Electro-deposited Cr Coating Layer to Hinder Fuel Cladding Chemical Interaction in a Sodium Fast Reactor (SFR)

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1. Introduction

The disposal of high level nuclear waste is a crucial problem in many countries that have nuclear plants. Small countries such as South Korea have especially limited disposal area, and have growing stockpiles of continuously produced nuclear waste. Because the sodium cooled reactor (SFR) has the ability to transmute nuclear waste, it has attracted significant attention.

The core of a SFR contains nuclear fuels and cladding. Unlike a conventional light water reactor, sodium reactors use metallic fuels, due to their high thermal conductivity, great compatibility with sodium, and ease of fabrication [1].

Cladding is an important structural component. Its role is to maintain the integrity of the nuclear fuel and prevent fission product release, and transmit thermal energy to the sodium coolant. Ferritic-martensitic steels are typically utilized for the cladding material. They have great resistance to swelling, up to 200 dpa, have high thermal conductivity, and low thermal expansion coefficient. HT9 has been adopted for the SFR research programs in many countries.

There is a crucial concern, however, regarding reaction between the actinides or rare earth elements in the metallic fuel and the ferritic-martensitic steel cladding [2]. This phenomenon is named Fuel Cladding Chemical Interaction (FCCI). Lower melting point eutectic compounds are formed by this FCCI, which decreases the integrity of the cladding and the nuclear fuel.

The coating of the inner surface of FM steel cladding has been widely researched, with the goal of reducing FCCI. The coated layer acts as a diffusion barrier to actinides and rare earth elements. Various research teams have used diverse methods and materials for this coating. For example, Fauzia Khatkhatay et al,. [3] performed pulsed laser deposition to coat TiN and ZrN onto the surface of the cladding, and then performed Fe/Ce diffusion couple tests. TiN and ZrN coating layers of only 500nm thickness perfectly prevented chemical reactions between cerium and steel. Vahid Firouzdor et al. [4, 5] developed Yttrium Stabilized Zirconia (YSZ) and TiO_2 coating layers on steel substrates using electrophoretic deposition. Fe/Ce diffusion couple experiments showed that the TiO_2 coating reduced the solid state inter diffusion up to 83%. Moreover, no diffusion was observed in the 8μm YSZ coated Fe and Ce diffusion couple tests.

In this study, a Cr coating layer was deposited on the HT9 substrate using the electro-deposition method. When compared to other candidate materials such as TiN, ZrN, YSZ, and TiO_2, Cr achieved a higher level of comparability with the cladding. For example, the thermal expansion coefficients of Cr and Fe are similar. As a result, the Cr coating is expected to sustainably prevent chemical diffusion between the metallic fuel and cladding. Our previous publications showed that FCCI was successfully prevented by a Cr coating layer. Electro-deposition is a simple and efficient method for Cr coating. Hexavalent chromium (Cr6+) in an aqueous solution is deposited on substrate by applied electric current.

Because the service environment of cladding is the core of an SFR, the cladding is exposed to elevated temperature ~650ºC for thousands of hours. Therefore, it is essential to investigate whether the microstructure and properties of the Cr coating layer are modified by exposure at the target temperature for a prolonged time. To the best of the authors' knowledge, information about the properties of a Cr coating layer at that temperature is limited.

In this study, Cr coated HT9 disks were heated at 650ºC for 50 and 250 hours. The microstructure of the Cr coating layers was then observed, to examine the influence of the thermal cycle on the properties of the Cr layers.
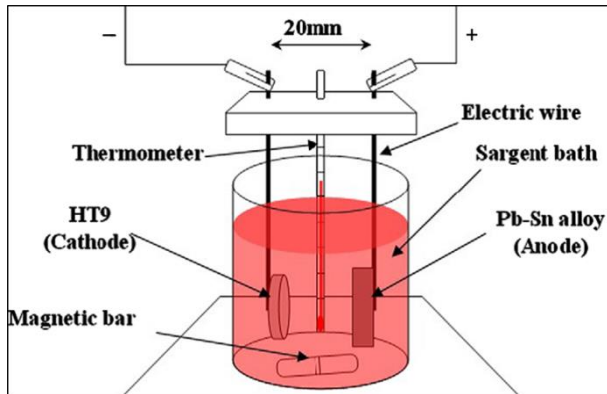
Fig 1. A schematic of the Cr electro-deposition setup [6].

