



Study on ZrSiO₄-aluminosilicate glass coating with high infrared emissivity and anti-oxidation properties



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ARTICLE INFO

Keywords:

Ceramics coating
Microstructure
Aluminosilicate glass
Anti-oxidation

ABSTRACT

A multicomponent coating composed of ZrSiO₄ and aluminosilicate glass was designed and prepared by slurry dipping and subsequent sintering on the surface of SiC substrate to improve the anti-oxidation ability of substrate. At appropriate temperature, the molten aluminosilicate glass carrying the stable ZrSiO₄ particles will spread over the substrate's surface to form a dense and stable coating. Among them, the flowing aluminosilicate glass will fill the crack and micro-pores of ZrSiO₄, which inform a dense coating. Glass phase and ceramic phase could inhibit the propagation of the cracks together. This coating shows high infrared emissivity at broad bands, reaching more than 0.93. Wonderfully, the coating was endowed high oxidation resistance performance at high temperature in the air, which was mainly attributed to the compact and stable structure of coating, coupled with the high thermal expansion coefficient.

1. Introduction

Stemed from excellent thermal conductivity, good thermal shock resistance, and high bearing strength, silicon carbide (SiC) ceramics support material has been widely studied and used in aerospace field, machinery industry, electronics and other fields [1–7]. Many studies have indicated that the poor oxidation resistance of SiC at high temperature oxidizing condition seriously affects their service life, becoming an urgent barrier for using SiC products in above harsh environment. Therefore, improving the high temperature oxidation resistance of SiC has become an important research direction of the researchers [5–7]. Many researches showed that an oxidation resistant coating, which generally has high reflectivity and low conductivity insulation, is a sensible choice for protecting substrate at high temperature [8,9].

Based on the excellent chemical and thermal stability (phase transformation temperature higher than 1973 K), as well low oxygen permeability rate and good erosion resistance, zirconium silicate (ZrSiO₄) has become one of the important material for the production of ceramic glaze layer [10,11]. In addition, it is important that SiC and ZrSiO₄ have similar coefficient of thermal expansion (they are 3.7–4.8 × 10⁻⁶/K, and 4.5 × 10⁻⁶/K, respectively), endowing them a good thermal expansion matching, which can avoid stress cracking phenomenon and improve the oxidation resistance performance. So, the

ZrSiO₄ can be considered as a good candidate for preparing antioxidant coating for SiC to improve the oxidation resistance of SiC at high temperature [8,12,13]. However, the following two factors put forward higher requirements for the preparation of ZrSiO₄ coatings. First, the pure ZrSiO₄ has a high melting point (2523 K), and the second, the pure ZrSiO₄ coating is not dense.

In this paper, as shown in the scheme in Fig. 1a, a composite coating composed of ZrSiO₄ and aluminosilicate glass (namely ZAG) was designed and prepared on the surface of SiC substrate by slurry dipping and subsequent sintering method. Herein, a flow mixture containing liquid aluminosilicate glass and ZrSiO₄ particles is formed at a point below the melting point of the ZrSiO₄ and above the melting point of the aluminosilicate glass. The flow of aluminosilicate glass can diffuse along the surface of SiC substrate and fill the crack and micro-pores of ZrSiO₄, at the same time, forming a dense coating layer. Excitingly, this excellent microstructure endowed the ZAG coating high oxidation protecting performance and high infrared emissivity.

2. Experimental

2.1. Specimens preparation

The ZrSiO₄ and amorphous aluminosilicate glass powders with the mass ratio of 4:1 were mixed and ball milled for 24 h to obtain a mother

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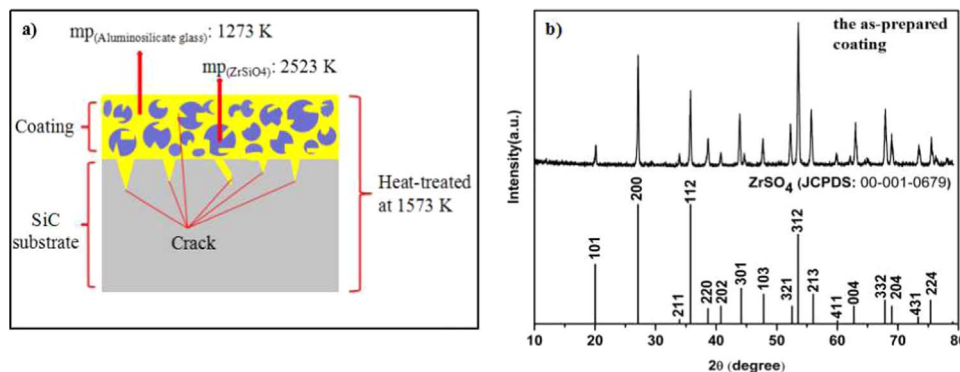


Fig. 1. a) Scheme illustration of the composition and preparation of the ZAG coating, and b) XRD patterns of the ZAG coating.

mixture. Then the mother mixture was added into alcohol to make the slurry. Subsequently, the binder Polyethylene Glycol was added into the slurry with a mass ratio of 1:100 to mother mixture. Finally, the slurry was painted directly onto the surface of SiC substrate, dried, and heat-treated at 1573 K for 2 h in argon atmosphere. Other samples were prepared with different feeding ratios of ZrSiO₄ and aluminosilicate powders in accordance with preceding operation. The thickness of the ZAG coating was controlled intentionally by the paint times.

2.2. Oxidation tests

The isothermal oxidation test was carried out in a tube furnace at 1773 K in air. Weight change of specimens was calculated by:

$$\Delta m = (m - m_0) / m_0 \times 100\% \quad (1)$$

where m_0 and m were the weights of the specimens before and after oxidation, respectively.

