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Substantial Reduction of High Level Radioactive Waste by Effective Transmutation of Minor Actinides in Fast Reactors Using Innovative Targets

Michio Yamawaki[1], Kenji Konashi[2], Koji Fujimura[1,3] and Toshikazu Takeda[1]
[1]Research Institute of Nuclear Engineering, University of Fukui
[2]Institute for Materials Research, Tohoku University
[3]Hitachi Research Laboratory, Hitachi, Ltd.,
Japan

1. Introduction

The problem of managing high-level long-lived radioactive wastes is one of the difficult issues associated with fission reactors. Long-term radiotoxicity is dominated primarily by minor actinides (MAs: Np, Am, Cm) and long-lived fission products (FPs: ^{99}Tc, ^{129}I, and so on). The potential radiotoxicity of an isotope is defined as the ratio of its radiotoxicity to the annual limit on intake (ALI) by ingestion. Calculations have shown that the potential radiotoxicity level of the waste would be reduced to that of natural uranium ore after 1,000 years provided that the MAs and long-lived FPs are removed from the waste and transmuted in reactors (Kondo & Takizuka, 1994). Extensive core design studies have been performed to assess the fast reactor (FR) capability for transmuting the MAs (Wakabayahi et al., 1995), (Kawashima et al., 1995). It was pointed out that while the MAs are to be charged to the core in FRs and an annual transmutation rate of more than 10% is possible, significant problems would be encountered in the core safety characteristics, such as the sodium void reactivity and the Doppler coefficient. Many concepts for transmutation of the MAs have been proposed using light water reactors (LWRs) (Takano et al., 1990), (Masumi et al., 1995) as well as FRs. In LWRs, the neutron fluxes are lower than those in FRs, but neutron spectra are so softer that the neutron cross sections of the MAs are larger. Thus, LWRs provide similar MA transmutation performance to FRs according to core analysis. However, more of the higher actinides are produced by MA recycling and the reactivity penalty for MA loading is larger than that in FRs.

A study on burning long-lived actinides, especially Am, was done in FRs using moderated targets in the periphery of the core or in the core region (Rome et al., 1996). The target consisted of AmO_2 surrounded by ZrH_2 or CaH_2 or mixtures thereof. On the other hand, uranium zirconium hydride (U-Zr-H) fuel has been used in TRIGA (Training Research Isotopes and General Atomics) research reactors for about forty years (Simnad, 1981) and the hydrogen absorption properties of U-Th-Zr and U-Th-Ti-Zr fuels have been examined

for their possible development as a new U-Th mixed hydride fuel (Yamamoto et al., 1995), (Yamawaki et al., 1998). 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.

In this chapter, we propose effective MA transmutation core concepts using an FR. Moderated regions containing Np and Am are constructed in a driver core region because Np and Am will be effectively transmuted into other actinides of shorter half-lives or effective Pu fuels in the high neutron flux with soft spectra. The MA-containing hydride fuel (U-MAs-Zr-H) is loaded in the target assembly of the moderated region, from the viewpoints of high hydrogen and MA concentration and MAs-H homogeneity, which mean good transmutation characteristics. The resulting Cm is partitioned and stored to decay for over 100 years, and the resulting Pu is recovered and recycled in the driver core region. Systematic parameter survey has been carried out to investigate the fundamental characteristics of MA transmutation and the core safety parameters such as sodium void reactivity in a 1,000 MWe-class fast reactor core.

In the second part of this chapter, the amount of MAs loaded in the MA burning core was increased by loading MA targets even in the radial blanket region, which increased the transmutation amount of the MA. And transmutation rate and incineration rate of MA in the MA once-through core were increased by being lengthened the irradiation period of MA targets. Based on the enhanced MA transmutation characteristics, a scenario for introducing the concept was investigated.

In the third part of this chapter, feasibility of the actinide hydride containing Np and Am as a transmutation target fuel to reduce the amount of long-lived actinides in the high level nuclear waste was studied by employing $UTh_4Zr_{10}H_x$ as a simulated actinide hydride fuel where Th is a surrogate for minor actinides. Irradiation tests of the simulated actinide hydride fuel target have been carried out in Japan Material Testing Reactor (JMTR) of Japan atomic Energy Agency (JAEA). After irradiations, both non-destructive and destructive examinations were carried out for each test. The integrity of the hydride fuel pellet was confirmed through irradiation, supporting the feasibility of the concept of hydride fuel targets.

2. Fast reactor core concepts for MA transmutation using hydride fuel targets

2.1 Actinide-hydride target and its features

Special target assembly, which contains actinide-hydrides, is considered to achieve high transmutation rate in fast reactor. The actinide-hydride target assemblies are loaded in core region of fast reactor containing mixed oxide fuels. Fast neutrons generated in the core fuel region are moderated in the actinide-hydride target assemblies and then produce high flux of thermal neutrons, which have large cross section of nuclear reaction with actinides. The target contains [237]Np, [241]Am and [243]Am with ratio of 77.4, 5.0 and 17.6, which corresponds to that for LWR UO_2 fuel irradiated with burnup of 45GWd/t. The Cm was excluded to be stored for over 100 years.

The U-(Np, Am)-Zr hydride composition has the atomic ratio of U:(Np, Am):Zr:H=1:4:10:27, which was determined based on the experimental study on the U-Th-Zr-H system

(Yamawaki et al., 1998). The Use of actinide-hydride target enables to increase amount of actinides loaded in the target assemblies. Fig.1 compares loading amount of actinides in the case of the actinide-hydride target with that in the case of mixed actinide oxide pins and $ZrH_{1.6}$ pins. The latter loading arrangement was discussed in previous report. In the condition of same densities of hydrogen atoms, the amount of loaded actinides by the former method is about six times larger than that in the latter method.

The actinide-hydride has very high hydrogen atom density, which is equivalent to that of liquid water. The moderation of fast neutrons in the actinide-hydride target has been simulated by the Monte Carlo code, MCNP (Briesmeister, 1993) with the JENDL-3.2 library. The calculations have been done for a slab of actinide-hydride with the length of 2m side and 20cm thick (Konashi et al., 2001). The source neutron with fission energy spectrum is incident at a point in center of a face of the actinide-hydride slab. The neutron spectra were averaged over the volume of each small cell (20cm side and 1cm thick), which is placed at the center of the slab. Fig.2 shows the results of the Monte Carlo code calculations. Incident fast neutrons are adequately moderated within 5cm depth of actinide-hydride target. Total flux decrease with increase of distance from the target surface, since the thermalized neutrons are effectively captured by actinide nuclides. The Monte Carlo calculation results show that the actinide-hydride target is an excellent integral target-modulator system.

(a) Hydride target (Np+Am=29kg)   (b) Oxide target and $ZrH_{1.6}$(Np+Am=5kg)

● U/MA/Zr/H=1/4/10/27   ● $(Np,Am)O_2$ ○ $ZrH_{1.6}$
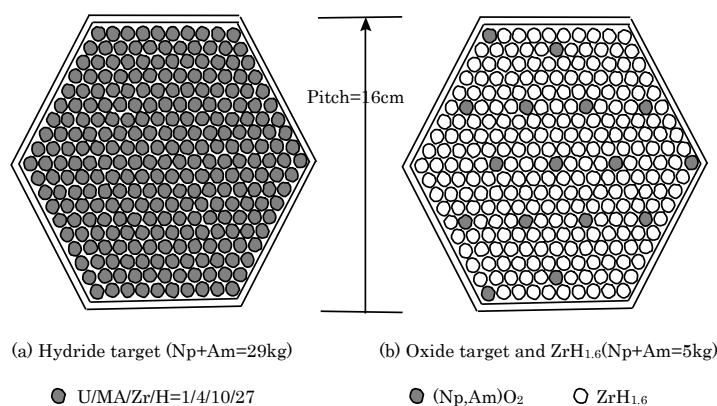
Fig. 1. Comparison of loading amount of actinides using hydride targets with that using oxide targets and $ZrH_{1.6}$ moderators (Konashi et al., 2001)
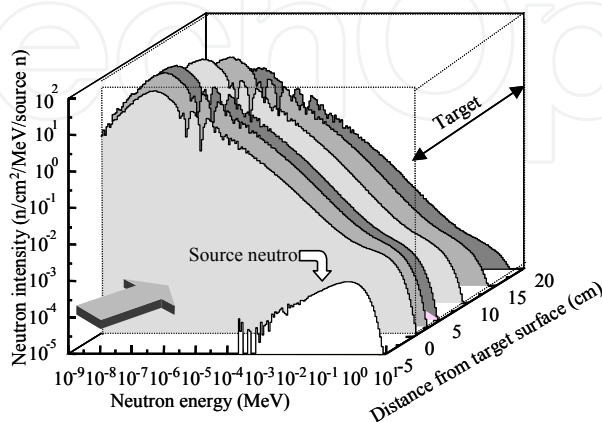

Fig. 2. Neutron energy distributions simulated by MCNP for each cell of target (Konashi et al., 2001)

2.2 Calculation conditions

Systematic survey calculations were implemented to investigate the basic characteristics of MA transmutation based on a 1000 MWe-class fast reactor core with mixed oxide fuel using hydride fuel targets. The design parameters are shown in Table 1. The core layout with 36 target assemblies is shown in Fig.3.

Core analyses were performed with the SRAC (Tsuchihashi et al., 1986) and CITATION codes using the JENDL-3.2 library. The 107-group effective microscopic cross sections in the target assembly, inner core, outer core, and blanket regions were calculated using the SRAC code. The effective cross section of the target assembly was calculated with a super cell model, which is a one-dimensional cylinder model consisting of the homogeneous target assembly surrounded by the inner core region. The effective cross sections of the other regions were calculated with the homogeneous cell model. Burnup calculations were made with the two-dimensional RZ model using the CITATION code with the 107-group microscopic cross sections. The burnup period was divided into some steps of about 180 days. The actinide transmutation chain was considered up to ^{245}Cm as shown in Fig.4. The MA transmutation characteristics such as the transmutation rate in the driver core were calculated with this model. Those in the central target assembly were calculated with this model and those in the off-center target assembly were evaluated by the following procedure. The RZ model is suitable for calculation of the MA transmutation characteristics for the central target assembly, but not the off-center target assembly. So, transmutation characteristics of the off-center target assembly were estimated based on those of the central target assembly by using the relative total neutron fluences, because the transmutation characteristics of the target are almost proportional to the total neutron fluences in the target. The total neutron fluences were calculated in the clean core without the target assembly at the corresponding positions of the center and off-center target assemblies. Strictly, these estimations need 3-dimensional calculations because the radial flux distribution varies with the irradiation. However, the total transmutation rate of all target assemblies could be estimated to a certain extent in the model described above, because the target assemblies are loaded throughout the driver core.

Item	spec.
Reactor electric power	1,000 MW
Reactor thermal power	2,600MW
Cycle length	1year (3 batch refueling)
Core concept	2 region-homogeneous
Core fuel element	PuO_2+UO_2
Blanket fuel element	Depleted-UO_2
Coolant material	Sodium
Core diameter/Height	3.00/1.00m
Pu enrichment (IC/OC)	15.8/20.2wt%

Table 1. Main design parameters of 1,000 MWe fast reactors

○ Inner core fuel   ⊖ Radial reflector
◉ Outer core fuel   © Control rod
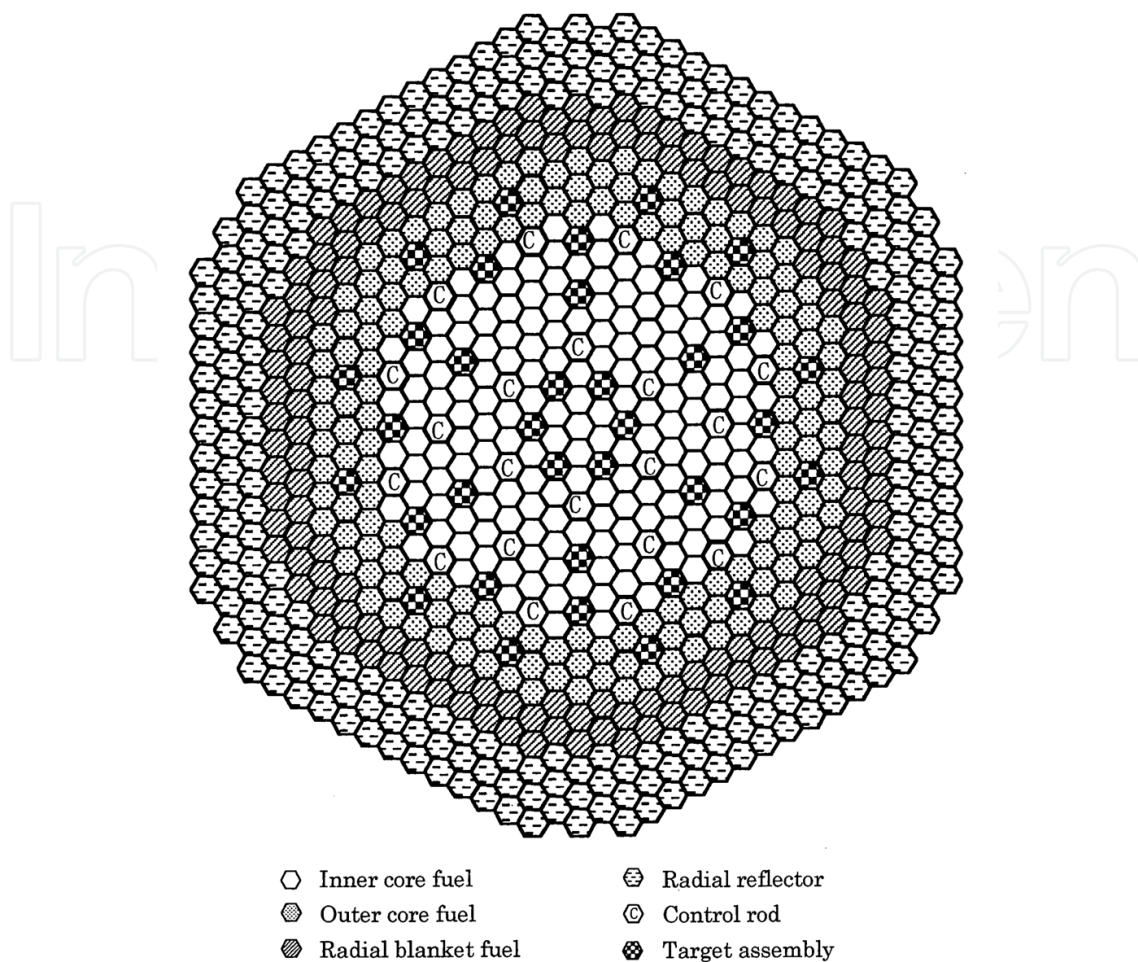▨ Radial blanket fuel   ✪ Target assembly

