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Effects of SiC content on microstructure and properties of carbon/ceramic conductive composites

Xiao Gao[a], Xin Zheng[a], Jie Gao[a], Juncheng Liu[a,*], Jiahai BAI[a] and Guoliang Yuan[b]

[a]*School of Material Science and Engineering, Shandong University of Technology, Zibo 255049, China*
[b]*Shandong Electric Shield Polytron Technologies Inc, Zibo 255000, China*

The effects of SiC content on the phase composition, microstructure, sintering performance, mechanical and electrical properties of the carbon/ceramic conductive composites were investigated. The results show that when SiC content increases from 1 wt% to 3 wt%, lamellar graphite structure can be seen in the microstructure of the composites, and the porosity and water absorption increases gradually. With further increase of SiC content to 6 wt% and 9 wt%, lamellar graphite disappears, and granular SiC crystals fill in the ceramic phase, thus increasing the densification degree of the composites. When SiC content is in the range of 1 wt% to 3 wt%, the shrinkage rate decreases sharply while the weight loss decreases a little. As SiC content increases from 3 wt% to 9 wt%, both the shrinkage rate and weight loss basically keep unchanged. With the increase of SiC content, the bending strength of the composites first decreases and then increases, and finally reaches the peak value at 9 wt% SiC. However, the resistivity presents an opposite variation trend, and the conductivity is best as SiC content is 9 wt%.

Key words: Carbon/ceramic conductive composites, SiC content, Microstructure, properties.

Introduction

Carbon/ceramic conductive composites have shown various applications as the typical functional materials in home appliances, building heating, health care [1] and other industries. At present, the research on these materials mainly focuses on the effects of the content of conductive filler on their conductivities [2-13]. Duan and Xie prepared carbon/ceramic conductive composites by adding graphite into composites, and they found that graphite could hinder the densification and strengthening of the composites. Qiao [6-7] studied the influences of Cr, TiC, SiC, and B_4C on the conductive properties of carbon/ceramic conductive composites. It was discovered that Cr doping was favorable to the conductivity of composites, whereas B_4C doping led to the significant increase of thermal shock resistance. Li prepared carbon/ceramic composites by respectively adding carbon black, graphite and their mixture into the kaolin powder. The results showed that the composites turned to be a good conductor when the content of conductive filler was more than 25% [8]. However, the effects of combination conditions between flake graphite and ceramic matrix on the microstructure and conductive properties of carbon/ceramic conductive composites have rarely been studied from the published literatures.

For carbon/ceramic conductive composites filled with graphite conductors, large pores can be easily found between graphite and ceramic matrix. As a result, the composites often present increased porosity and reduced bending strength. In this study, the effects of SiC content on the material microstructure, sintering performance, mechanical and electrical properties of carbon/ceramic composites were investigated. It was believed that our work could provide valuable references for the preparation of carbon/ceramic conductive composites.

*Corresponding author:
Tel : +86-533-2786998
Fax: +86-533-2786998
E-mail: jchliu@sdut.edu.cn

Experimental

Preparation of carbon/ceramic conductive composites

Carbon/ceramic conductive composites were prepared from albite, diopside, quartz, SiC and graphite. Four samples were prepared and their nominal compositions are shown in Table 1. The mixtures were milled, dried, granulated, and then dry-pressed and sintered at a low temperature. The forming pressure was 20 MPa and the sintering temperature was 1120 °C.

Characterization and property test of carbon/ceramic conductive composites

The grain phase composition was identified using an

Table 1. Compositions of various samples (wt/%).

Sample number	Ceramic matrix	Graphite	SiC	Balance
a	88	10	1	1
b	86	10	3	1
c	83	10	6	1
d	80	10	9	1

X-ray diffractometer (XRD, D8 Advance, Brucker, Germany), equipped with a Ni-filtered Cu Kα radiation source (λ = 0.154178 nm). The microstructures of the composites were characterized by a field emission scanning electron microscope (FESEM, Sirion 2000, FEI, Netherlands). Water absorption and porosity of the composites were determined by the Archimedes method using distilled water as liquid medium. Weight loss and linear firing shrinkage were simply calculated by measuring the length of specimens before and after sintering. Bending strength was tested via a three-point bending test (ATOGRAPH AG-I, Shimadzu) with a support distance of 30.0 mm and a cross-head speed of 0.50 mm/min. The electrical resistivity was measured using the four probe method.

