

Benchmark of evaluated nuclear data libraries using post-irradiation experimental data on fuel composition changes of the fast reactor JOYO

K. Yokoyama^{1,a} and T. Jin²

¹ Japan Atomic Energy Agency (JAEA), Reactor Physics Analysis and Evaluation Group, 4002, Narita, O-arai, Ibaraki 311-1393, Japan

² NESI Incorporation, 4002, Narita, O-arai, Ibaraki 311-1393, Japan

Abstract. Post irradiation experiment (PIE) data on depleted fuel composition of the experimental fast reactor JOYO MK-II have been accumulated since 1986 by the JNC (the former of JAEA). In the present study, all available PIE data of JOYO MK-II driver fuel were analyzed and integral data concerning ^{235}U depletion and ^{236}U generation were prepared. Both the integral data are sensitive to ^{235}U capture cross section and applicable to nuclear data benchmarks. The recent evaluated nuclear data libraries, JENDL-3.2, -3.3, JEFF-3.1 and ENDF/B-VII, have a tendency to overestimate the generation of ^{236}U . A cross section adjustment demonstrated that re-evaluation of ^{235}U capture cross section improved the overestimation.

1 Introduction

In fast reactor core design, it is important to improve the prediction accuracy of nuclear characteristics. A cross section adjustment technique using integral experimental data is an effective approach for the reduction of nuclear data induced uncertainties among analytical results. An adjusted cross section set, ADJ2000, based on a fast reactor experiment data base was developed in the past [1]. In the adjustments, a few hundred integral data were adopted; however, almost of these came from zero-power experiments. In order to improve reliability and applicability for power reactors, burn-up characteristics should be added to next version of the adjusted cross section library. At the JAEA, measurement of burn-up in post-irradiation experiment (PIE) is performed. In the experimental fast reactor JOYO MK-II, a total of 362 driver fuel and 69 irradiation test subassemblies have been irradiated. PIE data on depleted fuel composition of JOYO have been accumulated since 1986 [2]. In the present study, all the available PIE data of JOYO MK-II driver fuel were analyzed from the perspective of prediction accuracy evaluation of the isotopic composition change. The measured isotopic composition was translated to the rate of change of atomic number density (RC) in order to discuss prediction uncertainties induced from cross section data. The measured RCs were utilized for a benchmark calculation of the recent evaluated nuclear data libraries, JENDL-3.2 [3], JENDL-3.3 [4], JEFF-3.1 [5] and ENDF/B-VII [6].

2 Experiments

JOYO is Japan's first experimental fast reactor constructed at the O-arai Research and Engineering Center. The first criticality was achieved in 1977. There are three JOYO phases; namely, the MK-I, MK-II and MK-III. In the present study, the PIE data from driver fuels from the MK-II core were analyzed. JOYO MK-II, which is an irradiation core consisting

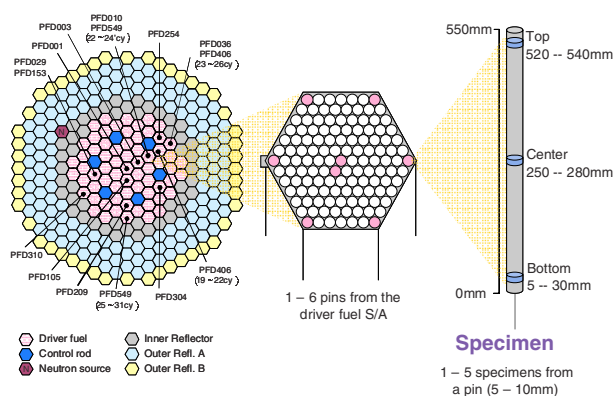


Fig. 1. JOYO MK-II core configuration and PIE specimens of the driver fuel subassemblies.

of U-Pu mixed oxide driver fuel and stainless steel reflector subassemblies, was in operation from 1982 to 1997. Typical U enrichment is ~ 12 or ~ 18 wt%, and Pu content is ~ 30 wt% [7]. Figure 1 shows the core configuration and an overview of PIE specimens. In the PIE, the isotopic composition of U, Pu and Nd at the end of life was measured for 75 specimens of driver fuel assemblies irradiated at various positions in the core region, whose burn-up was 0.3–8.7% FIMA (fission per initial metallic atom). Isotope dilution analysis is applied to the burn-up measurement. It is important to note that this experimental analysis method has been used consistently since 1984, because all PIE data from JOYO MK-II can be processed for a calculational analysis under the same condition.

