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Comparison of thermal shock behavior of SiC coating deposited on graphite substrates by chemical vapor reaction and physical vapor transport

Yootaek Kim[a,*], Changsub Jang[a], Junwon Choi[a] and Eung-Seon Kim[b]
[a]*Department of the Materials Engineering, Kyonggi University, Suwon 443-760, Korea*
[b]*Korea Atomic Energy Research Institute, Daejoen 305-353, Korea*

The thermal shock behavior of α-SiC coatings deposited using the chemical vapor reaction (CVR) and the physical vapor transport (PVT) methods on graphite substrates was investigated. The correlation between the physical properties such as crystallinity, surface roughness, etc. and the thermal shock behavior of the coated specimens was evaluated. Analyses of the SiC-coating layers deposited by CVR and PVT were carried out by means of X-ray diffraction (XRD), scanning electron microscopy (SEM), and electron probe microanalysis (EPMA). A better quality of crystal growth was observed on the surfaces prepared by PVT. The surface morphology of the α-SiC-coated substrate obtained by the PVT method was denser than that obtained by CVR. More crystal facets were observed on the surface coated by PVT, which indicates that the crystallinity of the surface coated by PVT is much higher than that by CVR. Judging from confocal laser scanning microscope (CLSM) observation, the surface roughness of the layer coated by CVR looks much smoother than that by PVT. In terms of the thermal shock behavior, the PVT specimen looked more stable as compared to the CVR specimen. The crystallinity and microstructure of the α-SiC-coated surface play an important role in the thermal shock properties of the SiC coatings; the greater the degree of crystallinity and the greater the surface roughness, the more resistant the coating is to thermal shock.

Key words: Thermal shock, SiC, Chemical vapor reaction, Physical vapor transport.

Introduction

Carbon and graphite materials are chemically stable and have low densities and high strength at elevated temperatures. They are widely used as engineering materials in heaters, electrical contacts, high-temperature heat exchangers, rocket nozzles, the leading edges of aircraft wings, etc. [1, 2]. In high-temperature gas-cooled reactors (HTGRs), graphite is used as a moderating material, a structural material, a reflector material, or a fuel-element matrix material. During operation, graphite materials are surrounded by an inert coolant with few active impurities such as carbon dioxide, oxygen, and water vapor. However, the use of graphite materials has been restricted due to its poor oxidation resistance at elevated temperatures in an oxidizing atmosphere. To improve the safety of HTGRs, the oxidation resistance is very important. Some methods have been developed to form SiC coatings on graphite in order to fabricate HTGR fuel-element matrices [3,4]. Oxidation protection for carbon materials has been studied over the past 60 years, and silicon carbide (SiC) is considered the best coating material owing to its good mechanical properties, a coefficient of thermal expansion close to that of carbon, and good resistance to oxidation [5, 6]. It is thus necessary to develop a convenient method for the mass production of SiC coatings on graphite [7]. Chemical vapor reaction (CVR) coating, in which molten silicon reacts at the surface of a graphite substrate to form SiC, is an effective way to produce SiC coatings [8]. However, this process often results in a high defect density, and because oxygen can corrode the substrate through these defects, the oxidation resistance of such coatings is not sufficient at elevated temperatures. Another problem is that the coating formation behavior can be quite different depending on the carbon material used [9]. The physical vapor transport (PVT) method, a crystal growth method using radio-frequency induction heating, is another way to obtain higher-quality SiC coatings [10-12]. The present study was conducted to investigate the relationship between thermal shock resistance and the microstructure of SiC coatings deposited by CVR and PVT on top of nuclear-grade graphite substrates.

*Corresponding author:
Tel : +82-70-4024-9765
Fax: +82-31-249-9774
E-mail: ytkim@kyonggi.ac.kr

Experimental Procedures

The nuclear-grade graphite substrates used in this experiment were provided by Toyo Tanso. The properties of the graphite substrates are summarized in Table 1.

The CVR SiC coating was deposited on the graphite substrate at a temperature of 1600 °C and a pressure of 1.3 Pa for 160 min in a vacuum furnace. Physical vapor transport (PVT) was conducted at a temperature of 2000 °C for 3.5 hrs at a pressure of 200 Torr (27 kPa)

Table 1. Properties of the nuclear- grade graphite substrates used in the present study.

