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A Nuclear Characteristics Study of Inert Matrix Fuel for MA Transmutation in Thermal Spectrum

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Abstract

The nuclear characteristics of a new conceptual fuel were analyzed as an option for incineration of plutonium extracted from nuclear warheads and spent fuel and transmutation of MAs. In order to eliminate uranium conversion to plutonium, the inert matrix fuel without uranium was selected to raise incinerating ratio. Erbia(Er_2O_3) and MAs as neutron absorbers were used for a reduction of large burnup reactivity swing and for a maximization of MA transmutation. The effects of plutonium and MA transmutation were compared with various neutron spectra which could be obtained on commercial PWR. As an option of inert matrix fuel with MA additives, it was confirmed that the feasibility of plutonium incineration in thermal spectrum and the transmutation of MAs is larger with hardened spectrum specially.

1. Introduction

For the incineration of excess plutonium derived from nuclear warheads and spent fuels in commercial nuclear power plant, some ideas have been proposed and are being studied vigorously in order to avoid proliferation risks. One of the ideas is the use of once-through inert matrix fuels(IMF) which substitute non-uranium oxide such as YSZ, Al_2O_3, ZrO_2, $MgAl_2O_4$ for uranium oxide base.

Using IMF without uranium, consists of only plutonium and inert matrix, there are several problems such as a large burnup reactivity swing, small fuel temperature coefficients and small delayed neutron fraction[1)]. Especially, because of a large burnup reactivity swing, to achieve compatible discharged

burnup rate with conventional LWRs', it is necessary to load large amounts of plutonium in IMF and to use burnable poison, Er_2O_3 so as to control the excess reactivity. About twice higher content of plutonium in IMF than that in mixed oxide fuel(MOX) may alter the incineration rate of plutonium and increase the production rate of Minor Actinides(MAs).

In this study, we proposed a new idea of IMF concept that utilizes MAs as burnable absorber instead of Er_2O_3. Although the absorption cross-sections of MAs are less than that of erbium, MAs could work as a burnable absorber effectively because their absorption cross-sections are mostly larger than that of plutonium. This concept gives another function of MA transmutation; MAs capture neutrons as a burnable poison material firstly, convert to fissile MAs and are destroyed by fission. For the maximization of transmutation in thermal spectrum, the spectrum effects were also tested. Therefore, we attempted to make the spectrum harder by reducing a fuel pin pitch. This is a well-known fact that fast spectrum is more effective in MA transmutation than thermal spectrum.

2. Fuel Composition and Calculation Model

The IMF composition in this study consists of three mixture oxides – Inert matrix, Weapon/Reactor grade plutonium and MAs. Aluminum oxide(Al_2O_3) which has good thermal conductivity was selected as an inert matrix base material. The research about the material property and characteristics of several inert matrix type – yttria stabilized zirconia(YSZ), magnesium spinel($MgAl_2O_4$) and corundum(Al_2O_3) - have been done in Japan Atomic Energy Research Institute(JAERI) [2]. JAERI study showed that corundum(Al_2O_3) had some problems in swelling and degradation of a material property after neutron irradiation. As a preliminary study, however, corundum(Al_2O_3) was selected for a base material. In order to maintain the reactivity and incinerate plutonium, pure plutonium was used without uranium. The compositions of plutonium were divided into two branches – weapon grade plutonium and reactor grade plutonium. Compositions of each plutonium grade were shown in Table 1. Because it is difficult to deal with weapon grade plutonium politically, this study was carried out with reactor grade plutonium.

Table 1. Isotopic composition of reactor grade and weapon grade Pu[3]

Pu-Grade	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	Am-241
Reactor Grade(a/o)	1.80	59.00	23.00	12.20	4.00	0.00
Weapon Grade(a/o)	0.00	94.00	5.30	0.70	0.00	0.00

The composition of MAs within spent fuel was evaluated by independent cell code calculations. Although many kinds of MA existed in the spent fuel of commercial PWR, 4 isotopes of MA which were crucial in transmutation because of their long half-lives and large amounts. The data of amount, half-life and number density of each chosen isotope were shown in Table 2.

