### ARTICLE IN PRESS

Ceramics International xxx (xxxx) xxx-xxx



Contents lists available at ScienceDirect

## Ceramics International



journal homepage: www.elsevier.com/locate/ceramint

# Thermophysical behavior of thermal sprayed yttria stabilized zirconia based composite coatings

S. Nath<sup>a</sup>, I. Manna<sup>b,c</sup>, A.K. Jha<sup>d</sup>, S.C. Sharma<sup>d</sup>, S.K. Pratihar<sup>e</sup>, J. Dutta Majumdar<sup>b,\*</sup>

<sup>a</sup> Department of Mechanical, Aerospace and Automotive Engineering, Coventry University, Coventry CV1 2JH, United Kingdom

<sup>b</sup> Department of Metallurgical and Materials Engineering, Indian Institute of Technology, Kharagpur 721302, West Bengal, India

<sup>c</sup> Indian Institute of Technology, Kanpur 208016, Uttar Pradesh, India

<sup>d</sup> Vikram Sarabhai Space Centre (VSSC), Thiruvananthapuram 695022, India

<sup>e</sup> Department of Ceramic Engineering, National Institute of Technology, Rourkela 769008, Odisha, India

#### ARTICLE INFO

Keywords: Composite coatings Yttria stabilized zirconia Porosity Coefficient of thermal expansion Thermal conductivity

#### ABSTRACT

The effective thermal conductivity of a composite coating depends on intrinsic thermal conductivity of the constituent phases, its characteristics (size, shape) and volume fraction of porosities. The present study concerns studying the effect of CoNiCrAlY and  $Al_2O_3$  content on the coefficient of thermal expansion and thermal conductivity of the YSZ (YSZ-CoNiCrAlY and YSZ- $Al_2O_3$ ) based composite coatings developed by thermal spray deposition technique. The coefficient of thermal expansion and thermal conductivity of the Composite coatings were measured by push rod dilatometer and laser flash techniques, respectively, from room temperature to 1000 °C. Variation in density, porosity, coefficient of thermal expansion, and thermal conductivity was observed in the composite coatings with the addition of different volume fraction of CoNiCrAlY and  $Al_2O_3$  powders in YSZ-CoNiCrAlY and YSZ- $Al_2O_3$  composites, respectively. Comparison between the theoretical and experimental thermal conductivities showed a mismatch varying from 4% to 58% for YSZ-CoNiCrAlY composite coatings and from 58% to 80% for YSZ-Al\_2O\_3 composite coatings. Model based analyses were used to understand the mechanism of thermal conductivity reduction in the composite coatings. It was concluded that the morphology of porosities varied with composition.

#### 1. Introduction

Thermal barrier coatings (TBCs) are useful in protecting and extending the operational life of the gas turbine engine components (burners, transition ducts, blades, and vanes) which are exposed to corrosive and oxidative environments operated at elevated temperature [1,2]. The efficiency of a turbine engine may be improved by increasing the turbine inlet temperature, and increasing the life of the coated component.

Conventional TBC consists of a two layer coatings where, the ceramic top coat (YSZ) is deposited on to the surface of pre-deposited bond coat (MCrAlY, M = Co, Ni, or both). Though, the superior thermal shock resistance of the Yttria stabilized Zirconia (YSZ) coating makes it a popular material for TBC application, however, the mismatch in the coefficient of thermal expansion (CTE) between the top coat and bond coat is the major cause of failure of TBC under cyclic environment [3,4]. To minimize the failure due to coefficient of thermal expansion (CTE) mismatch, in a MCrAlY/YSZ duplex TBC, and a MCrAlY/YSZ

functionally graded thermal barrier coating (FGTBC) has been proposed [4-10]. YSZ/Al<sub>2</sub>O<sub>3</sub> composite coatings have also been reported to improve the hot corrosion resistance and oxidation resistance of TBC [11–14]. Presence of Al<sub>2</sub>O<sub>3</sub> phase in the YSZ/Al<sub>2</sub>O<sub>3</sub> composite coating is beneficial in improving its thermal cycling resistance, hot corrosion and oxidation resistance properties [11–13]. However, a detailed study of the thermal properties of YSZ/Al<sub>2</sub>O<sub>3</sub> composite coating has not been undertaken. Thermal conductivity of any material is an important parameter which allows a quantitative as well as qualitative assessment of the heat transfer characteristics of the material. A significant reduction in thermal conductivity can be achieved by the introduction of microstructural defects in the form of porosities, micro-cracks, and interfaces. Yang et al. [15] compared the experimentally measured thermal conductivity of the sintered YSZ/Al<sub>2</sub>O<sub>3</sub> composites with its theoretical value to conclude on the role of interfacial thermal resistance on the effective thermal conductivity of the composites [15]. The deviation of the thermal conductivity from its intrinsic value is not only dependent on the content of porosity in the coating but also

\* Corresponding author.

