Inter-diffusion between U-Zr-Ce metallic fuel and HT9 cladding at high temperatures

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1. Introduction

U-Zr metallic fuels and ferritic-martensitic stainless (FMS) steel claddings have been considered as the most probable fuel for the sodium-cooled fast reactor (SFR) [1]. The lifetime of the metallic fuel is dependent on the cladding integrity. One of the most important degradation mechanism in metallic fuel is fuel-cladding chemical interaction (FCCI). FCCI is the inter-diffusion reaction between U based fuel and Fe based cladding.

FCCI results in the reduction of cladding wall thickness, and deteriorates the integrity of the fuel rod. This interaction between the fuel slug and cladding occurs during normal operation as well as transient conditions. The diffusion of Fe into U-Zr causes reaction layer formation in fuel and a group of lanthanides (La, Ce, Nd, Pr) which is produced from fission and migrates down the temperature gradient next to the cladding inner surface. Furthermore eutectic temperature of lanthanide and Fe is much lower than that of the U-Fe system, and thus lanthanides could increase these FCCI reaction significantly.

In this study, high-temperatures heating tests were performed for lanthanide containing U-10Zr metallic fuel with HT9 cladding in order to understand the FCCI behavior.

2. Experimental

2.1 Materials

The diffusion couples of U-10Zr-5Ce and HT9 were prepared for the FCCI tests. Among the lanthanides, Ce and Ne are most abundant element, therefore, the material used in this study are U-10Zr-5Ce fabricated by an injection casting method and HT9 cladding material [2]. HT9 was normalized at high temperature of 1050 °C for 1 h and final tempering was conducted at 780 °C for 2 h. After the heat treatment, HT9 was machined into disk specimens of 6 mm diameter and 1 mm thickness for the diffusion couple test.

2.2 Experiments

The surface of the disk type of U-10Zr-5Ce and HT9 material were polished and assembled in a special holder for FCCI. The holder was made of HT9. Figure 1 shows the schematic diagram of the holder for FCCI tests. As shown in the figure, the top and the bottom plates of the U-10Zr-5Ce specimens come into contacts with HT9 plates, which enable to induce eutectic reaction of fuel with HT9 at the both sides. The surface of U-10Zr-5Ce and HT9 was finally polished with 3 μm of diamond suspension on rotating cloths followed by grinding with silicon carbide paper up to grip P2000. The final polishing and assembling were conducted in inert He environment glove box to minimize oxidation during specimen preparation.2

Fig. 1. Schematic diagram of the holder and furnace

To evaluate the fuel cladding chemical interaction (FCCI) under the transient condition in reactor, the U-10Zr-5Ce fuel with HT9 were heated at 800 °C for 1,2 hour in vacuum environment of 10^{-5} torr. After the annealing, the holder was quenched in water. The microstructures of the fuel slug and cladding were observed using scanning electron microscope (SEM, JSM-6610LV JEOL) and elemental distributions and chemical compositions in reaction layer were analyzed by both the energy-dispersive X-ray spectroscopy (EDS, Oxford Instruments) in SEM .

3. Results and discussions
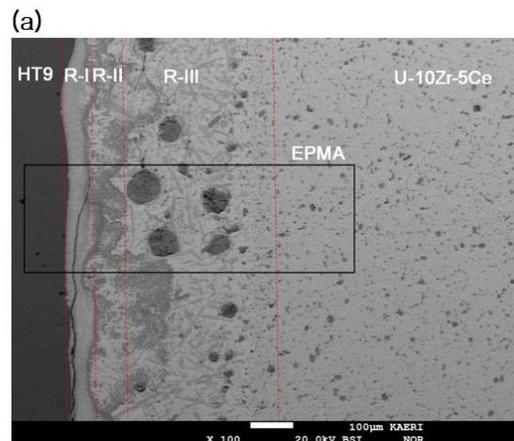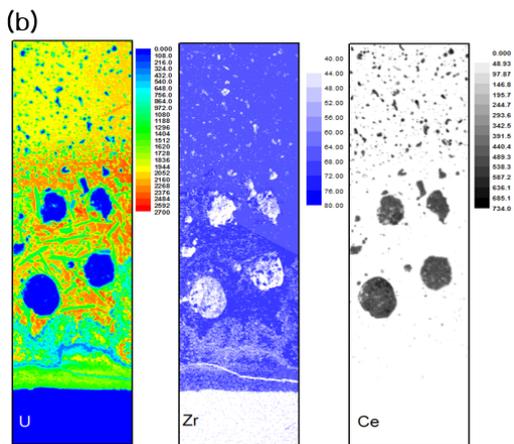
Fig. 2. (a) FCCI area, and (b) U, Zr, Ce distribution map in black box in Fig. 2. a