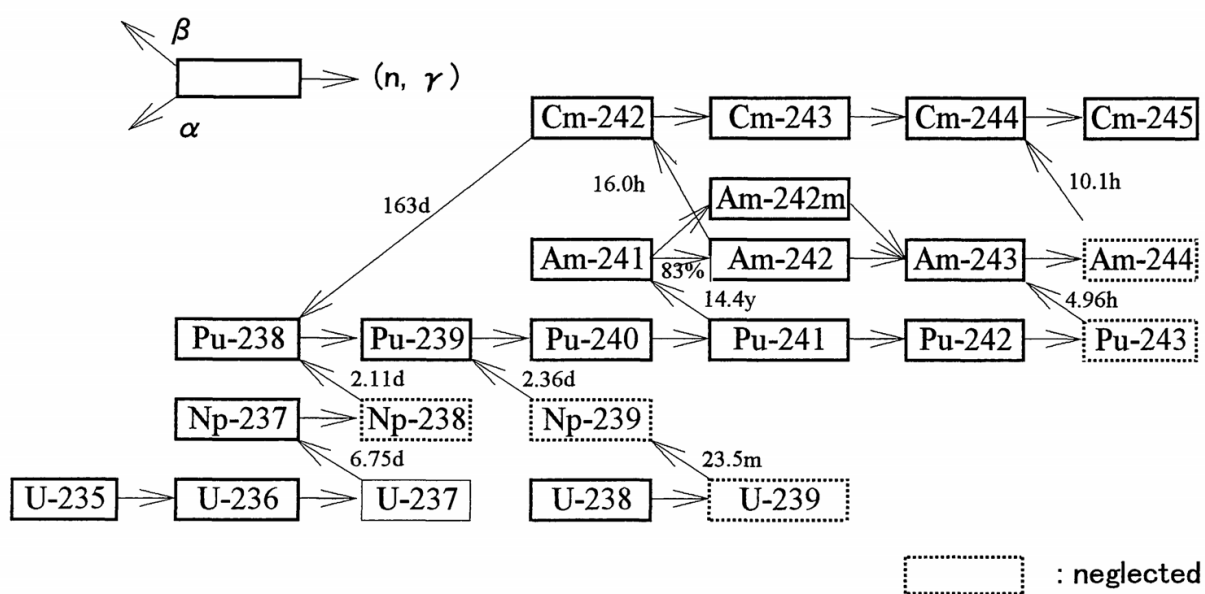Fig. 3. Core layout of 1,000 MWe fast reactor with 36 target assemblies (Sanda et al., 2000)

Fig. 4. Actinide transmutation chain employed in this chapter

In this chapter, both the MA transmutation and MA incineration were estimated. The MA transmutation is defined as the initial MAs minus the residual MAs, and the MA incineration as the incineration by fission of MAs and their daughter nuclides during the irradiation (Takeda & Yokoyama, 1997). So, transmuted MAs include incinerated MAs and the resulting Pu. The reactivity coefficients were directly calculated from the difference in multiplication factors between the reference and perturbed cores at BOC (Beginning of cycle) with the two-dimensional RZ model using the CITATION code. The target assemblies were modeled by some rings according to the number of target assemblies. Doppler coefficients in the core, blanket, and target assembly regions, and the sodium void reactivity in the core and target assembly regions were calculated.

The composition U/MAs/Zr/H in the hydride fuel was assumed to be 1/4/10/27 based on earlier study results (Yamamoto et al., 1995) for the case of the highest hydrogen and MA concentration, and the compositions were changed in order to evaluate the effect on MA transmutation characteristics. The density of U-MAs-Zr-H fuel was set at 7.15 g/cm^3 based on the preliminary measured data of U-Th-Zr-H fuel with the composition of 1/4/10/27. The amount of loaded MAs for the target assembly in this case could be about 7.6 times larger than that in the mixed MAO_2 and $ZrH_{1.6}$ target studied previously with the same numbers of hydrogen atoms. The hydride fuel target assemblies containing Np and Am were loaded in the core region. Cm was excluded, being instead stored to decay for over 100 years. The isotopic content $^{237}Np/^{241}Am/^{243}Am$ of the MAs in the target used for the reference analysis was 77.4/5.0/17.6. These values were based on the content without the cooling for the discharged LWR UO_2 fuel of about 45 GWd/t burnup, in which the Cm was assumed to be partitioned in the reprocessing process. The effect of the isotopic content of the discharged fuel with some cooling time on the transmutation was studied later.

2.3 Nuclear performance and core concepts

2.3.1 Parameter survey

(a) MA transmutation

To evaluate the fundamental MA transmutation characteristics, one MA hydride fuel target assembly was loaded in the center of the 1000 MWe-class FR core described above. Figure 5 shows the dependency of MA transmutation rate on hydrogen density in the hydride fuel target assembly loaded with 29.1kg of MAs. In this figure, 100 % hydrogen density fuel means U-MAs-Zr-H fuel with the composition of 1/4/10/27. The target assembly was irradiated in the core center for 3 years. The MA transmutation rate of U-MAs-Zr-H fuel is about two times larger than that of U-MAs-Zr fuel without hydrogen. Figure 6 compares the averaged neutron energy spectra in the inner core region and hydride fuel target assembly. Higher fluxes in the thermal and epithermal spectra are obtained in the target assembly, where the MA transmutation is better mainly because some MAs have the large resonance of fission cross sections in the epithermal energy region. Figure 7 shows the neutron flux radial distributions at typical energies near the target assembly loaded in the core center at BOC. Thermal and epithermal fluxes increase rapidly near the target assembly and have a flat distribution in the target assembly. The heavy nuclei composition and the transmutation characteristics in the target assembly with and without hydrogen loaded in the core center are listed in Table 2, where the transmutation rate means the transmuted MAs divided by

the initial MAs. Since Cm remaining in a MA-burning target is assumed to be partitioned and stored for decay for over 100 years, and the resulting Pu and the remaining Cm (mainly fissile ^{245}Cm) can be recovered and recycled in the driver core, Cm is excluded from the residual MAs in this table. The MA incineration rate is given in this table, which means the incinerated MAs by fission divided by the initial MAs. The ratio of the incineration rate to the transmutation rate as well as transmutation and incineration rates increase with the irradiation period (the operation cycle). As the ratio increases, the contribution of the MA incineration increases in the transmutation. The ratio in the hydride fuel is 0.51 for the 3-year irradiation. This is much larger than that (0.32) of the metal fuel without hydrogen, but the MLHGR (Maximum Linear Heat Generation Rate) is too large for the hydride fuel after the 3-year irradiation. In this study, the MLHGR limit of the hydride fuel target was assumed to be 500W/cm based on the metal fuel core design (Yokoo et al., 1995) in a sodium cooled FR. The MLHGR of the target in the middle of the second operation cycle exceeds the limit as shown in Table 2. This is due to the increase of fissile isotopes (^{239}Pu, ^{241}Pu, ^{242m}Am, ^{245}Cm) produced by neutron capture reactions of MAs during the irradiation and the large fission cross sections of these isotopes in the epithermal energy regions. Fast neutron fluence (≥ 0.1MeV) in the target is about 40% smaller than that in the driver core even after the 3-year irradiation, so cladding materials have significant design margins for neutron irradiation in the target. In the core design using the target, it is desirable that the MLHGR of the target be as large as possible within the limit for the higher MA transmutation, because higher neutron fluence will be obtained in the target. So, we studied shuffling of these target assemblies in each cycle and/or adjusting the composition of the MAs and hydrogen in the targets. The results are discussed in Section 2.3.2.

Next, the amount of MAs loaded in the target fuel was decreased by changing the MAs/Zr/H composition to study increase of the incineration rate, where U-free fuels were applied so as not to produce new MAs. By using these fuels, neutron fission reactions will increase because a large epithermal neutron flux is obtained in the target due to the little neutron flux depression at BOC . Table 3 shows MA transmutation and   incineration rates, and the MLHGR for 3-year irradiation for various targets loaded in the core center. The density of MAs-Zr-H fuel was set at 7.15 g/cm^3 for all cases. The MA incineration rate reaches over 90% for many cases. A core concept for the once-through option using these targets is discussed in Section 2.3.2.

The isotopic content of the MAs in the target was based on the content for the discharged LWR fuel without the cooling for the reference analysis. In order to get the effect of the isotopic content, transmutation characteristics were estimated for the central target assembly with different isotopic contents. The contents ^{237}Np/^{241}Am/^{243}Am were set at 59.6/28.2/12.2 and 41.9/49.6/8.5 for the discharged LWR fuel with 3-year and 10-year cooling times, respectively (At. Energy Soc. Jpn. Ed., 1994). As the cooling time increases, ^{237}Np decreases and ^{241}Am increases. Table 4 shows the MA transmutation and incineration rates of U-MAs-Zr-H fuel with the compositions of 1/4/10/27 and 1/4/10/0 for different isotopic contents of the MAs. This table shows the effect of isotopic contents is small for the transmutation rates, though the transmutation rates decreases a little with the cooling time. Also, the MA incineration rates increase remarkably as the cooling time increases. This is due to the fact that the fission cross section of ^{242m}Am transmuted by ^{241}Am is much larger than that of ^{237}Np in the epithermal energy region.
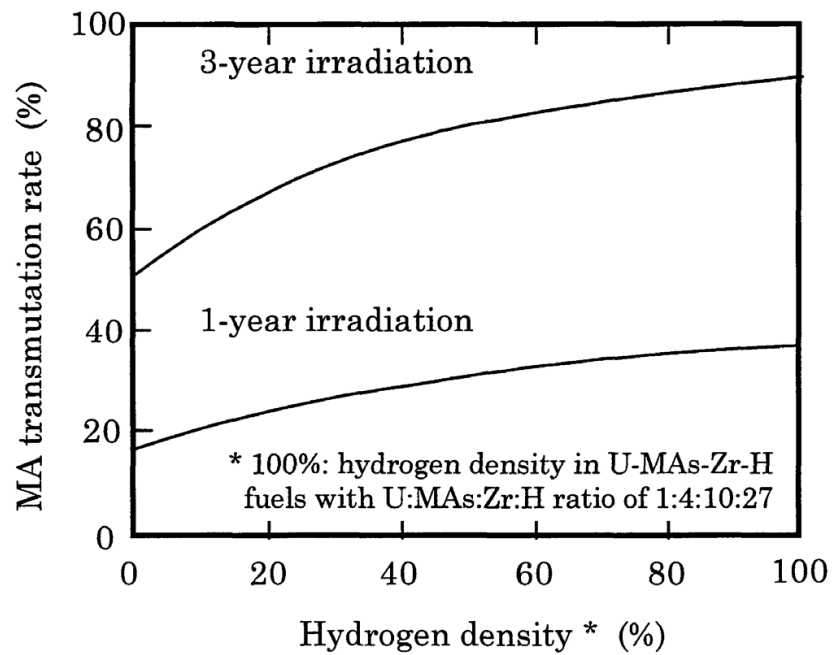
Fig. 5. Dependency of MA (Np+Am) transmutation rate on hydrogen density in a hydride fuel target assembly loaded   in core center (Sanda et al., 2000)
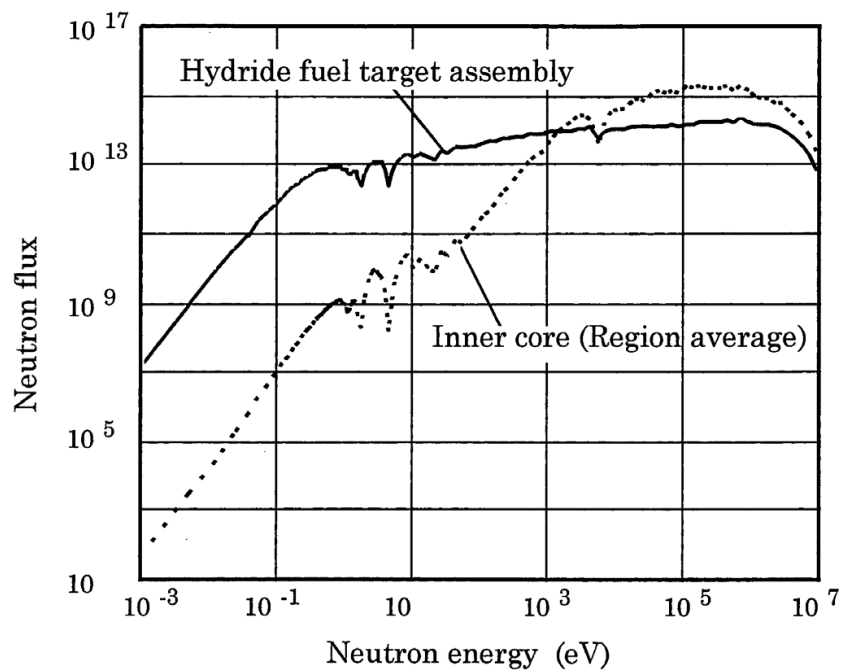

