Transactions of the Korean Nuclear Society Spring Meeting
Jeju, Korea, May 9-10, 2024

Evaluation of Chemical/Mechanical Diffusion Barriers for Mitigating FCCI in Nuclear Metallic Fuel

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***Keywords :** Metallic fuel, Fuel-cladding chemical interaction (FCCI), Cr electroplating, Zr-liner, diffusion couple experiment, Diffusion barrier.

1. Introduction

Fuel-cladding chemical interaction (FCCI) is a significant concern in sodium-cooled fast reactors (SFRs) and small modular fast reactors (SMFRs) using metallic fuels and claddings. SFRs and SMFRs are Generation-IV nuclear reactors aimed at recycling pyro-processed spent nuclear fuels [1]. Promising candidates for fuel and cladding, such as U-Zr-transuranium (TRUs) isotopes and HT9, possess excellent properties and compatibility with sodium coolants [2]. However, their interaction induces FCCI during reactor operation, resulting in the formation of brittle intermetallic compounds, known as wastage zones, which can significantly reduce reactor safety margins [3]. Mitigation strategies for FCCI include diffusion barrier coatings (chemical method), metallic liners (mechanical method), and various fuel alloys. Among these, research on fuel alloys has explored methods such as altering nuclear fuel design to an annular type or performing alloying to prevent FCCI. Despite its promising potential, this field demands extensive research due to its comparatively challenging objectives in contrast to diffusion barriers and metallic liners.

Cr-coatings and Zr-liners have emerged as the most promising diffusion barriers, demonstrating excellent compatibility with fuel and cladding, as well as desirable material properties. However, direct comparison of their diffusion barrier properties is challenging due to differing evaluation conditions in previous studies [3-7]. Therefore, comparative studies are essential for selecting suitable barriers in various reactor designs. Additionally, the applicability of barriers to the inner surface of HT9 cladding is crucial, considering its small diameter (under 8 mm). Most barrier property evaluation researches have been limited to laboratory-scale examinations conducted on disks, highlighting the need for homogeneous barrier properties and excellent adhesion to ensure sustainable FCCI limitation over prolonged cladding lifetimes [5, 6]. Investigation of the barrier effects on cladding mechanical properties is essential, particularly concerning various mechanical procedures and heat treatments during Zr-liner cladding fabrication. To address these challenges, a Cr-coating and Zr liner were applied to the inner surface of HT9 cladding, and their adhesion states were evaluated. Mechanical examinations, such as transient ramp and tensile tests, were conducted on the barrier-cladding specimens. Diffusion couple tests were also performed to compare barrier effectiveness under identical conditions.

2. Methodology

2.1 Fabrication of Cr-coated and Zr-liner cladding

The HT9 cladding, consisting of 12.0 wt% Cr, 1.0 wt% Mo, 0.6 wt% Ni, 0.6 wt% Mn, 0.52 wt% W, and 0.3 wt% V, with the remainder being Fe, was manufactured from an HT9 ingot through hot extrusion, cold pilgering, cold drawing, and multiple heat treatments. The as-fabricated HT9 cladding had dimensions of 5.6 mm inner diameter, 0.6 mm thickness, and 1 m length. Electroplating was employed to apply a Cr-coating to the inner surface of the cladding. The electroplating process involved using an electrolyte containing hexavalent chromium trioxide powder dissolved in distilled water, with sulfuric acid as a catalyst. The HT9 cladding was vertically fixed in a jig and connected to the electrolyte circulation pathway, while a Pb/Sn alloy wire (anode) was passed through the cladding (cathode) without physical contact. The electrolyte, circulated through Teflon tubes and the cladding's inner space using a pump, was heated to 70 °C, and the direction of circulation was automatically reversed every 20 seconds for uniform coating. The Cr-coating was applied by providing direct currents of 1.6 A/cm^2 for 500 minutes.

