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Influence of water vapour on structural and thermal conductivity of post-heat treated plasma sprayed LZ and YSZ coatings

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ABSTRACT

In this work, the role of water vapour content during the post-heat treatment on the microstructure and thermal conductivity of atmospheric plasma sprayed lanthanum zirconate (LZ) and yttria stabilized zirconia (YSZ) coatings were studied. For this purpose free standing LZ and YSZ coatings with thickness about 300–400 μm were prepared and post-heat treated at 1100 °C for 50 h and 100 h with and without water vapour content. Phase stability and microstructure were investigated using X-ray diffraction and scanning electron microscope respectively. Thermal conductivity of as-sprayed and post-heat treated coatings were measured by laser-flash thermal diffusivity method and results obtained show that the thermal conductivity of post heat treated coatings increased significantly in both coatings. Further, the thermal conductivity of the coatings increased with increasing heat treatment time. Overall observations show that the presence of water vapour during the post-heat treatment has significantly influenced the microstructures as well as thermal conductivity in both coatings. Thermal conductivity of post-heat treated lanthanum zirconate and yttria stabilized zirconia coatings with presence of water vapour is 2.12 W m^{-1} K^{-1} and 2.48 W m^{-1} K^{-1} respectively which is 46% and 41% higher than as-sprayed coatings.

1. Introduction

The durability of plasma-sprayed thermal barrier coatings (TBCs) was strongly depends on the thermo-physical properties of the selected materials and their microstructure architectures. For several decades, yttria-stabilized zirconia (YSZ) thermal barrier coatings are extensively used in land and aviation turbine industry. However, the yttria-stabilized zirconia coatings are failed to accomplish the requirements of new generation advanced TBC's capable of withstanding higher temperatures. The development of new generation TBC is required to drive the technology to elevated level of efficiency [1,2]. Recently, Lanthanum zirconate (LZ) with pyrochlore structure emerged as a promising TBC material and it has prospective to replace the conventional YSZ due to its low thermal conductivity and high temperature phase and structural stability [3–7]. Further the oxygen permeability of LZ based coatings is lower in comparison with that of conventional YSZ based coating systems [8].

Numerous investigations have been made to understand the fundamental behaviors and changes in thermo-physical and mechanical properties of LZ coatings under various simulated TBC's conditions including various hot corrosion environments [9]. Many attempts have been made on LZ to meet the requirements of advance TBC's at high temperatures. The thermal stability, hot corrosion behaviors of as-sprayed LZ based coatings at high temperature have been investigated by various researchers and few of them studied how the post annealing treatment affected the coating properties. Chen et al. [10] prepared LZ coatings by APS method and reported the thermo-physical properties. Zhou et al. [11] and Xiang et al. [12] prepared CeO_2 doped LZ thermal barrier coatings and studied the effects of co-doping on the phase structure and their thermo-physical properties particularly thermal conductivities and thermal expansion coefficients of the coatings.

Erdogan et al. [13] studied the influence of long time post annealing on thermal stability and thermo-physical properties of plasma sprayed $La_2Zr_2O_7$ coatings. Their results reveal that the sintering behaviour of coatings during the annealing affects the thermal conductivity severely by the reduction of microcracks, particularly in the first 50 h of heat treatment. Further their analysis revealed the decomposition of pyrochlore structure LZ coatings into meta-stable tetragonal and monoclinic ZrO_2 with long heat treatment time at 1150 °C. Wu et al. [14] reported on the phase

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stability of Ce-modified $La_2Zr_2O_7$ coatings and its chemical compatibility with YSZ. Their result shows that the pure LZ and Ce-doped LZ coatings had excellent phase stabilities and good chemical compatibility with YSZ. Also the excess amount of Ce-doping in LZ has weakened the phase stability and chemical compatibility with YSZ significantly.

Numerous earlier reports clearly illustrate the hot corrosion behaviour of LZ coatings against foremost corrosive elements at elevated temperatures and time duration [15]. Summary of the existing results reveals that the durability of the LZ exhibits superior hot corrosion resistance against V_2O_5. However the comprehensive resisting performance of LZ coatings against different kinds of harsh corrosive environments was not enough when compared with conventional YSZ coating for hot corrosion results [16]. Also attempts were made to improve the hot corrosion resistance of LZ coatings by adding different dopents in LZ structure, composite coating of LZ/YSZ and double layer or multilayer LZ/YSZ coatings [17,18].

