



# Characterization and thermal cycling behavior of $\text{La}_2(\text{Zr}_{0.7}\text{Ce}_{0.3})_2\text{O}_7/8\text{YSZ}$ functionally graded thermal barrier coating prepared by atmospheric plasma spraying



Sumei Zhao<sup>a,b</sup>, Yu Zhao<sup>a,b</sup>, Binglin Zou<sup>a</sup>, Xizhi Fan<sup>a,b</sup>, Jiaying Xu<sup>a,b</sup>, Yu Hui<sup>a,b</sup>, Xin Zhou<sup>a,b</sup>, Sanxi Liu<sup>c</sup>, Xueqiang Cao<sup>a,\*</sup>

<sup>a</sup> State Key Laboratory of Rare Earth Resources Utilization, Changchun Institute of Applied Chemistry, Chinese Academy of Sciences, Changchun 130022, China

<sup>b</sup> University of the Chinese Academy of Sciences, Beijing 100049, China

<sup>c</sup> Department of Chemical and Biological Engineering, Changsha University of Science and Technology, Changsha 410114, China

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## ABSTRACT

A five-layer ceramic-ceramic functionally graded thermal barrier coating (FG-TBC) of  $\text{La}_2(\text{Zr}_{0.7}\text{Ce}_{0.3})_2\text{O}_7$  (LZ7C3) and 8YSZ has been prepared by atmospheric plasma spraying. The thermal expansion coefficients (TECs) of the five ceramic layers of the graded coating from top layer to the inner layer are gradually increased. Thermal cycling behavior and failure mechanism of the coating have been studied. The abnormal oxidation of the bond coat and sintering of the LZ7C3 are the primary factors for the spot spallation of the graded coating. The effects of thermal expansion mismatch also accelerate the spot spallation. The spallation at the edge of the coating is due to the effect of thermal expansion mismatch and thermal stress produced during thermal cycling.

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## 1. Introduction

Thermal barrier coatings (TBCs) which protect substrate materials against thermal corrosion and oxidation have been widely used in the production of corrosion-resistant and high-temperature structural parts such as gas turbine engines, diesel engines and power generation systems [1–4]. The material typically used nowadays is yttria partially stabilized zirconia (YSZ), especially 8YSZ [5,6]. 8YSZ performs well up to about 1200 °C, but it cannot be used above 1200 °C for long time due to sintering and phase transformation which would lead to a reduction of the strain tolerance in combination with an increase of Young's modulus and a volume change during cooling [7,8].

In order to increase the operation temperature and lifetime of TBCs, new TBCs structures were designed and new materials were developed [9]. As a result, a ceramic (YSZ)–metal (bond coat) functionally graded TBC (FG-TBC) has been widely investigated to improve the TBC lifetime [10–15]. However, such a graded microstructure leaves the maximum metal surface area which would facilitate bond coat oxidation and cause catastrophic volumetric changes with crack nucleation and growth, finally leading

to the failure of coating system. So new TBCs structures were designed [16–18]. A typical ceramic–ceramic FG-TBC has also been proposed to solve the residual stress problem.  $\text{LaMgAl}_{11}\text{O}_{19}/8\text{YSZ}$  (LMA/8YSZ) FG-TBCs [19,20] and  $\text{La}_2\text{Zr}_2\text{O}_7/8\text{YSZ}$  (LZ/8YSZ) FG-TBCs [9,21] have been prepared to improve the thermal cycling lifetime of the TBCs.

Recently, some new candidate materials for TBCs' application at high temperature, such as LMA [19,20],  $\text{LaTi}_2\text{Al}_9\text{O}_{19}$  [22], metal-glass composite [23],  $\text{ZrO}_2\text{--Y}_2\text{O}_3\text{--La}_2\text{O}_3$  [24], LZ [25,26],  $\text{La}_2\text{Ce}_2\text{O}_7$  (LC) [27],  $\text{La}_2(\text{Zr}_{0.7}\text{Ce}_{0.3})_2\text{O}_7$  (LZ7C3) [28] and other rare earth oxides doped zirconia [29] have been investigated. Among these candidates, the rare earth zirconates and cerates were proposed as a promising TBCs material. However, LZ which was pyrochlore has a low thermal expansion coefficient (TEC) [30] and LC which was fluorite has a high sintering ability [31]. Doping  $\text{CeO}_2$  into LZ results in LZ7C3, which has a higher sintering resistance, high thermal expansion coefficient and low thermal conductivity ability. LZ7C3 has a mixture structure of pyrochlore and fluorite, and has high sintering-resistance due to its structural character of multiphase [32–34]. LZ7C3 is thermally stable after long-time annealing at 1573 K and no phase transformation. The linear thermal expansion coefficient of LZ7C3 is about  $10.66 \times 10^{-6} \text{K}^{-1}$ , similar to that of 8YSZ. The thermal conductivity of LZ7C3 is  $0.87 \text{ W m}^{-1} \text{ K}^{-1}$ , lower than that of 8YSZ and LZ [35]. The thermal conduction in

\* Corresponding author. Tel./fax: +86 431 85262285.

