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Concept of hydride fuel target subassemblies in a fast reactor core for effective transmutation of MA

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Abstract

U–Th–Zr alloys with four different compositions were hydrogenated and examined for their hydrogen holding capacities, microstructural and hardness changes with irradiation and thermal diffusivities. Considerably high hydrogen capacity was confirmed up to about 1173 K. A certain degree of irradiation stability was observed, and relatively high thermal diffusivity was ascertained for these hydrogenated ternary alloys. Based on these results, a new concept for effective transmutations of MA was proposed, where target assemblies containing Np and Am in hydrogenated form are loaded in a fast reactor core. This concept has a great potential to achieve the best transmutation of MA with improvement of safety characteristics in a fast reactor core. © 1998 Elsevier Science S.A.

Keywords: Minor actinides transmutation; Hydride reactor fuel; Fast reactor

1. Introduction

Studies on hydrogenation of uranium alloys [1–5] have led to a possibility of MA-containing hydride fission fuel which may be utilized as a MA burning target material. In this study, preparation of U–Th–Zr–H_x type fuel materials has been carried out, where MA are simulated by Th, and their microstructural observation and property measurements have been done. Irradiation of the hydrogenated alloys was carried out in a nuclear reactor and then their microstructural and hardness tests were performed. Thermal diffusivities were measured for the unhydrogenated and hydrogenated alloys.

Based on the studies of such MA containing hydrided fuels, a systematic parameter survey has been performed to investigate the fundamental characteristics of MA transmutation as well as the reactivity coefficients in a 1000 MWe-class fast reactor core. A new core concept was conceived for effective MA transmutation using a fast reactor, in which moderated regions containing Np and Am are constructed in a core region because Np and Am will be effectively transmuted into other actinides of shorter half-lives or effective Pu fissile fuels in soft neutron spectra. MA-containing hydride fuel(U–MA–Zr–H_x) is applied in a moderated target subassembly from the standpoint of high hydrogen and MA concentration and MA–H homogeneity.

2. Studies on hydrogenated U–Th–Zr alloys

2.1. Preparation

U–Th–Zr alloys with atomic ratios of U:Th:Zr=2:1:6, 1:1:4, 1:2:6 and 1:4:10 were prepared in a tri-arc melting furnace in an argon atmosphere. Hydrogenation was carried out in a hydrogen sorption Sieverts system at 1173 K and varied hydrogen pressures below 100 kPa. Hydrogenation was initiated at hydrogen pressure of about 35 kPa, then continued at 75 kPa and then at 100 kPa after equilibrium state was attained at each pressure. All the four alloys exhibited quite similar behaviour, i.e. the atomic ratio H/(Th+Zr) was almost unity at the first equilibrium at 35 kPa and increased to about 1.34 at 100 kPa and 1173 K as shown in Table 1.

After hydrogenation at 1173 K and then cooling to 773 K in 100 kPa hydrogen, the hydrogenated specimens obtained were 2:1:6:13.3, 1:1:4:9.5, 1:2:6:15.2 and 1:4:10:27 in U:Th:Zr:H atomic ratio. Hydrogen was further

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Table 1
Hydrogen capacity of the specimens at varied operating pressure at 1173 K and after cooling down to 1073 K and 773 K in 100 kPa hydrogen

Composition (M)		x in MH_x at 1173 K		Cooled[a] down to	
U:Th:Zr	35 kPa	75 kPa	100 kPa	1073 K	773 K
2:1:6	5.8	8.5	9.4	–	13.3
1:1:4	4.9	5.6	6.7	7	9.5
1:2:6	8.8	9.7	10.7	11.3	15.2
1:4:10	18.2	18.5	18.8	–	27

[a] In 100 kPa H_2.

absorbed by the specimens from the atmosphere during cooling. Fig. 1 shows hydrogen desorption property of the U–Th–Zr–H alloy with U:Th:Zr:H=1:1:4:9.5 at 1173 K [4]. There appeared two plateaus during desorption corresponding to ZrH–$ZrH_{1.4}$ equilibrium at 90 kPa and $ThZr_2H_{7-x}$ homogeneous range equilibrium. The two plateaus are in good agreement with those corresponding to $ThZr_2$–H and Zr–H systems [7,8].

The ternary alloys tested consisted mostly of δUZr_2 solid solution. On hydrogenation, uranium was rejected from this solid solution to form fine uniform dispersion in $ThZr_2H_{7-x}$ and ZrH_{2-x} compounds. Each phase was as fine as 1 μm or less in diameter and homogenously mixed with each other. This behaviour may suggest that these phases were formed from one solid solution with high zirconium content, as suggested by Badayeva and Kuznetsova [6].

