

# PIE Analysis for Minor Actinide

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Minor actinide (MA) is generated in nuclear fuel during the operation of power reactor. For fuel design, reactivity decrease due to it should be considered. Out of reactors, MA plays key role to define the property of spent fuel (SF) such as  $\alpha$ -radioactivity, neutron emission rate, and criticality of SF. In order to evaluate the calculation codes and libraries for predicting the amount of MA, comparison between calculation results and experimentally obtained data has been conducted. In this report, we will present the status of PIE data of MA taken by post irradiation examinations (PIE) and several calculation results.

## 1. Introduction

Minor Actinide (MA) has been a key issue in the nuclear data evaluation. One of the reasons is reactivity change due to MA should be considered in the fuel design, especially in high burnup MOX fuel. Another reason is MA is a main target in the strategy of partitioning and transmutation (P&T) of long-life radioactive isotopes. Of course, concerning the property of spent fuel (SF), amount of MA defines  $\alpha$ -radioactivity, decay heat, and neutron emission rate. It also affects the criticality of SF.

Validation of calculation codes and libraries has been carried out by comparison between calculation results and experimental data. However, required data taken by Post Irradiation Examinations (PIE) are not enough, and many analyses present discrepancies between calculations and PIE data. To improve the calculation, well-organized comparison using several codes and libraries are essential. This report describes a status of measured isotopic composition data of MA taken by PIE, and several studies to obtain useful information from comparison between calculations and experimental results. It includes examples of PIE data of UO<sub>2</sub> and MOX fuel, status of PIE analysis by French institute, and comparison of MA amount between calculations using JENDL-3.2[1] and JENDL-3.3[2].

## 2. Requirement of PIE Data

**Table 1** shows the typical amount of MA in spent UO<sub>2</sub> and MOX fuel without cooling time<sup>1</sup>. Since fresh MOX fuel contains plutonium and <sup>241</sup>Am, spent MOX fuel contains much <sup>241</sup>Am and curium isotopes. Higher burnup increases the amount of MA in SF. Current trend "higher burnup" and "using MOX" emphasize the importance of MA data.

**Table 2** shows the data concerning the MA generation. Since MA accumulates through several generation paths, to revise the calculation results, the chain analysis should be conducted to check the consistency of cross section data in each generation chain, and contribution of each path should be evaluated to determine the priority of revision of cross section data. For example, in the thermal reactor condition, most important generation path of <sup>238</sup>Pu is the path starting from (n,2n) reaction of <sup>238</sup>U, instead of <sup>239</sup>Pu (n,2n) reaction [3].

During the cooling time, some MA increase through the decay of parent isotopes.<sup>237</sup>Np accumulates during long cooling time by the  $\alpha$ -decay of <sup>241</sup>Am, and if cooling time is longer than several thousands years, all <sup>241</sup>Am become <sup>237</sup>Np. These facts imply that the improvement of one MA

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<sup>1</sup> MA represents actinides except for uranium and plutonium. However, considering the status of <sup>238</sup>Pu calculation results, we treat <sup>238</sup>Pu as MA in this report.

cross section has another effect and the importance depends on the considering time scale.

**Table 1** Amount [g/THM]\* of MA in PWR  
(Continuous Operation of PWR under constant specific power)

	PWR UO <sub>2</sub>		PWR MOX	
	40 GWd/t	50GWd/t	40GWd/t	50GWd/t
<sup>237</sup> Np	551.1	716.0	150.9	181.9
<sup>238</sup> Pu	184.6	310.2	2674	2829
<sup>241</sup> Am	43.64	56.30	1851	1726
<sup>243</sup> Am	106.8	209.4	1628	1903
<sup>244</sup> Cm	32.89	86.45	703.1	1004
<sup>245</sup> Cm	2.116	6.660	78.71	133.6

\* Gram per Ton Initial Heavy Metal

To collect basic data in order to contribute to develop nuclear fuel cycle facility, several post irradiation examinations were done from 1960's. **Table 3** summarizes the status of PIE data of MA. In the public-opened database on spent fuel composition SFCOMPO [4], 53 measurements of <sup>237</sup>Np from 7 reactors, 107 measurements of <sup>241</sup>Am from 10 reactors, 65 measurement of <sup>243</sup>Am from 7 reactors are archived. For curium isotopes, we have more than 100 data of <sup>244</sup>Cm.

