COMPARATIVE STUDY FOR MINOR ACTINIDE TRANSMUTATION IN VARIOUS FAST REACTOR CORE CONCEPTS

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Abstract

A comparative evaluation of minor actinide (MA) transmutation property was performed for various fast reactor core concepts. The differences of MA transmutation property were classified by the variations of fuel type (oxide, nitride, metal), coolant type (sodium, lead, carbon dioxide) and design philosophy. Both nitride and metal fuels bring about 10% larger MA transmutation amount compared with oxide fuel. The MA transmutation amount is almost unchanged by the difference between sodium and lead coolants, while carbon dioxide causes a reduction by about 10% compared with those. The changes of MA transmutation property by fuel and coolant types are comparatively small. The effects caused by the difference of core design are rather significant.

1. Introduction

Research and development of the fast reactor have been carried out in several nations with mixed oxide fuel and sodium coolant chosen as standard components of the reactor core. In addition, the transmutation of minor actinide (MA) nuclides using a fast reactor core has been investigated extensively from the viewpoint of reducing the environmental burden of long-lived radioisotopes, that is considered as one of the main features of the fast reactor.

Meanwhile alternative core concepts have been proposed to utilise liquid heavy metal or gas as a coolant, and the design works of these cores are performed widely to exploit the merit of each coolant. Concerning the fuel material, both mixed nitride and metal fuels are considered as the feasible candidates. The MA transmutation properties have also been investigated for these core concepts made from various fuel and coolant materials, and a good performance has been reported for each concept as a result of its hard neutron spectrum.

In order to provide basic information for selecting candidates of the commercial-use fast reactor core, this study presents a comparative evaluation of MA transmutation property for various fast reactor core concepts employing the same analytical method and nuclear data. The differences among oxide, nitride and metal fuels were examined, besides the three types of coolants, sodium, lead and carbon dioxide were compared.

2. Calculation conditions

2.1 Definition of investigated cores

Various fast reactor cores investigated in this study are described below. All of them were selected in terms of the practical feasibility prospected by conceptional design works.

Table 1 shows the three sodium-cooled large cores that examine the difference among oxide, nitride and metal fuels (referred as Na-MOX, Na-MN and Na-Metal cores). In order to extract the difference of fuel type the basic reactor performances (i.e. reactor power, cycle length, average fuel burn-up, coolant pressure drop, etc.) were not changed. Maximum linear heat rate was also conserved among these cores in the same level (limited up to about 430 W/cm). Fuel pin diameter, pin pitch and number of sub-assemblies were adjusted in order to satisfy these conditions.

It seems that the nitride and metal-fuelled cores have better core characteristics than that of the oxide-fuelled core. Their high heavy metal density causes a reduction in Pu enrichment as well as burn-up reactivity loss. It increases also the breeding ratio.

The reference oxide fuelled core was designed and investigated as a feasible candidate for the commercial-use fast reactor by JNC-Japan [1]. It is a conventional FBR core having homogeneous two fuel enrichment zones surrounded by fertile blanket. One remarkable feature is that automatically dropping absorber using a curie-point magnet is installed inside each of inner core sub-assembly. The number of fuel pins in inner core sub-assembly is reduced for placing a guide tube of the safety device. Nevertheless this does not influence on the MA transmutation property.

The investigated cores to examine the difference of coolants between sodium and lead are listed in Table 2. The BREST-300 reactor proposed by RDIPE-Russia [2] was selected as a reference lead-cooled power reactor. It has a safety-oriented core concept such as almost zero burn-up reactivity, low coolant pressure drop and low coolant void reactivity, which is totally different from a conventional FBR design. The superior core characteristics are achieved by means of high heavy metal density of nitride fuel, reflection effect of lead coolant and a sub-assembly without duct tube. BREST-300 has no fertile blanket but large heavy metal inventory in other words it can be said that the blanket is included into the active core.

