Validation of Pin-Wise Isotope Prediction Methodology by Comparing with Experimental Data

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1. Introduction

For a safe transportation and storage of spent nuclear it is required to evaluate criticality conservatively. However, burn-up credit is applied to cask design in order to remove unnecessarily high conservative assumptions. It is necessary to predict isotopic composition in a spent fuel for license evaluation for spent fuel cask design. Up to now, isotopic composition is predicted in an assembly-wise database. More detail evaluation of isotopic composition by pin-wise prediction may decrease uncertainty a lot for many applications; criticality evaluation, spent fuel recycling, cask shielding design, spent fuel repository design, etc. Accuracy of pin-wise isotopic concentration may play a critical role in adopting burn-up credit evaluation.

Therefore, checking up pin-wise isotope prediction methodology was performed through comparison with measurement data of OECD/NEA in this study. While objective error is under 5% for the major isotopes, more large errors are expected for some isotopes which has small amount and negligible impact to criticality.

2. Analysis Method

2.1 Pin-Wise Isotope Prediction Methodology

Pin-wise isotope prediction methodology is shown in Fig. 1 [1]. Pin-wise isotope prediction is conducted through three steps. Step 1 is to make Burnup-Isotope Number Density Data Base. In this step, data base is produced according to the type of fuel and fuel pin group(location). Step 2 is pin-wise burnup information. Step 3 is to predict isotope number density for each pin by interpolating.

Evaluation of two stages is needed for pin-wise isotope prediction methodology validation. First stage is to validate Burnup-Isotope Number Density Data Base. In this stage, reliability of data base produced by assembly calculation is validated. In second stage, prediction of pin burnup is evaluated. Pin-wise burnup prediction is performed through DeCART2D/MASTER. In this paper, validation of Burnup-Isotope Number Density Data Base is mentioned.

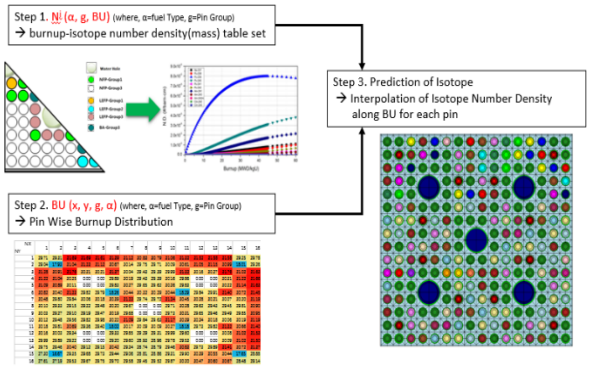

Fig. 1. Pin-Wise Isotope Prediction Methodology Scheme

2.2 OCED/NEA Measurement Data

Burnup-credit was applied for efficient spent fuel management [2,3]. code validation for adopting burn-up credit was performed by comparing calculated value with experimental data as licensing strategy. These measurement data were collected due to management of isotopic composition in spent fuel.

In this study, benchmark of three reactors is performed. These reactor measurement data are used to validate pin-wise isotope prediction methodology. Measurement data are listed in Table I. three reactor is selected because of three reasons. First reason is sufficient fuel assembly information. Second one is a variety of isotope data. Last one is reliability of measurement data. Because each experiment is done by using different analytical way for spent fuel assay data, uncertainty is different. Therefore, experimental data with lower uncertainty were chosen.

Table I: OECD/NEA Measurement Data List

Reactor	Assembly	Pin	Sample Name	Burnup (GWd/tU)
Takahama Unit 3	NT3G23	SF95	2	24.35
			3	35.42
			4	36.69
			5	30.4
		SF96	2	16.44
			3	28.2
			4	28.91
			5	24.19
	NT3G24	SF97	2	30.73
			3	42.16
			4	47.03
			5	47.25
			6	40.79

Calvert Cliffs Unit 1	D047	MKP 109	CC	37.12
			LL	27.35
			P	44.34
	D101	MLA 098	BB	26.62
			JJ	18.68
			P	33.17
Ohi Unit 1&2	G13	N13	91E07	52.434
	17G	C5	89G01	21.465
			89G03	28.717
		F4	89G08	30.172
			89G10	38.496
		O13	89G05	25.137