For the Zr-liner HT9 cladding, a mechanical process was employed instead of chemical deposition. This process involved using a Zircaloy-4 tube and an intermediate HT9 cladding product, which were pre-pilgered multiple times to achieve initial dimensions. A smaller-diameter pilgered Zircaloy-4 tube was inserted into the prepared HT9 intermediate product, and the couple passed through tungsten carbide-cobalt pilger rolls for four cycles of conjunction pilgering. Annealing at 750 °C for 1 hour after each cycle ensured a proper microstructure. The final Zr-liner product had HT9 and Zr-liner thicknesses of approximately 0.5 mm and 0.05 mm, respectively, and a length increase from 500 to 780 mm. The use of lubricant during pre-pilgering reduced friction, and ultra-sonic cleaning in an ethanol bath removed any residual lubricant before conjunction pilgering.

2.2 Characterization

The microstructural characteristics of the Cr-coating and Zr-liner and their interfacial contacts with the HT9 cladding were assessed using optical microscopy (OM) and micro-CT techniques. Radial sections of the Cr-coated and Zr-liner HT9 claddings were polished and examined using OM. Additionally, a non-destructive micro-CT technique was employed to create 3D models of the claddings. CT imaging was performed in the axial direction, and at least 10 CT images were obtained from different regions to evaluate barrier adhesion.

To assess the effects of the barriers on mechanical properties, various tests were conducted on untreated, Cr-coated, and Zr-lined HT9 claddings. Uniaxial and ring tensile tests were performed separately at 25 °C using the MTS 810 system. The dimensions of the specimens and examination procedures followed ASTM A370 standards. Creep rupture and transient ramp tests were conducted, excluding the Zr-liner cladding due to safety concerns arising from its thicker dimension. Schematics of the procedures for these tests were described, and the transient ramp tests were classified into transient hold and thermal tests.

The diffusion barrier property of the Cr coating and Zr liner was evaluated through diffusion couple tests at 650 °C for 25 hours using a Ce-Nd alloy as the simulated fuel. Specimens were prepared by cutting the barrier cladding into five regions, and SEM was used to observe the microstructures of the diffusion couple tested specimens.

3. Results and Discussion

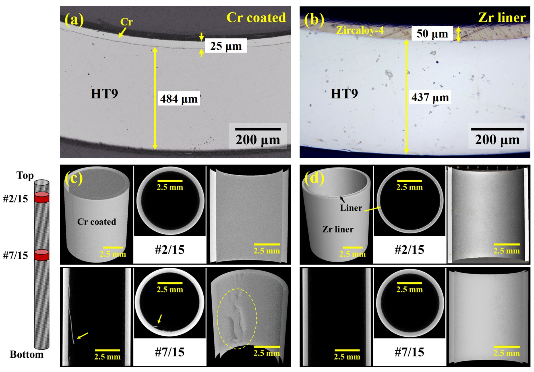

Fig. 1 OM images (a, b) and Micro-CT images (c, d) of Cr-coated and Zr-lined HT9.

Fig. 1 shows the cross-sectional microstructure and Micro-CT images of the barrier cladding formed by the Cr-coating and Zr-lining processes. The thickness of the formed barrier layers was confirmed to be 25 µm for the Cr-coating process and 50 µm for the Zr-lining process. Considering the operational environment of nuclear fuel cladding, it is advantageous to form the barrier layer with a minimum thickness to minimize its impact on thermal conductivity and tendencies of nuclear fission reactions. From this perspective, the Cr-coating process offers an advantage over the Zr-lining process due to its ability for flexible thickness control. Conversely, the Micro-CT imaging results revealed that the Zr-lining process formed a more uniform and crack-free barrier layer compared to the Cr-coating process. Since cracks in the barrier layer can serve as preferential pathways for FCCI in reactor operating conditions, the Zr-lining process is considered advantageous in terms of barrier performance.

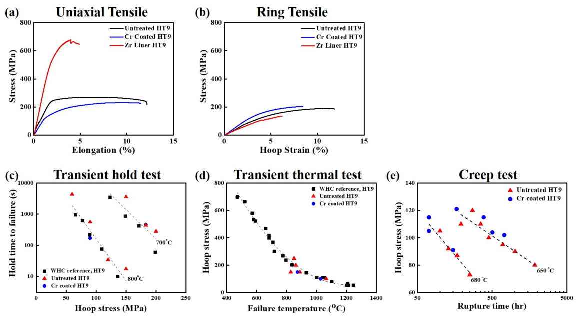

Fig. 2 Mechanical properties alteration resulting from the barrier formation process; (a, b) Tensile tests, (c, d) Transient tests, (e) Creep tests.

