

## Hydrothermal corrosion behavior of Cr and Al doped CVD-SiC by interdiffusion

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### 1. Introduction

SiC composite cladding is one concept of fuel cladding as an accident tolerant fuel system in light water reactors[1,2]. SiC has excellent mechanical properties at high temperature, chemical inertness, good irradiation resistance properties so that SiC is considered for various nuclear structural application[3,4]. The stability test of the SiC composite cladding under a normal operation and a severe accident condition is required to apply the SiC composites cladding to an accident tolerance fuel cladding in LWRs. The mechanical properties and the gas tightness is very important factor for fuel cladding. SiC composite cladding tube must be maintained without damaging the internal pressure generated by the fission product[5,6]. Another important issues of SiC composited cladding is hydrothermal corrosion. SiC have the disadvantage impeding their use as an ATF cladding due to hydrothermal corrosion under a normal operating condition of LWR. SiC is generally known as a corrosion resistant material. However, in LWR coolant environments, the corrosion rate of SiC is quietly high and silica dissolution and deposition problem is severe problem. Many researches have been studied recently to solve the hydrothermal corrosion of SiC composite cladding. The SiC modification for the passivation and the barrier coating to the SiC composite cladding have been studied. It is reported that the hydrothermal corrosion rate of CVD-SiC is significantly reduced in PWR simulating loop with the dissolved hydrogen control. However, there still the dissolved silica problem is remained due to the long usage time of the cladding. In this study, we decided to study improving the hydrothermal corrosion resistant of SiC own. Using interdiffusion between coating material and SiC, Cr and Al are doped in the surface of CVD-SiC. The hydrothermal corrosion results of Cr and Al doped CVD-SiC was analyzed. Doping of each element at the different interdiffusion condition was analyzed by SIMS. The hydrothermal corrosion resistance improvement of Al doped CVD-SiC was investigated.

### 2. Methods and Results

To investigate the effect of Cr and Al element doping to CVD-SiC on hydrothermal corrosion behavior, Cr and Cr/Al thin film were deposited by an arc ion plating method on CVD-SiC(TCK Co. Ltd.). Cr and Al element doping were conducted applying interdiffusion method

between the metal thin film and SiC by the heat treatment at 1800 °C for 2 hr, 4 hr. Cr and Al doping into CVD-SiC were observed by SIMS. The hydrothermal corrosion was evaluated as an accelerated experiments in the non-degassed environment using autoclave in the condition of 360 °C and 19 MPa.

Table 1. The hydrothermal corrosion condition and the weight changes of CVD-SiC and Cr doped CVD-SiC.

Specimen	Interdiffusion condition	Corrosion	Weight change (g/m <sup>2</sup> )
CVD-SiC	As-is	1 day	2.49
Cr coated CVD-SiC	As-is	1 day	2.28
Cr doped CVD-SiC	1800 °C, 2hr	1 day	2.52

Table 1 shows the hydrothermal corrosion results of CVD-SiC, Cr coated CVD-SiC and Cr doped CVD-SiC by interdiffusion. The weight change seems to be no significant differences between three specimens. Fig. 1 shows the microstructure of the specimens after hydrothermal corrosion test. The hydrothermal corrosion behavior of all three kinds of specimens reveals typical grain boundary preferential corrosion behavior, consistent with the conventional corrosion behavior of CVD-SiC[7]. The grain boundary corrosion accelerates as the corrosion progresses, it is observed that some grain appear to be shattered from the surface. Cr doping by interdiffusion does not seem to affect the hydrothermal corrosion of CVD-SiC.

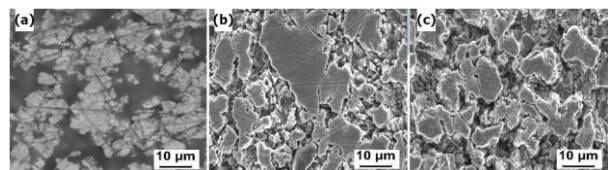


Fig. 1. Microstructure of (a) CVD-SiC, (b) Cr coated CVD-SiC, (c) Cr doped CVD-SiC after hydrothermal corrosion.

