3D Layer Coating Technology on Zirconium Alloy Cladding Tube Applied to Accident Tolerant Fuel

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1. Introduction

To obtain the high-temperature oxidation resistance during a plant accident, the development of accident tolerant fuel (ATF) is a major concern of the fuel cladding used in LWR's[1, 2]. Hydrogen, which is generated by the corrosion reaction of zirconium alloy at high-temperature, acts as an explosion source. Thus, a decrease in the oxidation rate of zirconium alloy at high-temperature is a very important factor to improve the accident tolerance of zirconium alloys used as a fuel assembly. The current method used to decrease the corrosion rate of zirconium alloy for a nuclear application adjusts the alloying elements such as Nb, Sn, Fe, or Cr, and their ratios [3-6]. However, the oxidation resistance of zirconium-based alloys at a high-temperature is not considerably improved by the addition of alloying elements [7, 8]. Thus, the improvement of oxidation resistance of zirconium-based alloy at a high-temperature is a difficult problem when using commercial alloying elements. Research on new materials and concepts has been suggested to overcome the acceleration of high-temperature oxidation rate of zirconium-based alloys [1, 2]. A 3D laser coating of in-corrodible materials on a zirconium alloy surface can be considered in this study.

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 3D laser coating techniques for both coating methods and coating materials to apply to accident tolerant fuel.

2. Methods and Results

Many coating methods such as a plasma spray, chemical/physical vapor deposition, and laser coating have been and applied in the commercial industry field. Among them, we considered direct application methods on the fuel assembly components having a complex shape (cladding tube, guide tube, and spacer grid). Since the length of the cladding and guide tube is 4 m, a coating method having a high vacuum control is not acceptable from an economical point of view. In addition, the coating can be applied to an irregular surface shape, because the coated area is not flat (tube and irregularly formed grid). For these reasons, a 3D laser coating method supplied with powder as a coated material was developed as shown in Fig. 1.

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.

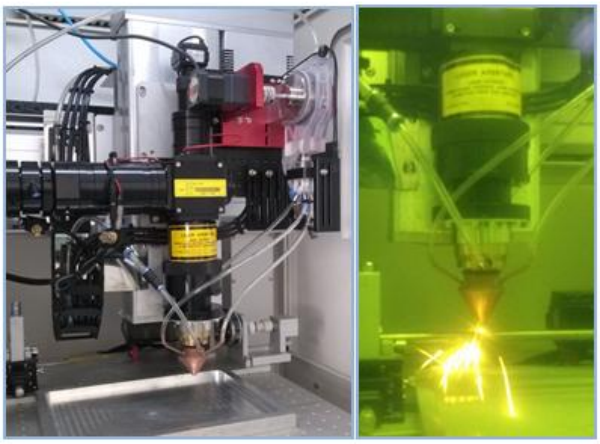

Fig. 1. A 3D laser coating equipment and processing image to make a Cr-coated layer on the zirconium cladding surface.

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

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 $50^{o}C$/min, and the temperature was maintained at $1200^{o}C$ 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-situ weight gain data were acquired every 5 seconds during the test.

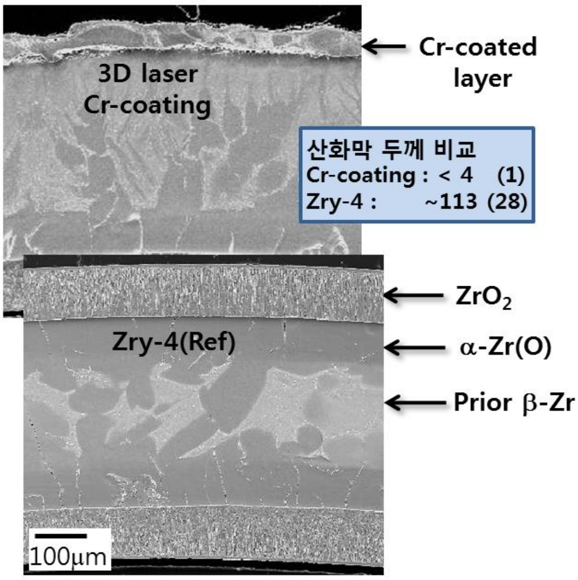

Fig. 2. Cross-sectional SEM observation of the Cr-coated Zircaloy-4 and none-coated Ziercaloy-4 cladding tubes after the high-temperature oxidation test.

After the coating of the Cr particles using the 3D laser coating technology, the oxidation behaviors were evaluated by the high-temperature corrosion test at 1200°C for 200 s. Fig. 2 shows the comparison result between Cr-coated and none-coated tubes for the cross-sectional direction. It is known from the high-temperature oxidation test that the oxidation resistance of the Cr-coated tube by a 3D laser coating is superior to that of the Zircaloy-4 tube. However, the coating thickness and the homogeneity for applying the fuel cladding will be determined after adjusting various factors such as the laser power, particle size, and coating speed.

3. Conclusions

From the Fukushima accident, it is now recognized that a hydrogen-related explosion, which is caused by the severe oxidation of zirconium alloy, is one of the major concerns of reactor safety. A coating technology for the zirconium alloy surface was considered to decrease the high-temperature oxidation rate of zirconium-based alloy.

The 3D laser coating technology using Cr powders to reduce the high-temperature oxidation rate in a steam environment was developed. The Cr-coated layer by this technology was successfully produced on the Zircaloy-4 cladding tube, and it was identified that the Cr-coated layer showed a good oxidation resistance without severe damage from the results of the high-temperature oxidation test and the microstructure analysis. From this study, the hydrogen generation of zirconium alloy caused by an excess oxidation reaction in a high-temperature steam environment can be considerably reduced by the application of the Cr coating technology using the 3D laser coating supplied with Cr powders.

ACKNOWLEGEMENT

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