J. Plasma Fusion Res. SERIES, Vol. 3 (2000) 233–237

# Preparation and Properties of the Composites Characterized by Grading SiC Coatings and Graphite Substrates

QUANGUI Guo*, LANG Liu, JINREN Song, JIANGANG Li[1] and YEXI He[1]
*Institute of Coal Chemistry, Chinese Academy of Sciences, P.O.Box 165, Taiyuan Shanxi, 030001, China*
*[1]Institute of Plasma Physics, Chinese Academy of Sciences, P.O.Box 1126, Hefei, Anhui, 230031, China*

(Received: 18 January 2000 / Accepted: 1 June 2000)

## Abstract

The composites with SiC coatings and graphite substrates are discussed in this paper. Microstructure observations by SEM and optical microscopes indicate the grading distribution of SiC coating. The thickness of SiC coatings and the distribution of SiC particles have a significant dependence on the reaction temperatures at which the coatings were formed. Test results show that the composites are of high density, high mechanical strength and low porosity, and the excellent performance of oxidation resistance at high temperatures up to 1200°C as well.

## Keywords:

carbon based composite, SiC coatings, graphite , microstructure

## 1. Introduction

Graphites and CFC (carbon fiber composite) are now widely used as plasma-facing materials (PFMs) in large fusion devices [1-3]. It is well known that carbon based materials have distinct disadvantages as PFM in the next generation fusion device, due to the chemical erosion (CE) [4-9] and radiation enhanced sublimation (RES) [10,11]. The CE and RES of graphite in high temperature plasma environment will result in the serious contamination of plasma and the short lifetime of PFM. To improve this behavior of graphite, some efforts have been made. One way is doping the graphite with the elements of B, Si and Ti [12-16], another way is to provide a low-Z, stable refractory material to the surface of graphite for PFMs. Boron and boron carbide based coatings have broadly been studied by Linke [17], Bolt [18] and Valentine [19]. SiC is also a low-Z, stable refractory material with many advantages when applied as a coating on the surface of carbon-based substrates. It dose not melt and is stable in an inert atmosphere up to 2900 K. While for SiC coating prepared by CVD method, thermal expansion mismatch between SiC coating and graphite substrate will cause the formation of microcracks and even spallation due to poor adherence of coating to carbon substrate [20].

The objective of this study was to develop a new kind of composites with SiC coatings and graphite substrates for plasma-facing materials using chemical vapor reaction (CVR) combined with chemical vapor infiltration (CVI). Previous efforts by the authors [21] and the other researchers [22,23] demonstrated grading SiC coatings obtained by the above fabrication methods were very adherent to graphite materials and had excellent high temperature oxidation resistance. To determine the applicability of the composites as PFMs, detailed knowledge of their properties and microstructure should be ascertained.

## 2. Experimental

### 2.1 Materials

The properties of graphite materials which were

*Corresponding author's e-mail: qgguo@21cn.com

Table 1 Basic properties of the graphite substrate

| Material type | Density ($g/cm^3$) | Porosity (%) | Bending strength(Mpa) | Compressive strength(MPa) | Electrical resistivity ($\mu\Omega m$) | Shore hardness | Ash content (ppm) |
|---|---|---|---|---|---|---|---|
| Graphite | 1.85 | 16 | 35 | 75 | 12 | 55 | 50 |

employed as substrates are listed in Table 1.

Graphite substrates were cut into cubic specimens of the size of $10 \times 10 \times 10$ mm, polished and cleaned in an ultrasonic bath with alcohol.

### 2.2 Microstructure characterization

The microstructure of the composites with grading SiC coating and graphite substrates was analyzed with a scanning electron microscope. The phase composition of the coatings was determined by XRD patterns. An optical microscope was used to determine the thickness of the coatings and the distribution region of SiC particles.

### 2.3 Properties testing

The weight of the samples before and after coating was determined with an analytical balance. Volume of samples and the composites was obtained using the Archimedes law. The density of the composites was calculated. For the samples with SiC coatings, the compressive strength was tested and oxidation resistance was performed in a muffle furnace in static air at 1200°C.

## 3. Results and Discussion

### 3.1 Preparation of the composites

The composites of SiC coatings and graphite substrates were obtained in a high-temperature furnace by the following ways: when cubic graphite samples were heated to the temperatures ranged from 1600–1800°C, vaporized silicon was formed and it reached the surface of graphite and the chemical reaction occurred: $Si + C \rightarrow SiC$. The reaction occurred not only at the surface, but also in the interior of substrates due to the infiltration of Si (g) through the pores, thus the composites of grading SiC coatings and graphite substrates were prepared.

The composites with the density of 1.88–2.28 g/$cm^3$ were prepared depending on the temperatures. As indicated in Table 2, the reaction temperature has a significant influence on the weight-gain and the density of the samples.

