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Dependence of microstructure and thermal conductivity of EB-PVD thermal barrier coatings on the substrate rotation speed

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Abstract

EB-PVD TBCs were deposited at nearly the same process parameters but substrate rotation speed to investigate the effect of rotation adjustment on microstructure and thermal conductivity. The microstructure was checked by scanning electron microscopy (SEM) and the thermal conductivity was calculated on the basis of determination of thermal diffusivity by laser flash method. The results showed the stationary deposition formed most dense coating and increasing rotation speed resulted in wider columnar grains and enlarged shadowing effect which meant much more volume of pores. The density and thermal conductivity at room temperature both decreased with increasing rotation speed. The lowest experimental value, 1.37 W/mK, was obtained for a 171μm thick free-standing coating, when the rotation speed was 20 rpm in this study.

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1. Introduction

During the past decades, TBCs had drawn more and more scientific and technical interest and meanwhile got more than ever employed in the hot section of gas turbine engines for protection against extreme service conditions [1, 2]. The current state-of-art material for TBCs was 6~8wt% YSZ and it was well-established the microstructure of TBCs could vary enormously depending on their manufacturing method of which was commonly described as plasma-spray (PS) or EB-PVD. TBCs produced by PS took on a layered structure which could effectively restrain the heat transfer inside of the coatings, while that by EB-PVD presented columnar microstructure with intercolumnar pores predominantly aligned paralleling to the heat conduction gradient, which was harmful for their thermal insulation ability. However, considering synthetically the superior adhesion resulted from the elevated-

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temperature diffusion during deposition and excellent strain tolerance [3] provided by their columnar morphology, EB-PVD TBCs are promising candidates for future advanced TBCs.

Many reports had dealt with the relationship between process management and microstructure and therefore performance of EB-PVD TBCs [4-6]. The most successful investigation was the influence of substrate temperature and there was a most widely accepted explanation based on the 'structure zone model' proposed by Movchan et al [7]. Meanwhile, the substrate rotation speed during deposition was another factor to be managed if the substrate temperature was fixed. And different substrate rotation speed indeed changed the microstructure of EB-PVD TBCs. Since closely dependence on microstructural features such as porosity and morphology [8, 9], thermal conductivity was then indirectly affected by the substrate rotation speed. So far, much less effort was focused on this aspect. Consequently, this paper tried to reveal the influence of substrate rotation speed on the microstructure and thermal conductivity of EB-PVD TBCs.

2. Experimental procedure

The coatings were fabricated within a laboratory EB-PVD unit; model UE-204, which was introduced previously [10]. Before mounted in the vacuum chamber, the samples were grit blasted for several minutes aiming at cleaning and activating their surface， following by the ultrasonic and alcohol cleaning. The source material for top coat was 7wt% Y_2O_3 stabilized ZrO_2 (7YSZ). Then the deposition was started when the background pressure was less than 5×10^{-3}Pa and the samples were preheated enough at about 1173K. The rotation speeds for the top coat were selected to be 0 (stationary), 5, 15 and 20 rpm. The as-deposited coatings were finally annealed at 1050℃ for 8 hours in a vacuum furnace.

The microstructure as well as the fracture morphology of the as-annealed coatings under different rotation speed was investigated by SEM. In order to investigate the thermal conductivity, single 7YSZ coatings were coated on the Φ12.6mm cylindrical carbon steel substrate. The details for the preparation of free-standing coatings were illustrated in Fig.1.

Fig.1 Route chart for preparation of free-standing coatings: (a) polishing of carbon steel substrate; (b) preparation of the single 7YSZ coatings;(c) erosion in a hydrochloric acid solution; (d) cleanness for the final free-standing coatings

Thermal conductivity (λ) was derived from the following equation, $\lambda = \rho \cdot C_P \cdot \alpha$, where ρ is the density, C_p is specific heat and α is the thermal diffusivity. Firstly, the density of each sample was calculated by measuring their mass and volume using an electronic balance and a micrometer, respectively. Then the specific heat capacity was obtained by differential scanning calorimetery (DSC) and the thermal diffusivity was determined by laser flash method on a LFA427 set [11]. Finally, the experimental value of thermal conductivity of each sample was determined according to the above-mentioned equation.

