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VALIDATION OF MINOR ACTINIDE CROSS SECTIONS BY STUDYING SAMPLES IRRADIATED FOR 492 DAYS AT THE DOUNREAY PROTOTYPE FAST REACTOR

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ABSTRACT

To evaluate neutron cross-section data of minor actinides, separated actinide samples and dosimetry samples were irradiated at the Dounreay Prototype Fast Reactor for 492 effective full power days. Irradiated samples were analyzed both at the Oak Ridge National Laboratory and at the Japan Atomic Energy Research Institute. The burnup calculations were carried out for minor actinides using the determined flux distribution and flux level. The calculated results were compared with the experimental data. This paper discusses the burnup calculations and the validation of minor actinide cross-section data in evaluated nuclear data libraries.

1. INTRODUCTION

The Japan Atomic Energy Research Institute (JAERI) has been developing technologies for partitioning and transmutation of long-lived nuclides in high-level radioactive liquid waste since the mid-1970's. These activities cover the development of an aqueous partitioning process, the design study of a dedicated transmutation system, the development of a high-power proton accelerator, the nitride fuel-cycle technologies, and the basic research on nuclear and fuel properties [1]. JAERI has developed the concept of double-strata fuel cycle, in which partitioning and transmutation is carried out in a dedicated and small-scale fuel cycle attached to the commercial fuel cycle [2]. The system study of transmutation has made it clear that the dedicated system could more effectively transmute minor actinides (MAs) than fast reactors for power generation. Two types of dedicated transmutation systems have been designed; an actinide burner fast reactor (ABR) [3] and an accelerator-driven subcritical system (ADS) [4]. In both systems, the major nuclear fuel material is minor actinide nitrides. Therefore, in these dedicated systems, reliable neutron data of MA are required to obtain a sound design.

The current status of MA nuclear data is not so satisfactory and, for some nuclides, measured data are nonexistent. For a transmutation study, significant efforts are needed not only to measure nuclear data of MAs but also to evaluate the reliability of existing data by using integral measurements. As such an effort, the radiochemical analysis data of the actinide samples

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irradiated at the Dounreay Prototype Fast Reactor (PFR) was used. The irradiation experiments were conducted under a joint research program between the United States (US) and the United Kingdom (UK). The samples were milligram quantities of actinide oxides of 18 different isotopes from uranium to curium. These oxide powders were encapsulated in vanadium holders and irradiated in the core of PFR for 492 effective full-power days (EFPD). Parts of sample solutions were transported to JAERI from the Oak Ridge National Laboratory (ORNL). In this work, the burnup calculations are performed and the calculated data are compared with the analytical data to evaluate the reliability of neutron cross sections of MAs in evaluated nuclear data files.

2. IRRADIATION OF THE ACTINIDE SAMPLES

We describe briefly the experimental information needed for the calculational analysis. The actinide samples [5] were prepared by ORNL and made into a fuel pin [6] by the Hanford Engineering Development Laboratory. The samples and dosimeters used in the experiment were encapsulated in a high-purity material that would not produce an undesirable background after irradiation. Capsules made from high-purity vanadium were used for this purpose. These capsules were then placed in four fuel pins. In this paper, we will discuss only the results for FP-4 (35 capsules consisting 9 dosimeter samples and 26 actinide samples). The pins were placed in two specially built cluster of 19 pins. The other pins in these clusters were standard fuel pins. In turn, six of these 19-pin clusters formed a demountable subassembly (DMSA). The location of the DMSAs in the core are shown in Figure 1. The cluster containing FP-4 was loaded at location 0706, 0702 and 0706. At final discharge, the cumulative total irradiation for FP-4 amounted to 492 EFPD. The total exposure and the total fluence were 295,483 MWd and $\sim 2\times 10^{23}$ n/cm^2, respectively.

