

SiC composite ceramic materials oxidation behaviour based on three kinds of C_p content

SiC material is a kind of material with excellent properties, in order to inhibit its oxidation behaviour and prolong the service life of the material during the process of using, the oxidation behaviour of three kinds of SiC materials containing different amounts of C_p in the three temperature ranges from 400°C to 1100°C is studied in this paper. It emphatically analyzes the relations among C_p /SiC's quality change fraction in air and oxidation temperature, oxidation time, C_p /SiC's apparent porosity and oxidation temperature, C_p /SiC's bending strength and oxidation temperature, oxidation time. It is concluded that the C_p content of SiC is the key factor to influence the strength and oxidation behaviour of composite materials and the kind of SiC composite ceramics which add C_p can improve oxidation resistance of the materials.

Keywords: Temperature range, oxidation behaviour, mass change rate, show porosity, Bending strength

1.0 Introduction

Second phase material introduces ceramic matrix composite, it is a kind of multiphase material on the strength and toughness, also known as multi-phase composites or composite ceramics (Wei, et al., 2013). This kind of material is a new kind of ceramic material, gradually developed in the 1980s, including fiber toughening ceramic matrix composite, heterogeneous particle dispersion strengthening phase in situ growth ceramic composite material, functionally gradient ceramic, composite ceramic and nanometer ceramic composite (Zhao, et al., 2013). The performance of the ceramic composite is closely related with its structure. Raw material, structure and craft are different, the performance of the material is also different. Constituting the constituent material of the ceramic composite includes fiber, matrix and interface, for the C_p /SiC ceramic matrix composite, material and structure of the interface is the key to influence its performance (Noël, et al., 2014).

Messrs. Hongwei Xing, Zhenchao Liua, Yuzhu Zhanga, Ministry of Education Key Laboratory with Modern Metallurgical Technology, College of Metallurgy and Energy, North China University of Science and Technology, Tangshan 063210, and Hongyan Zhang, Institute of Light Industry, North China University of Science and Technology, Tangshan 063210, China. Email: xlding2015@yeah.net

C_p /SiC is a kind of composite material with carbon granule as the second phase and silicon carbide as matrix, which has some excellent performance like high temperature resistance, easy to process and corrosion resistance (Simone, et al., 2016; Ruffer, 2015).

Based on the previous research, in this paper based on the (C_p /SiC) ceramic matrix composites (Tang, et al., 2017; Wang, 2013; Lu, 2013) as the application of high temperature glass jig material, requires the component material should have high temperature resistance (900-1100°C), and it does not bond with the part (Kaushal, 2013), It also has some bending strength (1000-350 MPa) and can be recycled, in this paper, It studys the oxidation behaviour of composite C_p /SiC ceramic matrix composite and breaks the limitations of the application of materials.

2.0 Experiment

The main raw materials needed for the experiment are nanometer carbon black (C_p), Si, and B4C. The type of nanometer carbon black (C_p) is N330 whose purity is greater than 99% and the particle size fraction is 100-120 nm. The purity of Si is greater than 99% and the diameter is 1.592 μ m. The oleic acid ($C_{18}H_{34}O_2$) and anhydrous ethanol (C_2H_5OH) are used on the analysis of purity.

The instruments and equipment needed for the experiment are vacuum free sintering furnace, field emission scanning electron microscope and atmosphere muffle furnace. The model of vacuum free pressure sintering furnace is syl-600 and the manufacturer is Zhuzhou Norbert High Temperature Equipment Co., Ltd. The model of field emission scanning electron microscope is SEM/JSM7500F and the manufacturer is Japanese electronics company.

As shown in Table 1, different pulp formulations are shown.

In this experiment, the experiment steps of using micron-grade silicon powder and nanometer carbon black granules are shown as follows:

Step 1: Through mechanical chemical synthesis SiC and the remaining nano-carbon particles, the quality of the preparation in situ is 15% and 25% nano-carbon particles and silicon carbide composite powder.

