Transactions of the Korean Nuclear Society Autumn Meeting
Gyeongju, Korea, October 24-25, 2013

Cr Layer Coating on Zirconium Alloy Cladding Tube Applied to Accident Tolerant Fuel

Hyun-Gil Kim*, Il-Hyun Kim, Yang-Il Jung, Dong-Jun Park, Jeong-Yong Park, Yang-Hyun Koo
LWR Fuel Technology Division, KAERI, 989-111 Daedeok-daero, Yuseong-gu, Daejeon, 305-353, Republic of Korea
**Corresponding author: hgkim@kaeri.re.kr*

1. Introduction

It has been recognized that hydrogen generation is one of the important concerns of reactor safety, since serious reactor damage can be caused by a hydrogen explosion, as determined by the Fukushima accident. Hydrogen is generated by the corrosion reaction of zirconium alloys such as the fuel cladding, spacer grid, and channel box, and the corrosion reaction can be considerably increased with an increase in the environmental temperature [1, 2]. Thus, a decrease in the high-temperature oxidation rate of zirconium alloys is a key factor in decreasing the hydrogen generation during a nuclear power plant accident.

The current method used to increase the corrosion resistance of zirconium alloy for a nuclear application basically adjusts the alloying elements such as Nb, Sn, Fe, or Cr, and their ratios. However, the oxidation rate of zirconium alloys at a high-temperature of 1200^{o}C is not considerably changed with the alloy composition [3, 4]. Thus, it is a problem that the decrease in the oxidation rate of zirconium-based alloys at high-temperature is difficult to achieve using commercial alloying elements. New materials and concepts have been suggested to overcome the acceleration of high-temperature oxidation of zirconium alloys [5, 6].

The coating technology is widely applied in other industrial materials to reduce the corrosion and wear damages, as the corrosion and wear resistances can be easily obtained by a coating technology without a change in the base material. Thus, surface coating technology on zirconium alloy was selected in this work after technical deliberation for a decrease in the high-temperature oxidation rate, near term application, easy fabrication, economic benefit, and easy verification, although the high-temperature strength was reduced more than for other suggested technologies of hybrid and full ceramic materials. However, an optimized technology for the coating materials and coating methods for the zirconium alloy cladding must be developed for nuclear application. Thus, this work is focused on the coating techniques for both coating methods and coating materials to apply to accident tolerant fuel.

2. Methods and Results

A Zircaloy-4 cladding tube was used as a substrate with an outer diameter of 9.5 mm and wall thickness of 0.57 mm. The selection of the coated materials was based on the neutron cross-section, thermal conductivity, thermal expansion, melting point, phase transformation behavior, and high-temperature oxidation rate. After considering this point, the metal base material of Cr was selected as a coating layer for the surface coating on the zirconium-based alloy.

Two applied coating techniques are used this: for the first, pure Cr powders are attached to the Zircaloy-4 sheet and tube surface by a plasma spray (PS) coating method, and for the second, a Cr-coated layer by a PS is treated by laser beam scanning (LBS) to increase the adhesion between the Zircaloy-4 matrix and Cr-coated layer. Fig. 1 shows the plasma spray coating process to make a Cr-coated layer on the Zircaloy-4 cladding surface. To increase the adhesion property between coating layer and cladding substrate, the cladding surface was treated by sand blasting. A thermal spray machine (LCP Rev. A model; Sulzer Metco Co.) was used as the plasma coating, and Ar was used as the inactive gas. With use of a feeder, Cr powders of 75 μm in mean diameter were propelled through an Ar inlet under a pressure of 100 MPa. Six PS coatings, each for 20 sec, were applied to control the coated layer thickness. The plasma gun and sample were maintained at a 100 mm distance from each other.

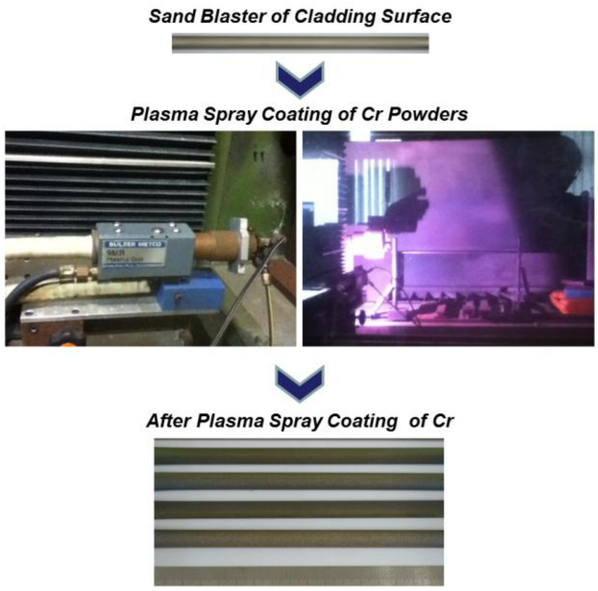

Fig. 1. Cr coating procedure by plasma spray method to make a Cr-coated layer on the zirconium cladding surface.

The microstructure and composition of a Cr-coated layer for a cross-sectional direction has been determined using optical microscopy (OM).