2.2. Irradiation effect

Two of the hydrogenated alloys with composition of 1:1:4:7.2 and 1:2:6:11.4 in U:Th:Zr:H ratio were irradiated up to thermal neutron flux of 7.4×10^{22}, 2.2×10^{23} and 7.4×10^{23} n m^{-3} in Japan Material Testing Reactor (JMTR) of Japan Atomic Energy Research Institute (JAERI). The burn up of the specimens was estimated to be 0.3, 0.9 and 3.1% of 235 U atoms, respectively.

Dimension and mass of the specimens exhibited little change before and after the irradiation. Nor was observed any difference in the microstructures of the specimens before and after irradiation. Only some neutron irradiation effect was found in the microhardness test of the specimens [5]. As shown in Fig. 2, microhardness of the specimens decreased slightly with increasing neutron fluence. Such a slight effect of neutron irradiation on mechanical property can be probably attributed to the fine microstructure because the defects produced in crystals by irradiation can sink to grain boundaries with large area. This result also supports attractiveness of the hydrogenated alloys as a fission reactor fuel material.

2.3. Thermal diffusivity

Thermal diffusivity was measured for the unhydrogenated specimens with the U:Th:Zr ratios of 1:2:6, 2:1:6 and 1:4:10 as well as for the hydrogenated specimens with the U:Th:Zr:H ratios of 1:4:10:27 and 1:2:6:15.2 by means of a laser flash thermal diffusivity measuring apparatus. The measured temperature rise curves were analyzed using the logarithmic method. The obtained thermal diffusivities of the unhydrogenated and hydrogenated specimens are shown in Fig. 3a and b, respectively. The hydrogenated specimens have shown a tendency of thermal diffusivity to decrease slightly with temperature up to about 800 K. Then, the thermal diffusivity of the specimens started to increase sharply to reach a plateau at 1073 K. These increases of the thermal diffusivities resulted from dehydrogenation of the specimens. It should be noted that during the laser flash method measurement the specimens were kept in high vacuum. This operating condition, i.e. the vacuum of about $9\sim11\times10^{-6}$ Pa and high temperature,

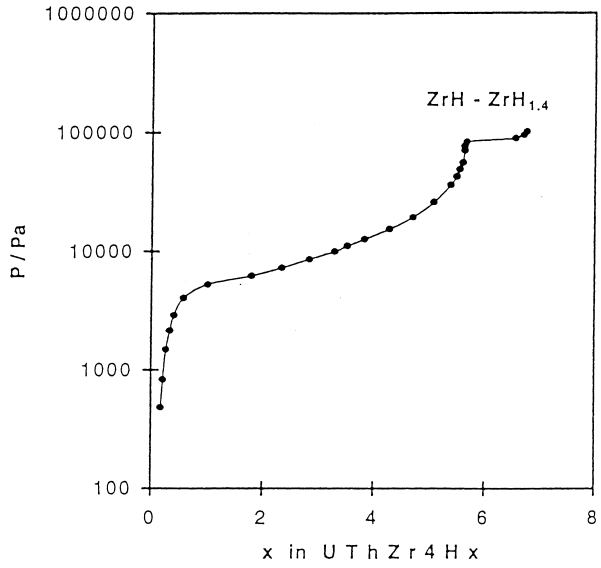

Fig. 1. Hydrogen pressure–composition isotherm of alloy with U:Th:Zr:H ratio of 1:1:4:9.5 at 1173 K.

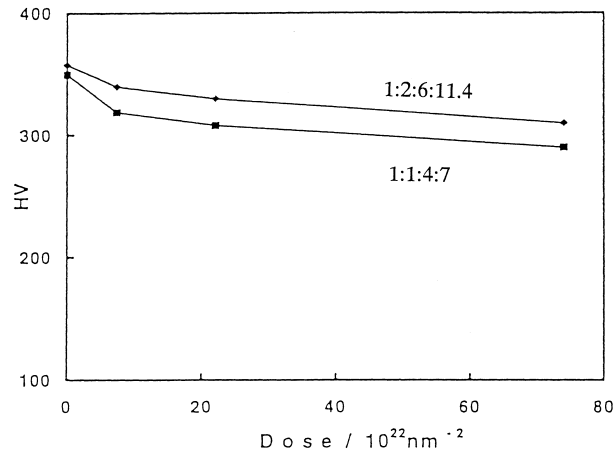

Fig. 2. Vickers' microhardness of unirradiated and irradiated U–Th–Zr alloy hydrides with U:Th:Zr:H ratio of 1:1:4:7.2 and 1:2:6:11.4 respectively.

