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TECHNICAL REPORT

Minor Actinides Incineration by Loading Moderated Targets in Fast Reactor

Hongchun WU*, Daisuke SATO**[†] and Toshikazu TAKEDA**

**Department of Nuclear Engineering, School of Energy and Power Engineering, Xi'an Jiaotong University*

***Department of Nuclear Engineering, Graduate School of Engineering, Osaka University*

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The effect of hydrogen concentration and loaded mass of minor actinides (MAs) in the target on the core performance and MAs transmutation rate was analyzed in this paper. An optimum core was proposed which has 96 MAs target assemblies of which MAs fuel pins per assembly is 38 with the composition ratio U/MA/Zr/H of 1/4/10/50. This optimized core offers good core performance and can transmute MAs very effectively, the transmutation rate was about 67% (939 kg) and the incinerate (transmute by fission) rate was about 35% (489 kg) through 3 years of reactor operation. It is about 2–3 times larger than current transmutation method that MAs are loaded homogeneously in the PWR and fast reactor core.

KEYWORDS: *minor actinides, transmutation, moderated targets, fast reactors, hydrogen, concentration dependence, fuel pins, reactor core, performance*

I. Introduction

Minor actinides (MAs) accumulated in discharged fuel of thermal reactors mainly contributes to the hazard indices over 100 years after the storage begins. So there are a lot of studies to incinerate MAs in conventional and advanced LWR, fast reactor (FR)^{(1)–(4)}. As for the application of thermal reactors, rather over-moderated (soft neutron spectrum) fuel cells were proposed⁽³⁾⁽⁴⁾. This is mainly because the capture reaction cross sections of ²³⁷Np and ²⁴¹Am are large in thermal energy and the resultant ²³⁹Pu and ^{242m}Am have large fission cross sections. On the other hand, as for FRs, rather hard spectrum cores were proposed⁽⁵⁾. The use of hard spectrum results in the increase of threshold fission reactions of ²³⁷Np and ²⁴¹Am.

However, in LWRs the total neutron flux is low, although the neutron spectrum is so soft that the neutron reaction cross sections of the MAs are large. On the other hand, FRs can provide higher flux, but the neutron spectrum is hard. So it seems a great potential to achieve the best transmutation of the MAs by loading moderated targets in FRs. Recently, loading of moderated targets in the FR was proposed⁽⁶⁾. It can incinerate the MAs very effectively because it can provide both the high flux level and soft spectrum.

Although the moderated targets loading in FR can achieve high transmutation rate the use of the moderated targets results in a significant variation of the power

distribution during burnup. So we consider an optimum content of hydrogen, MAs mass in the moderated target mixture and loading pattern to keep both good core performance and high transmutation rate.

Uranium zirconium hydride (U–Zr–H) fuel has been used in TRIGA research reactors for many years. The hydride fuel has several advantages such as high hydrogen atom concentration as well as inherent safety, low release of fission products, and high thermal conductivity. So we can use MA-containing hydride fuel or the MA-containing material surrounded by ZrH₂ metal or mixtures of them. We denote the atomic ratio of the target as U/MA/Zr/H, and change the atomic ratio of hydrogen, the mixture mass in the target and the target number in the core to survey their effect on the core parameters respectively.

Usually the MAs inventory difference before and after a burnup is used for the MAs transmutation rate. However it is important to know the incineration by fission, the Pu production, and the MAs production from Pu and other MAs. So we have decomposed the commonly used transmutation rate (TR) into four components⁽⁴⁾⁽⁵⁾: (1) The overall fission rate OF (containing the fission of Pu or other MAs after transmuting target MA nuclide), (2) The Pu production rate PuR (production rate of Pu isotope from target MA nuclide), (3) The MA production rate MAR (production rate of other MA nuclides from target MA nuclide), (4) The element production rate EPR (production rate of the target MA nuclide from other MA nuclide and Pu isotope).

II. Calculation Models

As for FRs, we considered 2 cases of which thermal

* Xi'an, 710049, P.R. CHINA.