3 Analysis

3.1 Experimental data translation

Rate of number density change

In the PIE, Pu content and U and Pu isotopic composition at the end of life (EOL) were measured. However, they have

^a Presenting author, e-mail: yokoyama.kenji09@jaea.go.jp

small sensitivity to cross section. In order to utilize these measurements for a nuclear data benchmark and/or cross section adjustment, the PIE data were translated to a rate of number density change per burn-up as follows:

$$RC = \frac{N_{BOL} - N_{EOL}}{N_{EOL}} \cdot \frac{1}{BU}, \quad (1)$$

which is more sensitive to nuclear data. Because the burn-up, BU , is measured by the Neodymium method in the PIE, the RC was normalized by the total number of measured fissions. In the case that the initial number density, N_{BOL} , was zero, such as ^{236}U , the number density generation per burn-up, N_{EOL}/BU , was evaluated instead.

Fitting evaluation

On the other hand, RC is also sensitive to the initial fuel composition and contains a systematic error induced by the initial composition uncertainty. Therefore, the least-square fitting method was applied to the evaluation of RC. The PIE data were classified for the fitting by 1) the initial fuel composition and 2) the neutron spectrum at the irradiation position. The neutron spectrum was evaluated by a spectrum index of $^{10}\text{B}(n,\alpha)$. This classification roughly corresponds to the irradiation position of the core. In the present analysis, an initial fuel composition of early cycles ($^{235}\text{U}/^{238}\text{U} = 12.2/87.8$ at%) was chosen as a fitting data set for uranium. Since both the content and isotopic compositions have variations; it is difficult to find a meaningful data set for plutonium. An initial fuel composition of middle cycles ($^{238}\text{Pu}/^{239}\text{Pu}/^{240}\text{Pu}/^{241}\text{Pu}/^{242}\text{Pu} = 0.7/68.8/21.4/6.7/2.3$ at%) was selected for plutonium. As for neutron spectrum classification, the hardest spectrum, i.e., the core center region, was selected in order to reduce the calculational model uncertainty.

Experimental uncertainty

It is necessary to evaluate the uncertainties of the derived experimental data. The random error of the RC was evaluated by the fitting error because the random errors of the experiments are reduced by the fitting. On the other hand, the systematic error is not reduced; therefore, the following systematic errors were taken into account: 1) systematic error of burn-up which comes from fission yield of Nd ($\sim 1.2\%$); and 2) experimental systematic error in isotope dilution analysis ($\sim 0.5\%$).

3.2 Calculational analysis

The calculational procedure is summarized in figure 2. The calculation was performed in three phases, whole core burn-up calculation, sample position burn-up calculation, and sensitivity analysis.

Burn-up calculation

In both the whole core and sample position burn-up calculation, ABBN-type 70-group constants based on JENDL-3.2 were utilized. In the whole core burn-up calculation, a neutron diffusion calculation with 3-dimensional Tri-Z geometry model was performed. In addition, the following model effects were corrected to cross section prior to the sample position burn-up calculation: 1) mesh effect;

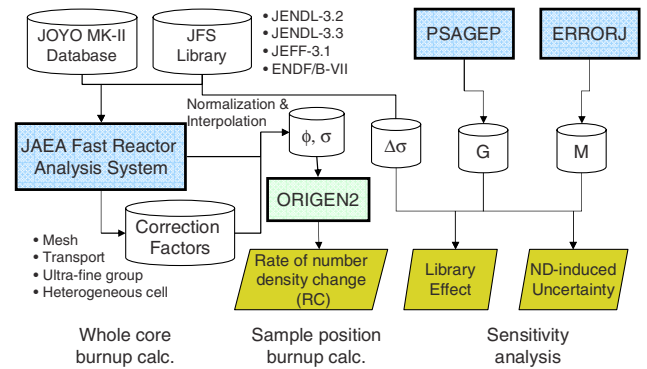


Fig. 2. Calculation methods and analysis procedure.

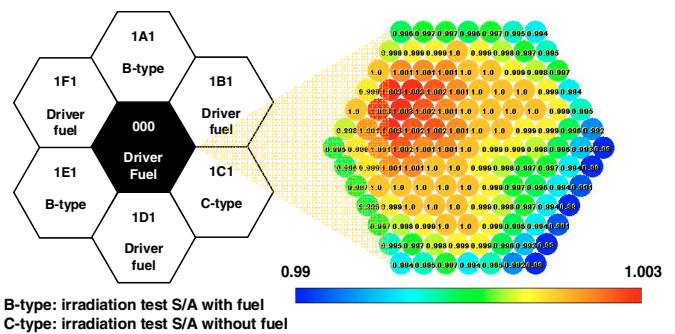


Fig. 3. Example of calculated flux distribution inside the driver fuel.