TABLE 1: LIST OF DIFFERENT SLURRY FORMULA DATA

	Slurry	Unit	SiC	15% C _p /SiC	25% C _p /SiC
1	C _p /SiC	g	300	300	300
2	ethylalcohol	mL	195	195	195
3	H ₂ O	mL	105	105	105
4	B ₄ C	g	3	3	3
5	dispersant	g	0.9	0.9	0.9
6	lubricant	mL	3	3	3

Step 2: The method of synthesis of SiC and the preparation of nano C_p/SiC composite powder can be compared to the literature (Kim, et al., 2016).

Step 3: In accordance with the formula as shown in Table 1, in turn, into the polyurethane lining after weighing grinding jar, add a diameter of 15-20 mm steel polyurethane ball, the ball material ratio of 2:1, add preconfigured liquid mixture to tank premix.

Step 4: After 6 hours on the planet type ball mill mixing through 80 mesh sieve, sieve pore diameter of 178 microns, made into slurry, after spray granulation pressure molding, placed C_p/SiC ceramic body in a graphite crucible with argon as shielding gas.

Step 5: According to the process route shown in Fig.1, the sintered body samples were tested and the voids rate was characterized, and it was cut into 3 mm × 4 mm × 36 mm standard samples to do the three-point bending strength test.

The process flow chart of C_p/SiC composite ceramics sintering is shown in Fig.1.

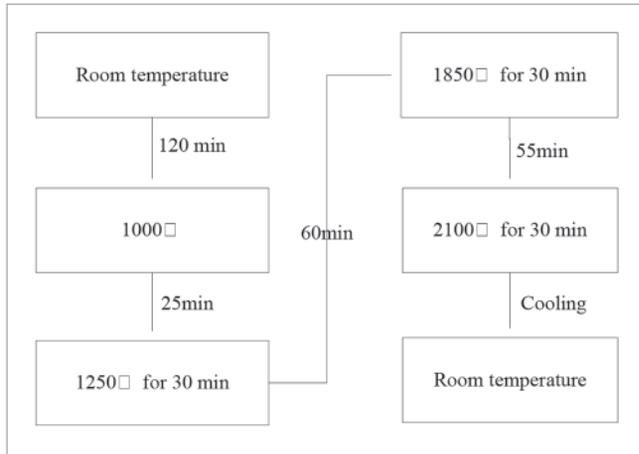
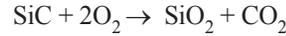
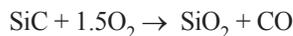
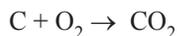


Fig.1 C_p/SiC process flow diagram of composite ceramic sintering process

3.0 Result and discussion

The oxidation mechanism of composite ceramic materials conforms to passive oxidation, and its reaction is shown below



The C_p/SiC composite ceramics are oxidized according to the equation shown above, and the actual oxidation process can be divided into several stages. The reaction of oxygen and the matrix is C_p. The oxygen diffuses through the SiO₂ membrane to the SiO₂/SiC interface. The reaction of oxygen with SiC in SiO₂/SiC interface. Reaction products CO and CO₂ gas diffusion is through SiO₂ membrane.

In the case of passive oxidation, silica amorphous phase was formed in the early stage of oxidation to fill the pores and reduce the porosity of the material; it makes the surface of silicon carbide material improve, some crack tip passivation, the material strength increases. Amorphous silica crystallization occurred above 1200°C, the higher is the temperature, the faster is the crystallization rate, then as the oxidation temperature increases the silicon dioxide film changes, the change of the phase makes the structure and the intensity decreases.

To explore the details of C_p/SiC's oxidation behaviour at different temperatures, this chapter from C_p/SiC in the air quality change rates ($\Delta m/m$) (%), oxidation temperature (°C), oxidation temperature (h), C_p/SiC show porosity of apparent porosity (%) and C_p/SiC bending strength (MPa) five quantities to analyze the problem, that is, the separate effect of oxidation temperature, oxidation time on $\Delta m/m$, the relationship between apparent porosity and oxidation temperature and the separate effect of oxidation temperature, oxidation time on bending strength.