Few number of <sup>245</sup>Cm measurements are stored in the database. This is because <sup>244</sup>Cm is main contributor to neutron emission from SF and relatively short half-life (18.1 year) and amount of <sup>245</sup>Cm is small in normal UO<sub>2</sub> SF. However, in some case, curium isotopes would become important in spent MOX fuel reactivity estimation. In MOX fuel of 60GWd/t using initial composition of multiple recycled Pu, curium isotopes (<sup>242,243,244,245</sup>Cm) have reactivity worth of approximately 900 pcm at 5 year cooling, even though MOX fuel of 40GWd/t using first generation Pu, it is approximately 60 pcm.<sup>2</sup> [5]

**Table 2** Basic Data Concerning the MA Generation  
(PWR 17×17; UO<sub>2</sub>; <sup>235</sup>U 4.1wt%; JENDL-3.3)

	Half life [year]	Main Generation chain	σ <sub>c</sub> [barn]*	σ <sub>f</sub> [barn]*
<sup>237</sup> Np	2.14×10 <sup>6</sup>	<sup>238</sup> U(n,2n) <sup>237</sup> U ⇒ β <sup>-</sup> (6.75day) <sup>236</sup> U(n, γ) <sup>237</sup> U ⇒ β <sup>-</sup> (6.75day) <sup>241</sup> Am ⇒ α (432year)	32.57	0.5367
<sup>238</sup> Pu	87.7	<sup>237</sup> Np(n,γ) <sup>238</sup> Np ⇒ β <sup>-</sup> (2.11day) <sup>242</sup> Cm ⇒ α (163day) <sup>239</sup> Pu (n,2n)	29.30	2.398
<sup>241</sup> Am	432	<sup>241</sup> Pu ⇒ β <sup>-</sup> (14.4year)	118.32	1.211
<sup>243</sup> Am	7,370	<sup>241</sup> Pu(n,γ) <sup>242</sup> Pu (n,γ) <sup>243</sup> Pu ⇒ β <sup>-</sup> (5h)	49.452	0.4439
<sup>244</sup> Cm	18.1	<sup>243</sup> Am(n,γ) <sup>244/244m</sup> Am ⇒ β <sup>-</sup> (10.1h/26m) <sup>243</sup> Cm (n,γ)	17.57	0.8197
<sup>245</sup> Cm	8,500	<sup>244</sup> Cm (n,γ)	17.80	117.5

<sup>2</sup> Reactivity worth is defined as pcm = 10<sup>5</sup>×ln(k<sub>reference</sub>/k) in the reference 3.

It seems that we have large amount of MA measurement data. However, many of them were taken in 1960's and 1970's and data that have higher burnup more than 40 GWd/t are scarce. Several decades ago, European countries had large and open program of PIE[6]. However, it is difficult to find opened activity in them now. In Belgium, Belgonucleaire (BN) has been organizing several PIE activities. ARIANE [7] is the typical case conducted under international collaboration managed by BN. In the program, PIE data of 15 samples were taken from SF irradiated in two PWR and one BWR. The example of analysis of ARIANE data is shown in the reference[8]. However, the experimental data have not been freely opened and published in unclassified reports.

It is well known that France has large PIE activities to verify their codes and libraries.[9] However, data have not been opened because they are funded by French industry.

In Japan, JAERI has been conducted PIE during 1980's and 1990's. The data taken in the activities are widely used especially for benchmarking of codes and libraries [10,11,12]. Especially, the data from Takahama-3 is only published PIE data set for PWR 17×17 UO<sub>2</sub> fuel of 4.1wt% enrichment close to 50 GWd/t. However, there is no further PIE program in JAERI. Considering the situation of experimental facilities, it seems difficult to obtain new measurement data from unclassified report.

**Table 3** Number of PIE Data Stored in SFCOMPO

	Number of Samples <sup>§</sup>			Number of Assembly	Number of Fuel Pin	Burnup [GWd/t]			
	Total	Taken from				0 - 19.99	20.00 - 29.99	30.00 - 39.00	40.00 - 60.0
		PWR	BWR						
<sup>237</sup> Np	53	29(5)*	24(2)	12	15	14	12	19	8
<sup>238</sup> Pu	128	88(7)	40(3)	18	41	27	55	38	8
<sup>241</sup> Am	107	69(7)	38(3)	17	38	26	41	32	8
<sup>243</sup> Am	65	47(6)	18(1)	11	20	14	25	20	6
<sup>244</sup> Cm	110	76(6)	34(2)	13	34	24	51	29	6
<sup>245</sup> Cm	28	11(1)	17(1)	2	4	5	5	12	6

\* Figures in ( ) are numbers of reactors.

