Target accuracy of MA nuclear data and progress in validation by post irradiation experiments with the fast reactor "JOYO"

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This paper presents a discussion about the target accuracy of MA nuclear data for fast reactor cycle system development, as well as the validation work on those nuclear data by PIE analyses. The PIE analyses are in progress on fuels and MA samples (^{237}Np, ^{241}Am, ^{243}Am, ^{244}Cm) irradiated at the experimental fast reactor "JOYO". The analysis result on the first examined MA sample suggested the necessity of re-evaluation of the isomeric ratio for ^{241}Am capture reaction both in ENDF/B-VI and in JENDL-3.3. The above result contributes to the uncertainty-reduction both of burnup reactivity loss and of gamma energy release from fuel assemblies.

1. Introduction

Japan Nuclear Cycle Development Institute is developing a commercialized fast reactor cycle system that involves the recycling of minor actinide (MA) nuclides. To develop a burnup calculation method and to validate MA nuclear data, we have launched isotopic composition analyses of post irradiation examination (PIE) results. The irradiations were performed at the experimental fast reactor "JOYO".

MA recycling in fast reactors brings about the reduction of burnup reactivity loss, the increase of sodium void reactivity, and the increase of decay heat and neutron emission from fuel assemblies. Before getting into the PIE analyses, nuclear-data-induced uncertainties for above-mentioned quantities were evaluated tentatively as described in **Section 2**. We discussed the relation between target accuracy of those quantities and that of MA nuclear data. Then, the influential MA nuclear data having a priority in validation work were selected.

PIE analyses are in progress on (1) JOYO MK-I core fuel, (2) JOYO MK-II driver fuel, and (3) MA samples (^{237}Np, ^{241}Am, ^{243}Am, ^{244}Cm) irradiated at JOYO MK-II core. The former two analyses are dedicated to the development of a burnup calculation method, as well as to the validation of capture cross-sections for major heavy metal nuclides (^{238}U, ^{239}Pu, ^{240}Pu, etc.). The third one is expected to be applied directly to the validation of MA nuclear data. The present status of those PIE analyses is reported in **Section 3**.

2. Target accuracy of MA nuclear data

In this section, we try to estimate how much the uncertainty of MA nuclear data influences on reactor core parameters and fuel-cycle-related quantities. The contributions from MA nuclear data uncertainty were compared with the target accuracy assumed for each quantity. Then we discussed the necessity of MA nuclear data improvement and its target accuracy. A demonstration-type sodium-cooled FBR core concept[1] was employed as a reference. The reactor thermal power is 1600 MW_{th}. The operation cycle length is 375 EFPD. Fuel exchange is performed by 3 dispersed batches. The average fuel burnup is 85700 MWd/t. Fuel type is mixed oxide, and the plutonium enrichment is about 19 weight % of heavy metal. MA extracted from LWR spent fuel (denoted as LWR-MA hereafter) was assumed to be mixed to the active core fuel homogeneously. The MA contents were ^{237}Np/^{241}Am/^{243}Am/^{244}Cm = 49/30/16/5 (weight %), and its amount in heavy metal was set to 2.7 weight %.

Nuclear-data-induced uncertainty was evaluated by the following formula:

$$Standard\ deviation = \sqrt{\mathbf{GMG}^T}.$$

The **G** and **M** stand for a vector of cross-section sensitivity coefficients and a cross-section covariance matrix, respectively. They are expanded in the space of nuclide, reaction, and energy group number. Sensitivity coefficients were calculated by using the SAGEP code[2]. For covariance matrix, the JENDL-3.2 covariance file[3] was used for major nuclides. Since covariance for MA nuclides was not available, variance of MA nuclear data was tentatively deduced from discrepancies among the following major nuclear data libraries: JENDL-3.3[4], ENDF/B-VI release 5[5], and JEF-2.2[6]. Note that there is no confidence that the discrepancy among those libraries is equivalent to actual uncertainty.