2) transport effect; 3) ultra-fine group cell calculation effect [8]; 4) heterogeneous cell effect; and 5) flux fluctuation inside the fuel subassembly. In the core calculation, the fuel assembly was separated using six triangle meshes, and the neutron flux at the irradiated position was evaluated by linear interpolation, as shown in figure 3. This figure presents the flux distribution inside the driver fuel at the core center.

Calculational model uncertainty

Calculational model uncertainty was evaluated by the correction factors because it is assumed that the calculational model uncertainty is proportional to the model effect. In the present study, the correction factors were assumed to have $\sim 30\%$ uncertainty, referring the adjustment study estimation [1]. The calculational model uncertainty (1σ) was evaluated as 1–2%.

Nuclear data induced uncertainty

In order to quantify integral nuclear data information included in the PIE data, a sensitivity analysis was applied to an evaluation of library effect and nuclear data induced uncertainty. Sensitivity coefficient with respect to nuclear data was calculated by generalized perturbation theory taking into account burn-up effects. Nuclear data induced uncertainty, U , is evaluated by the burn-up sensitivity coefficient, G , and covariance matrix, M .

$$U = GMG^t. \quad (2)$$

In order to calculate the burn-up sensitivity and to process the covariance matrix, PSAGEP code [9] and ERRORJ code [10] were utilized, respectively.

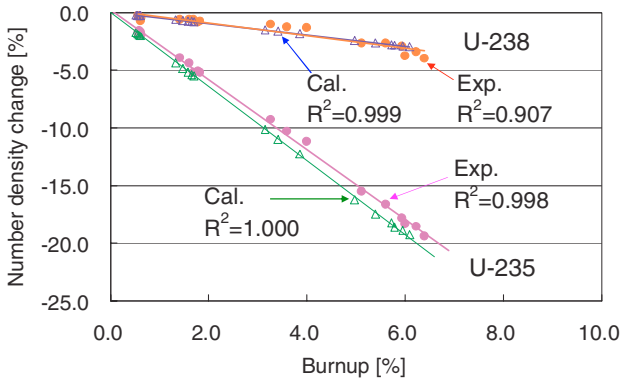


Fig. 4. Calculation and experimental results of number density change for ^{235}U and ^{238}U and fitting parameters.

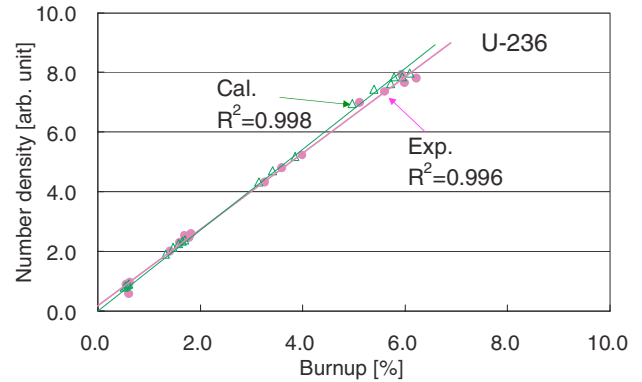


Fig. 5. Calculation and experimental results of number density change for ^{236}U and fitting parameters.

Library effect

The library effect caused by nuclide, x , and energy group, g , was calculated using the burn-up sensitivity coefficient as follows:

$$\frac{\Delta RC}{RC} = \sum_{x,g} G_{x,g} \left(\frac{\Delta \sigma_{x,g}}{\sigma_{x,g}} \right). \quad (3)$$

This method was numerically validated by comparing the direct calculation with two libraries. The direct calculation results agreed well with the sensitivity calculation results. For this purpose, ABBN type cross section libraries based on JENDL-3.3, JEFF-3.1 and ENDF/B-VII were utilized.

Cross section adjustment

In addition, cross section adjustments were performed. As a reference case, an updated version of ADJ2000; namely, ADJ2007, is used for the present study. It is an adjusted cross section library for fast reactors, which is based on JENDL-3.2 and uses the 237 integral data of ZPPR, FCA, MASURCA, BFS, as well as ADJ2000. As a comparative case, a cross section adjustment calculation with the 237 integral data and the PIE data; namely, ADJ2007+PIE, was performed. In both adjustments, it was confirmed that the cross section changes did not exceed 2σ of nuclear data uncertainty.

