Hydrothermal Corrosion Behaviors of Cr-Based Alloy and Nitride Coated Nuclear Fuel Cladding

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1. Introduction

In beyond design basis accidents (BDBA), such as the Fukushima-Daichi accident, zirconium fuel claddings have a significant safety risk of hydrogen explosion due to strong oxidation and hydrogen release [1-4]. Replacing Zircaloy cladding with accident tolerant fuel (ATF) cladding having high accident resistance, currently used as fuel cladding on pressurized water reactor, have been actively studied after the Fukushima Daiichi nuclear accident [5-7].

It is expected that the ceramic material with a low amount of hydrogen generated by reaction with a coolant during abnormal operation of the reactor and excellent in high temperature mechanical properties can be utilized as a next generation reactor cladding material. Among them, SiCf/SiC composites have attracted much attention due to its excellent corrosion resistance and high temperature strength as well as its high thermal conductivity and low neutron absorption cross section. Additionally, it does not suffer from fretting wear and hydrogen reactions. The SiC has a very good oxidation resistance due to the formation of a SiO2 protective coating in a high temperature gas environment, but mass reduction of the SiC is occurred by the dissolution of oxide layers of $SiO_2 / Si(OH)_4$ in a hydrochemical environment. high temperature Microstructural change and defects occurred during the SiCf/SiC synthesis process affect neutron irradiation deformation and corrosion behavior.

Also, deposition of protective coatings on Zircaloy cladding tubes has been considered as a near-term solution of enhanced ATF cladding. 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. Owing to these advantages, AIP has been widely used to efficiently form protective coatings on cutting tools, dies, bearings, etc. Thus, considering the advantages of AIP, we attempted to improve the oxidation resistance of Zircaloy cladding and the corrosion resistance of SiCf/SiC using AIP. For this purpose, we coated Cralloys and Cr-alloy nitrides on the claddings and confirmed their corrosion behavior in the simulated PWR primary water condition.

2. Methods and Results

Environmental barrier coatings (EBCs) can effectively prevent corrosion of fuel cladding during normal operation. Unlike metal-based ATF cladding, the SiC composite itself has excellent accident resistance. Therefore, EBC materials for SiC fuel cladding should focus on improving corrosion resistance in normal operating environments rather than improving accident resistance. In this study, we selected Cr-alloy and Cr-alloy nitrides for EBC materials which have relatively low thermal expansion coefficient, high corrosion resistance, and good mechanical properties. The Cr coating, Cr-Al coating and ternary Cr-Al-N coatings were deposited using hybrid PVD method.

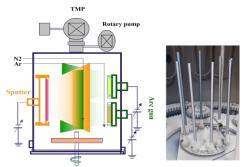


Fig. 1 Schematic diagram of hybrid PVD system

Fig.2 is shown comparison of the crystalline phases and microstructures on Cr coating and Cr-alloy coatings using the X-ray diffraction peaks and SEM images. The XRD result exhibits that all of the diffraction peaks can be indexed as the cubic phase of Cr on the Cr coating and as the various Cr-Al alloy phases on the Cr-alloy coating.

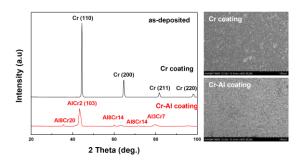


Fig. 2 X-ray diffraction patterns of Cr and Cr-alloy coatings

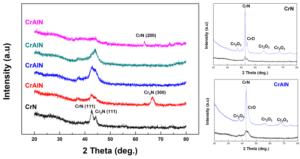
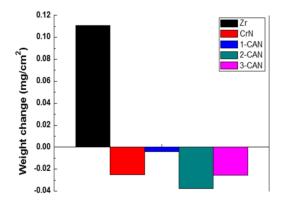


Fig. 3 X-ray diffraction patterns of CrAlN costings

Fig. 2 shows the X-ray diffraction peaks of asdeposited and stream-oxidized Cr-Al-N coatings. The high-temperature oxidation tests for CrN and CrAlN coatings were performed in a 1473 K steam environment using a thermo-gravimetric analyzer for up to 2000 sec [4]. After oxidation test, the oxidation behavior of the CrN and CrAlN coatings were compared by XRD.



Hydrothermal corrosion tests were carried out for 60 days in 310°C, 10 MPa water in static autoclave for the selection of coating materials. Fig. 4 shows the weight changes of Zr alloy, CrN coating and CrAlN coatings. Compared with a Zr alloy, CrAlN coatings showed weight loss due to the dissolution. In particular weight loss of CrAlN coating was extremely small at -0.0042 mg/cm² after 60 days.

Zr alloy and SiC-based fuel claddings are likely to be corroded in the high temperature pressurized water under neutron irradiation. Cr-based EBC can effectively prevent corrosion of Zr alloy and SiC-based fuel cladding. The CrAIN coating has excellent corrosion resistance. The EBCs will be deposited on the commercial zirconium alloy cladding tube and silicon carbide cladding tube by hybrid PVD technique and evaluate their hydrothermal corrosion behavior in a simulated PWR water loop at 360 °C and 18.5 MPa.

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