Fabrication	Bulk (g/cm^3)	Compressive (MPa)	Tensile (MPa)	Hardness (HSD)
Iso-molded	1.77	78	25	51

using a radio-frequency induction-heating furnace. During the coating processes, solid poly-silicone was thermally evaporated, reacted, and crystalized on the surfaces of the graphite substrates, thus forming a dense SiC coating layer.

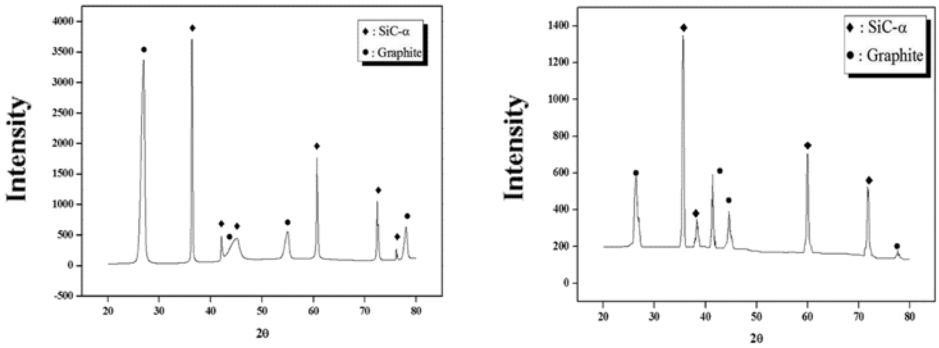
To determine the thermal shock resistance, the SiC-coated specimens were placed in a furnace preheated to 1000 °C and kept at that temperature for 1 hr. Afterwards, the specimens were taken out of the furnace and subjected to thermal shock by direct exposure to air at room temperature. Investigations of the SiC coatings prepared by each method were carried out by means of X-ray diffraction (XRD) for phase and compositional analyses, optical microscopy using a Camscope, scanning electron microscopy (SEM) for microstructural analysis, and confocal laser scanning microscopy (CLSM) for surface roughness analysis.

Results and Discussion

Fig. 1 shows the XRD patterns of the SiC coating samples deposited by CVR and PVT. The XRD analysis confirmed that the coating layers mainly consisted of α-SiC and graphite. Specimens coated by PVT showed relatively smaller graphite peaks than those coated by CVR. These results indicate that the surface density of the SiC coating layer by PVT is higher than that by CVR.

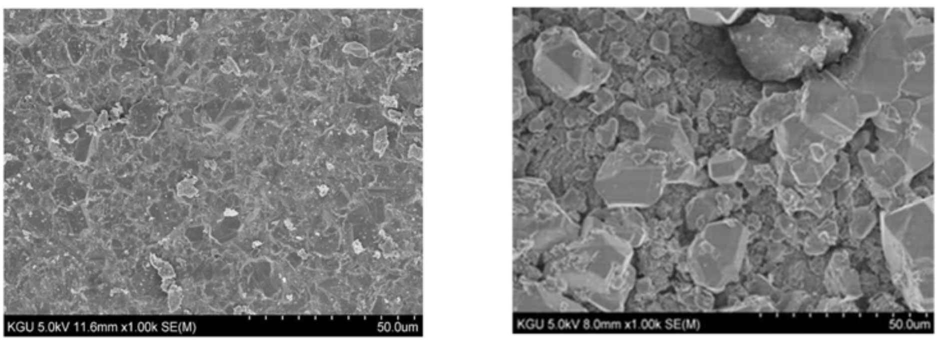
The SEM images and the surface roughness analysis with CLSM of the surfaces of the SiC-coated specimens are shown in Fig. 2 and Fig. 3, respectively. It is apparent that the SiC coating layer deposited by PVT was denser and had fewer pores than that deposited by CVR. In addition, a larger crystal size and more facets were observed on the surface of the specimen coated by PVT, which indicates that the crystallinity of the surface coated by PVT is much higher than that coated by CVR. These results are in good agreement with those of the XRD analysis. As expected from the SEM observation, the surface roughness of the specimen coated by CVR looks much smoother than that coated by PVT, as shown in Fig. 3.

