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Oxidation resistance and wettability of graphite/SiC composite

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Abstract: The core-shell structure of graphite@SiC was synthesized at different calcination temperatures using the raw materials of microsilica and carboxy methylated cellulose (CMC). The oxidation resistance and wettability (with water) of graphite@SiC were investigated. The results showed that CMC could react with microsilica to form a SiC shell on the graphite surface at elevated temperature. Consequently, the SiO_2 phase was totally converted into the SiC phase at above 1600°C. The microstructure of SiC coating on the graphite became denser with increasing temperature. Thermo-gravimetric curves revealed that the weight loss percent of graphite was about 97.3% while the value decreases to 29.78% when the SiC was formed. Differential scanning calorimetry analysis showed that the existence of SiC coating decreased the enthalpy of carbon oxidation reaction from 12.02 kJ/g to 1.14 kJ/g, confirming excellent oxidation resistance. At the same time, the water contact angle of graphite was about 78.5° while the core-shell structure of graphite@SiC reduced the wetting angle to 43°. The study of the formation mechanism of graphite@SiC showed that SiO_2 could be reduced by CMC to SiO(g), which would be deposited on the graphite to form SiC coating.

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Keywords: Surface modification ; SiC coating; Oxidation resistance; Wettability

1 Introduction

Owing to low thermal expansion coefficient, high thermal conductivity and non-wetting characteristics with the molten steel or slag [1-5], flaky graphite gives rise to the carbon containing composites with excellent thermal shock resistance, high temperature strength and excellent corrosion resistance. All these features lead to carbon-containing refractory composites possessing wide application in the steelmaking industries of the blast furnace, converter, ladle and Ruhrstahl Heraeus degassing processing (RH) [6]. However, the non-wettability with water of flaky graphite has greatly limited its wide application in the refractory castable because the surface with inactive flaky graphite increases the water content and leads to high porosity, which could ultimately deteriorate the strength and corrosion resistance to molten slag or steel. Additionally, the unmodified natural graphite can be easily oxidized in air atmosphere leading to significant degradation of physico-chemical properties [7]. Thus, the surface modification of flaky graphite has been the focus of researchers across the world.

Aimed at solving this problem, the researchers have been exploring the coating technology on the surface of the flaky graphite to improve its oxidation resistance and wettability. Most of the coating layers on the graphite were composed of ceramic phases that are either oxide or non-oxide [8-11]. For example, Ansar et al. [12] used $MgAl_2O_4$ spinel and mullite ($3Al_2O_3 \cdot 2SiO_2$) to form oxide coating on the graphite surface. Compared with uncoated flaky graphite, the oxidation weight losses of spinel and mullite coated graphite at 1200°C were obviously decreased to 22% and 29.7%, respectively. Meanwhile, the water wettability of coated graphite was improved by 8.0% and 5.0%, respectively, compared to uncoated graphite. In the same way, Yilmaz et al. [13] prepared boehmite (Al(OH)3 or Al2O3) sol-coated graphite. In terms of oxidation resistance, the weight loss of graphite coated with boehmite at 1100°C was as low as about 12%. Mukhopadhyay et al. [14-15] synthesized calcium aluminate ($CaAl_2O_4$) coating on the graphite surface and obviously improve both the oxidation resistance and wettability of flaky graphite. The weight loss of graphite at 1000°C reduced by 10 times and the value was decreased from over 90% to 9.07%. the water content in refractory castables with coated graphite was decreased from 10.8% to 7.5%, which was based on unconventional "ball-in-hand" test[15]. Sharif et al. [16] utilized gas phase reaction to prepare the coating with MgO/carbon nanocomposite on the surface of flaky graphite. Owing to MgO coating as a barrier, flaky graphite was protected from oxygen attack and the reaction of oxygen and carbon was inhibited. The initial oxidation temperature was increased from 700°C to 800°C, yet the weight loss of MgO coated graphite at 1300°C was still about 80%.

Compared with oxide coating, non-oxide SiC with excellent mechanical, chemical and water wetting properties behave well with suitable coating phases in high temperature industry [17-19]. Yun et al. [20] prepared SiC-coated graphite by chemical vapor reaction at above 1700°C, and the lowest weight loss of the coated graphite at 700°C was achieved at approximately 2 wt%. Yang et al. [21] synthesized SiC coating with excellent self-healing ability at temperatures from 1623 to 1823 K. The coating protected flaky graphite from ablation for 50s by an oxyacetylene torch test. Jafari et al. [22] obtained a dual-layer coating structure with nano-SiC/SiC on the surface of flaky graphite with cementation and electrophoretic deposition. In some of these composite structure, the dual-layer plays an important role on enhancing the oxidation resistance.

In addition to the above methods, molten salt technology is also considered as an alternative for SiC coating formation. Ding and Deng et al. [23] reported that flaky graphite with SiC coating had a weight loss of about 10% at 1200°C in an oxidizing atmosphere. When oxidant temperature rose to 1350°C, the weight loss of coated graphite was increased to about 15%[24]. Similarly, Masoudifar et al. [25] utilized the same method to obtain a SiC coating on the surface of flaky graphite with a mass loss of about 25% at 1200°C. Ye et al. [26] used raw materials of carbon black and Si to prepare SiC coating based on the molten salt method. Since the wettability of SiC was better with water, the dispersivity and flowability of flaky graphite-contained refractory castables were improved greatly in the castable system containing the carbon resource.

