Transactions of the Korean Nuclear Society Spring Meeting
Jeju, Korea, May 7-8, 2015

Oxidation resistant chromium coating on Zircaloy-4 for accident tolerant fuel cladding

Jung-Hwan Park*, Eui-Jung Kim, Yang-Il Jung, Dong-Jun Park, Hyun-Gil Kim, Jeong-Yong Park, Yang-Hyun Koo
LWR fuel Technology Division, Korea Atomic Energy Research Institute, 989-111 Daedeok-daero,
Yuseong-gu, Daejeon 305-353, Korea
**Corresponding author: Junghwan@kaeri.re.kr*

1. Introduction

After the Fukushima accident, accident tolerant fuel (ATF) has been widely studied, which is resistant even under a beyond-design accident. The attributes of such a fuel are approved reaction kinetics with steam, a slower hydrogen generation rate, and good cladding thermo-mechanical properties [1]. Many researchers have tried to modify zirconium alloys to improve their oxidation resistance in the early stages of the ATF development [2,3].

Corrosion resistant coating on cladding is one of the candidate technologies to improve the oxidation resistance of zirconium cladding. By applying coating technology to zirconium cladding, it is easy to obtain corrosion resistance without a change in the base materials.

Among the surface coating methods, arc ion plating (AIP) is a coating technology to improve the adhesion owing to good throwing power, and a dense deposit (Fig. 1). Owing to these advantages, AIP has been widely used to efficiently form protective coatings on cutting tools, dies, bearings, etc [4,5]. Thus, considering the advantages of AIP, we attempted to improve the corrosion resistance of zirconium using AIP. For this purpose, we coated Cr on Zircaloy-4 claddings and their corrosion behavior in high temperature and steam environment was also investigated.

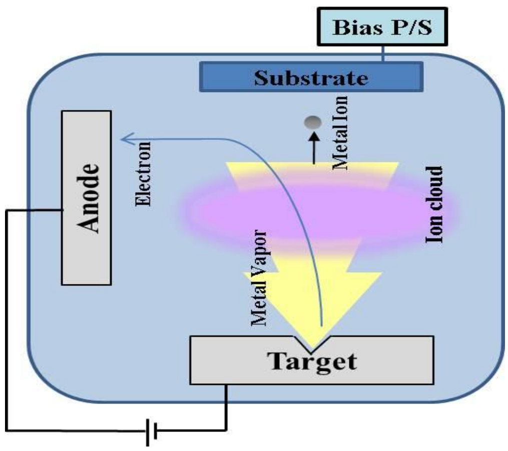

Fig. 1. Schematic representation of arc ion plating system.

2. Methods and Results

2.1 Growth and characterization of Cr film on cladding

Cr films were deposited on the Zircaloy-4 claddings by the AIP with Cr target (99.9% purity) in an argon (Ar) atmosphere. The claddings were cleaned ultrasonically in ethanol and acetone solution, and the claddings were then mounted in a vacuum chamber. The chamber was evacuated at a pressure of 1×10^{-6} Torr and heated to 120 °C to remove the residual gas absorbed on the chamber wall and claddings. When the deposition began, the Cr target was burnt by the triggers, and the arc current was kept at 80A. The partial pressure of Ar was kept at 1×10^{-2} Torr. The samples were negatively biased at 150 V during the deposition. The substrate temperatures were kept constant at 473 K.

Fig. 2 shows structure analysis of Cr film on Zircaloy-4. The XRD result exhibits that all of the diffraction peaks can be indexed as the cubic phase of Cr, and chromium oxide phases were not observed. A thin pure Cr coating was successfully formed on the Zircaloy-4 cladding by the AIP. Fig. 3 shows the cross-sectional SEM image of the Cr-coated Zircaloy-4 cladding. The Cr layer appears dense and continuous.