3.1 THE EFFECT OF OXIDATION TEMPERATURE AND OXIDATION TIME ON $\Delta m/m$

To separately explore the effect of oxidation temperature, oxidation time on $\Delta m/m$, it is needed to have a contrasting comparison. Firstly, the relationship between the conversion rate and the oxidation temperature of three ceramic materials after 2 hours in different temperatures is analyzed. Finally, the time of oxidation is analyzed in the case of three materials under the 1000°C. Fig.2 shows the separate effect of oxidation temperature, oxidation time on $\Delta m/m$.

It can be seen from the analysis of Fig.2 that before 900(°C) the quality has not changed, and the quality has increased between 900(°C) and 1100(°C). When it reaches 1100°C and $\Delta m/m$ % = 11%, this temperature is the maximum value of $\Delta m/m$. Between 600°C and 1100°C, 15% C_p/SiC and 25% C_p/SiC materials have similar quality loss curves. In the temperature interval of 600°C~700°C, the performance is the loss of oxidation quality, and the phenomenon of rapid increase of oxidative mass loss with the increase of temperature. In the 700°C, the maximum value of the loss rate of the mass appeared. At the temperature range of 700°C ~1100°C, the loss rate of mass shows the trend of decrease with the increase of temperature.

In the first 2 hours at 1000°C, the quality rate tends to

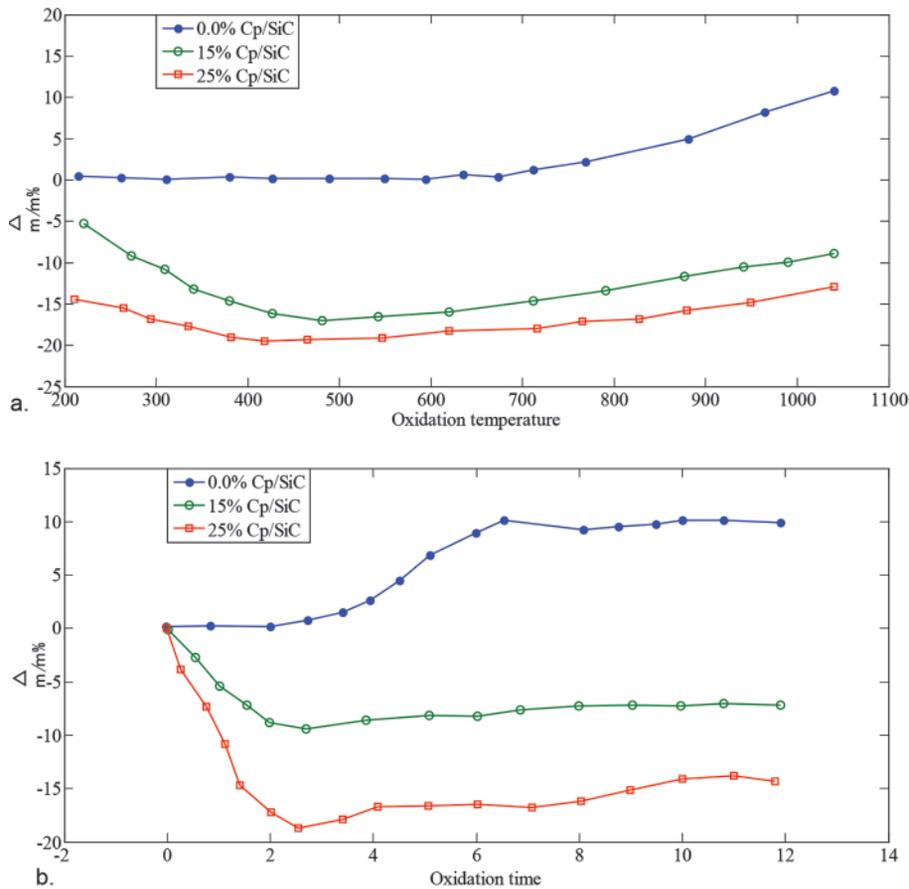


Fig.2 The separate effect of (a)oxidation temperature and (b)oxidation time on $\Delta m/m$

zero. During the 2 to 8 hours period, the quality change rate was significantly increased, and the quality change rate continued to increase during the 8 to 12 hours, but the rate of increase increased slowly. Due to Cp existence in the first 2 hours the two kinds of ceramic materials Cp/SiC (15%) and Cp/SiC (25%) at 1000°C make mass loss increased obviously. When the time between the 2 hours and 10 hours, the rate of oxidation mass loss gradually decreases and the quality rate tends to a constant value in 10 hours to 12 hours.

