

# Microstructures and properties of double-ceramic-layer thermal barrier coatings of $\text{La}_2(\text{Zr}_{0.7}\text{Ce}_{0.3})_2\text{O}_7/8\text{YSZ}$ made by atmospheric plasma spraying

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## ABSTRACT

A double-ceramic-layer (DCL) thermal barrier coatings (TBC) of  $\text{La}_2(\text{Zr}_{0.7}\text{Ce}_{0.3})_2\text{O}_7/8\text{YSZ}$  (LZ7C3/8YSZ) was prepared by atmospheric plasma spraying (APS). The phase structure, composition, thermal conductivity, surface and cross-sectional morphologies, adhesion strength and thermal shock behavior of the LZ7C3/8YSZ coating were investigated. The X-ray diffraction pattern showed that the phase structures of top coat LZ7C3 was different from the powder for spraying, which consists of pyrochlore LZ and fluorite LC structures. Main peaks between LZ and LC in as-sprayed LZ7C3 have almost overlapping diffracted angles and approximately equal diffracted intensity. Thermal shock lifetime and adhesion strength of the DCL LZ7C3/8YSZ coating are enhanced significantly as compared to single LZ7C3 coating, and are very close to that of single 8YSZ coating. The mechanisms of performance improvement are considered to be effective reduction of stress concentration between substrate and LZ7C3 coating by 8YSZ buffer effect, and the gentle thermal gradient initiated at the time of quenching in water. The DCL LZ7C3/8YSZ coating has lower thermal conductivity than 8YSZ, which was only 25% of 8YSZ bulk material and 65% of 8YSZ coating by APS.

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## 1. Introduction

Thermal barrier coatings (TBCs) are widely used in turbine engines to protect hot-section metallic components from corrosion and oxidation [1,2]. Typical TBC systems are composed of a zirconia stabilized with 8 wt. % yttria (8YSZ), a metallic bond coat and a Ni-based superalloy substrate. The 8YSZ is widely used as the standard material of TBCs due to its low thermal conductivity ( $2.1 \text{ W m}^{-1} \text{ K}^{-1}$ ), relatively high thermal expansion coefficient ( $11 \times 10^{-6} \text{ K}^{-1}$ ) and chemical inertness in combustion atmospheres [3]. However, above  $1200^\circ\text{C}$ , it could hardly be used for long-term application due to its low sintering resistance and low phase stability, which leads to volume increase and accelerates the spallation failure of TBCs [4–6].

In the next generation of advanced engines, further increase in thrust-to-weight ratio will require even higher gas turbine inlet temperature. To meet this requirement, two aspects are taken into consideration. The one is to develop new TBCs materials with higher temperature capability and lower thermal conductivity

than the current 8YSZ [7]. Recently, some materials have been evaluated as the candidates for TBCs, such as  $\text{LaMgAl}_{11}\text{O}_{17}$  (LMA),  $7.5 \text{ wt.}\% \text{ Y}_2\text{O}_3\text{-HfO}_2$  (YSH),  $\text{La}_2\text{Zr}_2\text{O}_7$  (LZ),  $\text{La}_2\text{Ce}_2\text{O}_7$  (LC) and  $\text{La}_2(\text{Zr}_{0.7}\text{Ce}_{0.3})_2\text{O}_7$  (LZ7C3) [5,8–12]. Among those interesting candidates, the LZ7C3 ceramic shows the promising thermophysical properties for high-temperature TBCs, but relatively low thermal expansion coefficient of LZ7C3 will lead to high thermal stress between LZ7C3 coating and substrate. The single LZ7C3 coating usually has a short thermal shock life [13]. The other is to design new structure of TBCs. The multilayer design could be effective to improving the thermal shock life of TBCs as Tamura and Takahashi suggested [14,15]. X. Q. Cao et al. prepared a five-layer  $\text{La}_2(\text{Zr}_{0.7}\text{Ce}_{0.3})_2\text{O}_7/8\text{YSZ}$  functionally graded thermal barrier coating prepared by atmospheric plasma spraying [16], and the lifetime reached to 880 thermal cycles. The multilayer system exhibits excellent thermal cycle property, but its preparation technology is very complex. A double-ceramic-layer (DCL) coating is newly developed, such as  $\text{La}_2\text{Zr}_2\text{O}_7/8\text{YSZ}$  (LZ/8YSZ) [17,18],  $\text{La}_2\text{Ce}_2\text{O}_7/8\text{YSZ}$  (LC/8YSZ) [6] and  $\text{La}_2(\text{Zr}_{0.7}\text{Ce}_{0.3})_2\text{O}_7/\text{La}_2\text{Ce}_2\text{O}_7$  (LZ7C3/LC) [19]. In DCL coating, the top ceramic layer should have a low thermal conductivity and high phase stability, and it acts as a thermal insulator to protect the inner layer [14–21]. Because the LZ7C3 has low thermal conductivity ( $0.79 \text{ W m}^{-1} \text{ K}^{-1}$ ) and high

