Journal of the European Ceramic Society xxx (2016) xxx–xxx

Contents lists available at www.sciencedirect.com

Journal of the European Ceramic Society

journal homepage: www.elsevier.com/locate/jeurceramsoc

Dominant effect of particle size on the CeO_2 preferential evaporation during plasma spraying of $La_2Ce_2O_7$

Li-Shuang Wang, Shan-Lin Zhang, Tao Liu, Chang-Jiu Li*, Cheng-Xin Li, Guan-Jun Yang

State Key Laboratory for Mechanical Behavior of Materials, School of Materials Science and Engineering, Xi'an Jiaotong University, Xi'an, Shaanxi 710049 China

ARTICLE INFO

Article history:
Received 21 August 2016
Received in revised form 1 November 2016
Accepted 8 November 2016
Available online xxx

Keywords:
Atmospheric plasma spraying
$La_2Ce_2O_7$
Powder particle size
Thermal barrier coatings (TBCs)
Preferential evaporation

ABSTRACT

The inhomogeneous composition distribution of atmospheric plasma sprayed $La_2Ce_2O_7$ limits its operation in thermal barrier coatings. Two types of $La_2Ce_2O_7$ powders were used to investigate the effect of the powder size and structure on the preferential evaporation of Ce during plasma spraying. The results indicate that powder particle size is dominant for the Ce evaporation loss. Such effect is significantly intensified with the decrease of particle size. When the particle size is larger than 30 μm, the Ce loss is less or can be neglected. Two mechanisms including the element diffusion mechanism and convection mass transfer mechanism are proposed to well explain the evaporation rule. Moreover, the loss of Ce is higher using the agglomerate-sintered powder due to its higher thermal conductivity than the as-agglomerated powder. Correspondingly, the CeO_2 loss can be neglected in the coatings as the agglomerated powders with a size <30 μm were used.

1. Introduction

Thermal barrier coatings (TBCs) have been widely used to protect metallic components of gas turbines from exposure to high temperature. TBCs together with internal cooling components could reduce the surface temperature of the superalloy by about 100–300 °C [1]. Yttria partially stabilized zirconia (YSZ) is widely used as TBCs material due to the low thermal conductivity and excellent mechanical property [1,2]. However, the high operation temperature of the next generation TBCs limits the long-term reliable operation of YSZ, due to the phase transformation [3–5]. Therefore, in recent years, some rare earth ceramic materials with fluorite structure, perovskite structure or pyrochlore structure are considered as promising candidates. These materials include $LaMgAl_{11}O_{19}$ [6,7], $La_2Zr_2O_7$ (LZO) [8,9], $Gd_2Zr_2O_7$ (GZO) [10,11], $SrZrO_3$ [12,13] and $La_2Ce_2O_7$ (LCO) [14,15], which have been widely investigated due to their phase stability and low thermal conductivity above 1473 K. Among these new materials, $La_2Ce_2O_7$, a solid solution formed by La_2O_3 in CeO_2 with a fluorite phase structure, has attracted much attention due to its low thermal conductivity, high phase stability at high temperature, and comparable thermal cycling life to 8YSZ [14–17]. The current fabrication method of LCO coatings included atmospheric plasma spraying (APS) and electron beam physical vapor deposition (EB-PVD). It has reported that the LCO coatings deposited by APS effectively prevented CMAS attack at 1250 °C [18]. Besides, the LCO coating deposited by EB-PVD showed good thermal cycling behavior [15]. As a result, the LCO coating was regarded as a promising TBCs material.

The ceramic materials such as LZO, GZO, $SrZrO_3$, LCO et al. consist of multiple ceramic constituents. During rapid heating to over their melting point by high temperature plasma jet in APS process, one of the constituents would experience preferential evaporation, resulting in deviation of coating compositions from the stoichiometry of original materials, due to the fact that the individual elements of these constituents have different vapor pressures. For plasma sprayed $La_2Ce_2O_7$, the preferential evaporation of CeO_2 leads to nonstoichiometric coating, due to higher vapor pressure of CeO_2 (2×10^{-2} atm, 2773 K) in comparison to La_2O_3 (8×10^{-5} atm, 2773 K) [5]. Consequently, the preferential evaporation of CeO_2 in spray process deviated coating composition from that of the original powders, resulting in the degraded performance of LCO TBC coatings. On the one hand, it was reported that the fluorite phase stably exists in a specific range of Ce/La molar ratio. The excess of La_2O_3 in the coatings will induce phase instability [19]. It has been proved that the LCO coating with the Ce/La ratio close to 1 shows the highest phase stability [20,21]. On the other hand, Cao et al. [14] reported that the thermal cycling life of the LCO coating

---

* Corresponding author.
E-mail address: licj@mail.xjtu.edu.cn (C.-J. Li).