The effects of the temperature on the weight-gain and the density of the composites can be illustrated by the formation mechanism of SiC. At the reaction temperature of 1600°C, due to the low pressure and the relatively low diffusion rate of Si (g) into the interior of the substrates, the formation of SiC is mainly on the surface and the penetration of Si (g) into the interior is very limited. At 1700°C, SiC is not only formed at the surface, but also at the center of the sample by the diffusion of Si through the inner pores. At higher temperature of 1800°C, due to the rapid reaction between Si and carbon substrate, the primarily formed SiC layer at the surface of substrate acts as a diffusion barrier against Si-mass transfer by the gas phase, resulting in the small weight-gain of the samples and the relatively low density of the composites.

Table 2 The effect of reaction temperature on the weight-gain and the density of the samples

| Reaction temperature(°C) | 1600 | 1700 | 1800 |
|---|---|---|---|
| Weight-gain(%) | 7.31 | 20.95 | 7.26 |
| Density of the sample($g/cm^3$) | 1.94 | 2.15 | 1.94 |

### 3.2 Phase composition and microstructure of the coatings X-ray diffraction method was used to determine the phase compositions of the coatings.

Because no graphite diffraction peaks and only SiC crystal diffraction peaks were observed, as shown in Fig. 1, from which it can be concluded that the whole surface of graphite substrate was covered with SiC particles. Fig. 2 was surface photography of the composites by the optical microscope prepared in the temperature of 1700°C. It revealed that the coatings on the surface of graphite substrate were mainly composed of SiC particles in the size of 5–10 $\mu$m.

Transversal sections of the composites were observed by scanning electron microscope and optical microscope to characterize the microstructure of the coatings. SEM images and optical microscope observation verified the microstructure of the grading

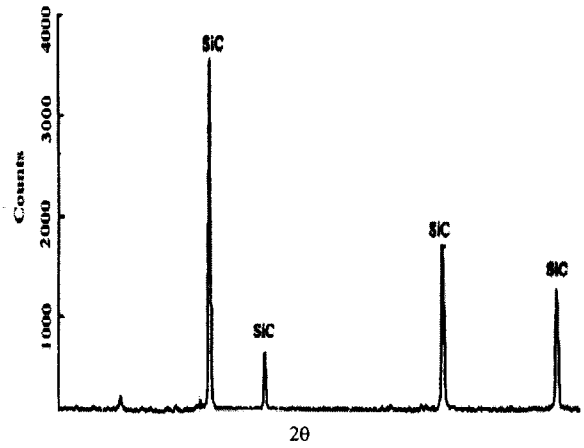


Fig. 1 XRD pattern of the coatings of the composites

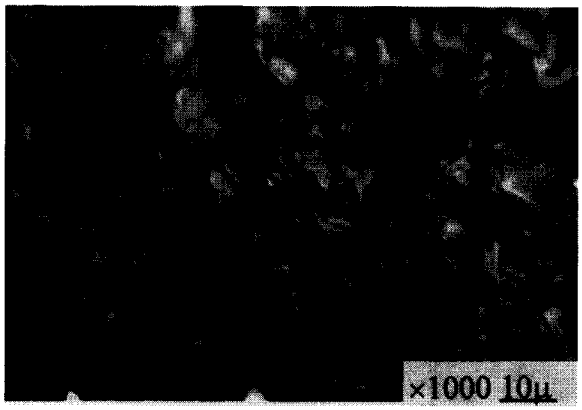


Fig. 2 Surface photography of SiC coating observed by an optical microscope

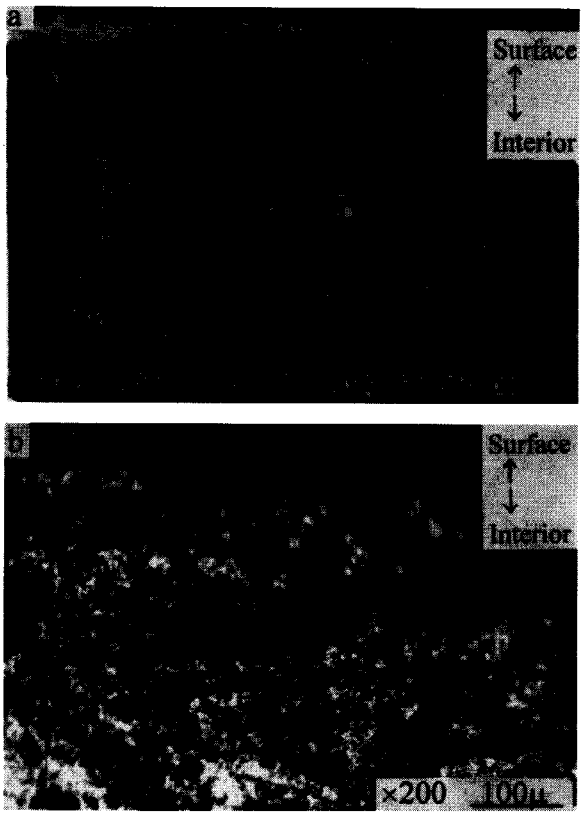


Fig. 3 Grading distribution of SiC coatings of the composites in the transversal section a-SEM image of the composite with the density of 1.94 g/cm$^3$; b-optical microscope image of the composite with the density of 2.05 g/cm$^3$