4 Analysis results

4.1 Results of fitting evaluation and C/E values

The results of the fitting evaluation for uranium are shown in figures 4 and 5. The fitting data sets for ^{235}U and ^{236}U were suitable for the fitting evaluation from the viewpoint of burn-up distribution and number of data. The fitting data set for ^{238}U has the same burn-up distribution; however, it exhibits relatively large variation. In the PIE, ^{238}U analysis requires an additional process due to the co-existence of mass number 238 in the isotopes of U and Pu. In addition, the original experimental errors can be enlarged by error propagation because the RC of ^{238}U is relatively small. The results of the fitting evaluation for plutonium are shown in figure 6. The fitting data set for plutonium were less suitable than that for uranium because of the small number of data. Although the data set for ^{241}Pu was corrected by decay effects due to the

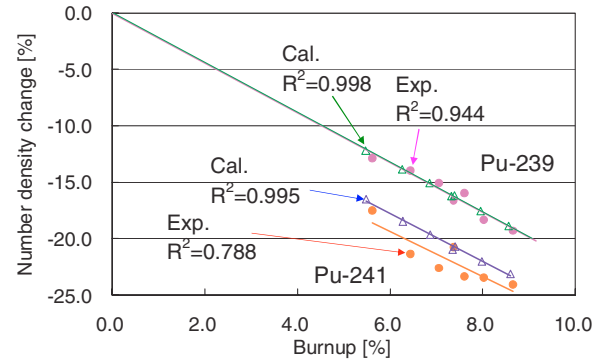


Fig. 6. Calculation and experimental results of number density change for ^{239}Pu and ^{241}Pu and fitting parameters.

operation history, the variation was quite large. ^{238}Pu , ^{240}Pu and ^{242}Pu were also evaluated by the same method; however, we were unable to accomplish a meaningful fitting evaluation.

Table 1. Analysis results of rate of number density change (RC) with JENDL-3.2 as a reference case.

	NDIU*	Calculation [FIMA ⁻¹]	Experiment [FIMA ⁻¹]	C/E
^{235}U	1.0%	$-3.20 \pm 1.2\%$	$-3.03 \pm 1.7\%$	1.06
^{236}U	2.1%	$+1.34 \pm 1.2\%$	$+1.27 \pm 2.0\%$	1.06
^{238}U	1.2%	$-0.48 \pm 1.6\%$	$-0.55 \pm 8.1\%$	0.88
^{239}Pu	1.2%	$-2.21 \pm 1.6\%$	$-2.21 \pm 10.9\%$	1.00
^{241}Pu	3.2%	$-2.11 \pm 1.6\%$	$-1.99 \pm 23.2\%$	1.06

* NDIU: nuclear data induced uncertainty.

The analysis results are summarized in table 1. The calculated RCs of ^{235}U and ^{236}U slightly overestimate the experimental RCs. The calculated RC of ^{238}U agreed with the measured RC within the uncertainty; however, it is not useful for nuclear data benchmarks because the experimental uncertainty was larger than the uncertainties induced by nuclear data and calculational model. C/E values for ^{239}Pu and ^{241}Pu were nearly equal to unity; however, their experimental uncertainty was larger than the other uncertainty as well as ^{238}U . For the present study, it is concluded that the PIE data of ^{235}U and ^{236}U are useful to nuclear data benchmarks.

4.2 Benchmark and adjustment

Table 2 shows benchmark results for the RC of ^{235}U and ^{236}U . All the evaluated nuclear data libraries overestimate the RC of ^{235}U by $\sim 5\%$. The cross section adjustment partly resolved the overestimation of the RC of ^{235}U by adjusting cross sections of ^{235}U capture/fission, sodium and steel inelastic scattering, as shown in figure 7. However, $\sim 3\%$ of discrepancy remained after the adjustment. The fact implies that there exist other reasons for the overestimation; i.e., unknown uncertainty for calculational model, experiment and nuclear data. As for the RC of ^{236}U , all evaluated nuclear data libraries overestimate. From the viewpoint of library effect, it is noteworthy that JENDL-3.3 overestimates by $\sim 12\%$ compared with JENDL-3.2. The cross section adjustment significantly improves the C/E value of ^{235}U . For this improvement, the effect of ^{235}U capture cross section adjustment is dominant because other reactions are not sensitive to the RC of ^{236}U . The energy-wise difference of the cross section among the libraries is shown in figure 8. There are no significant differences among the three libraries of JENDL-3.3, JEFF-3.1 and ENDF/B-VII in the energy region below 30 keV. However, only JENDL-3.3 has a larger cross section in the energy region above 30 keV.

