

Dependence of Corrosion Behavior of SiC Ceramics on PWR Water Chemistry

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1. Introduction

Silicon carbide fiber-reinforced silicon carbide matrix (SiC_f/SiC) composites have a great potential for critical components of fusion and advanced fission energy systems due to the excellent high temperature properties, irradiation tolerance, inherent low activation and other superior physical/chemical properties [1]. Recently, there have also been efforts on applying the SiC_f/SiC composites to the light water reactor (LWR) fuel cladding and guide tubes as well as channel boxes for fuel assembly of the boiling water reactor (BWR) [2,3].

In spite of potential benefits of the SiC composite cladding, there are a lot of technical issues that need to be clarified for the LWR application because the previous research on the SiC_f/SiC composite has mostly been focused on high-temperature application. Especially, the corrosion resistance in the PWR water is an important parameter to insure the cladding performance under normal operating condition.

Generally, SiC ceramics are highly corrosion resistant by forming a protective SiO₂ layer in an air atmosphere. However, the corrosion resistance of SiC is largely dependent on the fabrication route of SiC and the exact environmental condition [4,5]. For example, in a high-temperature and high-pressure water or a steam environment, the corrosion resistance of SiC is decreased because the protectiveness of the SiO₂ layer is deteriorated [4-7].

Until now, the exact corrosion behavior of SiC in high temperature water is not clear due to the differences in the test samples and conditions. The kinetics of the SiC corrosion in an LWR condition, therefore, needs to be defined to confirm the possibility of a burn-up extension and the cost-benefit effect of the SiC composite cladding. In this study, we evaluated the corrosion behavior of SiC-based ceramics fabricated by various routes. We also examined the effect of PWR water chemistry on the corrosion resistance of SiC.

2. Experimental Procedure

In order to evaluate the effect of fabrication methods of SiC on the corrosion behavior, we tested three kinds of SiC ceramics, reaction-bonded SiC (RBSC), sintered SiC (SSiC), and chemically vapor deposited (CVD) SiC. The RBSC ceramics contained different amount of residual silicon, 21.4, 29.9, and 36.6 vol% for RBSC(L), RBSC(M), and RBSC(H), respectively. The corrosion test was conducted at 360°C in a vessel with an internal

volume of 100 cm³ fabricated from SUS 316 type stainless steel.

Long-term corrosion tests were also performed to examine the effect of PWR water chemistry using CVD SiC specimens in static autoclave and PWR-simulating water loops, where the dissolved oxygen (DO) content was maintained at approximately 5 ppb. The corrosion behavior of CVD SiC was also performed in the PWR-simulating water loop with a control of dissolve hydrogen (DH) content as well as the DO content.

3. Results and Discussion

3.1 Effect of Fabrication Methods of SiC Ceramics

Fig. 1 shows the weight changes of the RBSC and CVD SiC specimens after corrosion at 360°C for 7 days in the mini-autoclave containing pure water. The weight loss increases as the content of residual Si in RBSC ceramics increases because the residual Si pockets were preferentially corroded. The CVD SiC shows much lower corrosion than the RBSC specimens. Detailed analyses on the microstructure and phase evolution can be found in our previous report [4].

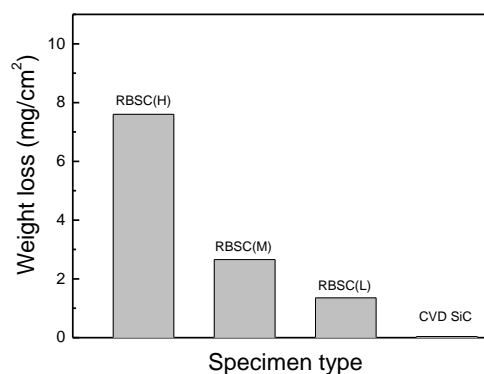


Fig. 1. Weight loss of the RBSC and CVD SiC specimens after corrosion at 360°C for 7 days in pure water [4].

We also compared the corrosion behavior of SSiC and CVD SiC specimens. Fig. 2 shows the weight changes of the SSiC and CVD SiC specimens after corrosion at 360°C for various times in pure water. In both cases, weight loss increases as the exposure time increases. The CVD SiC shows much higher corrosion resistance than the SSiC ceramic. The large deviation from the parabolic relation at the longest exposure times

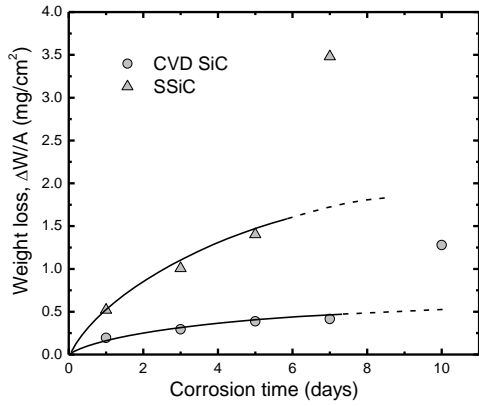


Fig. 2. Time dependence of the weight loss for the SSiC and CVD SiC ceramics after corrosion testing in 360°C water [5].

can be attributed to the preferential attack on grain boundaries [5]. The preferential intergranular corrosion leads to the disintegration of grains into water due to the weakening of grain boundaries at extended corrosion times.