Fig. 6. Comparison of neutron energy spectra between inner core and hydride fuel target assembly (Sanda et al., 2000)
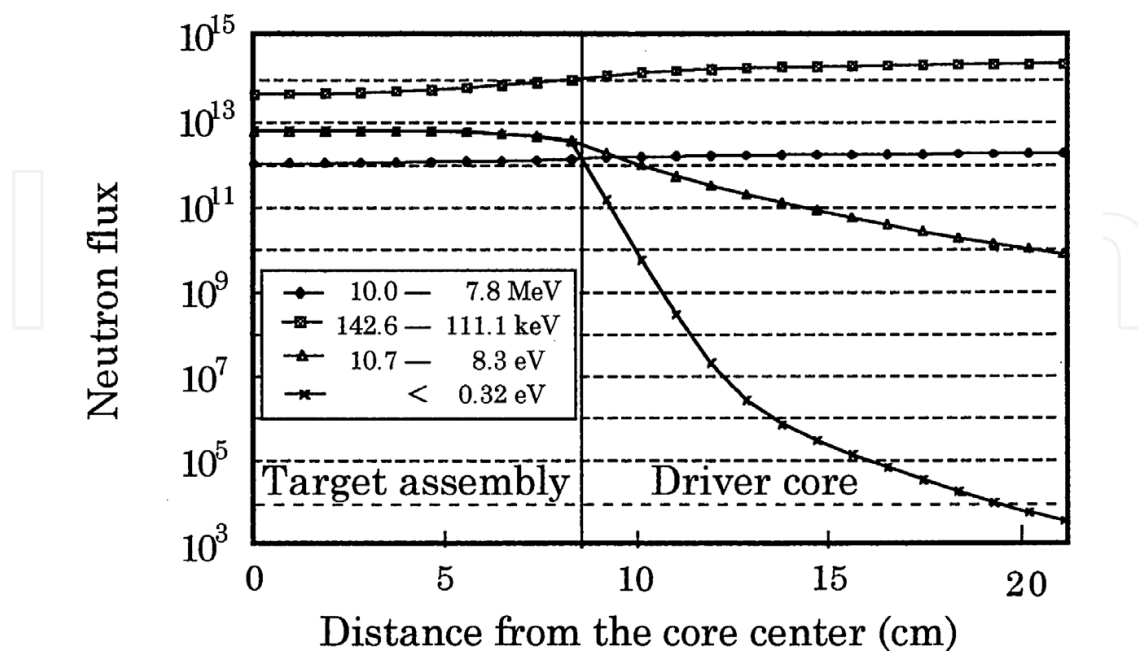
Fig. 7. Neutron flux radial distribution at typical energies near the target assembly loaded in core center
(Sanda et al., 2000)

	With hydrogen				Without hydrogen			
Irradiation period (yr)	0	1	2	3	0	1	2	3
Heavy nuclei mass (kg)								
U	7.27	6.35	5.23	3.94	7.27	6.81	6.69	6.35
^{237}Np	22.44	14.20	7.16	2.55	22.44	18.14	14.12	10.59
^{239}Np	0.00	0.01	0.01	0.01	0.00	0.00	0.00	0.00
Pu	0.00	8.08	13.04	12.97	0.00	3.51	6.25	8.03
^{241}Am	1.47	0.71	0.26	0.07	1.47	1.18	0.91	0.68
^{242}Am	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
^{242m}Am	0.00	0.04	0.02	0.01	0.00	0.03	0.04	0.04
^{243}Am	5.22	2.93	1.16	0.30	5.22	4.32	3.45	2.68
Cm	0.0	2.4	3.3	2.6	0.0	0.9	1.5	2.0
MAs (Np+Am) mass (kg)	29.1	17.9	8.6	2.9	29.1	23.7	18.5	14.0
MA transmutation rate TR (%)[†1]	0.0	38.6	70.4	89.9	0.0	18.7	36.4	52.0
MA incineration rate IR (%)[†2]	0.0	5.4	20.3	45.5	0.0	4.2	9.4	16.6
IR/TR	—	0.14	0.29	0.51	—	0.22	0.26	0.32
MLHGR[†3] of target (W/cm)	67	327	796	1,096	118	190	259	309

[†1](Initial MAs-Residual MAs)/Initial MAs
[†2]MAs and their daughters incinerated by fission/Initial MAs
[†3]Maximum Linear Heat Generation Rate

Table 2. Heavy nuclei mass and MA transmutation characteristics for two types of targets with and without hydrogen (U/MAs/Zr/H=1/4/10/27 or 0) loaded in core (Sanda et al., 2000)

These results show the incineration rates of the targets in the reference analysis are conservative when the isotopic content of the MAs in the target was based on the content for the discharged LWR fuel without cooling.

Case	Composition U/MAs/Zr/H	MAs loaded (kg/assembly)	MA transmutation rate (%)	MA incineration rate (%)	MLHGR of target (W/cm)	MLHGR[†] of core (W/cm)
A	1/4/10/27	29.1	89.9	45.5	1,096	558
B	0/1/10/15	13.6	98.6	79.3	728	710
C	0/0.7/10/15	9.5	99.3	91.7	600	790
D	0/0.5/10/15	6.8	99.7	97.6	535	851
E	0/0.5/10/10	6.8	99.3	90.4	411	700
F	0/0.1/10/15	1.4	~100.0	99.8	125	1,023

[†] Thermal power peak in fuel assembly adjacent to target assembly

Table 3. Comparison of MA transmutation characteristics for various targets loaded in core center for 3-year irradiation (Sanda et al., 2000)

Irradiation period (yr)	With hydrogen				Without hydrogen			
	0	1	2	3	0	1	2	3
MA transmuration rate (%)								
Case 1 (Cooling time=0 yr)[†]	0.0	38.6	70.4	89.9	0.0	18.7	36.4	52.0
Case 2 (Cooling time=3 yr)[†]	0.0	38.6	71.9	91.3	0.0	18.5	36.0	51.6
Case 3 (Cooling time=10 yr)[†]	0.0	38.6	72.9	92.7	0.0	18.2	35.7	51.2
MA incineration rate (%)								
Case 1 (Cooling time=0 yr)[†]	0.0	5.4	20.3	45.5	0.0	4.2	9.4	16.6
Case 2 (Cooling time=3 yr)[†]	0.0	10.0	30.6	57.0	0.0	5.7	12.8	21.5
Case 3 (Cooling time=10 yr)[†]	0.0	13.6	40.0	67.5	0.0	7.0	15.8	25.8

[†] Isotopic content $^{237}Np/^{241}Am/^{243}Am$ of MAs in targets
   Case 1: 77.4/5.0/17.6 (reference),   Case 2: 59.6/28.2/12.2,   Case 3: 41.9/49.6/8.5

Table 4. MA transmutation and incineration rates for two types of targets (U/MAs/Zr/H=1/4/10/27 or 0) with different isotopic contents loaded in core center (Sanda et al., 2000)

(b) Reactivity coefficient

As the MA enrichment increases in an FR, the neutron energy spectrum gets harder and the fast fission rate increases. Therefore, safety characteristics deteriorate; i.e. the value of the sodium void reactivity becomes more positive and the absolute value of the Doppler coefficient decreases. For our core concepts, the spectrum gets softer due to the hydride fuel targets and safety characteristics are improved. Figures 8 and 9 show the relations between reactivity coefficients and the number of target assemblies composed of U/MAs/Zr/H fuel with a composition of 1/4/10/27. In the figures, the corresponding reactivity coefficients are described for an FR core loaded with the same amount of MAs homogeneously. For a reference core without MAs, sodium void reactivity is 5.0 $ in the core region, and Doppler coefficient is -1.0×10^{-2} Tdk/dT in the core and blanket regions. In the proposed cores, the positive sodium void coefficient decreases with the number of target assemblies. In contrast, the negative Doppler coefficient shows little dependency on the number of target assemblies and the relative value slightly increases with the number. This little dependency is due to compensation from the positive effect of spectrum softening and the negative effect of higher Pu enrichment in the driver core by loading the target assemblies, because higher Pu enrichment means less ^{238}U, the main contributor of Doppler coefficient. However, these figures show safety characteristics can be improved by loading MA hydride targets, compared with an FR core loaded with MAs homogeneously.

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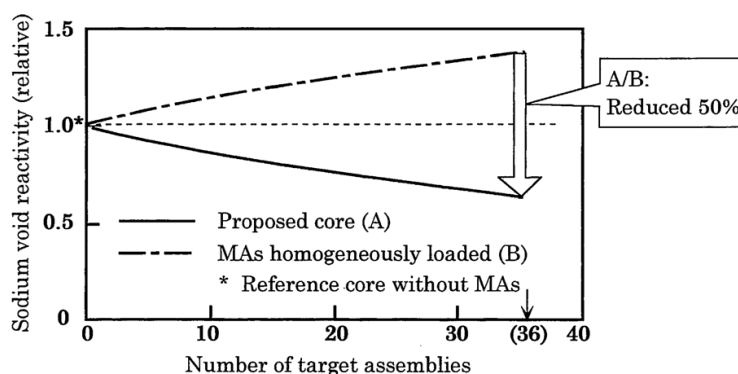173

Fig. 8. Relation between sodium void reactivity target assemblies (Sanda et al., 2000)
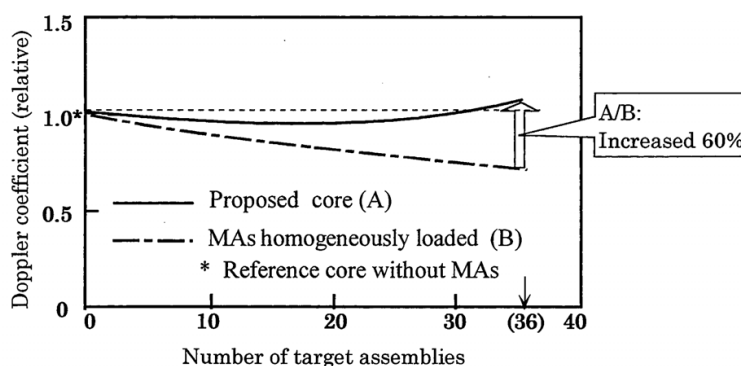

Fig. 9. Relation between Doppler coefficient and number of and number of target assemblies (Sanda et al., 2000)

(c) Thermal power peaking for fuel assembly adjacent to target assembly

Thermal power peaking is generated in the fuel assemblies adjacent to the target assembly by epithermal neutrons coming from the target. When the amount of MAs loaded is small in a target, the thermal power peaking causes a problem which must be considered in the core design as neutron flux depression is small in the target assembly due to fewer MA neutron capture reactions. In order to decrease the thermal peak, we studied an assembly composed of hydride fuels surrounded by ^{99}Tc, which is one of the long-lived FPs and makes a large contribution to long-term radiotoxicity. Figure 10 shows the radial power distribution near the target with and without ^{99}Tc after the 3-year irradiation in the case of target assembly composed of MAs/Zr/H fuel with the composition of 0.7/10/15. The density of this fuel was set at 6.0 g/cm^3 based on TRIGA type fuel data of U-Zr-H fuel with the composition of 0.7/10/27 (Simnad, 1981). The density of ^{99}Tc was set at the theoretical density of 11.5 g/cm^3. While the big thermal peak in the adjacent fuel assemblies is generated for the target assembly without ^{99}Tc rods, the MLHGR of the adjacent fuel assemblies comes within the limit by replacing two layers of hydride fuel rods with ^{99}Tc rods as shown in Fig. 10. The MLHGR decreases with the incineration rate of MAs for the target assembly. This is due to decreasing the amount of the fissile nuclei by fission after the 3-year irradiation with a larger fission incineration rate. Figure 11 shows the radial power distribution near the target with 2 layers of ^{99}Tc rods for different irradiation periods. In the target assembly, the LHGR (Linear Heat Generation Rate) increases in the central region up

to the 2-year irradiation. This is due to the increase of fissile nuclei such as [239]Pu, [241]Pu, [242m]Am, and [245]Cm, which are produced by neutron capture reactions of MAs with irradiation and have very large fission cross sections in the thermal and epithermal energy region. After the 2-year irradiation the LHGR decreases in the target assembly. This is due to the decrease of the fissile nuclei by the fission reaction. The transmutation and incineration rates in the discharged target decrease to 98.4 % and 82.4 % from 99.4 % and 92.6 %, respectively, by using 2 layers of ^{99}Tc rods. These target assemblies can be applied to the core concept for the MA once-through option due to the high incineration rate described below.
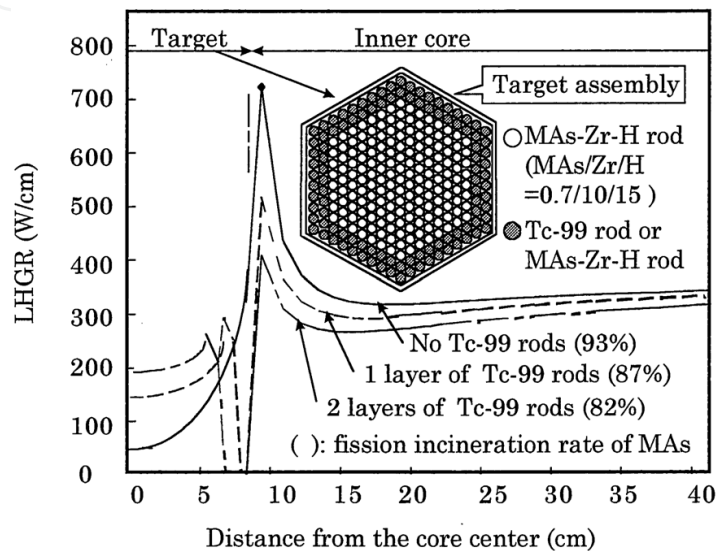
Fig. 10. Radial power distribution near target fuel assembly with and without ^{99}Tc rods after the 3-year irradiation (Sanda et al., 2000)
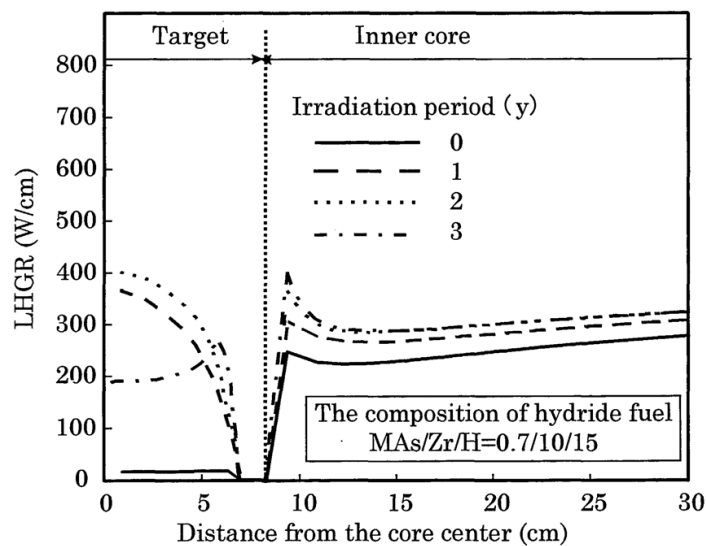
Fig. 11. Radial power distribution near target fuel assembly with 2 layers of ^{99}Tc rods for different irradiation periods (Sanda et al., 2000)

2.3.2 Proposed core concepts

We proposed two core concepts using MA-containing hydride fuel targets based on the survey results described above. One is for the MA burner option with Pu multi-recycling to transmute a larger amount of MAs, and the other is for the MA once-through option. The merit of the MA once-through option is that it can avoid some problems such as spent targets transporting and reprocessing, and new targets fabrication.