§ Number of Sample includes Cross Check Data.

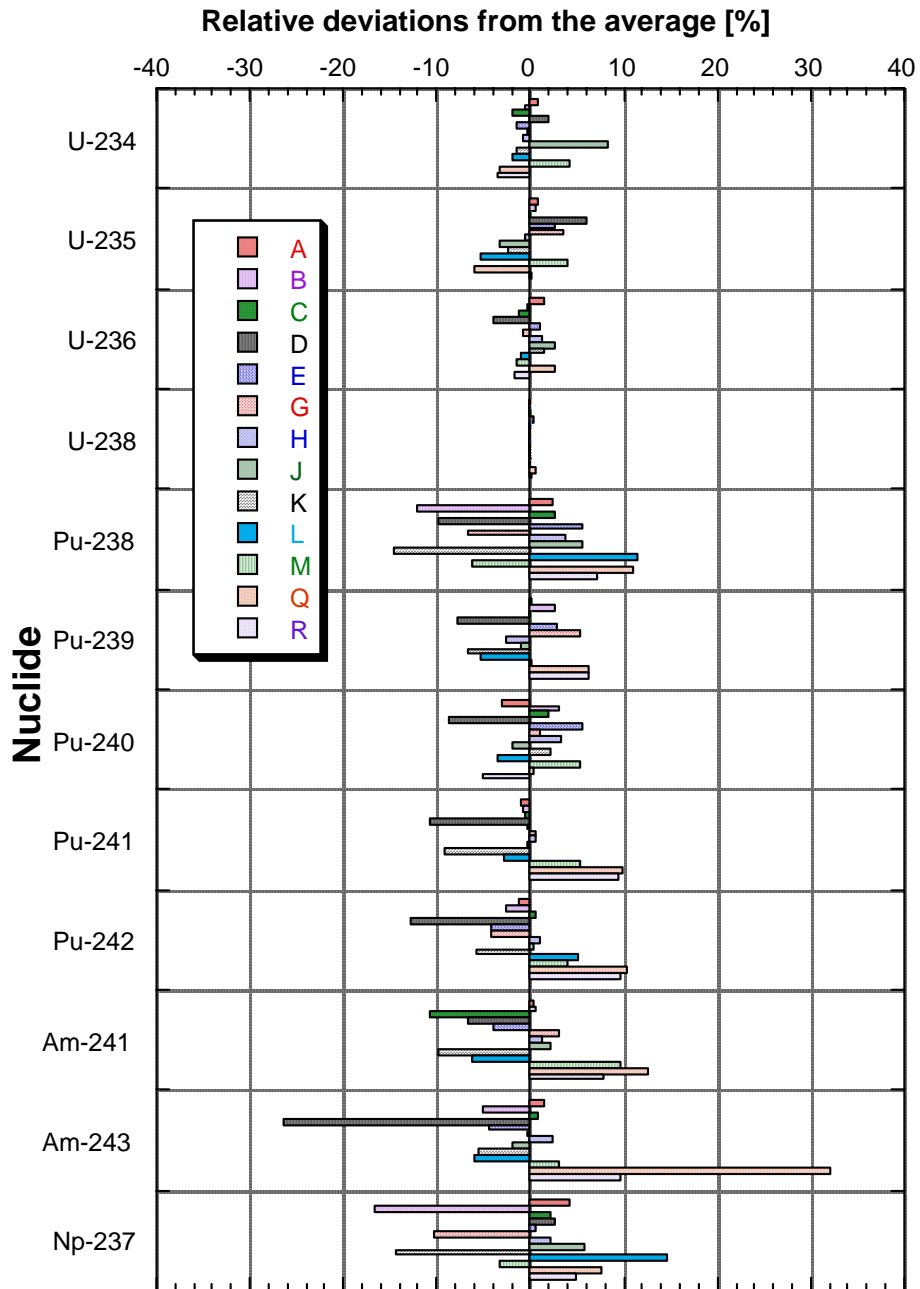
### 3. Status of Burnup Calculation

#### 3.1 Numerical Benchmark

Depletion calculation is one of the most interested problems in reactor physics community. OECD/NEA/NSC has been proposing several depletion calculation benchmarks for plutonium utilization and burnup credit application [13,14,15,16]. In the benchmark calculations, MA shows larger deviation among the participants. **Figure 1** is the example of difference of calculation among benchmark participants of OECD/NEA/NSC/WPNCS burnup credit criticality safety benchmark Phase-IIIB [15]. We can understand that the difference of MA among participants is larger than uranium and plutonium.

