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Engineered architectures of gadolinium zirconate based thermal barrier coatings subjected to hot corrosion test

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ABSTRACT

Gadolinium zirconate (GZ) is considered as a promising top coat candidate for high temperature TBC applications. Suspension plasma spray has shown the capability to generate a wide range of microstructures including the desirable columnar microstructure. In this study, two different TBC architectures were deposited using the axial suspension plasma spray. The first variation was a triple layered TBC comprising of thin YSZ base layer beneath a relatively porous GZ intermediate layer and a dense GZ top layer. The second variation was a composite TBC architecture of GZ and YSZ comprising of thin YSZ base layer and GZ + YSZ top layer. Cross sectional SEM analysis of the layered and composite TBCs revealed a columnar microstructure. The porosity content of the deposited TBCs was measured using two methods (Image Analysis and Water Intrusion). The as-sprayed TBCs were exposed at 900 °C for 8 h to a corrosive salt environment consisting of a mixture of vanadium pentoxide and sodium sulfate. XRD analysis on the as-corroded TBCs top surface showed the presence of gadolinium vanadate in both the layered and the composite TBCs. SEM/EDS analysis of the top surface and the cross-section of the layered and composite TBCs after hot corrosion test revealed the infiltration of the molten salts through the columnar gaps. The composite TBC showed a lower hot corrosion induced damage compared to the layered TBC where a considerable spallation was observed.

1. Introduction

Thermal barrier coatings (TBCs) are multilayered coating systems comprising of a base layer of metallic bond coat of MCrAlY composition and a ceramic top layer. TBCs are widely used in land based and aero engine applications for insulating the metallic components from high heat loads. With the rise in demands for higher engine efficiency, the turbine inlet temperature has to be increased [1–2]. 7–8 wt% Yttria stabilized zirconia (YSZ) is the standard top coat ceramic material used for TBC application due to its excellent properties such as low thermal conductivity, good fracture toughness and high coefficient of thermal expansion (CTE). However, at high temperatures (>1200 °C), YSZ is susceptible to CMAS attack and phase transformation which results in reduced TBC durability [3–11]. New ceramic materials which can overcome these challenges at high temperature are highly desirable.

During the past decade, potential top coat TBC candidates of new ceramic material compositions have been explored [12–16]. Among the new materials, pyrochlores of rare earth zirconate based compositions (lanthanum zirconate and gadolinium zirconate) have shown to be promising candidates due to their lower thermal conductivity compared to YSZ [12]. Lanthanum zirconate (LZ) has processing issues during plasma spraying due to its tendency to lose original stoichiometry [14,17]. Gadolinium zirconate (GZ) has demonstrated to possess excellent CMAS resistance capability compared to the standard YSZ [8,18,19]. However, GZ single layer TBCs have shown poor thermal cyclic performance compared to the YSZ single layer TBC [20]. The reason for inferior thermal cyclic life in GZ single layer TBCs was attributed towards the thermo chemical compatibility issues of GZ with the in situ grown thermally grown oxide (alumina) [21]. Engineering a multi-layered TBC, for instance, with YSZ as the base layer and GZ as the top layer, has shown superior thermal cyclic life compared to the single layer YSZ TBCs [22] [23].

Processing route of a TBC plays an important role in the microstructure obtained which in turn determines the functional performance (thermal cycling life, thermal conductivity, erosion resistance) of the TBC. Columnar microstructure favors good thermal cyclic life of the TBC compared to the conventional splat morphology due to its

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improved strain tolerance. In addition, TBCs with columnar microstructure were shown to possess superior erosion resistance compared to the splat morphology obtained by atmospheric plasma spraying (APS) [24, 25]. Electron Beam Physical Vapor Deposition (EB-PVD) is the conventional processing route to obtain a columnar structure. However, it is relatively expensive and has a slow deposition rate than the plasma spraying process (APS). With APS route, it is next to impossible to obtain a columnar microstructure. An alternative to EB-PVD, suspension plasma spraying technique can generate columnar microstructures by using sub-micron sized powders suspended in solvents such as water or ethanol [26].

TBCs used in land based gas turbines encounter molten salt attack (hot corrosion) which results in reduced TBC lifetime. Major source of the corrosive salts (sulfate and vanadate) are the impurities (sulfur, vanadium) present in low grade fuel used for such applications. The mechanism of TBC degradation due to the molten salt attack was reported to be due to the selective leaching of the stabilizer from zirconia which results in an undesirable martensitic phase transformation (tetragonal to monoclinic) resulting in a volume change of 3–5% upon cooling [27]. It is of interest to design TBCs which can resist the corrosive attack by opting for a low reactive top coat TBC material or a dense sealing layer in order to improve the TBC durability.

Previously, attempts were made by Batista et al. to deposit a sealing layer by post treatment of APS processed TBCs using laser glazing [28]. However, this additional TBC post treatment step with increased processing cost could not help in restricting the hot corrosion attack as it could not seal the cracks completely. Therefore, it is desirable to obtain a dense sealing layer by varying the process parameters.