Table 2. Isotopic composition of MAs

Isotopes*	Number Density (#/barn-cm)	Half Life (year)
Np237	1.2527E-05	2.1440E+06
Np239	2.1861E-06	2.3565(day)
Am241	1.0658E-06	4.3220E+02
Am242m	2.0411E-08	1.4100E+02
Am243	2.7070E-06	7.3700E+03
Cm242	3.6211E-07	162.79(day)
Cm243	7.8779E-09	2.9100E+01
Cm244	9.0272E-07	1.8100E+01
Cm245	5.3373E-08	8.5000E+03
Cm246	4.5768E-09	4.7600E+03

* after 40,000 MWD/MTU burnup in conventional PWR

Cell burnup calculation was accomplished by HELIOS code package[4]. Assembly geometry for the inert matrix fuel design was chosen to be the same with conventional 16 × 16 type PWR. A mixed fuel material is loaded at all of fuel rods homogeneously within an assembly. As the mass of each fuel composition, the same effective full power days(EFPDs) are targeted for the same thermal power. To verify the effect of spectrum hardening, the values of Vm/Vf are selected as 1.7, 1.3, 1.0 and 0.7 from PWR nominal case to the minimum value, which corresponds 1.285 cm, 1.198 cm, 1.129 cm, and 1.055 cm in pin pitch.

3. Calculation Results

3.1 Inert Matrix Fuel with Erbia for Plutonium Incineration

Before the calculation of IMF added with MAs, some calculations were performed about general IMF composition - Al_2O_3 matrix dispersed with PuO_2 and Erbia(Er_2O_3). To compare nuclear characteristics with UO_2 fuel and MOX, the discharged burnup length is designed by a linear reactivity model. The reactivity at about 900 EFPDs was adopted to be zero, resulting that the compositions that satisfies above condition are 4 w/o U-235 enrichment for UO_2 fuel, 6 % Pu volume fraction for MOX, and about 10-15 % Pu volume fraction with 1-1.5 % Erbia additive for IMF. Figure 1 shows the burnup reactivity changes of IMF loaded with weapon-grade and reactor-grade plutonium. The rapid k-infinite value changes of IMF with burnup are compensated by Erbia additive sufficiently and we could have flatted k-infinite curves than that of MOX.

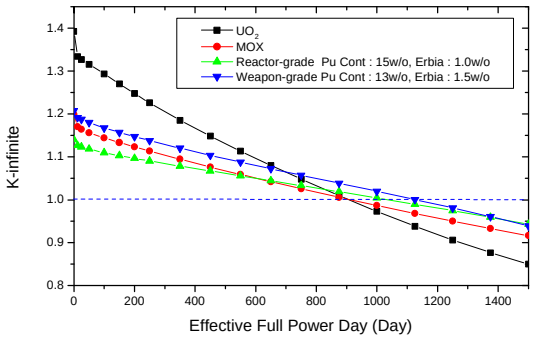

Figure 1. Burnup reactivity changes of UO2, MOX and IMF

The changes of incinerated plutonium fissile amount through the cycle length to 1,400 days were shown in Figure 2 and 3. Because of thermal spectrum, although the fraction vector of Pu-241 increases as shown in Figure 2, a large amount of Pu-239 could be incinerated. When the small amount of reactor-grade plutonium was used especially, the vector of Pu-241 decreased at EOC because of the lack of fissile materials as shown in Figure 3.

