Microstructural Evolution and Interface Stability of Cr/Mo-Coated Zirconium Alloys Near the Mo-Zr Eutectoid Temperature

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Abstract
This study is concerned with the interfacial evolutionary behavior of the Cr/Mo-coated Zr alloy, an accident-tolerant fuel cladding tube. The interfacial microstructure of Cr/Mo-coated Zr alloy and the mechanism of its formation were investigated at three temperatures: 700, 750, and 800 °C.
Keywords: ATF, Interface, Microstructure

1. Introduction

Zirconium (Zr)-based alloys have been employed as nuclear fuel cladding materials due to their low thermal neutron capture cross-section and oxidation resistance. The 2011 Fukushima Daiichi accident exposed the rapid oxidation and failure of Zr alloys at high temperatures, leading to hydrogen explosions. To enhance the accident tolerance of cladding tubes, particularly under LOCA conditions, Cr coatings on Zr cladding have emerged as a promising solution due to their robust corrosion resistance. However, the Cr/Zr interface's stability is challenged by interdiffusion, forming brittle ZrCr2 compounds and potential Cr-Zr eutectic reactions above 1330°C. Various barrier layers, including Molybdenum (Mo), have been explored to mitigate these issues. Mo is particularly promising due to its thermal properties and higher eutectic temperature with Zr. Prior studies have focused on high-temperature performance, but the behavior around the Mo-Zr eutectoid temperature (~750°C) remains less understood. This study investigates the microstructural evolution of Cr/Mo-coated Zr alloys in the 700-800°C range, focusing on the interface before and after the eutectoid reaction, aiming to enhance the safety margin of these cladding tubes.

2. Experimental process and results

Cr/Mo-coated Zr alloys were fabricated using pulsed laser deposition (PLD). The samples were annealed at three different temperatures: 700 °C, 750 °C, and 800 °C, each for 4 hours in a high vacuum environment. After annealing, the samples were cooled to room temperature within the furnace. Subsequently, specimens subjected to different heat treatments were prepared using a focused ion beam (FIB) and examined with a transmission electron microscope (TEM) equipped with an energy-dispersive spectrometer (EDS). This allowed us to analyze the microstructure and compositional distribution at the Cr/Mo-coated Zr alloy interfaces under various heat treatment conditions.

The results indicate that Mo serves as an effective diffusion barrier below 750 °C, preventing the formation of intermetallic compounds in the interfacial region. However, once the temperature reaches 750 °C, which is above the Mo-Zr eutectoid reaction temperature, significant reactions between Mo and Zr occur. This leads to the formation of four distinct phases in the interfacial region, including a large intermetallic compound, $Zr(Mo, Cr, Fe)_2$.

3. Conclusion

This study examined the microstructural evolution and composition distribution at Cr/Mo-coated Zr alloy interfaces near the Mo-Zr eutectoid temperature (700-800 °C). 730 °C, Mo effectively acts as a diffusion barrier, preventing intermetallic compound formation. However, above this temperature, Mo loses its barrier function, leading to the development of four distinct interfacial zones: β-Zr, large $Zr(Mo,Cr,Fe)_2$ intermetallic compounds, α-Zr, and the original Mo layer with fine $Zr(Mo,Cr,Fe)_2$, severely impacting interface performance.