Hot corrosion behavior of GZ based multi-layered TBCs was investigated in our previous work [29]. GZ based multi-layered TBCs exhibited higher corrosion induced damage compared to the single layered YSZ. The reason for spallation of GZ based TBC was attributed to the infiltration of molten salts in the columnar gaps leading to the formation of the corrosive product and thereby reducing the strain tolerance. Another reason was the low fracture toughness of GZ compared to YSZ [30]. This work is an attempt to improve the hot corrosion resistance of the GZ based TBC by opting for a double layer TBC comprising composite of GZ and YSZ (50:50 wt%) as the top layer and YSZ as the base layer. The reason for utilizing the composite of GZ and YSZ was to improve the fracture toughness. For reference, a triple layered TBC of GZ dense/GZ/YSZ was also included in the work. The dense GZ layer was deposited using a water based GZ suspension. Both the TBC systems were exposed to a mixture of sodium sulfate and vanadium pentoxide for 8 h at 900 °C and their interaction with the corrosive salts was analyzed using SEM/EDS and XRD.

2. Experimental details

2.1. Sample preparation

Hastelloy-X substrate discs of 25.4 mm diameter and 6.3 mm thickness were grit blasted using alumina of 220 μm grit size to achieve a surface roughness (Ra) of 3 μm. Commercially available bond coat powder, AMDRY-386 (Oerlikon Metco, Switzerland) (Ni 18Co 13Cr 10Al 0.1Y), was deposited using the high velocity air fuel (HVAF) process (UniqueCoat M3, UniqueCoat, Virginia USA). The bond coated substrates were again grit blasted using alumina particles of 220 μm grit size to achieve a surface roughness (Ra) of 5 μm. The surface was later air blasted to remove the loosely bonded alumina particles from the grit blasting process.

2.2. Suspension feedstock

Two production suspensions and two experimental suspensions (3 ethanol based and 1 water based) were provided by Treibacher Industrie AG, Austria, for this study. The first production suspension was the commercially available AuerCoat YSZ suspension in ethanol. The YSZ consisted of 8.1 wt% Y_2O_3, 1.9 wt% HfO_2, total other impurity oxides <0.05 wt% and the balance of ZrO_2. Particle size distribution of the suspension was D10 of 0.23 μm, D50 of 0.55 μm and D90 of 1.07 μm. The second suspension was the proprietary AuerCoat Gd-Zr suspension in ethanol. The material had comparable purity level and particle size to the YSZ suspension.

The experimental suspensions consisted of a 50:50 mixture of the YSZ and GZ suspensions with a solids load of 25%. The suspension was produced by mixing equal parts by weight of the single oxide suspensions. The final experimental suspension consisted of a water based GZ suspension manufactured from the same feedstock powder as the ethanol based suspensions. Solids load for the water based suspension was 40 wt%.

2.3. Top coat processing

The top coat was deposited by axial suspension plasma spray process using the Axial III plasma gun and Nanofeed 350 feeder system obtained from Mettech Corp., Canada. The specimens to be coated were mounted on a rotating fixture and a fixed rotating speed was used to obtain a uniform coating thickness on the mounted specimens. Prior to the top coat deposition, the surface of the bond coated substrate was heated (approximately 200 °C) in order to get rid of the volatile impurities. A plasma exit nozzle of 3/8″ was used and the suspension was axially injected into the plasma through a 250 μm diameter orifice. Two variations of top coats were deposited, as shown in Fig. 1. The suspension feedrate, standoff distance and the rest of the spray parameters used for the TBC processing are given in Table 1.

2.4. Coating characterization

2.4.1. Metallography

The as-sprayed TBCs were cold mounted using low viscosity epoxy and sectioned using slow speed diamond cutter. The sectioned samples were again cold mounted and polished to obtain a scratch free surface. Details of the polishing steps are discussed in our previous work [31]. The polished samples were gold sputtered and observed in a scanning electron microscopy from Hitachi TM 3000, Japan in back scatter electron mode. Additionally, elemental maps (Gd, Zr, Y) of the cross section of the TBC samples were analyzed by EDS (Bruker). For the top surface analysis, carbon tape was applied to make the surface conducting. After the hot corrosion test, the TBCs were mounted, polished and analyzed by SEM/EDS in a similar fashion.

2.4.2. Porosity measurement

The porosity content of the as sprayed TBCs was measured using an image analysis method and the water intrusion method.

2.4.2.1. Water intrusion method. The multilayered TBCs were treated as a single unit and their porosity content was measured by water intrusion method according to the ISO standard (ISO 18574:2003(E)) [32]. Free standing coatings were produced according to the method discussed in our previous work [31]. Later, the free standing coatings were cleaned with acetone and dried in the oven at 120 °C for an hour. Dry weight (m_1) of the free standing coatings was measured and later immersed in distilled water. The setup comprising of a beaker with TBCs immersed in distilled water was placed in a vacuum chamber and vacuumed several times to remove the entrapped air from the pores of the TBCs and allow water to infiltrate. The weight of TBCs immersed in water was measured as 'm_2'. The free standing coatings infiltrated with water were removed from the beaker and their wet weight (m_3) was measured immediately after wiping away the excess water (water entrapped on the rough TBC) from the surface. The apparent porosity