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A glass-ceramic coating with self-healing capability and high infrared emissivity for carbon/carbon composites

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ABSTRACT

Featured with the dense and stability under high temperature, a ZrSiO₄-aluminosilicate glass coating for the carbon/carbon composites was designed and prepared via a slurry painting-sintering process. The molten aluminosilicate glass can fill the crevices and cracks to improve the compactness, and play a self-healing role to restrain the propagation of the cracks caused by thermal mismatch under the high temperature. Furthermore, it has high infrared emissivity, which is helpful for cooling of composites to improve the antioxidant capacity. This coating has high antioxidation capability with a mass loss of 23.8 g/m² for substrates at 1773 K for 20 h.

1. Introduction

Carbon/Carbon (C/C) composites have a great potential for high performance structural materials due to their fine mechanical properties at high temperature, low density and low coefficient of thermal expansion (CTE), etc. [1–6]. However, C/C composite materials show rapid oxidation above 773 K in oxygen-containing atmosphere, resulting in the sharp decrease of 40–50% in weight within a minute, which limits their high-temperature applications [7–9]. So, antioxidant protection becomes an imperative means for C/C composite. Many investigations have shown that anti-oxidation coating is an effective strategy to protect the C/C composites in high-temperature environment [10–12], among which the high-temperature glass-ceramic coatings attracts much attention. Featuring with excellent chemical inertness, low thermal conductivity, high temperature stability and superior mechanical properties, the glass-ceramic coatings have been conceived as an effective thermal barrier coating for C/C composites [11,13–19]. A high temperature flow matter, such as SiO₂, is an important component of glass-ceramic coating. It can spread across the surface of substrate and fill the cracks to form a dense layer. Generally, as to silicon-based ceramics, a dense SiO₂ glassy film with low oxygen permeability will form on the surface of substrate at high temperature. For example, Walker and Corral [15] prepared the UHTC (Ultra high-temperature ceramics) filled C/C composite by vacuum infiltration, which can self-generate the high-temperature oxidation-resistant ZrO₂-SiO₂ glass-ceramic coatings at 1600 °C. This glass-ceramic coating is formed

by the spread of self-generating SiO₂ on C/C composite, showing an enhanced oxidation protection. R.Y. Luo team [18] fabricated a gradient self-healing coating consisting of three layers, SiC-B₄C/SiC/SiO₂. The outer layer SiO₂ was prepared by hydrolyzing tetraethylorthosilicate (TEOS) on the surface of SiC at an acid condition, and closing down the micro-cracks. K. Kobayashi et al. [19] prepared the carbon-B₄C-SiC composites with two different B₄C-SiC ratios. The B₂O₃-rich and SiO₂-rich borosilicate layer coating formed by preheating at 800 °C and 1200 °C for 10 h, respectively. The flowing borosilicate plays an important role in the formation of the coating. On the one hand, applying such coating can reduce the degradation of the C/C composites at high temperature. On the other hand, if the coatings have high infrared heat radiation coefficient, it can considerably reduce the surface temperature. As a consequence, the oxidation of substrate can be retarded.

In addition, SiC is usually applied as a bonding layer between C/C composites and outer layer of coating because of its good physical and chemical compatibility with C/C composites and ceramic coating [3–5,7,8,12–14,16,17]. Zirconium silicate (ZrSiO₄) has a high melting point (2523 K), and it has good chemical and thermal stability (phase transformation temperature higher than 1973 K), low oxygen permeability rate and good erosion resistance. Accordingly, ZrSiO₄ become a good candidate to prepare antioxidant coating for C/C composites [9,20–23]. Meanwhile, the thermal expansion coefficients of ZrSiO₄ and SiC are close ($4.5 \times 10^{-6}/\text{K}$, $3.7 \sim 4.8 \times 10^{-6}/\text{K}$, respectively). In other words, they have a good thermal expansion matching. So, SiC-

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ZrSiO₄ double layer coating can greatly improve the anti-oxidant performance of C/C composite material. However, the pure ZrSiO₄ coating has micro-pores and micro-cracks, which will seriously affect the anti-oxidant performance of coating at high-temperature. As mentioned above, using the high-temperature liquid component to fill these pores is necessary for the formation of dense coating [16,18,19]. Additionally, it is also noteworthy that in most preparation processes for coatings, gaseous impurities will be produced when the precursor reacts to form the component of coating, resulting in defects, such as pores or cracks.

In this paper, a glass-ceramic coating composed of ZrSiO₄ and aluminosilicate glass was developed. It was prepared on the surface of SiC-modified C/C preform by slurry painting and subsequent sintering method. The aluminosilicate glass was employed to act high-temperature liquid candidate, which does not decompose or react with other components to produce gaseous impurities at high temperature in the present work. During the sintering process, the molten aluminosilicate glass will spread across the surface of preform, carrying the stable ZrSiO₄ particles and filling the cracks at the same time to form a dense ceramic-glass coating with self-healing ability. In addition, the coating has high infrared emissivity under high temperature. The compact and stable structure, coupled with high infrared radiation ability at high temperature, endowed the ZrSiO₄-glass coating with enhanced anti-oxidation performance than single SiC film.