Most of the research work on LZ coatings concentrated on their thermal stability at high temperatures, hot corrosion resistance against corrosive environments and changes in thermo-physical properties due to the post-heat treatments [13,19]. However, until now there is no clear view about how the water vapour environment at high temperature affects LZ coating properties especially its thermal conductivity. The objective of the present work is to study the effect of water vapour content during the post-heat treatment on the phase stability, microstructure and thermal conductivity of atmospheric plasma sprayed lanthanum zirconate (LZ) coating and compared with conventional YSZ coating. For this purpose, free-standing LZ and YSZ coating specimens were prepared by atmospheric plasma spraying under optimum spraying conditions and subjected to post-heat treatment at 1100 °C for 50 h and 100 h with and without water vapour atmospheres. The effect of water vapour exposure on the microstructure, phases and thermal conductivity of the coatings were explored by using X-ray diffraction pattern (XRD), Fourier transform infrared spectroscopy (FTIR), Scanning electron microscope-energy dispersive spectroscope (SEM-EDS) observations and laser-flash thermal diffusivity analyzer.

2. Experimental method

Lanthanum zirconate powder (LZ) and conventional yttria stabilized zirconia (YSZ) were used in this study. Single phase pyrochlore structure of lanthanum zirconate powder (LZ) were produced by a mechanically activated solid state route from powder mixtures of ZrO_2 (Himedia, India), La_2O_3 (Himedia, India) in ethanol. Milled suspensions were then oven-dried and sintered at 1500 °C. The powders obtained were mechanically crushed and sieved with different sizes of mesh in order to obtain narrow particle size distributions. Average particle size distribution of both powders was evaluated by a laser diffraction particle analyzer. The as-prepared LZ powder exhibited a powder size distribution varying from approximately 25 to 37 μm. Conventional 7 wt% yttria stabilized zirconia (7YSZ) powder was procured from Cumi Ltd, India. The average size distribution of conventional 7YSZ powder is 45–63 μm.

As-prepared LZ powder and conventional 7YSZ powders were sprayed using a 40 kW DC plasma spray torch (Ion Arc Technologies, Coimbatore, India) under optimum conditions. The optimum spray parameters employed to deposit the above coatings were developed internally based on numerous trial coatings. In this study, the torch was operated at 30 kW input power with flowing of 30 slpm argon and 4 slpm nitrogen. Both the powders were sprayed on 25 mm diameter steel substrates. The maximum substrate temperature during the spraying was approximately 300 °C. Torch to substrate distance 120 mm was kept constant in both coatings. Thickness of the coating was approximately 300–400 μm. The powder feed rate is approximately 25 g/min used for coating deposition.

For post heat treatment, the steel substrates were removed from the coating microstructure by soaking the coated steels with HCl solution to produce free-standing coatings. The post heat treatment was carried out in tubular furnace at 1100 °C for periods of 50 h and 100 h with and without water environment. For water vapour treatment, the atmosphere inside the tubular furnace was maintained with water vapour environment by passing a continuous flow of 50% H_2O + 50% air mixture (in molar ratio) at the flow rate of 1 slpm using an in-house developed high temperature tube furnace.

The structural characteristics of powders and coatings were evaluated using a scanning electron microscope (SEM; Hitachi TM3000). The free standing coatings were first mounted in epoxy resin then cut using a diamond saw and subsequently polished using standard metallographic procedures in order to preserve and reveal the proper structural features of the coatings. The phase stability of as-sprayed and heat-treated coatings was investigated using Fourier-transform infrared spectroscopy (FTIR; Spectrum 100 model spectrophotometer) and X-ray diffraction (XRD; Bruker AXS, Karlsruhe, Germany) with Cu-$K_{\alpha 1}$ radiation at a scan rate of 5°/min. The porosity of the coating microstructures were estimated from cross-sectional microstructure images by using image J software. For this purpose, coating cross-sections were completely scanned from one end to another and five images from different locations in each coating were selected and the porosity values of the five images were averaged.

The thermal conductivity (λ) of as-sprayed and post heat treated samples were calculated by using the following equation:

$$\lambda = \rho \, \alpha \, Cp \tag{1}$$

where ρ is the density of the coating measured by the Archimedes method with an immersion medium of deionized water. Before these measurements, the specimens were pre-treated in boiling water for about 2 h in order to make the pores fully filled with water. Cp is the specific heat capacity of the coatings obtained using differential scanning calorimeters (DSC) for a powder specimen, working continuously at a scanning rate of 20 K/min from room temperature to 1200 °C (Model 404, Netzsch, Bayern, Germany) in an N_2 atmosphere. Here α is the thermal diffusivity of the coating. It was measured as a function of temperature in the range between 100 °C and 1200 °C by using the laser-flash method in a nitrogen atmosphere (DSC-1200, TA Instruments).

3. Results and discussion

3.1. Feedstock powders

The results of particle size distributions shows that the as-prepared LZ powder exhibits a particle size distribution considerably lower than that of typical plasma spray powders, i.e., almost 60% of the particles exhibit diameters lower than 30 μm. The surface morphology of the conventional YSZ and as-prepared LZ powders are shown in Fig. 1 (a) and (b) respectively. It is possible to distinguish the porous and ultrafine characters of the as-prepared LZ powders which were produced by mechanically activated solid state route followed by sintering and crushing. Meantime, in the conventional YSZ powders, the individual spray-dried particles were almost completely molten without any significant ultrafine characters due to their processing method.