Results and Discussion

Phase analysis

Fig. 1 shows the effect of SiC content on the phase composition of carbon/ceramic conductive composites. As can be seen, the diffraction patterns of the samples with 1 wt% and 3 wt% SiC are basically the same. Both of them display diffraction peaks of quartz, graphite, albite and diopside. In contrast, the samples with 6 wt% and 9 wt% SiC both display another two diffraction peaks at 34.8 º and 38.2 º, which are the characteristic peaks of SiC. In addition to the characteristic peaks of SiC, other diffraction peaks in XRD patterns of the four samples all correspond to quartz, graphite, albite or diopside. Hence, it can be concluded that the samples with 6 wt% and 9 wt% SiC have no other new phases besides SiC.

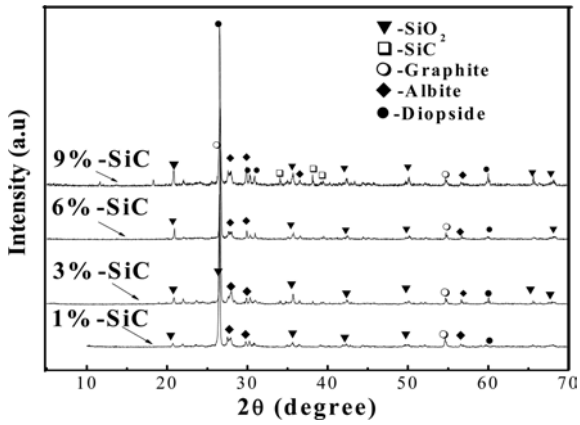

Fig. 1. Effect of SiC content on the phase composition of carbon/ceramic conductive composites.

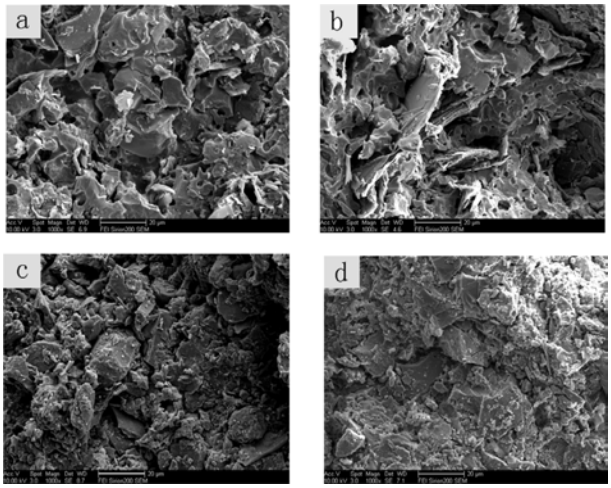

Fig. 2. Effect of SiC content on the microstructures of carbon/ceramic conductive composites: (a) 1 wt%, (b) 3 wt%, (c) 6 wt%, (d) 9 wt%.

Microstructure analysis

Fig. 2 shows the microstructures of carbon/ceramic conductive composites with different SiC contents. When the content of SiC is 1 wt% (Fig. 2(a)), flake graphite and smooth ceramic phase are combined closely, and flat pores distribute evenly in the ceramic matrix. With the increase of SiC content to 3 wt%, as Fig. 2(b) shows, the number of pores increases and the interstices between ceramic phases become larger. It can be clearly observed that flake graphite is inserted in the ceramic matrix. When the content of SiC increases to 6 wt% and 9 wt% (Figs. 2(c) and 2(d)), previous pores disappear, and flake graphite cannot be observed in the composites. In this case, a lot of granular crystals aggregate at the boundaries of ceramic phases, and the surface layer is covered with milky white material. This milky white material, as can be known from Fig. 1, is SiC.

The effect of SiC content on the microstructures of carbon/ceramic conductive composites can be explained as follows. When a small amount of SiC is added into the composites, since flake graphite cannot react with oxygen at this temperature, SiC powders rather than flake graphite would react with oxygen [14]. As the reaction product, SiO_2 can then change into smooth ceramic phases, as shown in Figs. 2(a) and 2(b). With the increase of SiC content to 6 wt%, there is no enough oxygen in the composite body to oxidize SiC powders during sintering so that more SiC grains remain in the sintered bodies. Consequently, more SiC grains come into being in the fracture surfaces of the composites, as shown in Fig. 2(c). When larger content of SiC is added into the composites, SiC covers the surface of the ceramic matrix and forms a layer of white material, as shown in Fig. 2(d). Therefore, it can be induced that SiC grains are mainly located at the boundaries of matrix grains.

