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Comparative study on high-temperature performance and thermal shock behavior of plasma-sprayed Yb_2SiO_5 and $Yb_2Si_2O_7$ coatings

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ABSTRACT

In this work, differences of microstructure and thermo-mechanical properties between air plasma sprayed Yb_2SiO_5 and $Yb_2Si_2O_7$ coatings were investigated. Thermal shock behavior between Yb_2SiO_5/Si and $Yb_2Si_2O_7$/Si EBC systems on SiC substrates were compared. Results show that the $Yb_2Si_2O_7$ coating exhibited significant microstructure evolution compared to Yb_2SiO_5 during high temperature thermal aging, which is attributed to the different activation energy of grain growth between the Yb_2SiO_5 (366 kJ/mol) and $Yb_2Si_2O_7$ (219 kJ/mol) coatings. The $Yb_2Si_2O_7$ coating exhibited lower CTEs, elastic modulus and hardness, however, higher thermal conductivity compared to the Yb_2SiO_5 coating. After thermal shock test for 40 cycles, partial delamination occurred in the Yb_2SiO_5/Si system, while the $Yb_2Si_2O_7$/Si system remained integral. The failure mechanism of these rare-earth silicates EBC systems were analyzed in detail. The results of this study may provide a reference for EBCs design and applications.

1. Introduction

Silicon-based ceramics, such as SiC and Si_3N_4, exhibit low density, superior fracture toughness, high strength and reliability at elevated temperatures, showing potential applications in gas turbine engines [1, 2]. However, they are prone to oxidation and corrosion when exposed to combustion environments containing water vapor, therefore, lacking environmental durability [3, 4]. Environmental barrier coating (EBC) systems have been developed to prevent silicon-based ceramics from corrosion by water vapor in combustion environments [5–7].

Recent works have demonstrated that rare-earth silicates, especially Yb_2SiO_5 and $Yb_2Si_2O_7$, are promising candidates of EBC materials [8–11]. Some efforts have been put to investigate the mechanical and thermal properties of bulk Yb_2SiO_5 and $Yb_2Si_2O_7$. The DOE-National Energy Technology Laboratory [12] reported that the thermal conductivity of Yb_2SiO_5 was 1.3–1.4 W/(m·K) at 200 °C–1400 °C, and the coefficients of thermal expansion (CTE) of Yb_2SiO_5 was $7.0–8.0 \times 10^{-6} K^{-1}$. Xiang et al. [13] reported that the intrinsic thermal conductivity of Yb_2SiO_5 decreased as 893.82/T and approached a minimum value (0.74 W/(m·K)) at high temperatures, and the CTE was $5.24 \times 10^{-6} K^{-1}$, theoretically. Zhou et al. [14] found that the thermal conductivities were almost inverse proportional to temperature and were extrapolated to be 4.6 and 2.0 W/(m·K), respectively, at 300 and 1400 K, and the linear CTE was $3.7–4.5 \times 10^{-6} K^{-1}$ (800–1600 K). Their results show that Yb_2SiO_5 and $Yb_2Si_2O_7$ are potential candidates of EBC materials.

Microstructure and properties of coatings are closely related to fabrication processes and quite different from bulk materials. It is well known that plasma sprayed coatings have unique microstructure characteristics, like laminar structure, interfaces between splats, pores and microcracks, which are of great influence on their performance, besides material itself. Lee et al. [8] sprayed Yb_2SiO_5/mullite + SAS/Si and Yb_2SiO_5/mullite/Si systems on SiC and Si_3N_4 substrates, respectively. Richards et al. [10, 11, 15–17] fabricated Yb_2SiO_5/mullite/Si, $Yb_2Si_2O_7$/mullite/Si and $Yb_2Si_2O_7$/Si systems on SiC substrates, respectively, using air plasma spray (APS) approach and thermal cycling tests of these systems were conducted. Some of our previous works have found that the plasma sprayed Yb_2SiO_5 and $Yb_2Si_2O_7$ coatings exhibited relative dense structures with defects such as pores, interfaces and microcracks. The amorphization and dissociation phenomena happened in the deposition processes of both coatings. In addition, the as-sprayed Yb_2SiO_5 coatings undergo microstructure changes when under high-temperature environments. And these changes can influence the thermo-mechanical properties of the coating, such as thermal

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conductivity, CTEs, Hv and elastic modulus [18, 19]. It is meaningful to investigate microstructure evolution and its influence on properties during thermal aging, which can help to increase the knowledge about the specific characteristics of the Yb_2SiO_5 and $Yb_2Si_2O_7$ coatings. Furthermore, the response of these coatings to thermal shock behavior would be enriched.

The objective of this preliminary study is to begin an investigation about the differences of microstructure and thermo-mechanical properties between the Yb_2SiO_5 and $Yb_2Si_2O_7$ coatings during thermal aging. Plasma-sprayed Yb_2SiO_5 and $Yb_2Si_2O_7$ coatings were thermally aged at high temperatures (1300–1500 °C) and their microstructure, mechanical and thermal properties were characterized. Yb_2SiO_5/Si and $Yb_2Si_2O_7$/Si systems were sprayed onto SiC substrates as they were subjected to thermal shock behaviors between 1400 °C and 25 °C, and the failure mechanisms of the EBC systems were explored. This study may provide a reference body for the differences in microstructure and properties between monosilicates and disilicates, which is essential to EBCs design and application.