Numerical benchmark is useful for peer review of calculation method and libraries. Unfortunately, since current international benchmark is toward to verification of complicated geometrical modeling and fuel design, such benchmark cannot give us clear information on MA generation. If we seek the possibility to check basic data and calculation method of MA, we should propose a simple problem using single pin-cell base problem or back to previous international

benchmarks.



**Figure 1** Relative Difference of atomic number densities of actinides (BWR Burnup calculation: No void, 40 GWd/tHM, no cooling.) [15]

### 3.2 PIE analysis

#### 3.2.1 Japanese PIE Analysis

Analyses of PIE data have been conducted to verify the computer code and libraries in Japan. In the reference [17], systematic comparison between calculation and experimental results for PWR 15×15 UO<sub>2</sub> fuel of 3.2wt% enrichment was carried out by Okumura et. al. The data are taken by JAERI from SF irradiated in Mihama-3. [10] **Table 4** shows the C/E values using MVP-BURN [17],

SWAT[18] and SRAC[19]. This analysis shows that underestimation of MA except for  $^{241}\text{Am}$  and  $^{243}\text{Am}$  irrespective of using libraries. However, it also implies that using continuous-energy monte carlo code can not improve the calculation result of MA. The authors pointed out the possibilities of small production of  $^{238}\text{Pu}$  and  $^{244}\text{Cm}$  due to small capture cross sections of  $^{237}\text{Np}$  or  $^{236}\text{U}$  and  $^{243}\text{Am}$ . For  $^{238}\text{Pu}$ , in spite of its importance, deviation from experimental results has not been solved. Previous studies in 1980's and 1990's show same underestimation of  $^{238}\text{Pu}$ . [20, 21]

Other PIE analysis [12] using JENDL-3.2 is shown in the **Table 5**. The data are taken by JAERI from SF irradiated in Takahama-3. This analysis also shows the underestimation of  $^{238}\text{Pu}$  and  $^{244}\text{Cm}$ . For  $^{237}\text{Np}$ , underestimation seems to be relaxed for samples of BWR-UO<sub>2</sub>. However, since its standard deviation of C/E value is large, we cannot conclude that the  $^{237}\text{Np}$  calculation results are improved definitively. The fact that  $^{238}\text{Pu}$ , products of capture reaction of  $^{237}\text{Np}$ , is underestimated should be considered.

For americium isotopes,  $^{241}\text{Am}$  is overestimated in the analysis. This is different from previous analysis of Mihama-3 by Okumura. Okumura also showed the overestimation in other analysis [22] using SRAC. Considering the analysis results by French code APOLLO2 [23] using JEF-2.2 [24] or JEFF-3.0 [25] shown in next sub-section presents better agreement with PIE data from Takahama-3, further discussion on this over estimation will present us important information regarding the  $^{241}\text{Am}$  generation.

**Table 4** Averaged C/E values of MA Calculation for Mihama-3 [17]

	JENDL-3.2			JEF-2.2	ENDF/B-VI
	MVP	SWAT	SRAC	SRAC	SRAC
$^{237}\text{Np}$	0.90	0.95	0.91	0.92	0.93
$^{238}\text{Pu}$	0.81	0.83	0.81	0.85	0.85
$^{241}\text{Am}$	0.97	0.99	0.97	0.96	0.98
$^{243}\text{Am}$	1.00	0.99	0.98	0.95	1.07
$^{244}\text{Cm}$	0.78	0.76	0.74	0.72	0.82
$^{245}\text{Cm}$	-	-	-	-	-