(a) MA burner option with Pu multi-recycling

An MA burner core was studied using target assemblies composed of U/MAs/Zr/H fuel with the composition of 1/4/10/27 without Tc. The core layout was shown in Fig.3, in which 36 target assemblies were loaded on 4 rings. The MA transmutation and the MLHGR of the targets were evaluated for two cases. The target assemblies were discharged after one and two cycle irradiations for the first and second cases, respectively. The MLHGR is much smaller than the limit for the first case, and nearly reaches the limit for the second. More of the MAs in the target assemblies on the inner ring are transmuted than those on the outer ring. The target assemblies were shuffled every irradiation cycle for the second case; the assemblies on the first and second rings were moved to the fourth ring at the beginning of the second cycle to obtain similar MA transmutation every irradiation cycle. This contributes to flattening the radial power distribution for each cycle. Table 5 shows heavy nuclei mass and MA transmutation of target assemblies for the proposed burner core concept. The transmutation and incineration rates are 30.6 and 3.7% in the discharged target for the first case, and 57.3 and 14.1% for the second case. For this core concept, the MAs produced in about 14 and13 LWRs can be transmuted every year in the target, respectively, for the first and second cases. Table 6 compares MA transmutation and reactivity coefficients in various core concepts. The MA transmutation and safety characteristics of the proposed cores are improved in comparison with conventional FR cores loaded the same amount of MAs.

Figure 12 shows a fuel cycle including the proposed MA burner and the heavy nuclei flow. In the MA burner core, Np and Am of the hydride fuel target are provided from reprocessing of the spent fuel of LWRs (FRs in future) and the MA burner core, and the discharged target. The reprocessed Pu is recovered and recycled in the driver core. As an option, long-lived FPs such as [129]I and ^{99}Tc are partitioned and mixed with a hydride such as $ZrH_{1.6}$. These are loaded in the radial blanket region in order to be transmuted into stable nuclides. When the radial blanket assemblies adjacent to the driver core region were replaced by ^{99}Tc target assemblies composed of ^{99}Tc and zirconium hydride pins with the same volume ratio, the annual transmutation mass is about 75kg /y (0.9%/y) which is equivalent to the amount produced by about 2 LWRs and a self-generated one.

(b) MA once-through option

An MA incineration core for the MA once-through option was studied using target assemblies composed of MAs/Zr/H fuel with a composition of 0.7/10/15 surrounded by two layers of ^{99}Tc as shown in Fig.10. The core layout is shown in Fig.3, and was the same as in the MA burner option. These target assemblies were shuffled every irradiation cycle and discharged after a 3-cycle irradiation within the limit of MLHGR. During shuffling the target assemblies on the inner ring were moved to the outer ring at each cycle to obtain a similar

MA transmutation for each target. Table 5 also includes heavy nuclei mass and MA transmutation characteristics of target assemblies for this proposed core concept. The transmutation rate and incineration rate are 93.0% and 63.9% in the discharged targets, respectively. For this core concept, the MAs produced in about 2 LWRs can be incinerated every year in the targets. For ^{99}Tc pins in the target assemblies, the annual transmutation mass is about 60kg /y (4.1%/y) which is equivalent to the amount produced by about one LWR and a self-generated one.

Similar core concepts are realized using the mixed MA and $ZrH_{1.6}$ target assemblies, but the MA transmutation characteristics would be inferior to those for the hydride fuels due to lower hydrogen atom concentration in the assembly as described in sec. 2.2, especially in the MA burner option.

Discharged cycle	Burner core				Incineration core	
	1 (1-year irradiation)		2 (2-year irradiation)		3 (3-year irradiation)	
	Charged	Discharged	Charged	Discharged	Charged	Discharged
Heavy nuclei mass (kg)						
U	261.68	235.51	130.84	102.53	0.00	0.00
^{237}Np	807.92	572.69	403.96	181.36	45.23	3.64
^{239}Np	0.00	0.24	0.00	0.17	0.00	0.00
Pu	0.00	230.63	0.00	197.80	0.00	18.61
^{241}Am	52.88	31.13	26.40	8.00	2.97	0.06
^{242}Am	0.00	0.04	0.00	0.02	0.00	0.00
^{242m}Am	0.00	1.12	0.00	0.54	0.00	0.01
^{243}Am	187.80	122.45	93.90	34.01	10.55	0.41
Cm	0.00	68.23	0.00	52.46	0.00	3.10
MAs (Np+Am) (kg)	1,048.6	727.7	524.3	224.1	58.8	4.1
MA transmutation (kg)	—	320.9	—	300.2	—	54.7
MA incineration (kg)	—	38.6	—	74.1	—	37.6
MA transmutation rate (%)	—	30.6	—	57.3	—	93.0
MA incineration rate (%)	—	3.7	—	14.1	—	63.9
MLHGR of target (W/cm)	65	227	65	500	16	286

Table 5. Heavy nuclei mass and MA transmutation characteristics of targets for burner and incineration core concepts (Sanda et al., 2000)

Type of MA loading	Conventional fast reactor core			Burner core	Incineration core
	Reference (No MAs)	Homo-geneous 3.8% MAs	Hetero-geneous MA target	36 MA hydride targets (U/MAs/Zr/H =1/4/10/27)	36 MA hydride targets (MAs/Zr/H =0.7/10/15, with 2 layers of ^{99}Tc)
MA loaded (kg)	0	1,050	1,050	1,050	176
Annual transmuta-tion rate (%/yr)	(—)	14[*1]	11[*1]	28[*2,†] (31)[††]	26[*3,†] (29)[††]	16[*1,†] (31)[††]
Sodium void reactivity (relative)	1.0	1.4	1.2	0.7	0.7
Doppler coefficient (relative)	1.0	0.7	0.8	1.1	0.9

[*1]: 3-year irradiation,   [*2]: 1-year irradiation,   [*3]: 2-year irradiation,   [†]: Driver core+Targets,   ( )[††]: Targets

Table 6. Comparison of MA transmutation characteristics and reactivity coefficient (Sanda et al., 2000)

2.4 Summary for section 2

Fast reactor core concepts were studied to reduce the long-term radiotoxicity of nuclear waste by using minor actinides (MAs) in the form of hydride fuel targets. The hydride fuel target assemblies containing Np and Am were loaded in the core region, and the Cm was partitioned and stored to decay for over 100 years. A systematic parameter survey was carried out to investigate the fundamental characteristics of MA transmutation and reactivity coefficients in a 1,000 MWe-class FR core. Results showed safety characteristics could be improved by loading MA hydride targets, as compared with the FR core loaded with MAs homogeneously. Two core concepts were proposed using 36 target assemblies. One was the MA burner core to transmute a larger amount of MAs by neutron capture and fission combined with Pu multi-recycling in FRs, where the MAs produced in about 13 LWRs could be transmuted every year with 58% transmutation rate (14% incineration rate) in discharged targets. The other was the MA once-through core to incinerate a small amount of MAs by fission, the MAs produced in about 2 LWRs could be incinerated every year with about 64% fission incineration rate (93% transmutation rate) in discharged targets. This study showed that the proposed core concepts using MA-containing hydride fuel targets have great potential to achieve good transmutation of MAs while providing the improved safety characteristics of an FR core.

MA transmutation characteristics using hydride fuel targets was studied from the viewpoint of reactor physics. However, future attention should be given to the following.

i.   Hydrogen is dissociated from hydride fuels at higher temperatures. When the fuel temperature rises under the transients, positive reactivity would be inserted by hydrogen dissociation in an FR. We estimated its effect roughly. The hydrogen dissociation pressures are about 0.1 and 16 atm at 700 and 1,000 deg. for $U-ZrH_{1.6}$ fuel, respectively Simnad, 1981. As a result, the hydrogen composition decreases from 1.6 to 1.5969 (0.2 % decrease) at 1,000 deg. under the assumption that the volume and temperature of the fuel are the same as those of the gas plenum in a fuel pin. And, the 0.2 % hydrogen decrease corresponds to about an inserted reactivity of 2 ¢ for the proposed core with 36 target assemblies composed of U/MAs/Zr/H fuel with a composition of 1/4/10/27. So, the hydrogen dissociation effect under the transients is expected to be little.

ii.   Hydrogen penetration increases in the stainless steel cladding material of the fuel pin at higher temperatures. Countermeasures, such as increasing the coolant flow rate for target assemblies so as to decrease the cladding temperatures and internal coating of the cladding material would be effective. Silicon carbide has a very good hydrogen retention capability. It is being used very successfully in HTGR (High Temperature Gas-cooled Reactor) fuel as the cladding against hydrogen (tritium) penetration even up to temperatures exceeding 1,000 deg. (Greenspan, 1997).

iii. It is necessary to develop new MA burning target materials, which are stable under high temperatures and irradiation conditions. The $U-ZrH_{1.6}$ fuel for the TRIGA reactor was reported by General Atomics to be stable at steady-state temperatures up to 700 deg. Under transient conditions the $U-ZrH_{1.6}$ fuel can withstand temperatures at high as 1,000 deg. (Simnad, 1980). The U-Th-Zr-H fuels have been fabricated and examined for thermal and mechanical properties through a preliminary irradiation examination (Yamawaki et al., 1998). These experiences are expected to contribute to development of Np and Am hydride fuels.
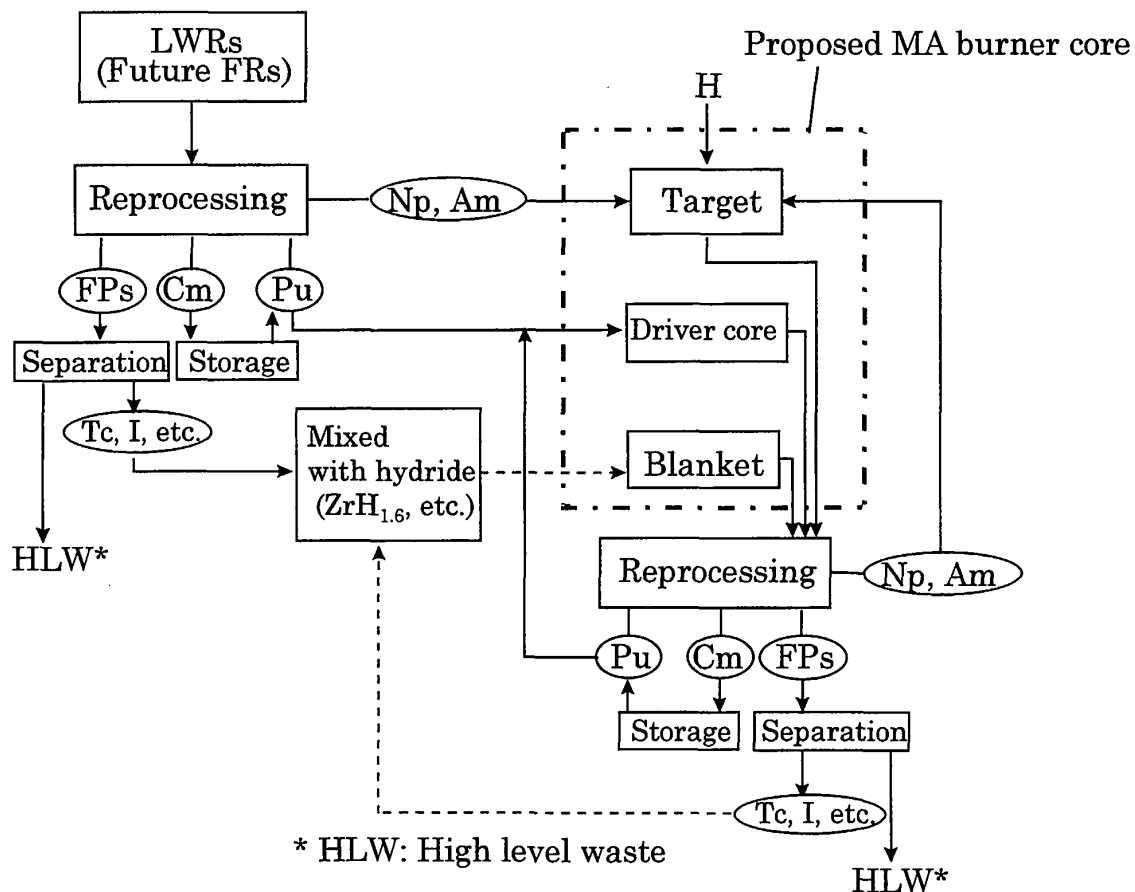
Fig. 12. Fuel cycle including the proposed MA burner core and heavy nuclei flow (Sanda et al., 2000)

3. Enhancement of transmutation characteristics of the MA transmutation fast reactor core concept using hydride fuel targets and its introduction scenario

3.1 Enhancement of transmutation characteristics

Two types of core concepts for the MA burning FR core using hydride fuel targets were proposed in Sec. 2. One is for an MA burner option to transmute a larger amount of MAs by neutron capture and fission combined with Pu multi-recycling in FRs. The other is for an MA once-through option to completely incinerate a smaller amount of MAs by fission. These concepts have great potential to achieve good transmutation of the MAs with improved of safety characteristics in the FR core.

In this study we try to enhance the MAs transmutation characteristics of both types of MA burning FR core using hydride fuel targets.

3.1.1 MA burner option with Pu multi-recycling

The U/MAs/Zr/H composition in the hydride fuel is assumed to be 1/4/10/27. In this option, a large amount of Pu is produced by the neutron capture reaction of MAs. In sec. 2,

the MLHGR limit of the hydride fuel target was assumed to be 500W/cm based on the metal fuel core design. Therefore the target assembly loaded in the core fuel region should be discharged after a 2-year irradiation. The MAs produced in about 13 LWRs could be transmuted every year by loading 36 target assemblies in the 1,000MWe-class FR core. The average transmutation rate in discharged targets was 58%. The main design parameters of 1,000 MWe-class FR were shown in Table 1.