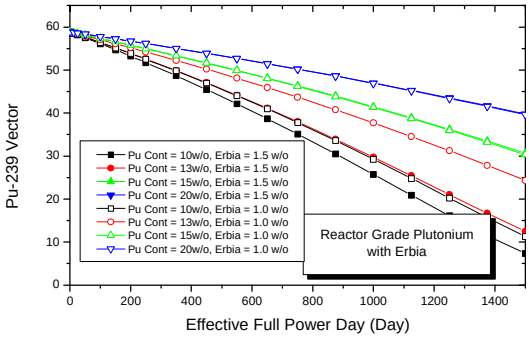

Figure 2. Pu-239 vectors with burnup in reactor-grade Pu

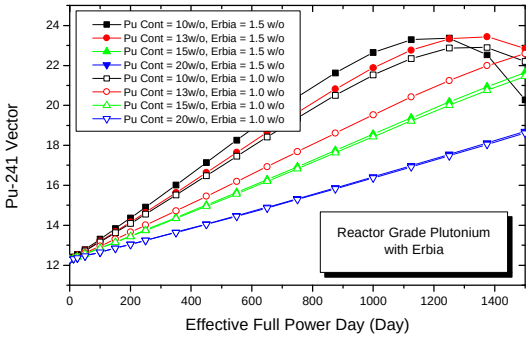

Figure 3. Pu-241 vectors with burnup in reactor-grade Pu

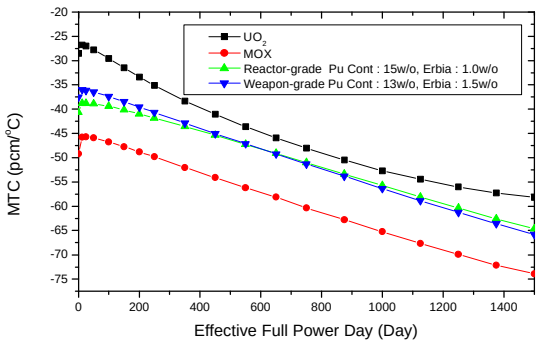

Figure 4. Comparisons of MTC with burnup

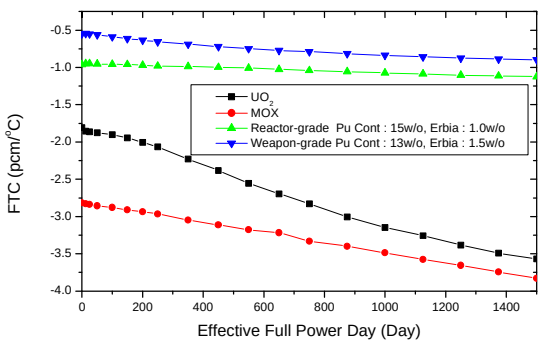

Figure 5. Comparisons of FTC with burnup

In the safety aspect, moderator temperature coefficient(MTC) and fuel temperature coefficient(FTC) and pin peaking factor of IMF were satisfied as shown by Figure 4, 5 and 6. Because of the absence of uranium, however, the delayed neutron fraction(β_tot) is much lower than that of MOX. Therefore, the reactor control can be a problem at startup and shutdown and it is also expected that a full IMF core could be an impractical option. Fortunately, the β values of IMF increased during irradiation because β value of Pu-241 is bigger than that of Pu-239 as shown in Figure 7.

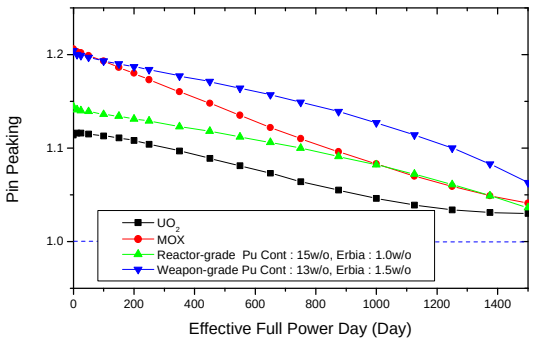

Figure 6. Comparisons of pin peaking.

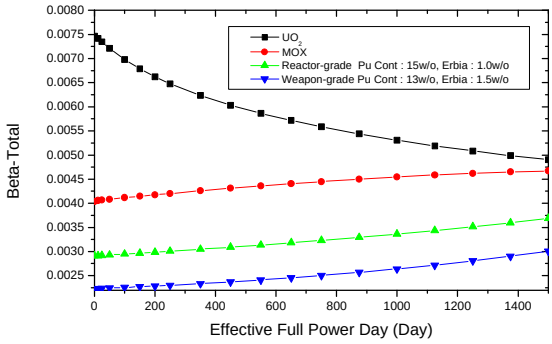

Figure 7. Comparisons of delayed neutron fraction

3.2 Inert Matrix Fuel with MAs additives for MA Transmutation

In this chapter, MA transmutation in thermal spectrum was tested with IMF design. A parametric study about composition choice of IMF was performed with various plutonium contents from 10 to 15 w/o and MA contents from 5 to 13 w/o. The value of Vm/Vf is also adjusted from 0.7, 1.0, 1.0 and 1.7. As shown in Figure 8., the trend of k-infinite slope is not changed with different Vm/Vf ratios when a small amount of MAs is added. Because of smaller absorption cross-section of MAs than that of Erbium, addition of large amount of MAs is good for the flattening of k-infinite curve, although the loss of reactivity might be too large.

