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Comparative Study of Cladding Corrosion with a Protective Film of Silicon Carbide

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1. Introduction

After the Fukushima nuclear accident, development of accident-tolerant nuclear fuel is being required as a solution for suppression of reaction between nuclear fuel cladding tubes and vapor as well as prevention of hydrogen explosion. This research has been conducted to prove the oxidation resistivity of protective coats in the situation of a critical accident by producing SiC composites coated nuclear as per two types of coating methods; formula of composites using precursor under low temperature process and formula of composites using a transcritical CO_2.

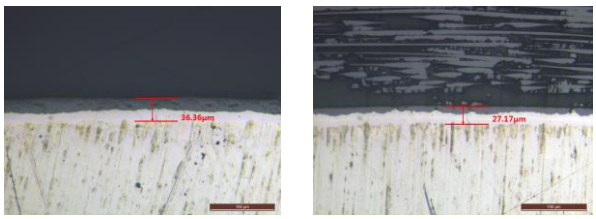
2. Method

The method of producing ceramic from polymer precursor was applied to create SiC_f/SiC composites in this study. Production process was as follows. Polycarbosilane-PCS was melted into organic solvent and became liquid state, and then was filled inside cracks of composites structure material for 24 hours.

The filled specimen was heated at 150℃ for 1 hour in hydrogen environment and then it was purified. The purified specimen was heated in Ar environment at room temperature initially and gradually elevated until 700℃ for 7 hours, then it was pyrolized by heating for 1 hour, after this, it was cooled down slowly again. Filling PCS using super critical CO_2 is the way utilizing super critical CO_2's excellent penetrating ability and solubilizing ability to non-polar organic material, specimen was produced using solvent of super critical CO_2 after covering SiC fiber outside tube of alumina. Experimental temperature was 800℃ and 1000℃, and the oxidation test of the specimen was conducted in vapor-Ar gas mixture. Oxidation was conducted for 30 minutes for the oxidation test at 800℃ while oxidation test at 1000℃ was oxidized for 15 minutes. Oxidized specimen went through mounting, grinding and polishing process and was observed for level of oxidation and microstructure using optical microscope.

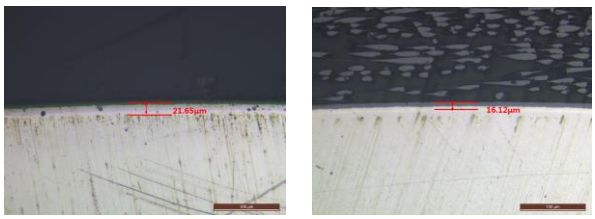
3. Results and Conclusion

To observe the changes of oxide layer thickness according to thickness of SiC fiber cover for both coating methods, specimens covered with 1 layer and 4 layers were prepared. First, Fig.1 was the specimen which was covered with 1 layer. Fig.1(a) on the left was showing the cross section of metal part without coating which had 36.36 ㎛ of oxide film and this was more oxidized compared to Fig.1(b) on the right side showing 27.17 ㎛ of oxide film. Fig.2 was the specimens covered with 4 layers. Fig.2(a) was the cross section of metal part without coating and it has 21.65 ㎛ of oxide film which was more oxidized compared to Fig.2(b), whose oxidize film at the place of fiber cover was 16.12 ㎛.



(a) (b)

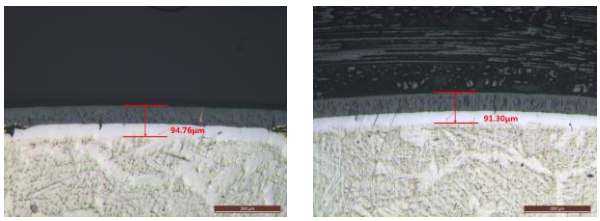
Fig.1 Cross section post oxidized of Zry-4 having SiC_f/SiC composite coating with 1 layer. (a) Metal part (b) Fiber part



(a) (b)

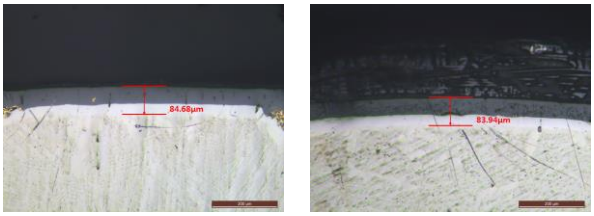
Fig,2 Cross section post oxidized of Zry-4 having SiC_f/SiC composite coating with 4 layers. (a) Metal part (b) Fiber part

Cross sections of specimen which was oxidized at 1000℃ for 15 minutes in vapor-Ar gas mixture were presented below in Fig.3 and 4. Fig.3 was specimens which were covered with 1 layer. Left side Fig.3(a) was the cross section of metal part without coating, and it had 94.76 ㎛ of oxide film. This was more oxidized than Fig.3(b), 91.30 ㎛ of oxide film at the place of fiber cover. Fig.4 was the specimens which were covered with 4 layers. Fig.4(a) is the cross section of metal part without coating and its oxide film was 84.68 ㎛ which was more oxidized than Fig.4(b) having 83.94 ㎛ of oxide film.



