Fuel-Cladding Chemical Interaction between U-Zr-X Metallic Fuel and Ferritic Martensitic Steels

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1. Introduction

Metallic fuel is being developed as a candidate fuel system for a sodium-cooled fast reactor (SFR). The advantages of metallic fuel include a high thermal conductivity, high heavy metal density, and a compatibility with a liquid sodium coolant. Ferritic martensitic steels (FMS) such as HT9 and T91 have been considered as candidate cladding materials for SFR fuel due to their excellent irradiation swelling resistance. Interaction layers are formed by a fuel-cladding chemical interaction (FCCI) during irradiation. Eutectic melting of these interaction products at a temperature much lower than a melting point of a fuel alloy is considered as one of the major performance-limiting factors for a metallic fuel system. FCCI behaviors of U-Zr and U-Pu-Zr alloys against Fe, Fe-Cr, D9 and HT9 have been investigated by an out-of-pile annealing of their diffusion couples[1-4]. Growth kinetics of the interaction layer thickness needs to be correlated because a thinning of a cladding by an interaction layer formation decreases the load bearing capability of a cladding tube. Recently, usage of a thin coating on the inside wall of a cladding as a diffusion barrier has been proposed in order to retard the interaction between fuel and a cladding.

Various fission products are produced in metallic fuel during irradiation. However, the effects of lanthanide elements on a FCCI have not been clearly understood as yet. In addition, minor actinides such as Np, Am, and Cm recovered from LWR spent fuel will be added to metallic fuel for the GEN-IV SFR in order to burn the long-lived minor actinide elements. Therefore, the effects of the addition of minor actinide elements on a FCCI need to be addressed, too.

In this study, FCCI behaviors of U-Zr-X alloys against FMS plates were investigated by observing the microstructures of the diffusion couples after out-of-pile high temperature annealing tests.

2. Methods and Results

2.1 Experimental Procedures

Small rods of U-Zr, U-Zr-Ce, U-Zr-Nd (U-Zr-X) alloys were fabricated by an induction melting of elemental ingots in zirconia crucibles. Each U-Zr-X rod was furnace-cooled after a melting. Nominal Zr content was 5, 10, 15 wt% and the lanthanide content was 2, 4, 6 wt%. As FMS cladding materials, plates of HT9 and T91 steels were used. Diffusion couple annealing tests were carried out in a muffle furnace. Mechanical clamps joining the diffusion couples of the U-Zr-X and FMS slices were vacuum sealed in quartz tubes. Annealing temperature was 700~800^oC.

Microstructures of the diffusion couple specimens were observed by using a scanning electron microscopy (SEM) and the elemental composition of the interaction layers were measured by using an energy dispersive X-ray spectroscopy (EDS).

2.2 Results

Liquid phase formation in the interaction layer can be identified by a microstructural characterization of the diffusion couples. According to liquefaction data, a liquefaction will occur at about 725^oC for a U-10Zr/Fe diffusion couple[4]. A cross-sectional micrograph of a U-10Zr/T91 diffusion couple annealed at 740^oC for 25 hours showed that various interaction layers were formed as shown in Figure 1. U-Fe binary phase diagram shows that the composition (at%) of the liquid phase forms by an eutectic melting is $U_{67}Fe_{33}$ at 725^oC. EDS analysis of the interaction layers showed that the interaction layer on the FMS side consisted of gray $(U,Zr)Fe_2$ and a bright liquid phase (air-quenched structure). On the U-Zr side, dark $(Zr,U)Fe_2$ and gray $(U,Zr)Fe_2$ were mixed in the $(U,Zr)_6Fe$ matrix. Zr-rich phase such as $ZrFe_2$ might decrease the eutectic melting temperature by lowering the Zr content in the interaction layer. The effects of the Zr content in the U-Zr-X alloy on the liquid phase formation behavior need to be analyzed.

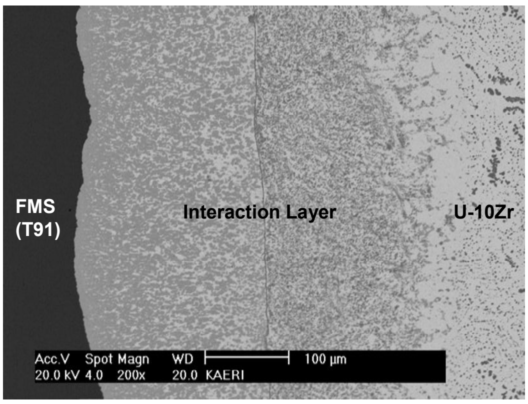

Figure 1. A scanning electron micrograph of a U-10Zr vs. T91 diffusion couple annealed at 740^oC for 25 hours.