First irradiation test with simulated actinide-hydride target has been done in JMTR. The U/Th/Zr/H type target material was used for the irradiation test (Konashi et al., 2000), since the hydrogen chemical potential of Th hydride is close to that of Np and Am hydrides. The MLHGR limit was set less than 200 W/cm in order that an enough thermal margin should be ensured to the actinide-hydride fuel. It is important to show the potential of the actinide-hydride as the target for MA transmutation reactor within a feasible condition. For that sake, a new loading scheme was proposed as follows.

All target assembly which are irradiated in the core fuel region within a year, are shuffled to the first row of the radial blanket (RB) region. As 72 target assemblies can be loaded in the first row of the RB region, 36 target assemblies can be irradiated within 2 years. As a result, 36 target assemblies will be discharged after a 3-year irradiation. Increase in number of the target brings the decrease in number of the core fuel, which will increase MLHGR of the core fuel within the condition shown in Table 1. Therefore, the number of target loaded in the core fuel region was limited to 36 according to sec. 2. Figure 13 shows the dependency on irradiation period of MA transmutation and MLHGR in a typical target assembly. In the RB region, the MA transmutation rate becomes small because the neutron flux level is smaller than in the core region. The average MA transmutation rate is 47%. On the other hand, the MLHGR becomes very small, which increases the margin of hydride fuel temperature. The total number of targets loaded in the core is 3 times more than the previous case, which increases the MA transmutation amount. As a result, the MA produced in about 21LWRs can be transmuted in the 1,000MWe-class FR core (Table 7).
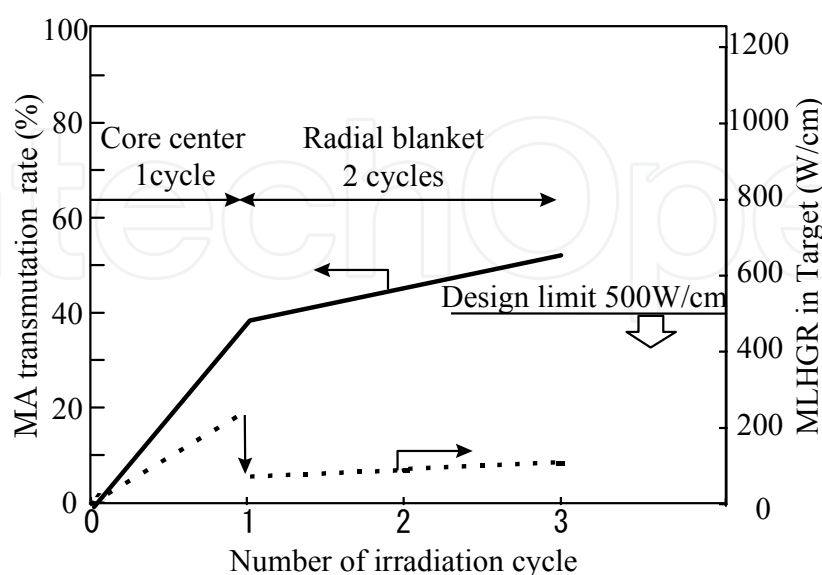

Fig. 13. Burnup characteristics of hydride fuel target of MA burner with Pu multi-recycling core (Fujimura et al., 2001)

Item	Unit	LWR	FBR[*1]		
			Without Targets	MA burner	MA once-through
Output	GWe	1	←	←	←
RB	—	—	2 layers	None	2 layers
BR	—	—	1.2	0.9	~ 1.0
Pu loaded	t/y	0	1.7	2.7	2.7
Pu produced	t/y	0.2	0.3	-0.3	~ 0
MA loaded	t/y	0	0.2	1.1	0.03
MA transmuted	t/y	-0.02	0.0	0.50 (47%)[*2]	-0.03 (~100%)[*3]
	[*4] LWR/y	-1	~ 0	21	1.3

*1 MAs self-generated in core fuel are recycled to core fuel,
*2 Including transformation of nuclei, *3 Incineration rate by fission,
*4 Divided by annual MAs produced in a LWR of same electric output

Table 7. Mass balance data of each type of core (Fujimura et al., 2001)

3.1.2 MA once-through option

In this option, the amount of MAs loaded in a target is small and the aim is for almost all of the MAs to be incinerated completely, including Pu which is produced by MAs transformation with neutron capture reaction. This option is free from reprocessing of spent target fuel. The target assembly is composed of MAs/Zr/H fuel with a composition of 0.7/10/15 according to sec. 2. The reason for no U is to minimize production of MAs. Hydride fuels are surrounded by two layers of ^{99}Tc rods to prevent thermal peaking at adjacent fuel rods. Figure 14 shows the dependency on irradiation period of MA transmutation rate and incineration rate in a typical target assembly. In the previous section, all target assembly in the MA once-through core was discharged after 3-year irradiation with core fuel assembles. Core average MA transmutation rate was 93% and incineration rate was 64%.

In this section, we let the irradiation period of a target assembly be twice as that of the core fuel to increase the MA transmutation rate and incineration rate. Both the MA transmutation rate and incineration rate is almost 100% after a 6-year irradiation as shown in Fig.14. In this option, MLHGR of a target will not be over its design limit because the inventory of MAs and Pu produced from transformation of MAs is very small. The merit of this option is that it avoids such problems as spent target transport, reprocessing and new target fabrication. Figure 15 shows the two options of the MA burning FR core using hydride fuel targets which we used to investigate an introduction scenario.
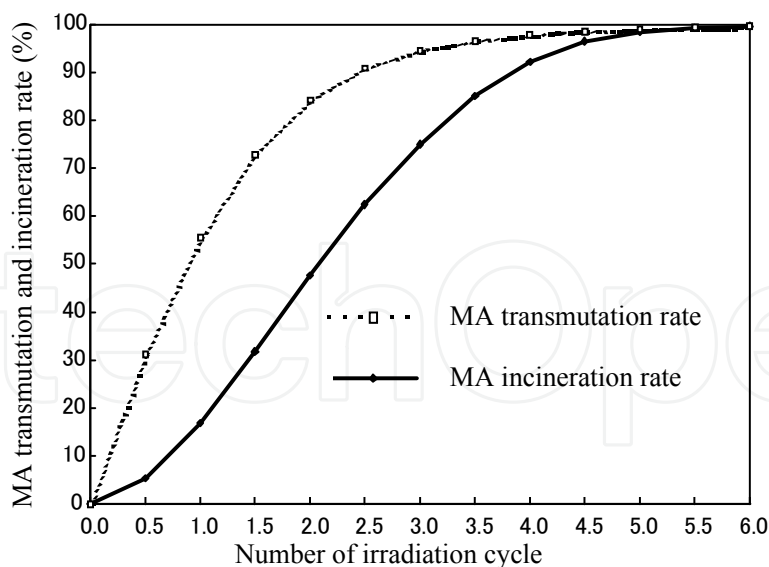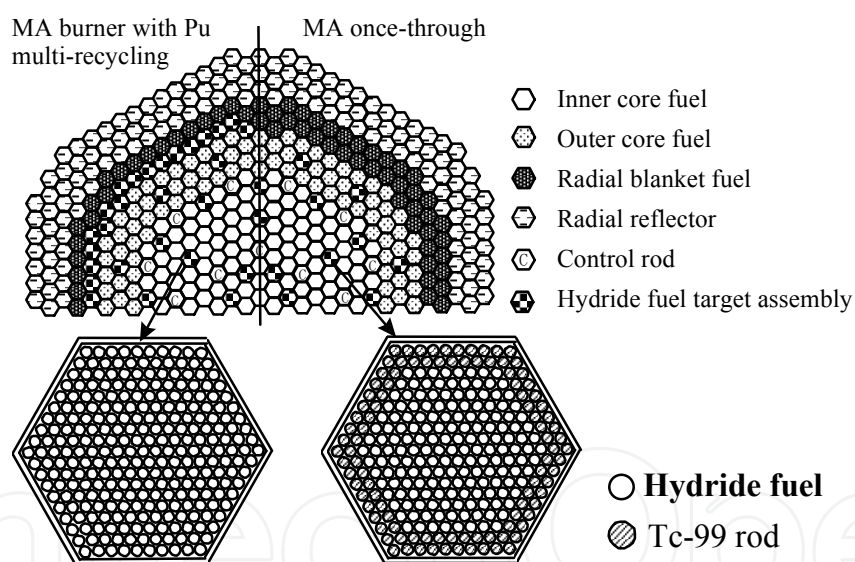
Fig. 14. Burnup characteristics of hydride fuel target of MA once-through core (Fujimura et al., 2001)

| Option	Hydride	Target position	Feature
MA burner	U/MA/Zr/H 1/4/10/27	Core(1 cycle)→ RB (2 cycles)	Relatively large mass of MAs is loaded
MA once-through	MA/Zr/H 0.7/10/15	Core (6 cycles)	Small amount of MAs completely incinerated →Tc-99 transmu-tation rate (～4%/y)

Fig. 15. Two options of MA burning fast reactor core using hydride fuel targets (Fujimura et al., 2001)

3.2 Investigation of introduction scenario

By introducing two options of the MA burning FR core using hydride fuel targets in the early stage of the fast breeder reactor period, the following scenario is feasible. The MA burning FR core using hydride fuel targets will be replaced by a self-generated MA transmutation FR core without the target after most of the MAs produced in the LWR are transmuted. In the reprocessing of the spent fuel of the self-generated MA transmutation FR core, MAs are not separated from Pu.

3.2.1 Nuclear power capacity and installation scenario of FBR

Nuclear power capacity and installation scenario of the FBR (Fast Breeder Reactor) are set with the high projection case as the reference (Wajima et al., 1996). Nuclear power capacity between the years 2010 and 2,100 is extrapolated based on its value of 45GWe in 2,000 and seventy GWe in 2010 which were given in the 1994's Long-Term Program for Development and Utilization of Nuclear Energy of Japan. Capacity growth rate between 2011 and 2050 is fixed at 14GWe per year which is equivalent to the minimum growth rate per 10 years between 1971 and 2010. The growth rate between 2051 and 2100 is fixed at 10 GWe/yr. Seventy GWe in 2010 is consistent with the value shown by the Electricity Industry Council's report for Japan given in July 1998. Above nuclear power capacity well coincides with that supposed in the paper (Hamamoto et al., 2001) which discussed the MOX cores for effective use of Pu in LWRs. The following installation projection of a large FBR is assumed. Installation of the FBR plant will start in 2010 when commercial LWRs will be replaced and installation of two commercial sized FBR plants should be completed prior to 2030. The FBR will be commercialized after 2030. The FBR plant installation rate is assumed to be relatively large in which the utilized amount of natural uranium ore in Japan will not be over 10 % of the world's ultimate resources (about 1.7 Mt). Figure 16 shows the yearly dependency of the nuclear power capacity for high projection case in Japan.
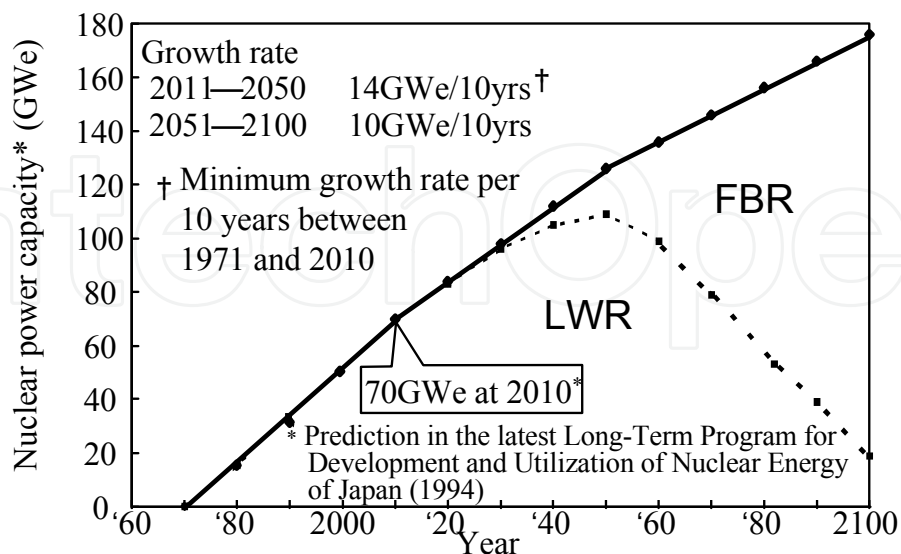
*High projection case - T. Wajima et al.: Journal of Japan Nuclear Society, Vol.38, No.2, 133(1996).

Fig. 16. Yearly dependency of the nuclear power capacity in Japan (Low projection case) (Fujimura et al., 2001)

Nuclear power capacity and installation scenario of the FBR with the low projection case as the above reference was also set. In this scenario, capacity growth rate after 2070 will be fixed at zero. The FBR will be installed after 2030 and commercialized at 2050. Figure 17 shows the yearly dependency of the nuclear power capacity for low projection case in Japan.
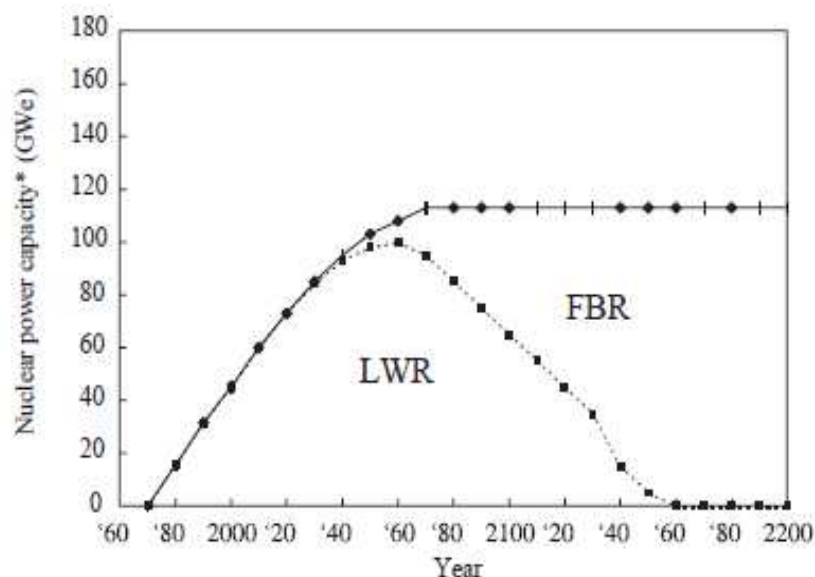
** Low projection case - T. Wajima, et. al.: Journal of Japan Nuclear Society, Vol.38, No.2, 133 (1996).