**Table 5** Averaged C/E values of MA Calculation for Takahama-PWR (SWAT with JENDL-3.2) [12]

	SF95	SF97	SF98
	PWR UO <sub>2</sub>	PWR UO <sub>2</sub>	BWR UO <sub>2</sub>
$^{237}\text{Np}$	-	0.96 (0.02)	1.12 (0.12)
$^{238}\text{Pu}$	0.84 (0.04)*	0.83 (0.02)	0.94 (0.06)
$^{241}\text{Am}$	1.14 (0.27)	1.21 (0.09)	1.17 (0.14)
$^{243}\text{Am}$	0.88 (0.03)	0.89 (0.01)	0.98 (0.04)
$^{244}\text{Cm}$	0.75 (0.05)	0.75 (0.02)	0.83 (0.03)
$^{245}\text{Cm}$	0.81 (0.07)	0.80 (0.03)	0.94 (0.08)

\* Values in ( ) is standard deviation

### 3.2.2 French PIE Analysis

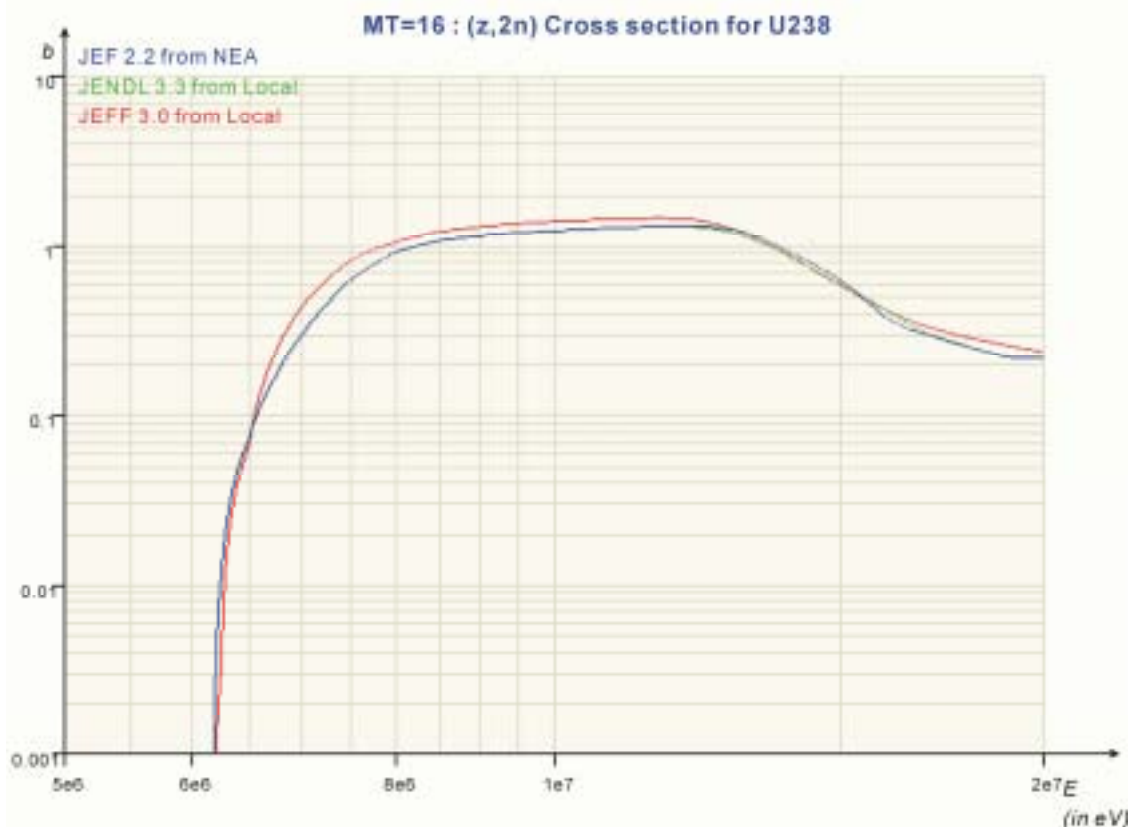
As stated in previous section, France is active in the field of research and development of fuel cycle technology. To validate their code, they have been conducting PIE in France. In the reference [26], instead of using French PIE data, SF97-4 sample of Takahama-3 were used for verification of APOLLO2 and new cross section data library based on JEFF-3.0. **Table 6** shows the results. It reports improvement of  $^{238}\text{Pu}$ ,  $^{237}\text{Np}$  and  $^{243}\text{Am}$ . For  $^{238}\text{Pu}$  and  $^{237}\text{Np}$ , the authors explained that increase of capture resonance integral of  $^{235}\text{U}$  by adopting evaluation by Leal et al [27] and