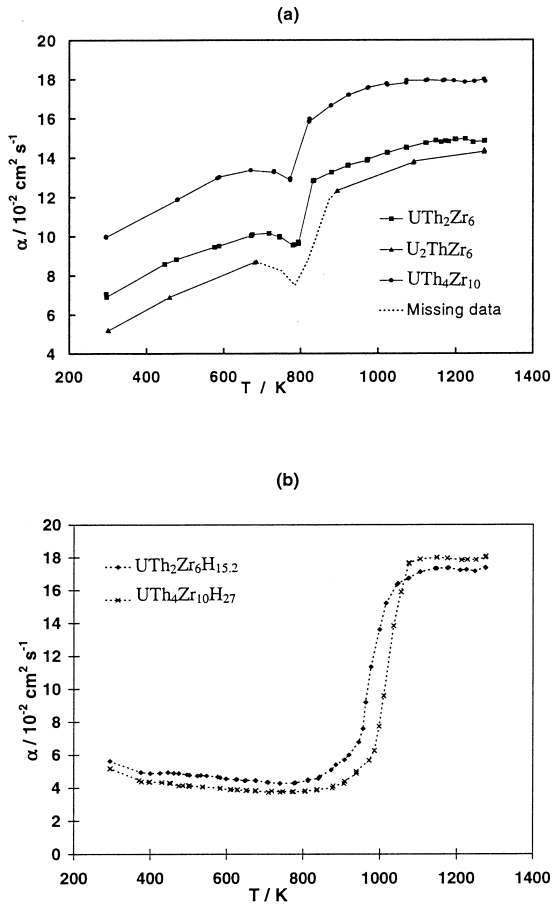

Fig. 3. Thermal diffusivities vs. temperature for unhydrogenated alloys (a) and hydrogenated alloys (b).

caused enhanced desorption of hydrogen from the specimens. After desorption was completed at 1073 K, the thermal diffusivity curve bent horizontally, corresponding to unhydrogenated alloys.

The obtained thermal diffusivity of hydrided specimens was about one third to one half of that of unhydrided alloys. This result leads to the conclusion that the hydrogenated specimens have higher thermal conductivities than UO_2 fuel, where the thermal conductivity of the oxide is equal to about one tenth that of the metal. This fact also supports the favourable characteristics of the hydrogenated U–Th–Zr alloys as a fission reactor fuel material.

3. Transmutation using hydride target assembly

Application of MA hydride to transmutation of nuclear wastes is discussed in this section. Many kinds of transmutation methods based on nuclear reactor concepts have been previously studied [9–11]. One of the most important factors for the transmutation methods is the transmutation rate. A high transmutation rate is necessary to reduce the inventory of nuclear wastes in nuclear reactors. The transmutation rate is determined by the values of neutron flux and nuclear reaction cross section. The target assembly, which contains MA hydrides, is considered to achieve a high transmutation rate in a fast reactor. The MA hydride target assemblies are to be loaded in the core region of a fast reactor with mixed oxide fuel. Fast neutrons generated in the core region are moderated in the MA hydride target assembly to produce a high flux of thermal neutrons, which have large cross sections of nuclear reaction with MA. The target contains ^{237}Np, ^{241}Am and ^{243}Am with a ratio of 77.4:5.0:17.6, which corresponds to that for LWR (light water reactor) UO_2 fuel irradiated up to a burnup of 45 GWD/t. The curium is to be excluded to be stored for over 100 years and the resulting plutonium is recovered to be recycled to the driver core region.

Calculational core analyses were performed with CITATION code [12] and SRAC code [12] using the JENDL-32 library. Fig. 4 shows the comparison of the neutron spectrum in the driver core and that in the MA hydride target assembly. Higher fluxes in the thermal and epithermal spectra are obtained in the target assembly. Fig. 5 shows neutron flux radial distributions for typical energies in the target assembly and an adjacent core fuel assembly. Thermal and epithermal fluxes increase steadily near the target assembly and show almost flat distributions in the target assembly.

