Long-term Thermal Aging Effects on the Inter-diffusion Behavior and Microstructural Evolution at the Cr-Barrier/FMS Cladding Interface

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1. Introduction

Metallic fuels considered promising for application in Sodium Fast Reactor (SFR) exhibit progressively increasing swelling as burnup accumulates, which can bring the fuel into contact with the cladding [1]. After contact, mutual diffusion and chemical reactions between fuel constituents and fission products particularly lanthanide species and alloying elements in the cladding give rise to fuel cladding chemical interaction (FCCI) [2]. FCCI promotes the growth of interfacial reaction layers and the formation of brittle phases, and it can induce localized wastage as well as degradation of mechanical performance [3]. These effects compromise cladding integrity and can substantially elevate the risk of reactor accidents. Accordingly, suppression of FCCI is regarded as a key safety issue for long-life, high-reliability operation of Generation IV reactor systems. A representative approach to mitigate FCCI is to introduce a barrier layer between the fuel and the cladding [4]. A chromium (Cr) barrier layer can effectively inhibit chemical interactions and, when formed as a conformal and well-adhered continuous film on the cladding surface, it provides this protection with only a limited increase in thermal resistance [5]. However, while prior studies have largely focused on FCCI mitigation at the fuel cladding interface, the degradation mechanisms that may develop at the Cr/cladding-substrate interface during long-term high-temperature exposure such as interdiffusion, phase transformation, precipitation, and void formation remain insufficiently understood [6]. Because such Cr/substrate interfacial phenomena can govern barrier continuity, adhesion, crack resistance, and long-term reliability, a quantitative understanding of these mechanisms is essential for ensuring the integrity of Cr-barrier-coated cladding during extended service. In this study, long-term thermal aging tests were conducted using disk-type diffusion-couple specimens to clarify elemental redistribution and microstructural evolution at the interface between an electrodeposited Cr barrier layer and 12Cr steel. The time-dependent development of interfacial features was examined to identify the dominant interfacial evolution processes and to provide mechanistic insight relevant to the long-term reliability of Cr barrier layers for SFR metallic-fuel systems.

2. Experimental Methods

Disk-type 12Cr steel specimens were used to investigate interdiffusion behavior at the interface between an electrodeposited Cr barrier layer and a 12Cr steel substrate. The disks were machined to a diameter of 2.4 cm and a thickness of 0.2 cm. Chromium electrodeposition was conducted using an electroplating system consisting of a 12Cr steel electrode electrically connected by Pb–Sn soldering (anode), a Pb–Sn wire electrode (cathode), a hot plate and magnetic stirrer for bath heating and agitation, and a power supply for current application. A schematic illustration of the electroplating configuration is shown in Fig. 1.

The plating bath was prepared using chromic acid (CrO_3, 250 g/L) and sulfuric acid (H_2SO_4, 2.5 g/L). Electrodeposition was performed at 55 °C under continuous stirring at 150 rpm. Two current waveforms were employed: direct current (DC) electrodeposition and pulse-reverse (PR) electrodeposition [7]. Three specimens were fabricated for each condition. To minimize oxidation during subsequent heat treatment, each coated disk was individually treated with an oxidation-prevention measure. Long-term thermal aging was conducted at 650 °C for 1000 h, 2000 h, and 3000 h. After aging, cross-sections were prepared and characterized using scanning electron microscopy coupled with energy-dispersive X-ray spectroscopy (SEM–EDS) and electron backscatter diffraction (EBSD) to evaluate microstructural evolution and interfacial compositional changes.

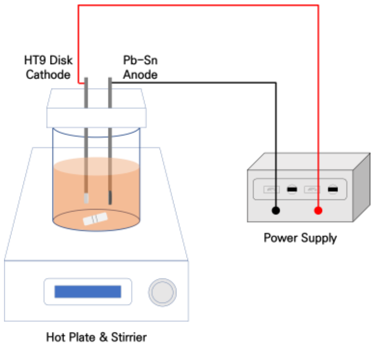

Fig. 1. Scheme of the electrodeposition configuration.

3. Results and discussion

Fig. 2 presents cross-sectional SEM micrographs and EDS elemental maps of the PR electrodeposited specimen after long-term thermal aging at 650 °C for 3000 h. At low magnification, the coating/substrate configuration appears macroscopically intact and the boundary between the Cr-rich layer and the Fe-rich 12Cr steel substrate remains distinguishable, indicating that a thick, continuous reaction layer is not evident at this scale. At higher magnification, the Cr barrier layer and the adjacent substrate region exhibit local thickness variations and compositional heterogeneity near the interface. This non-uniformity suggests spatially heterogeneous interfacial evolution, potentially governed by microstructure-dependent diffusion pathways such as grain boundaries and defects. The carbon (C) elemental map further indicates localized enrichment near the Cr/12Cr-steel interface. Because carbon is a critical solute influencing the mechanical strength of 12Cr steels through solid solution strengthening and carbide precipitation, carbon redistribution from the substrate toward the interface could be mechanically consequential. For example, carbon depletion in the adjacent substrate matrix may lead to local softening, whereas carbon accumulation near the interface may promote carbide formation and potentially alter interfacial integrity depending on the thermodynamic and kinetic conditions. It should also be noted that EDS-based carbon mapping can be sensitive to specimen-preparation artifacts therefore, the observed carbon enrichment should be validated by quantitative compositional profiling.