increase of (n,2n) cross section of  $^{238}\text{U}$  are the main reasons of improvement. **Figure 2** is the comparison of (n,2n) reaction cross section of  $^{238}\text{U}$ . Values of JEFF-3.0 is greater than those of JEF-2.2 and JENDL-3.3 between  $6.5\text{E}6$  to  $1.2\text{E}7$  eV. For  $^{243}\text{Am}$ , in the reference [28], the authors pointed out the capture cross sections of  $^{241}\text{Pu}$  in JENDL-3.2 and ENDF/B-VI are better than that of JEF-2.2, however, still small considering the integral experiments data. Finally, they included new evaluation<sup>3</sup> from thermal to 20 eV in JEFF-3.0 producing higher  $^{241}\text{Pu}$  capture cross section at the 0.26eV resonance, and  $^{243}\text{Am}$  is improved.

At last, we would like to mention on  $^{245}\text{Cm}$ . Both **Table 5** and **Table 6** present the difference is larger than other isotopes. As shown in **Table 5**, standard deviation of calculation results is also larger than  $^{244}\text{Cm}$ . This is same tendency with results in **Table 6**.

**Table 6** C/E-1 [%] of MA Calculation for SF97-4 from Takahama-3 PWR (APLLO2 with JEF-2.2/JEFF-3.0) [26]

	JEF-2.2	JEFF-3.0	total $1\sigma$
$^{237}\text{Np}/^{238}\text{U}$	-8.1	-3.7	0.7
$^{238}\text{Pu}/^{238}\text{U}$	-18.9	-14.1	3.8
$^{241}\text{Am}/^{238}\text{U}$	4.0	4.7	3.2
$^{243}\text{Am}/^{238}\text{U}$	-14.8	-7.7	4.7
$^{244}\text{Cm}/^{238}\text{U}$	-26.5	-19.3	6.4
$^{245}\text{Cm}/^{238}\text{U}$	-33.4	-22.9	7.7



**Figure 2** (n,2n) reaction of  $^{238}\text{U}$

<sup>3</sup> Reference 25 says that this evaluation is submitted to the Nuclear Science and Engineering by H. Derrien.

## 4. Comparison of Calculation Results using Several Evaluated Libraries

### 4.1 Example of Chain Analysis and Comparison among Libraries

The reason of difficulty of improvement of MA calculation is MA is generated through complicated long chain. For example, as shown in section 2,  $^{238}\text{Pu}$  is generated through four paths.

1.  $^{235}\text{U}(n,\gamma)^{236}\text{U}(n,\gamma)^{237}\text{U}(n,\gamma) \Rightarrow \beta^- \Rightarrow ^{237}\text{Np}(n,\gamma)^{238}\text{Np} \Rightarrow \beta^- \Rightarrow ^{238}\text{Pu}$
2.  $^{238}\text{U}(n,2n)^{237}\text{U}(n,\gamma) \Rightarrow \beta^- \Rightarrow ^{237}\text{Np}(n,\gamma)^{238}\text{Np} \Rightarrow \beta^- \Rightarrow ^{238}\text{Pu}$
3.  $^{242}\text{Cm} \Rightarrow \alpha \Rightarrow ^{238}\text{Pu}$
4.  $^{239}\text{Pu}(n,2n)^{238}\text{Pu}$

To determine the priority of evaluation and further discussion, chain analysis by burnup calculation results using several evaluated libraries was conducted. [29] **Table 7** presents the calculation results of MA using several evaluated nuclear data libraries. Authors confirmed most important reaction for  $^{238}\text{Pu}$  generation is (n,2n) of  $^{238}\text{U}$  and the difference of  $^{238}\text{Pu}$  depends on not only (n,2n) cross section, but also fission spectrum.

The reference also includes the discussion of  $^{244}\text{Cm}$  difference in **Table 7**. The authors explained that difference of capture cross section of  $^{242}\text{Pu}$  and  $^{244}\text{Cm}$  cause the difference of  $^{243}\text{Am}$  and  $^{244}\text{Cm}$  results. With the results shown in **Table 6** with APOLLO2, we could make further discussion to improve the cross section in the generation chain of  $^{244}\text{Cm}$ .

