TRU Transmutation in an LMFBR

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ABSTRACT

The characteristics of TRU transmutation in a 1000MWe-class LMFBR with mixed-oxide fuel were quantitatively studied to substantiate the feasibility.

We considered here two typical methods transmuting TRU in the LMFBR core. One is the method dispersing TRU homogeneously throughout the entire core (homogeneous TRU-loading method). It was found that the homogeneous method had no serious penalties of TRU loading to the reactor core performance, and the TRU transmutation rate reached approximately 11% per cycle with the loading of the weight ratio of 5% TRU in MOX fuel. The amount of the TRU transmutation in the LMFBR is almost six times as much as that of the TRU production from a 1000MWe-class LWR. As another desirable property, the TRU-loaded core was found to reduce the burnup reactivity loss by 40% compared with the reference **a** core without TRU. It can be concluded that the TRU transmutation in an LMFBR has no problems from the viewpoint of the core performance, provided that the homogeneous method could be employed.

The other possible method is the use of small number of subassemblies (target S/As) which concentrate TRU in fuel (heterogeneous TRU-loading method). The weight ratio of 50% TRU fuel was assumed to be loaded in the target S/As. The calculating results showed that the swing of power peaking near the target S/As became quite severe and critical for the thermal design criteria, although the TRU transmutation rate and other reactor core performances of the heterogeneous method were roughly comparable to the homogeneous method. It seems from the present study that the heterogeneous method has quite a serious problem to be overcome and much effort would be needed to make the method feasible.

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\$1. Introduction

The proper management of High Level Wastes (HLW) which are inevitably produced with reactor operations is one of the most important problems to complete the nuclear fuel cycle (Ref. 1). Most of fission products in the HLW have relatively short half-lives and decay to stable form within a few hundred years, while the transuraniums (TRU) like Np237 and Am243, the α -emitters, have extremely long half-lives of millions of years and contribute to the long-term radiotoxicity of the fuel cycle wastes. It is a strong social requirement not to leave these long-life HLW to future generations, and Power Reactor and Nuclear Fuel Development Corporation (PNC) has launched the project of the TRU transmutation study in LMFBRs from the view point of both reactor core characteristics and nuclear fuel cycle.

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It is generally acknowledged that LMFBRs would be superior TRU transmutation devices, mainly due to their hard neutron spectrum (Ref. 2). In the course of the TRU study of PNC, we first surveyed the relation between the TRU transmutation rates and the various reactor core parameters which included amount of TRU loading, fuel type, reactor core size, reactor operation length per cycle, power density, isotopic composition of TRU. The objective of the parametric study was to understand the basic characteristics of TRU transmutation in LMFBRs (Ref. 3). As the second stage, we chose a large LMFBR with mixedoxide(M0X) fuel as the TRU transmutation device and studied the characteristics of TRU transmutation in the large LMFBR quantitatively to substantiate the feasibility. The present report describes the summary of the second stage of the TRU transmutation study at PNC.

\$2. Survey of TRU Transmutation in a Large LMFBR **a**

First, a 1000MWe-class LMFBR core with MOX fuel was defined as the reference whose main parameters are shown in Table 2.1 and Fig. 2.1. The nuclear characteristics of TRU-loaded core was calculated by two-dimensional diffusion theory with depletion chain. The cross sections used were seven effective group constants condensed from the Japanese standard 70-group constant set, JFS-3-J2 (Ref. 4), which is based on an evaluated nuclear data library, JENDL-2 (Ref. 5).

The loaded-TRU material was assumed to come from LWR spent fuel with fiveyear cooling time before reprocessing. The isotopic composition of the TRU was calculated by the ORIGEN2 code (Ref. 6). Table 2.2 shows the initial composition and the burnup dependency of the fission reaction rate of each isotope in 1 kg of the TRU . It is found that the total fission rate increases with burnup and reaches the maximum value of one and half times of initial value at 4-year burnup time. The increase of the total fission rate is caused by the fission of Pu238 which is generated from the capture reaction of Np237.

We consider here two typical methods transmuting TRU in the LMFBR core. One is the method dispersing TRU homogeneously throughout the entire core (homogeneous TRU-loading method), 'which is expected not to affect the core characteristics seriously. The other possible method is the use of small number of subassemblies (target S/As) which concentrate TRU in MOX fuel (heterogeneous TRU-loading method), which can have the advantage of the fuel cycle because of handling the small number of TRU-included fuels in fabrication factory. The followings are the results of parametric studies regarding these two TRU-loading **a** methods.

