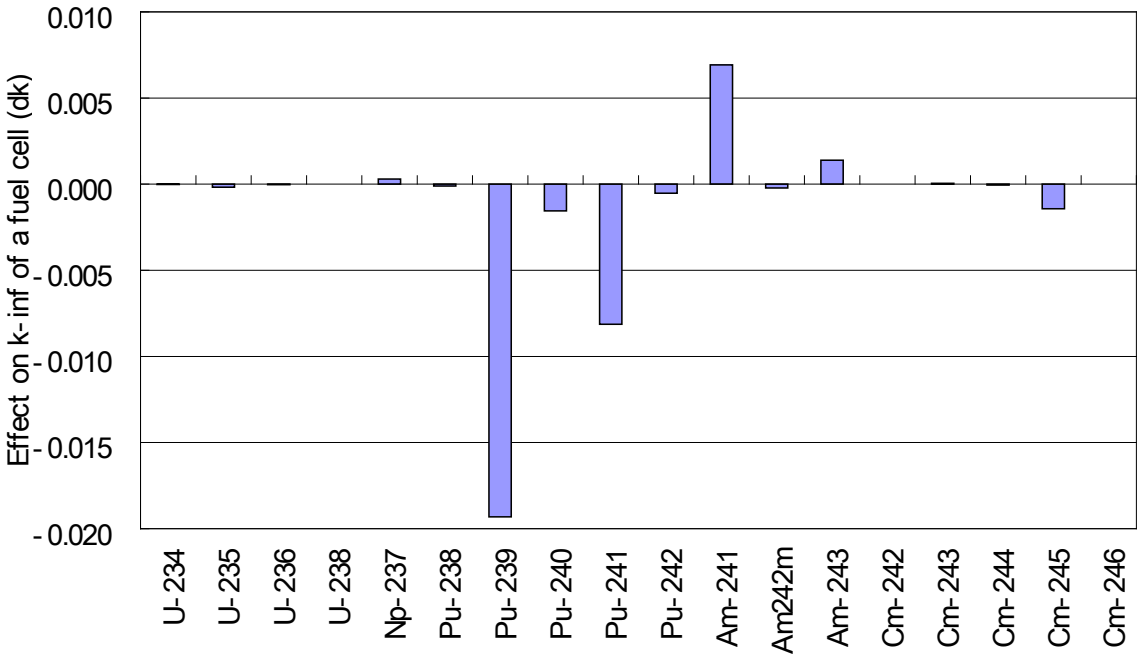


(a) Actinide

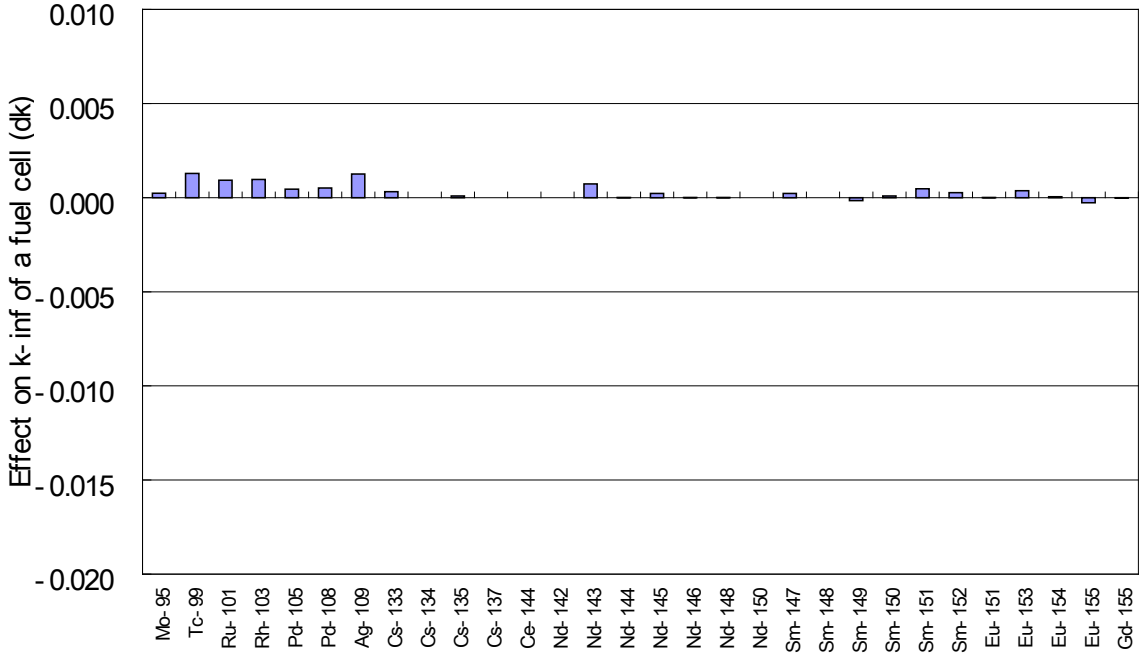


(b) FP

Fig. 6 Effect on k-inf of a fuel cell of the PWR UO2 bundle in the REBUS core



(a) Actinide



(b) FP

Fig. 7 Effect on k-inf of a fuel cell of the BWR MOX bundle in the REBUS core

3.3 Biases of Calculated Burnup Reactivity due to Inventory and Reactivity Calculations

The corrected inventories were applied to the core calculations of the irradiated fuel cores, and the values of k_{Hirr}^{irr} were calculated to obtain the burnup reactivity. The values of k_{Hirr}^{irr} was 0.99440 and 0.99620 for the irradiated bundle cores of the PWR UO2 and BWR MOX fuel, respectively. The change in k_{Hirr}^{irr} of the PWR UO2 fuel seemed to too small for the reactivity change of the infinite fuel cell, -0.70%dk. It was observed that the relative power of the irradiated fuel bundle are about half of the neighboring UO2 fuel rods of 3.3 wt% enrichment in the driver region, and decrease in the reactivity of the irradiated fuel bundle due to the inventory correction slightly increase the power of the neighboring 3.3 wt% UO2 fuel rods, which increase the core reactivity. The decrease in the reactivity of the fuel bundle and the increase of the neighboring driver region cancel each other to make the reactivity change by the corrected inventory small. On the other hand, the relative power of the irradiated fuel