

# A novel thermal barrier coating for high-temperature applications

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## Abstract

A new thermal barrier coating system based on  $\text{La}_{1.7}\text{Dy}_{0.3}\text{Zr}_2\text{O}_7$  (LDZ), which had a lower thermal conductivity for applications above 1773 K, was prepared by the air plasma spraying (APS). The phase composition, thermal expansion coefficient, thermal conductivity, the actual heat insulation and antioxidant ablation of the as-sprayed coatings were investigated. XRD results reveal that single pyrochlore phase LDZ coating is prepared and no new phase appears after ablation at 1573 K and 1773 K. Compared to 8 wt% yttria partially stabilized zirconia (YSZ) coatings, LDZ has better phase stability, lower thermal conductivity, better actual heat insulation, antioxidant ablation and heat resistance performance. These results imply that the LDZ ceramics can be explored as candidate material for the ceramic layer in TBC system.

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## 1. Introduction

Thermal barrier coatings (TBCs) are multilayered material systems deposited on metallic components of modern gas-turbine engines to thermally insulate them and to protect them against the hot and corrosive gas stream. TBCs consist typically of a ceramic top coat and a metallic bond coat [1].

The top coat works as thermal barrier by retarding the heat flow from hot gas to metallic substrate. For the next generation of advanced engines, further increase of gas turbine efficiency requires an even higher turbine inlet temperature. Up to now, the most successful TBC material in use is 6–8 wt% yttria partially stabilized zirconia (YSZ). However, it cannot be used for long-term application above 1473 K because of phase transformation and enhanced sintering [2–4]. In turn, the decomposition of tetragonal zirconia with the formation of monoclinic phase is usually accompanied by volume change and extended cracking.

As a result, a worldwide effort has been undertaken to identify new candidates for TBCs, including zirconates,

perovskites and hexa aluminates [5–8]. Among the interesting candidates for TBCs, lanthanum zirconate ( $\text{La}_2\text{Zr}_2\text{O}_7$ , LZ) with pyrochlore structure has received intense interest due to its low thermal conductivity and high phase stability [5,9]. So a number of investigations have been attempted to study the performance of LZ as a candidate for TBCs at high temperature [10–13]. However, some technical restrictions (relatively low thermal expansion coefficient, low fracture toughness) have been remarked for single LZ TBCs [3,14].

In recent studies, it has been reported that materials with lower thermal conductivity and higher thermal expansion coefficient can be prepared by doping with one or more oxides ( $\text{Yb}_2\text{O}_3$ ,  $\text{CeO}_2$ ,  $\text{Gd}_2\text{O}_3$ ,  $\text{Sm}_2\text{O}_3$ , and  $\text{Nd}_2\text{O}_3$ ) due to defect cluster formation, which indicates that the thermal conductivity and thermal expansion coefficient of  $\text{La}_2\text{Zr}_2\text{O}_7$  may be improved by doping with other elements in the cation of La or Zr [5,7,15,16].

Thus, it can be expected that  $\text{La}_{2-x}\text{Dy}_x\text{Zr}_2\text{O}_7$  may be a very promising TBC material. In our previous work, we have successfully prepared pyrochlore-type  $\text{La}_2\text{Zr}_2\text{O}_7$  nanocrystals and nanostructured lanthanum–zirconium coatings [17,18]. In this paper, we have prepared LDZ coating by the air plasma spraying (APS), the phase stability, the actual heat insulation

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Table 1  
Plasma spray parameters used for spraying bond coat and combustion synthesized LDZ powders.

Plasma spray parameters	YSZ	LDZ
Argon flow (NLPM)	40	45
Hydrogen flow (NLPM)	4	4.5
Amps (A)/volts (V)	600/60	600/64
Carrier gas flow (SCFH)	3	3
Powder feed rate (g/min)	35	25
Cooling air pressure (bar)	2.5	2.5
Spray distance (cm)	11	11
Gun speed (mm/s)	600	600

and the actual performance of LDZ ceramics were investigated in detail.

## 2. Experiment

LDZ and YSZ powders were synthesized by Molten Salts and hydrothermal method respectively. Then, LDZ and YSZ particles were reprocessed through spray drying to form granules with desired sizes in the range of 30–70  $\mu\text{m}$ . NiCrAlY bond coat was fabricated onto the substrates by air plasma spraying (APS-3000, Beijing, China), and then the LDZ coatings were deposited onto the bond coat using the same plasma spray system. The plasma spray parameters used are listed in Table 1.