2.1 Homogeneous TRU-loading Method

Reactor core characteristics evaluated here are amount of TRU transmuted, burnup reactivity loss and swing of power distribution with the TRU mass loaded in fuel as the parameter.

a. Amount of TRU Transmuted

Figure 2.2 shows the relationship between the amount of TRU transmuted and that of loaded-TRU in the FBR core. At least, one percent of TRU-loading to fuel would be needed to eliminate TRU, and the amount of TRU transmuted increases linearly with that of loaded-TRU. Since a 1000MWe-class LWR produces about 26 kg of TRU per year, an LMFBR with 5% TRU-loading can transmute the TRU mass from six LWRs in rough estimation.

b. Burnup Reactivity Loss

The TRU-loading to core results in significant decrease of burnup reactivity loss mainly due to Pu238 build-up. As seen in Fig. 2.3, the burnup reactivity becomes positive when the amount of TRU-loading is over lo%, therefore, the maximum amount of TRU-loading in the FBR core would be limited to about 10% from the aspect of reactor operational safety. From other viewpoint, a proper amount of TRU-loading might be advantageous to the extension of reactor operation period.

c. Swing of Power Distribution

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Figure 2.4 shows the swing of power distribution between the beginning and the end of equilibrium cycle (BOEC and EOEC) with the parameter of TRU mass loaded in the outer core region. It could be possible to minimize the

power swing by optimizing the ratio of loaded-TRU amount between the inner and the outer core region.

2.2 Heterogeneous TRU-loading Method

Parameters surveyed here are the mass of TRU contained in the target S/As, loading position of the target S/As and plutonium enrichment of the target S/As. The survey cases and the TRU transmutation ratio calculated are summarized in Table 2.3.

a. TRU Transmutation Rate

As seen in cases A30 through A100, or B30 through BlOO of Table 2.3, the TRU transmutation rate increases with the amount of TRU loading, but the tendency is not always linear and also depends on the position of the target S/As. About the dependence on loading position of the target S/As, the TRU transmutation rate in the case B50 where TRU is loaded at the periphery of the inner core is about four times as much as the case A50 of the core center loading with roughly identical TRU mass. This means that the target S/As perform high transmutation rate when they are loaded in a dispersive manner to avoid the depression of neutron flux.

In addition, quite a surprising fact was found from the case AB50, in which the target SIAs with 50% TRU were loaded at just the same positions with cases A50 and B50. The amount of TRU transmuted in the case AB50 was 147 kglcycle, which was by 80% larger than the summed value transmuted in the cases A50 and B50. This curious behavior of TRU transmutation was found to be caused by the increase of flux level in each TRU-loaded position due to the positive interference effect between two regions.

The case C50 shows the radial blanket region is another possibility to load TRU, since a large number of target S/As can be accepted there with no influence to core characteristics and transmute sufficient mass of TRU in spite of their low TRU transmutation rate.

Comparing cases A50, A50L and A50H, it is found that the TRU transmutation rate increases with plutonium enrichment of the target SlAs. This can be interpreted as the consequence of neutron flux increase by plutonium enrichment . We should consider, however, the swing power of target SlAs also increase, which is a large penalty for core thermal characteristics.

b. Swing of Power Distribution

Figure 2.5 shows the power distribution of the cases A30, A50 and A100 where the target S/As are loaded in the core center region with 30%, 50% and 100% TRU in the target SIAs, respectively. They are compared with no TRUloading case REF. At BOEC, the power of TRU-loaded region is quite depressed in these TRU-loading cases.

At EOEC, all three cases shows larger power in the TRU-loaded region than that of BOEC. The degree of the power swing tends to increase with the mass of loaded-TRU in the target SIAs, but the dependency on the mass of loaded-TRU is quite non-linear, especially in the case A100. This marked nonlinearity can be interpreted as the complicated effect of the dominant isotope Np237 in TRU nuclides, which acts as both strong poison and fertile of active Pu238 as shown in Table 2.2.

