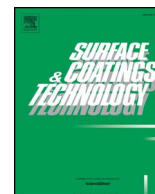




Contents lists available at ScienceDirect

## Surface &amp; Coatings Technology

journal homepage: [www.elsevier.com/locate/surfcoat](http://www.elsevier.com/locate/surfcoat)

# Fracture behavior and thermal durability of lanthanum zirconate-based thermal barrier coatings with buffer layer in thermally graded mechanical fatigue environments

Guanlin Lyu<sup>a</sup>, Bong Gu Kim<sup>a</sup>, Seoung-Soo Lee<sup>a</sup>, Yeon-Gil Jung<sup>a,\*</sup>, Jing Zhang<sup>b</sup>, Baig-Gyu Choi<sup>c</sup>, In-Soo Kim<sup>c</sup>

<sup>a</sup> School of Materials Science and Engineering, Changwon National University, Changwon, Gyeongnam 51140, Republic of Korea

<sup>b</sup> Department of Mechanical Engineering, Indiana University-Purdue University Indianapolis, Indianapolis, IN 46202, USA

<sup>c</sup> High Temperature Materials Research Group, Korea Institute of Materials Science, Changwon, Gyeongnam 51508, Republic of Korea

## ARTICLE INFO

## Keywords:

Thermal barrier coating  
Lanthanum zirconate  
Structural design  
Thermally graded mechanical fatigue test  
Thermal durability

## ABSTRACT

The effects of buffer layer on the fracture behavior and lifetime performance of lanthanum zirconate ( $\text{La}_2\text{Zr}_2\text{O}_7$ ; LZO)-based thermal barrier coatings (TBCs) were investigated through thermally graded mechanical fatigue (TGMF) tests, which are designed to simulate the operating conditions of rotating parts in gas turbines. To improve the thermal durability of LZO-based TBCs, composite coats consisting of two feedstock powders of LZO and 8 wt% yttria-doped stabilized zirconia (8YSZ) were prepared by mixing different volume ratios (50:50 and 25:75, respectively). The composite coat of 50:50 volume ratio was employed as the top coat, and two types of buffer layers were introduced (25:75 volume ratio in LZO and 8YSZ, and 8YSZ only). These TBC systems were compared with a reference TBC system of 8YSZ. The TGMF tests with a tensile load of 60 MPa were performed for 1000 cycles, at a surface temperature of 1100 °C and a dwell time of 10 min, and then the samples were cooled at room temperature for 10 min in each cycle. For the single-layer TBCs, the composite top coat showed similar results as for the reference TBC system. The triple-layer coating (TLC) showed the best thermal cycle performance among all samples, suggesting that the buffer layer was efficient in improving lifetime performance. Failure modes were different for the TBC systems. Delamination and/or cracks were created at the interface between the bond and top coats or above the interface in the single-layer TBCs, but the TBCs with the buffer layer were delaminated and/or cracked at the interface between the buffer layer and the top coat, independent of buffer layer species. This study allows further understanding of the LZO-based TBC failure mechanisms in operating conditions, especially in combined thermal and mechanical environments, in order to design reliable TBC systems.

## 1. Introduction

Thermal barrier coating (TBC) systems have been widely applied in hot section parts of gas turbine engines to provide thermal protection to metallic components from high temperature service environments [1,2]. Obviously, TBCs protect the underlying substrate against the high temperature oxidation and corrosion, enhance the operational life of hot section components, and increase the fuel efficiency of gas turbine engines by improving turbine inlet temperature (TIT). The fracture behavior and thermal durability of TBC systems are usually investigated in either thermal cycling or mechanical loading. Especially, in the case of moving parts such as turbine blades, not only is a thermal gradient generated between the heating and cooling surfaces of a blade, but the

centrifugal force also increases due to the rapid rotational speed of 3600 rpm [3]. Thus, the TBC system employed on turbine blades is subjected to both thermal and mechanical stresses, which may easily cause premature failures. Therefore, the lifetime performance of the TBC system, especially in the case of moving parts, should be evaluated in dual environments.

TBC systems usually consist of a superalloy substrate, an oxidation-resistant metallic bond coat, and a ceramic top coat. The classic top coat in a TBC system is 7–8 wt% yttria-stabilized zirconia ( $\text{ZrO}_2$ , 7–8YSZ) which has a high melting point (2680 °C) [4], relatively low thermal conductivity (2.0–2.3 W/m/K at ~1000 °C for a fully dense sample; 0.9–1.2 W/m/K for 10–15% porosity) [5–7], and favorable thermal stability [8,9]. As the TIT is continuously increasing in gas turbines,

\* Corresponding author.