** Yamada-oka, Suita-shi 565-0871.

[†] Corresponding author, Tel. & Fax. +81-6-6879-7903,
E-mail: dsato@nucl.eng.osaka-u.ac.jp

power is 2,600 MW: MOX fueled Na cooled reactor, and metal fueled Pb cooled reactor. The cycle length is one year, refueling batch is 3 and assembly pitch is 14.7 cm. Number of MA target assemblies is 96 which are loaded dispersedly in the core as shown in Fig. 1. Control rods are all withdrawn. Obviously, the maximum number of targets is 96 if we hope to load the targets without neighboring each other so as to avoid the local core power peaking. The residence time of targets in the core is 3 years. The main design parameters of this 1,000 MW-FBR are shown in Tables 1 and 2.

The composition of the MAs corresponds to that of the discharged fuel after 35 GWd/t burnup and 5-year cooling of currently used uranium fuel in PWRs: 49.2% ²³⁷Np, 30.0% ²⁴¹Am, 15.5% ²⁴³Am, 5.0% ²⁴⁴Cm⁽⁷⁾. The Pu vector for the fuel was assumed as 56.2% ²³⁹Pu, 24.4% ²⁴⁰Pu, 12.2% ²⁴¹Pu, 7.3% ²⁴²Pu⁽⁷⁾.

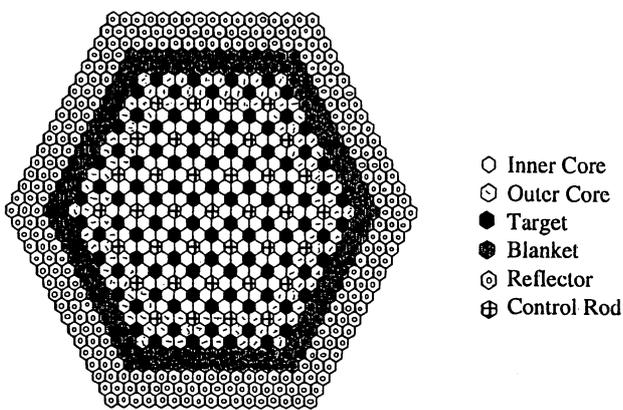


Fig. 1 FBR core layout

Usually core calculations of FRs are performed in 70 groups in Japan, and the accuracy is enough for estimation of core performance parameters. Though the use of moderator in target assemblies increases the thermal neutron spectrum, in the 70 groups structure contains only one energy group below 0.3 eV. Then for the simple case where one MA target assembly was loaded in the core center, we checked the accuracy of the 70 groups calculations by comparing with 107 groups calculations, where 30 groups are added below 0.3 eV. The 107 groups cross section was obtained by SRAC⁽⁸⁾ code based on JENDL-3.2⁽⁹⁾ library. The K_{eff} and transmutation rate of ²³⁷Np, ²⁴¹Am and ²⁴³Am in a target assembly evaluated from 70 and 107 groups calculations are shown in Table 3. The K_{eff} difference is less than 0.2% and the difference of transmutation rate is no more than 10%. The difference was rather small, so the calculations were performed in 70 groups.

Firstly, the calculations were performed by CASUP⁽¹⁰⁾ code to generate the 70 groups effective cross sections based on the JENDL-3.2 library. Secondly, few group cross sections of each nuclide were obtained by group collapsing using the neutron spectrum obtained from 70 groups two-dimensional *R-Z* diffusion calculations by the CITATION⁽¹¹⁾ code. In order to study the effect of targets loading pattern of the core, we used three-dimensional hexagonal nodal diffusion code ICOM3D code. Lastly, the burnup calculations were performed by the ORIGEN2⁽¹²⁾ code using the one group cross section. All these codes have been assembled into a big code package SPENDALL (Search of Plutonium Enrichment and Transmutation of Radiative Waste Calculation Code for FR). It can search the optimum Pu enrichment au-

Table 1 Main design parameters of FBR

Reactor thermal power	2,600 MW	Reactor electric power	1,000 MW
Cycle length	1 year (3 batch refueling)	Core concept	2-regional homogeneous
Fuel	MOX or metal	Coolant	Na or Pb
Core diameter/Height	300/100 cm	Pitch of assembly	14.7 cm
Number of fuel assembly (IC/OC/TA)	144/120/150	Number of target assembly	96