3. RADIOCHEMICAL ANALYSIS

The irradiated samples were originally dissolved and partitioned at ORNL and dried before transportation from ORNL to JAERI. These samples were analyzed independently at JAERI by chemical methods. The detailed compositions of the irradiated samples were determined by alpha measurements and gamma-ray measurements, and mass spectrometry. The main results obtained in this work are expressed in terms of the number of initial metallic atoms (IMA). The initial amounts of different actinides before irradiation can be obtained from the weight, concentration, composition, and average mass (based on the isotopic abundances) of each sample. Because of regulatory and transportation restrictions, the portions of the irradiated samples that were sent from ORNL to JAERI were kept deliberately small. Relying on the measured weights and liquid fractions for subsequent quantitative analyses would have introduced considerable uncertainties in the measured and reported data. Therefore, we adopt the following procedure based on a standard test method for atom percent fission in nuclear fuel (neodymium-148 method) which exploits the determination of stable fission product ^{148}Nd in irradiated fuel as a measure of fission [7]. The application of this method does require a knowledge of IMA before irradiation but not the measured weights and liquid fractions after irradiation. From the measured yields of the fission products, we can estimate the burnup of the samples. Burnup values of FIMA% (percentage of fission per initial metallic atoms) for the actinide specimens were obtained from the measured numbers of ^{148}Nd atoms, A, effective fission yields of ^{148}Nd, FY$_{eff}$, and the measured numbers of the actinide atoms, B, using

$$FIMA\% = \frac{A/FY_{eff}}{A/FY_{eff} + B} \times 100 \quad (1)$$

Effective fission yield of ^{148}Nd for each sample was calculated using the contribution of individual nuclides to the total fission rate and the fission yield from each contributor. The fission yield of ^{148}Nd for each nuclide was obtained from Ref. 8. For an isotope lacking a recommended value in Ref. 8, we assumed a value based on the values for nearby nuclides.

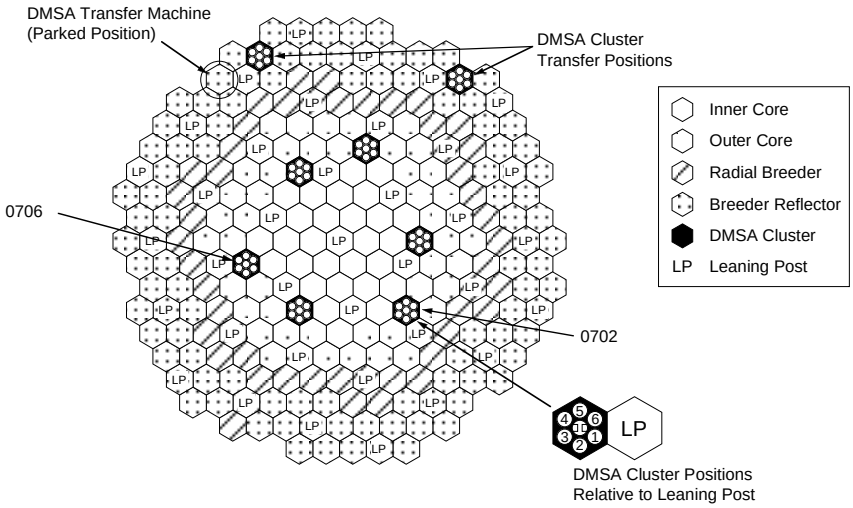

Figure 1. Core showing DMSA experimental positions

One way to assess the overall reliability of the data is to compare the ORNL and JAERI values for the amount of primary actinide present in each post-irradiated sample. This is done in Table I. Recall that the ORNL results are based on direct weight measurements while the JAERI results (especially the determination of the post-irradiation IMAs) are based on the ^{148}Nd method. The ORNL measurements were made using the best laboratory practices possible under the difficult constraints. Systematic errors were introduced by many conditions that could not be easily controlled such as target loading and encapsulation, contamination in hot cells, incomplete dissolution, occasional splattering, and aliquoting. Because of these problems, the ORNL analytical results were reported in Ref.9 without error bars. The uncertainties in the JAERI measured data became more traceable. On the negative side, we now had to contend with the uncertainties in the cumulative fission yields of ^{148}Nd for various actinides. The two sets of data are consistent with each other at the level of about 7%. This level of agreement persists concerning all actinides because the isotopic results obtained at ORNL and JAERI are virtually the same for all analyzed samples.