In this work, a novel synthesis using a simple and low cost process is introduced to modify the surface of flaky graphite forming graphite/SiC composite with core-shell structure. This way, SiC coating layer is formed in-situ on the surface of flaky graphite by reaction with carboxy methylated cellulose (CMC) and microsilica. Based on XRD and SEM analysis, phase evolution and microstructure are evaluated. The oxidation resistance is investigated by thermal analysis with TG and DSC. In addition, aiming to explore the wettability of the coated samples, the contact angle between solid and liquid are measured. Through thermodynamic calculation and predominance area diagrams, this study explored the in-situ growth mechanism of SiC.

2 Experimental

2.1 Materials and procedure

Flaky graphite (purity≥97.5 wt %, particle size≤88μm) and microsilica (purity≥95 wt %, particle size≤10μm) were used as raw materials. The CMC (purity≥97 wt %) was first dissolved in distilled water to obtain a 100g/L CMC solution. subsequently, flaky graphite and microsilica were added in sequence into the CMC solution to yield the precursor solution. The mole ratio of CMC, flaky graphite and microsilica was 1:1:5. The precursor solution was dried in an oven at 110°C for 24 h and then pulverized. Finally, the prepared precursor was heated at 1500°C, 1600°C and 1650°C, respectively, for 3 h in a coke-packed bed (where the atmosphere contains 35% CO and 65% N_2).

2.2 Sample characterizations

The phase evolution of specimens was determined by X-ray diffraction (XRD, Philips, X'Pert Pro). Microstructure characterization was performed by scanning electron microscope (SEM, FEI, Nova400Nano) equipped with energy dispersive spectroscopy (EDS). Meanwhile, transmission electron microscopy (TEM, JEM-2100UHR) was utilized to further explore the structure parameters. morphological information. To evaluate the oxidation resistance of the graphite/SiC core-shell composites, DSC and TG analysis were performed at a temperature of up to 1200°C with a heating rate of 10°C/min in air by a simultaneous thermal analyzer (NETZSCH, STA449C). The wettability of the specimens with water was investigated by a contact angle meter (JC2000A, China).

3 Results and discussion

3.1 Phase patterns of the synthesis of graphite/SiC core-shell composites

The XRD pattern of the specimens with different calcination temperatures is depicted in Fig.1. It can be seen that graphite is the main phase at different calcination temperatures. The relative intensities of β-SiC peaks and the SiO_2 peaks present opposite trend with the increasing calcination temperature. At 1500°C, the intensity of the diffraction peaks of SiO_2 is far greater than that of SiC, which is only detected at $2\theta=35.7°$. With increasing temperature to 1600°C, more intense characteristic peaks of β-SiC appear at $2\theta= 35.7°$,41.4°, 60.0°, and 71.8°, which refer to diffraction of cubic β-SiC (111), (200), (220) and (311) planes, respectively. At 1650°C, the peaks of SiO_2 completely disappear while only graphite and SiC phases appear to co-exist in the samples.

XRD results suggest that the calcination temperature is a key factor for the in-situ synthesis of graphite/SiC composite. When the temperature is lower than 1500 °C, only trace of SiC is formed. As the temperature increases to 1650°C, the SiO_2 at the surface of graphite has been completely carbonized into SiC.