Table 1. **Core design parameters for the fast reactors compared in this study (1)**
– Oxide fuel vs. nitride and metal fuels –

	Na-MOX Large-sized core	**Na-MN core**	**Na-Metal core**
Reactor power [MW_{th}]	3 800	←	←
Operation cycle length [EFPD]	540	←	←
Fuel exchange batch	5	←	←
Average fuel burn-up [GW_{th}d/t]	150	←	←
Core height [cm]	120	←	←
Coolant	Na	←	←
Coolant temperature [^{o}C] (outlet / inlet)	550/395	←	←
Coolant pressure drop [kg/cm^2]	~3	←	←
Fuel type	(U, Pu)$O_{1.98}$	(U, Pu)^{15}N	U-Pu-10Zr
Pu isotopic vector [wt%] (^{238}Pu/^{239} Pu /^{240} Pu /^{241} Pu /^{242} Pu /^{241}Am)	3/52/27/9.5/7/1.5	←	←
Fuel pin diameter [mm]	9.7	8.40	9.02
Smeared density [%TD]	82	80	75
Pin pitch/pin diameter	1.15	1.20	1.17
Number of fuel pins per sub-assembly (IC/OC)	234/271	←	←
Sub-assembly pitch [mm]	195.4	178.2	185.9
Number of sub-assemblies (IC/OC)	246/216	252/210	252/210
Heavy metal inventory (core) [t]	63	63	64
Pu enrichment [wt%] (IC/OC)	17.8/19.8	14.3/16.3	14.9/16.9
Burn-up reactivity loss [%dk/kk’]	2.9	1.4	1.9
Breeding ratio	1.04	1.14	1.11

Table 2. **Core design parameters for the fast reactors compared in this study (2)**
– Sodium coolant vs. lead coolant –

	Na-MOX Medium-sized core	**Pb-MOX core**	**Pb-MOX core (Low pressure drop)**	**Pb-MN core (Low pressure drop)**	**BREST-300 (Ref. [2]) (*IC/MC/OC)**
Reactor power [MW_{th}]	700	←	←	←	←
Operation cycle length [EFPD]	540	←	←	←	~300
Fuel exchange batch	5	←	←	←	←
Average fuel burn-up [$GW_{th}d/t$]	150	←	←	←	60
Core height [cm]	120	←	←	←	110
Coolant	Na	Pb	←	←	←
Coolant temp. [^{o}C] (outlet/inlet)	550/395	←	←	←	540/420
Coolant pressure drop [kg/cm^2]	~3	←	~1	←	←
Fuel type	$(U, Pu)O_{1.98}$	←	←	$(U, Pu)^{15}N$	←
Pu vector [wt%] ($^{238}Pu/^{239}Pu/^{240}Pu/^{241}Pu/^{242}Pu/^{241}Am/^{242m}Am/^{243}Am$)	3/52/27/9.5/7/1.5/0/0	←	←	←	0.5/64/28/3.1/ 1.7/2.1/0.1/0.5
Fuel pin diameter [mm]	9.7	9.7	9.7	8.87	9.1/9.6/10.4*
Pin pitch / pin diameter	1.15	1.27	1.40	1.45	1.49/1.42/1.31*
No. of fuel pins per S/A (IC/OC)	271	←	←	←	114
S/A pitch [mm]	195.4	216.0	238.2	226.3	150
No. of S/As (IC/OC)	30/54	30/54	30/54	30/54	57/72/56*
Heavy metal inventory (core) [t]	12	12	12	12	16
Pu enrichment [wt%] (IC/OC)	18.8/23.9	19.0/24.7	21.1/26.8	17.8/22.6	14.0/14.0/14.0*
Burn-up reactivity loss [%dk/kk’]	2.1	2.2	2.4	2.0	~0
Breeding ratio	1.11	1.08	1.02	1.09	~1

Table 3. **Core design parameters for the fast reactors compared in this study (3)**
– Sodium coolant vs. carbon dioxide coolant –