First, fuel-cycle-related quantities such as decay heat, neutron emission, and gamma energy release from a fuel assembly loaded in the central core region are discussed. We restricted our investigation to cross section uncertainty concerning the prediction of burnt amount of heavy metal sources, and did not consider the errors associated with decay constants, Q-values, neutron and gamma yields, and so on. The time point of evaluation was set to 4 years after the discharge. The results are summarized in **Table 1**. For decay heat and neutron emission, ^{243}Am capture and ^{244}Cm capture reactions were found to be the main contributors. They were related to the net generation of important source nuclide ^{244}Cm. For gamma energy release, both

Table 1 Nuclide and reaction-wise contribution to the uncertainty for fuel-cycle-related quantities (1σ, %)

(600MWe-FBR, LWR-MA 2.7% added, 4-year-cooled)

Source Nuclide	Contributed Nuclide and Reaction	Decay heat	Neutron emission	Gamma energy release
^{238}Pu	^{238}Pu Capture	1.2		
	^{241}Am Isomeric Ratio	1.2		
^{241}Am	^{241}Am Capture			1.4
^{242m}Am	^{241}Am Isomeric Ratio			4.7
^{243}Cm	^{241}Am Capture			1.5
	^{242}Cm Capture			6.9
	^{243}Cm Capture			1.0
	^{243}Cm Fission			1.7
	^{241}Am Isomeric Ratio			2.3
^{244}Cm	^{243}Am Capture	2.4	4.5	
	^{244}Cm Capture	2.3	4.4	
(cf.) Target accuracy (tentative value)		5〜10	5〜10	5〜10

* Contributions of MA were deduced from the discrepancy of nuclear data libraries.
** Contributions less than 1.0% are not indicated.

isomeric ratio of ^{241}Am capture reaction and ^{242}Cm capture reaction gave relatively large contributions, around 5% or more. If we assume the target accuracy is 5–10%, then the cross-sections of above contributed reactions turn out to have necessity of improvement.

As important reactor core parameters for recycling MA, we investigated effective multiplication factor (k_{eff}), sodium void reactivity, and burnup reactivity loss. The nuclear-data-induced uncertainty for those quantities is listed in **Table 2** together with the target accuracy. For k_{eff} and sodium void reactivity, the contributions from MA nuclear data were relatively small compared with other data such as ^{238}U inelastic scattering, ^{239}Pu fission, ^{239}Pu fission spectrum, and ^{238}U delayed neutron yield (ν_d). Meanwhile, we found a certain amount of contributions from both ^{241}Am capture and its isomeric ratio to the burnup reactivity loss. These are related to the generation of the efficient fissile material ^{242m}Am. It is also necessary to improve capture cross-sections of major heavy metal nuclides (^{238}U, ^{239}Pu, ^{240}Pu) for the burnup reactivity loss.

As a result, the following MA nuclear data were selected to have a higher priority for improving the accuracy: ^{241}Am capture, ^{241}Am isomeric ratio, ^{243}Am capture, ^{242}Cm capture, and ^{244}Cm capture. We also found that the present discrepancies of MA nuclear data had relatively small impacts on fast reactor cycle systems as long as MA abundance in fuel is less than a few percent of heavy metal. However, since there is no confidence that the discrepancy among libraries is equivalent to the actual uncertainty, necessity for measurement and validation works still exists. Preparation of covariance files for MA nuclear data is also required for a practical uncertainty evaluation in the future.

Table 2 Nuclide and reaction-wise contribution to the uncertainty for reactor core parameters (1σ, %)

(600MWe-FBR, LWR-MA 2.7% added)

Contributed Nuclide and Reaction	k_{eff} (BOC)	Sodium void reactivity (BOC)	Burnup reactivity loss
^{238}U Capture	0.24	0.8	3.6
^{238}U Inelastic Scattering	0.47	2.0	
^{239}Pu Capture	0.12		2.2
^{239}Pu ν_p	0.12	0.7	
^{239}Pu Fission	0.47	2.6	1.5
^{240}Pu Capture	0.10	0.5	2.1
^{241}Pu Capture	0.10		1.3
^{241}Pu Fission	0.10	0.5	1.0
^{241}Am Capture			1.3
^{241}Am Isomeric Ratio	-	-	1.9
^{242m}Am Fission			0.6
^{242}Cm Fission			0.5
^{244}Cm Capture			0.5
Lumped FP (^{239}Pu) Capture			1.4
Lumped FP (^{241}Pu) Capture			0.6
^{23}Na Capture		0.8	
^{23}Na Elastic Scattering		1.1	
^{23}Na Inelastic Scattering		1.7	
^{23}Na μ-average		0.6	
Fe Capture	0.10	0.7	
Fe Elastic Scattering		1.0	
Fe Inelastic Scattering	0.28	0.7	
^{239}Pu Fission Spectrum	0.40		
^{238}U ν_d	-	1.9	1.9
^{239}Pu ν_d	-	1.2	1.2
^{241}Pu ν_d	-	0.7	0.7
Others	0.28	1.4	1.0
Total (root of sum of squares)	0.94	5.2	6.5
(cf.) Target accuracy (tentative value)	0.3	10	5