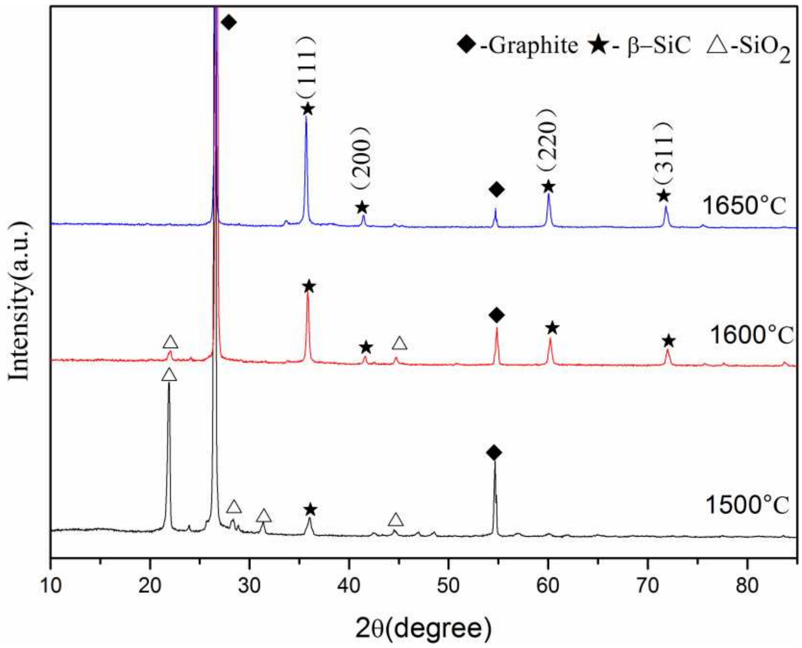


Fig.1 XRD patterns of specimens at different temperatures

3.2 Microstructure characterization of graphite/SiC core-shell composites

Fig. 2 depicts SEM images of specimens heat-treated at different calcination temperatures. It shows that the surfaces of raw flaky graphite are smooth (Fig. 2a) compared with the rough surfaces and edges of coated graphite after calcination at 1500°C (Fig. 2b). From the XRD results (Fig. 1), it is illustrated that the main phase at the surface of graphite are SiO_2 and a trace amount of SiC. When the temperature increases to 1600°C, the surface of graphite exhibits a significant change, which appears to be coated with a twisted whisker-like layer, as shown in Fig. 2c. The EDS analysis confirms that the whisker-like substance is SiC in which the weight percent of Si is 31.11%, as shown in Fig. 2d. Although it shows that several SiC whiskers are formed on the surface of graphite, the SiC layer is not completely coated on the graphite surface. Raising the calcination temperature to 1650°C, a dense coating layer has been formed on the surface of the flaky graphite, as depicted in Fig.2e. In contrast with Fig. 2c, the coating becomes denser and the free carbon content decreases. In addition, the EDS X-ray map of the specimens fired at 1650°C are shown in Fig. 2(f and g). The red color represents the distribution of carbon and the blue color denotes distribution of Si. Seen from both photographs, the carbon and Si elements are uniformly distributed on the surface of graphite. Combined with XRD analysis, it is confirmed that SiC mostly covers the graphite surface. From the above results, it is clear that the SiC coating grows gradually and then forms a dense coating on the surface of graphite with increasing temperature.

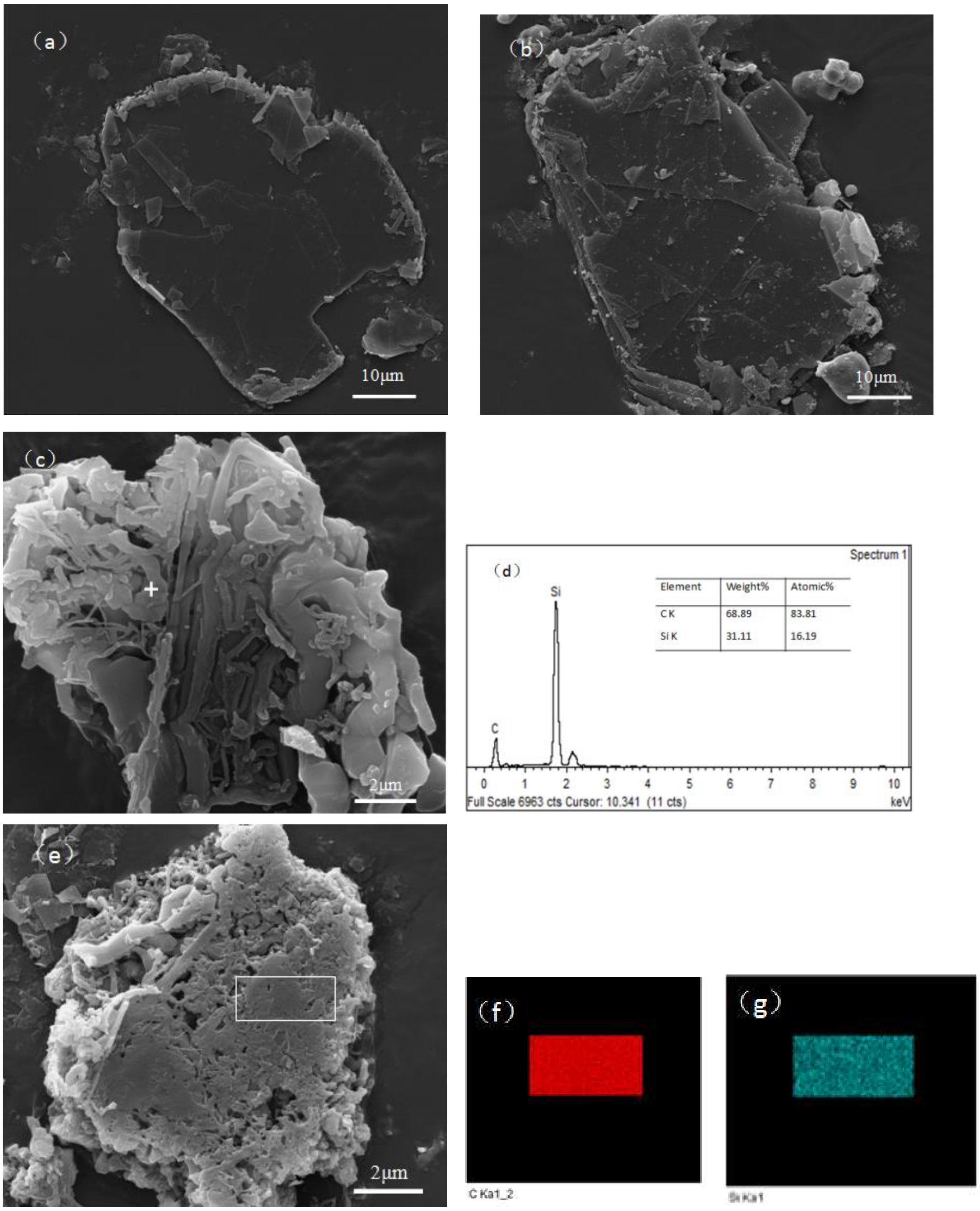


Fig. 2 SEM images of specimens heat-treated at different temperatures:

(a) raw graphite; (b) 1500°C; (c) and (d): SEM and EDS at 1600°C; (e), (f) and (g): SEM and EDS elemental map at 1650°C

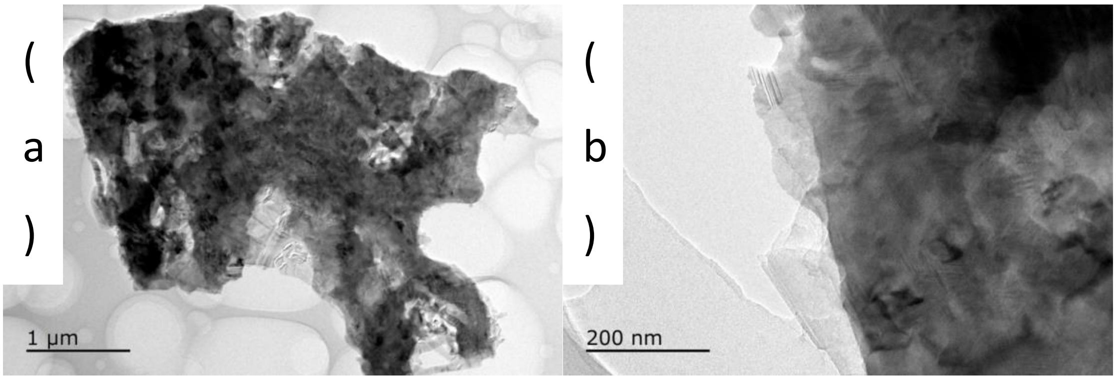
Morphological and structural analysis of the nano-structures of graphite/SiC is performed by TEM and is shown in Fig3. It is revealed from Fig 3a and 3b that the typical core-shell images of graphite/SiC is formed. Fig.3c depicts the corresponding lattice fringe image of coating surface. The spacing between adjacent lattice planes is 0.25nm. From the X-ray powder diffraction file No. 75-0254 for SiC, this interplanar spacing corresponds to the (111) plane of β-SiC.

The selected area electron diffraction (SAED) for the surface of specimens is shown in Fig.3d. It is found that the substance has a cubic structure crystal. In the SAED, each diffraction spot corresponded to certain crystal face. The interplanar spacing of the two selected crystal faces is measured respectively, which is 0.2268nm for one crystal face (R1) and 0.2472nm for the neighboring face (R2). At the same time, the angle of the two selected crystal faces is measured to be 53.48°. The interplanar spacing for faces of (200) and (111) are respectively 0.2179nm and 0.2516nm, which are quitely similar to the measured R1and R2 in Fig.3d. The angle between two crystal faces of cubic crystal can be also calculated from Eq.(1):

$$\cos\theta = \frac{h_1h_2+k_1k_2+l_1l_2}{\sqrt{(h_1^2+k_1^2+l_1^2)(h_2^2+k_2^2+l_2^2)}} \qquad (1)$$

where h, k, l defined the lattice planes.

Based on Eq. (1), the angle between the crystal faces (111) and (200) of β-SiC is calculated as 54.73°. It is worth noting that the value is close to the measured value in the Fig.3d.



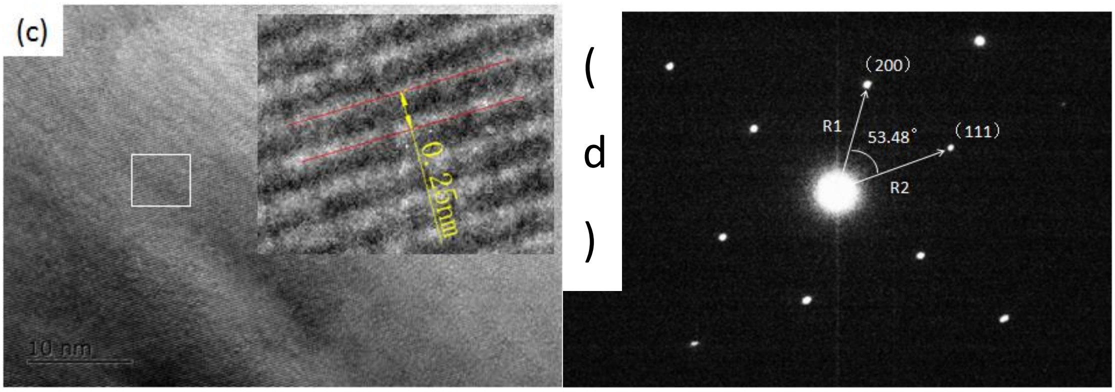


Fig.3 TEM images of specimens heat-treated at 1650℃

3.3 Oxidation resistance of graphite/SiC core-shell composites

The oxidation resistance behavior of graphite/SiC core-shell composites synthesized at different temperatures is evaluated by TG/DSC analysis. Fig. 4 shows TG curves of specimens heat-treated at different temperatures. According to the TG curves, initial oxidation temperatures of flaky graphite uncoated and coated are all in the range of 650−800°C, but the weight losses of the specimens are different. When the raw flaky graphite is heated from room temperature to 1200°C in air atmosphere, the weight loss is 97.30%, indicating that almost all the graphite has been oxidized. In contrast, the mass loss of specimens with coating decreases significantly with increasing heat treatment temperature. At 1500°C, the mass loss of graphite with SiC coating reduces to 44.17%. When the heat treatment temperature is 1600°C, the weight loss is further reduced to 40%. Similarly, up on heat treatment 1650°C, the weight loss of graphite/SiC core-shell composite reach its lowest value of 29.78%.