Fig. 2.a shows the magnified image of the interaction layer between U-10Zr-5Ce and HT9 after diffusion couple at 800 ℃ for 1h. The developed morphology of the reaction layers are different from the layered structures formed in the binary U-Fe system below the eutectic temperature.
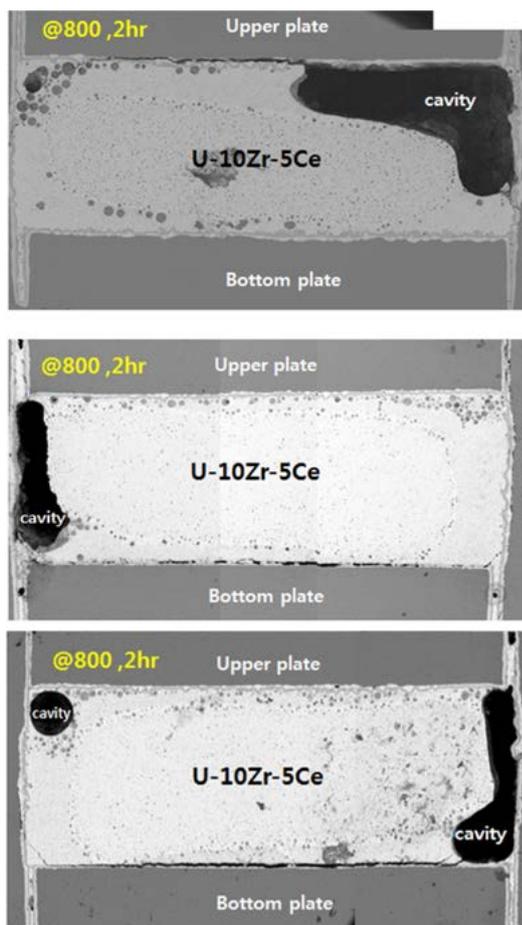
Fig. 3. BSE images of interaction area of U-10Zr-5Ce and HT9 at 800 ℃ for 2 hr.

Fig. 2.b shows the elemental distribution of the by EPMA. The concentrations of the major cladding constituents, Fe and Cr, in the reaction layers are lower than those on the cladding side. As shown in the figure, Fe was predominantly distributed in R-I and R-II, but distribution of Cr was concentrated at the layer boundary of R-I and R-II. In R-III, Cr was occasionally observed in some places while Fe was observed within the acicular type precipitations in R-III. On the other hand, U and Zr were observed in the overall original fuel side region excluding the circular precipitates in R-III. The circular precipitates were identified as Ce rich compound. Interestingly, the concentration of U in the matrix of R-III is higher than that of the original fuel matrix and a significant Ce agglomeration occurs only in this reaction zone.

Fig. 3 shows BSE images of the interaction layer of U-10Zr-5Ce and HT9 at 800 800 ℃ for 2 hr. As shown in the figure, a number of small Ce rich spheres were commonly observed in the phase boundary between un-reacted U-Zr-Ce matrix and reacted one. Also, Ce precipitates were concentrated at the upper corner of the specimens. It seems that the Ce-precipitates tend to agglomerate and migrate to upper position maybe due to the density differences. Such forms of the agglomeration can be found easily in the melt residue in the U-Zr-RE [3]

4. Summary and conclusions

Diffusion couple of U-10wt.%Zr-5wt.%Ce with HT9 was conducted at high temperature under vacuum conditions. The reaction layers of FCCI were examined using SEM and EPMA.

- The boundary between HT9 and U-Zr-Ce is move to the HT9 side after the reaction indicating that diffusion of Fe is faster than U.

- Ce agglomeration occurred in the reaction layer adjacent to the original fuel side; however, such agglomeration did not occur in the original fuel matrix.

- Thus, it is concluded that rapid agglomeration of Ce only occurs in the liquid phase of U and Fe. This supports liquid-like diffusion [4] of lanthanide in irradiated fuel

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