4. BURNUP CALCULATIONS

4.1. POWER HISTORY

The power history of the PFR is available in very fine details, i.e., the measurements of power levels are about 1-hour interval during times of significant power change. However, it was not practical to follow the details of the variations in power level in simulating the exposure of

Table I. Comparison between the weights of principal isotopes measured at ORNL and JAERI

Sample	Before irradiation Weight (mg) W(S)	After irradiation Weight(mg) ORNL W(O)[a]	After irradiation Weight(mg) JAERI W(J)	Ratio W(J) / Average of W(J) and W(O)
^{233}U	7.65	—[b]	3.86 ± 0.02	
^{234}U	3.50	3.33	2.83 ± 0.01	0.92 ± 0.01
^{235}U	8.37	4.96	4.81 ± 0.02	0.98 ± 0.01
^{236}U	7.96	7.23	7.27 ± 0.04	1.00 ± 0.01
^{238}U	9.81	8.40	9.08 ± 0.03	1.04 ± 0.01
^{237}Np	12.22	8.59	8.11 ± 0.24	0.97 ± 0.03
^{238}Pu	3.11	—[c]	2.18 ± 0.01	
^{239}Pu	8.47	4.90	5.02 ± 0.02	1.01 ± 0.01
^{240}Pu	10.84	10.26	8.84 ± 0.02	0.93 ± 0.01
^{241}Pu	4.25	1.13	1.14 ± 0.01	1.00 ± 0.01
^{242}Pu	2.05	1.94	1.78 ± 0.01	0.96 ± 0.01
^{241}Am	9.42	6.21	5.74 ± 0.06	0.96 ± 0.01
^{243}Am	9.83	6.79	6.45 ± 0.07	0.97 ± 0.01
^{243}Cm	0.39	0.22	0.16 ± 0.01	0.84 ± 0.01
^{244}Cm	8.25	4.48	5.53 ± 0.05	1.10 ± 0.01
^{246}Cm	6.69	4.92	5.81 ± 0.06	1.08 ± 0.01
^{248}Cm	1.76	1.45	1.58 ± 0.02	1.04 ± 0.01
				Average = 0.98 ± 0.07

[a] Taking into account the estimates made for losses in the ^{236}U, ^{238}U, ^{239}Pu, and ^{242}Pu samples (see Ref.9 for details.)

[b] Portion lost in dissolution.

[c] Severe sample loss in suspected.

the actinide samples. Rather, by taking account of periods of time when the variations were not great, it was more practical to simulate the reactor performance with time-averaged power levels for each of these periods. In such a calculation, how to divide the time periods and associate the averaged reactor power was important. The possible uncertainties resulting from such simulations have been discussed in Ref. 10. From the preliminary calculational results for fission products using the four different levels of detail for time steps, it was concluded in Ref. 10 that the ORNL simulation using 33 individual time steps was appropriate.The power history of these 33 time bins is shown in Figure 2. In this way, the exposure of the samples could be simulated by a succession of constant power level episodes whose time integral was equal to that of the actual power history.

4.2. ESTIMATION OF ONE-GROUP CROSS SECTIONS

We used the ORIGEN code [11] for the calculational analysis of the irradiated samples. The nuclear data were supplied by the JENDL-3.2 library [12]. The determination of the flux spectrum is an important first step in the production of the one-group cross-section library for the burnup calculation. For the detailed evaluation of the neutron flux spectrum, extensive information about reactor core (arrangement of subassembly, compositions of fuel pins, control rod positions, etc.) is required. Because of lacking this information, the neutron spectra and the flux values in the documents [10] published by ORNL were used for the calculational analysis.

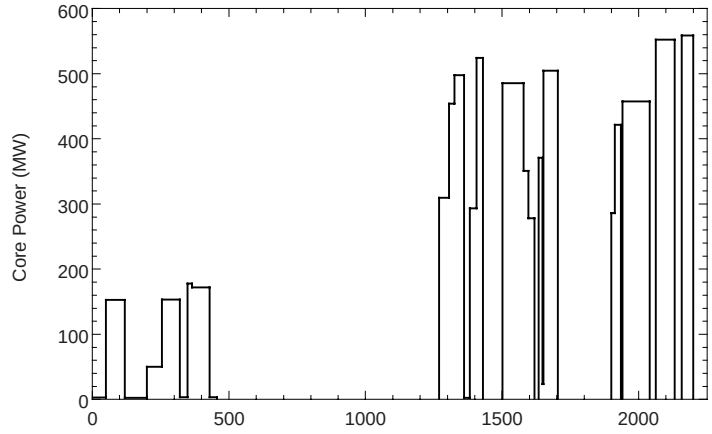

Figure 2 The power history simulation used in the calculations. Days are counted from July 1, 1982.