bundle of the BWR MOX is from 1.7 to 2.3 so that the decrease in the reactivity of the irradiated fuel bundle mainly affects on the power of the irradiated fuel bundle, and the cancellation observed in the PWR UO2 fuel is not brought. Therefore, neutron balance only in the region of the irradiated fuel bundle was considered for the change in k_{Hirr}^{irr} caused by the corrected inventory for the PWR UO2 fuel bundle. Adopting this treatment for the PWR UO2 fuel, the calculated burnup reactivity with the corrected inventories was shown in **Table 3** and also schematically shown in **Figs. 8** and **9**. From this analysis, the bias of the calculated burnup reactivity due to the inventory calculations are estimated to be 0.045 (=-2.522-(-2.567))and 0.102 (-2.437-(-2.539))%dk for the PWR UO2 and BWR MOX fuel bundles, respectively, which also means the bias due to the reactivity calculations -0.167 (=-2.567-(-2.40))$\pm$0.17 and -0.059 (=-2.539-(-2.48))$\pm$0.10%dk for the PWR UO2 and BWR MOX fuel bundles, respectively.

Table 3 Calculated burnup reactivity with corrected inventories

Unit: (%dk/kk')

Correction case/Fuel bundle	PWR UO2	BWR MOX
(a) Burnup reactivity with corrected inventories (Difference from that in Table 2)	-2.567 (-0.045)	-2.539 (-0.102)
(b) Burnup reactivity with corrected inventories (modifying Am and metallic FP) (Difference from that in Table 2)	-2.651 (-0.128)	-2.608 (-0.171)