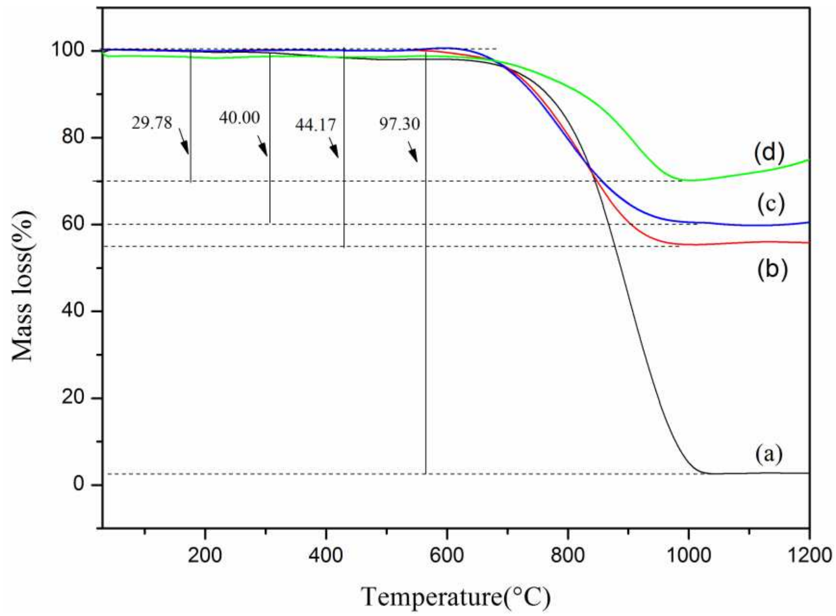


Fig.4 TG curves of specimens heat-treated at different temperature

(a) raw graphite; (b)1500°C; (c)1600°C and (d)1650°C

Considering the SEM micrographs and XRD analysis, the reduction of mass loss suggests that whether it is SiO_2 or SiC coating, the core-shell structure of specimen performs the role of an oxidation barrier to protect the graphite from oxygen attack, and hence improves the oxidation resistance of graphite.

The DSC curves of the graphite and graphite/SiC core-shell composites are shown in Fig. 5. It is found that the exothermic peak temperature of different specimens exists between 600°C and 1000°C.. The exothermic peaks of the samples with coating (specimens b~d) are flatter and wider than that of pure flaky graphite without any coating (specimen a), suggesting a retardation of oxidation kinetics. The detailed parameters of oxidation kinetics could be calculated from the DSC curves in Fig.5 and depicted in table 1. The onset temperature of specimens b~d with increasing heating temperatures is increased from 654.4°C to 800.8°C. At the same time, the final and peak temperature have the same trend as the onset temperature for specimens b~d.

The former value is increased from 823.1°C to 900.11°C and the latter is increased from 996.6°C to 1009.5°C, respectively. When the coating phases are transformed from SiO2 to SiC, the exothermic temperature, either the onset, peaking or final, become higher, which means that the SiO2 or SiC coating could play a key role in retarding the kinetics of graphite oxidation and decrease their weight loss(shown in Fig.4) correspondingly. The formation of SiC coating(in Figs.2 and 3) on the surface of flaky graphite exhibits the highest oxidation temperature and the minimum weight loss.

