

Environmental Coating Layer for the Metal-Ceramic Hybrid Fuel Cladding Tubes

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1. Introduction

For application in LWRs with suppressed hydrogen release, a metal-ceramic hybrid cladding tube has been proposed [1,2]. The cladding consists of an inner zirconium tube, and an outer SiC fiber-matrix SiC ceramic composite with surface coating as shown in Fig. 1. The inner zirconium allows the matrix to remain fully sealed even if the ceramic matrix cracks through. The outer SiC composite can increase the safety margin by taking the merits of the SiC itself. In addition, the outermost layer prevents the dissolution of SiC during normal operation.

It was reported that SiC dissolves continuously in water at about 360°C [3,4,5]. The dissolution is much faster as the composition is off the stoichiometry. Since the SiC preceramic polymer impregnation results in excess oxygen or carbon in the composite matrix, the outer environmental barrier is indispensable to prevent the expected corrosion.

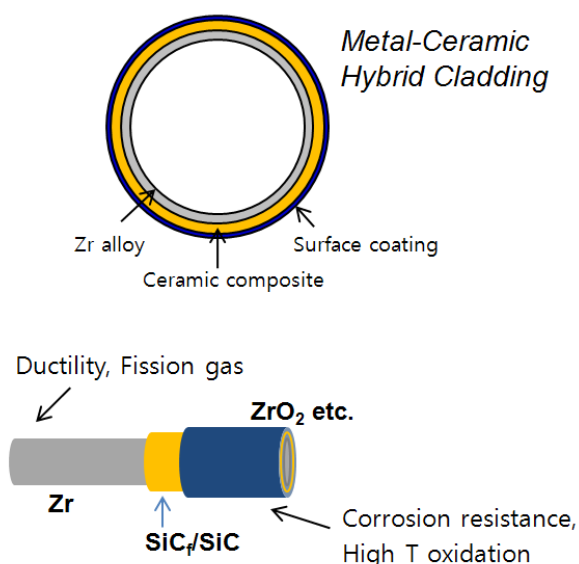


Fig. 1. Manufacturing process of the metal-ceramic hybrid fuel cladding tube. The first stage corresponds to the inner metal structure, and the second stage to the outer ceramic composite layer.

Several candidates were considered as a surface coating material. Oxides such as ZrO₂, ZrO₂-Y₂O₃, SiO₂, Cr₂O₃, and Ta₂O₅ are very stable in water. Also, the safety of fuel cladding against off-normal accidents could be increased, since the oxides can endure very high temperature. ZrO₂ in a nuclear environment has been studied a lot since it is naturally formed on conventional fuel cladding tubes [6,7]. In the case of SiO₂, the transformation of the crystal phase during irradiation was reported; however, their effect on the corrosion resistance was not investigated. Cr₂O₃ and Ta₂O₅ are quite good as a protective anti-corrosion layer, but inappropriate in terms of the neutron cross-section. A sol-gel based coating method and a synthesis of the precursor materials are being developed for the surface coating.

2. Methods and Results

ZrO₂ and ZrO₂-Y₂O₃ coatings were performed using metal-organic precursors. Precursors for the oxide coating were purchased from a commercial vendor (Kojundo, Japan). The precursors were diluted in a *n*-butyl acetate solution with a ratio of 2:3. The dilution was intended to decrease the gelation rate of the precursor, and thus increased the uniformity of the coating layer. The prepared precursors were coated on SiC plates with dimensions of 20 mm x 20 mm x 3 mm. SiC plates were dipped into the precursor for 30 sec, and then withdrawn at a rate of 3 mm/s. The coated samples were dried at 120°C for 5 min in air, and cured at 600°C for 5 min in air. After the curing process, ZrO₂ and ZrO₂-Y₂O₃ were formed on SiC plates.

A corrosion test was performed for the SiC plates with surface oxides in an autoclave with 360°C deionized water at 18.5 MPa. The surface microstructures were observed using a scanning electron microscopy (SEM).

2.1 Surface Coating

Fig. 2 shows the coated SiC plates. The uniformity of the coating was better in the case of ZrO₂-Y₂O₃. Variation in color and contrast was observed in the ZrO₂ coating, which revealed the thickness difference. Since the gelation rate of ZrO₂ was much faster than

that of $ZrO_2-Y_2O_3$, it was difficult to control the coating in the case of ZrO_2 . However, a homogeneous surface was observed in the $ZrO_2-Y_2O_3$ coating.

2.2 Corrosion Resistance

Fig. 3 shows the samples after a corrosion test. Coated SiC plates were put in a stainless steel basket during a corrosion test. The surface of SiC was changed to white. The white layer was easily removed after cleaning in acetone and ethyl-alcohol with ultrasonication. The surface of SiC revealed that the dissolution of SiC still occurred in the presence of ZrO_2 or $ZrO_2-Y_2O_3$ coating. However, the coating quality should be improved for reliable results.

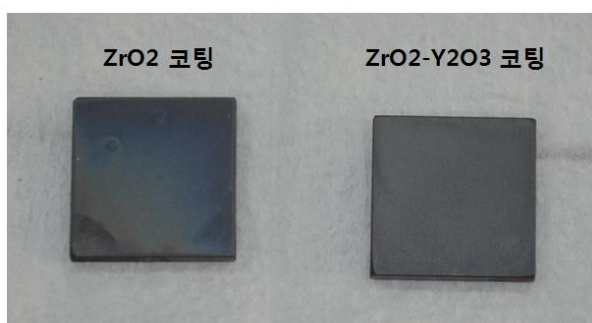


Fig. 2. Coated SiC plates with ZrO_2 (left) and $ZrO_2-Y_2O_3$ (right) by using a sol-gel dip coating method.

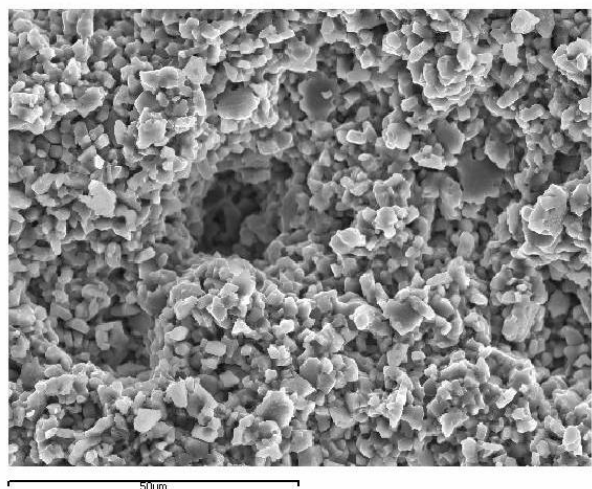
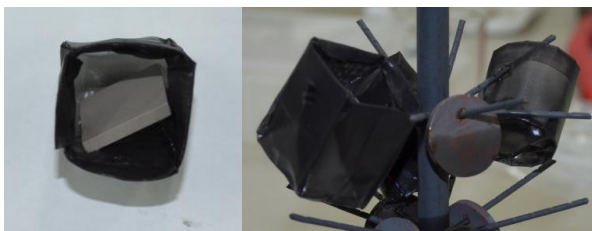


Fig. 3. Samples after the corrosion test at 360C water, along with high magnification of the SiC surface (bottom).

3. Conclusions

Surface coating of ZrO_2 and $ZrO_2-Y_2O_3$ on SiC was performed using a sol-gel dip coating method. Although the coated layer was impractical as a barrier of SiC dissolution, the preliminary test of coating materials can suggest the direction to further research.

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