E-mail address: [xcao@ciac.jl.cn](mailto:xcao@ciac.jl.cn) (X. Cao).

ceramic materials can be explained by the theory of phonon scattering [36]. As the additional  $\text{Ce}^{4+}$  ion has larger ionic radius and relative atomic weight than  $\text{Zr}^{4+}$  ion, the LZC material has a lower thermal conduction than LZ due to the reduction of free path length of the phonons. Taking all the necessary properties especially the thermal expansion coefficient and toughness into consideration, 8YSZ was chosen as the other ceramic composition in the graded coating system.

In our former work [37,38], the thermal cycling behavior and failure mechanism of the LZ7C3/8YSZ:Eu dual layer coating systems has been studied. The dual layer LZ7C3/8YSZ coating system has a much longer thermal cycling lifetime than other double-ceramic-layer (DCL) coating systems with new materials. Therefore, the LZ7C3/8YSZ graded coating was designed and the thermal cycling test was conducted in our research.

In our work, LZ7C3 and 8YSZ were chosen as the ceramic material. Ni-based superalloy with a bond coat was chosen as the substrate. The substrate was coated by double-component ceramic functionally gradient coatings consisting 100% 8YSZ, 75% 8YSZ + 25% LZ7C3, 50% 8YSZ + 50% LZ7C3, 25% 8YSZ + 75% LZ7C3 as the interlayer, 100% LZ7C3 as the top coat. The schematic description of LZ7C3/8YSZ functionally graded TBCs was shown in Fig. 1.

## 2. Experimental

### 2.1. Synthesis of LZ7C3 powder

LZ7C3 powder was synthesized by a solid state reaction method.  $\text{La}_2\text{O}_3$  (99.99%, Shenghua Chemicals of Hunan, China),  $\text{ZrO}_2$  (Zr (Hf)  $\geq$  99.5%, Dongfang Chemicals of Guangdong, China),  $\text{CeO}_2$  (99.99%, Shenghua Chemicals of Hunan, China) were selected as the starting materials. Rare earth oxides powders ( $\text{La}_2\text{O}_3$ ,  $\text{CeO}_2$ , and  $\text{ZrO}_2$ ) were heat-treated at 1000 °C for 1 h in a box furnace because rare earth oxides are hygroscopic. These three powders were mixed together in a proper ratio. A deionized water-based suspension of this mixed powder was ball-milled for 24 h using zirconia balls. After mixing, the slurry was dried and heated at 1400 °C for 12 h to obtain the LZ7C3 powder. For spray drying, the as-synthesized LZ7C3 powder was ball-milled together with Arabic gum, tri-ammonium citrate ( $(\text{NH}_3)_3\text{C}_6\text{H}_5\text{O}_7$ ) and deionized water for 72 h using zirconia balls. Then the obtained slurry was spray-dried (Jiangsu Yangguang Ganzao Co., Ltd), leading to the formation of LZ7C3 powder with free-flowing for plasma spraying.

### 2.2. Preparation of coating

A five-layer ceramic(LZ7C3)–ceramic(8YSZ) FG-TBCs was atmospheric plasma sprayed on the Ni-based superalloy substrate (30 mm in diameter and 3 mm in thickness) with a Ni–23.7Co–20Cr–8.7Al–0.6Y–3.5Ta (wt%) bond coat (100  $\mu\text{m}$  in thickness), using the Sulzer Metco plasma-spraying unit with a F4-MB gun. The

composition of each layer in the LZ7C3/8YSZ graded coating systems are respectively 100% 8YSZ, 75% 8YSZ + 25% LZ7C3, 50% 8YSZ + 50% LZ7C3, 25% 8YSZ + 75% LZ7C3 as the interlayer, 100% LZ7C3 as the top coat. Two independently controlled powder feeders were used to spray the intermediate composite coatings by adjusting the feeding rates of LZ7C3 and 8YSZ in proper ratio, respectively. The average thickness of each ceramic layer in the coating was about 50  $\mu\text{m}$ , and total thickness of the ceramic coating is about 250  $\mu\text{m}$ . The disk-shaped substrate had a beveled edge to minimize the effect of stressed originated at the free edge of the specimens.

### 2.3. Thermal cycling tests

Thermal cycling tests were carried out on a burner-rig facility with a coal gas/oxygen flame. The sample surface was heated from room temperature to  $1250 \pm 50$  °C for 5 min followed by quenching to room temperature within 2 min by a compressed air jet, each cycle lasted 7 min. The cycling process was repeated again and again until 5% of the ceramic coating was lost.

