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Phase stability of plasma sprayed YAG–YSZ composite beads/coatings at high temperature

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Abstract

Beads and coatings of YAG–YSZ composite ceramic were prepared by plasma spray. YAG was applied as an additive in the hope of improving the phase stability and oxygen impermeability of YSZ. To achieve this aim, some basic research about crystallization behavior and chemical compatibility of the composite were carried out. Plasma sprayed YAG tended to form amorphous state. With the different content of YAG in the composite, crystallization of YAG and YSZ was delayed by each other to different extent due to the barrier effect of heavy atoms. The relative low melting point of YAG led to dense coatings without obvious splat structure and further distinctive vertical cracks during crystallization within the coatings. The cracks became less severe when YAG content was lower. With the addition of YAG, the development of monoclinic ZrO_2 was suppressed while Y-rich phases were promoted at higher temperature.

Keywords: YSZ; Composite ceramic; Plasma spray; Crystallization; Chemical compatibility

1. Introduction

8 wt.% Y_2O_3 stabilized ZrO_2 (8YSZ) is the classical ceramic which is most widely applied as the topcoat of thermal barrier coatings in turbine engines. This material is preferred for its low thermal conductivity, high coefficient of thermal expansion (CTE) and high melting temperature. With the progress in aero-engine technology, the tendency to reach higher efficiency for turbines leads to increasing temperature in combustion chamber.[1–3] However, 8YSZ shows obvious shortcomings above 1473 K with long-term service under thermal cycling: phase transformation and high oxygen ion conductivity.[4–6] Metastable tetragonal 8YSZ (t' phase) gradually transforms to cubic phase (c) with ~14 wt.% Y_2O_3 and yttria-poor tetragonal phase (t) with ~4 wt.% Y_2O_3 in service condition. The latter finally transforms to monoclinic phase (m) at room temperature after a period of time. The phase transformation is accompanied by ~4% volume expansion. High oxygen ion conductivity at high temperature can accelerate the formation of thermally grown oxide (TGO) on metallic bond coat. All of the above are important factors for coating failure.

In recent years, reasearchers focus on new candidates for ceramic topcoat, such as zirconates[7–10] and aluminates. Materials with complex structure based on alumina, such as $Y_3Al_5O_{12}$ (YAG) and $LaMgAl_{11}O_{19}$ (LMA), have received many attentions.[11–16] YAG is superior to traditional Al_2O_3 as an oxygen diffusion barrier for its high phase stability, good mechanical property and acceptable thermal conductivity. Nevertheless, the CTE of YAG is lower compared with 8YSZ (~2000 °C and ~2700 °C, $\sim 9.5 \times 10^{-6}\,K^{-1}$ and $\sim 10.7 \times 10^{-6}\,K^{-1}$, respectively). Besides, YAG and LMA tend to form amorphous state with even lower CTEs because of their complex structure and fast cooling rate of plasma spray process.[17–19]

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Table 1
Parameters for spraying process.

Spraying parameters	Measurements
Current/A	600
Voltage/V	75
Flow rate of plasma gas (Ar, H_2)/L min^{-1}	46, 8
Spray distance/mm	100
Powder feed rate/g min^{-1}	25
Gun traverse speed/mm s^{-1}	1000

Su et al. and Ren et al. reported the application of YAG as an oxygen barrier interlayer between 8YSZ topcoat and metallic bond coat.[20,21] YAG suppressed the oxidation of NiCoCrAlY bond coat and the phase stability of 8YSZ near 8YSZ-YAG interface seemed to be improved. Chen et al. reported the performance of LMA-8YSZ composite ceramic for thermal barrier coatings.[22] The failure mode which differs from pure 8YSZ coatings was discovered after thermal cycling. In this paper, the study about the effect of YAG on the crystallization behavior as well as the chemical compatibility of YAG–YSZ composite are significant issues that can help in the successful design and understanding of these complex coatings. For thermally sprayed coatings, preferred orientation usually occurs during crystallization due to piling up of molten and re-solidified particles, which can result in biased data. Therefore, beads and coatings are comparatively studied to avoid preferred orientation and the interference of the substrate. This basic research might provide some useful information for possible application of YAG on thermal barrier coatings.