3.2 Effect of PWR Water Chemistry

In order to examine the effect of PWR water chemistry on the corrosion behavior of CVD SiC specimens, we performed corrosion tests in static autoclave without DO control and PWR-simulating water loop with DO control. Fig. 3 shows the weight loss of CVD SiC after corrosion for various times in 360°C water using static autoclave and loop facility.

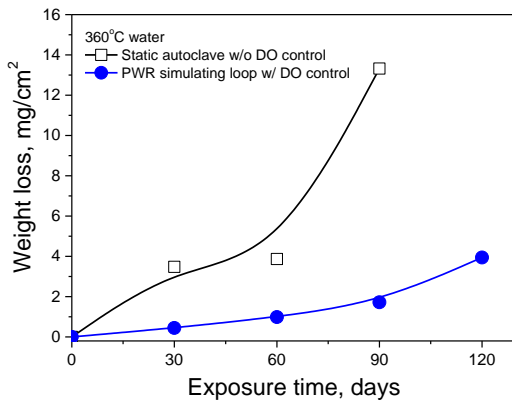


Fig. 3. Weight loss of CVD SiC specimens after corrosion for various times under 360°C water condition with and without the control of dissolved oxygen content.

It can be seen that the CVD SiC shows much higher corrosion resistance in the water loop test with the control of dissolved oxygen than in the static autoclave. However, the weight loss tends to be accelerated at extended corrosion times also in the water loop test. This was due to the preferential attack of grain boundaries [8]. These observations on the corrosion

behavior of SiC ceramics can be problematic for the application of SiC composites to the PWR fuel cladding. However, there are some dissolved hydrogen in the coolant water for current PWR fleets. Therefore, we have to also consider the effect of the dissolved hydrogen.

Fig. 4 compares the corrosion behavior of CVD SiC in PWR water loop with and without the control of DH content. The CVD SiC shows little weight loss up to 60 days and an extremely high corrosion resistance in the corrosion test with the DH control.

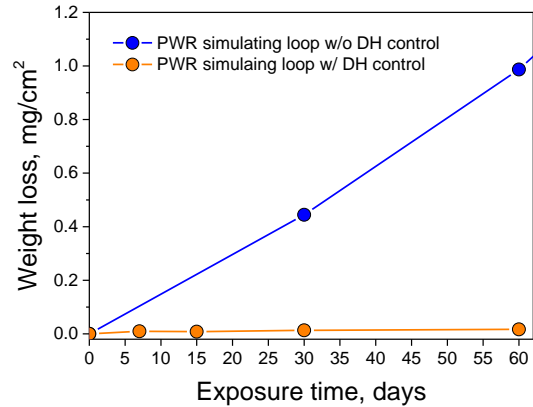


Fig. 4. Weight loss of CVD SiC specimens after corrosion for various times in PWR-simulating water loop with and without the control of DH content.

Fig. 5 shows SEM microstructure and AFM (atomic force microscopy) images for the surfaces of CVD SiC before and after corrosion test using the PWR-simulating water loop with DH control. The microstructure and surface roughness show little change after the corrosion test, suggesting a negligible corrosion of SiC under the test condition.

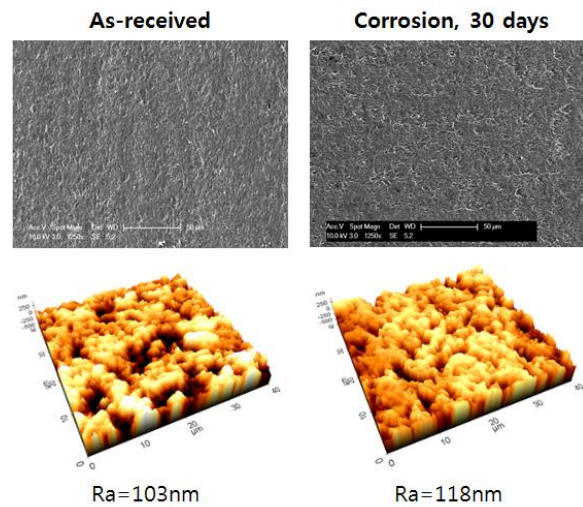


Fig. 5. SEM surface microstructures and AFM images before and after corrosion test in water loop with the control of DH content.

3. Summary

The highly pure CVD SiC ceramics exhibited a much better corrosion resistance than RBSC and SSiC specimens in high-temperature water. In the corrosion test of CVD SiC in 360°C water, the dissolution rate was much higher in the static autoclave, where the DO content was not controlled, than in the PWR-simulating water loops. Furthermore, the dissolution rate of SiC in the DH control environment was extremely low, indicating that the dissolved hydrogen significantly affects the corrosion behavior of SiC under PWR water condition, which is first observed in this study and will have a great impact on the application of the SiC composite to the accident-tolerant PWR fuel cladding.

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