Table 2. C/E values of RC evaluated by the recent evaluated nuclear data libraries and adjusted libraries with and without the PIE data.

	^{235}U	^{236}U
JENDL-3.2	1.056 (ref.)	1.059 (ref.)
JENDL-3.3	1.066 (+0.010)	1.121 (+0.062)
JEFF-3.1	1.060 (+0.004)	1.058 (-0.001)
ENDF/B-VII	1.054 (-0.020)	1.057 (-0.002)
ADJ2007	1.042 (-0.014)	1.037 (-0.022)
ADJ2007+PIE	1.032 (-0.024)	1.006 (-0.053)

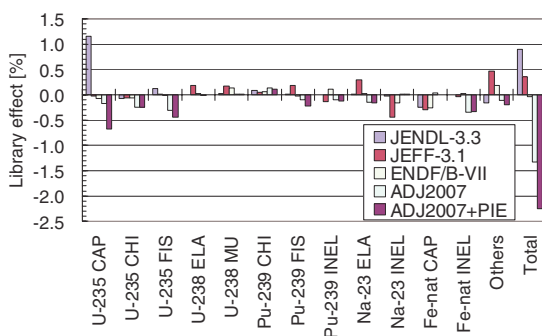


Fig. 7. Reaction-wise library effect for RC of ^{235}U .

This is the reason for the overestimation of JENDL-3.3. The adjusted library has a smaller cross section in the region

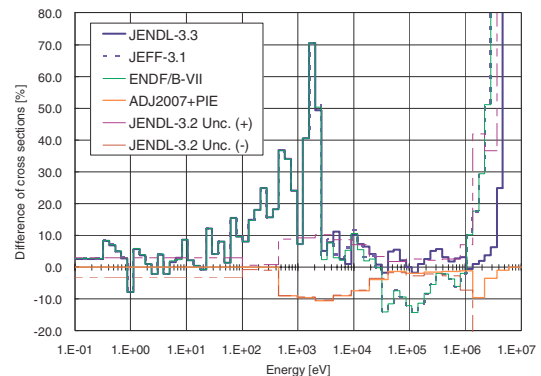


Fig. 8. Differences of ^{235}U capture cross section vs. JENDL-3.2.

below 30 keV, which improves the C/E value. This cross section change corresponds to 1σ uncertainty of JENDL-3.2.

5 Conclusions

The PIE data from JOYO MK-II driver fuels were analyzed, and integral data concerning ^{235}U depletion and ^{236}U generation were prepared. Both the integral data are sensitive to ^{235}U capture cross section and applicable to nuclear data benchmarks. All recent evaluated nuclear data libraries, JENDL-3.2, -3.3, JEFF-3.1 and ENDF/B-VII, have a tendency to overestimate the generation of ^{236}U . The cross section adjustment demonstrated that re-evaluation of ^{235}U capture cross section improved the overestimation.

The authors wish to thank Mr K. Numata at NESI for his help with burn-up calculation, and Dr S. Koyama and the JOYO staff for supplemental experimental information and fruitful discussion.

References

1. M. Ishikawa et al., J. Nucl. Sci. Technol. **2**, Suppl. 2, 1073 (2002).
2. S. Koyama et al., J. Nucl. Sci. Technol. **40**, 998 (2003).
3. T. Nakagawa et al., J. Nucl. Sci. Technol. **32**, 1259 (1995).
4. K. Shibata et al., J. Nucl. Sci. Technol. **39**, 1125 (2002).
5. A. Koning et al., JEFF Report 21 (2006).
6. M.B. Chadwick et al., Nucl. Data Sheets **107**, 2931 (2006).
7. Y. Ohkawachi et al., JNC TN9400 2003-029 (2003) (in Japanese).
8. T. Hazama et al., J. Nucl. Sci. Technol. **43**, 908 (2006).
9. K. Yokoyama et al., *Proceedings of Int. Conf. M&C 2005, Avignon, France* (2005), CD-ROM, No. 158.
10. G. Chiba, JAEA-Data/Code 2007-007 (2007).