http://dx.doi.org/10.1016/j.jeurceramsoc.2016.11.020

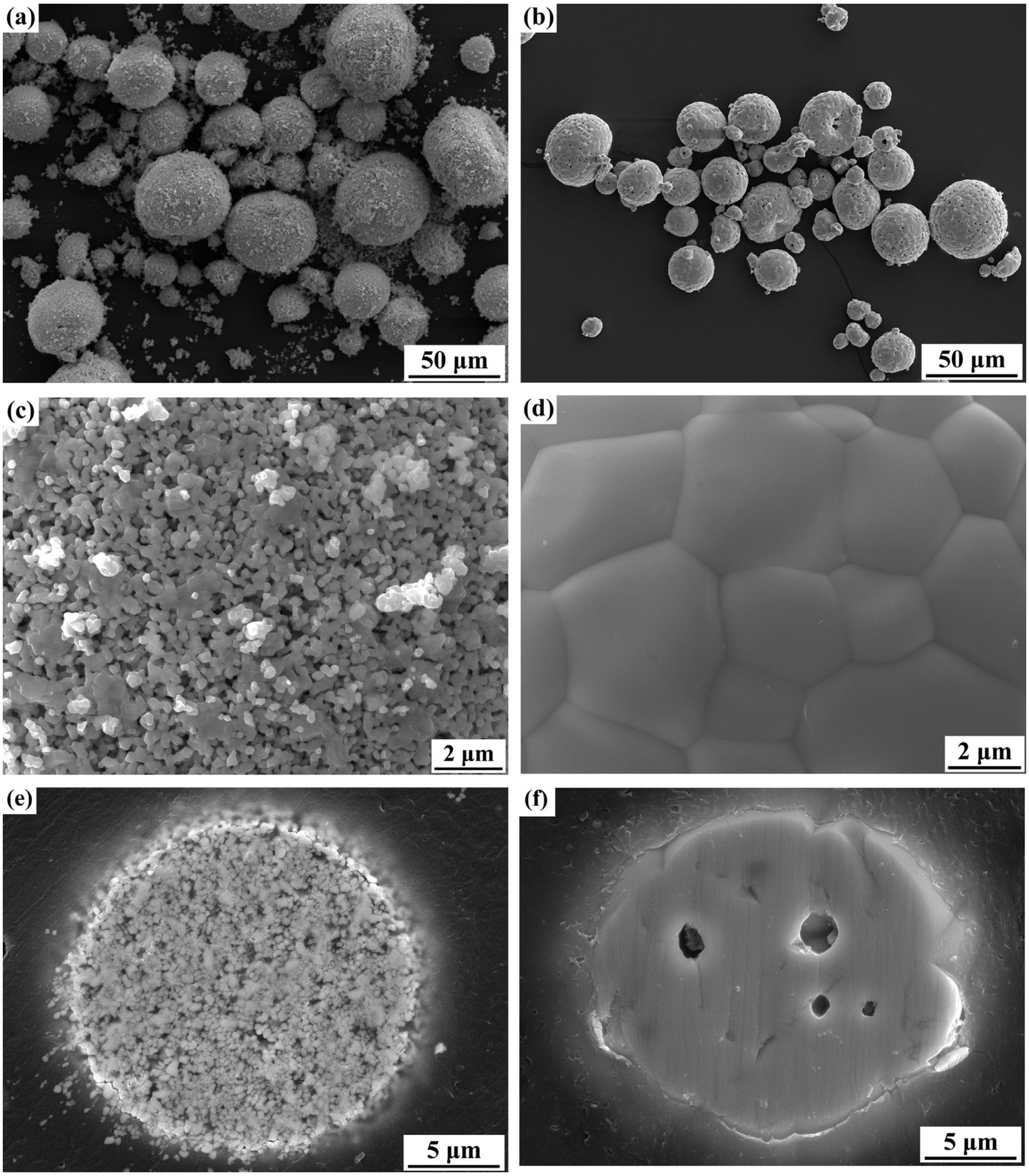

Fig 1. Surface morphologies of powders used in this study: (a) and (c) were the agglomerated LCO powders at low magnification and high magnification, respectively; (b) and (d) were the agglomerate-sintered LCO powders at low magnification and high magnification, respectively; (e) and (f) were the cross-sectional views of the agglomerated LCO powder and agglomerate-sintered LCO powder, respectively.

was sensitive to the CeO_2 content, and LCO presented long term thermal cycling lifetime when Ce/La ratio was 1.

Up to date, it is generally considered that the significantly preferential evaporation of constituent with higher vapor pressure is inevitable during plasma spraying of multiple constituent complex ceramics. Thus, one possible solution is to design spray powders such that powder contains excessive amount of high vapor pressure constituent over the stoichiometry. Dong et al. [22] reported that LCO coatings with a 1:1 Ce/La molar ratio can be prepared by the powders with a Ce/La molar ratio of 1:1.25.

However, since thermal spray powders usually have a wide particle size distribution, individual spray particles experience different heating to different temperatures due to the fact that heating degrees of spray powders is significantly influenced by particle size. Moreover, it was widely documented that higher degree heating of spray powders led to higher evaporation [23]. Thus, using the powders with a wide size distribution and even excessive amount leads to remarkably wide inhomogeneous distribution of compositions from one splat to another. For example, Mauer et al. showed that the La/Zr ratio can be varied from 1.06, being equal to that in the starting powder, down to 0.28 when plasma spraying of $La_2Zr_2O_7$ was performed using LZO powder with $d_{10} = 7\ \mu m$, $d_{50} = 44\ \mu m$, $d_{90} = 80\ \mu m$ [24]. Although by limiting the heating to spray powders the overall evaporation can be sigificantly reduced [22–24], it is difficult even impossible to supress the preferential evaporation induced inhomogeniety of compositions. To find the solution to such problem, it is essential to understand the dominant factors and their acting mechanisms for preferential evaporation.

Recently, according to our previous report [25], it has been found that the preferential evaporation of high vapor pressure constituent during plasma spraying was significantly influenced by particle size. The results indicated that particle size played a dominant