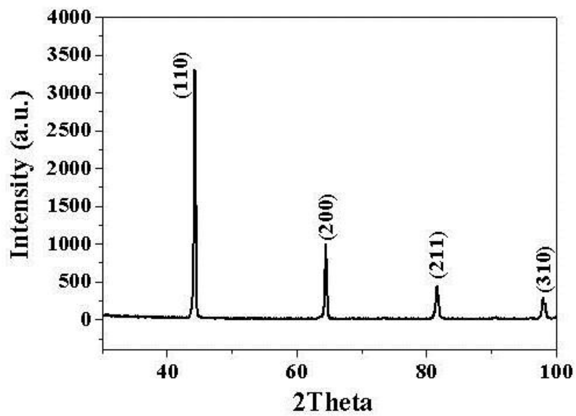

Fig. 2. Theta-2theta XRD pattern of the Cr-coated Zircaloy-4

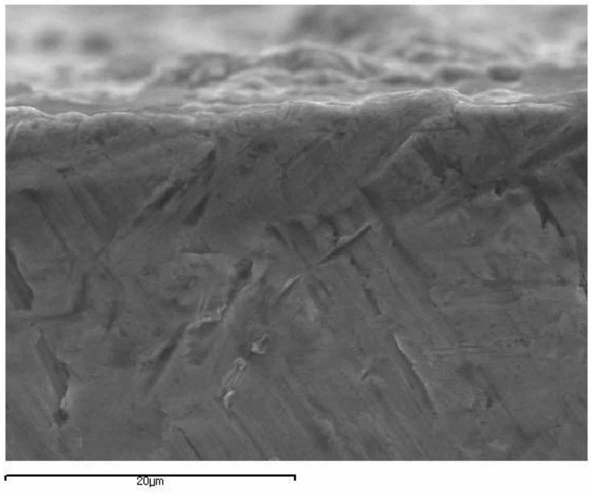

Fig. 3. SEM image of the Cr-coated Zircaloy-4 by AIP

2.2 Corrosion behavior of Cr coated Zircaloy-4

To investigate the effect of a protective Cr layer on the high-temperature oxidation behavior of Zircaloy-4, the high-temperature oxidation tests for the pristine and Cr-coated Zircaloy-4 were performed in a 1473 K steam environment using a thermo-gravimetric analyzer for up to 2000 s. The oxidation test specimens with an outer diameter, inner diameter, and length of 9.5, 8.3 and 30 mm, respectively, were cut from the longer tubes, deburred, grounded at the both ends, and cleaned in an ultrasonic bath of acetone and ethanol. The polished specimens were placed in a basket made of Pt inside the furnace. The temperature was increased at a heating rate of 50 K/min for up to 1473 K with Ar gas to prevent oxidation during the heating process. Steam was supplied into the furnace with Ar carrier gas immediately after the temperature reached 1473 K. The steam supply was maintained constant at 1473 K for 2000 s before the temperature was decreased by air cooling.

The weight gain of the pristine and Cr-coated Zircaloy-4 is shown in Fig. 4. The influence of the protective Cr layer was significant. The weight gain of the pristine Zircaloy-4 cladding was twice that of the Cr-coated Zircaloy-4 cladding. Moreover, the oxidation resistance of a Cr-coated surface is expected to be much better than the weight gain data, because the Cr-coated area is only outside the surface of the claddings. Therefore, the results of the high-temperature oxidation test showed that the Cr layer can effectively protect the claddings from a loss of coolant accident (LOCA).

The cross-sectional SEM observation of the surface region of the Cr-coated Zircaloy-4 after the high-temperature oxidation tests was executed to investigate the surface between Cr and Zircaloy-4. The results of SEM shows the spalling phenomenon was not observed after the high-temperature oxidation test. It is assumed that the adhesion property of the Cr layer by the AIP can be guaranteed regarding the thermal expansion point at up to 1473 K.

