



High-temperature corrosion behaviour of plasma sprayed lanthanum magnesium hexaluminate coating by vanadium oxide

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Received 25 May 2014; received in revised form 16 August 2014; accepted 25 August 2014

Available online 7 September 2014

Abstract

Plasma sprayed lanthanum magnesium hexaluminate ($\text{LaMgAl}_{11}\text{O}_{19}$, LaMA) coating degraded by molten V_2O_5 at 710–1050 °C was studied. LaVO_4 , $\text{Mg}_{0.388}\text{Al}_{2.408}\text{O}_4$ and $\gamma\text{-Al}_2\text{O}_3$ were the main corrosion products at 710 °C and 800 °C, while the corrosion products were LaVO_4 , MgAl_2O_4 , $\alpha\text{-Al}_2\text{O}_3$ at 950 °C and 1050 °C. AlVO_4 as an important intermediate corrosion product had strong influences on the formations of γ - or $\alpha\text{-Al}_2\text{O}_3$ at different temperature regions. Recrystallization and hot corrosion happened to LaMA coating simultaneously at 950 °C and 1050 °C. However, at 800 °C, LaMA coating underwent the most serious degradation along the coating thickness direction due to the presence of amorphous phase.

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Keywords: Thermal barrier coating; Hexaluminate; Plasma spray; Hot corrosion

1. Introduction

Thermal barrier coatings (TBC) are finding increasing applications in advanced aero- and land-based gas turbine and diesel engines. TBCs are frequently used to provide thermal, corrosion and erosion protections for the hot-section components to realize the growing demands for improved engine efficiency and durability. A typical TBC presents duplex layer structure comprising of a metallic bond coat (MCrAlY , $\text{M} = \text{Ni}$ and Co) as the oxidation and corrosion resistant layer, followed by a thermal insulation ceramic top coat fabricated either by atmospheric plasma spraying (APS) or electron beam-physical vapour deposition (EB-PVD). 6–8 wt% yttria partially stabilized zirconia (YSZ) is the state-of-the-art ceramic top coat material due to its low thermal conductivity and comparative thermal expansion coefficient with the superalloy substrate etc. During high-temperature service, YSZ coated components exposed to combustion gases

in gas-turbine engines usually encounter high thermal residual stress level due to the serious thermal and mechanical loadings which could facilitate the crack formation and spallation failure of YSZ top coat. Thermal aging of YSZ coating accompanied by phase transformation with the consequences of volume expansion, sintering with the increase in Young's modulus etc., limit its long-term application below 1200 °C.^{1–5} As a result, developing new TBC candidate materials with improved temperature capability and durability is strongly demanded. Rare earth zirconates ($\text{Ln}_2\text{Zr}_2\text{O}_7$, $\text{Ln} = \text{La}$ to Gd) with the pyrochlore-type structure,^{6,7} perovskite-type SrZrO_3 ,⁸ fluorite-type $\text{La}_2\text{Ce}_2\text{O}_7$ ⁹ etc., are recommended and extensively investigated as new TBC candidate materials for higher temperature applications.

Lanthanum magnesium hexaluminate ($\text{LaMgAl}_{11}\text{O}_{19}$, LaMA) with the magnetoplumbite-type structure is a newly developed TBC candidate material for high-temperature applications due to its excellent thermophysical properties.^{10–15} The thermal conductivity of sintered LaMA bulk which is not fully dense as reported by Schaefer et al., is about $\lambda = 0.8\text{--}2.6 \text{ W m}^{-1} \text{ K}^{-1}$ at room temperature to 1200 °C, depending on the specific porosity level,¹² its thermal expansion coefficient is about $9.6 \times 10^{-6} \text{ K}^{-1}$.¹⁵ Our former research

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indicated that plasma sprayed LaMA coating exhibited the longest thermal cycling lifetime among all of the rare earth hexaluminate oxides at the surface testing temperature of $\sim 1250^\circ\text{C}$.¹⁶ New functionally graded TBC system based on LaMA/YSZ even exhibited a thermal cycling lifetime which was almost two times of that of the LaMA/YSZ double ceramic layer TBC, and several times longer than that of single layer YSZ coating at the surface testing temperature about 1350°C .¹⁷ These preliminary results indicate that LaMA is a promising TBC candidate material for high-temperature applications.

During high-temperature service, there are many factors responsible for TBC failure. High-temperature chemical corrosion originating from the use of cost-effective fuels with the impurities such as V, Na, P and S,^{18–30} and as well as the ingested sand particles to form molten glass of calcium–magnesium–aluminium–silicates (CMAS)^{31–34} is one of the important factors responsible for the TBC premature failure. Hot corrosion behaviour and degradation mechanism of YSZ coating by molten vanadate, sulphate and P_2O_5 etc, have been extensively investigated. The main reason for YSZ coating corrosion failure is the chemical reaction between molten salts and Y_2O_3 stabilizer at high temperature, which gives rise to the leaching of yttria from the YSZ solid solution leading to the phase transformation and accelerated coating spallation failure.^{19,20,24–30,35} In the previous work, we investigated the corrosion behaviour of plasma sprayed YSZ/LaMgAl₁₁O₁₉ composite coating in molten sulphate–vanadate salt mixture at the fixed temperature of 950°C .³⁶ In this paper, the degradation mechanism of plasma sprayed free-standing LaMA coating exposed to molten vanadium oxide at 700 – 1050°C has been investigated in order to better understand the corrosion resistance of LaMA as a promising material for TBC applications.