2.3 Evaluation Procedure

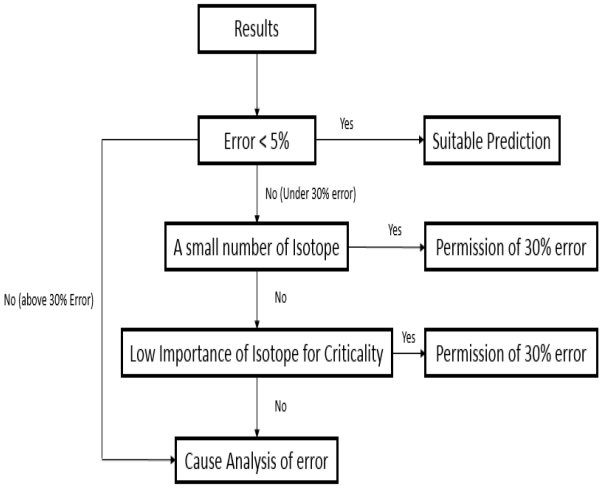

Fig. 2. Comparative analysis scheme

Fig. 2 shows comparative analysis scheme. Isotopic concentration [mg/gUi, milligram/gram Uranium initial] is selected as parameter and C/E ratio is used for comparison with experimental data.

$$C/E\ Ratio = \frac{Calculated\ Prediction}{Experimental\ Data} \quad (1)$$

Isotope under 5% error means to predict suitably. some isotopes are permitted up to 30% error through 2 step. In first step, number density in spent fuel is analyzed. A small number of isotope can be allowed up to 30% error. Unsatisfied isotope goes to second step. Second step is evaluation of importance for criticality. Although isotope concentration is large, impact for critical can be small because of low cross-section. Therefore, importance is evaluated through macroscopic XS like under equation. Isotope with low importance can be permitted up to 30% error.

$$\begin{pmatrix} Fissile = \frac{\Sigma_{f,Nj}^{Asy}}{\Sigma_{a}^{Asy}} \\ Absorber = \frac{\Sigma_{a,Nj}^{Asy}}{\Sigma_{a}^{Asy}} \end{pmatrix} \quad (2)$$

3. Results

3.1 Takahama Unit 3

Results of takahama unit 3 is shown in Fig. 3 and Fig. 4. Isotope distribution along error band is listed in Table II. Error under 5% means to predict suitably. Evaluation for number density and importance is conducted for isotope above 5% error.

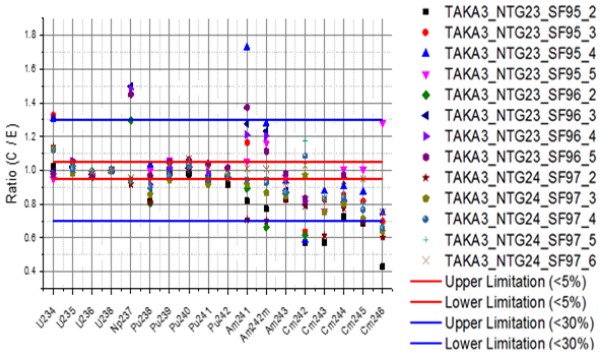

Fig. 3. C/E Ratio – Takahama Unit 3 (Actinide)

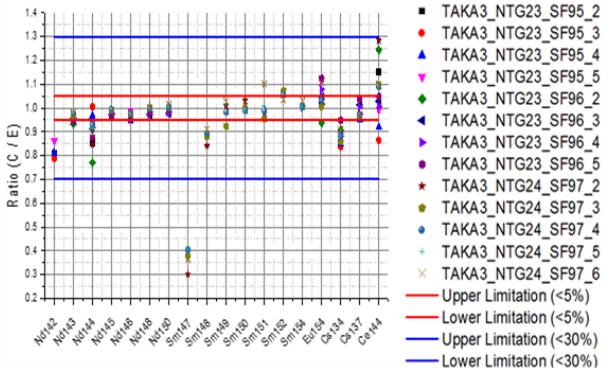

Fig. 4. C/E Riatio – Takahama Unit 3 (Fission Product)