2. Experimental procedure

2.1. Sample preparation

Yb_2SiO_5 and $Yb_2Si_2O_7$ powders were synthesized by solid-state reaction with Yb_2O_3 and SiO_2 as the starting materials. Details for powder synthesis can be found elsewhere [18, 19]. An air plasma spray system (A-2000, Sulzer Metco AG, Switzerland) with a F4-MB torch was then used to deposit the Yb_2SiO_5 and $Yb_2Si_2O_7$ coatings using the operating parameters listed in Table 1 onto water cooled aluminum substrates (128 mm × 84 mm × 2 mm) and on SiC substrates (35 mm × 5 mm × 4 mm) with a Si bond layer to prepare coatings for thermal shock resistance measurements. Free-standing samples were mechanically removed from the aluminum substrates. The free-standing coating samples were thermal aged at 1300 °C, 1400 °C and 1500 °C for 50 h, respectively, in a box furnace in air to investigate their microstructure and thermo-mechanical properties.

2.2. Thermal shock resistance measurements

Thermal shock tests were performed by first heating the samples in a tube furnace, and then quickly dropping into the cooling water (about 25 °C). It should be pointed out that the thermal shock tests were performed with the as-sprayed samples, which contained amorphous phase. Each cycle of this thermal shock test included the following steps: (1) heating up to 1400 °C and maintaining 20 min in a tube furnace, (2) rapid immersion in a water tank until a temperature of 25 °C reached, (3) imaging of the surface morphology. The thermal cycling process was repeated until peeling off occurred.

2.3. Specimens characterization

Phase compositions of the as-sprayed, thermal-aged and thermal shocked samples were identified by X-ray diffraction (XRD) using a Rigaku D/Max2550 diffractometer with Cu*Ka* (λ = 0.154056 nm) radiation. High temperature XRD (HT-XRD) was recorded at 1000 °C, 1200 °C and 1400 °C using Bruker D8 Advance XRD. All samples, including the as-sprayed, thermal-aged and thermal shocked samples, were cut to prepare the cross sections and then ground and finely polished using routine metallographic methods. The surface, fracture and polished cross sections of coatings were characterized using a scanning electron microscopy system (SEM, S-4800, Hitachi, Japan and Magellan 400, FEI, USA) equipped with energy-dispersive X-ray spectroscopy (EDS) and electron backscatter diffraction (EBSD) attachments (INCA SERIES, Oxford Instrument, UK).

Coating porosity was evaluated by an image analysis method. Five cross-sectional back-scattered electron images (BSEs) with 1000 × magnification were taken randomly in each sample for image analysis. The grain size of the thermal-aged samples was determined from SEM images using a rectangular procedure [20]. The average grain size (D) is given by

$$D = \sqrt{\frac{4A}{\pi(n_i + n_0/2)}} \tag{1}$$

where A is the area of rectangular, n_i and n_0 are the grain numbers in the rectangular and on the rectangular boundary, respectively.

Vickers hardness (Hv) was measured on the polished cross-sections of both the as-sprayed and thermal-aged coatings using an Instron Wilson-Wolpert Tukon 2100 B hardness tester under a load of 300 g with a dwell time of 15 s. The elastic modulus (E) was measured on the polished cross-sections by nano-indentation technique, using a G-200 nanoindenter (Agilent Technologies, Oak, Ridge, USA) equipped with a Berkovich indenter. The reported data corresponded to an average value of ten indents. They were representative of the mean values at a penetration depth of about 1000 nm.

Thermal expansion properties of the coatings were determined by measuring the temperature-dependent change of length of the samples using a Netzsch 402C high-temperature dilatometer. The as-sprayed coating samples were taken a heating process from 25 °C to 1400 °C before the measurement for the purpose of eliminating the influence of amorphous phase. The no-amorphous coating samples (20 mm × 5 mm × 0.5 mm) were used for coefficient of thermal expansion (CTE) measurements, which were done in flowing air atmosphere from 25 °C to 1400 °C at a heating rate of 5 °C·min^{-1}. The CTE was calculated by the formula:

$$CTE = \frac{\Delta L}{L \cdot \Delta T} \tag{2}$$

where ΔL and ΔT are the changes of sample length and temperature, respectively, L is the length of the sample at room temperature. Thermal expansion rate was calculated by ΔL/L.

Thermal diffusivity (α) of both coatings before and after thermal aging was measured using a laser flash method, as a function of temperature in the range from 200 °C to 1200 °C at intervals of 100 °C. And the specimen with 11 mm in diameter and 0.8 mm in thickness was used for thermal diffusivity measurements. The surfaces of specimens were coated with a thin layer of graphite in order to increase the absorption and emission efficiency of the sample surfaces. The specific heat capacity (C_p) as a function of temperature was calculated by the Neumann-Kopp rule with the heat capacity data of SiO_2 and Yb_2O_3 [21, 22]. Moreover, the specific heat capacities of Yb_2SiO_5 and $Yb_2Si_2O_7$ coatings thermal-aged at 1300–1500 °C were measured using diamond DSC in the temperature range from 25 °C to 500 °C in order to fitting the calculated values. The thermal conductivity κ values were calculated by Eq. (3) with the heat capacity (C_p), thermal diffusivity (α) and density (ρ) which was measured by the Archimedes method.

$$\kappa = C_p \times \alpha \times \rho \tag{3}$$

Table 1
Operating parameters used for air plasma spray.

APS layer	Power (kW)	Primary Ar (slm)	Secondary H_2 (slm)	Carrier Ar (slm)	Spray distance (mm)
Yb_2SiO_5	43	38	12	3	120
$Yb_2Si_2O_7$	43	38	12	3	120
Si	32	42	8	4	120