Americium-241 and metallic FP nuclides are the nuclides showing prominent trend in C/Es and relatively large contribution to the reactivity change of typical infinite fuel cells. The major inventory of Am-241 caused by decay of Pu-241, since the ratios of Pu-241 to Am-241 at the discharge of the fuel assembly, from which the pellet samples were taken, are 30.17 and 8.07 for the PWR UO2 and BWR MOX fuel, and the cooling times for the isotopic inventory measurement are 7.2 and 4.7 years, respectively. The values of C/Es for Am-241 are fairly larger than those of Pu-241, which indicate that systematic errors in the measurements of Am-241 are large. The values of C/Es for some of metallic FP nuclides seem to be too large to accept as they are. Therefore, as an alternative inventory correction, it is assumed that the C/E of Am-241 equal to that of Pu-241 and the C/Es of the metallic FP nuclides are 1.0. The calculated values of burnup reactivity were shown in Table 2 and schematically shown in Figs. 8 and 9. In this case, the bias of the calculated burnup reactivity due to the inventory

calculations are estimated to be 0.128 and 0.171%dk for the PWR UO2 and BWR MOX fuel bundles, respectively, which also means the bias due to the reactivity calculations -0.251± 0.17 and -0.128±0.09%dk for the PWR UO2 and BWR MOX fuel bundles, respectively. The arrows in the figures schematically show these biases.