Fig. 2 presents the comparison results of mechanical properties according to the barrier formation process. Mechanical property tests under high-temperature conditions, including the transient test, were evaluated only for the Cr-coating process due to lab safety concerns. For room temperature tensile tests, both uniaxial and ring type tests were examined to assess the mechanical property changes, including the anisotropy of the barrier cladding. While the Cr-coating process exhibited consistent room temperature tensile characteristics regardless of the direction, the Zr-lining process showed pronounced anisotropy and higher tensile properties. This is presumed to be a result of the strong anisotropic microstructure and grain refinement induced by the pilgering process. Additionally, it was observed that the relatively thicker Zr-liner had a greater influence on the mechanical properties compared to the Cr-coating barrier. Through the evaluation of properties under high-temperature conditions, including the transient test (Fig. 2 c~e), it was concluded that the Cr-coating process had minimal impact on the intrinsic properties of the cladding. In contrast, for the Zr-lining process, inconsistencies in high-temperature mechanical properties can be anticipated due to microstructural changes such as texture and grain refinement induced by the pilgering process. These findings will be investigated in the future.

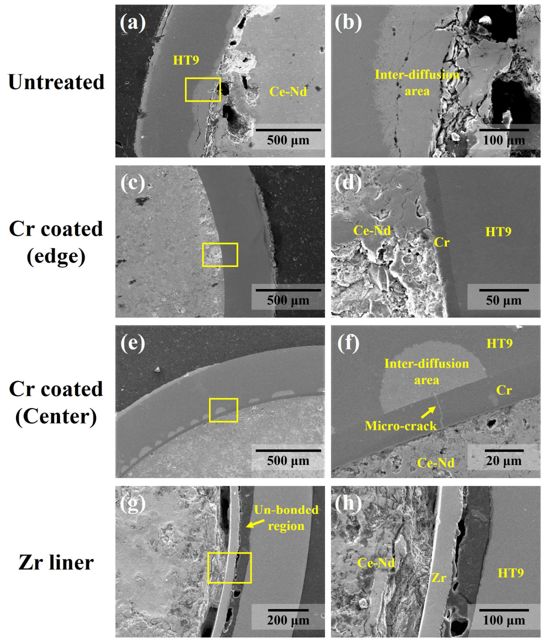

Fig. 3 Evaluation of FCCI resistance according to barrier formation process.

Fig. 3 depicts the evaluation results of FCCI resistance according to the barrier formation process. Initially, for Untreated HT9, a deep and wide interaction region was observed. In contrast, for the Cr-coating process, most of the FCCI phenomena were inhibited, but in areas with micro-cracks, some interaction regions were identified, suggesting micro-cracks acted as pathways for diffusion. Regarding the Zr-lining process, FCCI phenomena were suppressed in all areas. However, this is attributed to the presence of an un-bonded region between the cladding and Zr-liner, which prevented chemical interaction. Such un-bonded regions cause a significant decrease in the thermal conductivity of the cladding, indicating undesirable outcomes. At the current technological level, removing such un-bonded regions through mechanical processing methods like pilgering requires a high level of mechanical deformation, which is expected to directly impact the microstructure of the cladding. Thus, there exists a process dilemma wherein sacrificing the properties of the HT9 cladding is necessary for achieving excellent adhesion between the Zr-liner and HT9 cladding. In contrast, for the Cr-coating process, forming a crack-free barrier layer through controlled electroplating variables is feasible, making the chemical barrier formation process more advantageous than mechanical methods from a practical perspective.

4. Conclusions

This study provides a comparative analysis of process methodologies essential for mitigating FCCI phenomena in the application of nuclear metallic fuel. The findings from the comparison between the chemical Cr-coating process and the mechanical Zr-lining process are summarized as follows:

1) Zr-lining process demonstrates superior FCCI resistance.
2) Cr-coating process shows superiority in preserving the intrinsic properties (thermal, mechanical) of the cladding.
3) In terms of practical implementation, optimizing the Cr-coating process appears to be more feasible.

5. References

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