Table 2 Pu enrichment for each case studied

Fuel type	Core region	Pu enrichment (wt%)				
		I-1 [†]	I-2 [†]	I-3 [†]	I-4 [†]	I-5 [†]
MOX-Na	IC/OC	20.7/26.1	24.9/31.2	31.9/39.7	38.2/47.3	42.7/52.7
Metal-Pb	IC/OC	12.7/16.2	16.9/21.2	22.1/27.5	26.6/32.8	30.0/36.7
		II-1 ^{††}	II-2 ^{††}	II-3 ^{††}	II-4 ^{††}	II-5 ^{††}
MOX-Na	IC/OC	22.7/28.6	29.9/37.4	38.2/47.3	42.3/53.0	42.8/53.7
Metal-Pb	IC/OC	12.8/16.2	21.0/26.2	26.6/32.8	29.2/35.8	30.1/36.8

[†]Table 4, ^{††}Table 5

Table 3 Comparison of 70 and 107 calculations[†]

	70 groups	107 groups
K_{eff}	1.076308	1.077940
Transmutation rate (%)		
^{237}Np	94.8	82.2
^{241}Am	98.6	94.4
^{243}Am	93.2	82.5

[†]One MA target assembly loaded in core center.

tomatically which fulfills the condition that the K_{eff} at EOC becomes unity. At the same time, the difference of core power peaks appeared in the inner core and the outer core is minimized through adjusting the Pu enrichment ratio of inner core to outer core. This code can also give the four components of TR such as OF, PuR, MAR and EPR.

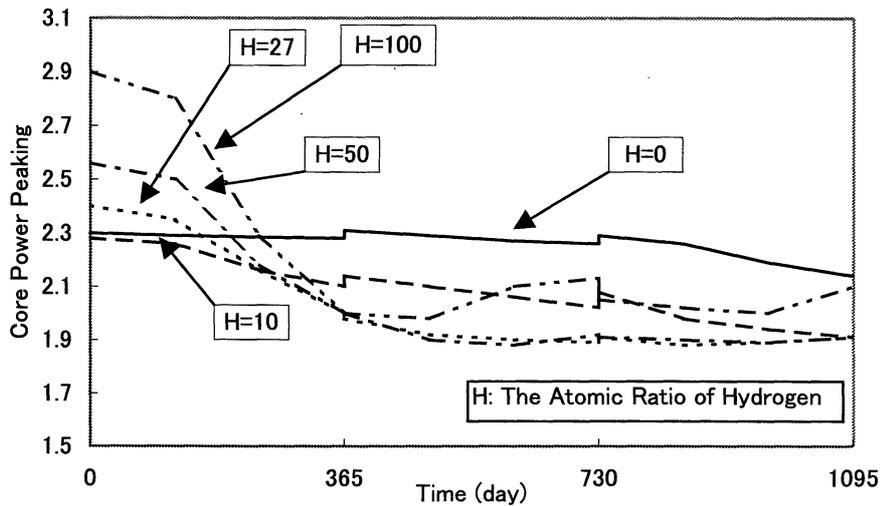
III. Calculation Results

1. Effect of Hydrogen Contained in Target

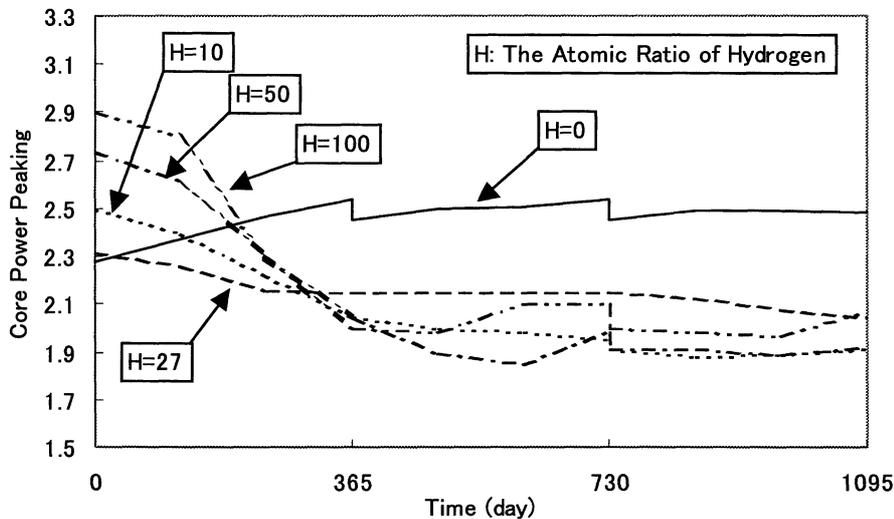
In this paper, target was considered as a homogeneous mixture of uranium, MAs, zirconium and hydrogen. The composition U/MA/Zr/H in this hydride fuel

