Key Engineering Materials *Online: 2006-08-15*
ISSN: 1662-9795, Vols. 317-318, pp 509-512
doi:10.4028/www.scientific.net/KEM.317-318.509

Thermal Conductivity of Nano-Pore Dispersed Y-PSZ Fabricated by EB-PVD

Byung-Koog Jang[a], Norio Yamaguchi[b] and Hideaki Matsubara[c]
Materials Research and Development Laboratory,
Japan Fine Ceramics Center, 2-4-1, Mutsuno, Atsuta-ku, Nagoya, 456-8587, Japan
[a]jang@jfcc.or.jp, [b]yamaguchi@jfcc.or.jp, [c]matsubara@jfcc.or.jp

Keywords: EB-PVD，Zirconia, Nano pore，Thermal conductivity, Columnar structure

Abstract. ZrO_2-4mol% Y_2O_3 coatings on zirconia substrate were deposited by EB-PVD. Influence of the coating thickness on thermal conductivity and thermal diffusivity of coated samples is examined. The fractured surface of the coated samples reveals a columnar microstructure consisting of feather-like structure. We have adopted a laser flash method for thermal diffusivity and specific heat capacity of coated samples. It was found that the thermal conductivity of the coating layers was strongly dependent on coating thickness. The thermal conductivity of coating layers shows increasing tendency with increasing the coating thickness.

1. Introduction

Electron beam-physical vapor deposition (EB-PVD) is a widely used technique for depositing thermal barrier coatings (TBCs) on Inconel substrates for high temperature applications, such as gas turbines, in order to improve the thermal efficiency. The EB-PVD process offers the advantages of superior strain and thermalshock tolerant behavior of the coatings due to their columnar microstructure [1-2]. The most durable TBCs for turbine parts are yttria-partially stabilized zirconia (Y-PSZ), which is an ideal candidate for TBCs because of low density, low thermal conductivity, high melting point and good thermal shock resistant properties.

Many studies were reported for developing TBCs with even low thermal conductivity to provide further improvement in engine performance. Therefore, the thermal conductivity is one of the most important properties for TBCs [3,4]. Especially, the thermal conductivity of porous coating layers is sensitive to the morphology, porosity and pore architecture [5-7]. The porous coating layers deposited by EB-PVD have a columnar structure, resulting in a non-uniform structure, making it difficult to measure the thermal conductivity, particularly for thinner coating thickness.

The purpose of the present work is to characterize the influence of YSZ coating thickness on the thermal conductivity of specimens deposited on zirconia substrate which is prepared by EB-PVD. We have therefore adopted laser flash method a means of determining the thermal conductivity of the coating layers from double-layer specimens consisting of coating and substrate [8-10].

2. Experimental

The coatings were deposited by EB-PVD using commercially available 4 mol% Y_2O_3 stabilized zirconia targets. We have used the zirconia substrates with ZrO_2-4 mol% Y_2O_3 composition and the disc shaped substrates consisted of 10.0 mm diameter and 1mm thick. The bond coating did not conducted to eliminate the influence of interface and bond coat layers on thermal conductivity. The substrates were first preheated to 900 ~ 1000°C in a preheating chamber using graphite heating element. The substrates were then moved to the coating chamber for deposition.

An electron beam evaporation process was conducted in a coating chamber under a vacuum condition using 45 kW electron gun. Deposition was generally conducted with rate of 5 μm/min and substrate rotation of 5 rpm. Total coating thickness is in the rage of 50 ~ 700 μm range for obtaining the different coating thickness. The quantitative analysis on pore size distributions in the coated layers was determined using mercury porosimeter.

The microstructure of coated specimens was observed by SEM. The density (ρ) of each coated sample was determined by measuring the mass and the volume of coated samples by a micrometer. To obtain the thermal conductivity ($\lambda=\rho\cdot\alpha\cdot C_p$), thermal diffusivity($\alpha$) and specific heat capacity(C_p) of coated specimens were measured at room temperature by the laser flash method.

3. Results and Discussion

The typical microstructures of center region of columnar grains in coating layers as function of coating thickness are illustrated in Fig. 1. The fractured surface of the coating layers deposited on ZrO_2 ceramic substrate clearly reveals columnar microstructure and all columnar grains were oriented in the same direction, i.e., perpendicular to the substrate. The thickness of columnar grains of each specimen shows some differences. The microstructure of specimen with 50 μm coating thickness shows more thinly columnar grains as compared to that of specimen with 500 μm coating thickness. It was found that columnar grains increased in size towards the top of column in comparison with the bottom of it. In addition, the intercolumnar gaps between columnar grains could be clearly observed, especially for sample with 500 μm coating thickness whereas these are not remarkable in the 50 μm coatings sample.