* Contributions of MA were deduced from the discrepancy of nuclear data libraries.

** Contributions less than 0.05% (1.0%) are not indicated for k_{eff} (for sodium void reactivity and burnup reactivity loss).

3. Progress in PIE analyses

(1) JOYO MK-I core fuel

So far, most of the analyses for JOYO MK-I core fuel have been completed. The ^{235}U enrichment ($^{235}U/U$) of the fuel was about 23 weight %, and the plutonium enrichment (Pu/(U+PU)) was about 18 weight %. We analyzed about 70 specimens up to 5% of burnup. The neutron flux history was calculated by 3-dimmensional whole core diffusion calculation taking into account the fuel exchange pattern through 11 operation cycles. The effective cross-sections were prepared from the fast reactor group constant set JFS-3-J3.2R based on JENDL-3.2[7]. Nuclide depletion calculation aiming at each PIE specimen was performed by the ORIGEN2 code with the neutron flux history and spectrum obtained by the preceding whole core calculation. The C/E values for main nuclides (^{235}U, ^{239}Pu, ^{241}Pu) didn't show any significant inconsistency. However, large dispersions were observed in C/E values for minority isotopes such as ^{238}Pu, ^{240}Pu, and ^{242}Pu. Those dispersions possibly came from measurement. Such a discrepancy was reduced in the latest MK-II driver fuel PIE data, for which refined experimental technique had been applied. We carry on finding out the cause of larger discrepancy of the MK-I C/E values, while proceed to the MK-II driver fuel analyses.

(2) MA sample irradiation

We have just started the PIE analysis of the MA samples (^{237}Np, ^{241}Am, ^{243}Am, ^{244}Cm) irradiated at JOYO MK-II core. Twenty-five MA samples were loaded at the two irradiation positions in the 3rd and the 5th assembly row (see **Fig. 1**). The irradiation was performed for 200-250 EFPD during the period of 1994 to 1999. PIE of the first sample (one of the ^{243}Am samples) was finished in October, 2003. The preliminary analysis result on the ^{243}Am sample is presented in the rest of this paper. The sample was loaded in the 3rd row, and the axial position was +350 mm upper from the core midplane (core height was 55 cm). That is, the sample was loaded in upper reflector region, and it was adjacent to control rod absorber. We suspected that the calculation modeling error could be large.

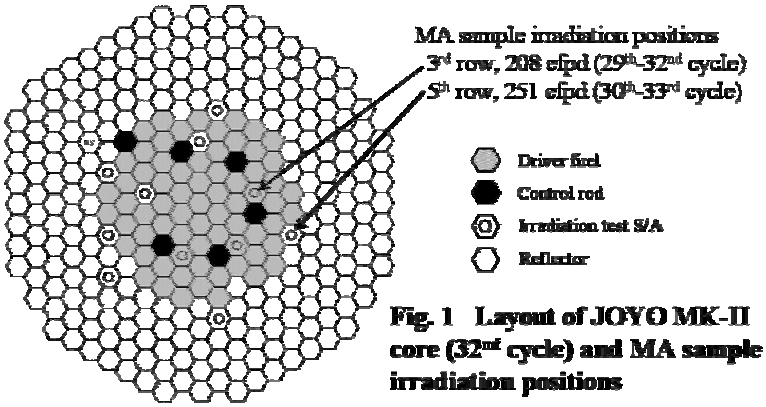

Fig. 1 Layout of JOYO MK-II core (32nd cycle) and MA sample irradiation positions

The ^{243}Am sample was initially composed of 12.2% of ^{241}Am and 87.8% of ^{243}Am. We focused on the generation of ^{242m}Am from ^{241}Am, as well as that of ^{244}Cm from ^{243}Am. Main purpose of the former transmutation process was to validate the isomeric ratio (IR) of ^{241}Am capture reaction. There exist only two IR evaluations with following large discrepancy: about 0.8 (ground/(ground + meta)) given from ENDF/B-VI under fast reactor spectrum, while about 0.7 from JENDL-3.3. This discrepancy influenced strongly on the generation amount of ^{242m}Am.