2. Experiments

The detailed experimental procedure has been described in [2, 6, 7]. Therefore, only a brief description is provided here. The Cr electro-deposition was performed on polished HT9 disks. A schematic of the electro-deposition process setup is shown in Fig. 1. The HT9 disk is coated by Cr at the cathode, and Pb-Sn alloy was selected as anode. The sargent bath is a water based solution composed of 250 g/l chromic acid (CrO_3) and 2.5 g/l sulfuric acid (H_2SO_4). The temperature of the sargent bath was maintained at 50℃ and 80℃ during the electro-deposition. These two temperatures were selected because the properties of the Cr coating layer are dramatically changed at those temperatures. While the hardness of the Cr coating layer is greater and cracks are observed at 50℃ coating, the 80℃ coating resulted in a crack-free and softer Cr layer. Moreover, the thickness of the Cr coating layer at 50℃ was almost twice that of the layer coated at 80℃ [6].

The HT9 disks Cr coated at 50 and 80℃ for 70 mins were vaccum sealed in glass to prevent oxidation at high temperature. The vaccum sealed glass was then inserted into a box furnace and heated at 650℃ for 50 and 250 hours. Specimens were cut in half after the heating tests to reveal their cross section. The microstructure of the polished cross sections was observed using Scanning Electron Microscopy (SEM, Model Tescan, VEGA easy probe).
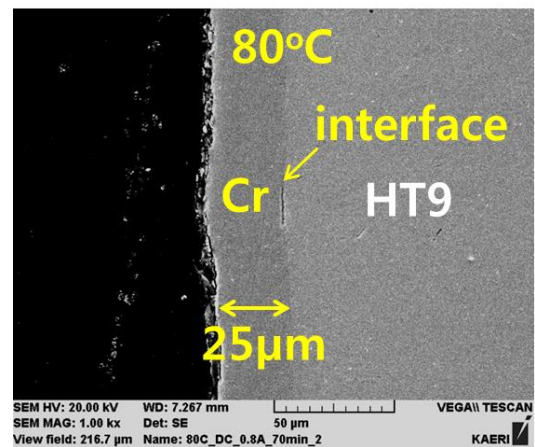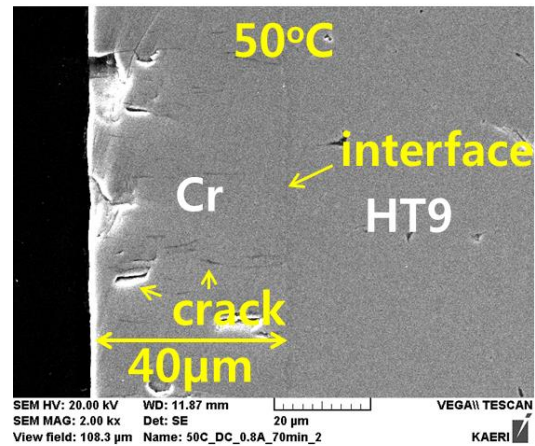

Fig 2. Cr coating layers deposited at 50℃ and 80℃, showing the difference in thickness and cracking.

3. Results and discussion

Fig. 2 shows SEM images of the Cr layers deposited on HT 9 disks at 50 and 80℃ for 70 mins. While the thickness of the Cr layer at 50℃ is 40µm, a thickness of only 25µm was obtained with the 80℃ coating. This difference in thickness implies that Cr deposition is more efficient at 50℃ than 80℃. Another important feature observed in the Cr layer at 50℃ was cracking, as indicated in the figure. The direction of most of the cracks is proportional to the growth direction of the Cr. Moreover, they seem to be indistinct, and appear to be slight defects. It is known that cracks are formed due to the volume shrinkage of chromium hydrides deposited in a Cr layer. The stress increases with the increasing thickness of the coating layers.

Fig. 3 shows SEM microstructure images of the Cr layer coated at 50℃ after heat treatment at 650℃ for 50 and 250 hours. The slight cracks observed in the as-deposited Cr layer (see Fig. 2) became more noticeable in the Cr layer after being heat treated for 50 hours. The cracks were even more obvious in the Cr layer heat treated for 250 hours. Interfacial debonding was observed at the interface between the HT9 and Cr layer. Considering the fact that actinides and rare earth

elements could easily diffuse through those deep cracks, it is strongly preferable to produce a Cr layer without cracks.