\$3. Nuclear and Thermal Characteristics of TRU-loaded Core

The influence of TRU-loading to the reactor core characteristics was quantitatively analyzed about a 1000MWe-class large LMFBR. Based on the previous survey, a typical TRU-loading core was assumed for each of the homogeneous and heterogeneous TRU-loading methods. The basic policies adopted here to set the loading patterns are: (a) as the main objective here is to understand physical mechanisms, a simple loading pattern is suitable rather than complicated one which might show better core performance, and (b) to compare the two loading methods, the total mass of loaded-TRU is set to be approximately identical between them, that is, 5% TRU in the whole core fuel in the case of the homogeneous method, and 37 target S/As which have 50% of TRU **a** in fuel in the case of the heterogeneous TRU-loading method. Figure 3.1 shows the loading pattern of TRU-loaded SIAs in the heterogeneous method. No TRUloaded core was also analyzed as the reference case.

3.1 Nuclear Characteristics

The basic nuclear characteristics was calculated by means of two-dimensional RZ diffusion-depletion theory. The three-dimensional distribution of power and burnup compositions were obtained with the combination of two-dimensional RZ and XY calculation. The plutonium enrichment of each core was determined to sustain criticality at EOEC, where the control rods are all withdrawn from the core. The characteristics evaluated here are power distribution, neutron spectrum, various reactivity coefficients, kinetics parameters and TRU transmutation rate.

Table 3.1 compares the calculated results about the nuclear characteristics of

the two TRU-loaded cores and the reference core.

a. Power Distribution

The swing of region-integrated power in the inner and outer core and blanket is almost identical among these three cores. The radial power distribution of the target S/A method , however, is quite different from that of the other cores as shown in Fig. 3.2. At BOEC, the power of TRU-loaded target S/As is very depressed compared with the reference core, while they get close at EOEC. This power swing would be a great obstacle for thermal characteristics.

b. Neutron Spectrum

Fig. 3.3 shows the spectra of neutron and adjoint flux in the three cores. It is found that the neutron spectrum gets harder and the high-energy importance becomes larger with TRU-loading mass.

c. Control Rod Worth

The control rod worth of the TRU-loaded cores decreases from the reference core by $10-20\%$. This may be caused by the hardening of neutron spectrum.

d. Burnup Reactivity Loss

Due to the production of Pu238 from Np237 in TRU, the burnup reactivity in the TRU-loading cores is 40% smaller than that of the reference core.

e. Reactivity Coefficients

The Doppler coefficients of the TRU-loading cores are $20 \sim 30\%$ smaller in absolute value, and the sodium density reactivity coefficients are 50% larger than the reference core because of the the spectrum hardening.

f. Kinetics Parameters

Similarly, the values of prompt neutron life time in the TRU-loaded cores become shorter by 20% than that of the reference core.

g. TRU Transmutation Rate

Table 3.2 summarized the TRU inventory and transmutation rate of these three cores. Both TRU-loaded cores can transmute TRU by 11-12% (about 180kg/cycle , and there is no differences between the two TRU-loading methods. In detail, Np and Am were eliminated after depletion, but Cm increased on the contrary. This would be a problem from viewpoint of the fuel handling and transportation in fuel cycle.

3.2 Thermal Characteristics

The coolant flow distribution was optimized for the no TRU-loaded reference core, and the influence of TRU-loading was evaluated about the thermal characteristics. The results were shown in Table **3.3.**

There is no thermal problems about the homogeneous TRU-loading core, since the power distribution hardly changes from the reference core. On the other hand, the position where maximum temperature of cladding occurs in the heterogeneous TRU-loading core moves from the reference core, and the hot spot temperature of the cladding reaches 775 degree-C which is higher than that of the reference core by 40 degrees.

Although there might be rooms for optimization of the flow distribution, the significant power swing is inevitable for the heterogeneous TRU-loading core. From the viewpoint of the thermal characteristics of the core, the homogeneous TRU-loading method is apparently superior to the heterogeneous TRU-loading method.

§4. Decay Heat and Neutron Emission from TRU-loaded Fuel S/A

The problem of the fuel handling is another aspect to be considered related to the TRU transmutation in FBR plant. Table 4.1 shows the decay heat and neutron emission rate from a fresh fuel SIA with 100% TRU-loaded, in comparison with a conventional MOX fuel S/A. In the case of 5% TRU-loaded fuel S/A, these values will be one-twentieth since these quantities are expected proportional to TRU mass.

If the TRU composition contains all of Np, Am and Cm, both the decay heat and neutron emission rate of the 100% TRU-loaded fuel S/A will exceed that of a conventional spent fuel. Especially, the neutron emission rate will be severe even in the case of a 5% TRU-loading fuel SIA. From these facts, it seems that some modifications of shielding and heat removal in the fuel handling system of a FBR plant, fuel fabrication factory and/or transportation vessels are needed in order to make the TRU transmutation feasible.