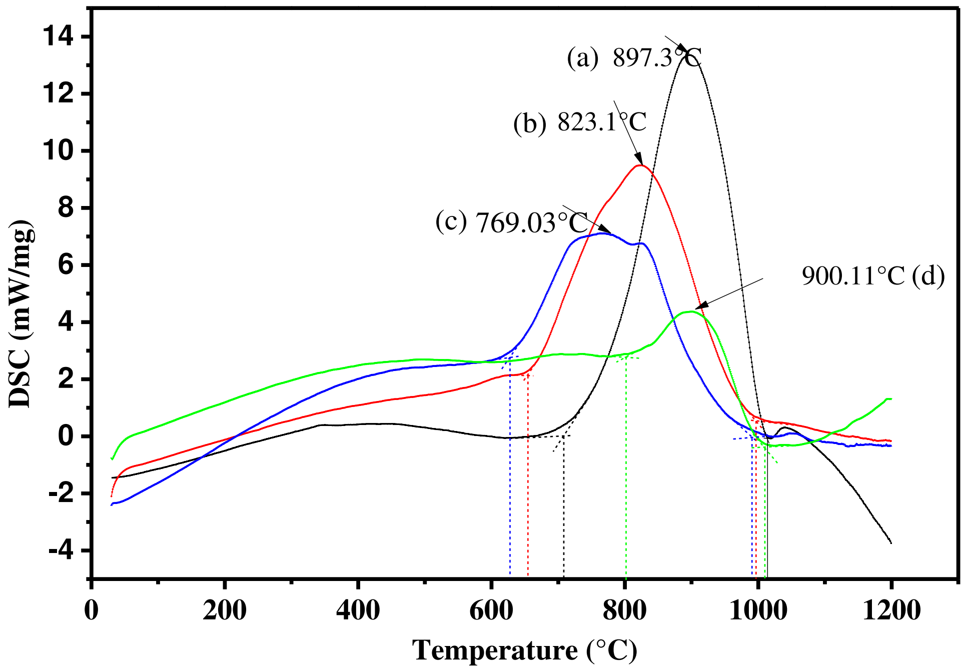


Fig.5 DSC curves of specimens heat treated at different temperatures

Table 1 The onset, peak and final temperatures of oxidant reaction kinetics of graphite/SiC core-shell structures

Specimens	Temperature (°C)		
	Onset	Final	Peak
a	708.1	1013.7	897.3

b	654.6	996.6	823.1
c	626.8	990.5	769.03
d	800.8	1009.5	900.11

Based on DSC curve in Fig.5, the peak area and enthalpy of graphite oxidation reaction could be calculated in table 2. It is interesting that the graphite with coating heat treated at three temperatures behaves lower exothermic peak intensity, compared with the pure flaky graphite, which also proves that the coating on the surface of flaky graphite retards the reaction of carbon and oxygen. The peak area presents a decreasing tendency and the enthalpy of graphite oxidation reaction decreases from 12.02kJ/g to 1.14 kJ/g, which means an excellent oxidation resistance of the graphite with coating. It is believed that SiC coating on flaky graphite is responsible for providing the oxidation resistance to the graphite/SiC core shell structure. The research results is agreement with Ref[22].

Table 2 The peak area and enthalpy of C-O oxidation reaction based on DSC curves

	a	b	c	d
peak area(J)	2003.25	1451.50	1150.51	189.69
Enthalpy/(kJ/g)	12.02	8.71	6.90	1.14

At the same time, it is interesting that the graphite samples with coating calcined at three temperatures behave well in the lower exothermic thermal peak compared with the pure graphite. It means that the coating in the surface of graphite postpones the reaction of carbon and oxygen. The detailed exothermic peak area and enthalpy of C-O oxidation reaction are calculated by the Origin software. The results are shown in Table 1. It shows that the peak area presents a decreasing tendency and the enthalpy of C-O oxidation reaction decreases from 12.02

kJ/g to 1.14 kJ/g, which depicts an excellent oxidation resistance of the specimens with coating. When the oxidation temperature reaches 1000°C in air atmosphere, the SiC coating on the surface of graphite can form a SiO_2 thin film to separate the oxygen attack [22]. This is a good explanation for the SiC coating with better oxidation resistance.

3.4 Wettability of graphite/SiC core shell structure

To evaluate the effect of SiC coating on the wettability of graphite, the contact angle between a drop of water and surface of the flaky graphite is measured in Fig. 6. It shows that the contact angles decrease with increasing heat treatment temperature. The contact angle of raw flaky graphite is 78.5°. When the heated treatment temperatures increase from 1500°C to 1650°C, the contact angle of the graphite with coating decrease significantly to 54.8°, 48° and 43°, respectively. The reduction of contact angle indicates that the coated surface has a hydrophilic nature so that water could wet the surfaces easily.

In particular, the water wettability of the three specimens with coating heat treatment at different temperatures has been improved. Along with increasing temperature over 1600°C, the SiC coating could be formed completely covering the surface of the flaky graphite. This gives rise to a polar surface of flaky graphite, thereby improving the hydrophilicity of the graphites[27,28]. The result is consistent with the report of Souza et al. [29] that the SiC coating can effectively improve the wettability of graphite.