Effect of SiC content on the porosity and water absorption of carbon/ceramic conductive composites

Fig. 3 shows the effect of SiC content on the porosity and water absorption of carbon/ceramic conductive composites. With the increase of SiC content, the porosity of the composites first increases and then decreases. To

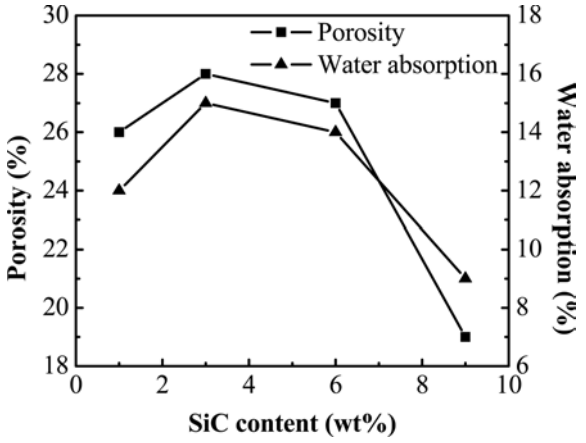

Fig. 3. Effect of SiC content on the porosity and water absorption of carbon/ceramic conductive composites.

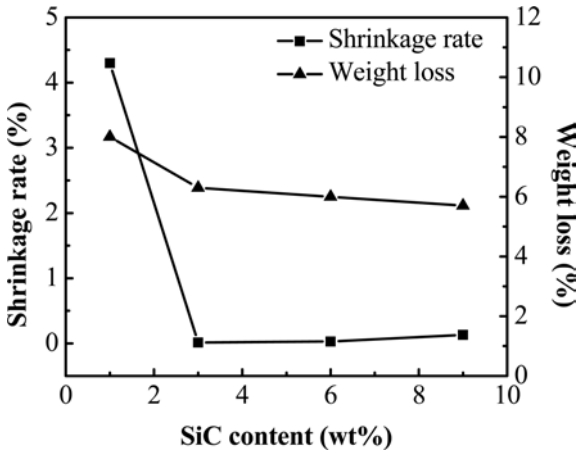

Fig. 4. Effect of SiC content on the shrinkage rate and weight loss of carbon/ceramic conductive composites.

be specific, the porosity of the composites increases when the content of SiC increases from 1 wt% to 3 wt%, and then begins to decrease with further increase of SiC content. This effect of SiC content on the porosity can be attributed to the following chemical reaction:

$$2SiC + 3O_2 \rightarrow 2SiO_2 + 2CO \quad (1)$$

When the content of SiC increases from 1 wt% to 3 wt%, more and more silica is generated. Because of the expansion of silica at high temperatures [15], the porosity of the composites becomes larger. When the content of SiC increases to 3 wt%, oxygen in the composite body is consumed completely and the pores between ceramic particles are filled with SiC particles. With further increase of SiC content from 3 wt%, the porosity decreases to a level lower than that of the composites with 1 wt% SiC [16].

As known from Fig. 3, the water absorption of carbon/ceramic conductive composites is positively correlated with the porosity.

Effect of SiC content on the sintering performance of carbon/ceramic conductive composites

Fig. 4 shows the effect of SiC content on the sintering performance, including linear firing shrinkage and weight loss, of carbon/ceramic conductive composites. When SiC content increases from 1 wt% to 3 wt%, the shrinkage rate of the composites sharply decreases to the lowest point at about 3 wt%. This is mainly due to the generation of SiO_2 from SiC oxidation. The as-generated silica expands during the sintering process, but the expansion can be partly compensated by the contraction of other ceramics phases. Thus, remarkable change of the firing shrinkage occurs when SiC content rises from 1 wt% to 3 wt%. With further increase of SiC content, no more SiC grains can be oxidized by the oxygen in the green body so that no more SiO_2 would be generated. Therefore, there is no appreciable decrease in the firing shrinkage of carbon/ceramic conductive composites, in spite of further addition of SiC. However, weight loss of the composites reduces gradually when SiC content increases from 1 wt% to 3 wt%, and then it basically keeps unchanged with further increase of SiC content. The main reason may be as follows:

When SiC content is in the range of 1 wt% to 3 wt%, chemical reactions shown in equations (1-3) may occur.

$$2C + O_2 \rightarrow 2CO \quad (2)$$

$$C + H_2O \rightarrow CO + H_2 \quad (3)$$

Based on the three reactions, it can be seen that the quality of the green body is degraded due to the dehydration of the green body and the oxidation of graphite and SiC. At the same time, more oxygen is consumed by SiC and less graphite is oxidized with the increase of SiC content, so that weight loss of the composites becomes less. As SiC content reaches 3 wt%, oxygen in the ceramic matrix is nearly completely consumed, so that the weight loss shows no remarkable change in spite of further increase of SiC content.