2.3. Characterization

The crystalline phase of the coatings was identified by X-ray diffractometer (D-max 2200, Rigaku). The microstructures were characterized by the scanning electron microscopy (SEM, JSM-6700F) and transmission electron microscopy (TEM, JEM-2100F). Infrared radiation rates were obtained by Dual-band emissivity measuring instrument (IR-2 type). Temperature was obtained by Infrared thermometers (TD 1800).

3. Results and discussion

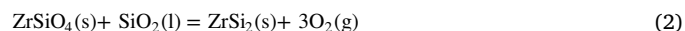
As shown in Fig. 1b, the XRD pattern of obtained coating indicated the existence of ZrSiO₄. Another ingredient aluminosilicate glass was not detected because of its amorphous characteristic. Its existence can be confirmed with HRTEM resolution and X-ray energy-dispersive spectroscopy (EDS) analysis (below section).

In order to explore the optimized ratio of ZrSiO₄ and aluminosilicate glass powder for ZAG, four samples with different mass ratios (ESI, Table S1) were prepared. The photographs of the four samples are shown in Fig. S1. It is shown that a great deal of micro-pores and cracks emerged on the surface of samples 1, 2, and 4, while sample 3 has no obvious flaws. Furthermore, the cross-sectional SEM images showed that the as-prepared ZAG coating with about 150 μm of thickness are closely contacted with SiC substrate (Fig. 2b). There is no obvious crack or gap between the coating and the substrate. So, the feed ratio of ZrSiO₄ and aluminosilicate glass powder was optimized at 4:1.

Fig. 2 showed the SEM and TEM characterizations of the as-prepared ZAG coating, as well the counterparts suffered isothermal oxidation at 1773 K for 10 min. As shown in Fig. 2a, the surface of the as-prepared coating is relatively flat, no obvious crack, and the ZrSiO₄ particles were evenly distributed in the aluminosilicate glass. The SEM

image of the counterpart after anti-oxidation test indicated that the surface of coating is in a shape of erosion, but it is still dense without crack (Fig. 2a'). Furthermore, the cross-sectional SEM photographs also illustrated the excellent anti-erosion ability of ZAG coating (Fig. 2b'). During the stage of anti-oxidation test at 1773 K, the viscosity of aluminosilicate glass reduced, and the mobility of aluminosilicate glass increased compared to the state at 1573 K, which is helpful to fill the micro-pores of ZrSiO₄ rapidly. No holes or cracks emerged in the coating at 1773 K for 10 min. The TEM/HRTEM resolutions in Figs. 2c and c' distinctly showed the stable coexistence of the ZrSiO₄ particle and the aluminosilicate glass, implying the good oxidation resistance of the coating. The crystalline particles can be identified as ZrSiO₄ from the resolution of HRTEM, which were surrounded by amorphous objective. The EDS analysis indicated that these powders can be identified as aluminosilicate (Fig. S2).

The formation mechanism of the dense and stable ZAG coating is discussed as follows (scheme in Fig. 1a). Firstly, the melting point of aluminosilicate glass is below 1273 K, while that of ZrSiO₄ is about 2523 K. When sintering the mixture of ZrSiO₄ and aluminosilicate glass at 1573 K in argon atmosphere, the appropriate amounts of aluminosilicate glass will fill the micro-pores of ZrSiO₄ when it melts. The molten aluminosilicate glass carrying the stable ZrSiO₄ particles will form a dense coating to protect the substrate. Besides, the thermal expansion coefficient of ZrSiO₄ is about $4.5 \times 10^{-6}/K$, and the thermal expansion coefficient of SiC substrate is $3.7-4.8 \times 10^{-6}/K$. So, they have a good thermal expansion matching, which can avoid stress cracking phenomenon and improve the oxidation resistance performance. Finally, a dense coating formed. Moreover, the thermodynamic functions calculation for assumed reaction of ZrSiO₄ and aluminosilicate glass (its main component SiO₂) at 1773 K was carried out as follows:



$$\begin{aligned} &G(\text{ZrSiO}_4) \quad G(\text{SiO}_2) \quad G(\text{ZrSi}_2) \quad G(\text{O}_2) \quad \text{kJ} \cdot \text{mol}^{-1} \\ &-2312.80 \quad -1048.09 \quad -353.77 \quad -346.39. \end{aligned}$$

$$\begin{aligned} \Delta G^0 &= \sum \nu_i G_i = G(\text{ZrSi}_2) + 3G(\text{O}_2) - G(\text{ZrSiO}_4) - G(\text{SiO}_2) \\ &= 1967.95 \text{ kJ} \cdot \text{mol}^{-1} > 0. \end{aligned}$$

Clearly, from the point of view of thermodynamics, this chemical reaction Eq. (2) couldn't take place at 1773 K.

High infrared emissivity of coating can significantly improve the cooling rate [14,15]. Here, the infrared emissivity data of the coating sample at 1–22 μm, 3–5 μm, and 8–14 μm bands are 0.93, 0.93, and 0.94, respectively. For a comparison, the infrared emissivity data of SiC substrate at 1–22 μm, 3–5 μm, and 8–14 μm bands are only 0.77, 0.74, and 0.78, respectively. Fig. 3a shows the temperature change curves of the ZAG-SiC sample and naked SiC substrates after isothermal oxidation from 1773 K to room temperature, indicating the cooling rate of the ZAG-SiC is obviously faster than SiC substrate.

The weight change of the antioxidant material under high temperature is an important evaluation of their antioxidant capacity. Due to the oxygen diffusion coefficient of coating, it may cause a little oxygen

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