Effect of Rare Earth Element on the Chemical Interaction between HT9 Cladding

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1. Introduction

Metallic fuel is considered as fuels for Sodium-cooled Fast Reactors (SFRs) because it can maximize uranium resources. It generates rare earth elements as fission products, where it is reported by aggravating Fuel-Cladding Chemical Interaction (FCCI) at the operating temperature [2-6]. Rare earth element forms as a multi-component alloy (Ce-Nd-Pr-La-Sm-etc.), where it shows higher reaction thickness than single element [3]. Previous experiment using binary model alloy of xCe-yNd revealed that reaction thickness reaches at a maximum when a ratio of rare earth is 1:1 [1]. The objective of this study is to evaluate the effect of rare earth elements on such synergistic behavior. A total of twelve rare earth alloy models are prepared, which of five of them are single system alloys and seven binary system alloys. Diffusion couple tests are performed with the HT9 (12Cr-1MoWV) cladding at 660 ℃ for 1, 6, 25 and 48 hours. The thickness of the reaction layer and the migration of each element are analyzed by scanning electron microscope.

2. Experimental Procedure

2.1 Specimen Preparation

The cladding material HT9 was processed into a disk with a diameter of 8 mm and a thickness of 1.5 mm. The rare earth specimens were prepared by selecting five elements from the fission product, which are Ce, La, Pr, Nd, and Sm. The ingots were produced by vacuum arc remelting using rare earths elements produced by Alfa Aeser Co. and processed into a square shape of 5 mm×5mm×1.5 mm. Ingots made into single element were produced. The binary alloy model consists of Ce, Nd, Pr, La and Sm elements by setting constant as 1:1 ratio, respectively. (50Ce50Nd, 50Ce50Pr, 50Ce50La, 50Ce50Sm, 50Nd50Pr, 50Nd50La, 50Nd50Sm)

2.2 Diffusion Couple Test

In this study, diffusion couple experiment of HT9 and rare earth alloy model was carried out. HT9 and rare earth alloy were inserted as shown in the Fig. 1 and Ta foil was applied at the top and bottom to prevent unnecessary reaction between specimen and jig. The specimen was contacted with a small jig, then put into a large one and fixed with a bolt. After fixing, argon gas was injected and sealed with a quartz tube. Experiments were performed for 1, 6, 25, and 48 hours successively at 660°C, which is 10 °C higher than the SFR operation temperature of 650°C. In order to secure reproducibility in the thickness of the reaction layers, the experiment was repeated 3-5 times. After the heat treatment, the specimen was cut in half to observe the cross section of the reaction layer.

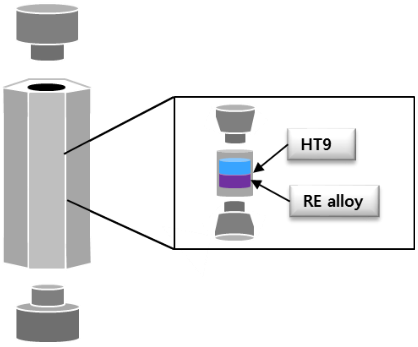

Fig 1. Schematic illustration of the diffusion couple test

3. Results and Discussion

3.1 Reaction phenomena between single RE and HT9

It is known that the thickness (Δx) of the reaction layer has a non-linear relationship with time (t), and the equation is as follows.

$$\Delta x = A \cdot t^{n} \tag{1}$$

In this formula, A is the constant and n is the reaction rate exponent. As the reaction rate exponent is closer to 0.5, the reaction is dominated by the diffusion mechanism, and the closer to 0.33, the greater the influence of the mechanism other than diffusion [5]. Rate exponent in Fig. 2 revealed that value of Nd was close to 0.5, which implies that diffusion occurred in Nd single element system.

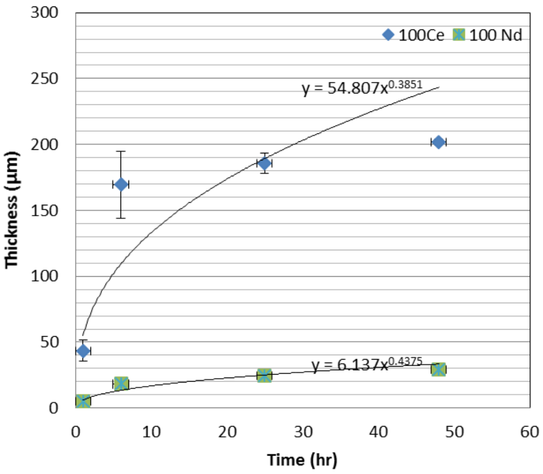

Fig. 2 Relationship between the thickness of reaction layer and experiment time

3.2 Synergistic effects between rare earth elements

The thickness of the reaction layer was defined as the depth penetrated into the cladding from the initial boundary. Depending on the rare earth alloy and time, the thicknesses of the reaction layers vary. The thickness of the reaction layers are summarized in the Table 1. In the single system, the thickness of the reaction layer was in the order of Ce, Pr, Nd, Sm, La, which coincides with the order of eutectic temperature. At the experimental temperature, Ce and Pr have eutectic reaction with Fe, and Nd, Sm and La diffused into Fe.

In the binary system, reaction layers of 50Ce50Nd, 50Ce50Pr, and 50Ce50Sm were formed over 300 μm and other alloys were formed below 100μm after test at 48 hours. There is an elemental combination that causes synergy and non-synergy effects between alloys. Ce-Nd system showed synergism when compared to the Ce or Nd single system. Sm-contained alloy showed synergism whereas La did not.

Table1 Thickness of interaction layers (μm) of binary rare earth alloys after diffusion couple test at 660°C for 48hour

48hr							
Ce	201.92	Ce	201.92	Ce	201.92	Ce	201.92
Nd	29.32	Pr	80.91	La	5.66	Sm	7.62
CeNd	318.16	CePr	413.49	CeLa	45.29	CeSm	363.30
	S		S		N		S
S : Synergism N : Non-synergism		Nd	29.32	Nd	29.32	Nd	29.32
		Pr	80.91	La	5.66	Sm	7.62
		NdPr	45.49	NdLa	9.66	NdSm	39.17
			N		N		S

4. Conclusion

This study was carried out to evaluate the compositional effect of rare earth element on the interaction behavior with HT9 at 660℃. Effects of rare earth element were tried by combining 5 elements (Ce, Nd, Pr, La, Sm) into binary alloys at a ratio of 1: 1. The thickness, composition, and elemental migration of the reaction layer were confirmed through the diffusion couple test. The results showed that all of rare earth elements reacted with HT9 to form intermetallic compounds. Reaction kinetics exhibited non-linear relationship, where the behavior of Nd in a single system is close to the diffusion. The specimens combined with Ce and Nd as well as alloys contained with Sm showed a synergistic effect on the reaction layer, while the specimens combined with La did not. Further research will be done by identifying the mechanism of synergy between elements and the microstructure, as well as the phase analysis and mechanical evaluation of the reaction layer.

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