The reason for the corresponding trend of the quality of SiC ceramic material has no add Cp and oxidation time. In the initial stage of oxidation, the amorphous silica is formed, with the increase of oxidation time, amorphous state silica is gradually crystallized into square quartz. As oxidation time increases, the substrate is completely covered by the oxide, and the oxidative product itself is known as the protective layer that blocks the diffusion of oxygen, making the later oxidation rate slow. Because 15% Cp/SiC and 25% Cp/SiC two kinds of the quality rate of ceramic materials tends to a constant value in 10-12 hours; is the formation of the stability of silica passivation

membrane, thus inhibiting the two ceramic materials continue to oxidation.

3.2 THE RELATIONSHIP BETWEEN APPARENT POROSITY AND OXIDATION TEMPERATURE

To explore the relationship between the quality change rates ($\Delta m/m$) of Cp/SiC in the air with oxidation temperature, there is also a comparison. In this paper, three kinds of ceramic materials SiC without Cp, 15% of Cp/SiC and 25% of Cp/SiC after the oxidation of 10 hours, the relationship between Cp/SiC's apparent porosity (%) and oxidation temperature (°C) is analyzed. The porosity of ceramic materials can be calculated as formula (1).

$$P_a = \frac{m_3 - m_1}{m_3 - m_2} \times 100\% \quad \dots (1)$$

As shown in Fig.3, the relationship trend chart between the apparent porosity and oxidation temperature are of three ceramic materials.

It can be seen from the analysis of Fig.3 there is no obvious change after oxidation that the voids rate of SiC ceramic material is without Cp in the range of 400-1100°C. The 15% of Cp/SiC and 25% of Cp/SiC ceramic materials showed an increased porosity in the range of 400-700°C, and the porosity decreased in the range of 700-1100°C.

The reasons for the change of porosity in the two ceramic materials 15% of Cp/SiC and 25% of Cp/SiC in the range of 400-1100°C. The increase of porosity in the interval between 400°C and 700°C is due to the rapid oxidation of Cp in this interval. In the range of 900-1100°C SiC begins to oxidize and silicon dioxide protect passivation film begins to form. It has

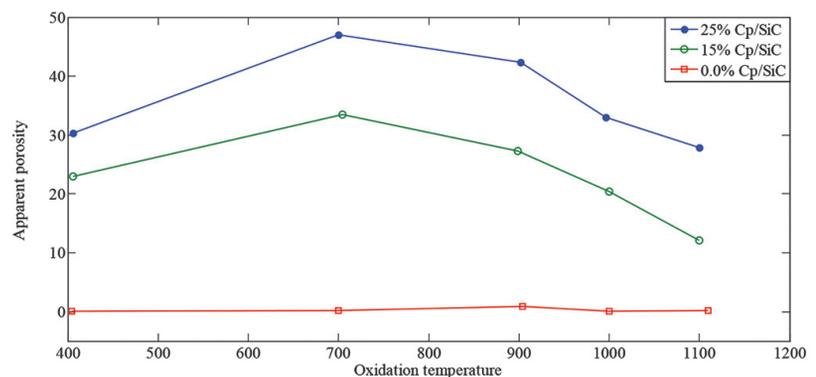


Fig.3: The relationship trend chart between the apparent porosity and oxidation temperature

the effect of filling the stomata and reducing the porosity of the material, so that the original defects in the SiC material have been improved, resulting in the decrease of the voids.