Phase composition was identified by X-ray diffraction (XRD, X-Pert, Panalytic, Netherlands). The porosity of coatings was obtained by a mercury intrusion instrument (AUTOPORE II 9220 V3.04, USA). Microstructure of the coatings was observed via a Field Emission Scanning Electron Microscope (SEM, FEI Quanta 200 FEG, Netherlands). The antioxidant ablation and heat resistance behavior for LDZ and YSZ coatings were carried out on Oxygen kerosene HVOF spray systems (EvoCoat-LF HVOF Liquid Fuel Controller, Sulze Metco). The actual heat insulation properties were appraised by the temperature of the substrate. The temperature of the substrate was recorded with the Real-time Computer-based Temperature Acquisition System which developed by our laboratory.

## 3. Results and discussion

Phase constituent, structural stability and microstructure: The XRD pattern of LDZ coatings is shown in Fig. 1A. It can be observed that the prepared coating is composed of the single pyrochlore LZ phase and no other phases exist in the products. XRD pattern of the LDZ coating ablated at 1573 K and 1773 K for 300 s are shown in Fig. 1B and C. Compared with Fig. 1A, after high temperature ablation (Fig. 1B and C), no significant difference among the XRD patterns is observed, indicating that LDZ coating thermally stable in the temperature range of interest for TBCs applications at least up to 1773 K.

Fig. 2 shows the typical microstructure and surface photographs of as-prepared coatings (LDZ and YSZ). As can be

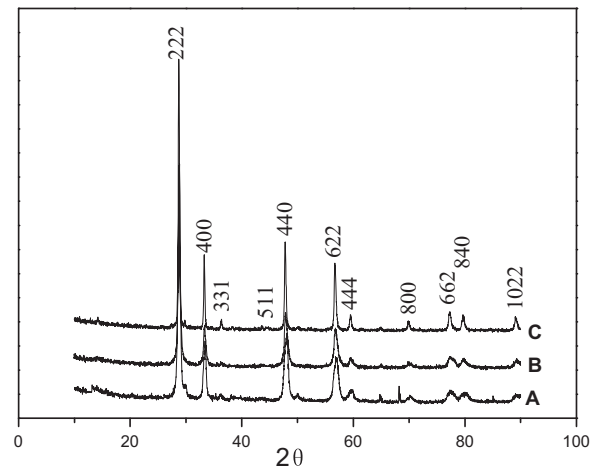


Fig. 1. XRD patterns: (A) LDZ as-prepared coatings, (B, C) are LDZ coatings being ablated for 300 s at 1573 K and 1773 K.

seen from Fig. 2, The surface of the as-prepared LDZ and YSZ coatings is smooth and the thickness is both about 0.52 mm. The measured surface-connected porosity of the LDZ and YSZ coatings is 7.72% and 7.02% respectively.

The thermal expansion coefficient (TEC) of  $\text{La}_{1.7}\text{B}_{0.3}\text{Zr}_2\text{O}_7$  coatings is presented in Fig. 3. The thermal expansion coefficient is proportional to the average distance between particles among the lattice, which is related to the strength of the ionic bonds [19]. The strength of the ionic bond is given in the following equation.

$$I_{A-B} = 1 - e^{\frac{\chi_A - \chi_B}{4}} \quad (1)$$

where  $I_{A-B}$  is the strength of the ionic bond between cations at sites A and B,  $\chi_A$  is the average electronegativity of cations at site A, and  $\chi_B$  is the average electronegativity of cations at site B. Therefore, the thermal expansion coefficients decrease with the electronegativity difference between cations at sites A and B decreasing. It can be seen that the TEC of LDZ coatings increases gradually with the temperature increases up to 1473 K, the value is  $10.3564 \times 10^{-6} \text{ K}^{-1}$  for LDZ. But there is a sudden decrease of the TEC at 586 K, just like the TEC of  $\text{La}_2\text{Ce}_2\text{O}_7$  [20]. For LZ, there are a large number of oxygen vacancies, and both the strength of vibration and the transverse vibration motions control the thermal expansion of the crystal.

Fig. 4 shows the typical microstructure and surface photographs of coatings (LDZ and YSZ) ablated at 1573 K for 300 s. Compared to Fig. 2, the surface of the coatings is still intact, no significant difference appeared through the microstructure after ablated at 1573 K for 300 s. This indicates that both LDZ and YSZ coatings all have very excellent antioxidant ablation, heat resistance performance and can protect the substrate effectively at 1573 K for 300 s.

The temperature of the substrate of coatings as a function of ablation time at 1573 K is plotted in Fig. 5. As shown in Fig. 5, the temperature of the substrate of YSZ coatings was about 1183 K, while that of LDZ coating was 1093 K, when the temperature reaches the balance. It indicated that LDZ coating has more excellent insulation properties than YSZ coatings.

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