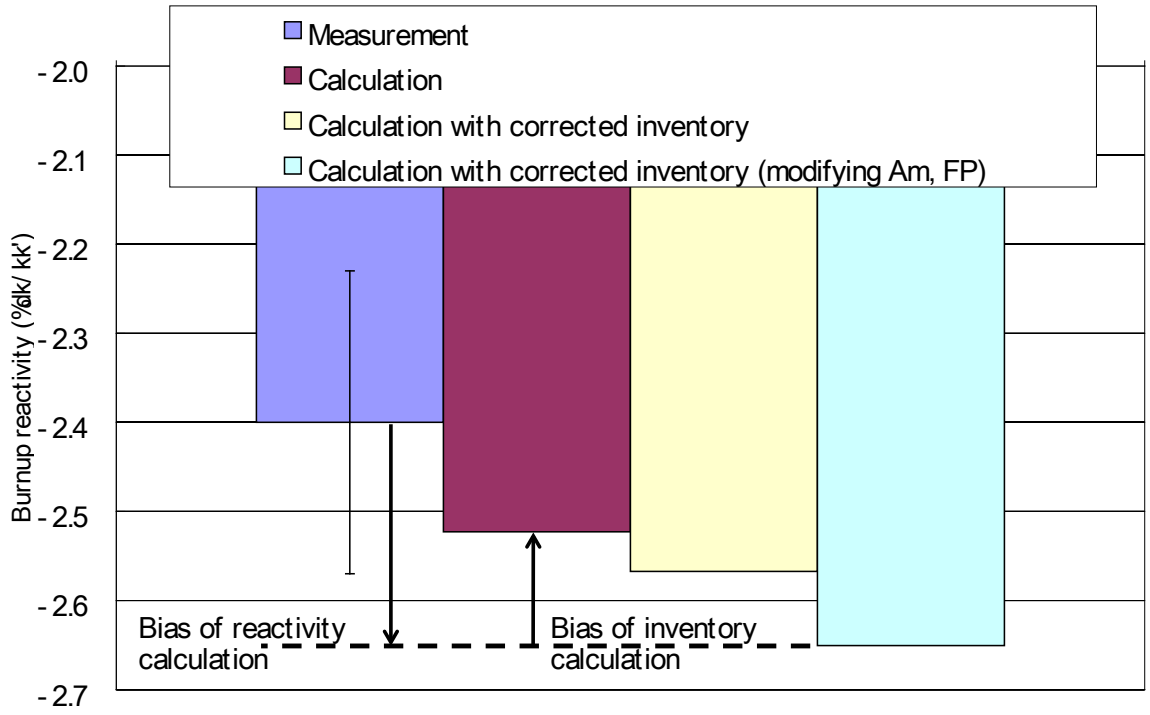


Fig. 8 Bias of burnup reactivity of the PWR UO2 fuel bundle

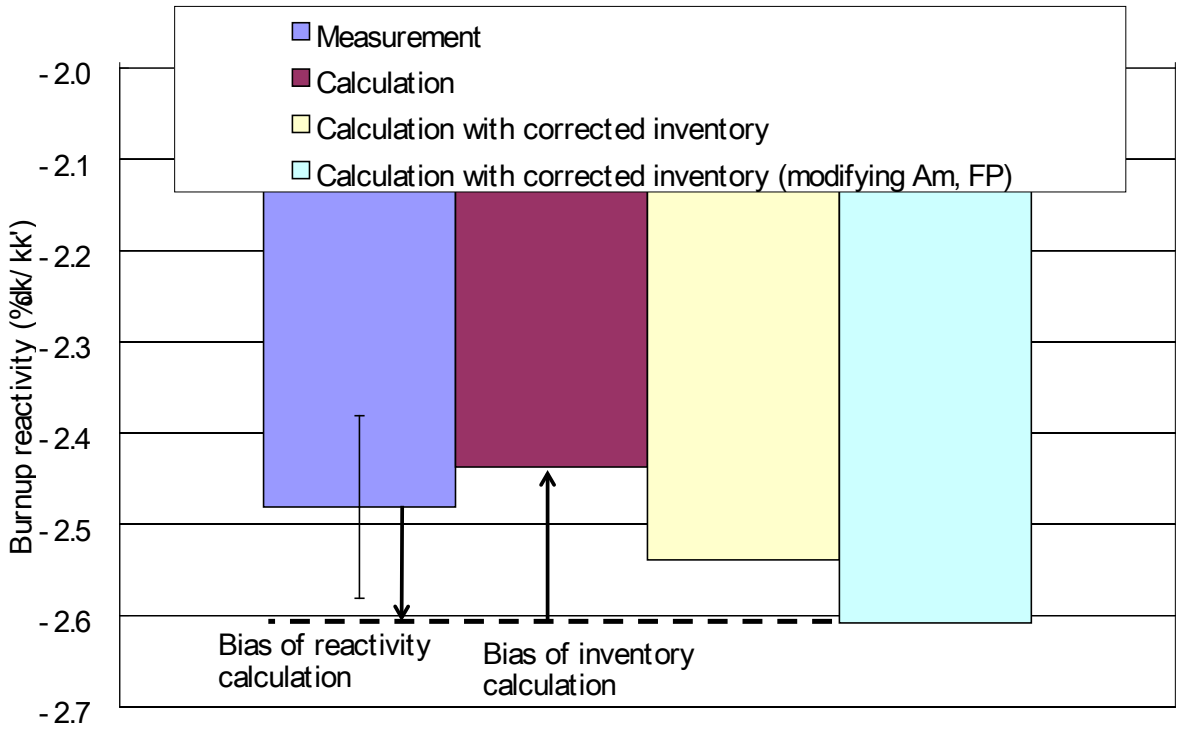


Fig. 9 Bias of burnup reactivity of the BWR MOX fuel bundle

4. Conclusions

The burnup reactivity of irradiated PWR UO2 (average burnup: 54.5GWd/t) and BWR MOX (average burnup: 60.6GWd/t) fuel bundles in the REBUS program was obtained using the measured critical water levels and water level reactivity coefficients of critical cores which contain the fresh and irradiated fuel bundles. Those are -2.40 and -2.48%dk/kk' for the PWR UO2 fuel and BWR MOX fuel bundles, respectively. The study for splitting calculation biases of the burnup reactivity into inventory and reactivity calculations was performed using measurement isotopic inventories of the irradiated fuel. The calculation biases for the PWR UO2 fuel bundle are +0.13 %dk/kk' for inventory calculation and -0.25 %dk/kk' for reactivity calculation, which makes total bias -0.12 %dk/kk'. Those for the BWR MOX fuel bundle are +0.17%dk/kk' for inventory calculation and -0.13 %dk/kk' for reactivity calculation, which makes total bias +0.04 %dk/kk'. The study shows that the calculation biases of the inventory and reactivity are in inverse relation to make the total biases smaller. The obtained information is useful to improve the analysis tools and models for further reduction of the calculation biases in the burnup reactivity.