Basically, the infinite dilution cross sections were used to estimate one-group cross sections for most nuclides. However, for some nuclides (^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu, and ^{241}Pu), we must consider the self-shielding effect because there is a large amount of fuel material containing these nuclides which surround the irradiated samples. To estimate the effective cross sections for these nuclides, we used the cell calculation code SLAROM [13] with the revised JAERI Fast Reactor Group Constant Set (JFS-3) [14]. For this purpose, detailed information about the standard fuel pins surrounding FP-4 is needed. Again, because of the lack of such information, we relied on the specification of the standard PFR fuel pin from Refs. 15 and 16. On the basis of the same reports, we also made a guess of the fuel composition. The influence of these effective values on the one-group cross sections will be discussed later.

The UK provided real flux data for the actual runs. These flux calculations were in six groups using three-dimensional full core plan with six triangles per subassembly and 17 axial mesh points of which 10 points were in the core height. The neutron spectra and the normalized fluxes for all axial positions in each run are listed in Ref. 10. The total flux was adjusted in such a manner that the integral reactor power was 600 MW total. The standard axial mesh points did not represent the actual heights of specific samples. The interpolations of the neutron spectrum and flux were performed for each run in which FP-4 was present. Dounreay provided a 37-group spectrum only for the outer-core center plane. We used this spectrum as a starting point in the calculation of the one-group cross sections to be used in the sample burnup calculations. This 37-group spectrum was subdivided to 73 groups because the revised JFS-3 has 73 groups. The upper energy limit was also extended from 10 MeV to 20 MeV. The subdivision was accomplished by assuming a 1/E spectrum shape within each group. The 73-group library was then collapsed to a 6-group library corresponding to the 6 groups described above. The one-group cross sections were calculated for each PFR run using this 6-group library with the 6-group flux spectra for the various reactor runs.

4.3. NEUTRON FLUX LEVEL

Before making detailed calculations, preliminary burnup calculations were performed for the verification of neutron flux level reported in Ref. 10. Based on the burnup calculations of major

actinide (^{235}U and ^{239}Pu) and dosimetry (^{235}U+^{238}U and ^{237}Np+^{239}Pu) samples, the neutron flux levels were determined at the locations where the minor actinide samples were irradiated. The cross sections and the fission yields of ^{148}Nd for these nuclides are well known. The results of preliminary calculations for the FIMA values are shown in Table II. The ratio of calculated-to-experimental (C/E) values for the three ^{237}Np+^{239}Pu dosimeters are higher than the values for the remaining four samples. The differences can be traced to an overestimation of FIMA for ^{237}Np in the calculations (see later discussion). We averaged the four C/E values (for ^{235}U, ^{239}Pu, and two ^{235}U+^{238}U dosimeter samples) to obtain C/E= 1.058; that is, the actual neutron flux is about 6% lower than that reported in Ref. 10. The burnup calculations were made after dividing the fluxes given in Ref. 10 by 1.06.

Table II. Results of preliminary burnup calculations for FIMA

Sample	FIMA(%)	C/E
^{235}U	32.2 ± 0.02	1.05
^{239}Pu	30.5 ± 0.02	1.07
Dosimetry		
^{235}U + ^{238}U	15.3 ± 0.02	1.06
^{235}U + ^{238}U	18.0 ± 0.02	1.05
^{237}Np + ^{239}Pu	18.6 ± 0.04	1.09
^{237}Np + ^{239}Pu	20.1 ± 0.05	1.08
^{237}Np + ^{239}Pu	19.4 ± 0.05	1.08

5. COMPARISON WITH EXPERIMENT

In Table III, we present the experimental FIMA values together with the C/E values for 17 actinide samples and 5 dosimetry samples. The uncertainties in the effective fission yields are mainly responsible for the uncertainties in the experimental FIMA values. The C/E values for the burned amount of each principal actinide is often a more demanding indicator of the agreement between theory and experiment than the C/E value for the final amount of the actinide itself. Therefore, in addition to the FIMA results, we present the results concerning the changes in the composition of principal nuclide in each actinide sample in Table III. These changes are expressed in terms of the loss of material divided by IMA. The uncertainties in the %change are the uncertainties in the measured values. In Figure 3, the C/E values for FIMA, for %change of the primary nuclide with mass number A, and for the %change of the A+1 nuclide are presented for each sample.