*E-mail addresses:* subhasisa.nath@coventry.ac.uk (S. Nath), imanna@iitkgp.ac.in (I. Manna), ak\_jha@vssc.gov.in (A.K. Jha), sharma\_sc@vssc.gov.in (S.C. Sharma), skpratihar@nitrkl.ac.in (S.K. Pratihar), jyotsna@metal.iitkgp.ernet.in (J. Dutta Majumdar).

http://dx.doi.org/10.1016/j.ceramint.2017.05.170

Received 15 March 2017; Received in revised form 22 May 2017; Accepted 24 May 2017 0272-8842/ © 2017 Elsevier Ltd and Techna Group S.r.l. All rights reserved.

on its morphology [16–18]. In a composite coating, the morphology and area fraction of the pores are dependent on the constituent phases present in the microstructure as they alter the effective thermal conductivity of the composite coatings. Process parameters also have a strong influence on the melting behavior of the powder particles during thermal spray deposition as they control the in-flight particle state (temperature and velocity) and hence, the microstructure [19]. Hence, engineering the composition as well as microstructure is a biggest challenge to enhance the service life of the coated component.

It is well understood that the inter-lamellar porosities have a dominant role in reducing the thermal conductivity more effectively than that of globular porosities due to the presence of porosities aligned perpendicular to the direction of heat flow. It is also reported that the morphology and orientation of porosities in the coating change the effective thermal conductivity of the thermal spray coatings [16-18]. The analyses are based on the available analytical models for different pore shapes [20-22]. From the reported results, it may be concluded that analytical models are helpful in establishing an understanding between microstructural features (structure) and thermal conductivity (property). Though, the reported results were based on the analysis of the 100% ceramic coatings, however, no discussions are available concerning the effect of composition (e.g. composite coatings) on the morphology of porosities and hence, the thermal conductivity behavior. Bakshi et al. [18] studied the thermal conductivity of the multi-walled carbon nanotubes (MWNT) - Al<sub>2</sub>O<sub>3</sub> composite coatings with varying MWNT content developed by plasma spray method and reported a good match between the theoretical thermal conductivity, calculated by the available theoretical models, and experimentally measured values for some models and poor match for others. However, extensive studies need to be undertaken to address the composition induced change in morphology of pores developed by thermal spray deposition technique.

In the present study, composite coatings consisting of CoNiCrAlY-YSZ and YSZ-Al<sub>2</sub>O<sub>3</sub> have been developed by thermal spray deposition technique. The thermal properties such as coefficient of thermal expansion and thermal conductivity of the composite coatings have been measured to understand the effect of composition on the thermal properties of the composite coatings. Finally, analytical model based thermal conductivity analyses have been undertaken to understand the effect of composition on the effect of immense importance owing to the fact that the pore morphology significantly alters the effective thermal conductivity of the composite coating.

#### 2. Experimental

#### 2.1. Materials

Commercially available CoNiCrAlY alloy powder (Co-32Ni-21Cr-8Al-0.5Y in wt%, MEC 9950AM, particle size 15-45 µm), yttria stabilized zirconia (7 wt% Y2O3-ZrO2, Amperit 831.007, particle size 15-85 µm) and Al<sub>2</sub>O<sub>3</sub> (Amperit 740.1, particle size 22-45 µm) ceramic powders were used as feedstock powders for the development of CoNiCrAlY-YSZ and YSZ- Al<sub>2</sub>O<sub>3</sub> composite coatings. Fig. 1 shows the scanning electron micrographs of (a) YSZ, (b) Al<sub>2</sub>O<sub>3</sub>, and (c) CoNiCrAlY powders used as feedstock for the development of CoNiCrAlY-YSZ and YSZ-Al<sub>2</sub>O<sub>3</sub> composite coatings. The shape of YSZ and CoNiCrAlY powders are spherical which ensure good flow characteristics of the powder particles. On the other hand, the Al<sub>2</sub>O<sub>3</sub> powder is irregular in shape due to the partial ionic bonding characteristics and a typical powder processing route. Fig. 2 shows the X-ray diffraction profiles of (a) YSZ, (b) Al<sub>2</sub>O<sub>3</sub>, and (c) CoNiCrAlY feedstock powders. The X -ray diffraction profiles of YSZ (Fig. 2(a)) and  $Al_2O_3$  (Fig. 2(b)) feedstock powders show presence of single phase tetragonal zirconia (t'-ZrO<sub>2</sub>) and α-Al<sub>2</sub>O<sub>3</sub>, respectively. On the other hand, the X-ray diffraction profile of CoNiCrAlY powder (Fig. 2(c))

shows the presence of  $\gamma'$  – Ni<sub>3</sub>Al and  $\beta$ –CoAl phases in  $\gamma$ –Co matrix. The  $\beta$ –CoAl phase acts as an aluminum reservoir which helps in the formation of Al<sub>2</sub>O<sub>3</sub> scales during high temperature exposure of TBC.