Preliminary calculation method is illustrated in **Fig. 2**. Burnup calculation of MA sample was carried out by the use of the ORIGEN2 code. The power history during MA irradiation was referred to the data from JOYO core management code system[8]. The absolute flux level and neutron spectrum were obtained by 70-group 3-dimmensional whole core static calculation, where the transport and mesh

effects were corrected. The effective cross section of control rod was prepared by the reaction rate ratio preservation (RRRP) method[9]. Infinitely-diluted cross-sections for MA nuclides were obtained from major nuclear data libraries (JENDL-3.2, JENDL-3.3, ENDF/B-VI release 5, and JEF-2.2), which were condensed into 1-group to be used in ORIEGN2 calculation.

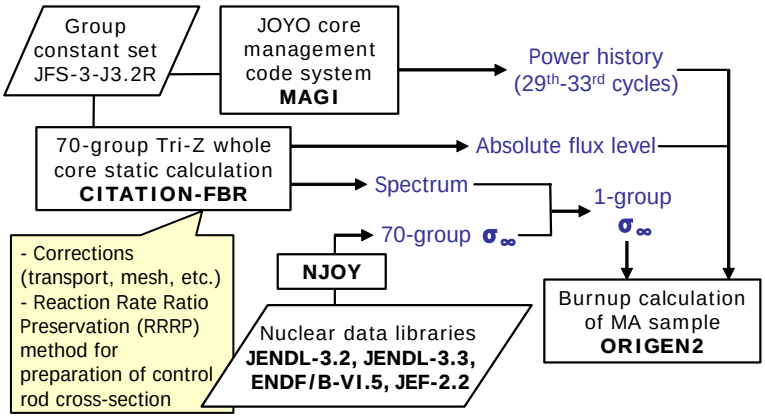

Fig. 2 Preliminary calculation method for the MA samples

The result of the preliminary calculation was summarized in **Table 3**. Assuming IR = 0.8 and using JENDL-3.2, we obtained the preliminary C/E value of 1.30 for the abundance ratio of ^{242m}Am/^{241}Am. In the case of IR = 0.85, the C/E value reduced to 1.00. To use other nuclear data libraries (IR was set to 0.85) changed the C/E value no more than 6%. The experimental error was about 2% via mass spectroscopy, which was small enough. The above results implied the possibility that the IR of ^{241}Am capture reaction lies around 0.85. This is consistent with the independent PIE results with PFR[10] and PHENIX[11]. Necessity of re-evaluation of the IR both in ENDF/B-VI and in JENDL-3.3 is suggested.

For the abundance ratio of ^{244}Cm/^{243}Am, the present calculations were systematically underestimated by 10-20%. We have met the difficulty in measuring the ratio of Cm/Am, in which relatively large experimental error (about 10%) arose from alpha spectroscopy. We now try to improve the measurement accuracy by means of the isotope dilution analysis.

Finally, calculation modeling dependency for the ^{243}Am sample was investigated. The heterogeneity effect of the sample-loaded assembly and the self-shielding effect of sample nuclides were found to be small. However, by the reason that the sample was loaded in reflector region with adjacent to control rod absorber, there was 5% of transport effect on neutron flux level, and the 3-4% of changes in one-group capture cross-sections for ^{241}Am and ^{243}Am took place from ambiguity of control rod modeling in the RRRP calculation. Significant uncertainty from nuclear data of structure material could also be anticipated. So, we had better concentrate on the other samples loaded in core region in future detailed analysis. Anyway, single result we have just obtained is never enough. We carry on the

Table 3 Comparison of C/E values for the isotopic abundance ratio of irradiated MA samples