Table 4 Hydrogen content for each case studied

Case-I	I-1	I-2	I-3	I-4	I-5
U/MA/Zr/H	1/4/10/0	1/4/10/10	1/4/10/27	1/4/10/50	1/4/10/100



(a) MOX FR



(b) Metal FR

Fig. 2 Core power peaking factor for various hydrogen content in MOX FR and metal FR

was changed in order to evaluate the effect on MAs transmutation rate and core performance. As advised by Ref. (6), U/MA/Zr is assumed as 1/4/10. Now we investigate the effect of hydrogen contained in the target; the ratio of hydrogen was varied between 0 and 100. We chose five cases as shown in Table 4 so as to survey the effect of hydrogen on the core parameters and MAs incineration rate. Next, we investigated the number of MAs fuel pins in each target and the value of core power peaking factor, and we let the core power peaking factor not exceeding the factor, which is 2.5, of the case that each target has one MAs fuel pin.

Because of the moderation by hydrogen, the thermal power peaking appears near the target assemblies, and the core power peaking at the beginning time becomes higher as the hydrogen content becomes higher as shown in Figs. 2(a) and (b). But after about one year, the local power peaking is reduced because of the bigger neutron

capture cross section of the targets.

Figures 3(a) and (b) show the transmutation rate of MAs after 3 years of burnup. Here the number of MAs fuel pins in each target assembly is 38 and the corresponding MAs mass is 14.54 kg. So the total loaded MAs mass in the core is about 1,400 kg. We found when hydrogen content is changed from 50 to 100, the difference of transmuted MAs was very small. So the case I-4, in which composition is 1/4/10/50, is considered as the upper limit for hydrogen content. Moreover, its core power peaking is acceptably small. So the optimum relative content of hydrogen in target is taken as 50 hereafter.

2. Effect of MAs Mass Loaded in Target

Based on the survey in the proceeding section, we found the optimum relative composition ratio of moderated target is U/MA/Zr/H=1/4/10/50. And we also found the difference between MOX fueled FR with Na