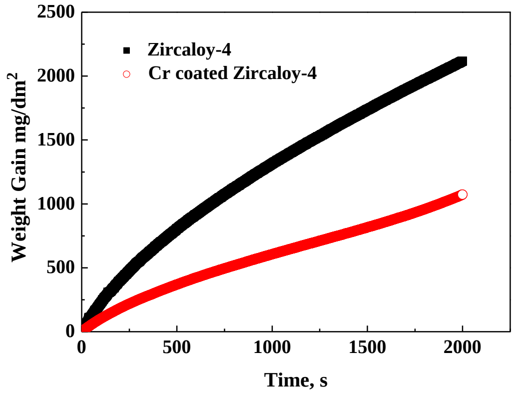

Fig. 4. Corrosion behaviors of the pristine and Cr-coated Zircaloy-4 in 1473 K steam for 2000 s.

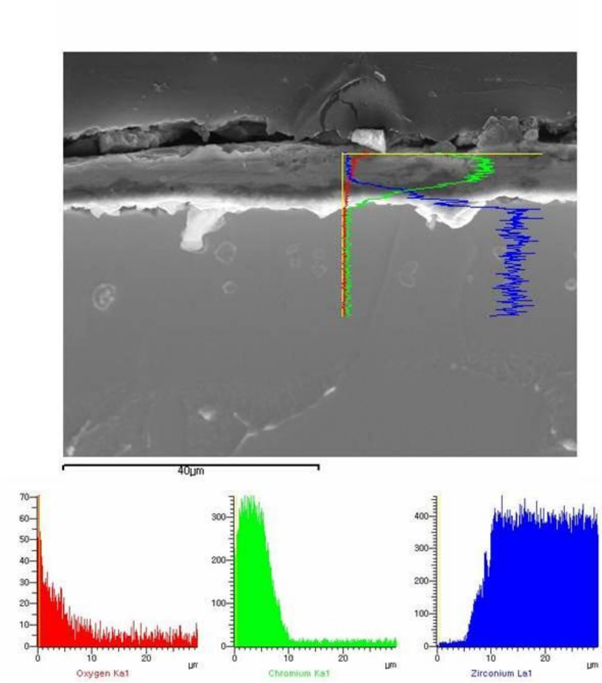

Fig. 5. Cross-sectional SEM observation of the Cr-coated Zircaloy-4 after the oxidation test.

3. Conclusions

In this study, The AIP technique for the protection of zirconium claddings from the oxidation in a high-temperature steam environment was studied. The homogeneous Cr film with a high adhesive ability to the cladding was deposited by AIP and acted as a protection layer to enhance the corrosion resistance of the zirconium cladding. It was concluded that the AIP technology is effective for coating a protective layer on claddings

ACKNOWLEGEMENT

This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea government (MSIP) (No. 2012M2A8A5000702)

REFERENCES

[1] S. Bragg-sitton, Development of advanced accident-tolerant fuels for commercial LWRs, Nuclear news, Vol. 57(3), p. 83, 2014.

[2] H. G. Kim, I. H. Kim, Y. I. Jung, J. Y. Park, Y. H. Jeong, Properties of Zr alloy cladding after simulated loca oxidation and water quenching, Nuclear Engineering and Technology (NET) Vol. 42(2), p. 193, 2010.

[3] J. H. Baek, Y. H. Jeong, Breakaway phenomenon of Zr-based alloys during a high-temperature oxidation, J. Nucl. Mater. Vol. 372, p. 152, 2008.

[4] M. Uchida, N. Nihira, A. Mitsuo, K. Toyoda, K. Kubota, T. Aizawa, Friction and wear properties of CrAlN and CrVN films deposited by cathodic arc ion plating method, Surface and Coatings Technology, Vol. 177, p. 627, 2004.

[5] S. Y. Yoon, J. K. Kim, K. H. Kim, A comparative study on tribological behavior of TiN and TiAlN coatings prepared by arc ion plating technique, Surface and Coatings Technology, Vol. 161, p. 237, 2002.