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Thermal shock behavior of mixed ytterbium disilicates and ytterbium monosilicates composite environmental barrier coatings

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ABSTRACT

The thermal shock properties of mixed $Yb_2Si_2O_7$ and Yb_2SiO_5 composite environmental barrier coating (EBC) topcoat, with various $Yb_2Si_2O_7$-to-Yb_2SiO_5 ratios, are investigated between room temperature and 1500 °C. The results show that, with more $Yb_2Si_2O_7$ introduced, the mixed $Yb_2Si_2O_7$ and Yb_2SiO_5 composite topcoats exhibit an increasing thermal shock property. This may be attributed to that, while $Yb_2Si_2O_7$ mole fraction increases, the thermal expansion coefficient of the mixed $Yb_2Si_2O_7$ and Yb_2SiO_5 composite ceramics becomes smaller, more matched to substrates and the fracture toughness of the mixed $Yb_2Si_2O_7$ and Yb_2SiO_5 composite ceramics becomes larger, both of which are favoring the improvement of thermal shock resistance.

1. Introduction

To achieve a higher thermal efficiency of aero-engines, an eternal goal in aviation industry, there exists enormous impetus to increase their operating temperature [1]. The traditional nickel based supperalloys, with an upper limit of about 1100 °C, cannot satisfy the requirements of a continuously increasing service temperature of novel aero-engines, which calls for the development of a novel structure material that can withstand much higher temperatures [2,3]. The silicon carbide based ceramic matrix composites (CMCs) are an ideal material to fulfill this task. They exhibit a combination of superior high temperature mechanical properties, excellent oxidation/thermal shock resistances, high reliability, and damage tolerance, all of which are desirable for the application in the hot section components of gas turbines [4–8]. Their major shortcoming is the rapid recession if exposed directly to a high temperature combustion environment with both water vapor and oxygen as the oxidizers [9–11]. By reacting with oxygen alone, a dense silica layer forms and prevents the underlying silicon carbide based CMCs from further oxidizing. However, this dense protective silica layer is reactive with water vapor to form a volatile species, the silicon hydroxide, thus resulting in a lack of environmental durability [12,13]. To prevent the reaction of silicon carbide based CMCs with water vapor, an environmental barrier coating (EBC) is required. EBCs are used as protective coatings for silicon carbide based components in gas turbine engines to inhibit surface reactions with water vapor released by hydrocarbon combustion [14–16].

According to the ytterbia-silica binary phase diagram (Fig. 1), there are two typical kinds of ytterbium silicates with different silica-to-ytterbia ratios, namely ytterbium monosilicate (Yb_2SiO_5) and ytterbium disilicate ($Yb_2Si_2O_7$). Recently, these two types of ytterbium silicates have been proposed as promising EBC topcoat materials due to a combination of desirable properties such as low thermal conductivity [17,18], high temperature stability [19,20] and excellent water vapor corrosion resistance [21,22].

Actually, study on the thermal shock behavior of Yb_2SiO_5 and $Yb_2Si_2O_7$ is beneficial to develop ytterbium silicate as EBC candidates as the thermal shock resistance is a comprehensive indicator to evaluate a coating. The thermal shock property of a material is related to a number of its mechanical and thermophysical properties, such as strength σ, elastic modulus E, thermal conductivity k and thermal expansion coefficient α, etc., and a higher strength and thermal conductivity and lower elastic modulus and thermal expansion coefficient are beneficial to the improvement of thermal shock property [23]. Yb_2SiO_5 has two factors favoring the improvement of thermal shock resistance, i.e. a lower elastic modulus ($E = 149$ GPa [24] in comparison to 168 GPa for $Yb_2Si_2O_7$ [25]) and a higher strength ($\sigma = 215$ MPa [24] in comparison to 159 MPa for $Yb_2Si_2O_7$ [25]); Whereas, $Yb_2Si_2O_7$ has two factors favoring the improvement of thermal shock resistance, i.e. a lower TEC ($\alpha = 4.1 \times 10^{-6}\,°C^{-1}$ [26] in comparison to 7–8 $\times 10^{-6}\,°C^{-1}$ for Yb_2SiO_5 [27]) and a higher thermal conductivity ($k = 1.8$ W/m·K [28] in comparison to 1.5 W/m·K for Yb_2SiO_5 [24] at 1200 K). As $Yb_2Si_2O_7$ and Yb_2SiO_5 respectively have two factors