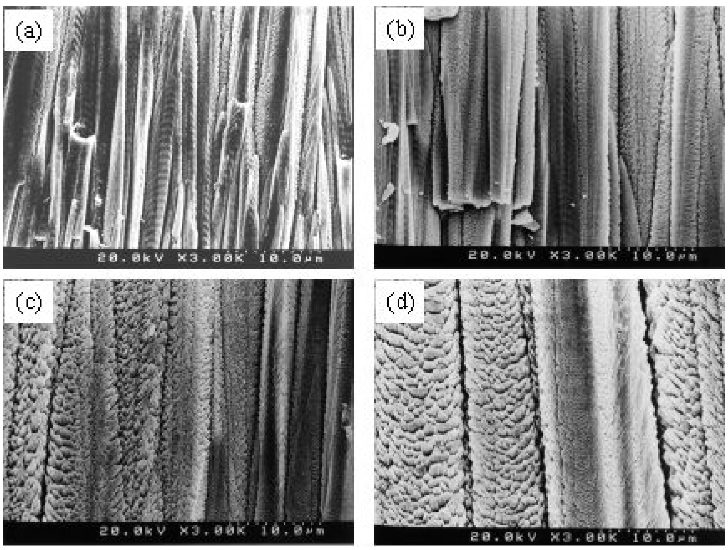

Fig. 1. SEM micrographs of fractured surface of ZrO_2-4 mol% Y_2O_3 deposited specimens with different coatings thickness: (a) 50 μm, (b) 100 μm, (c) 200 μm and (d) 500 μm.

Figure 2 shows the relationship between thermal conductivity, diffusivity and coating thickness for the combined double-layer specimens consisting of coating layers and substrate. Thermal conductivity and thermal diffusivity of thicker coated samples shows a lower tendency as compared to thinner coated samples. The thermal conductivity (λ_2) for coating layers alone can be derived according to Equation 1 [8,9].

$$\lambda_2 = \frac{d_2^{\,2} C_2 \rho_2 (3 d_1 C_1 \rho_1 + d_2 C_2 \rho_2)}{6 A_2 (d_1 C_1 \rho_1 + d_2 C_2 \rho_2) - d_1^{\,2} C_1 \rho_1 (d_1 C_1 \rho_1 + 3 d_2 C_2 \rho_2)/\lambda_1} \tag{1}$$

where d_1, C_1, ρ_1 and d_2, C_2, ρ_2 correspond to the thickness, specific heat and density of substrate and coating layers, respectively. λ_1 is the thermal conductivity of substrate. For double-layer specimens (coatings and substrate), the area bounded by the temperature rise curve and the maximum temperature line at the rear face of the coated specimen after the laser pulse heating, designated A_2 in Fig. 3, can be obtained by integration. This area is called the "areal thermal diffusion time" and it is given in Table 1.

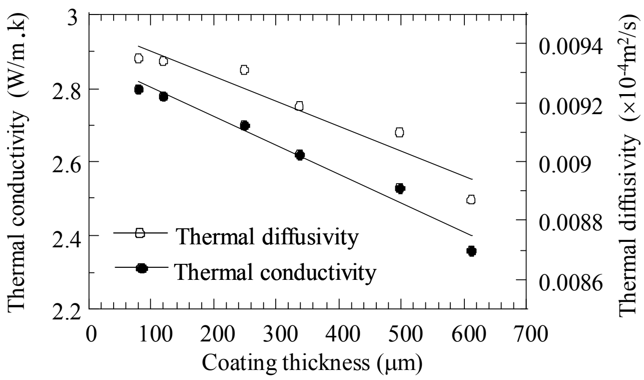

Fig. 2. Influence of coatings thickness on thermal conductivity and thermal diffusivity for combined coating and substrate specimens.

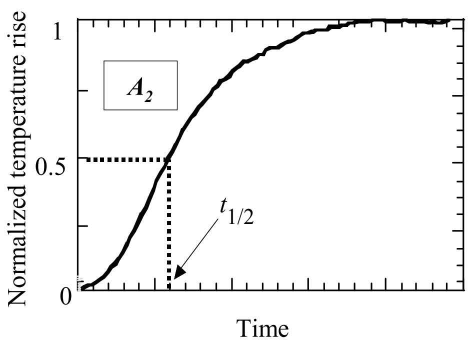

Fig. 3. Temperature response behavior as a function of time at the rear surface of coated specimen after laser pulse heating.

Table 1. Calculated areal thermal diffusion time values as function of coating thickness for coated specimens.

Coating thickness (μm)	Areal thermal diffusion time (A_2)
79	0.208
119	0.223
249	0.279
337	0.324
497	0.415
613	0.489

Figure 4 shows the correlation between the calculated thermal conductivities for coating layers alone with coating thickness based on Equation 2. The thicker coating layers reveal higher thermal conductivity than that of thinner coated specimens. This reason can be considered that low thermal conductivity at thinner coating layer depends on the microstructural nature of columnar grains. Figure 5 shows the number of column boundaries per same area for each ZrO_2-4 mol% Y_2O_3 layers. The number of column boundaries between columns decreased with increasing the coating thickness. The width and size of columns for thinner coating layers are small and fine, resulting in many columns as shown in Fig.1. On contrary, the width and size of columns for thicker coating layers are relatively large and coarse. Consequently, the thinner coating layers have many column boundaries in comparison with that for thicker coating layers. Generally, phonons interact with imperfections such as dislocation, vacancy, pore and grainboundary. Therefore, many column boundaries in thinner coating layers significantly make to reduce the mean free path by phonon scattering, resulting in the reduction of thermal conductivity. Figure 6 indicates the result of pore distribution for coating layer with 200 μm coating thickness. According to pore distribution analysis, the volume of the nano pores (for consideration of size < 100 nm in present work) is approximately 40 % and an average pore size is about 0.3 μm. The nano pores portion is easily formed during competitive columns growth of first stage by the shadowing effect after nucleation of columnar grains on substrate [4,5]. The nano pores are thought to be contribution for increase of phonon scattering because they can intersect the heat flow path.