Table II: Isotope List according to Relative Error

Relative Error	Isotope
Under 5%	^{235}U, ^{236}U, ^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{143}Nd, ^{145}Nd, ^{146}Nd, ^{148}Nd, ^{150}Nd, ^{149}Sm, ^{150}Sm, ^{151}Sm, ^{152}Sm, ^{154}Sm, ^{137}Cs
From 5% to 30%	^{234}U, ^{238}Pu, ^{241}Am, ^{242m}Am, ^{243}Am, ^{244}Cm, ^{245}Cm, ^{142}Nd, ^{144}Nd, ^{148}Sm, ^{154}Eu, ^{134}Cs, ^{144}Ce
Above 30%	^{237}Nd, ^{242}Cm, ^{243}Cm, ^{147}Sm, ^{106}Ru, ^{125}Sb

Evaluation of number density was performed on 50GWd/tU burnup. Number density is shown in Table III. Except for ^{144}Nd and ^{144}Ce, isotopes have small number density. Therefore, these isotopes are accepted up to 30% error. However, there are much number density for ^{144}Nd and ^{144}Ce, so importance evaluation has to be done. Table IV shows importance for criticality.

while ^{144}Nd and ^{144}Ce have a large number of number density, importance is low because of low XS. Therefore, 30% error is permitted for ^{144}Nd and ^{144}Ce.

Table III: Fuel Pin Number Density – 50GWd/tU

	Isotope	Number Density		Isotope	Number Density
1	U238	2.1430E-02	21	Pu238	7.4531E-06
2	U235	1.8755E-04	22	Sm152	5.0241E-06
3	Pu239	1.4265E-04	23	U234	4.8103E-06
4	U236	1.3013E-04	24	Am243	4.2682E-06
5	Cs137	7.0714E-05	25	Sm147	3.8790E-06
6	Pu240	6.5680E-05	26	Cm244	1.9880E-06
7	Nd144	5.8388E-05	27	Sm154	1.9858E-06
8	Pu241	4.1129E-05	28	Eu154	1.5008E-06
9	Nd143	4.0688E-05	29	Nd142	1.3353E-06
10	Nd146	3.8602E-05	30	Am241	1.2836E-06
11	Nd145	3.5367E-05	31	Sb125	8.1856E-07
12	Nd148	2.0039E-05	32	Sm151	5.7244E-07
13	Pu242	1.9741E-05	33	Cm242	5.6097E-07
14	Np237	1.5468E-05	34	Sm149	1.4852E-07
15	Ce144	1.5419E-05	35	Cm245	1.3374E-07
16	Sm150	1.5346E-05	36	Am242m	2.9900E-08
17	Ru106	1.1559E-05	37	Cm243	1.9222E-08
18	Nd150	9.6805E-06	38	Cm246	1.4228E-08
19	Sm148	8.3964E-06			
20	Cs134	8.3225E-06			

Table IV: Importance for Criticality – 50GWd/tU

	Isotope	Importance		Isotope	Importance
1	U238	2.6333E-01	21	U234	1.0486E-03
2	Pu239	1.5645E-01	22	Cm244	4.5129E-04
3	U235	8.1254E-02	23	Nd144	2.7590E-04
4	Pu240	7.6283E-02	24	Cm245	2.3868E-04
5	Pu241	5.0495E-02	25	Am242m	2.2667E-04
6	Nd143	1.1440E-02	26	Nd148	1.4737E-04
7	U236	9.6123E-03	27	Sm148	1.3824E-04
8	Pu242	6.6572E-03	28	Nd146	9.6285E-05
9	Np237	6.2807E-03	29	Nd150	6.5580E-05
10	Sm149	6.2133E-03	30	Cm242	4.5848E-05
11	Sm152	5.1523E-03	31	Sm154	3.8387E-05
12	Sm151	4.3474E-03	32	Cs137	2.6230E-05
13	Eu154	3.8636E-03	33	Ce144	2.4606E-05
14	Nd145	3.6388E-03	34	Nd142	2.3695E-05
15	Am243	3.0888E-03	35	Sb125	2.1928E-05
16	Pu238	2.2487E-03	36	Cm243	2.1789E-05
17	Sm150	2.1295E-03	37	Ru106	1.2698E-05
18	Am241	1.6682E-03	38	Cm246	7.9081E-07
19	Cs134	1.2551E-03			
20	Sm147	1.1605E-03			

3.2 Calvert Cliffs Unit 1 and Ohi Unit 1&2

Calvert Cliffs Unit 1 and Ohi Unit 1&2 also apply same method for evaluation. Results of Calvert Cliffs and Ohi is shown in Fig. 5 and Fig. 6. ^{237}Nd, ^{242}Cm and ^{243}Cm is matched well in Calvert Cliffs and Ohi reactor while error is large in Takahama reactor.