### 2.4. Characterization

The phase structures were analyzed by X-ray diffraction (XRD) (XRD, Bruker D8 Advance) with  $\text{Cu K}\alpha$  radiation at a scan rate of 8°/min.

Thermal expansion coefficients (TECs) of bulk material of each ceramic layer (LZ7C3, 75%LZ7C3 + 25%8YSZ, 50%LZ7C3 + 50%8YSZ, 25%LZ7C3 + 75%8YSZ, 8YSZ) were recorded with a high temperature dilatometer (Netzsch 402C, Germany). The mixtures of LZ7C3 and 8YSZ powder in ratio which is the same with each coating layer were prepared. A deionized water-based suspension of these mixed powders was ball-milled for 24 h using zirconia balls. The stoichiometlets LZ7C3–8YSZ mixture was uniaxially pressed into pellets at 20 Mpa and further compacted by cold isostatic pressing with a pressure of 200 MPa for 5 min, and then pressureless sintered at 1600 °C for 12 h in air atmosphere.

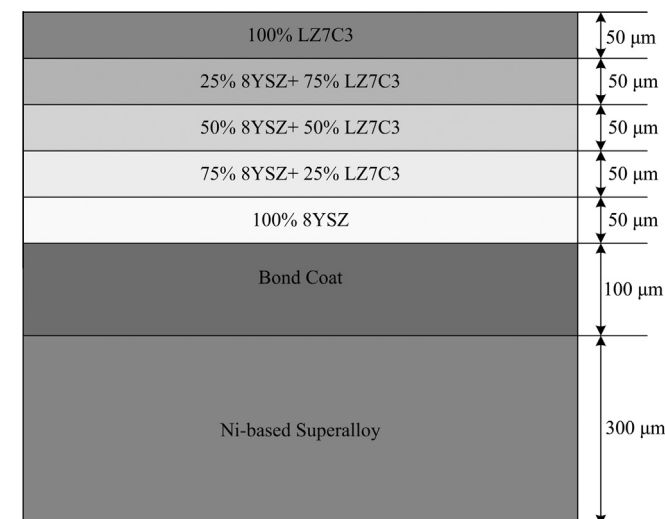


Fig. 1. Schematic description of LZ7C3/8YSZ functionally graded TBCs produced by atmospheric plasma spraying.

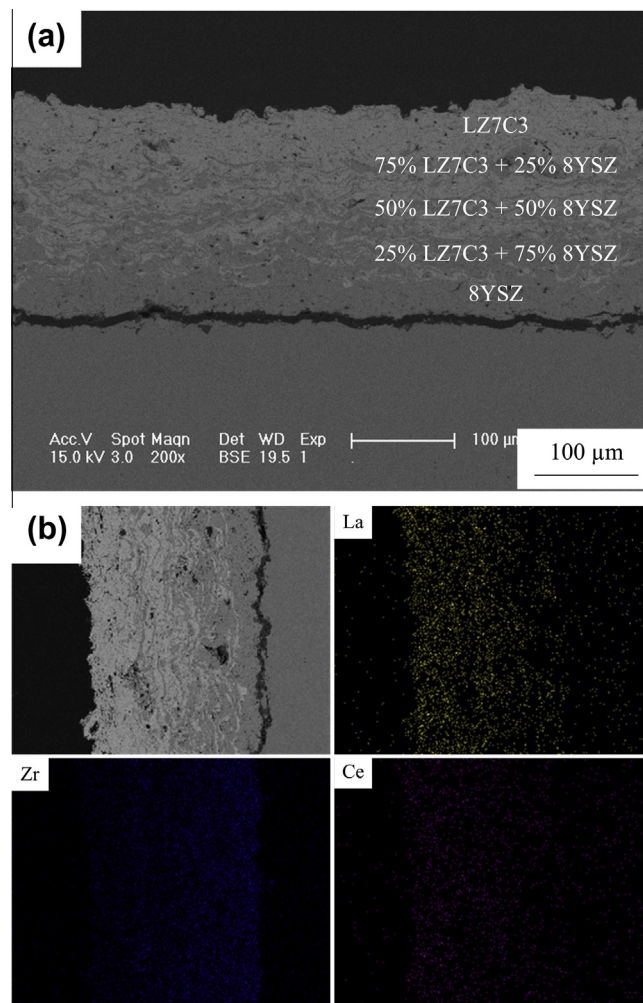


Fig. 2. The cross-sectional microstructure of the as-sprayed LZ7C3/8YSZ graded coating (a) and the corresponding elements distribution maps of the La, Ce and Zr (b).

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