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Novel thermal barrier coatings based on $La_2Ce_2O_7$/8YSZ double-ceramic-layer systems deposited by electron beam physical vapor deposition

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Abstract

Novel thermal barrier coatings based on $La_2Ce_2O_7$/8YSZ double-ceramic-layer (DCL) systems, which were deposited by electron beam physical vapor deposition (EB-PVD), were found to have a longer lifetime compared to the single layer $La_2Ce_2O_7$ (LC) system, and even much longer than that of the single layer 8YSZ system under burner rig test. The DCL coating structure design can effectively alleviate the thermal expansion mismatch between LC coating and bond coat, as well as avoid the chemical reaction between LC and Al_2O_3 in thermally grown oxide (TGO), which occurs above 1000 °C as determined by differential scanning calorimetry (DSC) analysis. The failure mechanism of LC/8YSZ DCL coating is mainly due to the sintering of LC coating surface after long-term thermal cycling.

Keywords: Thermal barrier coatings; CeO_2; ZrO_2; Electron beam physical vapor deposition

1. Introduction

Thermal barrier coating (TBC) systems typically consist of four components: superalloy substrate, metallic bond coat, TGO layer and ceramic topcoat. A metallic bond coat layer protects the substrate from oxidation and corrosion, as well as improves the bonding strength between the substrate and the ceramic topcoat. The ceramic topcoat is deposited on the bond coat as an insulation layer to protect the underlayers against temperature attack at high inlet temperatures of turbine engines. The TGO layer is developed during operation, which mainly consists of metallic oxides (mainly Al_2O_3) and can effectively reduce the oxidation rate of the bond coat. During the last decade, a lot of efforts were devoted to the development and manufacturing of ceramic TBCs for turbine parts, because the traditional turbine materials have reached the limits of their temperature capabilities. The state-of-the-art topcoat material, which is being used for TBC applications, is yttria partially stabilized zirconia (YSZ) [1,2]. YSZ performs well up to about 1200 °C, but it cannot be used above 1200 °C because of sintering and phase transformations, which lead to the reduction of strain tolerance in combination with an increase of the Young's modulus and a volume change during cooling [3,4]. As a consequence, a considerable reduction of thermal cycling lifetime is observed.

Some new ceramic materials, such as $LaMgAl_{11}O_{19}$, ZrO_2-Y_2O_3-La_2O_3, metal-glass composite, 7.5 mass% Y_2O_3-HfO_2 (7.5YSH) oxides, $La_2Zr_2O_7$ (LZ), $La_2Ce_2O_7$ (LC) and other rare earth doped zirconia, have been evaluated as potential TBC materials to further increase the operation temperature of turbine engines [5–12]. For selection of good TBC materials there are some important requirements which must be satisfied like low thermal conductivity, minimal coefficient of thermal expansion (CTE) difference, high melting point, good phase stability, low sintering rate, and low Young's modulus. No single material has satisfied all requirements for TBCs, however, it seems that the layered topcoat can overcome the shortcoming (short thermal cycling lifetime) of many new TBC materials [13–15].

The LC is used as an outer layer due to its lower thermal conductivity and good phase stability at high temperatures

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compared to 8YSZ. In this work, a newly developed DCL coating of 8YSZ and LC deposited by EB-PVD is presented. The thermal cycling behavior of the DCL coating was evaluated by the use of a burner rig test. The properties of single ceramic layer coatings of 8YSZ and LC are also presented and compared, as well as the failure mechanisms of the coatings.

2. Experimental procedure

Powders for preparing ingots for evaporation used in this work were La_2O_3, CeO_2, ZrO_2 and Y_2O_3 (purity 99.99%, Grirem Advanced Materials Co. Ltd., Beijing, China).

A 150-kW four-electron-gun EB-PVD equipment (Model UE205, Paton Electric Welding Institute, Kiev, Ukraine) with three water-cooled copper crucibles was used for preparing the TBCs. A Ni-based superalloy was chosen to be the substrate material and its chemical composition is given in the previous paper [16]. The substrate specimens for thermal cycle tests were machined into disks with diameter of 30 mm and 3 mm thickness [17]. NiCoCrAlY ingot was used as evaporation source of the bond coat, while LC and 8YSZ ingots sintered at 1550 °C for 2 h with diameter of 50 mm and 150 mm length were used as evaporation sources for the ceramic topcoats. The chemical composition of the NiCoCrAlY ingot is Ni-20Co-20Cr-8Al-1Y and for its coating is Ni-24Co-25Cr-8Al-1Y in wt.%. A NiCoCrAlY bond coat was first deposited onto the substrate by EB-PVD followed by vacuum heat treatment at 1000 °C for 4 h prior to the deposition of the ceramic topcoat. The 8YSZ coating was first deposited onto the bond coat followed by LC coating. The thicknesses of both 8YSZ and LC coating are about 40 μm and 70 μm, respectively. During deposition, the average substrate temperature was adjusted to 950 ± 25 °C. The substrates were then rotated at 14 rpm and no oxygen was introduced into the operation chamber.