Results in **Table 7** show good agreement of  $^{241}\text{Am}$  between JENDL-3.2 and JEF-2.2 calculations, and PIE analysis using JENDL-3.2 shows large difference from experimental data shown in **Table 5**. However,  $^{241}\text{Am}$  results with JEF-2.2 in **Table 6** shows relatively good agreement with experimental results. Concerning this problem, we would like to point out that standard deviation of  $^{241}\text{Am}$  is large as shown **Table 5**. We suppose decay correction adopted to the PIE data from Takahama-3 has effect to some extent. It will be useful if we are able to compare data and calculation without decay correction. In order to determine the reason definitively, we should also back to the difference of calculation method between used codes. This fact implies that using same code system having detail chain treatment is necessary to evaluate different libraries, and collaborative work with experimental group measuring isotopic composition is helpful to do cross check the data and calculation.

**Table 7** Ratios of calculated weights using ENDF/B-VI and JEF-2.2 to those using JENDL-3.2 [29]

	ENDF/B-VI (release 6)	JEF-2.2
$^{237}\text{Np}$	1.03	1.01
$^{238}\text{Pu}$	1.07	1.05
$^{241}\text{Am}$	1.01	0.99
$^{243}\text{Am}$	1.10	0.98
$^{244}\text{Cm}$	1.11	0.97
$^{245}\text{Cm}$	0.95	0.90

### 4.2 Improvement by adopting JENDL-3.3

JENDL Community may interest difference between calculations using JENDL-3.2 and JENDL-3.3. An example of comparison between PIE analysis using JENDL-3.2 and JENDL-3.3 is shown in **Table 8**. The objective data is SF97-4 selected from Takahama-3 PIE, since it was analyzed using APOLLO2.

**Table 8** C/E of MA Calculation for SF97-5 from Takahama-3 PWR  
(SWAT with JENDL-3.2 /JENDL-3.3)

	JENDL-3.2	JENDL-3.3	JENDL-3.3/JENDL-3.2
<sup>237</sup> Np	0.97	1.01	1.04
<sup>238</sup> Pu	0.82	0.86	1.05
<sup>241</sup> Am	1.14	1.10	0.96
<sup>243</sup> Am	0.89	0.90	1.01
<sup>244</sup> Cm	0.75	0.75	1.00
<sup>245</sup> Cm	0.80	0.79	0.99

<sup>237</sup>Np and <sup>238</sup>Pu are improved using JENDL-3.3 approximately 4%. This is because JENDL-3.3 using capture cross section of <sup>235</sup>U evaluation by Leal et al as JEFF-3.0, and the fission spectrum of <sup>235</sup>U became harder than JENDL-3.2. For <sup>241</sup>Am, because of larger capture cross section of JENDL-3.3, decrease in C/E values of several percents is obtained. Smaller capture cross section of <sup>243</sup>Am in JENDL-3.3 than JENDL-3.2 results in small increase of <sup>243</sup>Am. Generally, MA calculation result is improved using JENDL-3.3. However, it is not sufficient. This is same conclusion with the analysis using JEFF-3.0.

## 5. Conclusion

MA calculation would be one of the most interested problem in future program of nuclear energy. However, number of PIE data of MA is not sufficient, especially for high burnup more than 40 GWd/t and we cannot access all data taken in other country. To overcome this situation, we should carry out mutual comparison among calculation results using several libraries to obtain related information.

Comparisons among codes and experimental data had been conducted. Some efforts lead to the improvement of MA calculation. However, underestimation of several MA is not solved definitively. Since MA generates by multiple paths, it is difficult to determine the main reason of the discrepancy. Using the chain analysis and sensitivity analysis, we are able to point out the main contributor to improve the analysis results.

PIE data of UO<sub>2</sub> and MOX for higher burnup UO<sub>2</sub> and MOX (40 to 60 GWd/t) should be obtained utilizing international collaborative work. But it is difficult to carry out it because of several administrative problems. And theoretical work is required to point out the important chain and isotope. To facilitate the efficiency of nuclear data evaluation, we would like to propose following system to be used in quick check of nuclear data.

- Automated system to generate reactor constants for several neutronics codes,
- System to replace selected data (selection of isotope, reaction) in libraries to conduct sensitivity analysis of selected reaction data, and
- Development of database to compare calculation results and PIE data.

Since we are not able to access all data used in other countries, combination of international collaborative work for indirect comparison of codes and library, and efficient information/experience exchange should be pursued continuously for the improvement of MA calculation results.



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