Fig. 18. Yearly dependency of the nuclear power capacity in Japan (High projection case) (Fujimura et al., 2001)

3.2.2 Fuel cycle flow

Fuel cycle flow of each reactor being considered in this capter is shown in Fig. 18. All the Pu and MAs being extracted from reprocessing of LWR spent fuel are assumed to be utilized and transmuted by the FBR. No utilization of Pu in the LWR is assumed. In this study, all FBRs which will be installed after the residual amount of MAs from LWR spent fuel are transmuted, are assumed to have a self-generated MA transmutation FR core. MAs included in the spent fuel are not separated from Pu . They are utilized as fuel and transmuted. Concentration of the MAs in heavy metal is about 1-2 % in the equilibrium cycle core which does not affect nuclear performance of the core so much. On the other hand, it was shown that a 1,300MWe commercial size FBR can transmute self-generated six long-lived FPs (^{79}Se, ^{99}Tc, ^{107}Pd, ^{129}I, ^{135}Cs, ^{151}Sm) in the radial blanket and part of the axial blanket (Kobayashi et al., 1997). Fuel cycle flow of the core fuel of the MA burning FR using hydride fuel targets is the same as that of the self-generated MA transmutation FR core. And all MAs from the LWR spent fuel are loaded in hydride fuel targets. About half of the MAs will survive in a spent target fuel in the MA burner option core and they will be reloaded in the reactor as targets. In the once-through option core, most of the actinides are incinerated by fission when being discharged. Therefore a simple reprocessing will be needed for extracting FPs.
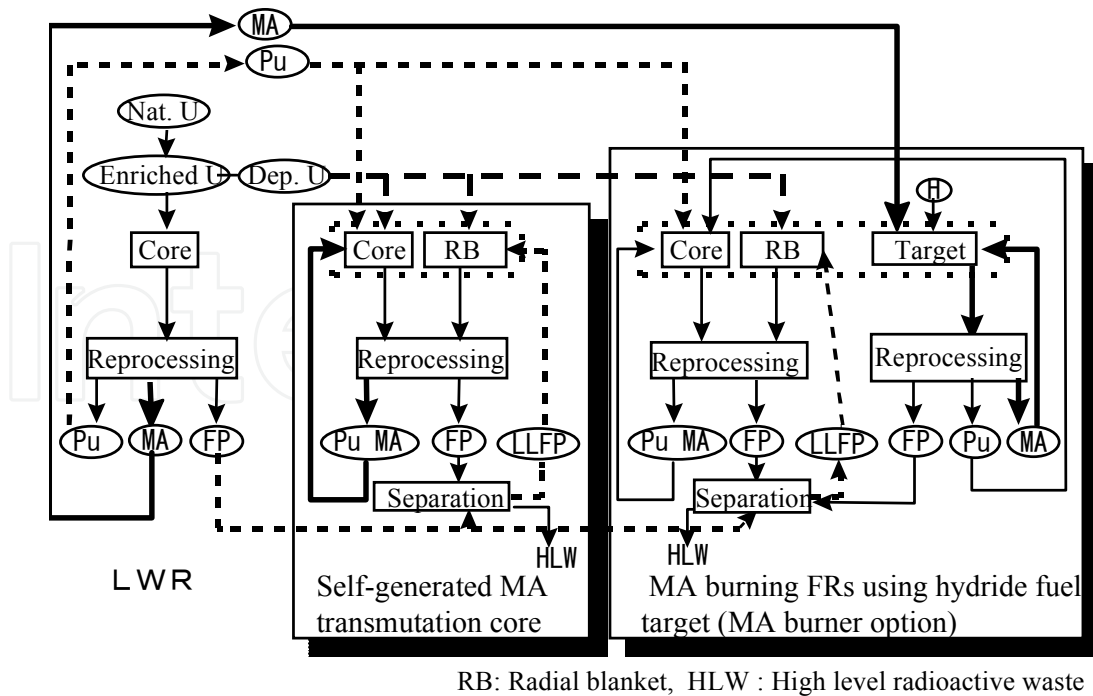
RB: Radial blanket, HLW : High level radioactive waste

Fig. 18. Fuel cycle flow (Fujimura et al., 2001)

3.2.3 Mass balance data for nuclear material

Table 7 shows the mass balance data for investigating the introduction scenario. Electric output of each core is 1GWe.

(a) Mass of MAs produced and transmutated

Twenty three kg of MAs are generated in an LWR per year. The FR without targets in table 7 is the self-generated MA transmutation FR core. Where transmuting no MAs, 28 kg of MAs are generated in an FBR per year. In the MA burner option core, 36 targets in the core region and 72 in the radial blanket region, for a total of 108 targets are loaded. Targets in the core region are shuffled every irradiation cycle and moved to the first row of the radial blanket region. They will be discharged after two more years irradiation. The MAs transmutation rate is 47% in the discharged target and the annual transmutation mass is about 500kg /yr. In the once-through option core, the target assemblies are discharged after six years of irradiation and shuffling in the core region. The core fuel assemblies are discharged after three years. Both MA transmutation and incineration rates are about 100% in the discharged targets. The mass being transmuted and incinerated is about 30kg /yr.

(b) Amount of MAs produced and transmuted

The produced mass of Pu in an LWR is about 20 kg/yr. Because some assemblies in the radial blankets will be exchanged with target assemblies, the breeding ratio will be less than 1 and Pu will be consumed. The consumed mass is about 300 kg/yr. Although depleted uranium is loaded in the radial blanket region of the once-through option core, high Pu enrichment due to the moderating effect of hydride in targets makes its breeding ratio about 1. Therefore annual Pu production and consumption becomes about zero. Target assemblies are discharged after six years of irradiation and shuffling. Core fuel assemblies are discharged after three years irradiation.

3.2.4 Introduction scenario

Transmutation characteristics of the MA burning FR core using hydride fuel targets were enhanced in sec. 3.1. In this section, the effectiveness of thr MA accumulation and the total number of spent target ass being reprocessed for the following cases.

(a) High projection case

(1) Without MA transmutation

MA accumulation was evaluated when an LWR and FBR without MA transmutation function were assumed to be installed according to the nuclear power capacity shown in Fig. 16. MA accumulation in 2100 is about 320 t as shown in Fig. 20.

(2) With MA transmutation

Case-0 LWR→the self-generated MA transmutation FR core

In an FBR, the self-generated MAs are transmuted. This does not contribute to an increase of MA accumulation out of reactors. Therefore MA accumulation in 2100 is about 190 t as shown in Fig. 20. The decreasing rate of MA accumulation is about 40% for Case-0.

Case-1 LWR→MA burner option→the self-generated MAs transmutation FR core

All FBRs being installed after the 2010s were assumed to be the MA burner with Pu recycling option core. New FBR plants, which will be installed after the 2050s when the accumulation mass of MAs out of reactors becomes about zero, will be replaced by the self-generated MAs transmutation FR core as shown in Fig. 19. In this scenario, MA accumulation out of reactors will be kept at zero after 2060 as shown in Fig. 20. As mentioned above, the spent target assemblies should be reprocessed. In this case, the total number of spent target assemblies to be reprocessed is about 14,000.
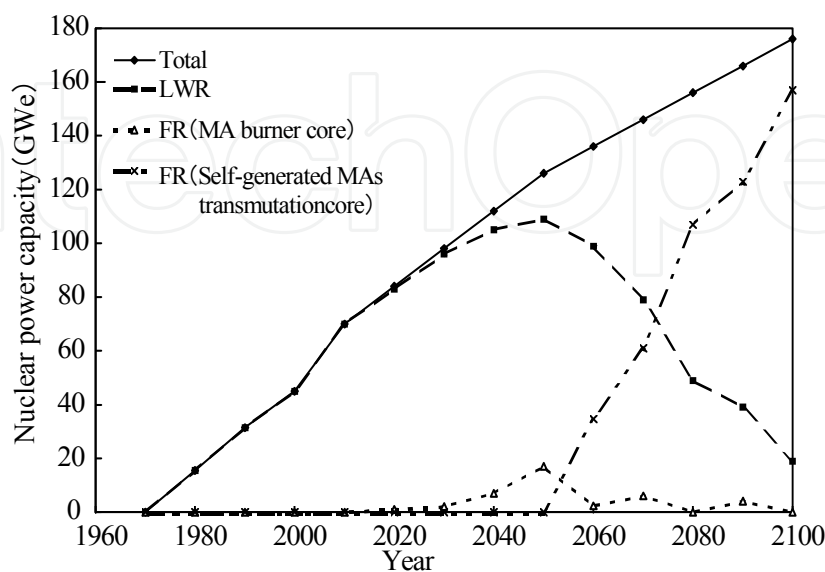
Fig. 19. Projection of installed nuclear capacity (Case-1) (Fujimura et al., 2001)

The number of installation plants for each type of FBR (the MA burner with Pu recycling option core, the self-generated MA transmutation FR core) are set so that the amount of Pu and MAs needed to start up their initial core will not be larger than their accumulation out of reactors. The same applies for Case-2.

If MAs would not be transmuted, more than 300 t of MAs should be disposed as HLW. In this scenario of Case-1, MA accumulation out of reactors will be zero at 2060 which reduces potential radiotoxicity so much in relatively short period and cost for managing the HLW.
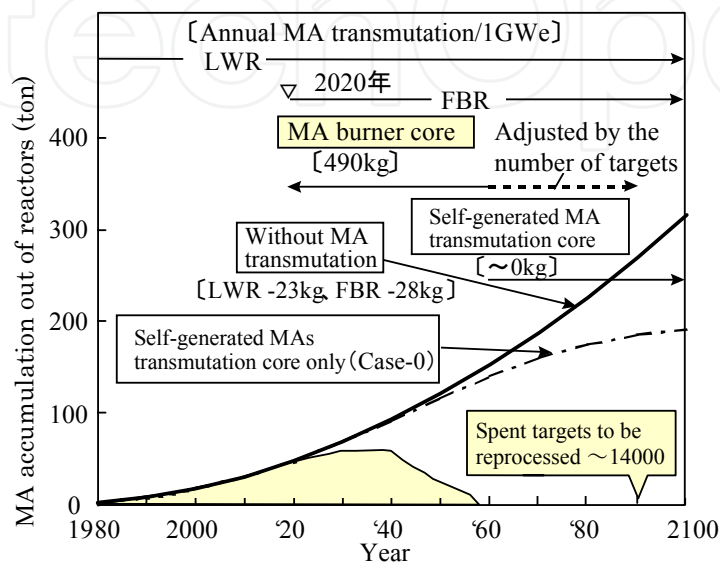
Fig. 20. MA accumulation out of reactors (Case-1) (Fujimura et al., 2001)

Case-2 LWR→MA burner option→Once-through option→the self-generated MA transmutation FR core

All FBRs being installed after the 2010s were assumed to be the MA burner with Pu recycling option core until the ratio of FBR plant numbers to the LWR number is 1 to 21 as shown in Fig. 21.
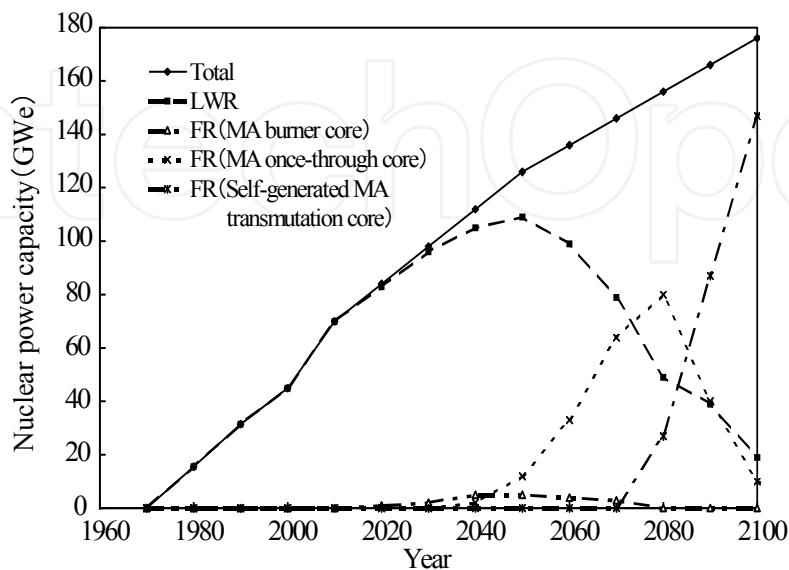
Fig. 21. Projection of installed nuclear capacity (Case-2) (Fujimura et al., 2001)

The once-through option core will be installed if the accumulation of MAs newly produced by LWRs would not increase after 2040. After the 2070s, the number of target assemblies loaded in the once- through option will be decreased in accordance with the decrease of the increasing rate of cumulative amount of MAs due to the decrease in number of LWRs plants. The once- through option core will be replaced by the self-generated MA transmutation FR core. MA accumulation out of reactors will be kept zero in the year 2090 as shown in Fig. 22. Total number of MA burner option cores with Pu multi-recycling is less than that of Case-1 and also the total number of spent target assemblies being reprocessed is about 7000 which is half of that in Case-1. In this scenario of Case-2, the cost for reprocessing the spent fuel will be decreased.
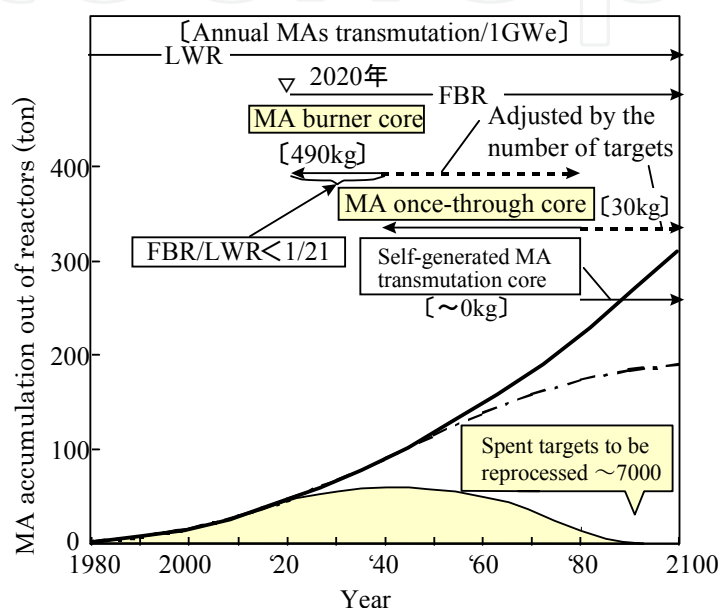
Fig. 22. MA accumulation out of reactors (Case-2) (Fujimura et al., 2001)

(b) Low Projection Case LWR→FR (No target)→MA burner option→the self-generated MA transmutation FR core

Projection of installed nuclear capacity is shown in Fig. 23. In this figure, FR with no target includes conventional type FBR core without MA transmutation and the self-generated MAs transmutation FR core. Installation of the FBR will be started at the 2030s as conventional type core without MA transmutation. The capacity growth of FBR is set to be about 1 GWe or 3 GWe per 10 years between 20231 and 2060, and 10 GWe per 10 years between 2061 and 2160. Installation of the MA burner with Pu recycling option core will be started at the 2040s. The capacity growth of the core is set to be about 1 GWe or 3 GWe per 10 years between 2041 and 2070. Its maximum capacity will be 18 GWe at 2080 as shown in Fig. 23. The accumulation mass of MAs out of reactors becomes about zero at 2080 as shown in Fig. 24. After this year, the number of MA targets loaded in the MA burner with Pu recycling option core will be reduced and some of the new FBR plants will be replaced by the conventional type core without MA transmutation to keep the MA accumulation out of reactors zero. Even though the number og spent target assemblies to be reprocessed was increased being compared with that of Case-1, it is feasible that MA accumulation out of reactors would be held to about zero within the 21st century.