In Table I, we showed that the ORNL and JAERI results for the weights of the principal actinides agree to within ±7%. This level of agreement is also what we would expect and given by the difficulties of the experiment and the proven performance of the methods employed. At present, there are three major nuclear data libraries (JENDL-3.2, ENDF/B-VI, and JEF-2.2) containing MA data and several less extensive evaluations [17]. There are considerable differences among these libraries concerning the cross-section values recommended for reactions of interest in this work. For the purpose of validating the JENDL-3.2 file, the calculated values were considered exactly. We then set the acceptable range of the C/E values at ±7%(1σ). We show this range and the 2σ limits in Figure 3.

Table III Results of calculations for total fission per initial metallic atom (FIMA in %) and %change in primary isotopes in the samples

Sample	FIMA (%)		%change	
	Experiment	C/E	Experiment	C/E
^{233}U	45.2 ± 0.6	0.95	-49.5 ± 0.5	0.95
^{234}U	8.63 ± 1.38	1.00	-19.2 ± 0.5	0.91
^{235}U	32.2 ± 0.2	1.00	-42.4 ± 0.3	0.97
^{236}U	5.44 ± 0.43	1.05	-7.56 ± 0.40	1.06
^{238}U	1.99 ± 0.01	0.93	-7.41 ± 0.30	0.89
^{237}Np	8.07 ± 0.69	1.16	-33.6 ± 2.0	0.98
^{238}Pu	16.3 ± 3.3	1.28	-39.5 ± 1.3	1.11
^{239}Pu	30.5 ± 0.2	1.02	-40.4 ± 2.4	0.98
^{240}Pu	9.61 ± 0.15	1.07	-18.4 ± 0.2	1.06
^{241}Pu	25.1 ± 0.7	1.09	-81.1 ± 0.7	1.00
^{242}Pu	5.70 ± 0.10	1.03	-13.1 ± 0.3	1.14
^{241}Am	8.99 ± 1.44	1.09	-39.4 ± 0.9	0.90
^{243}Am	5.12 ± 0.82	1.04	-34.4 ± 0.8	0.93
^{243}Cm	26.2 ± 3.4	1.13	-37.8 ± 1.2	0.95
^{244}Cm	8.75 ± 1.58	1.26	-52.3 ± 0.9	0.98
^{246}Cm	6.81 ± 1.02	1.17	-8.70 ± 0.61	0.92
^{248}Cm	5.68 ± 1.08	1.28	-9.41 ± 0.81	1.05
Dosimetry				
^{235}U + ^{238}U	15.3 ± 0.2	1.01		
^{235}U + ^{238}U	18.0 ± 0.2	1.01		
^{237}Np + ^{239}Pu	18.6 ± 0.4	1.04		
^{237}Np + ^{239}Pu	20.1 ± 0.5	1.03		
^{237}Np + ^{239}Pu	19.4 ± 0.5	1.03		

5.1. URANIUM SAMPLES

The C/E values for FIMA [see Figure 3(a)] and for %change in primary nuclides [see Figure 3(b)] in the uranium samples are reasonably close to unity within the acceptable range for all isotopes except for ^{238}U. Figure 3(c) shows that the calculation underestimates the buildup of nuclides in the uranium samples. There is thus the possibility of underestimation of capture cross sections for uranium isotopes in the JENDL-3.2 library. Calculation of the amount of ^{238}U remaining in the ^{238}U sample after irradiation is strongly dependent on the capture cross section. In our calculations, we used the effective cross section. The difference between this lower value and the higher infinite dilution cross section is about 10% for ^{238}U. These differences are much smaller (1-2%) for the other nuclides. Definite conclusions concerning the influence of self shielding in the ^{238}U sample (which affects the effective cross section) requires such information that is not currently available.

