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The effects of carbon/carbon composites blasting treatment and modifying SiC coatings with SiC/ZrB_2 on their oxidation and cyclic ablation performances

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ABSTRACT

To enhance the oxidation and ablation performances of SiC coating, C/C composites with a porous surface layer were designed via blasting treatment and then followed by ZrB_2 additive in the process of pressure-less reactive sintering. Cyclic ablation test using oxyacetylene torch from 1750 °C to room temperature showed that the coating possessed good ablation performance accompanying with the formation of Zr-O-Si glassy protective layer. The good sintering performance combined with the increased coating/substrate interfaces contributed to reduce the cyclic thermal stress and enhance the stability of the Zr-O-Si layer to the denudation of oxyacetylene torch, thus promoting the ablation performance.

1. Introduction

Carbon/carbon (C/C) composites have shown attractive application advantages due to their good properties including low density, low thermal expansion coefficient (CTE), sufficient thermal shock resistance, good designability and high specific strength [1–5]. However, oxidation is a major problem that hinders C/C from wide applications [2,3,6–9].

Coating technology has been proved a simple and effective way to address the problem [8–12]. Due to the self-sealing performance of glassy SiO_2 (the oxidation product of SiC), SiC is frequently used to protect carbon fiber reinforced composites against oxidation at high temperatures [2,11–13]. However, the thermal mismatch together with the adhesive strength of SiC coating and C/C substrate is the key issue that needs to be focused on. Unless the aforementioned problems are properly handled, cracking and spallation of the SiC coating will occur during thermal cycles of high and low temperatures. Interface optimization is proved to be a good choice. Feng.et.al [14] used low-density C/C composites (1.2 g/cm^3) to make use of their porous structures, which could provide the diffusion paths to the coating raw materials and then result in the increased coating/substrate interfaces. The increased interfaces could promote the oxidation performance of the produced coating. However, to ensure the thermal-structural components with favorable mechanical performances, C/C composites are required with sufficient density (> 1.7 g/cm^3) and low porosity. Thus, this method is difficult to take in practical applications. Finding an efficient way to induce a porous surface layer on high-density C/C (in the premise of minimum mechanical property loss) is becoming particularly important. As a result, pre-oxidation treatment of C/C was developed to construct a porous surface layer [15]. The results showed that the treatment time was relatively longer (up to 6 min), resulting in the inevitable damage of the mechanical performance of C/C. For real applications, the implementation of this method is also constrained by the size of the C/C, and more importantly, the oxidized regions of C/C are very difficult to choose, which cannot meet the requirements of C/C thermal structural components.

As an alternative method of pre-oxidation treatment, blasting operation of C/C was proposed to enhance the property of SiC coating [16,17]. Compared with pre-oxidation treatment, blasting treatment is much more easy-operated to construct an inlaid coating/substrate interface. This method is not limited by the shape as well as the size of C/C and can significantly reduce the processing time. And more importantly, the treated region is easy to choose, which can construct a porous surface layer on a specific region. But the performance improvement of SiC coating was still limited due to the poor thermal stability of SiO_2 glass when subjected to cyclic ablation test using oxyacetylene torch at temperatures ranging from 1750 °C to room temperature [16]. Introducing ultra-high temperature ceramics (UHTCs) into SiC coating might be an appropriate choice to enhance its oxidation and ablation properties at high temperature (> 1700 °C). So,

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HfC was attempted to be introduced into SiC coating by pressure-less reactive sintering to promote the thermal stability of the SiO_2 glass [18]. However, when the ablation time further extended, the performance enhancement was restricted due to the coating brittleness caused by its limited sintering performance, which affected the thermal stability of the formed HfO_2-SiO_2 protective layer to the denudation of oxyacetylene torch, and then resulted in the performance degradation. So, it is recognized that pretreatment of the C/C substrate and structure optimization of the coating should be combined.

ZrB_2, as one of the UHTCs, is also regarded as a promising material for thermal protection system because of its sufficient retained strength at high temperature and high melting point [19–23]. It has been reported that ZrB_2 addition is beneficial for reducing the SiO_2 consumption [8,20,21,23], and the glass formed by B_2O_3 and SiO_2 (the oxidation products of ZrB_2 and SiC) also possesses good self-sealing performance in a larger temperature range. Furthermore, the CTE difference between ZrB_2 and SiC is smaller than that of HfC and SiC ($\alpha_{ZrB2} = 5.9 \times 10^{-6}$/ °C [24], $\alpha_{SiC} = 4.5 \times 10^{-6}$/ °C [25], $\alpha_{HfC} = 6.6 \times 10^{-6}$/ °C [26], $\alpha_{C/C}$ =1-2 $\times 10^{-6}$/ °C [18,26]), which could benefit for the improvement of sintering performance and then make it easier for the coating preparation. But previous studies about ZrB_2 modified SiC coating are mainly focused on its oxidation behavior in static air or ablation property obtained in a single ablation test [20,23,27–29]. As a promising candidate for reusable thermal structural component, the coated C/C would be subjected to repeated ablative environment [30,31], so it is deemed important to evaluate the cyclic life of the prepared coating in dynamic environments. However, few studies have been performed to investigate the cyclic ablation performance of the ZrB_2 modified SiC coating on C/C [20,23,27–30,32], which is limited by the coating/substrate interface and the thermal stability of the formed oxidation products. So, blasting treatment combined ZrB_2 additive is expected to possess a better application prospect to promote the performances of SiC coating under the condition of dynamic combustion environment. In addition, whether blasting treatment will cause the severe mechanical performance degradation of C/C needs to be studied, which was not discussed before [16–18].