As shown above, the scenario with MA accumulation out of reactors as zero is feasible within the 21st century by introducing the MA burning FR core using hydride fuel targets.
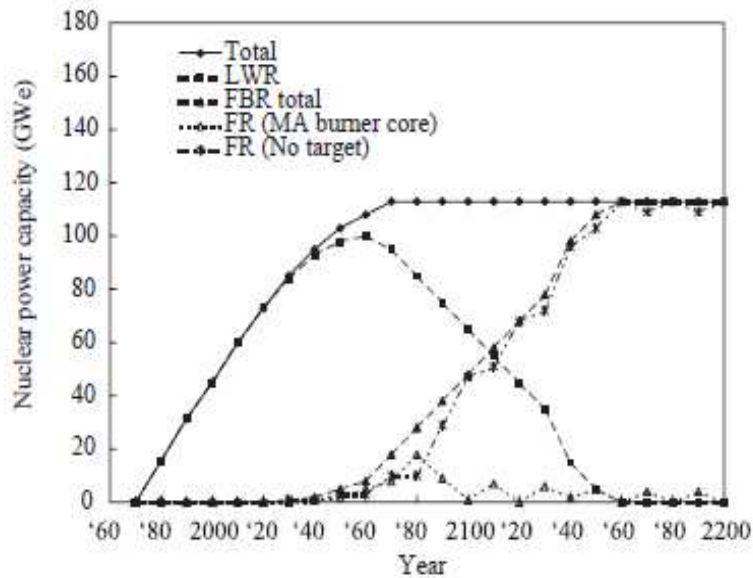

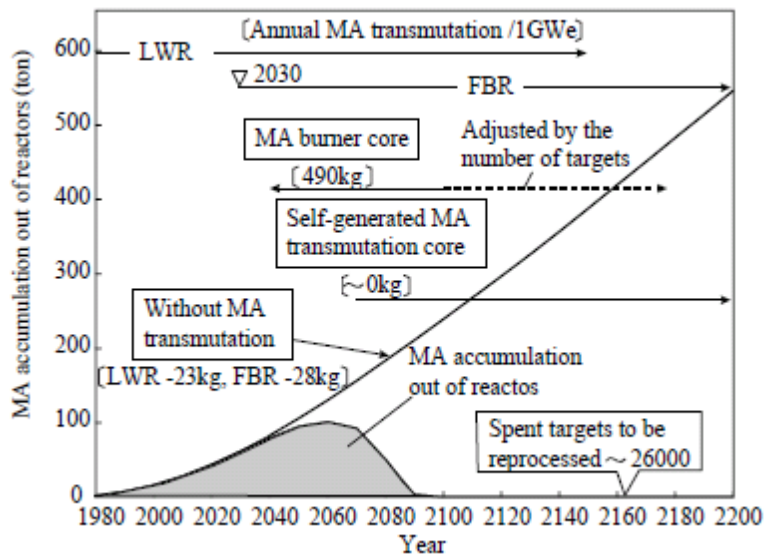Fig. 23. Projection of installed nuclear capacity (Case-3) (Fujimura et al., 2001)

Fig. 24. MA accumulation out of reactors (Case-3) (Fujimura et al., 2001)

3.3 Summary for section 3

Transmutation characteristics of MA burning FR core using hydride fuel targets were enhanced and an introduction scenario for the core was investigated.

1. The MA burner core with Pu multi-recycling could transmute the MAs produced in about 21 LWRs each year. The targets were shuffled after the 1-year irradiated in the core region and further irradiated for 2 years in the first row of the radial blanket region. MA transmutation amount could be increased compared with that in the sec. 2 while being increased thermal margine of MA hydride fuel.

2. The MA once-through core could incinerate almost all MAs loaded in targets by fission during a 6-year irradiation in the core region. The merit of this option is that it could avoid such problems as reprocessing of spent target.

3. By introducing all FRs with the MA burner core after the year of 2020, the following scenario would be feasible. The residual amount of MAs from LWR spent fuel would be held to about zero within the 21st century and all FRs would be changed to the self-generated MA transmutation core without targets.
When the MA once-through cores are introduced with MA burner cores, the total number of targets to be reprocessed could be reduced by about 50%.

4. Even in the low projection case while the MA burner cores are installed at the 2030s after conventional fast breeder reactor without MA transmutation, MA accumulation out of reactors would be held to about zero within the 21st century.

4. Feasibility of the actinide hydride containing Np and Am as a transmutation target fuel

4.1 Feasibility study using a simulated actinide hydride fuel

Feasibility of the actinide-hydride containing [237]Np, [241]Am and [243]Am as a transmutation target fuel to reduce the amount of long-lived actinides in the high level nuclear waste was studied by employing $UTh_4Zr_{10}H_x$, as a simulated actinide hydride fuel. Th was used as a surrogate for minor actinides from the viewpoint of handling radioactive material as well as its similar thermodynamic stability as those of minor actinide hydrides.

4.2 Fabrication and property measurement of the simulated actinide hydride fuel

The pellets of the simulated actinide hydride fuel were successfully fabricated through alloying and hydrogenation within expected diameter errors. Figure 25 shows that the U-Th-Zr hydride consists of three phases: U-metal, $ThZr_2H_x$ and ZrH_x. As shown in Fig. 26, the U-Th-Zr hydride can hold more hydrogen at temperatures above 1173K than the U-Zr hydride (Yamamoto et al., 1994). This is realized due to the higher thermodynamic stability of $ThZr_2H_x$ phase formed in the U-Th-Zr hydride than the U-Zr hydride used in the TRIGA reactors. This finding led to the present concept of the hydride fuel target containing [237]Np, [241]Am and [243]Am for effective transmutation (Yamawaki et al., 1997).
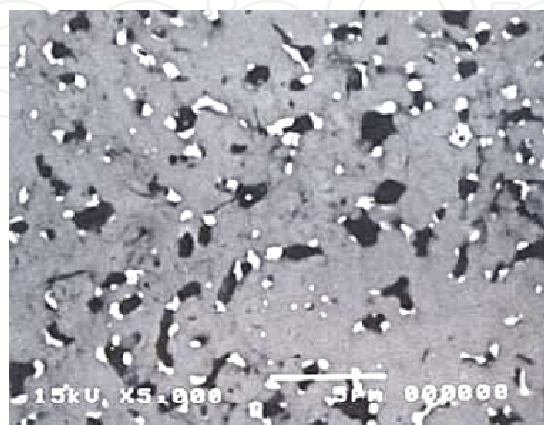

Fig. 25. Back-scattered electron image of $UTh_4Zr_{10}H_{20}$. The black areas consist of Zr hydride; the gray region of $ThZr_2H_x$; the white areas of U metal (Yamamoto et al., 1994)
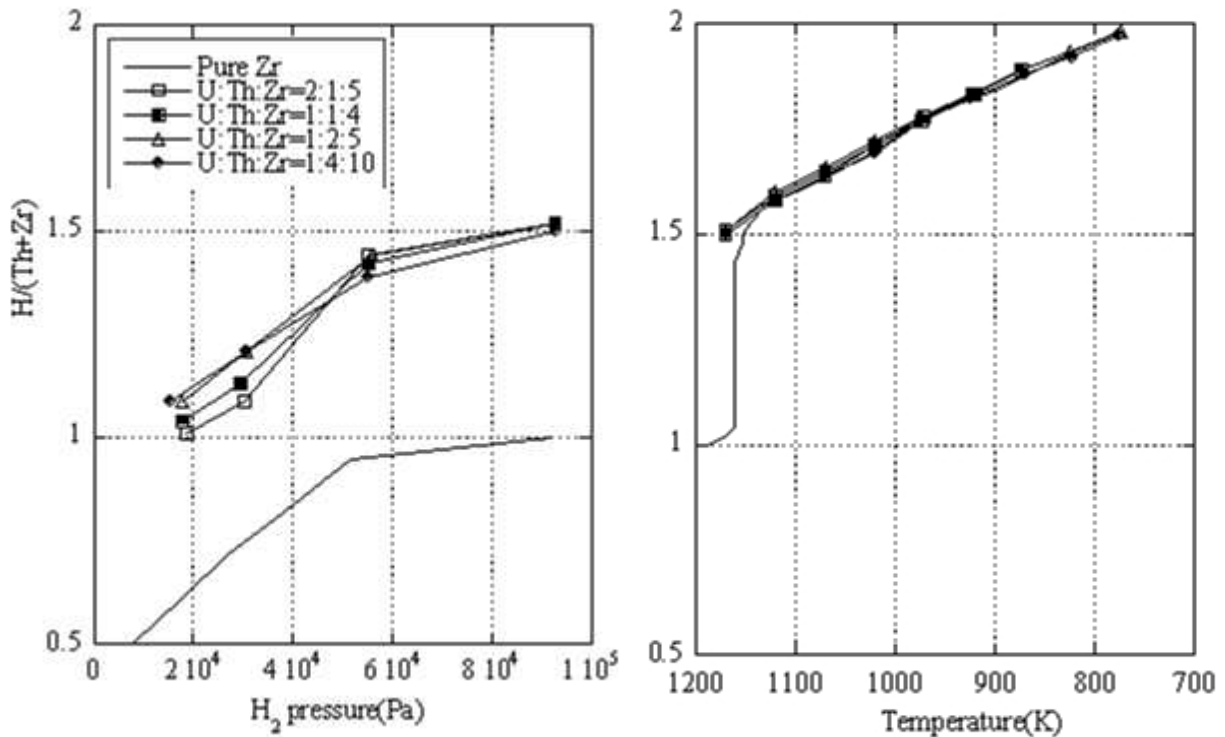
Fig. 26. Equilibrium hydrogen concentration in U-Th-Zr alloys under various hydrogen pressures at 1173K (left figure) and that at various temperatures under 10^5 Pa (right figure). Those of unalloyed Zr are shown as solid lines without symbols attached (Yamamoto et al., 1994)

Fundamental properties such as thermal diffusivity (Fig.27) and thermal expansion (Fig.28) have been measured for the hydride fuel pin design. The thermal diffusivities, α, of $UTh_4Zr_{10}H_{18-27}$ were measured using a laser-flash method (Tsuchiya et al., 2002) in the temperature range from room temperature to 950 K as shown in Fig.27. The thermal diffusivities have been measured both during increase and decrease of the temperature. The results of the respective values were in good agreement. This indicates that the hydrogen release from the specimens was negligible during the measurement. The thermal diffusivity is described as the sum of the lattice contribution and the electronic contribution. The defects due to hydrogen losses in the crystal structure of the hydride increase with decrease of hydrogen content. The marked decrease of the thermal diffusivity at temperatures lower than about 650K seems to be attributed to the effect of such hydrogen defects on the lattice contribution. The thermal conductivity, λ, of $UTh_4Zr_{10}H_x$ was calculated from the following relation of the measured α, the literature data of the density, ρ, and the estimated value of specific heat Cp:

$$\lambda = \alpha \times Cp \times \rho \ (W/cm \cdot K), \tag{1}$$

where
$Cp = -0.110 + 6.87 \times 10^{-4}T + 6.36 \times 10^{-3}x$ (J/g·K),
$\alpha = (1.11x - 21.2)/T + 2.29 \times 10^{-2} + (-3.18 \times 10^{-6}x + 7.59 \times 10^{-5})T$ (cm²/sec),
$\rho = 8.4 - 2.99 \times 10^{-2}x$ (g/cm³).

The thermal conductivity evaluated in this study as shown in Fig. 29 and the thermal expansion measured in this study are essentially important to estimate the temperature and the mechanical integrity of the hydride fuel during irradiation. The hydride fuel decomposes at high temperature, so that the temperature evaluation and mechanical behavior estimation are especially important for this fuel.