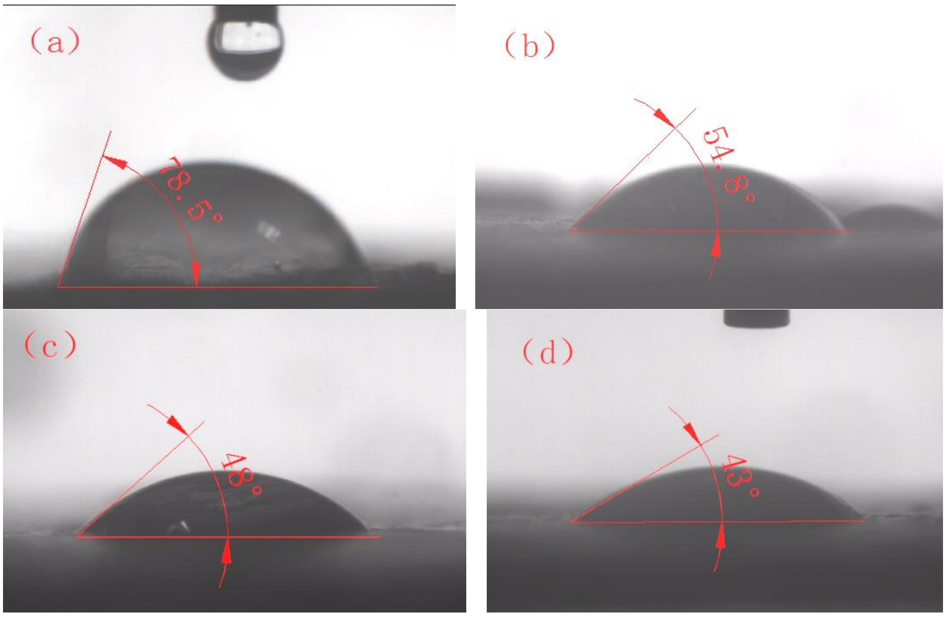


Fig.6 the contact angles of specimens with different temperature:

(a) raw graphite; (b)1500°C; (c)1600°C and (d)1650°C

3.5 In-situ growth mechanism of graphite/SiC core shell structure

The predominance diagrams of the Si–C–O systems at 1500°C, 1600°C and 1650°C in a coke-packed bed have been calculated using the FactSage software and the results are shown in Fig. 7. It depicts the predominance area diagram of Si-C-O system at 1500°C (Fig.7a) and SiC phase is stable when the value of $\ln(P_{CO}/P^0)$ is lower than 0.401, which is greater than that found in a coke-packed bed where $\ln(P_{CO}/P^0) < -1.04$. The predominance area diagrams of 1600°C and 1650°C are shown in Fig. 7b and c, and the values of the $\ln(P_{CO}/P^0)$ are 1.512 and 2.024, respectively. The diagrams show that the stability region of SiC and Carbon co-existing becomes wider; in other words, the SiC coating on the surface of flaky graphite is likely more stable at higher temperature.

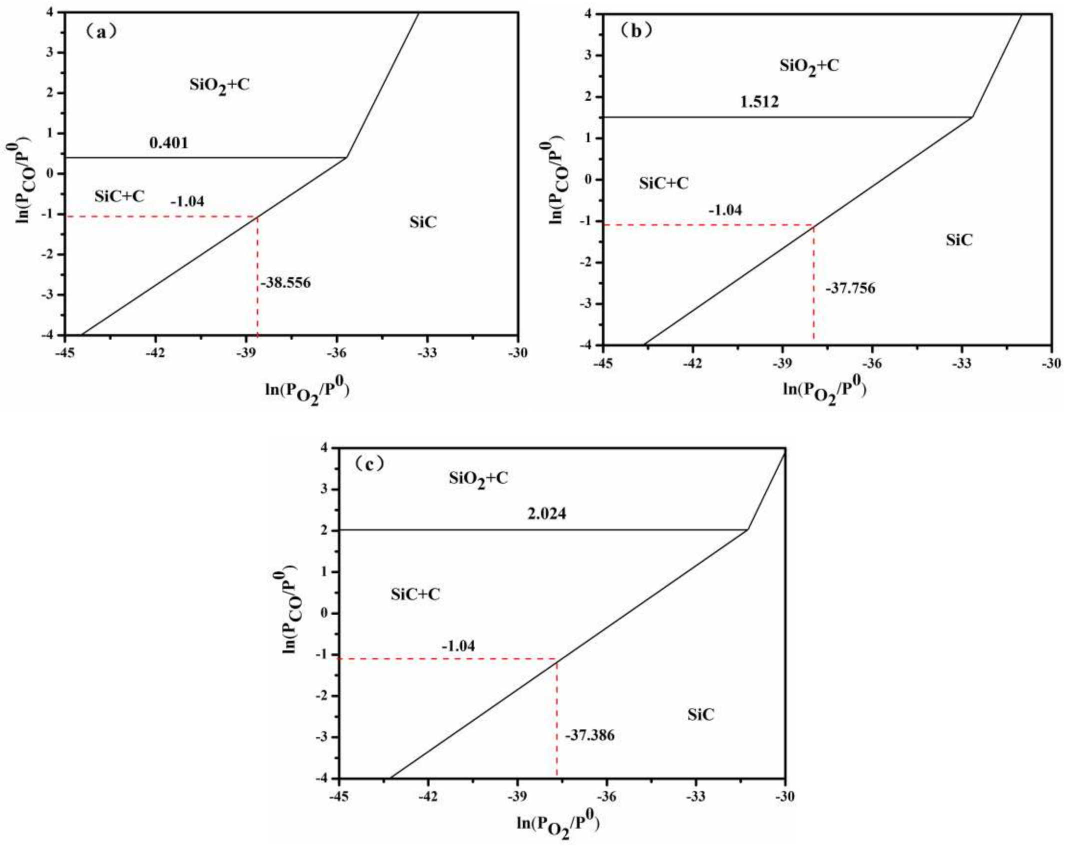


Fig.7 Predominance area diagrams of the system Si–C–O (a)1500°C, (b)1600°C and (c)1650°C. Red line is corresponding with the partial pressure of coke-packed bed atmosphere

Considering the coating conditions, a series of chemical reaction during the coating process could take place between carbon and SiO_2. The possible reactions in this system are as follows [30] in table 3.

