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Effects of temperature and duration on oxidation of ceramic composites with silicon carbide matrix and carbon nanoparticles

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ABSTRACT

This paper was to investigate the oxidation behavior of three pressureless sintered ceramic composites of silicon carbide (SiC) matrix with various contents of carbon nanoparticles (C_p) (i.e., 0, 15 and 25 wt%) at various temperatures (i.e., 600–1100 °C) and durations (i.e., 2–12 h). The results indicate that the oxidation of SiC/C_p ceramics in air is uniformly controlled by the oxidation of C_p at <700 °C, and the strength of the samples reaches a minimum value at 700 °C. At 700–900 °C, the oxidation is controlled via the unequal diffusion of O_2 through the micro-cracks. The oxidation of SiC/C_p is controlled by O_2 diffusion through the structural defects of the matrix at 900–1100 °C, and the strength of the composites increases at 1000 °C and reaches a maximum value during the initial period of 2 h due to the formation of SiO_2 layer and the healing of the matrix defects.

1. Introduction

The ceramic composite of silicon carbide matrix with carbon nanoparticles as a modified secondary phase (SiC/C_p) possesses an excellent thermo-stability and a corrosion resistance. Also, the addition of carbon nanoparticles into the matrix composite could improve the machinability [1]. These composite materials are attractive due to their potential applications in the glass industry [2]. It is well known that non-oxide ceramics are easily oxidized in air at increased temperatures and durations, leading to the eventual breakage and restricting the use of SiC/C_p composites [3–8]. Therefore, it is important to clarify the oxidation behavior of this non-oxide material as a high-temperature structural material at different temperatures and durations. Some recent studies on the fabrication, oxidation and anti-oxidation of SiC/C_f composites with carbon fiber (C_f) were carried out [9–13], but little information on the SiC/C_p composites with carbon particles (C_p) were reported.

As glass holder and clamp, the SiC/C_p composite must possess a thermal stability in the temperature range of 900–1100 °C and an appropriate strength. Also, this composite should not adhere to the glass component, withstand to be sustained and reuse.

This paper was to investigate the oxidation behavior of SiC/C_p composites with different contents of carbon nanoparticles as a promising ceramic composite material of glass holder and clamp in glass industry at various temperatures (i.e., 600–1100 °C) and durations (i.e., 2–12 h).

2. Experimental

2.1. Starting materials

The starting materials used were a carbon nanopowder (C_p) (N330, purity $\geq 99\%$, $d_{50}=0.091$ μm, SSA=89 m^2/g), a silicon micropowder (purity $\geq 99\%$, $d_{50}=1.592$ μm), B_4C as a sintering additive (Qianjin Boron-carbide Ltd. Co., China), oleic acid as a lubricating agent (analytical grade), and ethyalcohol (analytical grade).

2.2. Procedure and methods

The silicon powder was firstly mixed with a certain amount of carbon nanoparticles (i.e., from 0.0 wt% to 25 wt%), and then ground in a mode QM-1 planetary stirred ball mill (Nanjing Scientific Apparatus Research Institute, China) for 24 h to synthesize β-SiC

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and simultaneously prepare a SiC/C_p composite powder. The composite powders were rinsed with acid (i.e., hydrofluoric acid and muriatic acid) and distilled water for 5 times, and dried in a dryer at 80 °C. The detailed information of preparing the composite powder via a mechanochemical route was given in our previous work [14].

The composite powders with various contents of carbon nanoparticles (i.e., 0.0 wt%, 15.0 wt% and 25 wt%) were firstly mixed with B_4C, oleic acid, ethylalcohol and chemical disperser in a certain of ratio, respectively. B_4C was used as a sintering assistant to enhance the combination property of SiC/C_p ceramic composite during the pressureless sintering. The proportion ranges of C_p contents in the composite used in this study were selected based on our previous work [15]. It was found that the mechanic strength of the composite with C_p could be dominantly reduced at the C_p content of >25 wt%. The mixed materials were then wet-ground in the mill for 6 h. Table 1 shows the composition of each mixture. The as-prepared mixed slurries were dried by a spray dryer to obtain the hollow spherical particles available for molding by dry pressing. The green bodies with the size of 50 mm × 50 mm × 10 mm were obtained after the hollow spherical particulate materials were firstly dry-pressed in a molding press. The green bodies in a graphite crucible were then pressureless-sintered in a furnace (Zhuzhou Norbert High Temperature Instrument Ltd. Co., China) with Ar atmosphere. Table 2 shows the experimental design with various C_p contents, oxidation temperatures and durations. Fig. 1 shows the sintering process of the SiC/C_p composite.