Figure 2 shows a thinning of the FMS by the interaction layer formation. Thickness of the FMS after an interaction was decreased by a penetration of an interaction layer when compared with the initial thickness (1.0 mm). Thickness growth kinetics of the interaction layer can be expressed as a function of the temperature and time as follows:

$$T^2 = kt \quad (1)$$

$$k = A\exp(-Q_{eff} / RT) \quad (2)$$

where T is the thickness of an interaction layer, k is the kinetic constant, t is the annealing time, A is the pre-exponential constant, Q_{eff} is the effective activation energy for an interdiffusion, R is the gas constant, and T is the absolute temperature (K). Correlation constants can be obtained by the data from the multiple test results with different sets of temperatures and annealing times.

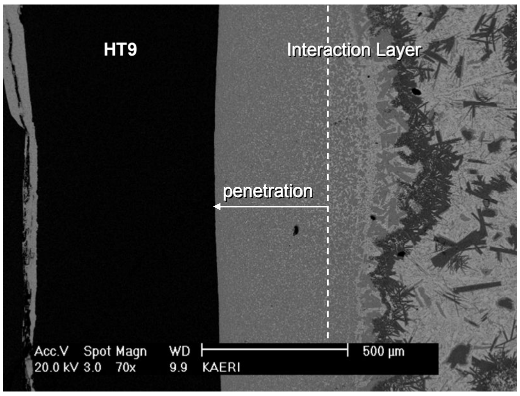

Figure 2. A scanning electron micrograph of a U-10Zr vs. HT9 diffusion couple annealed at 800°C for 7 hours.

Nakamura et al. constructed a U-Fe-Zr ternary phase diagram for 580°C, 700°C, and 800°C as shown in Figure 3 [5]. When the average composition of the interaction layers are connected on the phase diagram, diffusion paths of the diffusion couple can be obtained as superposed in Figure 3.

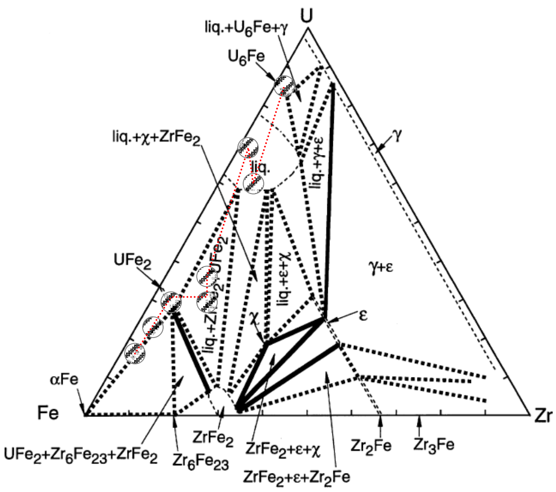

Figure 3. Ternary phase diagram of U-Zr-Fe at 800°C proposed by Nakamura et al.[5]. A diffusion path obtained in this study was superposed.

Effects of the addition of lanthanide elements to U-Zr-X fuel were analyzed by comparing the diffusion couple test results of U-Zr/FMS, U-Zr-Ce/FMS, U-Zr-Nd/FMS. In addition, performance of the diffusion barrier was compared by considering the phase diagram of each diffusion couple system. Thin foils of Zr, Cr, V, Nb, Ti, Mo were used as surrogates for the diffusion barrier coatings on the cladding. Although the diffusion couple test showed microstructures resulting from an eutectic melting, more exact eutectic melting temperature can be obtained by a dilatometer analysis. The total length of a diffusion couple decreases abruptly by deviating from its thermal expansion curve when an eutectic melting occurred on a heating.

3. Conclusion

Diffusion couple tests of U-Zr-X vs. HT9 or T91 showed that various interaction layers were formed. Liquid phase formation by an eutectic melting was observed in the high temperature (740~800°C) annealing tests. Diffusion paths were analyzed based on microstructural observations of the interaction layers. An experimental setup by using a dilatometer was designed to measure the eutectic melting point of the fuel-cladding interaction systems.

Acknowledgment

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