3. Results and discussion

The microstructure of top coat observed from polished cross-section presented typical columnar morphology as shown in Fig.2. Since the substrate temperature and melting point of 7YSZ were about 1173 and 3003K, respectively, the columnar characteristic here was confirmed by the classical 'structure zone model'. Secondly, it was clear the microstructure can be regulated by different rotational speeds without altering the deposition temperature. Under stationary deposition, a denser coating structure was developed because of more vapor particles reached the samples and there was few shadow effect there. The diameter of the columnar grain was estimated as 1.4μm. In contrast, the coatings exhibited marked columnar morphology and open microporosity (see Fig.3. (b)) when the substrates rotated. Well-developed columnar grains could be clearly observed for the 15 and 20 rpm samples, but not distinct in the 5 rpm case. The diameters of the columnar grains for the 5, 15, 20 rpm samples were estimated at 2.9, 3.7 and 5.5μm, respectively.

Fig.3 illustrated the fresh fracture morphology for another 15 rpm sample deposited when the substrate temperature was about 1273K. The observation was carried out under secondary electron mode. This specimen had grown for 40 minutes and the average deposition rate was estimated to be 4.1μm /min. The microstructure of such rotational specimen exhibited the coarse (at the bottom) and fine (at the top) columnar morphology as shown in Fig.3.(a) with feather-like structures in an enlarged area (Fig.3.(b)). It was clear from the observation the columns grew wider from the bottom to the top and the fracture mechanism was concluded to be brittle fracture.

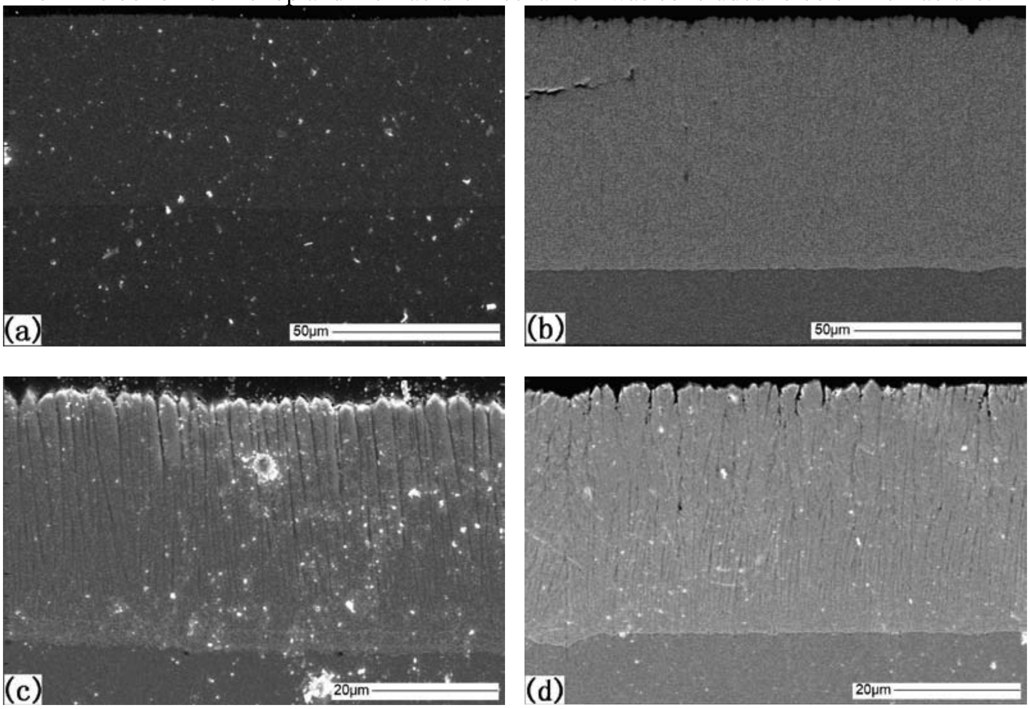

Fig.2 SEM images of cross-sectional for different substrate rotation speeds: (a) 0 rpm; (b) 5 rpm; (c)15 rpm; and (d) 20 rpm