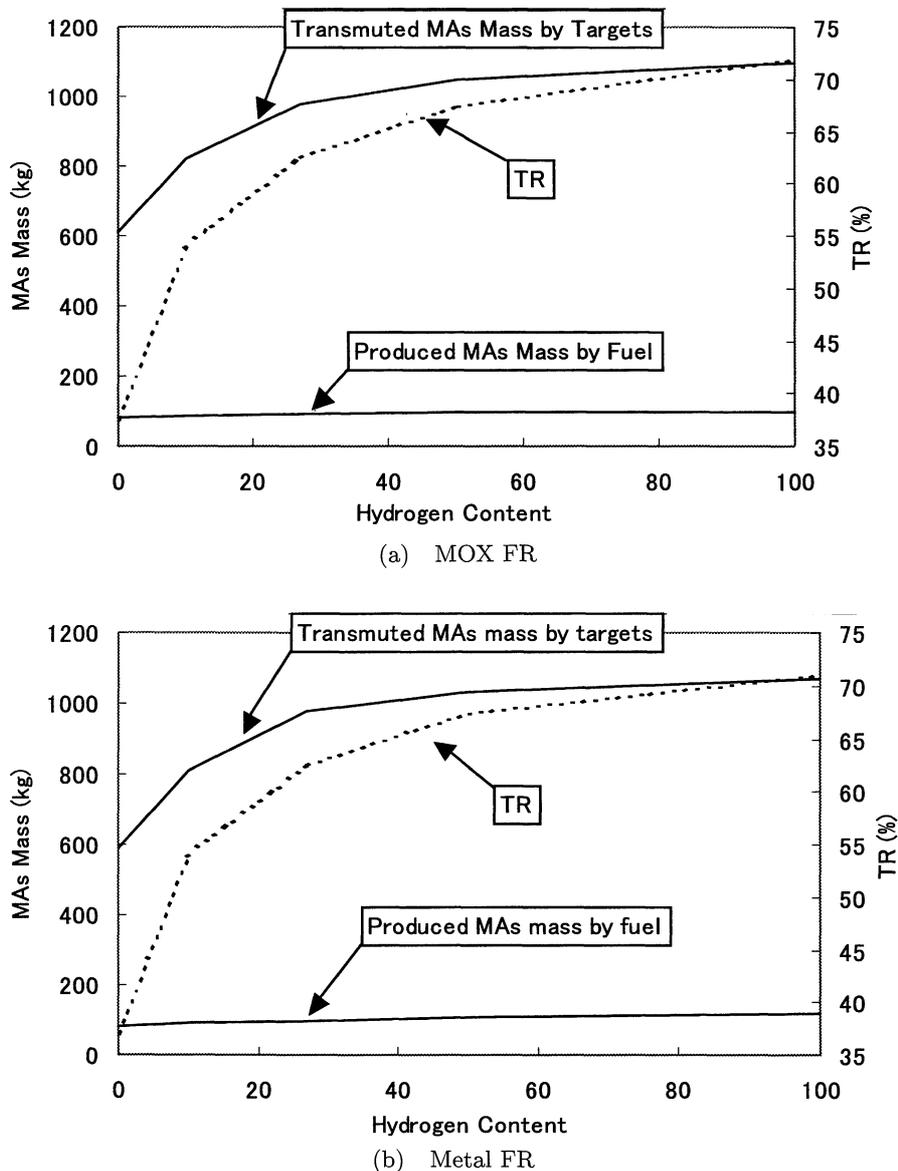


Fig. 3 Transmutation rates for various hydrogen content in MOX FR and metal FR

coolant and metal fueled FR with Pb coolant is small. So hereafter we will discuss the MOX fueled FR only. Now in order to survey the effect of absolute target mass loaded in the core, we keep the relative composition ratio (1:4:10:50) while the total number of target assemblies (96) is constant. We alter the mass of each target only by changing the number of MAs fuel pins in the target assemblies as shown in **Table 5**. Here the maximum number of MAs fuel pins in each assembly is 127. We assumed that if MAs fuel pins in the targets are withdrawn, the coolant Na fill the vacancy.

As shown in **Fig. 4**, when the loaded MAs mass is larger, the absolute transmuted MAs mass is larger. Of

course, the produced MAs mass should also be larger because the Pu enrichment must be raised to compensate the neutron absorption of MAs. So the transmutation rates are smaller as shown in **Fig. 5**. From **Fig. 5** we found if the MAs mass in each target is less than about 7 kg, the transmutation rate is negative because the produced MAs mass by the fuel is larger than the transmuted MAs mass by the targets. Considering both the efficiency and the core power peaking as shown in **Fig. 5**, the optimum case is Case II-3 where loaded MAs mass in each target is 14.54 kg; it can achieve almost the highest transmutation rate about 67.3%, transmute MAs about 939 kg and the maximum core power peaking is 2.56.

Table 5 Target mass for each case studied

Case-II	II-1	II-2	II-3	II-4	II-5
Number of MAs pin cell in each target	1	19	38	57	76
Corresponding MAs mass in each target (kg)	0.38	7.27	14.54	21.81	29.07