Table 2 shows the hydrothermal corrosion results of CVD-SiC, Cr/Al coated CVD-SiC, and Cr/Al doped CVD-SiC by interdiffusion. Cr/Al coating was used to dope rapidly Al element at sufficiently high temperature (1800 °C), because the melting temperature of Al coating was low(660 °C). The weight change of the Cr/Al doped CVD-SiC significantly decrease compared to the weight changes of the CVD-SiC and the Cr/Al

coated CVD-SiC. The weight change of the Cr/Al coated CVD-SiC is larger than that of the CVD-SiC. It is thought that very thin Cr/Al coating layer (~ 170 nm) fall off from the CVD-SiC by the oxidation during the hydrothermal corrosion. Fig. 2 shows the SEM surface images of each specimens after the hydrothermal corrosion test. All specimens reveal the grain boundary corrosion behavior. In the CVD-SiC and the Cr/Al coated CVD-SiC sample, the grain boundary corrosion has progressed so that the some grain fall off the surface. In the Cr/Al doped CVD-SiC sample, the grain boundary corrosion slightly occurred and the grain brake-off was not observed.

Table 2. The hydrothermal corrosion condition and the weight changes of CVD-SiC and Cr/Al doped CVD-SiC.

Specimen	Interdiffusion condition	Corrosion	Weigh change (g/m <sup>2</sup> )
CVD-SiC	As-is	12 hr	1.49
Cr/Al coated CVD-SiC	As-is	12 hr	2.40
Cr/Al doped CVD-SiC	1800 °C, 4 hr	12 hr	0.69

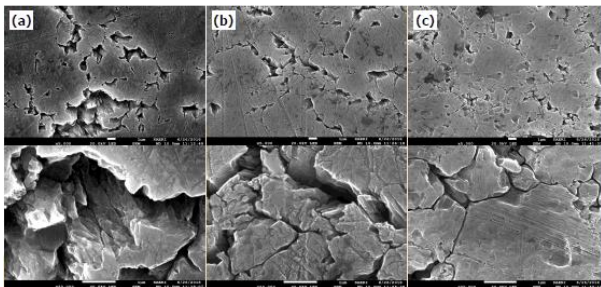


Fig. 2. Microstructure of (a) CVD-SiC, (b) Cr/Al coated CVD-SiC, (c) Cr/Al doped CVD-SiC after the hydrothermal corrosion.

In order to investigate the doping concentration and the depth, SIMS analysis was carried out. Fig. 3 shows the SIMS results of Cr/Al coated CVD-SiC and Cr/Al coated CVD-SiC by interdiffusion at 1800 °C for 4 hr. In SIMS results of the Cr/Al coated CVD-SiC, it was known that Cr and Al coating was well coated on the CVD-SiC. Cr and Al element was detected at depths after the coating due to the surface roughness of the specimen. In the SIMS results of the Cr/Al doped CVD-SiC, Cr and Al element detected up to a depth of about 10 μm. Cr element shows the depletion on the surface, while Al element was steadily diffused from the surface. It is thought that Al doping on CVD-SiC by interdiffusion affects the hydrothermal corrosion behavior. Maeda et al. reported that Al-doped SiC has a better tolerance against a corrosion due to the strong binding of Al with oxygen.[8] More detail research is needed to investigate the effect of Al doping on the hydrothermal corrosion of CVD-SiC.

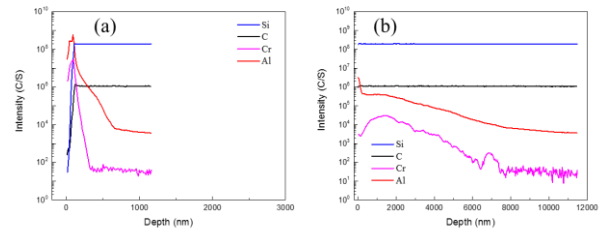


Fig. 3. SIMS results of (a) Cr/Al coated CVD-SiC, (b) Cr/Al doped CVD-SiC by interdiffusion.

### 3. Conclusions

The hydrothermal corrosion behavior of Cr and Al doped CVD-SiC was investigated. Cr and Al element was doped into CVD-SiC by interdiffusion through 1800 °C heat treatment in Cr coating CVD-SiC and Cr/Al coating CVD SiC, it is observed by SIMS analysis. The hydrothermal corrosion test of Cr and Al doped CVD-SiC was conducted in the condition of 360 °C, 19 MPa. The hydrothermal corrosion behavior of the Cr doped CVD-SiC was not different form the CVD-SiC. The hydrothermal corrosion rate of the Cr/Al doped CVD-SiC is quite less than that of the CVD-SiC. It was observed that the grain boundary corrosion is less occurred due to the Al element doping from the microstructure analysis.

### ACKNOWLEDGEMENT

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