HYDROGEN CONCENTRATION DEPENDENCE ON THERMAL AND ELECTRICAL CONDUCTIVITIES OF METAL-HYDRIDE COMPOSITE MATERIALS

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Introduction

A transmutation method of actinide radioactive wastes with irradiation hydride targets, which are loaded with the formation of pellets in the core region of fast breeder reactors containing mixed oxide fuels, has been proposed recently[1]. The hydrides are composed in the target materials and play a role as a neutron moderator to gain high flux of thermal neutron. During the irradiation, the gradient of temperature takes place between the parts of center and edge in the targets and the distribution of hydrogen concentration changes with hydrogen diffusion. The thermal and electrical conductivities for various hydrogen concentrations are ones of the most important parameters for designing of the hydride targets. So far, the thydrogen concentration and temperature dependences on the thermal and electrical conductivities of zirconium hydrides[2] and titanium hydrides[3] have been estimated by our group. In the present study, thermal diffusivities of uranium-zirconium hydrides ($UZrH_x$: x=1.6 and 1.9) have been measured within the temperature range from room temperature to 820-900 K by means of a laser flash method and the thermal conductivities have been evaluated with the experimental data of the thermal diffusivity and the reference data of the specific heat and the density[4]. Moreover, the electrical resistivity measurement has been performed and the heat conductions due to free electrons and phonons are discussed.

Results and discussion

A hydrogenation of 45 wt%U-Zr alloys was performed with Sieverts apparatus[5]. It was observed from Scanning Electron Microscope (SEM) that the $UZrH_x$ specimens were two phases composite materials, where ZrH_x phase of 1μm in diameter was dispersed in the bulk of U phase. The structures of $ZrH_{1.6}$ and $ZrH_{1.9}$ have face-ectered cubic (δ-phase) and face-centered tetragonal (ε-phase), respectively.

Fig. 1 shows the results of an isochronal annealing for 10 min at several temperatures of 298□973 K for $UZrH_{1.6}$, $UZrH_{1.9}$, $ZrH_{1.6}$ and $ZrH_{1.9}$.

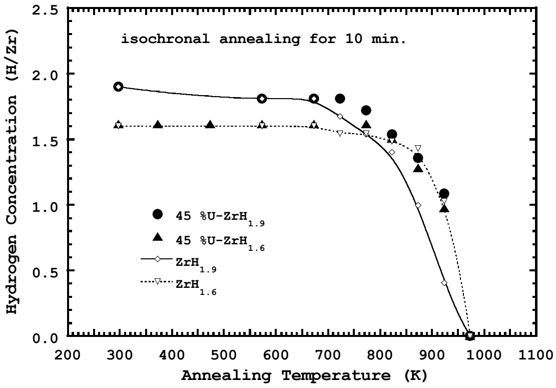

Fig. 1 Isochronal annealing of $UZrH_{1.6}$, $UZrH_{1.9}$, $ZrH_{1.6}$, $ZrH_{1.9}$ at several temperatures for 10 min

The values of H/Zr ratio are obtained by the mass changes. The interesting results are shown in **Fig. 1** that the decomposition temperature of 823 K for $UZrH_{1.6}$ is the same with $ZrH_{1.6}$, while that of 773 K for $UZrH_{1.9}$ is higher than that of $ZrH_{1.9}$. The diffusion of hydrogen atoms is prevented, because $ZrH_{1.9}$ phase is covered with U.

The thermal diffusivities of $UZrH_{1.6}$ and $UZrH_{1.9}$ were measured with increasing (•, ▲) and decreasing (○, □) the heating temperature by means of the laser-flash method, as shown in **Fig. 2**. To avoid the changes of the hydrogen concentration in the hydrides, special specimen containers, made of sapphire, were used in the

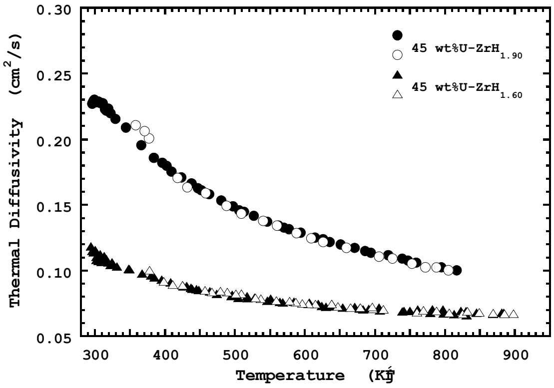

Fig. 2 Thermal diffusivities of $UZrH_{1.6}$ and $UZrH_{1.9}$.

present study. The heating temperatures were successful in elevating up to 900 and 840 K for $UZrH_{1.6}$ and $UZrH_{1.9}$, respectively, during the thermal diffusivity measurements. The thermal diffusivity of $UZrH_x$ increased with increasing hydrogen concentration and with decreasing the temperature. The thermal diffusivity of $UZrH_x$ greatly depends on that of ZrH_x[2], because the thermal diffusivity of U is nearly constant in the experimantal temperature range[6].