distribution of SiC coatings on graphite substrate, which were shown in Fig. 3. The grading distribution of SiC coatings possess the following characteristics: the surface layers of SiC with the thickness ranged from several μm to hundreds μm; the distribution of SiC particles along the distance from surface to the interior of substrates with the various penetration depths from several μm to the center of the composites, depending on the siliconization temperatures.

The distribution of SiC coating is closely related with the density of the composites. For the composites with the density of 1.88–1.95 g/cm$^3$, the thickness of the coatings is about several μm, and the distribution of SiC is limited to the depths of several tens μm; for the composites with the density of 1.96–2.10 g/cm$^3$, the thickness of the coatings is about several tens μm, and the distribution region of SiC is increasing to hundreds of μm; while for the composites with the density of 2.14–2.28 g/cm$^3$, the thickness of the coatings is up to hundreds μm, and the distribution region front of SiC reaches the center of the composites and almost all pores of substrates are filled with the SiC particles (see Fig. 4).

The porosity is calculated assuming that the true density of graphite and SiC is 2.2 g/cm$^3$ and 3.21 g/cm$^3$ respectively. A linear decrease for the porosity and a linear increase for the compressive strength with the density can be seen in Fig. 5. For the composites with the density of 2.28, the porosity is only 6.85 and the compressive strength is 166 Mpa, showing a decrease of 57% of porosity and an increase of 122% of strength in comparison with that of graphite substrates. The decrease of porosity is also an evidence that the SiC has formed in the interior of the substrates, which can be attributed the volume expansion accompanied with the reaction of Si and carbon.

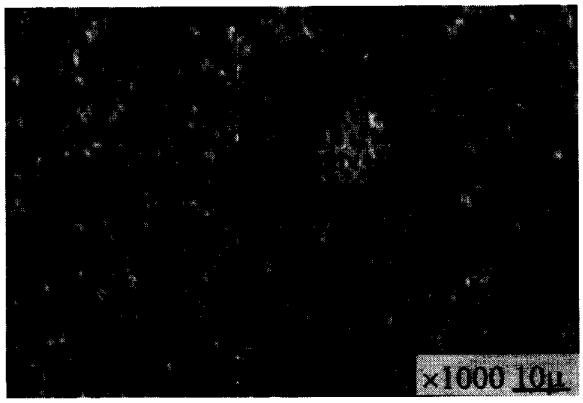


Fig. 4 The distribution of SiC particles in the interior of the composites with the density of 2.22 g/cm$^3$

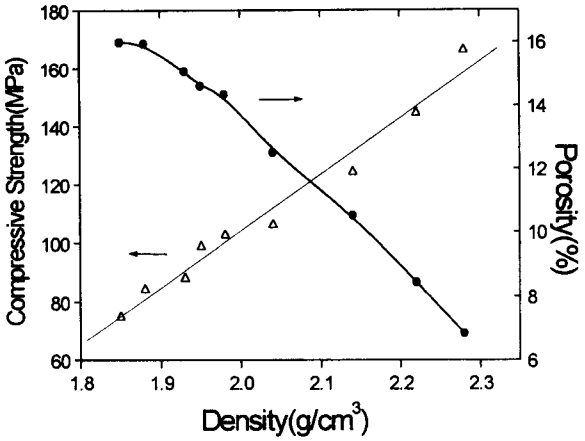


Fig. 5 The curves of compressive strength and porosity change with the density of the composites

### 3.3 Physical and mechanical properties of the composites Fig. 5 depicts the dependence of porosity and the compressive strength on the density of the composites.