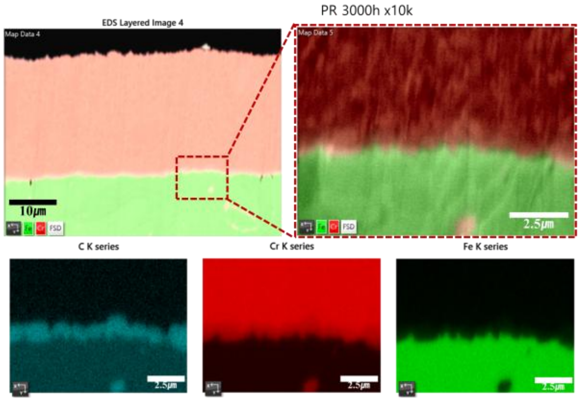

Fig. 2. Cross-sectional SEM micrograph and EDS elemental maps of the PR electrodeposited Cr-coated 12Cr steel after thermal aging at 650 °C for 3000 h.

Fig. 3 shows cross-sectional EBSD characterization of PR electrodeposited specimens aged at 650 °C for 1000 h, 2000 h, and 3000 h. After 1000 h, pronounced grain coarsening is observed near the Cr/12Cr-steel interface, suggesting that recovery and/or recrystallization followed by grain growth was locally activated during the early stage of exposure, likely driven by relaxation of residual stresses and defects introduced during electrodeposition. The interfacial coarse-grained region continues to develop up to 2000 h. After 3000 h, a region appears that is not consistently indexed as the Cr phase, implying progressive phase alteration and/or formation of reaction-derived constituents in the coating/interfacial area during prolonged aging.

In the 12Cr steel substrate, the extent of grain-boundary regions exhibiting degraded EBSD indexing quality and/or non-indexed pixels increases with aging time. This trend suggests that grain boundaries act as preferential diffusion pathways and are associated with localized compositional modification, precipitation, and/or defect accumulation. When considered together with the interfacial carbon enrichment suggested in Fig. 2, the results imply that carbon redistribution toward the interface may occur concurrently with elemental transport and may contribute to the formation of secondary compounds.

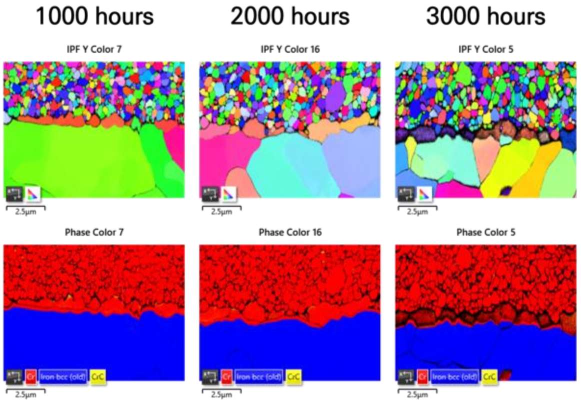

Fig. 3. Cross-sectional EBSD characterization of PR electrodeposited disks as a function of aging time at 650 °C (1000–3000 h).

4. Conclusions

This study examined the interfacial evolution of pulse-reverse electrodeposited Cr barrier layers on 12Cr steel disks during long-term thermal aging at 650 °C for up to 3000 h using cross-sectional SEM–EDS and EBSD analyses. SEM observations indicated that the coating substrate configuration remained macroscopically intact after aging, although local non-uniformity in microstructure and composition developed near the interface. EBSD results revealed pronounced grain coarsening of the Cr barrier layer adjacent to the interface at 1000 h, which continued to evolve up to 2000 h, implying recovery/recrystallization and grain growth during exposure. After 3000 h, an altered interfacial region not consistently indexed as the Cr phase was observed, suggesting progressive phase alteration and/or formation of reaction-derived constituents. In addition, EDS mapping suggested carbon enrichment near the interface, implying possible carbon redistribution and the potential formation of carbide-containing secondary products, which may affect long-term mechanical stability. Future work will focus on quantitative interfacial compositional profiling and detailed phase identification of fine precipitates to determine reaction products and to elucidate the mechanisms governing interfacial evolution during long-term aging.

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