Hydrothermal Corrosion Behaviors of Various Silicon Carbide Ceramics

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1. Introduction

Silicon carbide (SiC) ceramics synthesized by chemical vapor deposition (CVD) not only have excellent mechanical properties and oxidation resistance at high temperature environment, but also have a very small neutron absorption cross section (thermal neutron absorption coefficient = 0.09 barns). Moreover, the SiC ceramics as a nuclear fuel cladding have outstanding corrosion resistance and the low hydrogen liberation rate in hot steam under the pressurized water reactor (PWR) condition [1]. In recently, there have also been efforts on applying the SiC ceramics to the PWR fuel cladding and guide tubes [2]. In this study, we investigated the corrosion behaviors of various CVD SiC ceramics under simulated PWR primary water condition.

2. Experimental Procedure

Commercial CVD β -SiC samples (A, B, C and D)in a size of 10 x 10 x 2 mm were used for the corrosion tests. Corrosion tests were carried out for up to 30 days using a simulated PWR water loop in which water was deoxygenated and pressurized at 360°C under 18 MPa. To simulate the PWR primary water chemistry, the dissolved oxygen was maintained to be below 5ppb by weight. Deionized water was treated at pH 6.4 with 2.2 ppm Li and 650 ppm B. The specimens were weighed periodically using an electronic balance with an accuracy of 0.01 mg for the weight change before and after the corrosion test. The grain size were investigated using an electron backscatter diffraction (EBSD) before the corrosion test. To obtain good quality EBSD patterns, the samples were machined by mechanical polishing with diamond suspensions and colloidal silica suspension, sequentially. Resistivity of CVD SiC samples were measured by four point probe method. The surface microstructure before and after the corrosion test was analyzed using a scanning electron microscope (SEM).

3. Results and Discussion

Table 1 shows the resistivity of the CVD SiC samples measured by using 4 point probe technique. The samples have various resistivity characteristics ranging from sub-milliohm centimeter to sub-hundred kilo ohm. The electrical properties of CVD SiC ceramics were controlled by deposition temperature, doping elements, etc..

Table I: Resistivity of SiC ceramics

Sample	Resistivity ($\Omega \cdot cm$)
А	0.91 m
В	2.05 m
С	0.43 k
D	53.3 k

Microstructures of the CVD SiC ceramics were investigated by the EBSD technique. Fig. 1 shows EBSD micrographs for the top surfaces of the as-polished SiC samples. The EBSD micrographs show microstructures of equiaxed grains at the surfaces of the all SiC samples. The average grain sizes of the four samples determined by the EBSD on the surface areas were similar between 2.1 μ m and 3.5 μ m.



Fig. 1. EBSD micrographs for the surfaces of as-polished CVD SiC samples before corrosion test; (a) sample A, (b) sample B, (c) sample C and (d) sample D.

Fig. 2 shows the weight losses of the various CVD SiC ceramics after corrosion tests in 360°C under 18 MPa deoxygenated water. Generally, it was reported that the weight loss was occurred on the hydrothermal corrosion of the SiC [3]. It can be seen that there is a considerable amount of corrosion was occurred in the loop test depending on electrical characteristic of the SiC ceramics as indicated in Fig. 1. As decreasing the resistivity of SiC ceramics, the corrosion weight loss was dramatically increased more than 3.8 mg/cm^2 from 0.09 mg/cm² after 30 days of corrosion test, in the PWR-simulating water loop.



Fig. 2. Weight losses of various CVD SiC samples at 360°C in PWR-simulating water loops as a function of the resistivity.



Fig. 3. SEM micrographs on plane-view and cross-section of the SiC samples after corrosion test for 30 days in the PWR-simulating water loops; (a) sample A, (b) sample B, (c) sample C and (d) sample D.

Fig. 3 shows the SEM micrographs on the plane-view (PV) and cross-section (CS) of the various CVD SiC samples after the corrosion tests in the simulated PWR

water environment. A considerable amount of corrosion were occurred on the sample A and B with sub-milli ohm of resistivity, while the sample C and D with very high resistivity had good hydrothermal corrosion resistance. In particular, the microstructures of sample D on the PV and CS are hardly changed after corrosion tests. A preferential corrosion of grain boundary which is a typical corrosion behavior of SiC ceramics, although there are differences. It could be associated with dissolution of the native oxide of which SiO₂, Si(OH)₄ and SiO_xC_y phases have been known to be very weak to hydrothermal corrosion. The electrical property of material affects the rate of negative oxide formation in a corrosive environment. The charge exchange rate of material is faster in the corrosive environment as increasing the electrical conductivity. Therefore, the amount of the dissolved negative oxides is increased on the SiC with low resistivity due to oxides are rapidly formed, and the corrosion rate is accelerated.

4. Summary

Hydrothermal corrosion behaviors of various CVD SiC ceramics were investigated under simulated PWR primary water conditions. The weight losses of CVD SiC samples were measured after 30 days corrosion test at 360°C under 18 MPa as a function of resistivity. As increasing electrical conductivity, the corrosion weight loss was dramatically increased at hydrothermal condition. The corrosion rate of CVD SiC was accelerated in PWR-simulating water loops, due to the low resistivity of SiC induced the high rate of dissoluble negative oxide formation in the corrosive environment.

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