Furthermore, it is also'found in Table 4.1 the dominant element of these fuel properties is Cm. If it is possible to remove Cm from the TRU-loaded fuel, the decay heat value will decrease by one order, and the neutron emission rate by three orders. Since the dominant isotope, Cm244, has a relatively short half-life of 18 years, there might be another possibility of the fuel cycle, that is, partitioning of Cm from TRU in the reprocess and storing of Cm for a period separately. Some study will be needed to estimate the trade-off between the plant modification and the reprocessing.

\$5. Conclusions

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In the present study, the characteristics of TRU transmutation in a 1060MWeclass LMFBR with MOX fuel were quantitatively investigated to substantiate the feasibility.

There would be two typical methods transmuting TRU in the LMFBR core. One is the homogeneous TRU-loading method. It was found that the homogeneous method had nc serious penalties of TRU loading to the reactor core performance, and the TRU transmutation rate reached approximately 11% per cycle with the loading of the weight ratio of 5% TRU in MOX fuel. The amount of the TRU transmutation in the LMFBR is almost six times as much as that of the TRU production from a 1000MWe-class LWR. As another desirable property, the TRUloaded core was found to reduce the burnup reactivity loss by 40% compared with the reference core without TRU. It can be concluded that the TRU transmutation in an LMFBR has no problems from the viewpoint of the core performance, provided that the homogeneous method could be employed.

The other possible method is the heterogeneous TRU-loaded method. The weight ratio of 50% TRU fuel was assumed to be loaded in target S/As. The calculating results showed that the swing of power peaking near the target SIAs became quite severe and critical for the thermal design criteria, although the TRU transmutation rate and other reactor core performances of the heterogeneous method were roughly comparable to the homogeneous method. It seems from the present study that the heterogeneous method has quite a serious problem to be overcome and much effort would be needed to make it feasible.

As another aspect, the decay heat and neutron activity of the TRU-loaded S/A were estimated. As a result, it is recognized that the fuel handling system in an LMFBR plant, a fuel fabrication factory and/or fuel transportation vessels should be modified to treat the TRU-loaded fuel with high decay heat and strong neutron activity.

Finally, we list up the remaining items of researches to accomplish the TRU transmutation in an LMFBR,

Accumulation of data related to material property and irradiation characteristics of TRU-loaded fuel,

Improvement of nuclear data for TRU isotopes,

Establishment of reactor core design method including uncertainties of nuclear and thermal data of TRU,

Optimization of TRU-loading method in FBR cores,

and, Consideration of FBR plants to treat TRU, especially fuel handling system.

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Design parameters	Data			
1. Plant parameters				
Reactor thermal power	2517 MWt			
Coolant temperature	530/375 °C			
(Reactor outlet/inlet)				
Coolant flow rate	1.27×104 kg/s			
Operation cycle length	456 days			
Plant life time	40 years			
2. Core parameters				
\cdot Core concept	2-region homogeneous core			
Core layout	see $Fig.2.1$			
Number of core fuel S/As	175/180			
(inner/outer)				
Number of radial blanket S/As	72			
Number of control rods	18/6			
(primary/backup)				
Number of shielding	78/277			
(Stainless Steel / B_4C)				
Core diameter / core height	3.68/1.00 m			
Thickness of axial blanket	0.20/0.20m			
(upper/lower)				
3. Core fuel parameters				
·Fuel composition	$PuO_2 \cdot UO_2$			
\cdot Pu isotope ratio $(239/240/241/242)$	58/24/14/4			
·Pu enrichment(inner/outer)	15.3 /19.3 wt%			
Pattern of fuel exchange	3 dispersed batches			
4. Blanket fuel parameters				
Fuel composition	UO ₂			
·U isotope ratio(235/238)	0.3/99.7			
Pattern of fuel exchange	4 dispersed batches			

