Diffusion Behavior between the Rare Earth Element and Ferritic-Martensitic Steel

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1. Introduction

Metallic fuel has been considered as one of the probable fuel candidates for a sodium cooled fast reactor (SFR) in terms of maximizing fuel utilization, minimizing amount of spent fuel, and so on. However, metallic fuel reacts the constituent of cladding such as Fe above the reactor operating temperature to form eutectic compounds, which reduces the cladding thickness so that it affects fuel safety [1]. As the fuel burnup proceeds, rare earth elements such as Ce, La, Pr, Nd and Sm generate as fission products and then migrate at the fuel surface by the thermal gradient. Such rare earth elements are known to deteriorate a fuel cladding chemical interaction (FCCI) by accelerating cladding wastage, lowering the eutectic temperature, and so on [2].

The objective in this study is to investigate the interaction process between a rare earth element and clad material to demonstrate the effect of the rare earth element on the FCCI behavior. A diffusion couple test between the rare earth element and the cladding was performed at an SFR operating temperature, then a microstructual analysis was carried out. In this study, Misch metal was used, which contains the mixture of the rare earth element so that it is expected to be a good alternative in simulating the interaction between the rare earth and the clad material.

2. Experimental Procedure

2.1. Specimen preparation

Commercial Misch metal, produced by the Alfa Aeser company, was used as the rare earth sample in this study. Ce and La are mainly composed as 70.48% and 29.27% in weight percent, rspectively. Other elements such as Nd, Pr, Mg, and Fe were also incorporated in this material. Commercial ASTM A182 Gr. 92 steel (9Cr2W0.5MoVNb), produced by the POSCO specialty steel company, was used as the clad material in this study. Heat treatment of the Gr. 92 steel was normalized at 1040^{o}C followed by tempering at 760^{o}C for 8 hours in order to have a tempered martensite structure. Specimens were machined to an 8 mm-diameter disk, and the surface was polished down to 1200 grit using emery paper prior to the test.

2.2. Diffusion couple test

To assess the chemical reaction between the rare earth element and the steel, a diffusion couple test was carried out. The Misch metal and the clad disk were contacted with each other. The contacted specimen was wrapped with a tantalum foil to avoid any unnecessary reaction between the specimen and the jig. The coupled specimen was inserted into the screw-type jig made of 316-type stainless steel, and then the jig was tightened. After clamping, it was evacuated at a pressure of 0.01kgf/cm^2 and encapsulated using the quartz, and then the diffusion couple tests were performed. Tests were at a temperature of 660^{o}C for 1, 2, 6, 25, 48, and 120 hours. A temperature of 660^{o}C was chosen based on the designed operation temperature of an SFR, 650^{o}C, which counts 10^{o}C at the inner side of the cladding based on the temperature gradient of the fuel rod. After the tests, the specimens were pulled out of the furnace and cooled in an air environment. Then they were sectioned, mounted, and polished, followed by an optical microscopy (OM) and an SEM/EDX analysis.

3. Results and Discussion

3.1. Diffusion couple test result

Fig. 1 shows the SEM image of the diffusion coupled specimen tested at 660^{o}C for 2 hours, where it is shown that the interaction layer between the Misch metal and the clad specimen developed regardless of the test time, encroaching the clad thickness. The thickness of each interaction layer was measured by optical microscopy, and the growth of the interaction layer with the time was plotted, as shown in Fig. 2. Apart from the point at 25 hours, a relationship could be drawn as follows;

$$\Delta x^n = A \cdot t \qquad (1)$$

Where Δx denotes the penetration depth from the clad interface in ㎛, t is the exposure time in hours, and A and n are constants, which revealed that A and n are 3.90×10^5 and 3.51, respectively. This implied that the rate of growth in the interaction layer with time followed the cubic law. Generally, parabolic rate law prevails when a pure diffusion process dominates, whereas when the process other than the diffusion one controls the rate-determining step, the cubic rate law prevails [3].

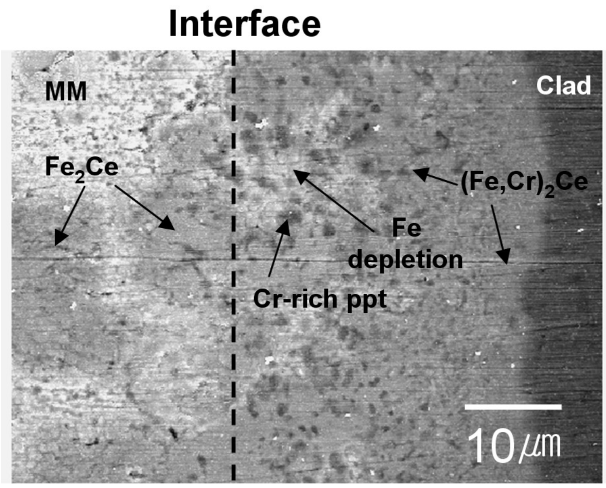

Fig. 1 SEM image of the Misch metal (left)-Gr.92 (right) coupled specimen tested at 660^{o}C for 2 hours

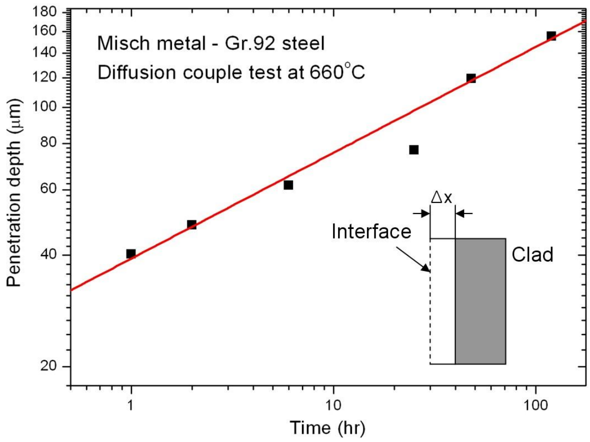

Fig. 2 Relationship between the interaction layer and the testing period

3.2. Interaction behavior between the Misch metal and the FMS at 660^{o}C

SEM observation revealed that a phase formation appeared as a result of an element migration inside the reaction layer. It was observed that Fe diffused outside the interface to form Fe_2Ce phase, and depleted inside the reaction layer as shown in Fig. 1. It seems that such a depletion of Fe may have originated from the excess diffusion of the Fe outside the initial interface. Also, a high intensity of the Cr was observed inside the reaction layer, which implied that Cr-rich precipitation was generated inside the interaction layer. It seems that Cr-rich precipitation resulted from the Fe depletion inside the layer as the region of the Cr-peak corresponded exactly to that of the Fe depletion. While Ce showed a stable distribution, little La was detected across the reaction layer. This means that Ce rather than La was the main constituent of the reaction layer.

4. Conclusions

A diffusion couple test between Misch metal and Gr.92 material was carried out to evaluate the interaction behavior of the rare earth element with the clad material under the FCCI condition. Fe in the clad side and Ce, rather than La, in the Misch metal diffused and reacted to form an interaction layer. As Fe diffused outside the clad interface, depletion of Fe caused by an excess diffusion as well as the formation of Cr-rich precipitation took place inside the interaction layer. Such process controlled the whole interaction so that it resulted in the cubic rate law of the layer growth kinetics.

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