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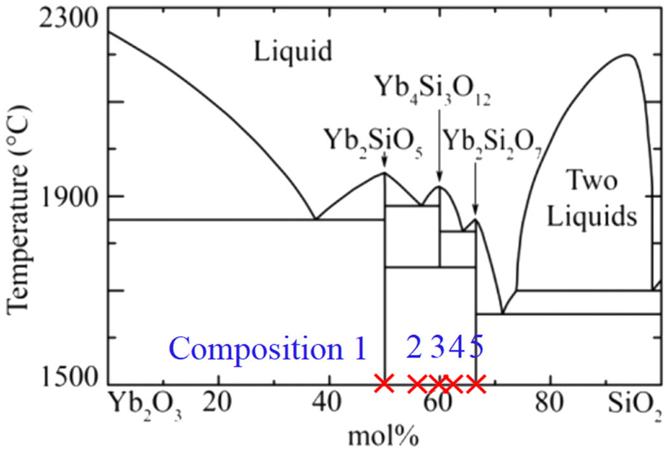

Fig. 1. Ytterbia-silica binary phase diagram. Five selected compositions, labeled as Composition 1 to 5, are marked accordingly.

desirable for thermal shock resistance, this indicates an intermediate composition, i.e. a mixture of Yb_2SiO_5 and $Yb_2Si_2O_7$, could probably offer a maximized thermal shock property. Therefore, the thermal shock behavior of EBC topcoats composing of mixtures of Yb_2SiO_5 and $Yb_2Si_2O_7$ is also worth further investigation.

In this study, we carry out the thermal shock test of Yb_2SiO_5, $Yb_2Si_2O_7$ and their mixtures EBC topcoats fabricated from an air plasma spray (APS) method. In addition, considering a relatively low temperature capability of Si bond coat (1414 °C), a silicon carbide bond coat is developed to replace the traditional silicon one. Five selected compositions (including end members) as shown in Fig. 1 of mixed Yb_2SiO_5 and $Yb_2Si_2O_7$ composite topcoats are deposited directly on a SiC bond coat made by a chemical vapor deposition (CVD) route and their thermal shock cycles are tested between room temperature and 1500 °C.

2. Experiments

Fig. 1 shows the ytterbia-silica binary phase diagram [29] with five selected compositions Yb_2SiO_5, $3Yb_2Si_2O_7$•$4Yb_2SiO_5$, $3Yb_2Si_2O_7$•$2Yb_2SiO_5$, $3Yb_2Si_2O_7$•Yb_2SiO_5 and $Yb_2Si_2O_7$. The raw powders, $Yb_2Si_2O_7$ and Yb_2SiO_5, (both of which have a purity of 99.9% and are purchased from Kai-Star Electro-Optic Materials, Wuxi) were mixed by ball milling (24 h, 300 r.p.m.) with zirconia media in alcohol in different molar ratios as shown in Fig. 1. They were then granulated by spray drying and then passed through 200 and 400 meshes to obtain powders suitable for plasma spraying. Topcoats of all ytterbium silicates were deposited on SiC bond coat by atmospheric plasma spraying device (APS-3000, AVIC Manufacturing Technology Institution, China) using processing parameters shown in Table 1. The silicon carbide bond coat was deposited on substrate from CVD at 1050 °C for 100 h by employing methyl trichlorosilane (MTS) as precursor. All substrates are 30 mm × 30 mm SiC_f/SiC CMCs with a thickness of 5 mm made from the precursor infiltration pyrolysis (PIP) route.

The prepared samples were then stabilization annealed (to transform metastable phases) at 1400 °C in air for 20 h at a heating rate of 8 °C/min and a cooling rate of 10 °C/min. The thermal shock properties of the mixed Yb_2SiO_5 and $Yb_2Si_2O_7$ topcoats with a CVD SiC bond coat on SiC_f/SiC CMCs were tested on a thermal shock testing rig. The test was conducted between room temperature and 1500 °C. For each cycle, samples were first placed in furnace (KF1700, Boyuntong, China), maintained at 1500 °C for 15 min and then cooled to room temperature within 5 min by a cooling air jet. The test was terminated when partial spallation of the top layer (more than 5% area) was detected.