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sintering-resistance [11,13], and the 8YSZ has high thermal expansion coefficient ( $11 \times 10^{-6} \text{ K}^{-1}$ ) and high shock-resistance [3], it is hopeful that the LZ7C3 coating layered on top of 8YSZ coating as a topcoat will have better thermal protection and thermal shock lifetime.

Currently, two processes are used to produce thermal barrier coatings, namely the atmospheric plasma spraying (APS) and the electron beam physical vapor deposition (EB-PVD). The DCL coatings prepared by EB-PVD usually have long thermal cycling lifetime because the EB-PVD TBC has high strain compliance [6,17–19]. The major disadvantage of EB-PVD coating are the relatively higher thermal conductivity, which induces poor heat insulation effect, compared with coating prepared by APS as reported by J.R. Nicholls [22,23]. M. Han et al. [24–26], reported optimization method of structure design on DCL LZ7C3/8YSZ by numerical simulation. It was found that 8YSZ coating eases effectively the thermal stress by the expansion mismatch between LZ7C3 coating and substrate. However, no report about the DCL coating of LZ7C3/8YSZ prepared by APS have been published up to now, and no thermal shock performance and thermal conductivity of LZ7C3/8YSZ coating are available.

In the present work, the APS was used to prepare the DCL coating of LZ7C3/8YSZ with lower thermal conductivity and better heat insulation effect. Meanwhile, the phase structure, composition, surface and cross-sectional morphologies, adhesion strength and thermal shock behavior of the LZ7C3/8YSZ coating were studied, and its thermal conductivity was also measured.

## 2. Experimental procedure

### 2.1. Synthesis of LZ7C3

The LZ7C3 with desired composition was synthesized by solid state reaction at  $1400^\circ\text{C}$  for 12 h using  $\text{La}_2\text{O}_3$ ,  $\text{CeO}_2$  (GRIPM Advanced Materials Co., Ltd., purity  $\geq 99.99$ ) and  $\text{ZrO}_2$  (Shanghai st-nano science and technology Co., Ltd purity  $\geq 99.99\%$ ). The APS spraying powders of LZ7C3 for plasma spraying were produced by spray drying process, and its morphology is shown in Fig. 1. Commercial 8YSZ powders (GRIPM Advanced Materials Co., Ltd.) and Ni-30Cr-12Al-0.3Y (wt. %) were chosen for spraying the bottom ceramic layer and the bond coat, respectively.