Fig. 4 shows the microstructure of the Cr layer coated at 80ºC after heat treatment at 650ºC for 50 and 250 hours. Unlike the Cr layer deposited at 50ºC (see Fig. 3), the 80ºC Cr layer maintained a crack-free microstructure even after heat treatments.
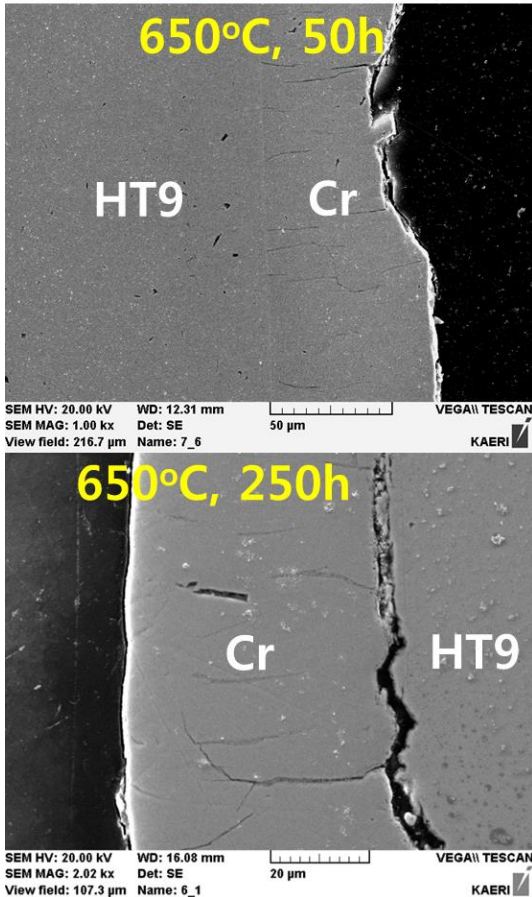

Fig 3. SEM images of the Cr layer coated at 50ºC after thermal treatment at 650ºC for the indicated durations.
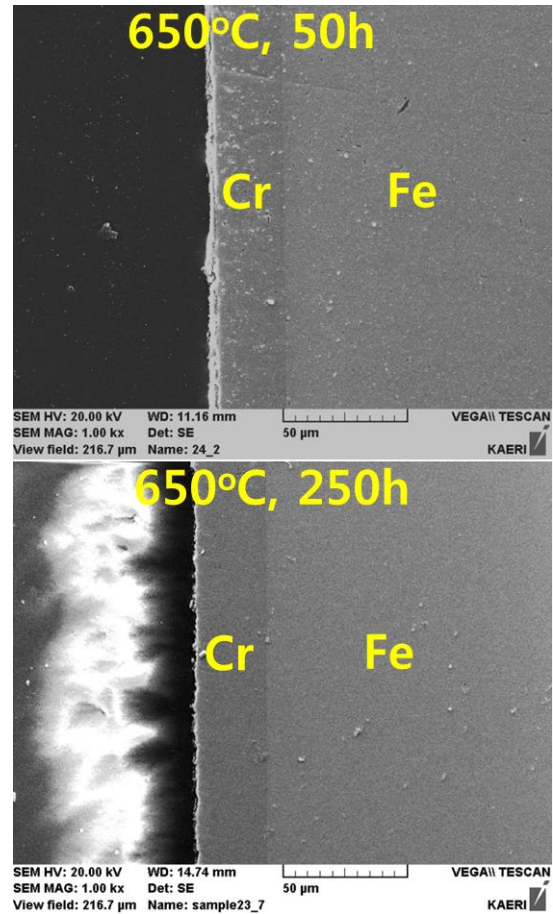

Fig 4. SEM images of the Cr layer coated at 80ºC after thermal treatment at 650ºC for the indicated durations.

4. Conclusions

Electro-deposition was performed to coat Cr layers onto HT9 disks at both 50ºC and 80ºC to hinder FCCI. Cracks were observed only in the Cr layer coated at 50ºC, due to its thickness. The observed cracks became more severe after heat treatment at 650ºC for 50 and 250 hours. Meanwhile, the Cr layer coated at 80ºC maintained a crack-free microstructure, even after the heat treatments. Because cracks are diffusion paths for actinides and rare earth materials, the 80ºC bath temperature seems to produce a better Cr coating layer to prevent FCCI.

REFERENCES

[1] I. Kim, F. Khatkhatay, L. Jiao, G. Swadener, J.I. Cole, J. Gan, H. Wang, Journal Of Nuclear Materials, 429 (2012) 143-148.

[2] J.H. Kim, H.J. Ryu, J.H. Baek, S.J. Oh, B.O. Lee, C.B. Lee, Y.S. Yoon, Journal Of Nuclear Materials, 394 (2009) 144-150.

[3] F. Khatkhatay, J. Jian, L. Jiao, Q. Su, J. Gan, J.I. Cole, H. Wang, Journal Of Alloys And Compounds, 580 (2013) 442-448.

[4] V. Firouzdor, J. Brechtl, L. Wilson, B. Semerau, K. Sridharan, T.R. Allen, Journal Of Nuclear Materials, 438 (2013) 268-277.

[5] V. Firouzdor, J. Brechtl, B. Hauch, K. Sridharan, T.R. Allen, Applied Surface Science, 282 (2013) 798-808.

[6] S.W. Yang, H.J. Ryu, J.H. Kim, B.O. Lee, C.B. Lee, Journal Of Nuclear Materials, 401 (2010) 98-103.

[7] H.J. Ryu, B.O. Lee, S.J. Oh, J.H. Kim, C.B. Lee, Journal Of Nuclear Materials, 392 (2009) 206-212.