Table 1
Air plasma spray parameters employed for current mixed ytterbium disilicate and ytterbium monosilicate EBC topcoat.

APS layer	Arc current (A)	Primary Ar flow (l/min)	Secondary H_2 flow (l/min)	Carrier Ar flow (l/min)	Powder flow (r/min)	Deposition power (kW)
EBC topcoat	300	36	5.5	2.6	30	18

The phase compositions were examined by X-ray diffraction analysis (XRD, Bruker D8 Advanced, Cu Kα radiation). Microstructures were analyzed using scanning electron microscope (SEM, Philips XL 30) by the backscattered electron (BSE) mode on polished cross sections and surfaces of EBCs. The average porosity of EBC top layer were evaluated from SEM images by linear intercept measurement [30]. For each composition, at least 5 SEM images were measured.

In order to identify the thermal expansion coefficients (TECs) and fracture toughness of corresponding compositions, which are closely related to the thermal shock behavior of EBC topcoats currently investigated, dense ytterbium silicate ceramic compacts were prepared by a spark plasma sintering (SPS) method at 1500 °C under a pressure of 40 MPa for 10 min, with a heating and cooling rate at 100 °C/min and 300 °C/min respectively. Thermal expansion coefficients were obtained from temperature-dependent changes on specimen lengths from room temperature to 1400 °C in air using a vertical high-temperature optical dilatometer (ODHT, Modena, Italy). The fracture toughness of samples was measured by three-point bending of single-edge-notched beams (SENB) on an electronic universal testing machine (WDW-200, Hengruijin, China) with a loading rate of 0.05 mm/min. For each composition, at least 5 samples were tested. The fracture toughness is calculated according to the following equations [31]:

$$K_{IC} = \frac{3PL}{2B\omega^2} a^{1/2}\ Y \tag{1}$$

$$Y = 1.93 - 3.07\frac{a}{\omega} + 13.6\left(\frac{a}{\omega}\right)^2 - 23.98\left(\frac{a}{\omega}\right)^3 + 25.22\left(\frac{a}{\omega}\right)^4 \tag{2}$$

where P is the maximum load, L, B, ω are the span, width and height of samples respectively and a is the depth of the notch.

3. Results and discussions

3.1. Phase compositions and microstructures of mixed $Yb_2Si_2O_7$ and Yb_2SiO_5 composite topcoats

Fig. 2 shows the XRD patterns of EBC topcoat surface with various compositions before and after thermal annealing at 1400 °C for 20 h. There are no additional peaks found except for those peaks of $Yb_2Si_2O_7$ and Yb_2SiO_5 in the annealed EBC topcoat, demonstrating that no other phases have been formed. It also shows that $Yb_2Si_2O_7$ and Yb_2SiO_5 have different monoclinic structures. With an increase of the $Yb_2Si_2O_7$ mole fraction in the mixed $Yb_2Si_2O_7$ and Yb_2SiO_5 composite topcoats, the peaks of the I2/a $Yb_2Si_2O_7$ monoclinic structure are getting stronger.

Fig. 3 exhibits the backscattered electron images of polished cross sections of $3Yb_2Si_2O_7$•$4Yb_2SiO_5$ topcoat and $3Yb_2Si_2O_7$•Yb_2SiO_5 topcoat respectively. Plenty of pores can be found on the cross section of top layers, which is a typical feature of a coating deposited by the APS technique. As shown in Table 2, the current mixed $Yb_2Si_2O_7$ and Yb_2SiO_5 composite topcoats have almost the same porosity level, around 7%. The existence of pores in EBC topcoat has the following benefits. First, the pores provide spaces for expansion and thus reduce the TEC of the topcoat, which helps to reduce the thermal stresses caused by TEC mismatch between topcoats and substrates [32]. Second, the porosity reduces the elastic modulus, imparting the topcoat a better stress tolerant property. Therefore, the existence of pores in EBC topcoats helps to improve thermal shock property. As the porosity in topcoats of all compositions current investigated has the similar level, the porosity effects on thermal shock property can be comfortably