	Na-MOX Large-sized core	**Na-MOX core (Equivalent to ETGBR)**	**ETGBR (Ref. [3])**
Reactor power [MW_{th}]	3 800	→	3 600
Operation cycle length [EFPD]	540	→	344
Fuel exchange batch	→	→	5
Average fuel burn-up [$GW_{th}d/t$]	150	→	120
Core height [cm]	120	→	150
Coolant	Na	←	CO_2
Coolant temperature [°C] (outlet/inlet)	550/395	←	525/252
Coolant pressure drop [kg/cm^2]	~3	←	~3?
Fuel type	→	→	$(U, Pu)O_{1.98}$
Fuel pin diameter [mm]	9.7	7.84	8.2
Pin pitch/pin diameter	1.15	1.30	1.55
Number of fuel pins per sub-assembly (IC/OC)	234/271	271	169
Sub-assembly pitch [mm]	195.4	185.2	180.6
Number of sub-assemblies (IC/OC)	246/216	183/161	334/216
Heavy metal inventory (core) [t]	63	49	50
Pu enrichment [wt%] (IC/OC)	17.8/19.8	17.5/21.5	18.7/26.7
Burnup reactivity loss [%dk/kk’]	2.9	2.5	2.4
Breeding ratio	1.04	1.08	1.08

In the present investigation, a series of lead cooled cores (Pb-MOX, Pb-MN cores) was prepared starting from a conventional medium-sized Na-MOX core as shown in Table 2. These cores provide the effects of a replacement of sodium to lead, a reduction of coolant pressure drop and an employment of nitride fuel under keeping the reactor performance in the same level. It is found that employment of the lead coolant and the reduction of coolant pressure drop make the core characteristics worsen while the nitride fuel improves them. The rest of the differences between Pb-MN core and BREST-300 are to be considered all together as a difference of design philosophy.

The sub-assembly specification of the medium-sized Na-MOX core is the same as the large Na-MOX core appeared in Table 1, excepting that the safety devices are not placed inside the inner core sub-assemblies.

Table 3 presents the cores prepared for comparing coolant effects between sodium and gas. The ETGBR designed by NNC-United Kingdom [3] was chosen as a feasible concept of gas cooled fast reactor. It has a two-region homogeneous core employing MOX fuel and carbon dioxide gas coolant. Its active core height is larger than that of the reference large-sized Na-MOX core because there is no need to concern about the coolant void reactivity, this is one of the merits of gas cooled fast reactor. The other design parameters (reactor power, cycle length and average fuel burn-up) are also different. To compare these cores under the same condition, a Na-MOX core equivalent to ETGBR was prepared as shown in Table 3. Comparison among the three cores can classify the observed differences into the effects of coolants and design parameters. Almost the same burn-up characteristics are obtained for both the equivalent Na-MOX core and ETGBR except that a fairly high Pu enrichment is needed in the outer core of ETGBR.

2.2 Representation of MA transmutation

The MA treated in this study was assumed to come from LWR spent fuel with five-year cooling time before reprocessing. The isotopic composition of the MA is shown in Table 4, which was calculated by the ORIGEN2 code [4]. It consists mainly of fertile MA nuclides such as ^{237}Np, ^{241}Am and ^{243}Am.

Table 4. **Compositions of minor actinides from LWR waste***

Nuclide	**Composition (wt%)**
^{237}Np	49.14
^{241}Am	29.98
^{242m}Am	0.08
^{243}Am	15.5
^{242}Cm	0.0
^{243}Cm	0.05
^{244}Cm	4.99
^{245}Cm	0.26

* Discharged from PWR (35 GWd/t) and cooled for 5 years before reprocessing.

MA nuclides were homogeneously distributed into all the core fuel in replacement of heavy metal nuclides. A content of MA was considered up to 5wt% of the fuel heavy metal amount. The Pu enrichment was adjusted to assume the same minimum-required reactivity all through the operation cycle.

Following net MA transmutation amount per cumulative power was used in this study as a quantity representing the transmutation property:

MA transmutation amount = (MA inventory at BOC [kg] – MA inventory at EOC [kg])
[kg/GW$_{th}$/year] /Reactor power [GW$_{th}$]/Operation cycle length [year],

where the MA transmutation amount was divided by reactor power and cycle length in order to compare various reactor cores of different specifications. Note that the above MA transmutation amount has a dimension of transmuted amount per energy emission from a reactor.