#### 2.2. Development of composite coatings

Prior to thermal spray deposition, the substrates were grit blasted using alumina grits followed by simultaneous cleaning in acetone and isopropyl alcohol. For the development of CoNiCrAlY/YSZ composite coatings, CoNiCrAlY and YSZ powders were initially mixed in the volume ratio of 70:30, 50:50, and 30:70 using a planetary ball mill for 4 h at 300 rpm to ensure proper mixing of powders without altering their original shapes. The 100% CoNiCrAlY and 100% YSZ coatings were deposited on the grit blasted substrates using high velocity oxyfuel spray (HVOF) and atmospheric plasma spray (APS) techniques, respectively. Similarly, for the development of YSZ-Al<sub>2</sub>O<sub>3</sub> composite coatings,  $Al_2O_3$  and YSZ powders were mixed in the volume ratio of 70:30, 50:50, and 30:70 using a planetary ball mill for 4 h at 300 rpm to ensure proper mixing of powders. Table 1 summarizes the process parameters employed for the plasma spray deposition of composite coatings.

#### 2.3. Characterization of thermal barrier coatings

Followed by the development of coating, a detailed characterization of the microstructure of the coated surface was carried out by field emission scanning electron microscopy (SUPRA 40, Zeiss SMT AG, Germany). The grain size of the YSZ and  $Al_2O_3$  coatings were measured by linear intercept method (ASTM E112). A detailed phase analysis of the coating was carried out by X-ray diffraction (XRD) technique (Bruker D8 Discover, Germany) using Cu K $\alpha$  radiation (wavelength ~ 0.15418 nm) at a scanning speed of 0.05°/s. The X-ray source was operated at an accelerating voltage of 40 kV and current of 40 mA.

Free standing coatings of CoNiCrAlY/YSZ were obtained by depositing the coatings onto the polished steel substrates followed by carefully cutting out the coating using a slow speed diamond cutter. On the other hand, YSZ-Al<sub>2</sub>O<sub>3</sub> composite coatings were deposited onto the polished steel substrates followed by immersing it in a 40% HNO<sub>3</sub> solution for 3 h to get the free standing coatings. The free standing coatings were then oven dried for measuring the dry weight. The density of the free standing coatings is calculated by Archimedes' principle as given by Eq. (1).

$$\rho = w_1 / w_2 - w_3 \tag{1}$$

where,  $\rho$ ,  $w_1$ ,  $w_2$ , and  $w_3$  represent the density of the coating, initial dry weight of the coating, water saturated weight of the coating, and weight of coating in de-ionized water, respectively. The saturated weight of the coating was obtained by soaking the free standing coating in the boiling water for one hour followed by measuring its weight using a precision weighing balance. The total porosity content can be evaluated from Eq. (2) as follows:

Total porosity in the coating, 
$$P_{total} = 1 - (\rho_{measured} / \rho_{theoretical})$$
 (2)

The theoretical densities,  $\rho_{\text{theoretical}}$  of YSZ, Al<sub>2</sub>O<sub>3</sub>, and CoNiCrAlY are 5.96 g/cm<sup>3</sup> [15], 3.98 g/cm<sup>3</sup> [15], and 7.24 g/cm<sup>3</sup> [23], respectively. The theoretical densities of CoNiCrAlY-YSZ and YSZ-Al<sub>2</sub>O<sub>3</sub> composite coatings were calculated using rule of mixture which are presented in Table 2.

The coefficient of thermal expansion of the free standing as-sprayed coatings (with dimension of  $10 \times 5 \times 1 \text{ mm}^3$ ) was measured in air from 27 °C to 1000 °C using a dilatometer (NETZSCH DIL 402 C, Germany) at a heating rate of 10 K/min. Fractional change in length,  $\Delta L/L$  as a function of temperature was measured and the coefficients of thermal expansion,  $\alpha$ , was measured from the slope of the curve.

Thermal diffusivity of the freestanding coatings  $(10 \times 10 \times 1 \text{ mm}^3)$  was measured using laser flash technique (LFA 427, Netschz,

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