The microstructure characteristics of the composites are preferable for their application as plasma-facing materials because of their high density and small porosity. It can be anticipated that the behavior of chemical sputtering and hydrogen isotope retention can be greatly improved because the whole surface including the surface of pores was covered by SiC particles, which exhibit thermal and chemical stability as low-Z materials under plasma environment. In addition, the formation of SiC in the composites dose not destroy the structure of graphite skeleton, high thermal conductivity and thermal shock resistance of

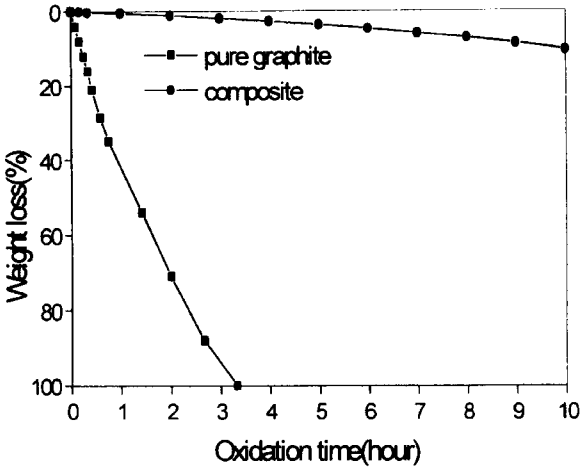


Fig. 6 Weight-loss of the composite and pure graphite as a function of oxidation time at 1200°C

graphite substrates would not deteriorate greatly, in comparison with the doped graphites [24]. On the contrary, some basic properties such as porosity and mechanical strength have been improved to some extent.

### 3.4 Oxidation resistance

Fig. 6 showed the weight-loss of the composite with the density of 2.15 g/cm$^3$ and pure graphite as a function of oxidation time at 1200°C. Pure graphite was burn out in about 3.3 hours, while the weigth-loss for the composites is only about 10% at the end of 10hours. It is obvious that the oxidation behavior of graphite substrate was greatly improved by the grading SiC coating.

The high performance of oxidation resistance of the composite was closely related with its microstructures:

- the good adherence of SiC coatings to graphite substrate because of the in-situ formation of SiC by the reaction of Si with carbon of the substrate;
- the grading distribution of SiC coatings from outer surface to the interior provides a grading expansion behavior that have relaxed the thermal mismatch strain to ensure the integrality of the coatings and the crack-free microstructure even after thermal shock;
- during the exposure of the composite to the oxidative atmosphere, a silica layer has formed and acts as a diffusion barrier against the oxygen transfer reaching the surface and the interior of substrate, thus rapid oxidation of graphite at high temperatures was highly restrained.

## 4. Conclusion

The composites prepared in this paper were

characterized by grading SiC coatings and graphite substrates. The thickness of SiC coatings and the distribution of SiC particles were dependent on the reaction temperatures at which the coatings were formed. The prepared composites are of high density, high strength and low porosity, and the excellent performance of oxidation resistance at temperatures up to 1200°C as well. Further research of investigations on the behavior of the composites under plasma environment need to de done to determine their applicability to PFMs.

## References

[1] A. Miyahara *et al.*, J. Nucl. Mater. **155-157**, 49 (1988).
[2] V. Barabash *et al.*, J. Nucl. Mater. **233-237**, 718 (1996).
[3] L.B. Begrambekov, J. Nucl. Mater. **233-237**, 754 (1996).
[4] J. Roth, J. Nucl. Mater. **266-269**, 51 (1999).
[5] B.V. Mech *et al.*, J. Nucl. Mater. **255**, 153 (1998).
[6] M. Wittmann *et al.*, J. Nucl. Mater. **227**, 186 (1996).
[7] J.W. Davis *et al.*, J. Nucl. Mater. **241-243**, 37 (1997).
[8] C. García-Rosales, J. Nucl. Mater. **211**, 202 (1994).
[9] J. Roth, J. Nucl. Mater. **145-147**, 87 (1987).
[10] V. Phillips *et al.*, J. Nucl. Mater. **179-181**, 25 (1991).
[11] Y. Ueda *et al.*, J. Nucl. Mater. **227**, 251 (1996).
[12] M. Balden, Physica Scripta. **81**, 64 (1999).
[13] J.W. Davis *et al.*, J. Nucl. Mater. **195**, 166 (1992).
[14] Reinhold Wolf, J. Nucl. Mater. **212-215**, 1174 (1994).
[15] R. Schwörer *et al.*, J. Nucl. Mater. **241-243**, 1156 (1997).
[16] M. Rubel *et al.*, J. Nucl. Mater. **258-263**, 787 (1998).
[17] J. Lin ke *et al.*, J. Nucl. Mater. **176-177**, 859 (1990).
[18] H. Bolt *et al.*, J. Nucl. Mater. **212-215**, 1239 (1994).
[19] P.G. Valentine *et al.*, J. Nucl. Mater. **212-215**, 1146 (1994).
[20] G. Savage, Carbon-carbon composites, Chapman & Hall, London, 1993.
[21] Q.G. Guo *et al.*, Carbon, **37**,147 (1999).
[22] Kimio Fujii *et al.*, J. Nucl. Mater. **203**,10 (1993).
[23] Junichi Nakano *et al.*, J. Nucl. Mater. **217**, 110 (1994).
[24] B.N. Enweani *et al.*, J. Nucl. Mater. **224**, 245 (1995).