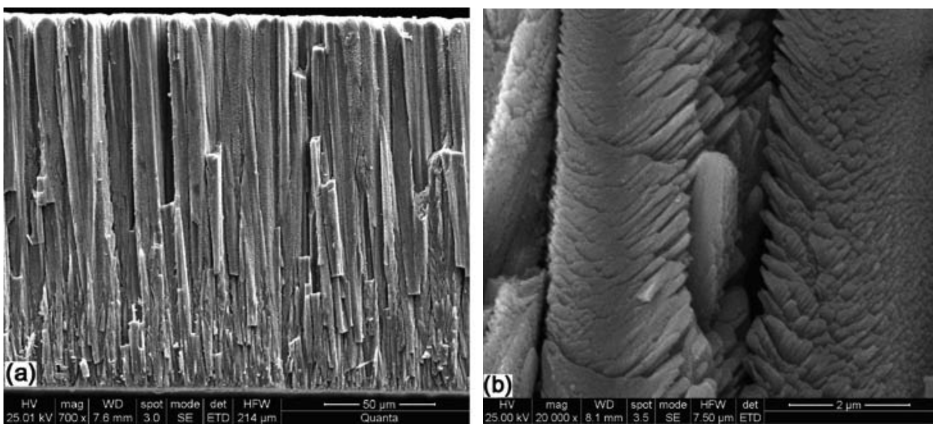

Fig.3 Fractured cross section of EB-PVD TBCs(a) with columnar morphology; (b) with feather-like structures and intercolumnar pores

The effect of substrate rotation on the diameter of the columnar grain was revealed in Fig.4. It can be concluded that, at the near substrate temperature and other processing parameters, the diameter of the columnar grain increased nonlinearly with increasing substrate rotation speed, which was consistent with the report by U.Schulz et al [12]. This phenomenon could be explained as the total influence of the amount of vapor particles arriving at the column top and the interval for the surface diffusion. The higher the substrate rotated, the less vapor particles reached the column top and the fewer intervals for the surface diffusion.

When the single 7YSZ coatings were about 165μm thick for the 40min deposition at about 1173K, the density exhibited a strong dependence on the substrate rotation speed as show in Fig.5 (a). The density decreased with increasing substrate rotation speed, which indicated a looser coating, that was a higher porosity volume then, was expected to get at a higher rotation speed. Since the theoretical density of the bulk 7YSZ material was 6.05g/cm^3, the calculated pores volume increased from 9% to 24% with increasing rotated speed in the range of 0-20 rpm.

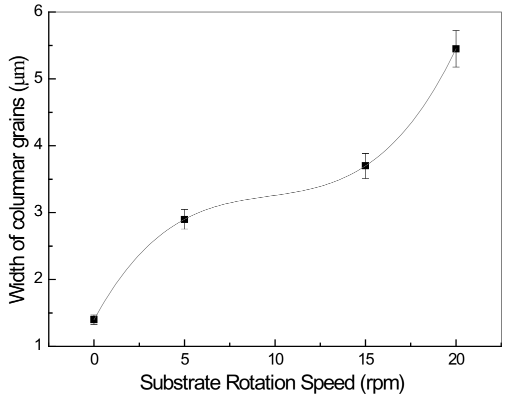

Fig.4 Dependence of width of columnar grains on the substrate rotation speed

It was known the density strongly depended on the volume of pores inside the coatings. The formation of porous columnar morphology was always explained by the so-called shadowing effect arose from the continuous variation of vapor incident angle of vapor flux during rotation [13]. With increasing rotation speed, the coating density decreased, which meant enlarged shadowing effect and therefore formation of many more gaps and/or pores in the shadow region as shown in Fig.3 (b). On the other side, the competitive growth of columns in their preferred growth direction resulted in wider grains which reduced the pores volume. However, the output in Fig.5 (a) indicated the enlarged shadowing effect under increasing rotation speed contributed much more in the total volume of pores and gaps.

