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Interaction Behavior between Binary Lanthanide Alloy and HT9

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1. Introduction

Metal fuel in Sodium-cooled Fast Reactor (SFR) has the advantages of high thermal conductivity, excellent compatibility with the coolant sodium, and superior proliferation resistance when recycling spent nuclear fuel in connection with the pyroprocessing. However, when the U in metal fuel and Fe in fuel cladding come into contact at temperatures above 780^{o}C, the resulting eutectic phenomenon causes mutual fusion, and when Pu is added, the eutectic temperature is lowered to 650^{o}C [1,2] (Fuel Cladding Chemical Interaction, FCCI). Among the nuclear fission products included in the light water reactor fuel rod, a small amount of rare earth metals exist, such as Ce, La, Pr, Nd, Sm, and so on. These elements are initially added to the SFR metallic fuel through pyroprocessing, to about 1 wt%. Also, as SFR operation continues, additional amounts of rare earth elements are newly produced as a fission product. These elements are moved by the temperature gradient and accumulate at the surface of the fuel rod, where these rare earth elements contribute negative effects, such as decreasing the temperature at which the eutectic phenomenon occurs and increasing the reaction layer [3,4].

In this study, the effects of Ce and Nd, which dominantly influence FCCI, were evaluated in order to qualitatively analyze the effect of each element on such interactions. In order to conduct experiments, a binary model alloy was fabricated by changing Ce and Nd content, and after conducting diffusion couple tests with the SFR cladding tube material HT9, the effect of elemental content was assessed through microstructure analysis.

2. Experimental Procedure

2.1. Material preparation

The binary Ce-Nd model alloy was fabricated using the vacuum arc melting (VAR) process. By quantifying Ce and Nd, 4 types of binary Ce-Nd alloy (80Ce-20Nd, 60Ce-40Nd, 40Ce-60Nd, 20Ce-80Nd, wt% basis) ingots were fabricated to be 200g each. For comparison, ingots of 100wt% Ce (hereafter 100Ce) and 100wt% Nd (hereafter 100Nd) were also manufactured. The melted alloy was processed into disk specimens of 6 mm diameter and 1.5 mm thickness for the diffusion couple tests. The cladding material used in the experiments was HT9, which includes 12 wt% Cr, 1 wt% Mo, and microelements. HT9 was fabricated using vacuum induction melting and hot rolling. After 1 hour of normalizing at 1050^{o}C and tempering for 2 hours at 780^{o}C, the material was finally given a tempered martensite structure. After the heat treatment, the cladding material was processed into disk specimens of 8 mm diameter and 1.5 mm thickness for the diffusion couple tests.

2.2. Diffusion couple test

After manufacturing the experimental specimens, the diffusion couple test was conducted. After arranging the pre-polished Ce-Nd alloy and HT9 cladding material to touch each other, the materials were joined together using screws to induce interdiffusion at the test temperatures. After joining, the specimens were placed in a quartz tube and sealed with a vacuum of 10^{-2} Torr. The diffusion couple test was performed by inserting the sealed specimens in a heating furnace for a specific amount of time. The test conditions were 1, 2, 6, and 25 hours at 660^{o}C, which is 10^{o}C higher than the maximum design temperature 650^{o}C by assuming the metallic fuel-cladding tube interface.

After test, the specimens were cut, mounted and polished then the microstructural observation was carried out using scanning electron microscope (SEM) equipped with energy dispersive spectra (EDS). The reaction layer thickness was measured based on the boundary surface between two materials before the test by averaging thickness values, and the variation in reaction layer thickness according to the alloy element and test time was investigated.

3. Result and Discussion

3.1. Interaction behavior

Fig. 1 shows the growth of reaction layer thickness according to the alloy element. The 1 hour diffusion couple test results showed that a reaction layer of 100Ce approximately 65.9μm was formed and the reaction layer thickness began to increase as the Nd content was increased. In the case of the 40Ce-60Nd alloy, the reaction layer thickness reached its maximum of approximately 205μm. Based on the 40Ce-60Nd case, the reaction layer thickness began to decrease as the Nd content was increased. In the case of 100Nd, a reaction layer formation of approximately 8.05μm was observed. This trend was also observed in the 25 hour diffusion couple tests. In terms of the compositional effect, synergism was observed, where the thickness was at its maximum when their ratio reached nearly 1:1. This effect showed a consistent tendency regardless of the test time. This implies that not only Ce but also Nd has a significant impact on the formation of the interaction layer.