2. Experimental procedures

LaMA powders were prepared by a solid state reaction method. Three commercially available high purity oxides of La_2O_3 (99.99%, Guangdong Chenghai Chemicals Co., Ltd.), Al_2O_3 (99.99%, Tangshan Huatai Functional Ceramic Materials Co., Ltd.) and MgO (99.2%, Wuzi Zehui Chemicals Co., Ltd.) were selected as the starting materials. Such three oxides mixed in proper ratio were heated at 1650°C for 24 h to obtain the final product. Fig. 1(a) shows that the as-synthesized LaMA powder has a good purity, its XRD pattern matches the corresponding LaMA JCPDS card no. 26-0873 very well. The typical platelet-like hexagonal grains can be observed in the SEM micrograph of LaMA powder as shown in Fig. 1(b). The as-synthesized LaMA powders mixed with arabic gum and deionized water were subjected to ball-mill mixing again and were later spray-dried (Jiangsu Yangguang Ganzao Co., Ltd.). The obtained free-flowing LaMA powders with the particle size of 20 – $100\ \mu\text{m}$ were sieved for plasma spraying. The spray-dried powders were plasma-sprayed onto a graphite substrate using the Sulzer Metco plasma spray unit with a F4-MB gun to produce a LaMA coating with a thickness of $\sim 1.2\ \text{mm}$. The as-sprayed LaMA coating exhibited a density of $\sim 3.0\ \text{g cm}^{-3}$ (relative density 70% compared with the theoretical density $4.285\ \text{g cm}^{-3}$ of LaMA).

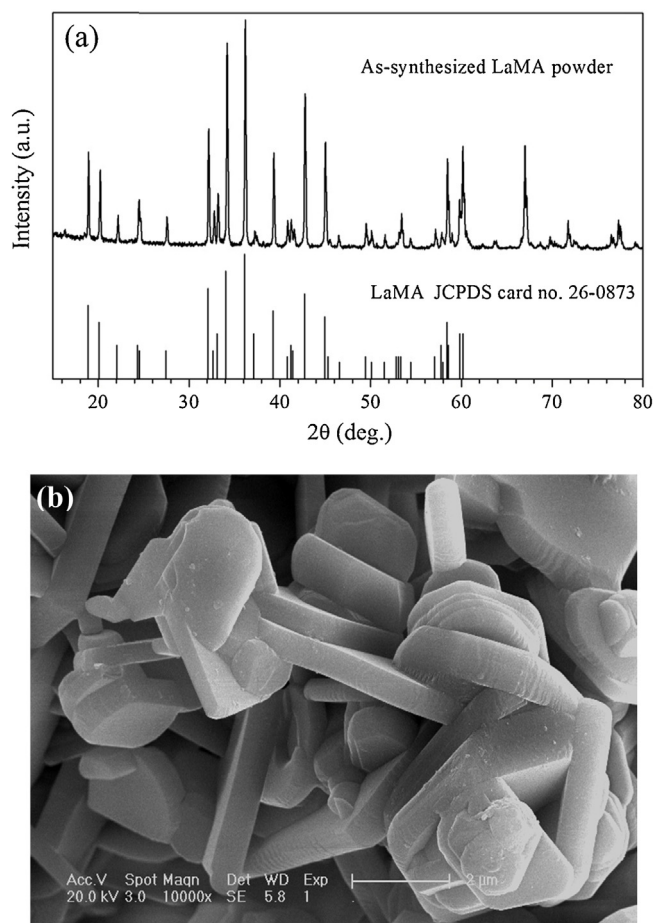


Fig. 1. XRD pattern of as-synthesized LaMA powder (a) and morphology of platelet-like LaMA grains.

For high temperature hot corrosion tests, the free-standing as-sprayed LaMA coating was obtained by mechanical removing from the graphite substrate, followed by cutting them into bars with dimensions of $2\ \text{cm} \times 2\ \text{cm} \times 1.2\ \text{mm}$. All the bars were cleaned by acetone and ethanol, and dried for 12 h at 200°C . After that, fine V_2O_5 powder was spread uniformly on the surfaces of the coating bars at an approximate loading of $20\ \text{mg/cm}^2$ to perform an accelerated high temperature hot corrosion tests on coating samples compared to the TBCs coated blades of the industrial gas turbines. Hot corrosion tests were carried out in a muffle furnace with an ambient atmosphere at the temperatures of 710°C , 800°C , 950°C and 1050°C for 2 h and 10 h, respectively. The temperature regions selected here are under considerations of complex service conditions of TBCs, and to get more details about the degradation mechanism of LaMA coating. After isothermal corrosion, samples were cooled down to the room temperature in the furnace.

After hot corrosion, corroded surfaces of samples were examined by X-ray diffraction (XRD, Bruker D8 Advance diffractometer, $\text{Cu-K}\alpha$ radiation, $\lambda = 0.15406\ \text{nm}$) at the range of $2\theta = 15$ – 80° , with a step size of 0.02° and a counting time of 0.5 s per step, to identify the corrosion products. Surface and cross-section of the corroded samples were coated by a thin Au layer to make them electro-conductive prior to examinations by

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