3.3 THE EFFECT OF OXIDATION TEMPERATURE AND OXIDATION TIME ON BENDING STRENGTH

To separately explore the effect of oxidation temperature, oxidation time on bending strength, it is needed to have a contrasting comparison. The separate effect of oxidation temperature and bending strength of the two ceramic materials-15% C_p/SiC and 25% C_p/SiC on flexural strength is

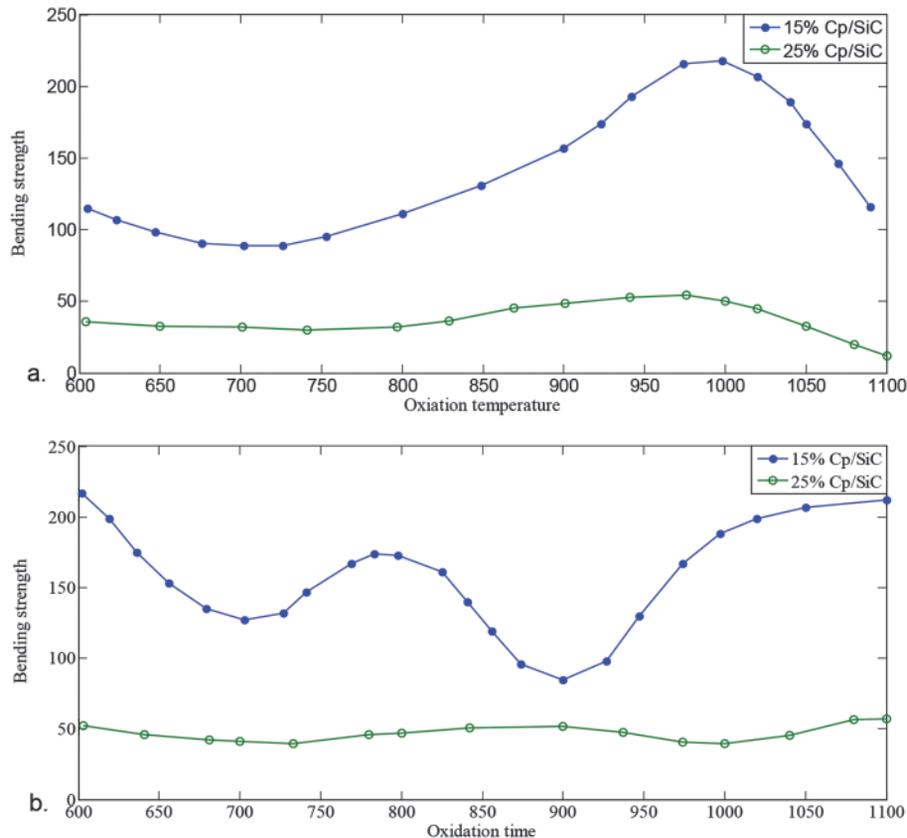


Fig.4 The separate effect of (a)oxidation temperature and (b) oxidation time on bending strength.

analyzed in this paper. Fig.4 shows the separate effect of oxidation temperature, oxidation time on bending strength.

By analyzing Fig.4, It is clear that when the temperature of the ceramic material is between 600°C and 700°C there is a obvious loss on ceramic quality and decrease on bending strength. When the temperature is between 700°C and 1000°C, the SiC of 15% C_p/SiC composite ceramics gradually oxidizes into silicon dioxide protective film, then resulting in an increase in the bending strength of the materials. When the temperature is 1000°C, the maximum value is reached, and then the oxidized passage is easily formed with the increase of the temperature, which makes the bending strength decrease sharply. Due to increased carbon content, the 25% of C_p/SiC composite ceramic will delay the formation of silica film, which will be consistent with the change of 15% of C_p/SiC material after 800-1000°C, SiC oxidized gradually and formed a silica coating, lead to the increase of bending strength, and reaching the maximum when the temperature is 1000°C, and then it gets oxidized more with the rise of temperature, and it is easy to form oxidation channel and all of which makes the bending strength reduced sharply.