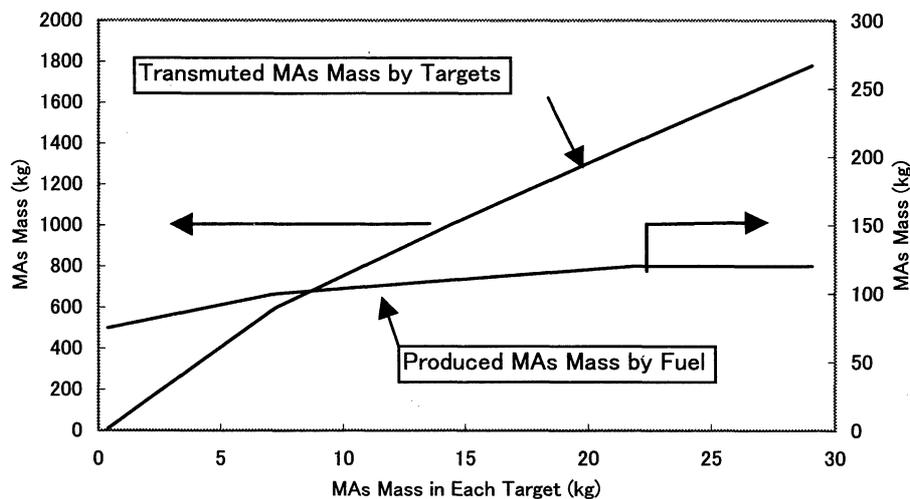


Fig. 4 Transmuted mass and produced mass for various target mass

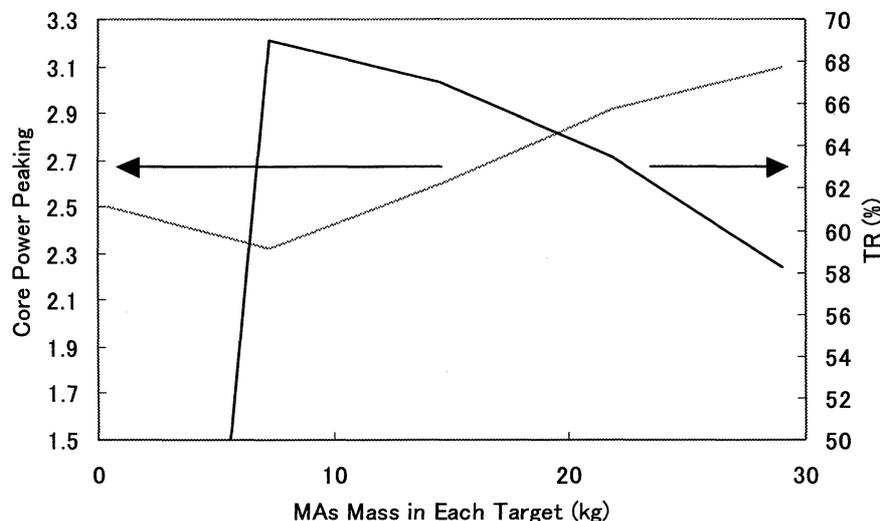


Fig. 5 Transmutation rates and core power peaking for various target mass

3. Effect of Loaded Target Number

In order to evaluate the effect of number of targets on the core performance and transmutation rate, we changed the number of targets. We kept the target composition U/MA/Zr/H to be 1/4/10/50 and the MAs mass loaded in each target to be 14.54 kg, which has been proved to be the optimum case in the proceeding sections. All targets were loaded dispersedly in the core so as to avoid the power peaking problem.

As shown in Fig. 6, when the number of target is 96, the core power peaking achieves the lowest, because this case is nearly the homogeneous loading pattern. In fact, from Fig. 1 we can find the maximum number of target is 96 if we have to load the target without neighboring each other. If we load more targets in this core, there must be 3 targets adjacent each other. Due to the symmetry of the core, there should be 6 groups of such targets in the core, when the total number of target in the core is 102. The "grouping" of targets will result in a big shifting of the core power distribution. As shown in Fig. 6 the maximum core power peaking will be raised from 2.56 to 2.93. From Fig. 6 we found the transmutation rate of 96 targets case achieved the maximum. So it is undesirable if the number of target exceeds 96.

4. Comparison of Transmutation and Incineration Rate

Table 6 shows the comparison of the optimum case of this paper to the optimum cases of HMPWR (High Moderated PWR) and FR⁽⁵⁾. It indicates the "Moderated Targets" case of this paper is very effective. It is about 2-3 times larger than the current method by PWR and FRs. As shown in Fig. 7, the Pu production rate (PuR) of MAs occupies about 40% of transmutation rate. This means the "waste" MAs can be transmuted effectively and used as fissionable material using the proposed method.

IV. Conclusion

Calculation results of this paper indicate some conclusions: The optimum composition ratio of the U/MA/Zr/H of moderated target is 1/4/10/50. If the hydrogen is more than 50 there is not much effectiveness on minor actinides (MAs) transmutation but will result in bigger core power peaking. In both MOX fueled and metal fueled fast reactor (FR) of this paper, the optimum MAs mass loaded in each target assembly is about 14.54 kg. The targets are loaded in all over the core without making "target groups" can achieve a good core performance and high transmutation rate. In the sample FR of this paper the maximum number of target is 96.