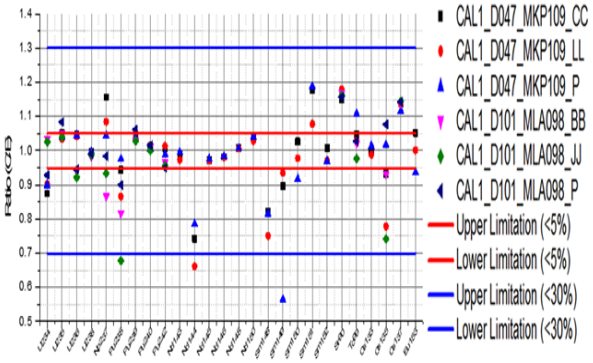

Fig. 5. C/E Ratio – Calvert Cliffs Unit 1

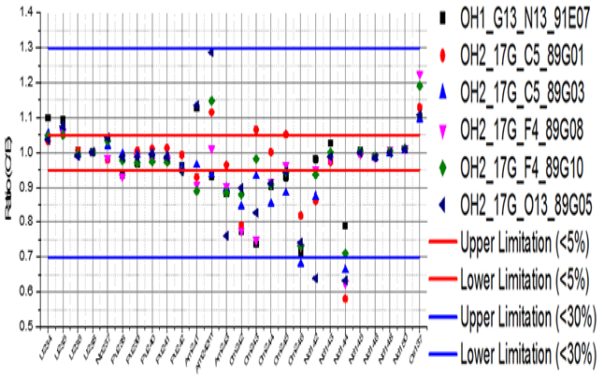

Fig. 6. C/E Ratio – Ohi Unit 1&2

However, there are overestimated isotopes. These isotopes are listed in Table V. Overestimated isotopes have short half-life. While calculated value is results when fuel assembly is discharged, measurement data is value after sufficient cooling was conducted. Because of cooling time, radioactive decay happens. For this reason, some isotopes are overestimated. Half-life is shown in Table VI.

Table V . Over Estimated Isotope

Calvert Cliffs Unit 1						
Isotope	MKP 109_CC	MKP 109_LL	MKP 109_P	MLA 098_BB	MLA 098_JJ	MLA 098_P
Pu241	1.347	1.372	1.323	1.391	1.380	1.413
Eu155	2.618	2.607	2.422	--	--	--
Ohi Unit 1&2						
Isotope	N13_ 91E07	C5_ 89G01	C5_ 89G03	F4_ 89G10	F4_ 89G08	O13_ 89G05
Eu154	1.727	1.545	1.635	1.887	1.894	1.604
Sb125	8.020	7.497	7.100	8.758	8.876	7.600
Cs134	5.419	5.128	4.864	5.485	5.280	4.897
Ru106	36.036	31.488	28.657	34.144	31.255	28.589
Ce144	109.996	88.012	86.157	94.216	81.858	81.806

Table VI. Half Life of Isotope

	Isotope	Half-life (yr)		Isotope	Half-life (yr)
1	Am242m	0.001829	12	Cs137	30.08
2	Cm242	0.446027	13	Pu238	87.7
3	Ce144	0.780575	14	Am241	432.6
4	Ru106	0.870685	15	Cm246	4706
5	Cs134	2.0652	16	Pu240	6561
6	Sb125	2.7586	17	Am243	7370
7	Eu155	4.753	18	Cm245	8423
8	Eu154	8.601	19	Pu239	24110
9	Pu241	14.29	20	Pu242	375000
10	Cm244	18.01	21	Np237	2144000
11	Cm243	29.1	22	U236	23420000

4. Conclusions

In this study, validation of Burnup-Isotope Number Density Data Base was performed by comparing with measurement data. relative error is 5% for major isotopes and 30% for a small number of isotope which is not important for criticality. Also, cooling time must be considered when isotope with short half-life is predicted.

In this study, assembly average macroscopic XS is used when importance is evaluated. For detail information, calculation using ORIGEN or MCNP will be conducted. Also, validation of burnup prediction calculated by DeCART2D/MASTER is supposed to be performed.

Acknowledgement

This work was supported by KOREA HYDRO & NUCLEAR POWER CO., LTD (No. 2017-Tech-12)

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