2.3 Method of calculation

Neutron flux and depletion calculations were carried out by the CITATION code [5] in diffusion approximation where core geometry was modeled in two-dimensional RZ representation and a 7-group energy structure was used. Seven-group effective cross sections were collapsed from the adjusted cross section library ADJ98 [6] based on the evaluated nuclear data JENDL-3.2 [7]. The depletion calculation was performed until the core compositions settled down to a state of fuel cycle equilibrium. Neutron flux was normalised at each burn-up step using the values of fission energy emission recommended by Sher [8], where the contributions from capture gamma heat were also taken into account.

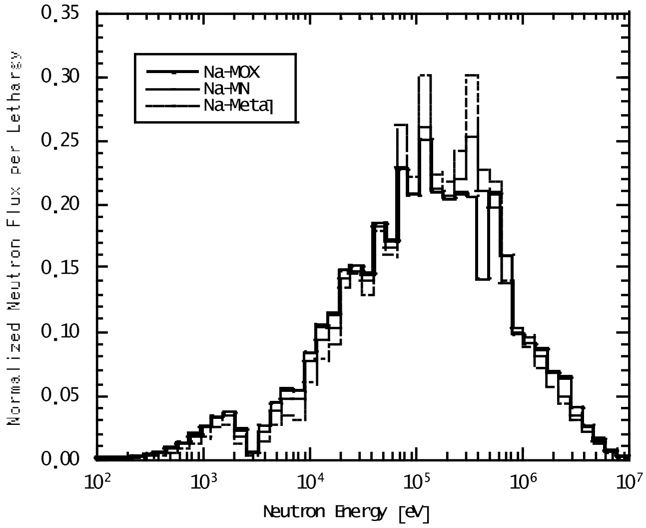
2.4 Comparison of neutron spectra

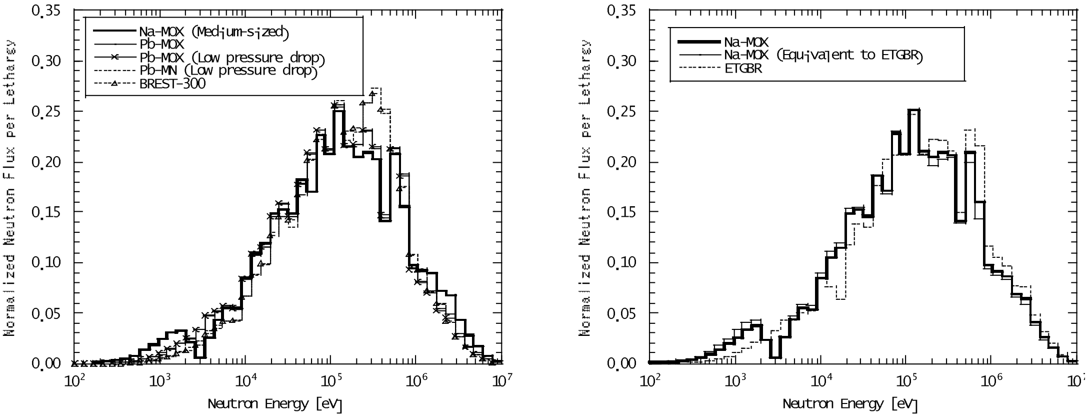
The neutron spectra for the fast reactor cores investigated in this study are shown in Figure 1. Transmutation of MA includes capture reactions (^{237}Np, ^{241}Am), fissile-type fission reactions (^{242m}Am, ^{245}Cm) and threshold fission reactions (^{237}Np, ^{241}Am, ^{243}Am, etc.). One-group cross-sections for these transmutation reactions depend on the shape of neutron spectrum. The former two types of reaction are enhanced by the flux in keV energy region, the latter one is determined by the flux in MeV energy region.

Concerning the fuel types, a harder neutron spectrum is observed in the order of metal, nitride and oxide fuels (Figure 1(a)). For nitride and metal fuels, it should be noted that the decrease of flux in MeV energy region occurs after the normalisation due to the lack of resonance of oxygen around 500 keV.