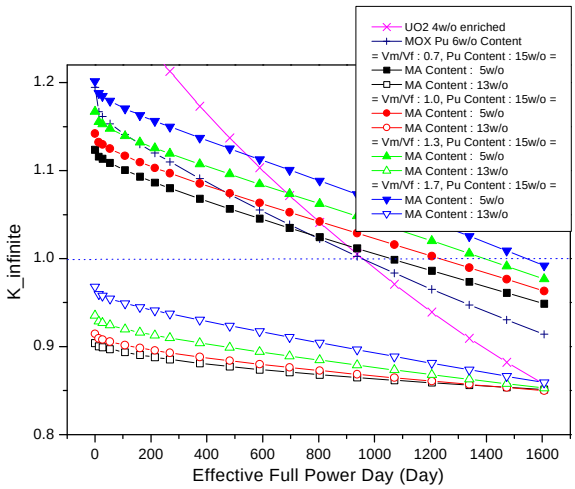

Figure 8. The trend of k-infinite curves with various MAs additives

The change of isotopic amounts of plutonium and MAs with different neutron spectrum were calculated and analyzed. As shown in Table 3, in a soft spectrum when Vm/Vf ratio is 1.7, the efficiency of incineration plutonium was high, 6.93 kg in soft spectrum compared with 5.86 kg in harder spectrum. On the contrary, a hard spectrum when Vm/Vf ratio is 0.7 is good to transmute more MAs, 4.42 kg in harder spectrum compared with 3.34 kg in soft spectrum. However, it infers that the most of Np-237 was transmuted to Pu-238 unfortunately. It means that Pu-238 that also has to eliminate because of high radioactivity and decay heat was produced during the transmuting Np-237.

Table 3. The mass difference of TRU isotopes between BOC and EOC (in unit of kg/Assembly)

Vm/Vf=0.7	Np237	Np239	Am241	Am242m	Am243	Cm242	Cm243	Cm244	Cm245	Cm246	Total
BOC	12.6160	0.0000	1.0916	0.0000	2.7955	0.0000	0.0000	0.9360	0.0000	0.0000	17.4391
EOC	7.8638	0.0000	0.9836	0.0378	1.8637	0.1166	0.0057	1.6976	0.4256	0.0213	13.0156
Diff. of Mass	-4.7522	0.0000	-0.1080	0.0378	-0.9318	0.1166	0.0057	0.7616	0.4256	0.0213	-4.4235

	Pu238	Pu239	Pu240	Pu241	Pu242	Total
BOC	0.6685	22.0056	8.6143	4.5884	1.5108	37.3876
EOC	4.8586	12.3658	7.9988	4.5623	1.7395	31.5250
Diff. of Mass	4.1901	-9.6398	-0.6155	-0.0260	0.2287	-5.8626

Vm/Vf=1.7	Np237	Np239	Am241	Am242m	Am243	Cm242	Cm243	Cm244	Cm245	Cm246	Total
BOC	12.6160	0.0000	1.0916	0.0000	2.7955	0.0000	0.0000	0.9360	0.0000	0.0000	17.4391
EOC	8.9433	0.0000	1.0408	0.0292	2.0606	0.1143	0.0035	1.6309	0.2633	0.0133	14.0992
Diff. of Mass	-3.6727	0.0000	-0.0508	0.0292	-0.7349	0.1143	0.0035	0.6949	0.2633	0.0133	-3.3399

	Pu238	Pu239	Pu240	Pu241	Pu242	Total
BOC	0.6685	22.0056	8.6143	4.5884	1.5108	37.3876
EOC	4.0523	11.5873	8.2716	4.6389	1.9113	30.4613
Diff. of Mass	3.3837	-10.4182	-0.3427	0.0505	0.4005	-6.9262

When fuel pitch is reduced to 0.7 in Vm/Vf value from 1.7, neutron spectrum was not hardening enough because of H_2O moderator as shown in Figure 9. As results, the variations of fission/capture cross-section have negative values belong to neutron spectrum change.

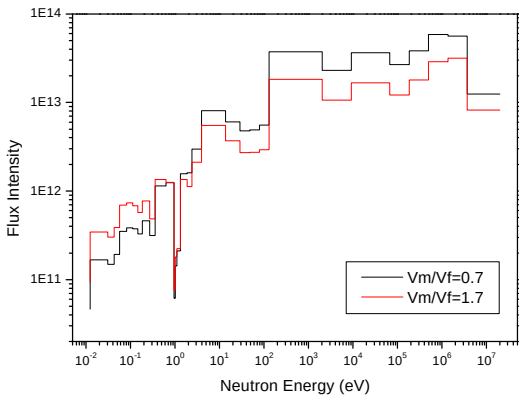

Figure 9. Neutron spectra of two different Vm/Vf value

3.3 Inert Matrix Fuel with MAs additives in D_2O moderator for MA Transmutation

As a neutron spectrum is hardened, the fission cross-section of fertile MAs becomes larger. However, the fission cross-section change with H_2O moderator was not considerable. In order to confirm a hardening effect, heavy water moderator was compared. As the change of moderators, harder spectra could be acquired as shown in Figure 10. When harder spectra were used, a amount of transmuted MAs is also larger than with H_2O moderator even though plutonium incineration ratio decreased as shown in Table 4. The effect of harder spectra was also shown in Table 5 about the cross-section of MA fertile.