2. Experimental

2.1. Specimens preparation

The reagents silicon (Si) (300 mesh), graphite (300 mesh), ZrSiO₄, Polyethylene Glycol 6000 (PEG-6000 for short, the chemical formula is expressed in HO(CH₂CH₂O)_nH, where *n* represents the average number of oxygen vinyl groups, and the average chemical formula weight is about 5400-7800. It is soluble in water and alcohol, serving as dispersant, adhesive, softener, and so on.), and ethanol are analytical grade and used without further purification, which are provided by Sinopharm Chemical Reagent Co., Ltd, China. As to glass powder, the aluminosilicate glass is firstly heat-treated at 1773 K for 2 h. After water quenching, it is crushed and ground. Finally, the aluminosilicate glass powder is screened by 800 mesh. Here, it needs to mention that the composition of the aluminosilicate glass powder is roughly determined by XRF (Section I, Table S1). It is mainly consisted of the silicon oxide, alumina, and a few other common oxides such as iron oxide, sodium oxide, etc. The hemispherical point temperature test shows that the glass powder is melted into a hemispherical shape at about 1253 K, which can be considered to the melt temperature of glass powder (Section II, Fig. S1). The XRD result of glass powder indicated they were amorphous (Section II, Fig. S2).

3D C/C preform with 1.90 g/cm³ in density was chosen as substrates. After sanding, cleaning, drying, the preform is buried in the mixture of graphite powder and silicon powder with the weight ratio of 4:1, as illustrated in Fig.1-I. Subsequently, the samples were heat-treated at 1773 K for 2 h in argon atmosphere, forming the inner SiC-coated C/C preform, which was appointed as the substrate in the following procedure. The ZrSiO₄ and amorphous aluminosilicate glass powder were employed for building outer glass-ceramic cladding. As shown in Fig.1-II, the construction of it included the slurry painting and subsequent sintering processes. Firstly, the ZrSiO₄ and aluminosilicate glass powders were mixed and milled, and then added into alcohol with 1:8 of the weight ratio of the powder mixtures and alcohol to obtain the slurry. Subsequently, the PEG-6000 with a mass ratio of about 1:50 to glass powders is added into the slurry to achieve a suspension solution. Next, the process of painting is carried out by immersing the SiC-coated C/C preform in the slurry, and subsequent draining and drying (373 K). And this process of painting was repeated 5 times. Finally, the as-coated specimens were heat-treated at 1573 K for 2 h in argon atmosphere.

Above procedure was repeated 3 times for construction of outer coating (the ZrSiO₄ and aluminosilicate glass coating, namely ZAGC). So far, the final double layer coated C/C composites with SiC and ZAGC were achieved.

Aiming to fabricate the dense coating, the optimal ratio of ZrSiO₄ and aluminosilicate glass powder was screened out with 4 parallel groups. Their ingredients were listed in Table 1.

2.2. Characterization

The X-ray diffraction meter (D-max 2200, Rigaku) was used to identify the crystalline phase of the coatings. The electron microscopies (SEM/TEM, JSM-6700 F/JEM2100 F) were employed to characterize and resolve the microstructures of the double layer coated C/C composites, including inner and outer coatings.

The dual-band emissivity measuring instrument (IR-2 type) was utilized to give the infrared radiation rates of the SiC-coated C/C composites and ZAGC/SiC-coated C/C composites at ambient temperature, respectively. In order to reduce the experimental error caused by the sampling variation and the measurement deviation, nine parallel experiments were carried out for the each group, respectively, that is, 9 sampling and 9 measurements for each group. The sample variances of the SiC-coating and ZAGC/SiC-coating are calculated to evaluate the data reliability, respectively. Then take the average of these measurements to discuss the infrared radiation performance.

The cooling rate was measured with infrared thermometers (TD 1800). The SiC-coated C/C composites and ZAGC/SiC-coated C/C composites are first heated to 1773 K in air, respectively. Subsequently, when the temperature was stable at 1773 K, these samples were taken out from the furnace to cool down from 1773 K to RT naturally. In the meantime, the sample temperature changes with time were recorded at 10-second intervals. And the parallel experiment was repeated 5 times for every sample, and the averages of them were taken to discuss the contribution to oxidation resistance.

2.3. Oxidation tests

The oxidation tests for the double layer coated C/C specimens were carried out in a tube furnace at 1773 K in air. As well, the SiC-coated perform was employed as a contrast to the oxidation kinetics experiment. In the study of high temperature oxidation of coating, usually using the weight change per unit area.

The average weight change per unit area (dotted as $w_{c_{sq,m}}^2$) of the coating material was employed to evaluate their antioxidant capacity. The $w_{c_{sq,m}}^2$ can be calculated as

$$w_{c_{sq,m}}^2 = (m - m_0) / \sum sq_i \quad (1)$$

where m_0 and m are the weights of the specimens before and after oxidation, respectively, and the sq_i represents the surface areas of each surface of the C/C composites.

3. Results and discussion

The macrographs of the four samples are shown in Fig. 2. It is obvious that a large number of cracks and micro-pores emerged on the surface of the coating 1[#], 2[#], and 4[#], as shown in Fig. 2a, b and d, while the surface of the coating 3[#] is smooth without obvious above defects (Fig. 2c). It can be seen that the appropriate ratio of ZrSiO₄ and aluminosilicate glass powder is very important for the formation of dense coating. The present experimental results show that the 4:1 of mass ratio of ZrSiO₄ and aluminosilicate glass powder (the volume ratio is about 2:1) is suitable to form a dense coating. In the latter study, the sample No. 3 was used as the experimental subject.

Referring to the schematic diagram (Fig. 1), the formation mechanism of the double layer coatings is respectively discussed as

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