2.3. Characterization

The crystalline phase of the composite samples was identified by X-ray diffraction (mode XRD-6000, Shimadzu Co., Japan). The formation of the composite samples was observed by scanning electron microscopy (mode SSX-550, Shimadzu Co., Japan).The flexural strength of the composite samples was determined byan electronic universal testing machine (mode CMT5305, Shenzhen Xinshansi Measurement Technique Co. Ltd., China). The particle size of the ground powder was determined by a laser particle size analyzer (mode MicrotracX-100, Honeywell Co., USA). The specific surface area of the samples was measured by BET nitrogen adsorption analyzer (mode HM1201, Beijing Diline instrument Co. Ltd., China). The density of the sintered composite samples was measured based on the Archimedes principle. The microhardness of the sintered composite samples was determined by a microhardness tester (mode 432SVD+LEVEL2, Walter Potter Co., USA). The apparent porosity (P_a) of the sintered composite samples was determined by

$$P_a = \frac{m_3 - m_1}{m_3 - m_2} 100\% \quad [1]$$

where m_1 is the apparent mass of the dried samples, m_2 is the mass of the saturation samples and m_3 is the mass of the saturation samples in air.

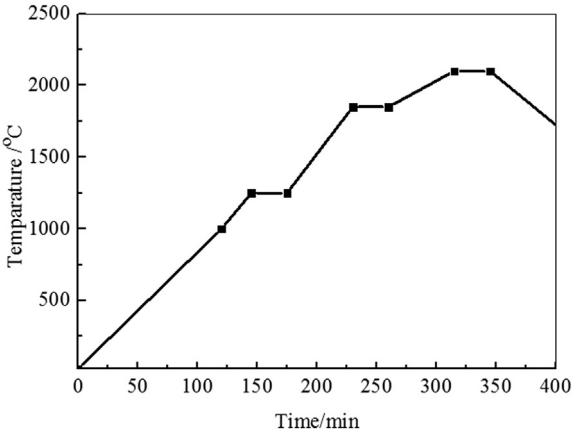

Fig. 1. Sintering process of the SiC/Cp composites.

Table 1
Compositions of the mixtures.

No.	SiC/C_p/g	Ethylalcohol/ml	H_2O/ml	B_4C/g	Disperse agent/g	Lubricant/ml
1#(SiC)	300	195	105	3	0.9	3
2#(15 wt% SiC/Cp)	300	195	105	3	0.9	3
3#(25 wt% SiC/Cp)	300	195	105	3	0.9	3

Table 2
Experimental design with various C_p contents, oxidation temperatures and durations.

No.	Cp/%	Oxidation temperature /°C	Oxidation duration/h	No.	Cp/%	Oxidation temperature /°C	Oxidation duration/h
1#-1	0	600	2	3#-6	25	1100	2
2#-1	15	600	2	1#-7	0	1000	4
3#−1	25	600	2	2#-7	15	1000	4
1#-2	0	700	2	3#-7	25	1000	4
2#-2	15	700	2	1#-8	0	1000	6
3#-2	25	700	2	2#-8	15	1000	6
1#-3	0	800	2	3#-8	25	1000	6
2#-3	15	800	2	1#-9	0	1000	8
3#-3	25	800	2	2#-9	15	1000	8
1#-4	0	900	2	3#-9	25	1000	8
2#-4	15	900	2	1#-10	0	1000	10
3#-4	25	900	2	2#-10	15	1000	10
1#-5	0	1000	2	3#-10	25	1000	10
2#-5	15	1000	2	1#-11	0	1000	12
3#-5	25	1000	2	2#-11	15	1000	12
1#-6	0	1100	2	3#-11	25	1000	12
2#-6	15	1100	2				