To evaluate the high-temperature oxidation behavior, the prepared samples were mounted in the test equipment, and a mixed gas of steam and Ar was then flowed at a 10 ml/min flow rate. The temperature of the samples rose 50°C/min, and the temperature was maintained at 1200°C for 2000 s. During the heating ramp, only Ar was continuously supplied to prevent the oxidation, and a mixed gas of steam and Ar was supplied at the target test temperature. The high-temperature oxidation test was performed on two samples for each material, and the in-suit weight gain data were acquired every 5 seconds during the test.

Fig. 2 shows a cross-sectional OM observation of the Cr-coated layer by a PS on the Zircaloy-4 cladding tube. The thickness of Cr-coated layer was determined from an OM image analysis. In this figure, the thickness of the Cr-coated layer was formed on the cladding surface after six repeated passes of spraying numbers. The mean thickness of the Cr-coated layer was about 180 μm after the surface polishing to remove a surface contamination and to make a smooth surface from an irregular surface. Thus, a uniform layer thickness can be obtained by a surface polishing process for the Cr-coated tube by a PS. In the high-magnification, it was observed that many pores having irregular shape were formed in the Cr-coated layer.

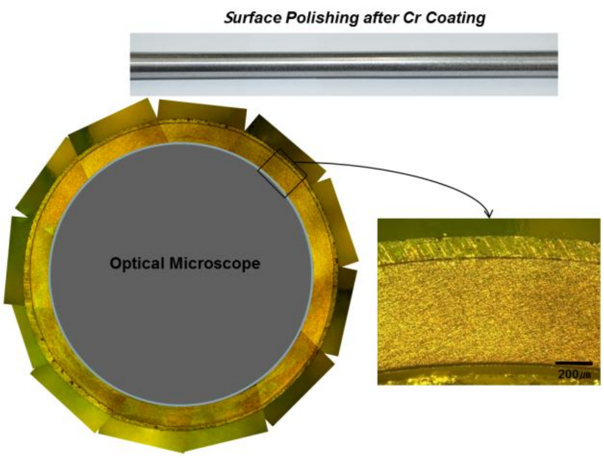

Fig. 2. Cross-sectional OM observation of the Cr-coated layer on the Zircaloy-4 cladding tube by a PS.

After the coating of the Cr particles using the PS methods, the oxidation behaviors were evaluated by the high-temperature corrosion test at 1200°C for 200 s. It is known from the high-temperature oxidation test that the oxidation resistance of the Cr-coated tube by a PS is superior to that of the Zircaloy-4 tube. From the OM observation after the oxidation test, the oxide layer of the Cr-coated tube was formed at the inside of the cladding tube. Thus, it is known that the weight gain of the Cr-coated tube is caused by the inner side oxidation of the tested tubes.

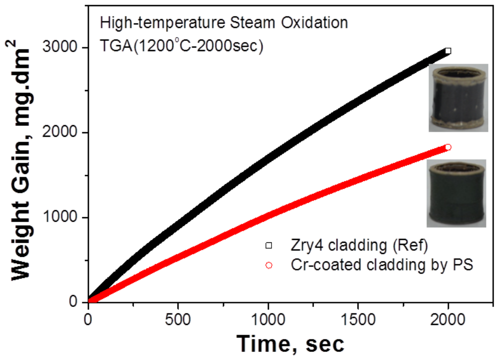

Fig. 3. Comparison of weight gain behavior between coated and un-coated Zircaloy-4 cladding tube.

3. Conclusions

The Cr-coating method by a PS was studied to reduce the oxidation rate of zirconium-based alloy in a high-temperature steam environment. The Cr-coated layer on the surface of Zircaloy-4 cladding tube is successfully obtained using the PS, and the oxidation resistance of the Cr-coated tube by a PS is superior to that of the Zircaloy-4 tube from the high-temperature oxidation test.

REFERENCES

[1] Baker, L., Just, L. C., "Studies of metal-water reactions at high-temperatures III. Experimental studies of the zirconium-water reaction." ANL-6548, 1962.
[2] Cathcart, J. V., Pawel, R. E., McKee, R. A., Druschel, R. E., Yurek, G. J., Campbell, J. J., Jury, S. H., "Zirconium metal-water oxidation kinetics. IV. Reaction ratestudies." ORNL, ORNL/NUREG-17, 1977.
[3] Kim, H. G., Kim, I. H., Jung, Y. I, Park, J. Y., Jeong, Y. H., "Properties of Zr Alloy Cladding after Simulated LOCA Oxidation and Water Quenching." Nucl. Eng. Technol., Vol. 42, No. 2, (2010) pp 193–202.
[4] Baek, J. H., Jeong, Y. H., "Breakaway phenomenon of Zr-based alloys during a high-temperature oxidation." J. Nucl. Mater., 372 (2008) pp 152-159.
[5] Hallstadious, L., Johnson, S., Lahoda, E., "Cladding for high performance fuel." Prog. Nucl. Energy, 57 (2012) pp 71-76.
[6] Cheng, B., "Fuel behavior in service accidents and Mo-alloy based cladding designs to improve accident tolerance." TopFuel 2012, Manshester, United Kingdom 2 – 6 Sep., 2012.