References

1) P. Baeten et al., "The REBUS experimental programme for burn-up credit," *Proc. Int. Conf. on Nuclear Critical Safety, ICNC2003*, Tokai, Ibaraki, Japan, Oct. 20-24 (2003).
2) K. Kawashima, T. Yamamoto, K. Kamimura, "Analysis of reactor physics experiment for the irradiated LWR MOX fuels," *Proc. Int. Conf. ICAPP06*, Reno, NV, USA, June 4－8, 2006, Paper 6077 (2006).
3) T. Yamamoto, K. Kawashima, "Preliminary analysis of irradiated UO2 fuel core in REBUS program," *Proc. Int. Conf. ICNC2007*, St. Petersburg, Russia, May 28-June 1 2007 (2007).
4) B. Lance, A. Renard, D. Marloye *et al.*, "Status of the REBUS-PWR international programme," *Proc. Int. Conf. PHYSOR2002*, Seoul, Korea, Oct. 7－10, 2002 (2002).
5) B. Lance, D. Marloye, A. Renard *et al.*, "Preliminary analysis of the REBUS-PWR results," *Proc. IAEA Technical Meeting on "Advances in Applications of Burnup Credit to Enhance Spent Fuel Transportation, Storage, Reprocessing and Disposition,"* London, UK, Aug. 29- Sept. 2, 2005 (2005).
6) C. Venard, I. Duhamel, J. Raby *et al.*, "International programme REBUS french comparisons of the experimental data with the results of various criticality codes," *Proc. Int. Conf. ICNC2007*, St. Petersburg, Russia, May 28-June 1 2007 (2007).
7) F. Attale, H. D. Berger, W. Stach *et al.*, "Evaluation of the REBUS experiment on the Neckarwestheim II fuel for burn-up credit applications," *Proc. Int. Conf. ICNC2007*, St. Petersburg, Russia, May 28－June 1 2007 (2007).
8) C. Riffard, P. Marimbeau, B. Roque *et al.*, "International program REBUS experimental validation of spent fuel isotopic predictions for a 3.8% UO2 PWR with the "DARWIN" code system," *Proc. Int. Conf. ICNC2007*, St. Petersburg, Russia, May 28－June 1 2007 (2007).
9) A. Alander, S. Misu, S. Thareau *et al.*, "AREVA NP burnup credit investigation on irradiated MOX fuel within the REBUS BWR programme," *Proc. Int. Conf. PHYSOR 2008*, Interlaken, switzerland, Sep. 14-19 (2008)
10) T. Yamamoto, Y. Ando, Y. Hayashi, "Analysis of core physics experiments on irradiated BWR MOX fuel in REBUS program," *ibid*.
11) T. Yamamoto, K. Kawashima, Y. Ando *et al.*, " Analysis of core physics experiments on fresh and irradiated BR3 MOX fuel in REBUS program, *J. Nucl. Sci. Technol.*, **46**[5], 484 (2009).

12) Y.Nagaya, K.Okumura, T.Mori, M.Nakagawa, "*MVP/GMVP II: Genaral Purpose Monte Carlo Codes for Neutron and Photon Transport Calculations based on Continuous Energy and Multi-Group Methods*" JAERI-1348, (2005).
13) K. Okumura, T. Kugo, K. Kaneko *et al.*, *SRAC2006: A Comprehensive Neutronic Calculation Code system*, JAERI-Data/Code 2007-004 (JAERI) (2007).
14) K. Shibata, T. Kawano, T. Nakagawa et al., "Japanese evaluated nuclear data library version 3 revision-3: JENDL3.3," *J. Nucl. Sci. Technol.*, 39[11], 1125 (2002).
15) R. E. Alcouffe, *DANTSYS: A Diffusion Accelerated Neutral Particle Transport Code System*, LA-12969-M (June 1995).
16) T. Nakagawa, K. Shibata, S. Chiba et al., "Japanese evaluated nuclear data library version 3 revision-2," *J. Nucl. Sci. Technol.*, 32, 1259 (1995).