4.3 Irradiation tests

Irradiation tests of the simulated actinide hydride target have been conducted in Japan Material Testing Reactor (JMTR) of JAEA. Two irradiation tests of the U-Th-Zr hydride fuel were carried out. The irradiation conditions of the first test were burnup of 0.2% FIMA (Fraction per Initial Metal Atom), linear heat rate of 140 W/cm , fast neutron dose of 1.10×10^{19} n/cm^2 and thermal neutron dose of 1.23×10^{20} n/cm^2. The irradiation conditions of the second test were changed to burnup of 1.1% FIMA, linear heat rate of 178 W/cm, fast neutron dose of 4.66×10^{19} n/cm^2 and thermal neutron dose of 6.43×10^{20} n/cm^2. After irradiation, non-destructive and destructive examinations were performed in each test. It was confirmed that the integrity of the hydride fuel was kept intact through irradiation, supporting the feasibility of the present concept for the hydride fuel target.
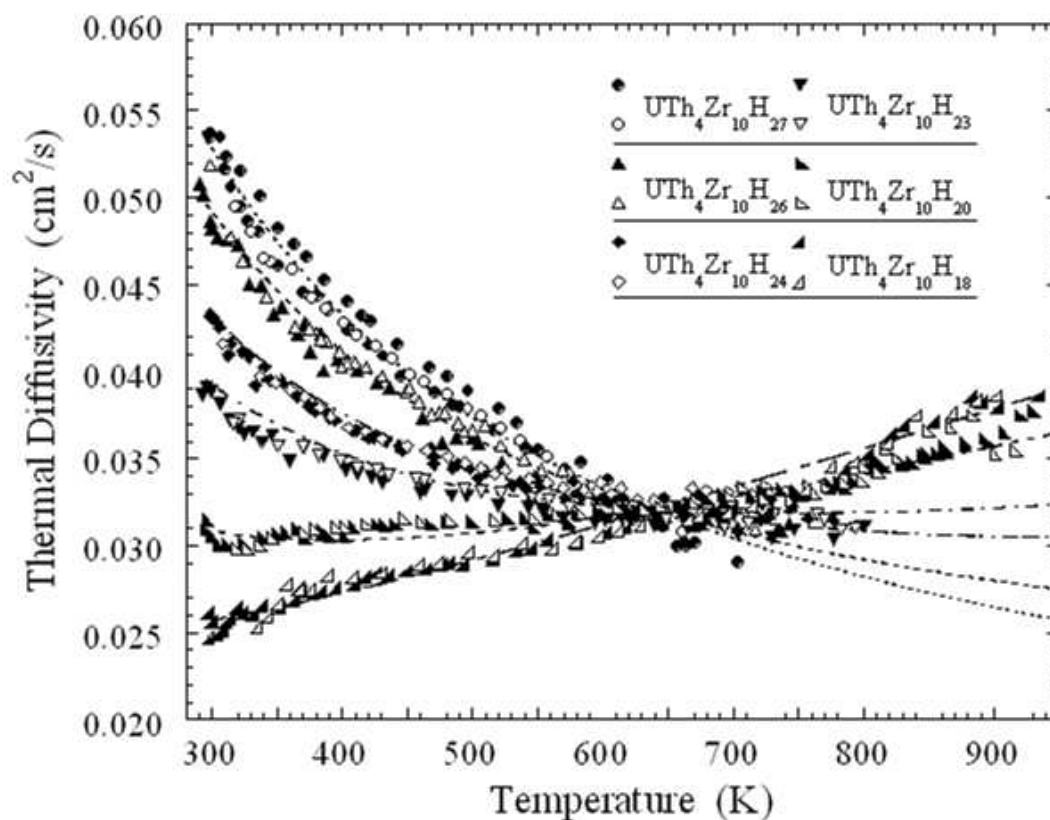

Fig. 27. Thermal diffusivity of $UTh_4Zr_{10}H_{18-27}$ (Open symbols: increasing temperature; Solid symbols: decreasing temperature) (Tsuchiya et al., 2002)
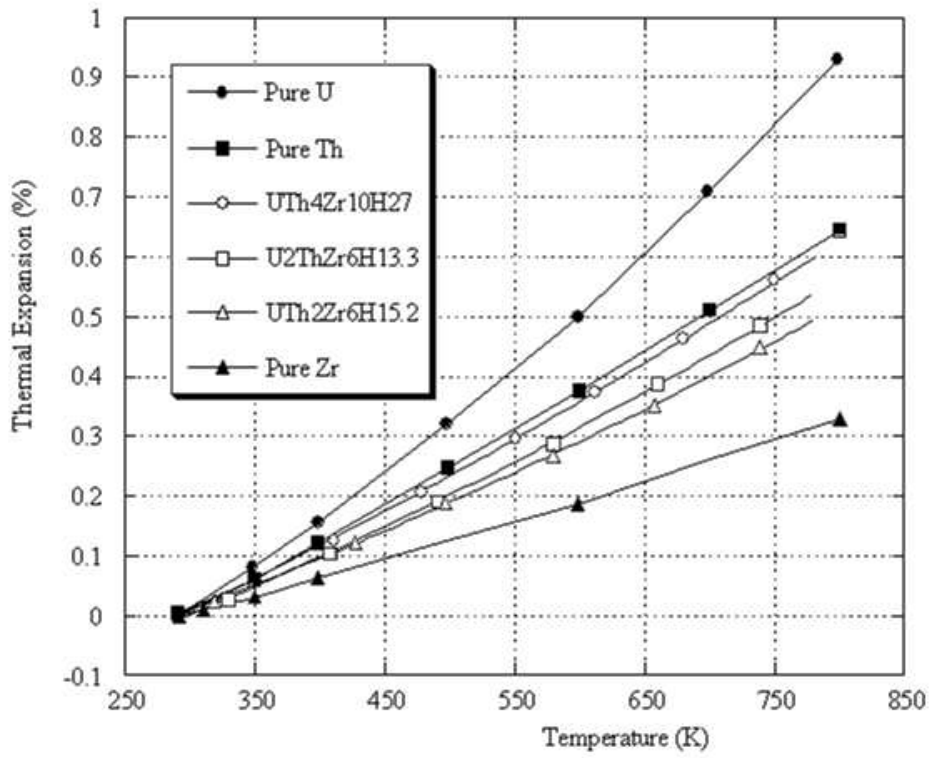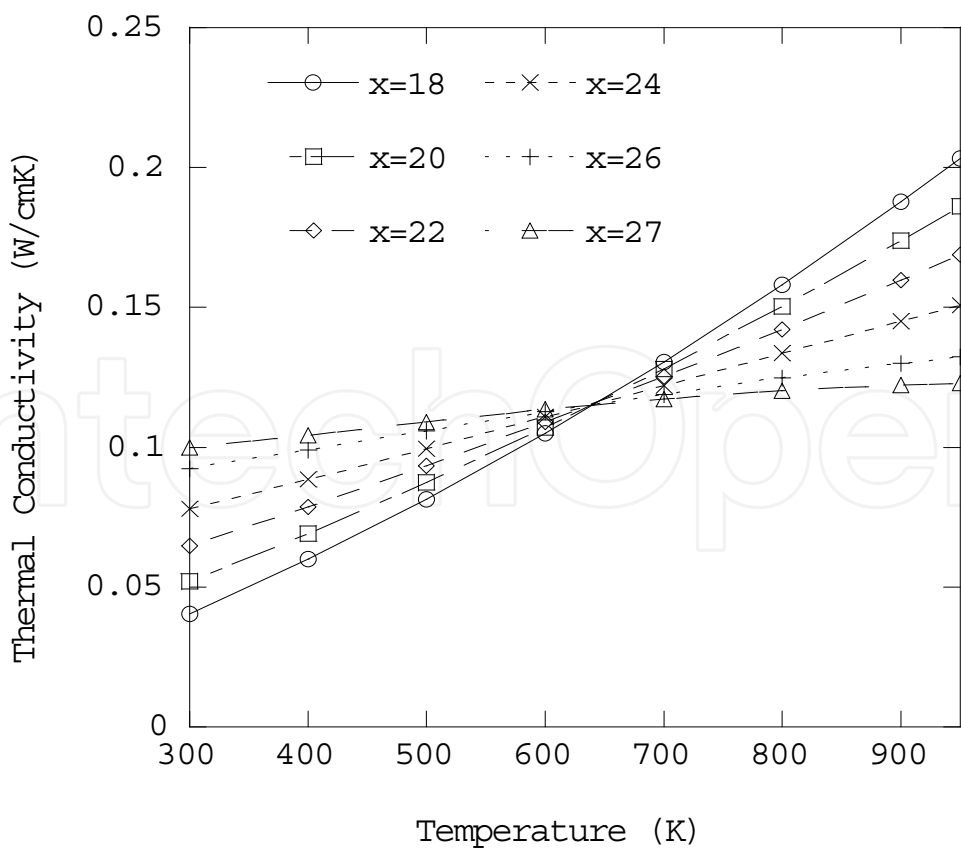
Fig. 28. Thermal expansions of U-Th-Zr hydrides

Fig. 29. Thermal conductivity of $UTh_4Zr_{10}H_x$ (x=18-27)

4.4 Summary for section 4

Feasibility of the actinide hydride containing ^{237}Np, ^{241}Am and ^{243}Am as a transmutation target fuel to be used to reduce the amount of long-lived actinides in the high level nuclear waste has been studied by employing $UTh_4Zr_{10}H_x$ as a simulated actinide hydride fuel, where Th is a surrogate for minor actinides. The pellets of the simulated actinide hydride fuel were successfully fabricated through alloying and hydrogenation within expected diameter errors. It was shown that the U-Th-Zr hydride fuel has higher stability at high temperature than U-Zr hydride. Fundamental properties of $UTh_4Zr_{10}H_x$ such as thermal diffusivity and thermal expansion have been measured and then thermal conductivity was evaluated. These properties are important to evaluate the temperature and mechanical integrity of the hydride fuel during irradiation. Irradiation tests of the simulated hydride fuel were carried out in JMTR of JAEA. It was found that the integrity of the simulated hydride fuel was kept intact through irradiation. This suggests the integrity of the actual hydride fuel to be kept through irradiation, supporting the feasibility of the present concept of using the hydride as transmutation target.

5. Conclusion

High-level radioactive wastes are generated after reprocessing of spent fuels from nuclear reactors, which include long-lived radioactive nuclides of actinides and fission products. The currently available method for final disposal of the high-level wastes is to vitrify them, to store them and to dispose them underground. To reduce the work needed to carry out this type isolation job, a number of transmutation methods have been proposed and studied. This chapter reports an innovative efficient method for transmutation of minor actinides (MAs) by applying hydride targets to fast reactors.

Fast rector core concepts have been studied to reduce long-life radiotoxicity of nuclear waste by applying MA-containing zirconium hydride fuel targets. Systematic parameter survey has been carried out to investigate the fundamental characteristics of MA transmutation and the core safety parameters such as sodium void reactivity in a 1,000 MWe-class fast reactor core. Two core concepts were proposed, using 36 target assemblies, by adjusting the composition of hydride fuels. One is the MA burner core to transmute a large amount of MAs in a short time combined with Pu multi-recycling in fast reactors, whereby the MAs produced in about 13 light water reactors (LWRs) can be transmuted every year with a 58% MA transmutation rate in discharged targets. The other is the MA once-through core to incinerate a small amount of MAs by fission, whereby the MAs produced in about 2 LWRs can be incinerated every year with a 64% MA incineration rate due to 93% transmutation rate in discharged targets. These concepts have been shown to have great potential to achieve good transmutation characteristics of MAs while providing the improved safety characteristics of a fast reactor core.

A scenario to reduce the long-life radiotoxicity of nuclear waste by applying fast reactor cores loaded with hydride fuel targets have been investigated. The MA burner core with Pu multi-recycling can transmute MAs as much as produced in 21 LWRs a year. The targets are shuffled after irradiated 1 year in the core region, then further irradiated for 2 years in the radial blanket region. The MA once-through core can incinerate almost all of the MAs in

targets by fission during 6-year irradiation in the core region. Introduction of the MA burner core for all fast reactors (FRs) after the year 2020 allows the following scenario. The residual amount of MAs from LWR spent fuel can be held nearly zero within the 21st century and all FRs will be changed to the self-generated MA transmutation core without the targets. When the MA once-through cores are introduced with MA burner cores, the total number of targets to be reprocessed can be reduced by 50%. Even in the low projection case while the MA burner cores are installed after conventional FRs, MA accumulation would also be held to nearly zero within the 21st century.

Feasibility of the actinide hydride containing Np and Am as a transmutation target fuel to reduce the amount of long-lived actinides in the high level nuclear waste was studied by employing $UTh_4Zr_{10}H_x$ as a simulated actinide hydride fuel where Th is a surrogate for minor actinides. The pellets of this simulated actinide hydride fuel were successfully fabricated through alloying and hydrogenation within expected diameter error. The U-Th-Zr hydride can hold a larger amount of hydrogen above 1173K than the U-Zr hydride due to higher thermodynamic stability of the former than the latter. Thermal diffusivity and thermal expansion coefficient of $UTh_4Zr_{10}H_x$ were measured, from which thermal conductivity was evaluated.

Irradiation tests of the simulated actinide hydride fuel target have been carried out in Japan Material Testing Reactor (JMTR) of JAEA. Two tests were conducted up to the burnup of 0.2% and 1.1% FIMA (Fission per initial metal atom) each. After irradiations, both non-destructive and destructive examinations were carried out, which showed that the integrity of the hydride fuel was kept intact through irradiation. This result supports the feasibility of the concept of the hydride fuel as a transmutation target.

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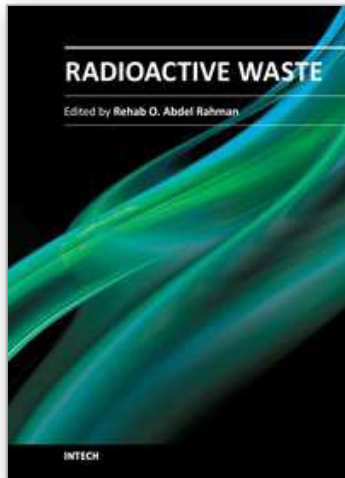
Radioactive Waste

Edited by Dr. Rehab Abdel Rahman

The safe management of nuclear and radioactive wastes is a subject that has recently received considerable recognition due to the huge volume of accumulative wastes and the increased public awareness of the hazards of these wastes. This book aims to cover the practice and research efforts that are currently conducted to deal with the technical difficulties in different radioactive waste management activities and to introduce to the non-technical factors that can affect the management practice. The collective contribution of esteem international experts has covered the science and technology of different management activities. The authors have introduced to the management system, illustrate how old management practices and radioactive accident can affect the environment and summarize the knowledge gained from current management practice and results of research efforts for using some innovative technologies in both pre-disposal and disposal activities.

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