Effect of SiC content on the mechanical and electrical properties of carbon/ceramic conductive composites

Fig. 5 shows the effect of SiC content on the mechanical and electrical properties, including bending strength and resistivity, of carbon/ceramic conductive composites. As can be seen, the bending strength of the composites first decreases and then increases while the resistivity shows an opposite trend, namely, it first increases and then decreases. When SiC content is less than 3 wt%, SiC reacts with oxygen to generate silica. Because of the expansion of silica at high temperatures, the porosity of the composites becomes larger so that the effective cross-sectional area of the composites is reduced. As a result, stress concentration occurs and the intensity is reduced. At the same time, silica also has a function of reducing the shrinking percentage of

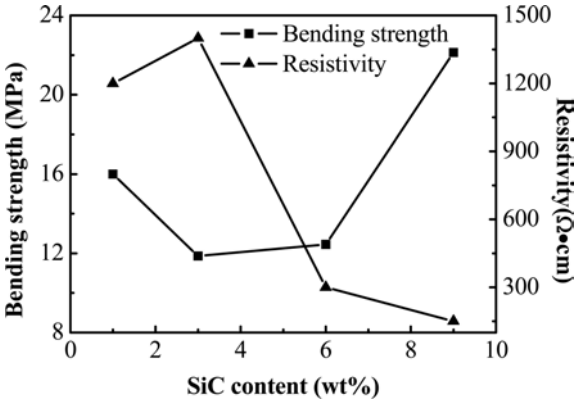

Fig. 5. Effect of SiC content on the bending strength and resistivity of carbon/ceramic conductive composites.

carbon/ceramic conductive composites, which leads to the increase of the spacing of graphite and thus partial disconnection of the conductive network [17]. Obviously, this effect can increase the resistivity of the composites.

When the content of SiC increases to 3 wt%, oxygen in the green body has been consumed completely. With further increase of SiC content, the pores between ceramic particles are filled with SiC particles, which may reduce the porosity of the composites. It is noticed that the bending strength of composites is closely related to the porosity [13]:

$$\sigma = \sigma_0 e^{-k\alpha} \quad (4)$$

where σ is the bending strength of a material with certain porosity, σ_0 is the bending strength of the material without the porosity, α is the porosity, and κ is a constant. Eq. (4) indicates that there is an exponential relation between the bending strength of the composites and the opposite number of the porosity. That is, the porosity can directly affect the bending strength of the composites. The larger the porosity is, the lower the bending strength is.

When the content of SiC is higher than 3 wt%, the pores between ceramic particles are filled with the SiC particles that cannot react with oxygen and the conductive network is connected again because SiC has semi-conducting properties. Hence, the resistivity of the composites becomes lower. With further increase of SiC content, the texture of the composites becomes more compact, and the number of conductive network is increased accordingly. As a result, the resistivity decreases again and finally reaches the lowest level when the content of SiC is 9 wt%.

Conclusions

In summary, carbon/ceramic conductive composites were successfully prepared, and the effects of SiC content on the microstructure and properties of the composites were investigated. Some conclusions can be drawn as follows:

Firstly, when SiC content increases from 1 wt% to 3 wt%, lamellar graphite structure can be seen in the microstructure of the composites, and the porosity and water absorption increases gradually. With further increase of SiC content to 6 wt% and 9 wt%, lamellar graphite disappears, and granular SiC crystals fill in the ceramic phase, thus increasing the densification degree of the composites.

Secondly, when SiC content is in the range of 1 wt% to 3 wt%, the shrinkage rate decreases sharply while the weight loss decreases a little. As SiC content increases from 3 wt% to 9 wt%, both the shrinkage rate and weight loss basically keep unchanged.

Thirdly, with the increase of SiC content, the bending strength of the composites first decreases and then increases, and finally reaches the peak value at 9 wt% SiC. At the same time, the resistivity presents an opposite variation trend, and the conductivity is best when SiC content is 9 wt%.

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