### 2.2. Preparation of LZ7C3/8YSZ coating

The LZ7C3/8YSZ coatings were prepared by APS with Hang-TianZhenBang plasma spray units. High velocity oxygen fuel

(HVOF) spraying was used to spray NiCrAlY bond coat onto disk-shaped Ni-based superalloy substrate (DZ125) with a diameter of 30 mm and a thickness of 3 mm. Then the 8YSZ coating was firstly prepared onto the bond coat followed by the spraying of LZ7C3 coating. The main parameters used for spraying LZ7C3/8YSZ coating are listed as follow: Current is 510 A, Voltage is 70 V, Distance is 70 mm, Ar/H<sub>2</sub> is 45/12 slpm, Feed rate is 30 g/min and Gun velocity is 500 mm/s.

### 2.3. Thermal conductivity

The thermal diffusivity ( $\lambda$ ) of LZ7C3/8YSZ coating (the thickness of LZ7C3 and 8YSZ is 0.82 mm and 0.87 mm respectively) was measured using laser flash method (Netzsch LFA427). The specific heat capacity ( $C_p$ ) was measured using differential scanning calorimetry (DSC Netzsch DSC204-F1). The density ( $\rho$ ) was measured by the Archimedes principle. The thermal conductivity ( $k$ ) of coating was calculated by Eq. (1) with specific heat capacity ( $C_p$ ), density ( $\rho$ ) and thermal diffusivity ( $\lambda$ ):

$$k = \lambda \cdot \rho \cdot C \quad (1)$$

### 2.4. Thermal shock and adhesive strength test

The adhesive strength of LZ7C3/8YSZ coatings was performed by tensile adhesion test on the CMT5504 test machine. Thermal shock tests were conducted in a programmable furnace in air. When the temperature of the furnace reached to  $1100^\circ\text{C}$ , the samples were put into the furnace. The holding time at high temperature was 10 min and then the samples were directly quenched into water, the water temperature throughout the thermal shock test was between  $20^\circ\text{C}$  and  $30^\circ\text{C}$ . This method of thermal shock testing was also used by other researchers [27–30]. Three parallel specimens were conducted in the test. The lifetime of the coating was defined as the number of thermal shock when the spallation region of the coating was more than 5% of the coating surface area.

### 2.5. Microstructure and phase

The sprayed coatings were characterized by both optical and scanning electron microscopy (ESEM, FEI Quanta 250). The composition of the coatings was analyzed by energy dispersive spectroscopy (EDS, EDAX GENESIS). Phase analysis of top coating was determined by X-Ray diffractometry (XRD, Rigaku D/Max 2500) with Ni filtered Cu  $K\alpha$  radiation (0.1542 nm) at the scanning rate of  $4^\circ/\text{min}$ .

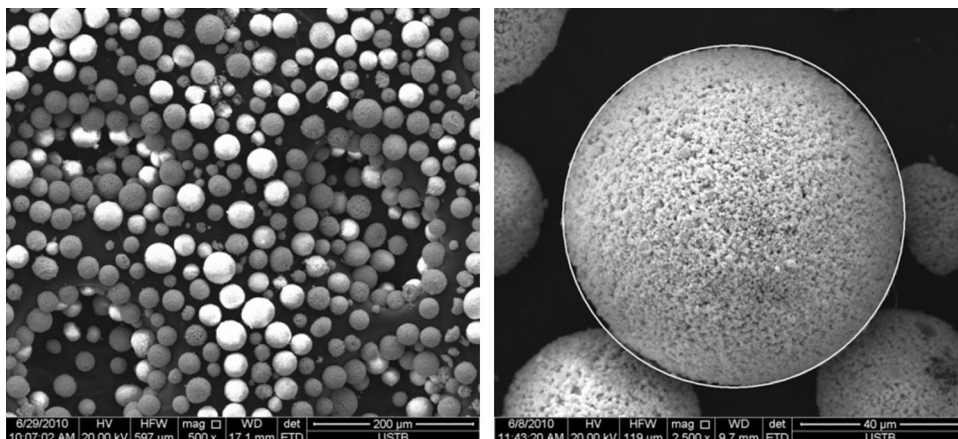


Fig. 1. Macro (a) and micro (b) morphology of spray-dried LZ7C3 powders for plasma spraying.

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