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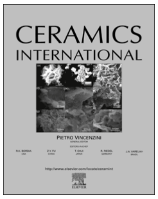

Microstructure and thermal properties of nanostructured gadolinia doped yttria-stabilized zirconia thermal barrier coatings produced by air plasma spraying

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ABSTRACT

In this article, the nanostructured 2 mol% Gd_2O_3-4.5 mol% Y_2O_3-ZrO_2(2GdYSZ) coating was developed by the atmospheric plasma spraying technique. And the microstructure and thermal properties of plasma-sprayed 2GdYSZ coating were investigated. The result from the investigation indicates that the as-sprayed coating is characterized by typical microstructure consisting of melted zones, nano-zones, splats, nano-pores, high-volume spheroidal pores and micro-cracks. The 2GdYSZ coating shows a lower resistance to destabilization of the metastable tetragonal (t′) phase compared to the yttria stabilized zirconia(YSZ). The thermal diffusivity and thermal conductivity of the nano-2GdYSZ coating at room temperature are 0.431 $mm^2\ s^{-1}$ and 1.042 W/m K, respectively. Addition of gadolinia to the nano-YSZ can significantly reduce the thermal conductivity compared to the nano-YSZ and the conventional YSZ. The reduction is mainly attributed to the synergetic effect of gadolinia doping along with nanostructure.

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1. Introduction

Ceramic coatings, deposited onto metallic components, are extensively used to combine the structural integrity of metals with wear-resistance, corrosion resistance and heat insulation property of ceramics [1–5]. Thermal barrier coatings (TBC) should be included in this category as they are widely used in the gas turbine engine environment to increase the turbine efficiency and service life of engine components [6]. Up to now, plasma sprayed (3.5–4.5) mol% yttria-stabilized zirconia (YSZ) is one of the most commonly used TBC because of its low thermal conductivity and phase stability at operating temperatures under 1200 °C. However, at higher temperatures the metastable tetragonal (t′) phase undergoes a partitioning to t and c phase, with the t phase transforming to m phase on cooling. The phase transformation could lead to a significant change in the volume of the coatings and possibly result in disastrous flaking of TBCs [7–9].

Due to the limitations of YSZ, alternatives have been searched for advanced TBC applications. Nano-TBCs are of interest because of their lower thermal conductivity, improved toughness and ductility compared to that of coarser-grained ceramics [10–12]. Trivalent oxides, the addition of which can significantly reduce the thermal conductivity because of the generation of oxygen vacancies, are widely investigated recently. A. Rauf and coworkers have shown that the addition of La_2O_3 in nano-YSZ decreases the thermal conductivity of YSZ as a result of the formation of oxygen vacancies and nanostructure [13]. Similarly, Jin at el. [14] reported that the addition of TiO_2 to nano-YSZ can improve comprehensive performance of YSZ. Jang et al. [15] found that the thermal conductivity of sintered ZrO_2-4 mol% Gd_2O_3 decreased with increasing Gd_2O_3 addition. Ji et al. [16] calculated the lattice distortion and bond population of ZrO_2 doped with rare earth elements by the first principles. The result indicated that Gd-O has the small bond population and weak bond covalent among the rare earth oxides. Weak crystal lattice vibration will occur due to weak bond covalent, which leads to small thermal conductivity. However, the thermal properties of the nano-GdYSZ coatings have not been reported so far.

In the present paper, microstructure and thermal properties of nanostructured gadolinia doped yttria-stabilized zirconia thermal barrier coatings by air plasma spraying are investigated. The combined effect of gadolinia doping along with nanostructure on the thermal properties of YSZ coating has been discussed.

2. Experimental procedures

2.1. Preparation of nanostructured 2GdYSZ coatings

The nano-sized particles composed of 2 mol% gadolinia and 4.5 mol% yttria stabilized zirconia (2GdYSZ) were agglomerated to

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Table 1
Parameters of plasma-spraying

Power (kW)	36
Current (A)	600
Voltage (V)	60
Primary gas (Ar) (slpm)[a]	60
Secondary gas (H_2) (slpm)[a]	15
Carrier gas (Ar) (slpm)[a]	4.5
Spraying distance (mm)	80
Feed rate (g min^{-1})	25

[a] splm: standard liters per minute.

a size of ~30–100 μm for plasma spraying by the spray-drying technique. Then the powders were subjected to the drying treatment at temperature of 150 °C for 1 h.

Ni based super alloy DZ-125 was used as a substrate. The substrates were cut into coupons of 13 mm diameter and 2 mm thickness. In order to improve the adherence of the coatings, these coupons were grit-blasted to obtain a sharp-peaked surface roughness average of 4–5 μm. The powders were plasma sprayed onto the substrates and then removed from the alloy. The coatings detached from the substrate were ~1 mm thick. The plasma-spraying parameters are provided in Table 1.

2.2. Phase composition and phase stability

The stability of the metastable t′ phase in the as-sprayed 2GdYSZ coatings was investigated by heating the samples in a high-purity ZrO_2 crucible to 1400 °C, and holding for 10, 20, 40, 60 and 80 h, after which they were cooled to room temperature. Different samples were used for each run and the annealing temperature was chosen to produce significant transformation.