Figure 1. **Comparison of neutron spectra at core center**

(a) Oxide fuel vs. nitride and metal fuels





(b) Sodium coolant vs. lead coolant

(c) Sodium coolant vs. carbon dioxide coolant

Figure 1(b) shows the cases for the comparison of sodium and lead. It is found that the lead coolant reduces the neutron flux in MeV energy region, which is caused by inelastic scattering of lead. Neutron hardening by lead coolant seems smaller than that obtained by nitride fuel. In addition, the differences of reactor size, design parameters as well as the design philosophy do not drive any significant change on the neutron spectrum. It is possible to say that the fuel and coolant types mainly determine the neutron spectrum.

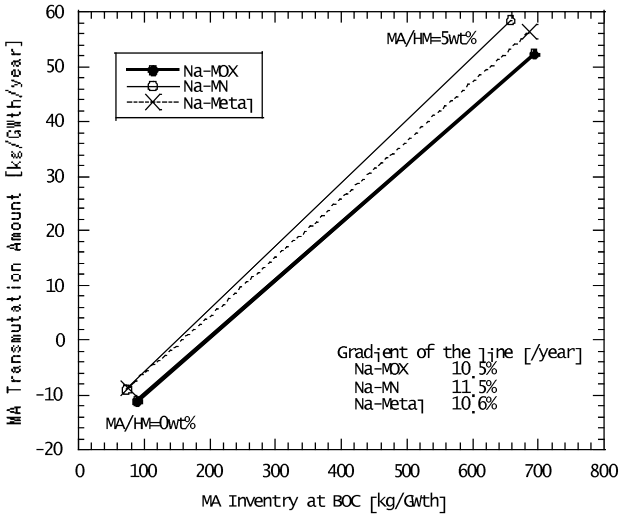
Carbon dioxide makes the neutron spectrum harder compared with sodium (Figure 1(c)). The comparison also shows that the difference of design parameters in the two Na-MOX cores does not alter the neutron spectrum so much.

3. Results and discussion

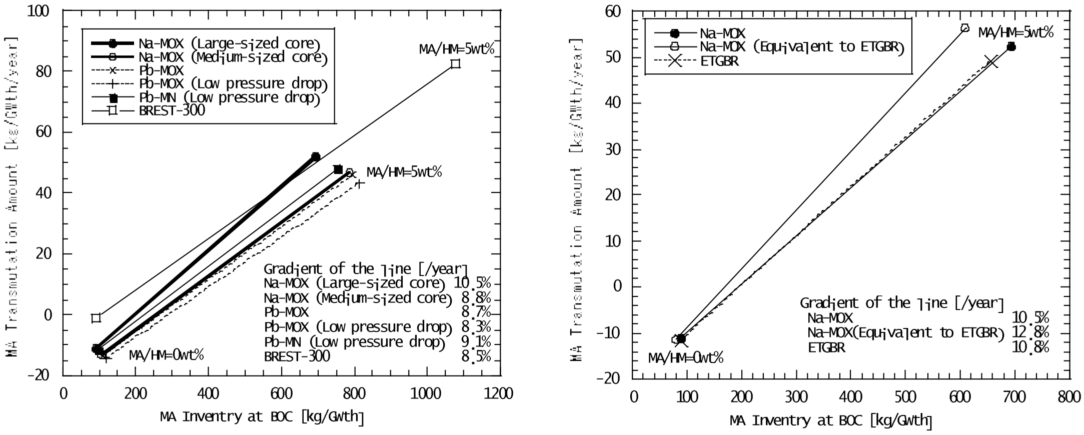
3.1 *MA transmutation properties*

The results of calculation for MA transmutation characteristics are indicated in Figure 2 below.

Figure 2. **Comparison of MA transmutation properties (LWR discharged MA)**



(a) Oxide fuel vs. nitride and metal fuels



(b) Sodium coolant vs. lead coolant

(c) Sodium coolant vs. carbon dioxide coolant

The vertical and horizontal axes represent the MA transmutation amount defined in the previous section and MA inventory at BOC, respectively. The gradient of the line stands for MA transmutation rate.