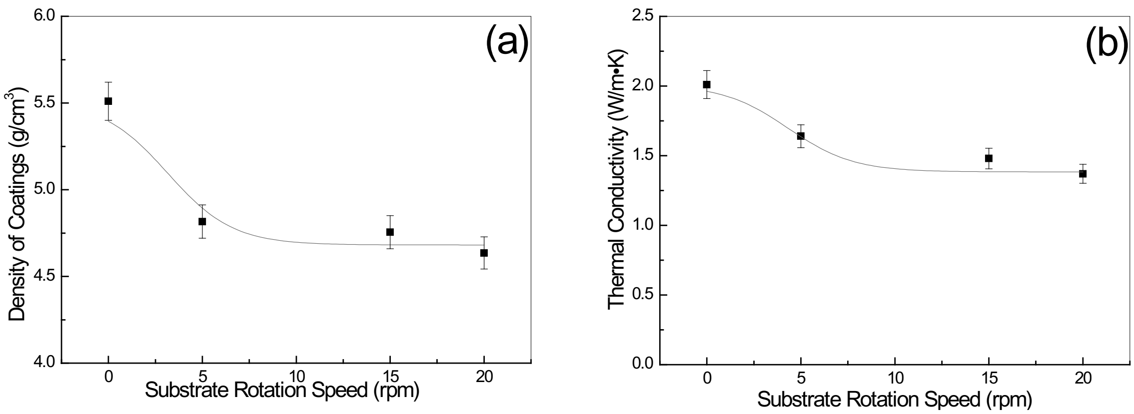

Fig.5 (a)Density of free-standing coatings; (b)Thermal conductivity at room temperature as a function of the substrate rotation speed

Besides, a higher density was proved for higher-temperature deposited TBCs [13], and the rotation of substrate can bring a temperature fluctuation during one revolution [6]. This temperature amplitude was in the size of $\Delta T=0.94*\tau$, where ΔT is the total temperature amplitude measured in K and τ is the time needed for one revolution in s. When the revolution was 12s, 4s and 3s, respectively, the real ΔT was 15±5K, 5±2K and 4±1K, a little higher than that according to the above relationship. In nature, such variation in substrate was small enough to ignore its influence on the coatings density.

After the determination of specific heat capacity and thermal diffusivity, the experimental value of thermal conductivity was finally calculated. Fig.5 (b) displayed the thermal conductivity at room temperature as a function of substrate rotation speed. The denser coatings deposited quietly possessed the largest thermal conductivity which could attribute to its least porosity volume. For the rotated samples, their thermal conductivity decreased with increasing rotation speed up to 20 rpm, and therefore the lowest value, 1.37 W/mK, was obtained then for a 171μm thick free-standing coating in this study. Such a result was similar with that reported by B.K. Jang et al [15], almost the same for the stationary sample yet a little larger for the rotated ones.

It had been proved the presence of porosity played the most important role in the reduction of thermal conductivity of TBCs by decreasing the mean free path because of the phonon scattering at pores [9]. In this investigation, the experimental trend of thermal conductivity was consistent with that of the coatings density and the fundamental mechanism was the formation of pores during deposition. Hence, the substrate rotation caused a variation of coating density and thermal conductivity by alternation of pore volume which was most derived from shadowing effect when other process parameters were nearly the same.

4. Conclusions

EB-PVD TBCs were fabricated at nearly the same process parameters but substrate rotation speed. The effect of substrate rotation speed adjustment on microstructure and thermal conductivity can be summarized as follows:

1) The specimens under different rotation speed all presented the columnar morphology, which was coincident with the 'structure zone model'. The stationary deposition formed denser coating and increasing rotation speed resulted in wider columnar grains which indicated the formation of more distinct columnar morphology.
2) The thermal conductivity at room temperature decreased with increasing rotation speed up to 20 rpm. Therefore, the lowest experimental value, 1.37 W/mK, was obtained for a 171μm thick free-standing coating in this study.
3) The enlarged shadowing effect derived from increasing rotation speed contributed most on the total volume of pores inside the coatings, which verified the formation of pores.

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