5.2. ^{237}Np AND PLUTONIUM SAMPLES

The C/E values for ^{237}Np shown in Figures 3(b) and 3(c) fall within reasonable bounds. The FIMA C/E [see Figure 3(a)] is outside but carries a large uncertainty. For ^{238}Pu, the calculation

overestimates the burnup of ^{238}Pu in the ^{238}Pu sample and underestimates the buildup of ^{238}Pu in the ^{237}Np sample. From these facts, we conclude that the fission cross section of ^{238}Pu is probably overestimated in the JENDL-3.2 library. In a transmutation system, ^{238}Pu is an important nuclide because relatively large amounts of ^{238}Pu are produced through neutron capture by ^{237}Np and ^{241}Am (and subsequent alpha decay of ^{242g}Am). These latter nuclides are main nuclides in MAs, and the fission cross section of ^{238}Pu in a fast spectrum is relatively high. Therefore, a reevaluation of nuclear data for ^{238}Pu is desirable. There is also the possibility of overestimation in the fission yield of ^{148}Nd for ^{238}Pu from the fact that the C/E for FIMA is high [see Figure 3(a)]. The contribution of ^{238}Pu fission in the ^{237}Np sample is about 35%, so this is one of the reason for the overestimation of FIMA in the ^{237}Np sample. For ^{242}Pu, the calculation overestimates the burnup of ^{242}Pu [see Figure 3(b)] and the buildup of ^{243}Am [see Figure 3(c)] which is a decay product of ^{243}Pu produced by ^{242}Pu+neutron. These results indicate possible overestimation of the capture cross section of ^{242}Pu. The C/E values are reasonable for all other plutonium isotopes, except for neutron capture by ^{239}Pu leading to ^{240}Pu [see Figure 3(c)]. The reason for the overestimation of the buildup of ^{242m}Am in the ^{241}Pu sample (C/E=1.17) is discussed later.

5.3. AMERICIUM AND CURIUM SAMPLES

The FIMA and %change C/E values for ^{241}Am and ^{243}Am are reasonably close to unity. The C/E values for the buildup of ^{238}Pu, ^{242}Pu, and ^{242m}Am through neutron capture by ^{241}Am in the ^{241}Am sample lie outside the acceptable range. The branching ratio of the ^{241}Am(n,γ)^{242g}Am and ^{241}Am(n,γ)^{242m}Am reactions is important in burnup calculations, and this ratio depends on the incident neutron energy. The existing data for this branching ratio are sparse [17]. In the case of the ^{241}Am and ^{241}Pu samples, in which the reactions involving ^{241}Am are dominant, we studied the dependence of the %change C/E values on the isomer ratio defined as the ratio of the ^{241}Am(n,γ)^{242g}Am cross section to the total capture cross section. Unacceptable C/E values are obtained for ^{242m}Am in both samples when an isomer ratio less than 0.7 is used in the calculations. Therefore, we prefer a higher value; a value of 0.8 was used in our calculations. No definitive conclusions concerning ^{241}Am can be drawn at this time by pending a better knowledge of the isomer ratio as a function of neutron energy.

The C/E of FIMA for curium samples nearly overlaps with unity if the large uncertainties in the cumulative fission yields of ^{148}Nd for curium isotopes are taken into consideration. The %change C/E values are close to unity for all curium isotopes. These results indicate that there is the possibility of overestimation in the fission yields of ^{148}Nd of curium isotopes. However, the uncertainty of the fission yield of ^{148}Nd of curium isotopes is large, so the measurements and reevaluation for curium isotopes are needed to reduce these higher values in the future. Also, The fission and capture cross sections of ^{246}Cm are poorly known and need to be measured.

6. CONCLUSION

For the study of minor actinide transmutation, reliable neutron cross section data are inevitable. This paper discusses the validation of nuclear cross section data of minor actinide. The cross-section data are evaluated by comparison of calculated integral reactor physics parameters and corresponding chemically analyzed data of irradiated minor actinide samples from ^{233}U to ^{248}Cm in a fast reactor.

Minor actinide samples were irradiated for the period of 492 effective full power days in the core of the 600-MW Dounreay Prototype Fast Reactor (PFR) under the US and UK Fast Rector Program. These irradiated samples were analyzed both at ORNL and JAERI in order to obtain highly reliable data of chemical analysis. Measured data are rates of depletion, transmutation, and fission-product generation. Obtained data would provide most valuable integral data for reactor physics calculation of minor actinides from the point of high quality of separated samples, wide range of samples and long irradiation period. These reactor physics integral data were calculated using the ORIGEN code. The cross-section data were obtained from the JENDL-3.2 library and the fission-yield data from the ENDF-349library.

