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Crack behavior of oxidation resistant coating layer on Zircaloy-4 for accident tolerant fuel claddings

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1. Introduction

Ever since the Fukushima accident, accident tolerant fuel (ATF) has been widely studied, and a coating technology for a fuel cladding surface has been considered to decrease the high-temperature oxidation rate of zirconium-based alloy. Recently, Terrani *et al.* reported the oxidation resistance of Fe-based alloys for protecting zirconium alloys from the rapid oxidation in a high-temperature steam environment [1]. Kim and co-workers also reported the corrosion behavior of Cr-coated zirconium alloy using a plasma spray and laser beam scanning [2].

However, in the operating environment, coatings are exposed to various external stresses that might cause coating damage. In particular, cracks are developed by tensile stress, and this significantly deteriorates the oxidation resistance. This tensile stress is possibly generated by the thermal cycle or bending or the irradiation growth of zirconium.

In this study, the crack behavior of Cr as protective coating layers on Zircaloy-4 was investigated under uni-axial tensile strain, and there critical strain to crack was determined in terms of applied and actual strain during the tensile test. kaer

2. Methods and Results

The crack initiation and propagation of the coating were observed with an optical microscope during the tensile force loading. The strain of the as-deposited coating was evaluated by the iso-inclination X-ray diffraction method, and the actual strain of the coating during the tensile test was calculated from the applied and initial strain of the as-deposited coating. The crack density was measured in terms of the applied strain of the coating during the tensile test.

2.1 Coating procedure

Cr films were deposited on the Zircaloy-4 plates by the arc ion plating (AIP) with Cr target (99.9% purity) in an argon (Ar) atmosphere. The plates were cleaned ultrasonically in ethanol and acetone solution, and the plates 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 200 °C. Deposition time was 2h, and the rotation speed of substrate was 2rpm.

2.2 Characterization of Cr film on cladding

The X-ray diffraction pattern of the as-deposited Cr Cr film on Zircaloy-4 is shown in Fig. 1. All of the diffraction peaks can be indexed as the cubic phase of Cr, and chromium oxide phases were not observed. A strong (200) preferred orientation was observed for Cr. Cross-sectional SEM image of Cr coating is shown in Fig.2. Smooth and featureless cross-section, suggesting a well-densified coating, was observed for Cr.

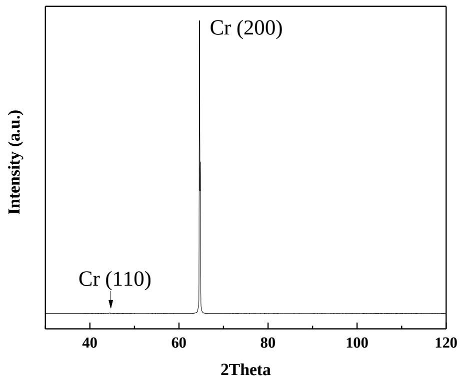

Fig. 1. XRD pattern of the Cr-coated Zircaloy-4

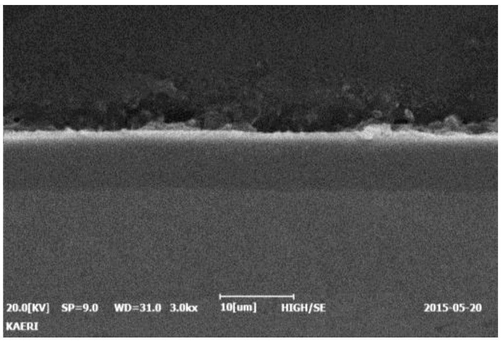

Fig. 2. SEM image of the Cr-coated Zircaloy-4 by AIP

2.2 Crack behavior under tensile stress

We measured the X-ray diffraction pattern for Cr coatings, and estimated the residual stress by the iso-inclination method [3,4]. The optic system was set up in the stress radial direction for σ_x as shown in Fig. 3. The coating strain of the as–deposited state was calculated from the following well-known formulae (1) and (2), and the resulting Eq. (3)

$$\sigma = \frac{E}{2(1+\nu)} \cot\theta_0 \frac{\pi}{180} \frac{\partial(2\theta)}{\partial(\sin^2\psi)} \quad (1)$$

$$\sigma = \varepsilon_0 E \quad (2)$$

$$\varepsilon_0 = \frac{1}{2(1+\nu)} \cot\theta_0 \frac{\pi}{180} \frac{\partial(2\theta)}{\partial(\sin^2\psi)} \quad (3)$$