Table 2.1 Main design parameters of the 1000MWe-class reference LMFBR

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Nuclide	TRU composition discharged from LWR (1024atm/cc)	1-group cross section (barns)		Fission reaction rate vs. irradiation time (flux: 4×10^{15} nv) (Units: 1015 fissions/sec/1kg of initial TRU amount)					
		Capture	Fission	Initial	1 years	4 years	7 years	10 years	
Np237	4.55×10^{-3}	1.44	0.39	1.92	1.53	0.77	0.39	0.19	
Pu ₂₃₈	0.0	0.70	1.18	$\tilde{}$	1.04	2,35	2.06	1.45	
Pu ₂₃₉	0.0	0.47	1.82	$\ddot{}$	0.06	0.56	0.84	0.82	
Pu ₂₄₀	0.0	0.49	0.42	\blacksquare	0.01	0.04	0.07	0.10	
Pu241	0.0	0.44	2.44	\overline{a}	$\overline{}$	0.02	0.05	0.08	
Pu242	0.0	0.42	0.30	$\ddot{}$	0.02	0.05	0.06	0.05	
Am241	2.73×10^{-3}	1.33	0.34	1.02	0.80	0.38	0.18	0.09	
Am242m	6.93×10^{-6}	0.37	3.91	0.03	0.27	0.32	0.19	0.10	
Am 243	1.40×10^{-3}	0.051	0.27	0.41	0.35	0.22	0.15	0.10	
Cm242	7.95×10^{-8}	0.31	0.20	۰.	0.04	0.02	0.01	0.01	
Cm243	4.57×10^{-6}	0.23	2.60	0.01	0.02	0.03	0.02	0.01	
Cm244	4.49×10^{-4}	0.80	0.48	0.20	0.27	0.29	0.25	0.20	
Cm245	2.31×10^{-5}	0.30	2.60	0.07	0.16	0.34	0.38	0.35	
TRU total	9.17×10^{-3}			3.66	4.56	5.39	4.65	3.55	

Table 2.2 Isotopic composition of TRU fuel loaded and burnup dependency of fission reaction rate of each nuclide

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Table 2.3 Effect of loading region and loaded amount of TRU upon the transmutation characteristics (Heterogeneous TRU-loading Method)

Table 3.1 Comparison of nuclear characteristics of TRU-loaded cores

*) Values in parentheses denote relative control rod worth

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Table 3.2 Summary of TRU inventory and transmutation rate

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Table 3.3 Comparison of thermal characteristics of TRU-loaded cores									
Core	flow region number in which max. hot spot cladding temperature occurred	Assembly flow rate	Max. assembly power*	Max. hot spot cladding temperature*					
Homogeneously									
TRU-loaded core									
• Inner core(EOEC)	$\mathbf{1}$	31.9 kg/s	9.89 MW	723 °C					
			(9.65 MW)	(714 °C)					
• Outer core(BOEC)	66	29.2 kg/s	8.84 MW	727 °C					
			(8.87 MW)	(728 °C)					
Heterogeneously									
TRU-loaded core									
\bullet Inner core(EOEC)	$\overline{2}$	31.5 kg/s	9.86 MW	756 °C					
			(9.08 MW)	(724 °C)					
\bullet Outer core(BOEC)	7	27.1 kg/s	8.98 MW	775 °C					
			(8.06 MW)	(732 °C)					

Table **3.3** Comparison of thermal characteristics of TRU-loaded cores

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*) Values in parentheses denote the power for the assembly at the same position

as in the reference core

H) \sim \sim $\frac{1}{2}$ ($\frac{1}{2}$

Table 4.1 Decay heat and neutron emission rate of fresh TRU fuel

Fig. 2.1 Core layout of 1000MWe-class reference LMFBR

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Ratio of TRU amount loaded in fuel (%)

Fig. 2.2 Relationship between Amount of TRU Transmuted and Ratio of TRU Amount loaded in Fuel (Homogeneous TRU-loading Method)

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Ratio of TRU amount loaded in fuel (%)

Fig. **2.3** Relationship between Burnup Reactivity Loss and Ratio of TRU Amount loaded in Fuel (Homogeneous TRU-loading Method)

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Ratio of TRU amount loaded in the outer core region (%)

Fig. 2.4 Relationship between Power Peaking Swing and Ratio of TRU amount loaded in the Outer Core Region with Fixed TRU Amount in the Inner Region (Homogeneous TRU-loading Method)

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Fig.2.5 Power distribution of heterogeneous TRU-loaded cores

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@ Target *SIA* including TRU

 $\sim 10^{11}$ km $^{-1}$

Fig. **3.2** Radial power distribution of TRU-loaded cores

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Fig. **3.3** Neutron and adjoint spectrum in the TRU-loaded cores