Crystal structure was characterized by X-ray Diffraction (XRD) (Model D/MAX 2200, Rigaku Co. Ltd., Tokyo, Japan) at a wavelength of 1.5406 Å. To evaluate the thermal shock resistance of the coatings, thermal cycle tests were carried out with a methane/oxygen flame, and the coating surface was cycled from room temperature to 1250 ± 10 °C within 20 s. The total heating time was 5 min, followed by quenching to room temperature within 90 s by a cooling air jet from both sides of the sample. The peak temperature of the substrate was 970 ± 10 °C. The surface temperature was measured with an infrared pyrometer while the substrate temperature was measured using a NiCr/Ni thermocouple inside a hole in the specimen. Cycling test was stopped when obvious degradation of the coatings (delamination failure, large cracking) occurred. The cycling number was then defined as the lifetime of the TBCs.

The coating structure and composition were investigated by a scanning electron microscope (SEM) (Model Hitachi S-3500N, Hitachi Ltd, Tokyo, Japan) with energy dispersive spectrometer (EDS) (Model Inca, Oxford Instruments, Oxfordshire, United Kingdom). The chemical compatibility of LC and Al_2O_3 mixed powders was analyzed by a simultaneous thermal analysis apparatus (Model STA 449C, Netzsch, Selb, Germany). The weight fraction of the two component powders in the mixture is 50%.

3. Results and discussion

LC is a solid solution of La_2O_3 in CeO_2 with fluorite type structure [18]. The XRD pattern is similar to that of CeO_2 with a small change of the lattice parameter (CeO_2, $a=5.4113$ Å; $La_2Ce_2O_7$, $a=5.5718$ Å). Fig. 1 shows the XRD patterns of LC coating in as-deposited state (Fig. 1(a)) and after 300 cycles (Fig. 1(b)), as well as LC/8YSZ DCL coating after 2007 cycles (Fig. 1(c)). The XRD pattern of LC coating after 300 cycles is unchanged compared to that of as-deposited one, indicating that no new phases formed. As to the LC/8YSZ DCL coating, there are some new diffraction peaks which appeared after 2007 cycles as shown in Fig. 1(c). La_2O_3 and CeO_2 phases were observed due to the partial decomposition of the LC coating, which is attributed to the chemical composition deviation of LC coating from $La_2Ce_2O_7$ upon deposition by EB-PVD [11,16]. The different vapor pressures of La_2O_3 (8×10^{-5} atm, 2500 °C) and CeO_2 (2×10^{-2} atm, 2500 °C) lead to the chemical composition deviation of LC coating from that of the ingot during the EB-PVD process [19]. Some ZrO_2 diffraction peaks are also observed in Fig. 1(c), which is due to the partial spallation of LC coating resulting in the exposure of 8YSZ coating into the air. It is should be noted that the small m-ZrO_2 diffraction peak appeared, which means that the phase transformations occur for 8YSZ coating during thermal cycling.

The surface morphology and the cross-section micrographs of LC/8YSZ DCL coating after 2007 cycles are shown in Fig. 2. About 4.5% of surface area of DCL coating spalled after 2007 cycles (Fig. 2(a)), but only the LC coating partially spalled from the 8YSZ coating surface, which can be clearly seen in Fig. 2(d). The surface micrograph of DCL coating after thermal cycling (Fig. 2(b)) reveals that the LC coating surface separated into some small isolated areas, and some of the isolated areas also spalled. This is caused by the sintering of LC coating during thermal cycling. The sintering effect of LC coating also can be seen in Fig. 2(c), about 10 μm thickness of LC coating near the surface has no clear column structure, which is the intrinsic microstructure of EB-PVD coatings. However, the

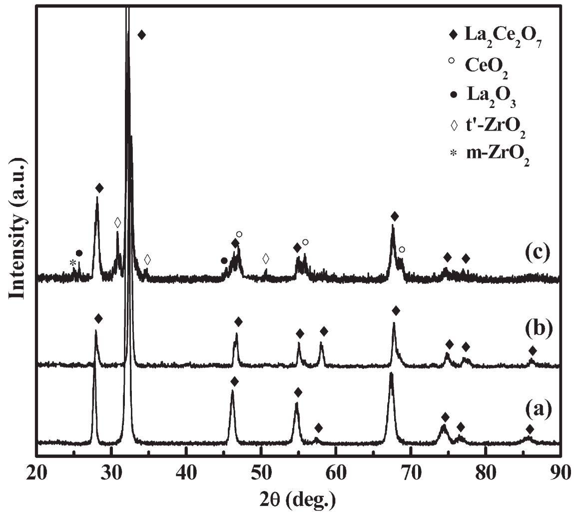

Fig. 1. XRD patterns of LC coating (a) as-deposited, (b) after 300 cycles, and LC/8YSZ DCL coating (c) after 2007 cycles.