Where
σ = residual stress
E = elastic modulus of coating
ν = Poisson ratio of coating
ψ = inclination angle

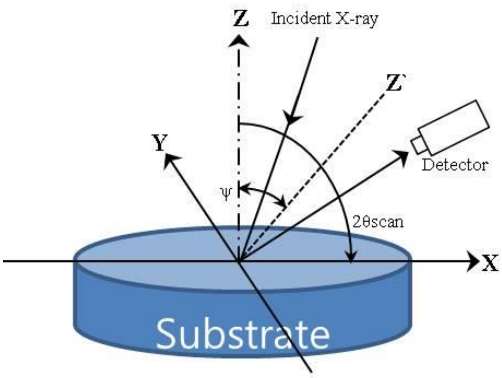

Fig. 3. The optic system for X-ray stress measurement.

The 2θ-sin^{2}ψ diagram of Cr coated Zircaloy-4 is shown in Fig. 4. From the slope of 2θ-sin^{2}ψ diagram, the residual stress and strain can be estimated using Eq (1) and (2). As results of calculation, the estimated residual stress is a compressive stress of 165MPa, and residual strain is 0.06%

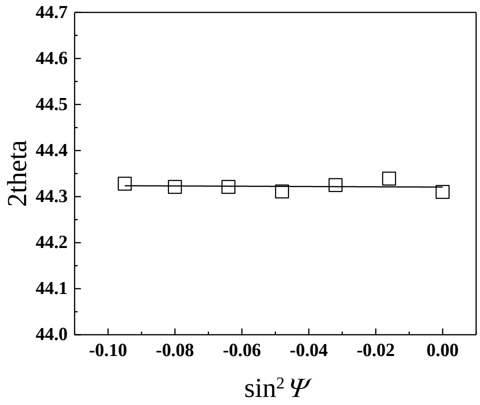

Fig. 4. 2θ-sin^{2}ψ diagram measured for the Cr coated Zircaloy-4.

Fig. 5 shows the load-displacement curves of a Cr coated and an uncoated specimen. These two loading curves were almost identical, and it was concluded that the effect of the coating process of the coating itself on mechanical property of substrate was negligible.

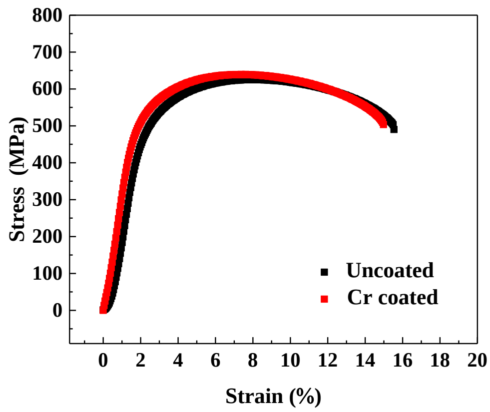

Fig. 5. Tensile test curves for a Cr coated and an uncoated specimen.

Typical surface images of the Cr coated Zircaloy-4 under tensile test are shown in Fig. 6. When applied strain was 4%, it started to crack at the edge. The number of cracks tended to increase as the strain was increased. These cracks developed normal to the applied tensile strain.

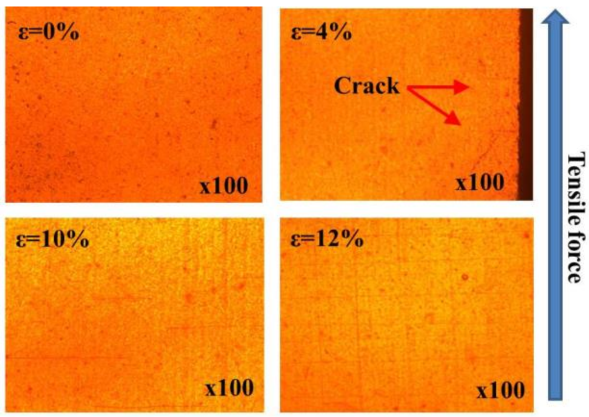

Fig. 6. Surface images of the Cr during the tensile test.

3. Conclusions

In this study, Cr was deposited by AIP on to Zircaloy-4 plate, and the crack behavior of Cr coated Zircaloy-4 under uni-axial tensile strain was observed. In addition, the strain of the as-deposited state was calculated by iso-inclination method. Coating began to crack at 8% of applied strain. It is assumed that a well-densified structure by AIP tends to be resistant to cracking under tensile strain.

ACKNOWLEGEMENT

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