Table 3 Chemical reaction and Gibbs free energy of Si–C–O system

Reaction	Gibbs free energy	ΔG(J/mol)		
		1773K	1873K	1923K
(1) $C(s)+0.5O_2(g) \rightarrow CO(g)$	$\Delta G=-110541-89.287T+8.314T\ln\{(P_{CO}/P^0)/(P_{O_2}/P^0)^{1/2}\}$	-5.56316	-0.03876	-6.87923
(2) $3C(s)+SiO_2(s) \rightarrow SiC(s)+2CO(g)$	$\Delta G=614044-353.005T+8.314T\ln(P_{CO}/P^0)^2$	-42494.6	-79524.4	-98039.3
(3) $2CO(g)+2SiO_2(s) \rightarrow 2SiC(s)+3O_2(g)$	$G=1891334-170.288T+8.314T\ln\{(P_{O_2}/P^0)^3/(P_{CO}/P^0)^2\}$	-84955.8	-159049	-196037

The analysis of thermodynamic calculation in table 3 illustrate that with the increase of heat treating temperature, the Gibbs free energy of reaction (1~3) becomes lower and lower and the value of reaction (3) exhibits the minimum value, which prove that this reaction is easy to take place. The results is agreement with the experimental results in Fig.1~3 that the higher treatment temperature could benefit the formation of SiC coating phase. The formation of SiC phases is due to the generation of SiO vapor from the microsilica-CMC reaction (1 and 2). Based on the atmosphere possesses lower CO pressure, SiC will be stable from predominant area diagram analysis in Fig. 7.

The progressive formation process of SiC coating layer is depicted in Fig.8. Firstly, microsilica is coated on the surface of flaky graphite and forms a precursor in the CMC solution. After drying the precursor, SiO_2 phases are attached on the surface of flaky graphite. When heat treated at 1500°C, the shape of SiO_2 coated on the flaky graphite surface becomes twisted and a minor SiC whisker is formed, shown in Fig.2b. At 1600°C, SiC whiskers become more and coat the surface of flaky graphite partly(Fig.2c). Meanwhile, a small amount of residual SiO2 exits. Raising the heat treatment temperature to 1650°C, the SiC layer becomes denser and the thickness of the coating increases because the whiskers grow further, which would result in the minimum weight loss, highest exothermic temperature and the minimum contact angle with water for the graphite/SiC core shell structure.

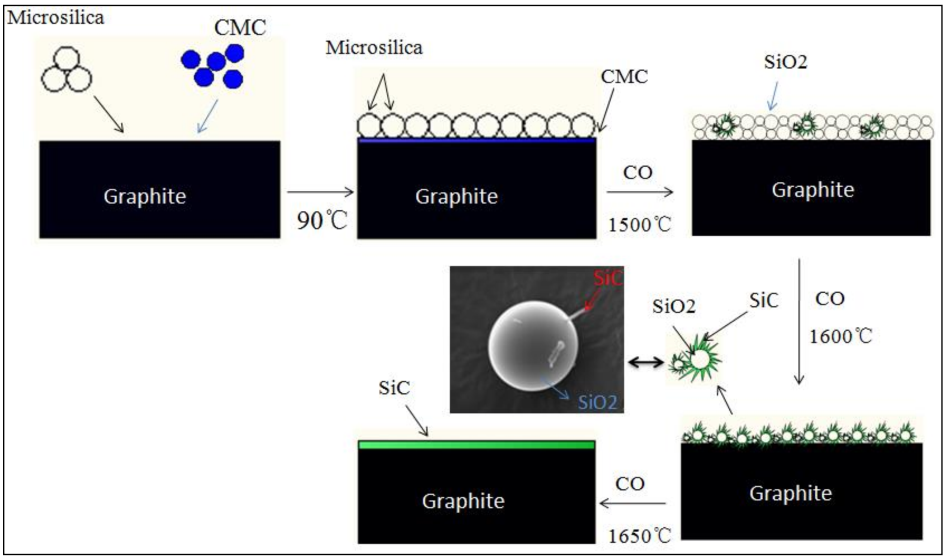


Fig.8 the formation process of SiC coating

4 Conclusions

In the present study, graphite/SiC core shell structure is successfully prepared by a novel method. Moreover, the specimen coated SiC whisker showed a remarkable improvement in terms of oxidation resistance and wettability. The conclusions obtained in this work can be summarized as follows:

(1) The SiC whisker formed on the surface of the graphite is mainly caused by the reaction of CMC and SiO_2 with increasing temperature. The carbonization of CMC effects a change of the partial pressure of CO. According to the thermodynamic calculation of Si–C–O system, the SiC coating is easily formed at lower CO pressure.

(2) The SiC coating can effectively enhance the oxidation resistance of graphite. Based on the results of TG and DSC analysis, SiC coating could decrease the mass loss of the graphite from 97.73% to 29.78%. At the same time, the enthalpy of carbon oxidation reaction is decreased from 12.02 kJ/g to 1.14 kJ/g.

(3) Graphite/SiC core-shell structure behaves excellently in terms of wettability, and the

contact angle with water is decreased from 78.5° to 43°.

Acknowledgments

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