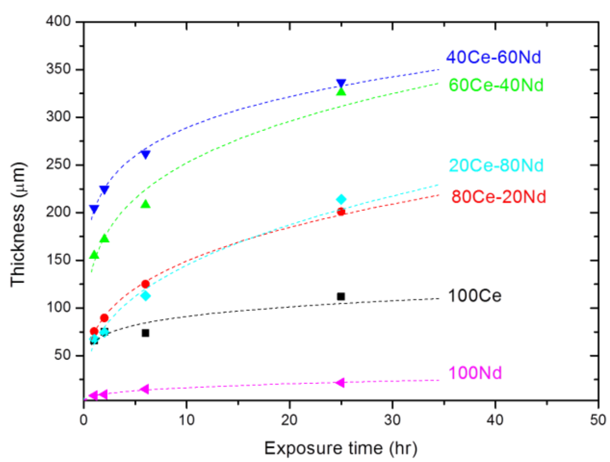

Fig. 1 Growth of interaction layer with time between binary xCe-yNd alloy and HT9 at 660°C

3.2. Analysis of interaction mechanism

Previous diffusion couple study using Mischmetal (70Ce-30La) with Gr.92 (9Cr-2WVNb) revealed that Ce mainly migrates into Fe side to form Fe_2Ce type intermetallic compound, leaving a Fe depleted zone and subsequent Ce substitution [5]. In the case of diffusion couple study with Nd and Gr.92 (9Cr-2WVNb), Nd gradually distributed inside reaction layer to form $Fe_{17}Nd_x$ (x=2, 5) type intermetallic compound [6]. The xCe-yNd alloy and HT9 cladding material interaction showed overlapping individual effects for Ce and Nd, as shown in Fig.2. The Fe distribution in the interaction layer was stabilized by the Nd, allowing continuous Fe movement toward the rare earth alloy, resulting in larger interaction layer thicknesses compared to the diffusion couple test with pure Ce and Nd.

4. Conclusion

Studies were conducted to assess the effects of rare earth elements produced during fission process. The binary model alloy xCe-yNd was fabricated using the vacuum arc melting (VAR) process, and the diffusion couple tests with cladding material HT9 was carried out at the temperature of 660°C. The result showed that Ce and Nd reacted with the Fe of the cladding material to form an interaction layer, and synergetic phenomena significantly affecting the interaction layer thickness was observed when their composition reached nearly 1:1, which is due to the overlapping reaction mechanisms between Ce and Nd with Fe. Further tests regarding synergistic effect on binary lanthanide elements (Ce, Nd, Pr, La, Sm) will be conducted by fixing element contents, 1:1.

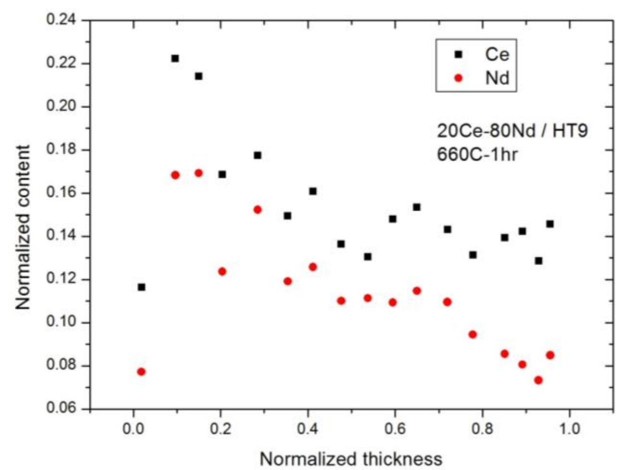

Fig. 2 Normalized element distribution of the test specimen by dividing initial nominal element content after diffusion couple test of 20Ce-80Nd at 660°C for given hour. (Left : 20Ce-80Nd, Right : HT9) Normalized thickness obtained by dividing interaction layer thickness.

Acknowledgement

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