Hydrothermal Corrosion and Degradation of Mechanical Properties of Silicon Carbide and Its Composites

Daejong Kim*, Hyeon-Geun Lee, Ji Yeon Park, and Weon-Ju Kim

Nuclear Materials Development Division, KAERI, 989-111Daedeok-daero, Yuseong-gu, Daejeon 305-353 ^{*}Corresponding author: <u>dkim@kaeri.re.kr</u>

1. Introduction

In recent years, there have been efforts to apply the SiC ceramics consisting of SiC monolith and SiC_f/SiC composite as a nuclear fuel cladding for the pressurized water reactors (PWR) because they have the outstanding corrosion resistance and the low hydrogen liberation rate in hot steam, which promises the larger safety margins under the severe accident conditions of PWRs [1]. In this study, the triplex SiC composites were examined for corrosion resistance in the simulated PWR primary water condition and change in mechanical strength after corrosion was evaluated.

2. Experimental Procedure

Two kinds of the triplex SiC composite tubes which have outer layers deposited at different temperatures of 1000°C (CVI) and 1200°C (CVD) were examined for hydrothermal corrosion and its influence on mechanical properties. The tubular specimens for corrosion tests have a dimension of a length of 10 mm, an inner diameter of 8.5 mm, and an outer diameter of approximately 10 mm. Corrosion tests were carried out for up to 120 days using the simulated PWR water loop in which water was deoxygenated, pressurized at 360°C under 20 MPa. Water chemistry contained that the dissolved oxygen (DO) below 5 ppb and hydrogen (DH) at approximately 2.7 ppm (35 cc/kg H_2O). Deionized water was treated at pH 6.4 with 2.2 ppm LiOH and 1200 ppm H₃BO₃. After corrosion test, hoop strength of the composite tubes was measured via internal pressurization of the polyurethane plug at room temperature using screw driven universal testing machine.

3. Results





Fig. 1. Corrosion rate of the triplex SiC composite tubes in the simulated PWR coolant environment with \sim 2.7 ppm of dissolved hydrogen.

Fig. 1 shows weight loss of the triplex SiC composite tubes consisting of monolithic CVD SiC inner layer, $SiC_{f'}SiC$ composite intermediate layer, and CVD or CVI SiC outer layer after corrosion for up to 120 days. Although they were exposed to hydrogen-containing water in which CVD SiC has been known to be excellent corrosion resistance [2], both triplex SiC tubes went through severe weight loss.

3.2 Hoop strength of SiC composite tubes after corrosion

Fig. 2 shows the load vs. displacement curves during the hoop test of the triplex SiC composite tubes. Maximum load of the triplex SiC composite tubes was dramatically reduced after corrosion.



Fig. 2. Load-displacement curves of the triplex SiC composite tubes during hoop test.

4. Discussion

Fig. 3 shows microstructure of the triplex SiC composite tubes after corrosion test in hydrogencontaining water at 360° C. An inner SiC layer which was deposited by chemical vapor deposition at 1200° C has good corrosion resistance in the simulated PWR environment as shown in Fig. 3(a). Also, there is no preferential corrosion of grain boundary which is a typical corrosion behavior of SiC ceramics [1]. After 120 days, however, a small gap was formed in the inner SiC layer. It could be associated with dissolution of the native oxide of which SiO₂ and SiO_xC_y phases have been known to be very weak to hydrothermal corrosion. The native oxide could be formed during exposure to air between two step deposition processes.

On the other hand, a SiC matrix phase suffered severe degradation after corrosion as shown in Fig. 3(b). Especially preferential corrosion occurred at grain boundary which is closely related to the formation of amorphous phase along grain boundary. Eventually many microcracks were generated in the SiC matrix phase and some SiC grains were detached from matrix. The reason of microcracks and detachment is unclear, but there is a possibility that the SiC matrix phase is contracted during the severe grain boundary corrosion.



Fig. 3. Microstructure of the triplex SiC composite tubes after corrosion showing (a) an inner SiC layer and (b) a SiC matrix phase of the SiC_{f}/SiC composite layer after 120 days and 60 days, respectively.

Fig. 4 shows microstructure of the SiC outer layers that deposited at 1000°C and 1200°C after corrosion, as shown in Fig. 4(a) and 4(b), respectively. The severe corrosion phenomena such as grain boundary corrosion, granulation, and spallation are observed in the SiC outer layer of CVI SiC triplex like the SiC matrix phase. On the other hand, the outer SiC layer, deposited at 1200°C, has still smooth surface after corrosion and there is no preferential corrosion.

Fig. 4. Microstructure of the triplex SiC composite tubes after corrosion showing the outer layers that deposited at (a) $1000^{\circ}C$ (CVI triplex) and (b) $1200^{\circ}C$ (CVD triplex) after 120 days and 30 days, respectively.

5. Conclusions

According to our previous results on hydrothermal corrosion of SiC ceramics, they have excellent corrosion resistance under the simulated PWR water although the influence of the radiolysis of water under irradiation was not considered. However, the CVI-processed SiC matrix phase of the SiC_f/SiC composite layer was severely damaged in high-temperature water that caused a significant loss of strength. Therefore, environmental barrier coatings (EBC) will play a important role for an use of SiC cemamics in PWR. It is thought that reliable EBC materials should be developed to use the SiC composite.

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