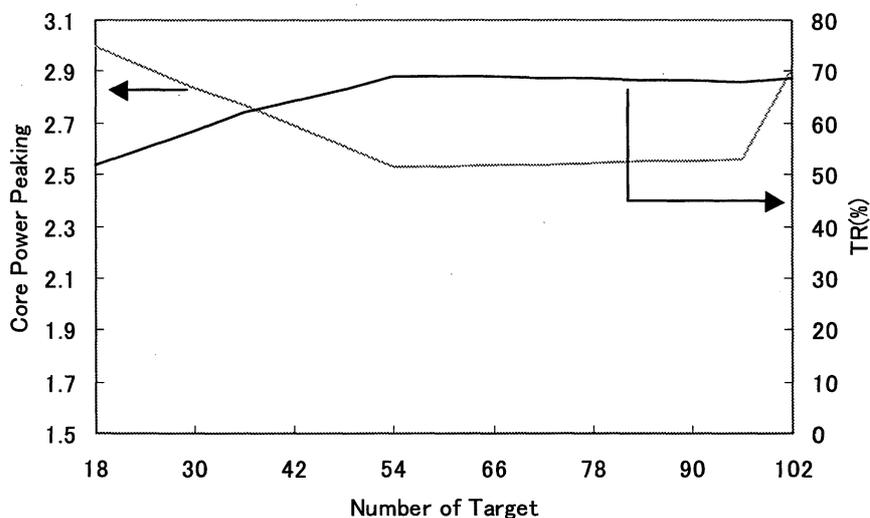


Fig. 6 Transmutation rates and core power peaking for various target number

Table 6 Transmutation rates comparison of optimum case of different reactor

Reactor type	TR		OF	
	(%/GWth·yr)	(kg/GWth·yr)	(%/GWth·yr)	(kg/GWth·yr)
High moderated PWR	3.36	27.50	0.69	5.60
Metal-Pb-FR	3.10	55.70	1.45	26.00
Moderated targets loading in FR				
MOX-Na	8.63	120.41	4.47	62.40
Metal-Pb	8.67	121.04	4.46	62.19

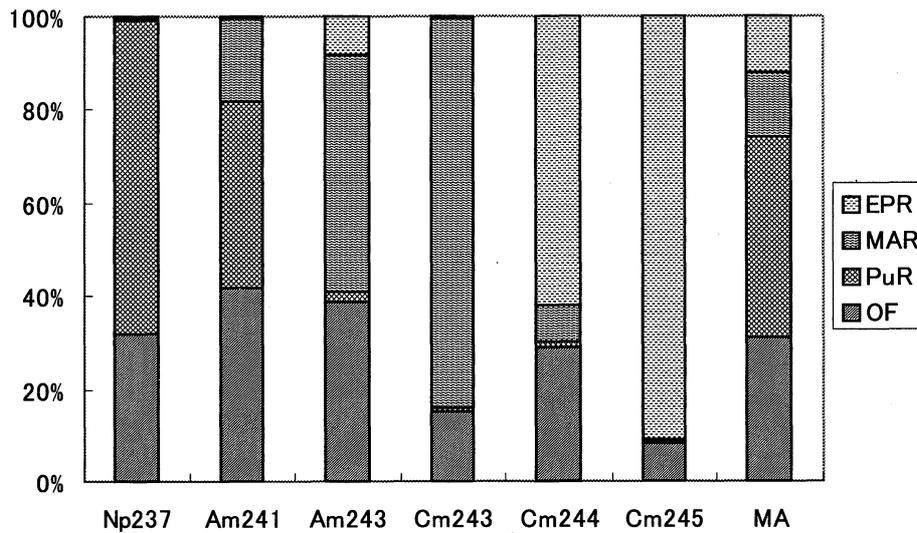


Fig. 7 Components of transmutation rate for optimum case

It has been proved that the optimum case of moderated targets loading in FR has great potential to incinerate MAs effectively. In the example of this paper, it can transmute MAs about 939 kg and incinerate MAs about 489 kg through 3 years of reactor operation. It is about 2–3 times larger than general method that loads MAs homogeneously in the PWR and FR.

We proposed the optimum core loading moderated targets from a viewpoint of only reactor physics in this study. Furthermore, as the future work, to realize this optimum core concept, experimental research should be done about the irradiation performance and fabrication.

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