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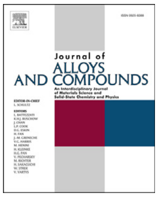

$SrCeO_3$ as a novel thermal barrier coating candidate for high–temperature applications

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ABSTRACT

In this study, perovskite-type $SrCeO_3$ was proposed and investigated as a novel thermal barrier coating (TBC) material for applications above 1200 °C. Results indicated that $SrCeO_3$ has a low thermal conductivity, decreasing from 2.84 W/(m•K) to 1.74 W/(m•K) with increasing temperatures from room temperature to 800 °C. The thermal expansion coefficients are in a range of $9.67 \times 10^{-6}\ K^{-1}$-$10.6 \times 10^{-6}\ K^{-1}$, which are comparable to those of the traditional yttria stabilized zirconia (YSZ). The results of thermogravimetric-differential scanning calorimeter (TG-DSC) analyses and long-term annealing tests indicated that $SrCeO_3$ has excellent phase stability from room temperature to 1300 °C. The above merits suggested that $SrCeO_3$ has a good potential for TBC applications at temperatures higher than 1200 °C. Therefore, plasma sprayed $SrCeO_3$ coating was further produced and characterized. However, coated samples failed very early during thermal cycling. The low fracture toughness (1.09 MPa $m^{1/2}$), high sintering activity and coating composition deviation are deemed to be the major factors which lead to the early spallation of $SrCeO_3$ coating. Finally, methods for the improvement of the thermal shock life of $SrCeO_3$ TBC were proposed.

1. Introduction

In the past few decades, thermal barrier coatings (TBC) have been increasingly applied in aero and land-based industrial turbines to protect their hot-section metallic components (e.g. transition pieces, combustion lines, first-stage blades and vanes) against oxidative and corrosive attacks [1]. It is reported that a large temperature drop (~100 °C) can be achieved across the ceramic layer with the application of TBC system, thereby remarkably enhancing the durability of the metallic components. On the other hand, the use of TBC can also allow higher gas inlet temperatures at a given metal temperature, leading to a sharp rise in engine efficiency [2–5].

Generally, TBC is a duplex system, at least consisting of two layers—a bondcoat layer and an insulating ceramic topcoat. The bondcoat is often a metal and has two major functions. It alleviates thermal stresses producing from thermal expansion mismatch, as well as improves the bonding strength between the substrate and the topcoat. The upper layer is a ceramic topcoat with the prime function to provide heat insulation by its low thermal conductivity and it must withstand the most extreme temperature, steep temperature gradients, fast temperature transients and complex stress conditions [6]. Therefore, the selection of topcoat materials is constrained by some basic requirements: (1) high temperature capability (>2000 K); (2) low thermal conductivity (<2.5 W/(m•K)); (3) phase stability between room temperature and operating temperature; (4) moderate thermal expansion coefficient closed to that of the metallic substrate ($>10 \times 10^{-6}\ K^{-1}$); (5) poor oxygen ion conductor; (6) in good conjunction with the metallic substrate and (8) sintering resistance of the porous microstructure; (9) superior fracture resistance [7]. Nowadays, state-of-the-art topcoat is zirconia partially stabilized with 6–8 wt% yttria (6–8 YSZ) fabricated either by atmospheric plasma spraying (APS) or by electron beam physical vapor deposition (EB-PVD) [8–10]. It possesses high thermal expansion coefficient (TEC ~$11.3 \times 10^{-6}\ K^{-1}$) and low thermal conductivity (~2.1 W/(m•K)) at elevated temperatures [11,12]. However, when YSZ coating is used at temperatures over 1200 °C for long-term applications, diffusion-induced phase

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transformations occur from metastable tetragonal phase (t'-phase) to tetragonal phase (t-phase) and cubic phase (c-phase). Subsequently, monoclinic phase (m-phase) emerges which would give rise to a volume change and possibly result in the formation of cracks in the coating [13,14]. Moreover, YSZ coating experiences an accelerated sintering above 1200 °C, which is accompanied by a reduction in porosity and an increase in Young's modulus. The serious sintering and phase transformation limit the long-term service of YSZ coating over 1200 °C. Thus, relentless efforts have been made to explore alternative materials for high-temperature performance above 1200 °C.

Within the past few years, some promising TBC materials have been evaluated, such as: rare-earth oxides stabilized zirconia [15,16], pyrochlore type ($Re_2Zr_2O_7$ (Re= La→Gd)) [17,18], fluorite type ($La_2Ce_2O_7$) [19,20], magnetoplumbite type ($LaMgAl_{11}O_{19}$) [21], rare-earth phosphates ($LnPO_4$, Ln = rare earth element) [22,23] and perovskites type ($SrZrO_3$, $BaZrO_3$, $BaLa_2Ti_3O_{10}$, $La(Al_{1/4}Mg_{1/2}Ta_{1/4})O_3$) [24–27] etc. Among these potential materials, the perovskite-type oxides (ABO_3) are characterized by high melting points, low Young's modulus and high thermal expansion coefficients. $SrZrO_3$ and $BaZrO_3$ were the two perovskite materials which were firstly evaluated as TBC candidates [28]. $BaZrO_3$ has a high melting temperature of 2600 °C, but the thermal service lifetime of $BaZrO_3$ is minimized by relatively low TEC, interior chemical stability as well as relatively high thermal conductivity. While $SrZrO_3$ was found to be a promising TBC candidate for high-temperature applications above 1250 °C with respect to its high melting temperature (2800 °C), relatively low thermal conductivity, and high TEC of ~10.9×10^{-6} K^{-1} (30–1000 °C) [28]. Besides, the sintering rate and Young's modulus of $SrZrO_3$ are lower than those of YSZ as well, which are beneficial to mechanical responses. Thermal cycling test performed with a gas burner showed that the $SrZrO_3$/YSZ double-ceramic-layer (DCL) coating had a lifetime more than 3200 cycles at 1250 °C, which was comparable to that of typical YSZ coating [29]. At operating temperatures higher than 1300 °C, the $SrZrO_3$/YSZ DCL coating even showed a better performance than YSZ coating. Unfortunately, it has been reported that $SrZrO_3$ undergoes temperature-induced phase transformations at intermediate temperatures, which has a detrimental effect on the performance of $SrZrO_3$ TBC [29]. The structure transition of $SrZrO_3$ is summarized as [30]:

$$\mathrm{Orthorhombic(Pnma)} \xrightarrow{1023K} \mathrm{pseudo-tetragonal(Imma)} \xrightarrow{1113K} \mathrm{Tetragonal(I4/mcm)} \xrightarrow{1403K} \mathrm{cubic}\left(\mathrm{Pm\bar{3}m}\right)$$

The aforementioned disadvantage of $SrZrO_3$ under thermal cyclic conditions leads to an intense search for new TBC materials in perovskites. S. Yamanaka et al. and M.A. Prévost have systematically studied the thermochemical and thermophysical properties of Ba and Sr series perovskites [31,32]. Based on their results, $SrCeO_3$ showed a glass-like thermal conductivity. The theoretical minimum thermal conductivity of $SrCeO_3$ obtained from semi-empirical molecular dynamic calculation is around 1 W/(m·K), which is much lower than that of $SrZrO_3$. Besides, $SrCeO_3$ also showed a larger TEC (~11.1×10^{-6} K^{-1}) than $SrZrO_3$, which is comparable to that of YSZ. Unlike $SrZrO_3$, $SrCeO_3$ was reported to have excellent phase stability and no phase transformation occurs from room temperature to its melting point [33]. Those preliminary results indicate that $SrCeO_3$ have potential for TBC applications. Thus the phase stability and related thermal properties of $SrCeO_3$ definitely need to be fully understood. However, to the best of our knowledge, $SrCeO_3$ has not been evaluated as a coating and its properties as TBC material have not been systematically reported in literature.

In this paper, $SrCeO_3$ was proposed as a novel TBC material. The thermophysical properties, phase stability and mechanical properties of $SrCeO_3$ were investigated. In addition, $SrCeO_3$ coating was prepared via APS, and its thermal cycling durability was evaluated. The associated failure mechanism was also studied.

2. Experimental

The two commercially available powders of $SrCO_3$ (purity ≥ 99.0%) and CeO_2 (purity ≥ 99.99%) were selected as original materials to synthesize $SrCeO_3$ powders by solid-state reaction. The two powders in proper ratio were ball-milled by zirconia milling balls and were heated at 1200 °C for 12 h to obtain the final product. The as-synthesized powders mixed with Arabic gum (adhesion agent), ammonium citrate (dispersing agent) and deionized water were subjected to ball-mill mixing again and were later spray-dried (SFOC-16, Shanghai-Ohkawaa Dryers Co., Ltd.). The free–flowing $SrCeO_3$ powders with particle size of 32–125 μm were collected for plasma spraying.

The resultant $SrCeO_3$ powders were pressed into pellets and bars by the cold isotropic pressing (CIP) method under a pressure of 200 MPa. Some green pellets were exposed at different temperatures (1100 °C, 1200 °C, 1300 °C, 1400 °C and 1500 °C) for 6 h to analyze the sintering resistance by measuring volume shrinkages. Those remaining compacts were pressureless-sintered at 1500 °C for 20 h in air for densification and afterward the bulk density was measured on the basis of Archimedes' method. When the density of $SrCeO_3$ ceramic reaches up to 95% of theoretical density, mechanical and thermophysical properties were evaluated. The sintered specimens were thermally etched at 1300 °C for 2 h in air for microstructural observations. The YSZ and $SrCeO_3$ powders were isothermally aged at 1300 °C for 12 h, 24 h, 36 h, 48 h, 96 h, respectively, to investigate phase stability. A homogeneous mixture of Al_2O_3 and $SrCeO_3$ powders in a molar ratio of 1:1 was also heated at 1200 °C for 12 h to study the chemical compatibility of $SrCeO_3$ against thermally grown oxide (TGO).

A metallic bond coat (Ni-25.17Cr-5.03Al-0.51Y (wt%)) was sprayed onto Ni-based superalloy (DZ125, 30 mm × 12 mm × 2 mm) via APS (Sulzer-Metco F4 gun, Switzerland) technique, about 150 μm in thickness. The porous $SrCeO_3$ ceramic coating (typically about 200 μm thick) was then deposited on the substrates by APS method, the detailed spray parameters are listed in Table 1. Prior to spraying, the substrates were primarily grit blasted with alumina particles for adhesion enhancement. Additionally, free-standing coating was also fabricated on the graphite substrates with a thickness of about 1 mm. After burning off the graphite substrates, the individual layer coating was cut into small bars for measuring phase stability.

Table 1
Processing parameters for spraying $SrCeO_3$ coating.

coatings	Current (A)	Power (kW)	Distance (mm)	Ar/H_2 (slpm)[a]	Feeding rate (g/min)	Gun velocity (mm/s)
Bond coat	530	42	100	35/12	17	800
Top coat	430	30	100	35/12	18	800

[a] slpm: standard liter per minute.