When the temperature is 1000°C, with the prolongation of oxidation time, the change of flexural strength of two kinds of ceramic materials is complicated. The bending strength decreases during 2-4 hours of heat preservation, and the bending strength increase within 4-6 hours. With the increase of holding time, the bending strength decreases and the

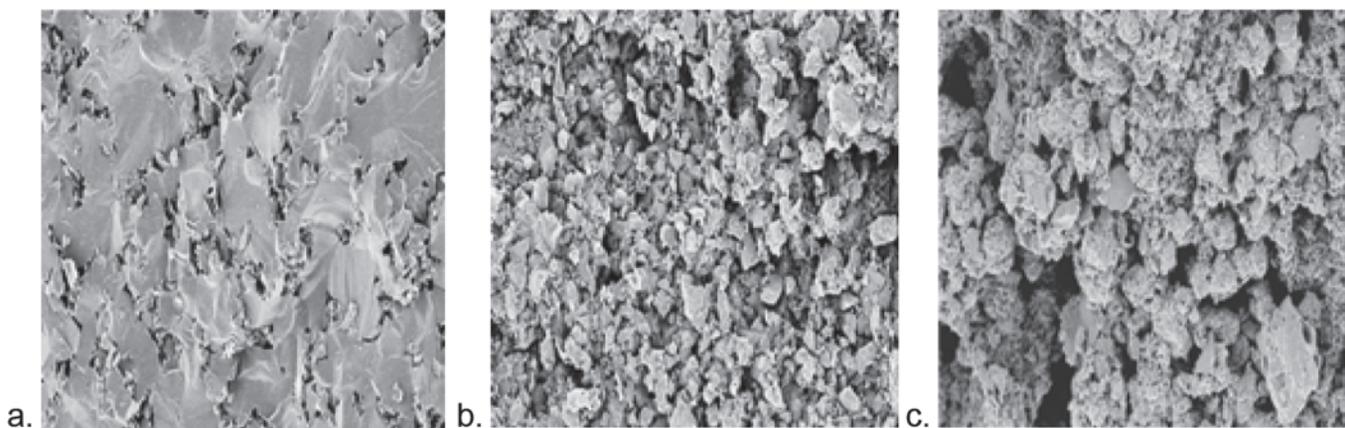


Fig.5 SEM photos comparison of three ceramic materials (a) 0% and b)15% and (c)25%

minimum value is 8 hours. Then as the holding time continues to increase, the bending strength obviously increases and the increase gradually slows down. In the initial stage of oxidation, the oxidation of carbon increases the porosity of the composite ceramics, and bending strength decreases. The oxidation of SiC ceramic matrix occurs with the increase of oxidation time. In the early stage of oxidation, the silica passivation film is formed, which fills the pores and reduces the porosity of the material, thus improving the surface defects of the SiC material.

It can be seen from Fig.5 that the ceramic structure in SiC without C_p is compact and the porosity is close to zero; in the 15% of C_p /SiC, the nano-particles are distributed evenly around the SiC crystals, forming a compact C_p /SiC composite ceramics; in the 25% of C_p /SiC, nano-carbon is easy to reunite, forming the network hole structure of conveying, making the strength of ceramic materials drop sharply. Obviously, with the increase of nanometer carbon content, the porosity of composite materials significantly increases under 900°C.

Conclusions

In this paper, the oxidation behaviours of C_p /SiC ceramic matrix composite in different temperature ranges have been studied. In order to highlight the characteristics of the oxidation behaviour of the material, three ceramic materials are compared and analyzed by SiC. The mass change rate, apparent porosity, three point bending strength and SEM photos of three kinds of materials at different temperatures are analyzed. When the temperature range is from 400-700°C, the oxidation behaviour of C_p /SiC composite ceramic in air is controlled by the reaction of carbon and oxygen. The composite ceramics show uniform oxidation and increase the apparent porosity, the bending strength decreases with the increase of the oxidation temperature, and the minimum value is at 700°C. When the temperature range is from 700-1000°C, the oxidation process is controlled by the diffusion of oxygen gas and the diffusion of oxygen through the micro cracks to control the oxidation process of the material. With the formation of silica phase, the apparent porosity decreases, the bending strength increases, and reaches maximum at 1000°C. When the temperature range is 1000-1100°C, the diffusion of oxygen through the SiC defect controls the oxidation process of the composite ceramics. The apparent porosity increases and the bending strength decreases. It is proved that the content of C_p in SiC is the key factor affecting the strength and oxidation behaviour of composite. The addition of C_p to SiC composite ceramics can enhance the oxidation resistance of the material.

Acknowledgments

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