Conclusions from the comparison between experimental data and numerical calculations are as follows;

1. We can obtain reliable FIMA values by using the ^{148}Nd method except that the uncertainties in the FIMA values are large for ^{234}U, ^{238}Pu, Am isotopes, and Cm isotopes because the ^{148}Nd yields are known poorly forthese isotopes and are probably overestimated. For these isotopes, measurements to improve the fission-yield data are needed.

2. In general, the reliability of JENDL-3.2 nuclear data for the minor actinides are at an adequate level for the conceptual design study of transmutation systems. But, there are some nuclides for which new measurements are needed particularly if the minor actinides constitute a major part of the nuclear fuel of a dedicated transmutation system. Such nuclides are ^{238}Pu and ^{242}Pu.

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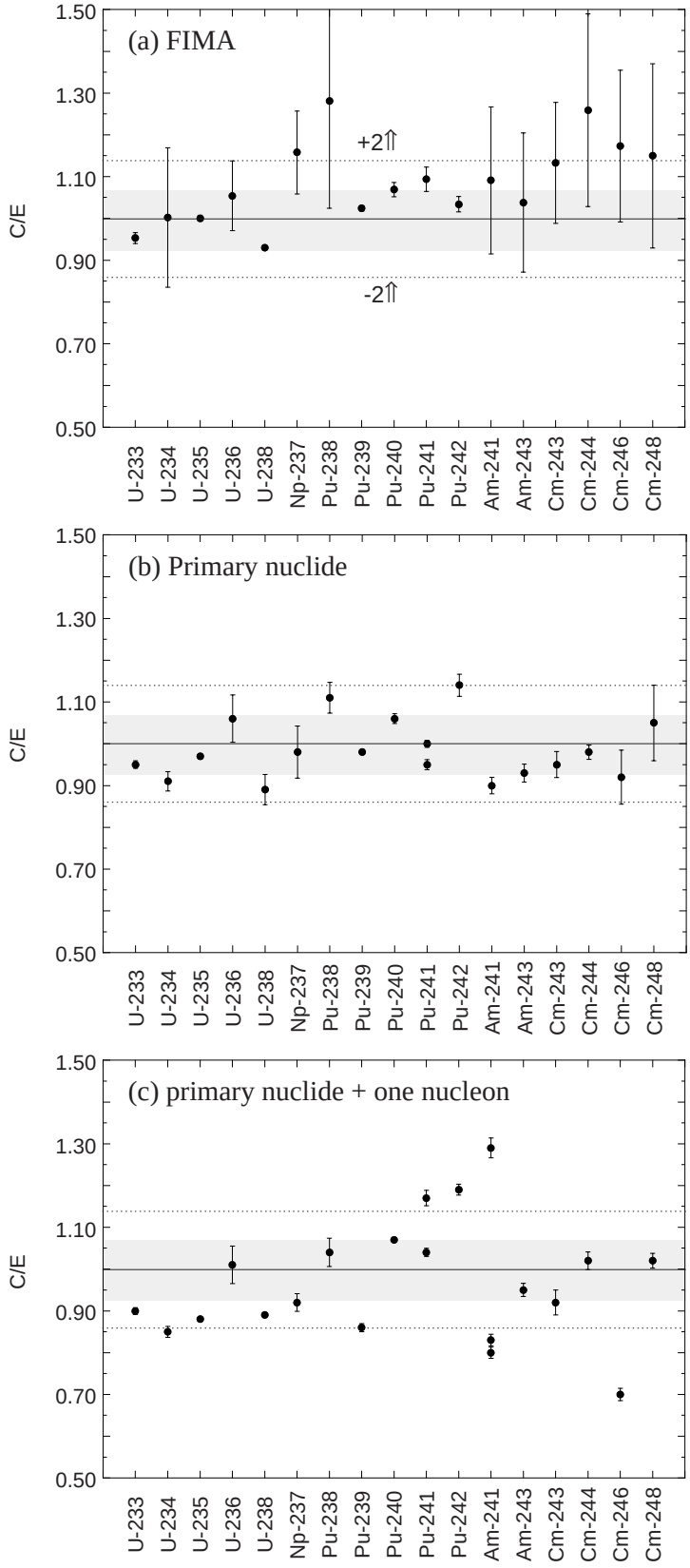

Figure 3. Summary of the calculated-to-experimental (C/E) values.