It is found that the nitride and metal-fueled cores bring about 10% larger MA transmutation amount compared with the oxide-fueled core (Figure 2(a)). In addition the effect of reduction of Pu enrichment appears as an upward transfer of the graphic lines at the point of MA/HM=0wt%.

From a comparison of sodium and lead cooled cores in Figure 2(b), it is shown that the MA transmutation amount is almost unchanged by the substitution of sodium to lead, reduced by the decrease of the coolant pressure drop, and increased by the utilization of nitride fuel. However the differences caused by a change of reactor power and the unique core concept of BREST-300 are rather significant. Especially, large heavy metal inventory of BREST-300 enables to obtain a larger MA transmutation amount. The transmutation rate of BREST-300 is almost same as that of Pb-MN core, there might be a compensation effect arise from the differences of reactor specifications such as lead reflector, Pu isotopic composition, cycle length, fuel burn-up, etc.

Figure 3(c) presents the difference between sodium coolant and carbon dioxide coolant. It is found that the MA transmutation amount decreases about 10% from sodium to carbon dioxide when compared in an equivalent reactor condition. It should be noted that the difference in core specification between the two Na-MOX cores has caused considerable changes.

As a result, it is found that the differences in MA transmutation property arising from the variation of fuel and coolant types are comparatively small, that is within ±10%. The effects caused by the core design difference are rather significant. Since it is possible to construct various types of reactor core even using the same fuel and coolant material, the reactor design philosophy might be more important factor which changes the MA transmutation property. If we put more priority on MA transmutation efficiency rather than core characteristics, it is possible to say that nitride and metal fuels have more potential for MA transmutation.

3.2 Breakdown of the changes in MA transmutation

More detailed examination of the changes in MA transmutation more showed some interesting results. The difference of MA transmutation amount can be divided into the effects of neutron flux level, neutron spectrum and MA inventory. When we compare the MA transmutation rate, only the factors of flux level and spectrum are to be considered. Result of the analysis is shown in Table 5, where the effects are extracted from comparing the cores in the equivalent reactor condition.

Table 5. **Analysis of the changes in MA transmutation amount**
(LWR discharged MA, MA/HM = 5wt%)

	Fuel type		**Reactor power**	**Coolant type**	
	MOX→MN	MOX→Metal	Large→ Medium	Na→Pb	Na→CO_2
Total flux level	+23%	+31%	-27%	+10%	-14%
Neutron spectrum*	-3%	-22%	-1%	-13%	-9%
MA inventory per reactor power	-8%	-2%	+17%	+1%	+10%
Net	+12%	+8%	-10%	+2%	-13%

* Including the effect of Pu enrichment change.

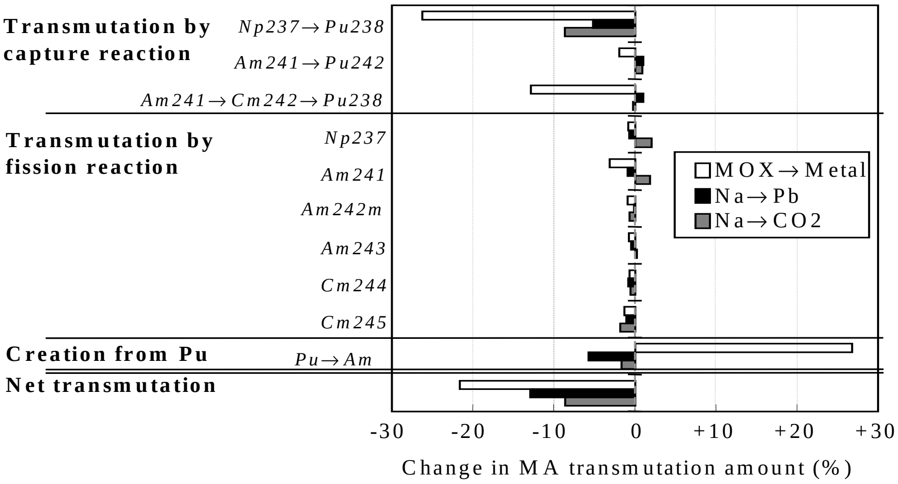
It is found that the higher MA transmutation amount observed in nitride and metal-fueled cores is the consequence of the increase in neutron flux level.