In this work, the effects of C/C composites blasting treatment and modifying SiC coating with SiC/ZrB_2 on the oxidation and cyclic ablation performances were investigated. The mechanical performance evolution of C/C via blasting treatment was studied by compression test. Cyclic ablation test was performed using oxyacetylene torch at temperatures ranging from 1750 °C to room temperature. As a supplement, thermogravimetric analysis (TGA), adhesive strength and CTE test were also performed.

2. Experimental procedure

2.1. Construction of a porous surface layer on C/C and introduction of ZrB_2 into SiC coating

Cylinder C/C specimens (Ø10 mm × 10 mm) with the density of 1.7–1.75 g/cm^3 were used. They were hand-polished by 400 grit SiC papers, cleaned with water in ultrasonic bath and then dried in electrothermal dry box. The procedure of blasting treatment as well as the processing of ZrB_2-SiC coating is exhibited in Fig. 1. The process was divided into two stages. In the first step, blasting treatment using oxyacetylene torch was performed to induce a porous layer on C/C. As shown in Fig. 1, the distance from oxyacetylene gun to C/C was 10 mm. During blasting treatment, gas flux and pressure of O_2 were 1.12 m^3/h and 0.4 MPa, and the gas flux and pressure of C_2H_2 were 0.83 m^3/h and 0.095 MPa. The processing time was 25–30 s. In step 2, ZrB_2 was introduced into SiC coating by pressure-less reactive sintering (or pack cementation). Powder composition was as follow: 45–65 wt. % Si (Jiuling Smelting Co., Ltd., Shanghai, China), 10–15 wt. % ZrB_2 (Dingdong Chemical Engineering Institute Co., Ltd. P.R.China) and 8–30 wt. % graphite (Carbon Plant, Xi'an, China). The mixed powders were well stirred in a ball grinding mill. The blasting treated C/C specimens (obtained in the first step) together with the prepared mixed powders were placed into a graphite crucible. After that, the graphite crucible was heat-treated in argon atmosphere. The crucible was firstly heated to 2000–2100 °C with the heating rate of 5 °C /min, then held for 2 h and finally cooled to room temperature with the cooling rate of 5 °C /min.

2.2. Characterization

2.2.1. Compressive strength test and cyclic ablation test

The compression tests were conducted using an electronic universal testing machine (CMT5304-30 kN). The sample size was Ø10 mm × 10 mm. During compression test, the rate of loading was 0.5 mm/min. Compression strength (σ) was calculated as follow,

$$\sigma = \frac{P}{F} \tag{1}$$

where P is the maximum load; F represents the cross-sectional area of the sample. The final strength is calculated by the mean of three specimens.

To obtain an environment of high temperature accompanying with combustion gas corrosion, oxyacetylene torch was adopted to conduct the cyclic ablation test. During the cyclic ablation test, gas flux and pressure of O_2 were 0.88 m^3/h and 0.4 MPa, and the corresponding gas flux and pressure of C_2H_2 were 0.65 m^3/h and 0.095 MPa. A detailed description of the cyclic ablation test is demonstrated in Fig. 2. Firstly, the mixed gas of O_2 and C_2H_2 was ignited. With the help of infrared radiation thermometer (accuracy of 0.75%, Raytek MR1SCSF), the distance from the oxyacetylene gun to the tested sample was adjusted to 10–14 mm to make the surface temperature reach 1750 °C. Inset in bottom-left corner of Fig. 2 exhibits the increasing temperature curve of the tested sample during ablation. During cyclic ablation test, the surface temperature increased rapidly, and the maximum temperature fluctuated around 1750 °C. After every 20 s (the ablation time at maximum temperature 1750 °C was about 16 s), the oxyacetylene gun was moved away, and the coated sample was cooled naturally before the next ablation cycle. The coated C/C without undergoing blasting treatment was referred as Z-S-1. The coated C/C underwent blasting treatment was labeled as Z-S-2. In each ablation cycle, the mass loss per unit area (ΔW) was measured to evaluate the ablation performance of the prepared coating, which was obtained by the following formula (2) [17,18]:

$$\Delta W = \frac{m_0 - m_1}{A} \tag{2}$$

where A represents the ablation surface area; m_0, m_1 are the mass before and after cyclic ablation test; The final ΔW is calculated by the mean of three samples.

2.2.2. Microstructure analysis, phase composition, TGA, CTE and adhesive strength test

Scanning electron microscopy (SEM, VEGA 755136XM) combined with energy dispersive spectroscopy (EDS) was adopted to observe the microstructures and morphologies. X-ray diffraction (XRD) was adopted to analyze the phase compositions on a Philips X'Pert MPD diffractometer. Spherical sample with the radius about 1.7 mm was used for TGA test, which was performed in air condition by thermal analysis apparatus (Mettler Toledo Star TGA/SDTA 851). CTE was performed by a Dilatometer (DIL402C). The adhesive strength test of the prepared coating before cyclic ablation test was measured with the help of an adhesive method, which was described in [18]. With the help of a confocal laser scanning microscope (C130, Lasertec Corp., Yokohama, Japan), the average surface roughness (Ra) of C/C before and after blasting operation was measured.