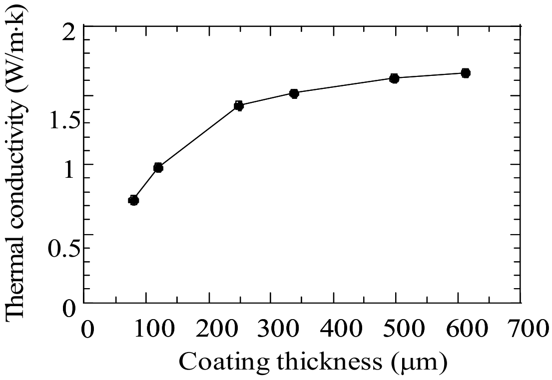

Fig. 4. Thermal conductivity as a function of coating thickness for ZrO_2-4 mol% Y_2O_3 coating layers.

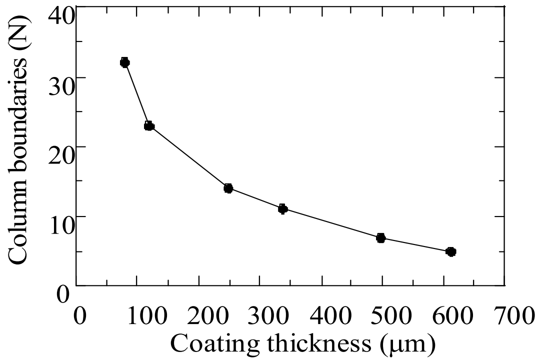

Fig. 5. Number of column boundaries per 30 μm width as a function of coating thickness.

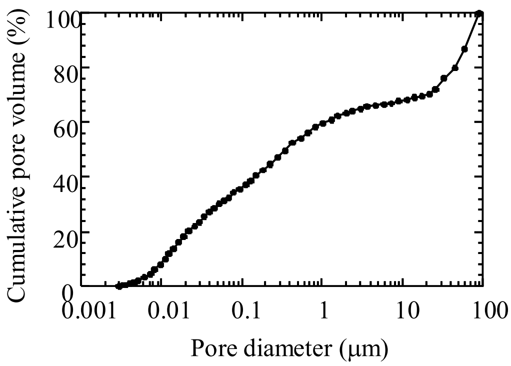

Fig. 6. Pore size distribution of ZrO_2-4 mol% Y_2O_3 coating layers.

4. Conclusions

A ZrO_2- 4 mol% Y_2O_3 coating layer was deposited on YSZ substrate by EB-PVD and found to consist of columnar grains. The thermal conductivity and thermal diffusivity of the combined coating and substrate specimens decreased with increasing the coating thickness. However, thermal conductivity of coating layers alone increased with increasing coating thickness. The thinner coating layers have many column boundaries than that of thicker coating layers. Increase of column boundaries in coating layers mainly leads to reduced thermal conductivity by the reduction of mean free path by phonon scattering.

Acknowledgments
The authors acknowledge the financial support of the New Energy and Industrial Technology Development Organization (NEDO), Japan.

References

[1] U.Schulz, S.G. Terry and C.G.Levi: *Mater. Sur. Eng. A*, 360 (2003) 319.
[2] C.A. Johson, J.A. Ruud, R.Bruce and D.J. Wortman: *Sur. Coat. Tech.*, 108-109 (1998) 80.
[3] D.D. Hass, P.A.Parrish and H.N.G. Wadley: *J.Vac. Sci. Technol.*, 16 (1998) 3396.
[4] C.G. Levi: *Current Opinion in Solid State Mat. Sci.*, 8 (2004) 77.
[5] T.J. Lu, C.G. Levi, H.N.G. Wadley and A.G. Evans: *J. Am. Ceram. Soc.*, 84 (2001) 2937.
[6] J.R. Nicholis, K.J. Lawson, A.Johnstone, D.S. Rickerby: *Sur. Coat. Tech.*,151-152 (2002) 383.
[7] J.Singh, D.E. Wolfe, R.A. Miller, J.I. Eldridge and D.M.Zhu: *J. Mater. Sci.*, 39 (2004) 1975.
[8] B.K.Jang, M.Yoshiya, N. Yamaguchi and H. Matsubara: *J. Mater. Sci.*, 39 (2004) 1823
[9] B.K.Jang, and H. Matsubara: *Trans. of Mater. Res. Soc. Jpn.*, 29 (2004) 417 .
[10] T. Baba and N. Taketoshi: *Proc. 2000 21th Jpn. Symp. on Thermophysical. Prop*. Tokyo, p.229.