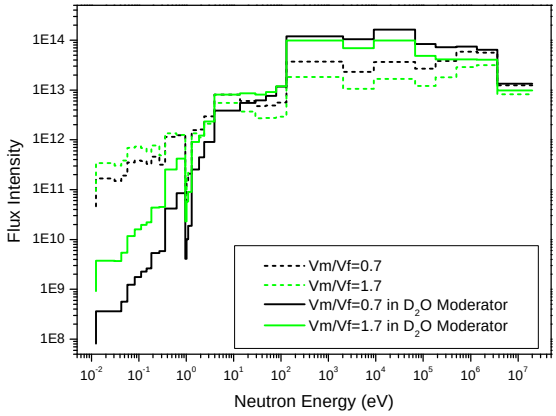

Figure 10. Neutron spectra belong to Vm/Vf values at BOC and EOC in D_2O moderator

Table 4. The mass difference of TRU isotopes between BOC and EOC in D_2O moderator

Vm/Vf=0.7	Np237	Np239	Am241	Am242m	Am243	Cm242	Cm243	Cm244	Cm245	Cm246	Total
BOC	12.6160	0.0000	1.0916	0.0000	2.7955	0.0000	0.0000	0.9360	0.0000	0.0000	17.4391
EOC	6.8494	0.0000	0.9493	0.0451	1.8160	0.1019	0.0105	1.5254	0.4014	0.0200	11.7191
Diff. of Mass	-5.7667	0.0000	-0.1423	0.0451	-0.9795	0.1019	0.0105	0.5895	0.4014	0.0200	-5.7200

	Pu238	Pu239	Pu240	Pu241	Pu242	Total
BOC	0.6685	22.0056	8.6143	4.5884	1.5108	37.3876
EOC	5.1959	12.6513	9.9028	3.2770	1.7925	32.8195
Diff. of Mass	4.5274	-9.3543	1.2885	-1.3114	0.2817	-4.5680

Vm/Vf=1.7	Np237	Np239	Am241	Am242m	Am243	Cm242	Cm243	Cm244	Cm245	Cm246	Total
BOC	12.6160	0.0000	1.0916	0.0000	2.7955	0.0000	0.0000	0.9360	0.0000	0.0000	17.4391
EOC	6.6340	0.0000	0.9166	0.0461	1.7414	0.1118	0.0102	1.6264	0.5482	0.0305	11.6651
Diff. of Mass	-5.9820	0.0000	-0.1750	0.0461	-1.0541	0.1118	0.0102	0.6905	0.5482	0.0305	-5.7740

	Pu238	Pu239	Pu240	Pu241	Pu242	Total
BOC	0.6685	22.0056	8.6143	4.5884	1.5108	37.3876
EOC	5.6247	12.6211	9.1961	3.7067	1.7128	32.8614
Diff. of Mass	4.9561	-9.3844	0.5818	-0.8817	0.2020	-4.5261