2. Experimental procedure

The powder of 8YSZ was 204NS purchased from Sulzer Metco, whose composition was designated as S_0. YAG was synthesized by solid-state reaction at high temperature. The composition of YAG was designated as S_1. Raw materials $Al(OH)_3$ and Y_2O_3 were purchased from Tangshan Huatai Functional Ceramic Materials Co., Ltd. and Guangdong Chenghai Chemicals Co., Ltd., respectively. The raw materials were calcined at 1000 °C for 2 h to decompose hydroxides and carbonates. They were mixed in stoichiometric ratio by ball milling and heated at 1500 °C for 6 h to synthesis YAG. Then the powders of YAG and 8YSZ were mixed by wet milling with zirconia balls for 12 h. The molar ratio of YAG to 8YSZ in the composites was 1:1 and 1:6. These two compositions were designated as S_2 and S_3, respectively. The milled powders were spray-dried as described in former research.[17] The free-flowing powders with particle size of 20–120 μm in diameter were sieved for plasma spray.

The powders were sprayed by an atmospheric plasma spray unit (Unicoat, F4 spraying gun, Sulzer Metco) with Ar–H_2 as plasma gas. The spraying parameters were listed in Table 1. Powders were melted in plasma jet forming beads and collected by empty bottles, which were ~2 m away from the spraying gun. The powders were also deposited onto smooth aluminum plate forming coatings with a thickness of ~400 μm. The aluminum substrate was simply pre-heated by the plasma jet. The heating mode was nearly the same as the spraying mode except that the heating time was 1/10 of the spraying time and no powder was supplied. The coatings can be easily peeled off from the substrate because of the mismatch of thermal expansion. Pure YAG and 8YSZ beads and coatings were also prepared as references.

The thermal properties of as-sprayed coatings were analyzed by a differential scanning calorimeter (DSC, STA 449F3, Netzsch) with a heating rate of 10 °C/min in air atmosphere. The beads and coatings were also thermally treated in a tube furnace at 920 °C, 1050 °C and 1200 °C for 0.5 h separately to study the crystallization behavior. Besides, the coatings were heated at 1200 °C for 12 h to achieve complete crystallization. An X-ray diffractometer (XRD, D8 Advanced, Bruker, Cu-Kα radiation) was employed for phase identification with a scanning rate of 8°/min and a step width of 0.04°. The microstructure and composition of the samples were examined by an environmental scanning electron microscope (SEM, XL-30 FEG, FEI) equipped with an electron diffraction spectrometer (EDS). Backscattered electron images (BSE) were also obtained from this microscope. The thermal expansion behavior of some as-sprayed coatings (with the dimensions of 25 mm × 4 mm × 0.4 mm) was measured by a high-temperature dilatometer (402C, Netzsch).

3. Results

3.1. Composition

The composition of as-sprayed coatings is listed in Table 2. The results are average values obtained from 5 times of EDS test for as-deposited coatings. Large areas were selected for EDS test to reduce the influence of inhomogeneity. YAG content is low in S_2 and 8YSZ content is low in S_3, compared with the ideal ratio. S_1 and S_2 are rich in Y while Y is nearly balanced in S_3. The composition of S_0 is close to the stoichiometric ratio.

3.2. Thermal analysis

DSC results are shown in Fig. 1. No obvious heat release is detected from 700 °C to 1200 °C for S_0. S_1 shows a sharp exothermic peak at 924 °C related to YAG crystallization. Peaks for S_2 locate at 928 °C, 943 °C and 1048 °C. A very small peak at 926 °C and a larger peak at 940 °C are observed for S_3. The DSC results provide useful information for understanding the crystallization process. Thermal treatment in furnace was then carried out at 920 °C, 1050 °C and 1200 °C.

3.3. Morphology of beads and coatings

Fig. 2 shows the plasma sprayed beads before and after thermal treatment for 0.5 h at different temperatures. YSZ should appear brighter than YAG in BSE image since its average atomic mass is larger. For as-sprayed beads, S_1 and S_2 show smooth and homogeneous spherical surface. In comparison, some white spots, i.e. primary crystals of YSZ, emerge on the surface of S_3. After heating at 920 °C for 0.5 h, the surface of S_1 shows pores