The phases present in the powder and the as-sprayed 2GdYSZ coatings before and after annealing were identified by D/max 2200pc X-ray diffractometer (Cu Ka radiation; Rigaku, Tokyo, Japan). The samples were scanned in the range of $2\theta = 10–90°$, with speed of 6° per minute. The {111} and {400} peaks, in the range of $2\theta = 27.5–32.5°$ and 72–75°, were scanned at 0.03° per minute.

2.3. Microstructure analysis

The microstructure, morphology of the nanostructured coatings were studied by S-3500 scanning electron microscope which are equipped with EDS (SEM, Hitachi, Tokyo, Japan) and transmission electron microscope (TEM). The mesopore size distribution was tested by Autosorb-iQ2.

2.4. Thermal diffusivity measurement

Thermal diffusivity of standalone 2GdYSZ was measured by the laser flash technique (LFA427, NET-ZSCH). Prior to the thermal diffusivity measurements, a carbon film was deposited on the coating samples. To evaluate the thermal diffusivity, the solution proposed originally by the following relationship [13].

$$\alpha = 0.\ 1388\frac{L^2}{t_{1/2}} \tag{1}$$

where L is the thickness of the sample (cm), and $t_{1/2}$ is the half time (s) that required for the rear surface to reach half the maximum temperature rise. Three values were taken at each temperature and the average of these values was used.

The thermal conductivity of each sample was calculated using the following equation:

$$\kappa\,(T) \;=\; \alpha(T)\,\rho C_{\mathrm{p}} \tag{2}$$

where κ stands for thermal conductivity, α is thermal diffusivity, C_p is heat capacity, and the density ρ of the coating was measured using Archimedes' principle. Density was assumed to be constant at all temperatures.

3. Results and discussion

3.1. Microstructure of as-sprayed coating

The XRD patterns of initial 2GdYSZ powder and as-sprayed 2GdYSZ coating are displayed in Fig. 1. The pattern of the as-sprayed 2GdYSZ coating, when compared to that of initial 2GdYSZ powder, displays the absence of Gd_2O_3 peaks. The result reveals that Gd^{3+} is completely in solid solution with ZrO_2. In {111} region ($2\theta = 27–33°$) and {400} region ($2\theta = 70–76°$), no m and c peaks are found in the pattern for the as-sprayed coating. In {400} region ($2\theta = 70–76°$), the pattern shows double peaks, t′(004) and t′(400), which are attributable to the t′ phase. The formation of non-equilibrium t′ phase in the as-sprayed 2GdYSZ coating is mainly ascribed to quenching of droplet after impacting on the substrate during the plasma spraying process.

The average grain size of the 2GdYSZ coating can be evaluated by using the following Scherrer equation [17]:

$$B_p(2\theta) = \frac{0.9\lambda}{D\cos\theta} \tag{3}$$

where D is the average dimension of the crystallite, $B_p(2\theta)$ is the line broadening measured at the half maximum intensity (FWHM), and λ(0.154 nm) and θ denote the wavelength of the X-rays and Bragg diffraction angle, respectively.

Instrumental broadening in the measurement of peak broadening was taken into consideration and Gaussian correction was applied by comparing the widths at half maximum intensity (FWHM) of X-ray reflection between the sample and the single crystalline Si standard. Then the true crystal broadening is obtained via:

$$B_p^2(2\theta) = B_h^2(2\theta) - B_f^2(2\theta) \tag{4}$$

where $B_p(2\theta)$ is the true half maximum width, $B_h(2\theta)$ and $B_f(2\theta)$

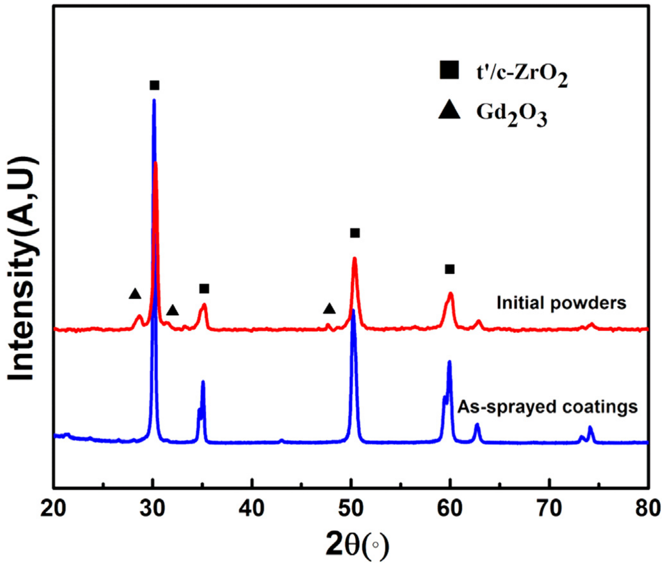

Fig. 1. XRD patterns of initial 2GdYSZ powders and as-sprayed 2GdYSZ coating.

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