Fig. 4 shows the results of the thermal shock test. Many cracks were found at the surface of the specimen prepared by CVR. By contrast, fewer cracks and less damage were found at the surface of the specimen prepared by PVT. These results are in good agreement with those from the analyses with XRD, CLSM, and SEM. This means that the factors affecting the thermal



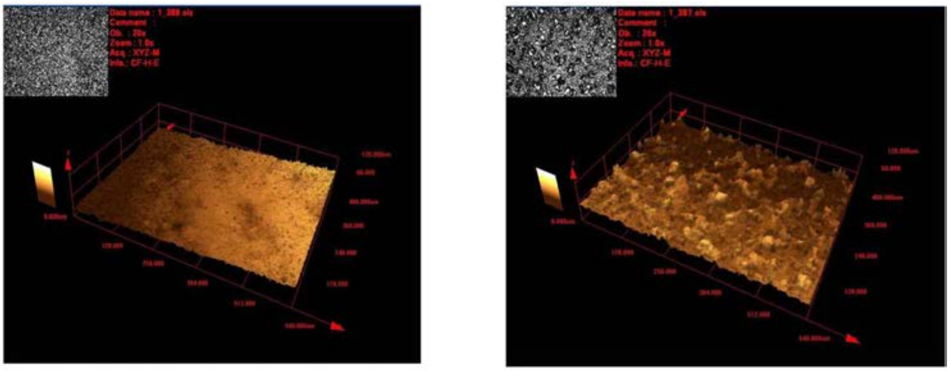
(a) (b)

Fig. 1. XRD patterns of the SiC-coated specimens deposited by (a) CVR and (b) PVT.



(a) (b)

Fig. 2. SEM images of the surfaces of SiC-coated specimens deposited by (a) CVR and (b) PVT.



(a) (b)

Fig. 3. Surface roughness analysis with CLSM of the SiC-coated specimens deposited by (a) CVR and (b) PVT.

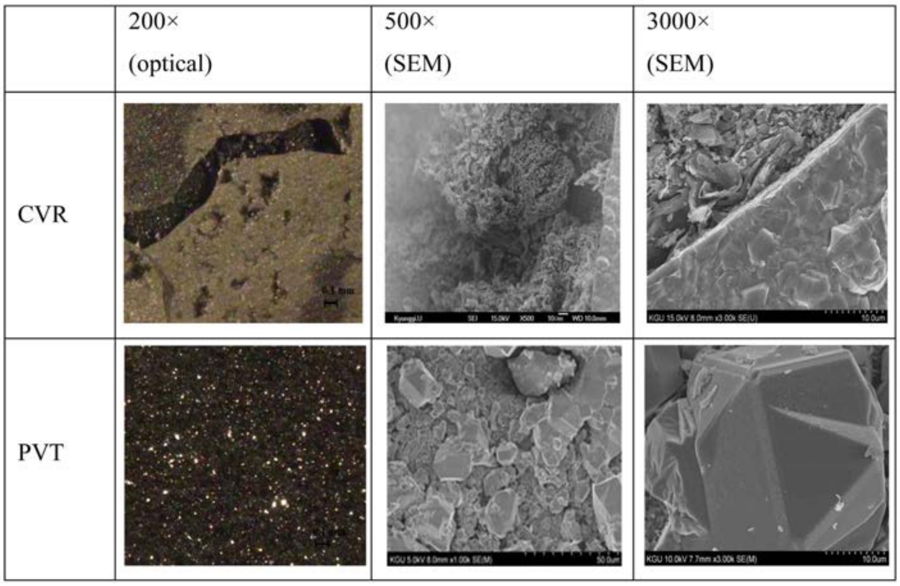


Fig. 4. Micrographs and surface morphologies of coated specimens prepared by CVR and PVT after the thermal shock test.

shock properties of the coating layer are crystallinity, surface roughness, size of crystallites, and number of surface defects such as voids and microcracks. Further research on the relationship between thermal shock resistance and oxidation resistance is anticipated for the future.

Conclusion

The surface morphology of the SiC-coated substrate obtained by the PVT method was denser than that obtained by the CVR method. More crystal facets on the coated surface were observed on the PVT specimen, which means that the crystallinity of the surface coated by PVT is much higher than that coated by CVR. The surface roughness of the CVR specimen looked much smoother than that of the PVT specimen. In terms of the thermal shock behavior of the α-SiC-coated specimens, the PVT specimen looked more stable as compared to the CVR specimen. From the microstructural observation of the coating layers after the thermal shock resistance test, less surface damage and fewer defects and modifications resulting from thermal shock stress was observed on the coating layers deposited by the PVT method. This result is in good agreement with the XRD, CLSM, and SEM analyses. Therefore, it may be speculated that the thermal shock resistance of SiC-coating layers may be enhanced by increasing the crystallinity, surface roughness, and size of crystallites on the surface and by decreasing the number of surface defects such as voids and microcracks.

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