In order to clarify electronic heat conduction for $UZrH_x$, the electrical resistivity measurement was perfomed at the tempearure up to 780 K using four-contact DC method. It is seen in **Fig. 3** that the electrical resisitivities of $UZrH_x$ incresed as the temperature increased and the hydrogen concentration decreased. The concentration dependence corresponds to the resistivity for ZrH_x and is caused by electron scattering due to hydrogen vacancy in the hydrides. The resistivity behavior at high temperature dominates by scattering of electrons by acoustic phonons as well as optical phonons[2].

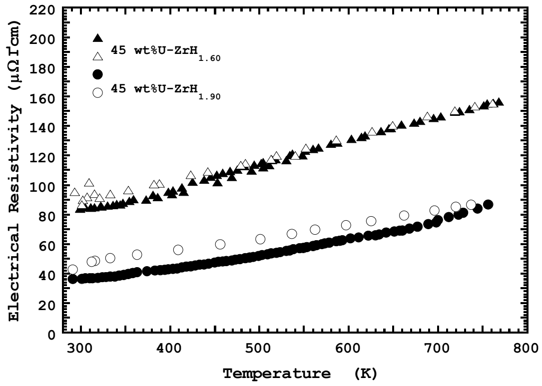

Fig. 3 Electrical resistivities of $UZrH_{1.6}$ and $UZrH_{1.9}$.

Fig. 4 shows the thermal conductivities (λ) of $UZrH_{1.6}$ and $UZrH_{1.9}$ at the temperatures up to 760 K which are calculated from the relation of $\lambda=\alpha C_p\rho$, where C_p and ρ represent the specific heat and the density, respectively. It has been already demonstrated that the values of C_p and ρ are the simple functions of temperature and composition[4]. The high thermal conductivity for the target induces to the safety at high linear power in reactors.

The thermal conductivities of $UZrH_{1.6}$ and $UZrH_{1.9}$ by electronic conduction (λ_e) were estimated from the relations of $\lambda_e=L_e\sigma T$, according to the Wiedemann-Franz rule. σ are the electrical conductivities, L_e are the Lorenz numbers for the electronic conduction, which are assumed as $L_e=(\pi^2/3)(k_B/e)^2$□2.45×10^{-8} [$W\Omega/K^2$], where k_B and e are the Boltzmann constant and elementary electric charge.

Finally, the thermal conductivities of $UZrH_{1.6}$ and $UZrH_{1.9}$ by phonon conduction (λ_p) were determined by subtracting λ_e from the experimental theramal cunductivity λ. At lower temperatures, the contribution by the phonons was greater than that by the electrons, while both electrons and phonons play an important role in the thermal conductivity above 500 K.

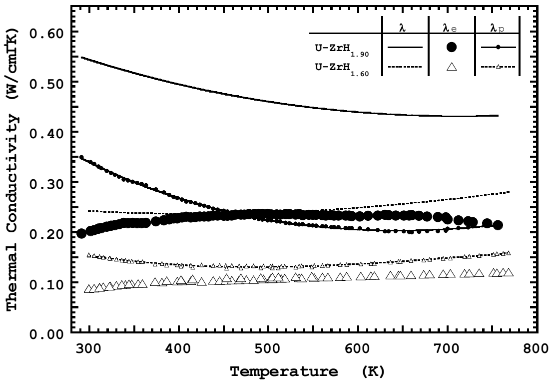

Fig. 4 Thermal conductvities of $UZrH_{1.6}$ and $UZrH_{1.9}$. λ, λ_e and λ_p represent the experimental thermal conductivity, thermal conductivities due to electrons and phonons.

Conclusions

The thermal diffusivities of the metal-hydride composite materials were measured and their thermal conductivities were estimated by taking into account the density and the specific heat. The thermal conductivity greatly depended on one of hydrides and changed with the hydrogen concentration. The heat conductions due to electrons and phonons were separated using Wiedemann-Franz rule and the electrical conductivity. The phonon-phonon scattering is dominant for room temperature, while both the electronic and phonon heat conductions play an important role for the thermal conductivity of $UZrH_x$ at high temperature above 500 K.

References

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