E-mail address: [jungyg@changwon.ac.kr](mailto:jungyg@changwon.ac.kr) (Y.-G. Jung).

<http://dx.doi.org/10.1016/j.surfcoat.2017.09.066>

Received 27 April 2017; Received in revised form 27 August 2017; Accepted 2 September 2017  
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there is a limitation in application of 8YSZ-coating systems, especially above 1200 °C. YSZ transforms from the  $t'$  phase to the tetragonal and cubic phases ( $t$  and  $c$  phases, respectively) by repetitive heating and cooling process above 1200 °C, and then to the monoclinic ( $m$ ) phase with a volume expansion of about 3–5 vol% in further operation, resulting in the spallation or delamination of the coating systems [4,7]. To solve these problems, lanthanum zirconate ( $\text{La}_2\text{Zr}_2\text{O}_7$ , LZO) has been proposed as a new material for TBC application. Compared to 8YSZ, LZO has a lower conductivity (1.5–1.8) W/m/K at 1000 °C, no phase transition from room temperature to melting point, a lower oxygen ion diffusivity, which protects the bond coat and the substrate from oxidation [4,9], a lower coefficient of thermal expansion (CTE) ( $9.1 \times 10^{-6}$ – $9.7 \times 10^{-6} \text{ K}^{-1}$  for LZO, compared to  $10.5 \times 10^{-6}$ – $11.5 \times 10^{-6} \text{ K}^{-1}$  for YSZ, bulk materials and coatings at 30–1000 °C), and lower sintering ability [10].

LZO does have shortcomings, such as lower CTEs and mechanical properties than 8YSZ. TBC systems with LZO can be used if the CTE of the LZO is selected to give a TBC system that is similar to a system with an 8YSZ coating, and the mechanical properties of the TBC systems with LZO are improved to be comparable with those of TBC systems with 8YSZ. However, there is no single material that satisfies all requirements for TBCs. Multilayer concepts have been put forward, which include an erosion-resistant layer as an outer layer, a thermal barrier layer, a corrosion–oxidation-resistant layer, a thermal stress control layer, and a diffusion-resistant layer [11]. However, multilayered structures can produce artificial defects at the layer interfaces, which can cause delamination or cracking. Therefore, a double-layer top coat (DLC) system, based on the multilayer concept, has been shown to be an effective method to meet the demands of developing TBC systems [10,12]. The DLC includes a ceramic top coat layer, a ceramic buffer layer, a bond coat layer, and a superalloy substrate. The buffer layer is introduced between the top and bond coats, which can reduce stresses generated at the interface due to the CTE mismatch between the top and bond coats during cyclic thermal exposure. In addition, in a previous study [13], layered LZO-based TBCs were already investigated through furnace cyclic thermal fatigue, thermal shock, and jet engine thermal shock tests. However, these methods evaluated the thermal durability in the cyclic thermal exposure, but not both thermal and mechanical stress environments.

In this study, LZO-based TBC systems with a layered structure in top coat were designed using blended feedstock of LZO and 8YSZ powders prepared by mixing in different volume ratios. Three types of the layered LZO-based TBCs were tested at room temperature and 1100 °C under a tensile load of 60 MPa to simulate a gas turbine blade subject to centrifugal force and high temperature gas flow, using a thermally graded mechanical fatigue (TGMF) test. The thermal durability and fracture behavior of the layered LZO-based TBCs were investigated and compared with those of the single-layer TBCs. The effects of the buffer layer structure and species at the interface on the lifetime performance of TBCs were also investigated through TGMF tests.

## 2. Experimental procedure

### 2.1. Starting materials and sample preparation

The substrate was an approximately 5.5 mm thick nickel-based superalloy (Hastelloy-X, nominal composition of 47Ni–22Cr–18Fe–9Mo, in wt%). The dimensions of the sample are shown in Fig. 1(A). The surface was blasted using alumina powder with a particle size of 60 mesh and then cleaned before deposition. The bond coat was prepared by an air-plasma spray (APS) method (9 MB; Sulzer Metco Holding AG, Switzerland) using an AMDRY 962 (hereinafter 962, nominal composition of Ni–22Cr–10Al–1.0Y in wt% and particle size of 56–106  $\mu\text{m}$ ; Sulzer Metco Holding AG). The feedstock for the top coat was blended with 8YSZ and LZO. These powders were employed as METCO 204C-NS (hereinafter 204C-NS, particle size of 45–125  $\mu\text{m}$ ;