Table 5. Two Group Cross-section of TRU in D_2O moderator

Isotopes	Fission Cross-section					
	Fast Group			Thermal Group		
	Vm/Vf=0.7	Vm/Vf=1.7	Diff.	Vm/Vf=0.7	Vm/Vf=1.7	Diff.
Np237	3.1562E-01	2.9016E-01	2.5460E-02	8.5719E-03	8.0628E-03	5.0910E-04
Np239	3.8118E-01	3.3650E-01	4.4680E-02	0.0000E+00	0.0000E+00	0.0000E+00
Pu238	1.6106E+00	1.7188E+00	-1.0820E-01	1.3377E+00	1.5657E+00	-2.2800E-01
Pu239	4.5544E+00	6.4507E+00	-1.8963E+00	3.5607E+02	3.7379E+02	-1.7720E+01
Pu240	3.8394E-01	3.5988E-01	2.4060E-02	2.9682E-02	2.8628E-02	1.0540E-03
Pu241	8.2882E+00	1.3136E+01	-4.8478E+00	1.9690E+02	2.3054E+02	-3.3640E+01
Pu242	2.7030E-01	2.4925E-01	2.1050E-02	2.0166E-04	2.1090E-04	-9.2400E-06
Am241	3.4197E-01	3.5532E-01	-1.3350E-02	2.9176E+00	2.8559E+00	6.1700E-02
Am242m	1.6453E+01	2.7096E+01	-1.0643E+01	1.3890E+03	1.5434E+03	-1.5440E+02
Am243	2.6039E-01	2.5636E-01	4.0300E-03	5.5424E-02	5.1540E-02	3.8840E-03
Cm242	1.9568E-01	1.8738E-01	8.3000E-03	4.8181E-01	5.1540E-01	-3.3590E-02
Cm243	1.2630E+01	2.5301E+01	-1.2671E+01	1.6582E+02	1.6916E+02	-3.3400E+00
Cm244	5.1615E-01	5.8756E-01	-7.1410E-02	9.1105E-02	9.7906E-02	-6.8010E-03
Cm245	9.3268E+00	1.5154E+01	-5.8272E+00	2.2239E+02	2.4603E+02	-2.3640E+01
Cm246	3.2671E-01	3.4422E-01	-1.7510E-02	1.4415E-02	1.4780E-02	-3.6500E-04
Isotopes	Capture Cross-section					
Np237	7.1299E+00	1.1388E+01	-4.2580E+00	3.0553E+02	2.7493E+02	3.0599E+01
Np239	8.6768E+00	1.3353E+01	-4.6757E+00	2.2422E+01	2.3909E+01	-1.4870E+00
Pu238	3.3518E+00	4.7426E+00	-1.3908E+00	4.3902E+01	5.1430E+01	-7.5280E+00
Pu239	2.7664E+00	4.1503E+00	-1.3839E+00	2.0995E+02	2.2083E+02	-1.0880E+01
Pu240	3.1225E+00	6.6300E+00	-3.5076E+00	1.4384E+02	1.3812E+02	5.7189E+00
Pu241	1.9658E+00	3.3710E+00	-1.4052E+00	7.9960E+01	9.0190E+01	-1.0230E+01
Pu242	2.9574E+00	7.4108E+00	-4.4534E+00	4.8472E+00	4.9274E+00	-8.0191E-02
Am241	7.5741E+00	1.2487E+01	-4.9126E+00	7.0244E+02	6.6528E+02	3.7158E+01
Am242m	2.5850E+00	4.3090E+00	-1.7240E+00	2.7900E+02	3.1180E+02	-3.2800E+01
Am243	7.2079E+00	1.3704E+01	-6.4957E+00	3.1730E+01	3.0654E+01	1.0751E+00
Cm242	2.6411E+00	4.0001E+00	-1.3590E+00	2.7238E+00	2.9094E+00	-1.8561E-01
Cm243	1.8430E+00	3.6620E+00	-1.8190E+00	1.3700E+01	1.3990E+01	-2.9000E-01
Cm244	5.2343E+00	1.0744E+01	-5.5102E+00	2.0153E+00	2.1042E+00	-8.8899E-02
Cm245	1.2272E+00	2.0730E+00	-8.4580E-01	2.5950E+01	3.0160E+01	-4.2100E+00
Cm246	1.5701E+00	2.5859E+00	-1.0158E+00	2.9235E-01	3.0012E-01	-7.7750E-03

4. Conclusions

The nuclear feasibility of IMF was shown with weapon-grade/reactor-grade plutonium and Erbia. When the volume fraction of plutonium is 13-15 % with 1.0-1.5 % Erbia additive dependant on the composition of plutonium grade, the same discharged burnup of 4 w/o enriched UO_2 fuel could be attained and the vector of Pu-239 decreased from 60% to 30% in reactor-grade plutonium and from 95% to 55% in weapon-grade plutonium. Because total delayed neutron fraction of IMF is lower than MOX, the reactor control might be a problem at startup and shutdown operation.

In order to add another ability of MA transmutation, MAs can be mixed in IMF as substitute for Erbia BP. In soft spectrum, it is confirmed that the most isotope of transmuted MAs was Np-237 and Np-237 was transmuted to Pu-238. The spectrum hardening effect and the change of moderator material were tested to maximize MA transmutation. Operating conditions which used 0.7 in Vm/Vf value and D_2O moderator could transmute about twice amount of MA, 5.7 kg MAs per assembly, than that on the commercial PWR condition and decrease the accumulation of higher MAs such as Curium. The ratio of the production of Pu-238 to the transmutation amount of Np-237 also decreased on above condition.

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