When the reactor power is reduced, the worsened neutron economy causes the decrease in flux level and the increase in MA inventory. This can also be seen in the substitution from sodium to carbon dioxide, which seems to be resulted from the relatively poor neutron economy due to high neutron leakage.

Thought the net MA transmutation properties are the same between sodium and lead cooled cores, there exits a cancellation of the flux level and the neutron spectrum effects.

The effect of neutron spectrum depends on MA nuclide composition. Then a decomposition of the neutron spectrum effect into individual reaction process was carried out as shown in Figure 3.

Figure 3. **Effect of neutron spectrum change for individual MA transmutation process (LWR discharged MA, MA/HM=5wt%)**



It turns out that the spectrum effect on the transmutation of LWR discharged MA consists mainly from the capture reactions of ^{237}Np and ^{241}Am, as well as the creation of Am from Pu. Especially for the replacement of oxide fuel by metal fuel, the large negative contribution of capture reaction processes is compensated by the large positive contribution from a reduction of MA creation from Pu.

The tendency of the variation of each reaction process is consistent with the changes of the neutron spectrum and the Pu enrichment. For most nuclides, transmutation components by capture and fission reactions are getting reduced by the spectrum hardenings, excepting the increase in the threshold fission reactions by the substitution from sodium to carbon dioxide. It can be said that a hardening of neutron spectrum does not always give a positive contribution to the MA transmutation performance.

3.3 Effects on the core characteristics

Effects on the core characteristics by the MA loading were reviewed. Changes of core characteristics by 5% of MA introduction are shown in Table 6. By the role of fertile material of MA there occur the decreases in Pu enrichment and burn-up reactivity loss. However the coolant void reactivity and the Doppler constant are changed towards not-safe direction due to the change of direct and adjoint flux. Relatively large increase of the coolant void reactivity on BREST-300 is caused by a larger amount of lead in the core. Since the tendency for the changes in core characteristics looks similar for every type of the cores, it is possible to say that neither fuel nor coolant types brings a prominent penalty to the core characteristics induced by MA loading.

Table 6. **Changes in the core characteristics by MA loading**
(LWR discharged MA, MA/HM=0.5wt%)

	Na-MOX large-sized core	**Na-MN core**	**Na-Metal core**	**Pb-MOX core**	**BREST-300 (*IC/MC/OC)**	**ETGBR**
Change in Pu enrichment [wt%] (IC/OC)	-1.2/-1.4	-2.0/-1.5	-1.7/-1.4	-1.1/-1.0	-0.1/+0.1/+0.2*	-1.1/-0.4
Change in burn-up reactivity [%dk/kk’]	-1.7	-2.2	-2.0	-1.4	-0.7	-1.1
Change in breeding ratio	+0.01	+0.04	+0.03	0.00	-0.06	-0.01
Change in coolant void reactivity [%dk/kk’]	+0.47	+0.49	+0.44	+0.55	+0.81	Not available
Change in Doppler constant [10^{-3}Tdk/dT]	+1.8	+1.6	+1.1	+1.0	+1.6	+1.3

4. Conclusions

MA transmutation properties for various fast reactor cores were compared in the equivalent condition as a power reactor. The observed differences were classified into the effects of fuel, coolant type and design philosophy separately. It is concluded that there shows no significant difference in MA transmutation amounts and rates arising from the variation of fuel types (oxide, nitride, metal) and coolants (sodium, lead, carbon dioxide). The effects caused by the difference of core design are rather important. If we stand on the viewpoint that core characteristics should be compensated for improving the MA transmutation efficiency, it is possible to say that nitride and metal-fueled cores have more potential for MA transmutation than oxide fueled core. By means of breaking down the MA transmutation amount into the effects of flux level, spectrum and MA inventory, the differences of MA transmutation property were analyzed more in detail.

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