(a) (b)

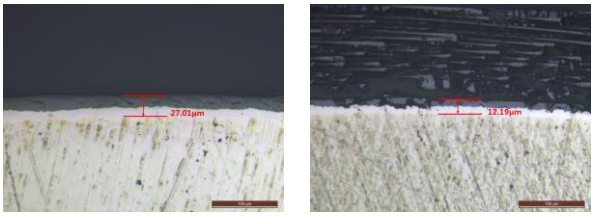
Fig.3 Cross section post oxidized of Zry-4 having SiC_f/SiC composite coating with 1 layer. (a) Metal part (b) Fiber part



(a) (b)

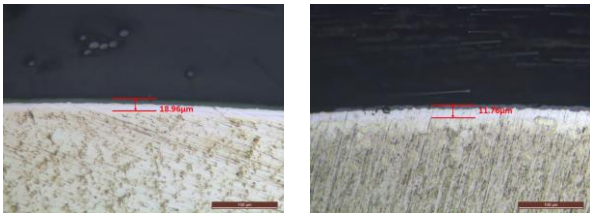
Fig.4 Cross section post oxidized of Zry-4 having SiCf/SiC composite coating with 4 layer. (a) Metal part (b) Fiber part

In the case of SiCf/SiC composites coating cladding tubes which was made as per the charge of supercritical CO2, Fig.5 is specimens which were covered with 1 layer. Fig.5(a) was the cross section of metal part without coating which had approximately 27.01 ㎛ of oxide film. Compared Fig.5(b), Fig.5(a) was more oxidized as shown that oxide film of fiber cover was 12.19 ㎛. Fig.6 is specimens with 4 layers. Fig.6(a) on the left was the cross section of metal part without coating and its oxide film was approximately 18.96 ㎛. It is more oxidized than Fig.6(b) on the right as it has 11.76 ㎛ of oxide film.



(a) (b)

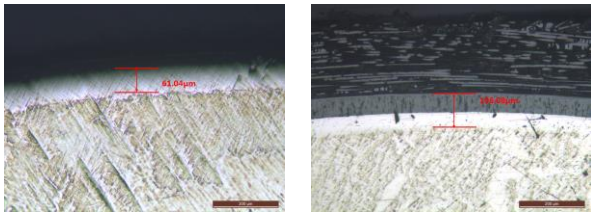
Fig.5 Cross section post oxidized of Zry-4 having SiCf/SiC super critical coating with 1 layer. (a) Metal part (b) Fiber part



(a) (b)

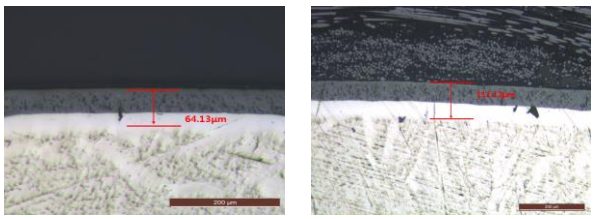
Fig.6 Cross section post oxidized of Zry-4 having SiCf/SiC composite coating with 4 layers. (a) Metal part (b) Fiber part

The pictures of cross section of specimens which was oxidized at 1000℃ for 15 minutes in vapor-Ar gas mixture were presented below in Fig.7 and 8. Fig.7 shows specimens covered with 1 layer. Fig.7(a) was the cross section of metal part without coating and its oxide film was approximately 61.04 ㎛ and it was less oxidized compared to Fig.7(b) which had 106.08 ㎛ of oxide film at the place of fiber cover. Fig.8 shows specimens with 4 layers thickness of cover. Fig.8(a) is the cross section of the metal part without coating and its oxide film is approximately 64.13 ㎛. This is less oxidized than Fig.8(b) on the right which has got 111.12 ㎛ of oxide film at the fiber cover.



(a) (b)

Fig.7 Cross section post oxidized of Zry-4 having SiCf/SiC composite coating with 1 layer. (a) Metal part (b) Fiber part



(a) (b)

Fig.8 Cross section post oxidized of Zry-4 having SiCf/SiC composite coating with 4 layers. (a) Metal part (b) Fiber part

Following table1 is a summary of suppression rates of protective coating-oxidation by two coating methods.
Suppression rate of protective coating-oxidation

$$= \frac{\text{thickness of oxide film of specimen with protective coating}}{\text{thickness of oxide film of specimen without protective coating}}$$

As the suppression rate of protective coating - oxidation is low, it has better ability of suppression of oxidization.

Table. 1 Results of high temperature oxidation experiment

Form of specimen			Suppression rate of protective coating-oxidation
SiCf/SiC composite protective coating cladding tubes made with PCS	800℃	1 layer	$\frac{27.17\ \mu m}{36.36\ \mu m} = 0.74$
		4 layers	$\frac{16.12\ \mu m}{21.65\ \mu m} = 0.74$
	1000℃	1 layer	$\frac{41.97\ \mu m}{42.34\ \mu m} = 0.99$
		4 layers	$\frac{45.65\ \mu m}{47.38\ \mu m} = 0.96$
SiCf/SiC composite protective coating cladding tubes made with Filling of super critical CO2	800℃	1 layer	$\frac{12.19\ \mu m}{27.01\ \mu m} = 0.45$
		4 layers	$\frac{11.76\ \mu m}{18.96\ \mu m} = 0.62$
	1000℃	1 layer	$\frac{106.08\ \mu m}{61.04\ \mu m} = 1.74$
		4 layers	$\frac{111.12\ \mu m}{64.13\ \mu m} = 1.73$

References

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[2] G. Amirthan, A. Udaya Kumar, and M. Balasubramanian, "Thermal Conductivity Studies on Si/SiC Ceramic Composites," Ceram. Int., 37 [1] 423-26 (2011). fa