Sulzer Metco Holding AG) and LAO-109-1 (Praxair Surface Technologies, Indianapolis, USA), respectively. Surface and side photographs of the as-prepared LZO-based TBC sample is shown in Fig. 1(B). The blended powders of 8YSZ and LZO were prepared by mixing 50:50 and 75:25 volume ratios for the top coat and buffer layers, respectively. Note that the buffer layer was introduced in the LZO-based TBC for reducing thermal mismatch between the bond and top coats. The thicknesses of the bond and top coats were designed to be 150  $\mu\text{m}$  and 430  $\mu\text{m}$ , respectively. Three kinds of LZO-based TBCs were prepared, including a single-layer top coat (SLC), a DLC, and a triple-layer top coat (TLC) with double buffer layers. The SLC consists of the bond coat and the top coat with 50:50 volume ratio in LZO and 8YSZ, the DLC consists of the bond coat, the buffer layer with 8YSZ (204C-NS), and the top coat with 50:50 volume ratio, and the TLC consists of the bond coat, the two buffer layers with 8YSZ (204C-NS) and with 50:50 volume ratio in LZO and 8YSZ, and the top coat with 50:50 volume ratio. A reference sample was designed using 8YSZ feedstock powder to compare with LZO-based TBCs. Furthermore, the top coat of the reference sample was deposited on the bond coat by using METCO 204C-NS (particle size of 45–125  $\mu\text{m}$ ; Sulzer Metco Holding AG) with an APS method. The feedstock powder of the bond coat for the reference sample was AMDRY 997 (nominal composition of Ni–22Co–22Cr–10Al–1.0Y in wt% and particle size of 56–106  $\mu\text{m}$ ; Sulzer Metco Holding AG), and the bond coat was prepared by using a vacuum plasma spray (VPS) process. The thicknesses of the top and bond coats were designed as 600 and 150  $\mu\text{m}$ , respectively. The schematic diagrams of the structural designs and its cross-sectional microstructures of as-prepared samples are shown in Fig. 2.

### 2.2. TGMF test

The delamination and fracture behaviors of TBCs in a combined thermal and mechanical environments, particularly for a turbine blade with a specific amount of the mechanical and thermal stresses, were investigated through TGMF tests. The apparatus, schematic diagram, and test condition of the TGMF test are shown in Fig. 3(A), (B), and (C), respectively. A uniaxial tensile load ( $\delta_a$ ) of 60 MPa was applied to a sample with a rate of 1.5 mm/min and strain was controlled with a mechanical strain range of 0.2%. The applied stress  $\delta_a$  is defined as  $\delta_a = P_a/S_0$ , where  $S_0$  is the entire cross-sectional area of the coated sample ( $S_0 = w (h_{\text{TBC}} + h_{\text{BC}} + h_{\text{S}})$ ), and  $h_{\text{TBC}} = 430 \mu\text{m}$ ,  $h_{\text{BC}} = 150 \mu\text{m}$ , and  $h_{\text{S}} \approx 5.5 \text{ mm}$  are the thicknesses of the top coat, bond coat, and substrate, respectively. TGMF tests were performed for 1000 cycles at a surface temperature of 1100 °C for a dwell time of 10 min, using liquefied petroleum gas, and then the sample was cooled at room temperature for 10 min in each cycle. The surface temperature of sample at the center and edge regions of flame was different due to the localized flame. Therefore the temperatures at the center and edge were measured by a R-type thermal couple and Infrared-temperature measurement device (Wavelength: 3.9  $\mu\text{m}$ , CTlaser MT, Optris, Germany), respectively. The criteria that were adopted for failure in the TGMF tests were > 50% spalling of the region, or cracking in the top coat and/or at the interface.

### 2.3. Characteristics

For microstructure analysis before and after TGMF testing, the samples were divided into three portions due to the difference in thermal fatigue phenomenon. In this investigation, the sectional views of chosen portions are perpendicular and parallel to the tensile load applied to the samples. The selected portions of samples were pre-processed to observe the cross-sectional microstructure before and after the TGMF tests. The samples were cold-mounted using epoxy resin, polished using silicon carbide paper and then final polished using 3  $\mu\text{m}$  and 1  $\mu\text{m}$  diamond pastes. The cross-sectional microstructures of the TBC samples were observed using a scanning electron microscope

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