	PFR [Ref.10]	JOYO (preliminary result on the ^{243}Am sample)					PHENIX [Ref.11]
Nuclear data library	JENDL-3.2	JENDL-3.2	JENDL-3.2	JENDL-3.3	ENDF/B-VI.5	JEF-2.2	JEF-2.2
^{241}Am Isomeric ratio	0.80	0.80	0.85	0.85	0.85	0.85	0.85
^{242m}Am / ^{241}Am	1.29	1.30	1.00	1.02	0.94	1.03	1.03
^{244}Cm / ^{243}Am	0.95	0.84	0.84	0.84	0.83	0.88	0.96

analyses for the remaining samples, increase the number of results, and apply detailed calculation modeling. The error estimation for both experiment and calculation will work out for practical utilization of the analysis results.

4. Summary

To develop a commercialized fast reactor cycle system involving the recycling of MA nuclides, we have launched the isotopic composition analyses of PIE results for fuels and MA samples (^{237}Np, ^{241}Am, ^{243}Am, ^{244}Cm) irradiated at the experimental fast reactor "JOYO". For reducing the uncertainty of burnup reactivity loss, as well as neutron emission and gamma energy release from fuel assemblies, the following MA nuclear data are selected to have a higher priority in accuracy-improvement: ^{241}Am capture, ^{241}Am isomeric ratio, ^{243}Am capture, ^{242}Cm capture, and ^{244}Cm capture. The analysis result on the first examined MA sample (one of ^{243}Am samples) implies the possibility that the ^{241}Am isomeric ratio lies around 0.85, which suggests the necessity of re-evaluation of the data both in ENDF/B-VI and in JENDL-3.3. We carry on the analyses for the remaining samples, increase the number of results, and apply detailed calculation modeling in order to sophisticate the interpretation of analysis results.

References

[1] H. Hayashi, *et al.*, *Progress Report of the Design Study on a Large Scale Reactor – Design Study on a 600MWe Class Plant* –, PNC TN9410 92-137 (1992), [in Japanese].

[2] T. Takeda and T. Umano, "Burnup Sensitivity Analysis in a Fast Breeder Reactor – Part I: Sensitivity Calculation Method with Generalized Perturbation Theory", *Nucl. Sci. Eng.*, **91**, 1 (1985).

[3] K. Shibata, *et al.*, "JENDL-3.2 Covariance File", *J. Nucl. Sci. Technol.*, **Suppl. 2**, 40 (2002).

[4] K. Shibata, *et al.*, "Japanese Evaluated Nuclear Data Library Version 3 Revision-3: JENDL-3.3", *J. Nucl. Sci. Technol.*, **39**, 1125 (2002).

[5] Cross Section Evaluation Working Group, *ENDF/B-VI Summary Documentation*, BNL-NCS-17541 (ENDF-201) (1991).

[6] OECD/NEA (Ed.), *The JEF-2.2 Nuclear Data Library*, JEFF Report 17 (2000).

[7] T. Nakagawa, *et al.*, "Japanese Evaluated Nuclear Data Library Version 3 Revision-2: JENDL-3.2", *J. Nucl. Sci. Technol.*, **32**, 1259 (1995).

[8] Y. Ohkawachi, *et al.*, *JOYO MK-II Core Characteristics Database – Update to JFS-3-J3.2R* –, JNC TN9400 2003-029 (2003), [in Japanese].

[9] K. Sugino and T. Iwai, *Advances in Methods of Commercial FBR Core Characteristics Analyses – Investigations of a Treatment of the Double-heterogeneity and a Method to Calculate Homogenized Control Rod Cross Sections* –, PNC TN9410 98-067 (1998), [in Japanese].

[10] K. Tsujimoto, *et al.*, "Validation of Minor Actinide Cross Sections by Studying Samples Irradiated for 492 Days at the Dounreay Prototype Fast Reactor – II: Burnup Calculations", *Nucl. Sci. Eng.*, **144**, 129 (2003).

[11] R. Soule and E. Fort, "Contribution to the Validation of JEF2 Actinide Nuclear Data: Analysis of Fuel and Sample Irradiation Experiments in PHENIX", *Proc. Int. Conf. on Future Nuclear Systems, Global '97*, Yokohama, Japan, Oct. 5-10, 1997, p.1332 (1997).