The fundamental characteristics of MA transmutation have been evaluated in the case of loading of 36 MA hydride assemblies in a 1000 MWe-class fast reactor core [13]. A high transmutation rate of 28%/year has been obtained. The transmutation amount of MA is equivalent to the annual production of MAs from thirteen 1000 MWe-class LWRs. Table 2 shows the comparison of transmutation characteristics of this concept with other proposed methods. MA transmutation has been studied using LWR [9] as well as FBR (fast breeder reactor) [10],

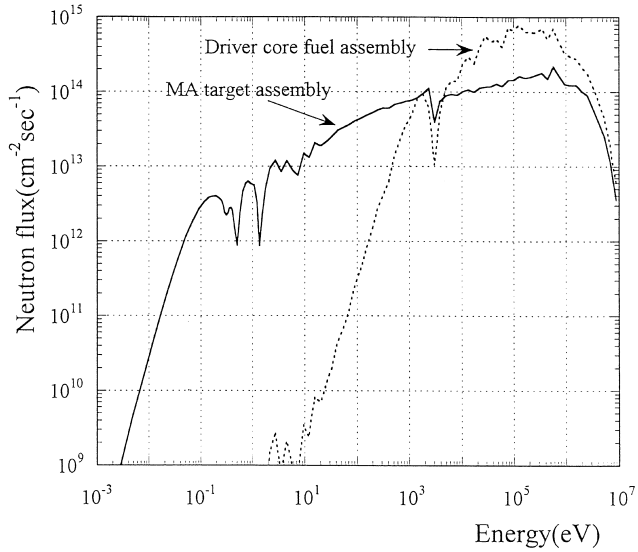

Fig. 4. Comparison of neutron energy spectra in driver core and MA hydride assembly.

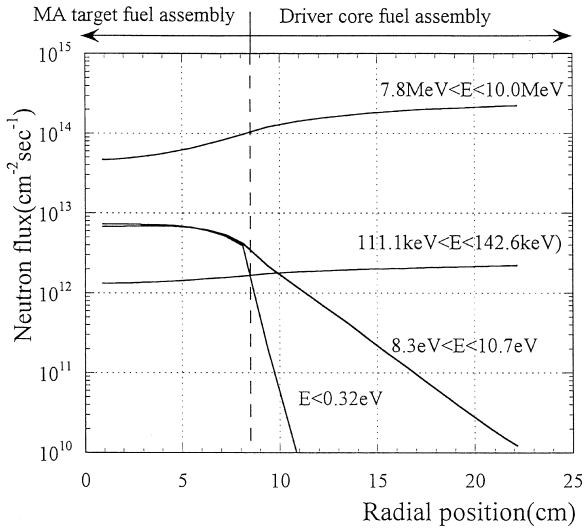

Fig. 5. Neutron flux radial distribution at typical energies in target assembly and adjacent core fuel assembly.

where oxide or metal MA has been considered. The transmutation rate of the present method is much greater than that of the method using LWR or FBR. The burner reactor [11], which is specially designed for MA transmutation, also presents a high transmutation rate.

The MA loaded FBR core and the burner reactor core have problems in the safety characteristics. Since energy spectrum of neutron is getting harder by loading MA, the value of the sodium void reactivity becomes more positive and the absolute value of the Doppler coefficient decreases. In Table 3, the safety characteristics of the proposed core are compared with those of conventional FBR core, where the same amount of MAs is homogeneously loaded in driver fuels. Reactivity coefficients in Table 3 are relative values to those of reference FBR core without MA. It is concluded that safety characteristics can be improved by loading MA hydride target assemblies in the proposed core.

4. Conclusion

Hydrogen absorption properties of U–Th–Zr alloys, where thorium simulates minor actinides, shows a high hydrogen capacity of the specimens up to a relatively high temperature of 1173 K, supporting the concept of hydrided alloy as MA transmuting target material. Microstructural examination and hardness tests after irradiation in a nuclear reactor revealed a certain degree of stability of the hydrided alloys. The thermal diffusivity measurement exhibited relatively high thermal conductivities of the hydrided alloys. Based on these results, a new concept for effective transmutation of MA was proposed, where target assemblies containing Np and Am in a hydrogenated form are to be loaded in a fast reactor core. This concept has been shown to have a potential to achieve the best transmutation of MA with improvement of safety characteristics in a fast reactor core.

Table 3
Improvement of safety characteristics by loading MA hydride target assemblies

	Homogeneously loaded FBR core	Present core
Sodium void reactivity	1.4[a]	0.7[a]
Doppler coefficient	0.7[a]	1.1[a]

[a] Relative values to reactivity coefficients of reference FBR core without MA.

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Table 2
Comparison of transmutation rates in various reactors

	PWR[a] [9]	FBR [11]	Burner reactor [11]	This work
Reactor power(MWth)	3410	2600	1200	2600
MA initial loading(kg)	2700	1450	1865	1050
Transmutation rate(% per year)	13	9	23	28

[a] Volume ratio of fuel to moderator (V_M/V_f)=3.