

Impermeable thin Al₂O₃ overlay for TBC protection from sulfate and vanadate attack in gas turbines

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ABSTRACT

To improve the hot corrosion resistance of YSZ thermal barrier coatings, a 25 μm and a 2 μm thick Al_2O_3 overlay were deposited by HVOF thermal spray onto to the surface of YSZ coating. Oxidation at high temperature and hot corrosion tests showed that Al_2O_3 overlay deposited on the YSZ TBCs surface can not only reduce the hot corrosion rate, but also significantly prevents the bond coat from oxidation.

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1. INTRODUCTION

Plasma sprayed thermal barrier coatings (TBCs) are widely used in gas turbine hot section components such as burners, transition ducts, shrouds, blades and vans. The most common TBC materials is Y₂O₃ (8wt%)-stabilized ZrO₂ type (YSZ) which has been developed over many years because of its high temperature stability, low thermal diffusivity and high coefficient of thermal expansion (CTE) [1,2]. However, when exposed to acidic molten salt, stabilizer yttria will be leached out from the zirconia solid solution, resulting in destabilization of the zirconia from tetragonal to the monoclinic phase and destruction of the coating.

The major failure mechanism that causes TBC spallation in gas turbine is bond coat oxidation and the growth of the thermally grown oxide (TGO), while hot corrosion of TBC will dominate coating failure in diesel engines which are usually operated with low quality fuels containing lots of impurities such as sulfur and vanadium [2].

Molten sodium salts of vanadium and sulfur oxides condense on to the TBCs at the temperature of 600-1000°C [3, 4]. Although zirconia itself shows good resistance to the molten sulfate or vanadate compounds arising from fuel impurities, yttria is leached out of the zirconia by the reaction with V₂O₅ or NaVO₃ to form YVO₄, causing structural destabilization of ZrO₂ (i.e., transformation of the zirconia from the tetragonal and/or cubic to monoclinic phase upon cooling, which is accompanied by a large destructive volume change) [5-10]. Stresses resulting from destabilization of the zirconia eventually cause the delamination and spalling of the coating.

Thus, extension of the benefits of TBCs to such impurity-containing environments requires the development of hot corrosion resistant coating. Based on Lewis acid-base concept, zirconias stabilized with indium (In₂O₃) [11, 12], scandia (Sc₂O₃) [13] and ceria (CeO₂) [8,14] as well as Ta₂O₅ [6,15] and YTaO₄ [15] have been evaluated for their hot corrosion resistance. On the other hand, over the years there have been, and still continue to be, efforts to close the

surface of zirconia TBCs by laser-glazing and arc lamp [16-18] or various “seal coats” [18-25] to prevent penetration of molten deposits into the porous YSZ coating.

Alumina has a high melting point and stability without showing phase transition at high temperature like the ZrO_2 ceramics. Al_2O_3 has a small solubility particularly in molten salts and is expected to show an excellent corrosion resistance [26]. The hot corrosion tests of TiAl with Al_2O_3 coating in the sulfate melt at $900^\circ C$ have shown that the Al_2O_3 coating is very stable in the sulfate melt and effectively prevent intermetallic TiAl from hot corrosion attack [27]. Chen et al’s experiment has demonstrated that the Al_2O_3 coating could resist hot corrosion attack of molten Na_2SO_4 salt for longer time than the YSZ coating [28]. In addition, Al_2O_3 - ZrO_2 composite coatings have been explored as thermal barrier applications, showing better resistance in NaCl molten salt than YSZ [29]. This allows the potential application of Al_2O_3 in gas turbines. On the other hand, Al_2O_3 barrier layer was also deposited between the top coat and bond coat by chemical-vapor deposition (CVD) to suppress the oxidation rate of the bond coat. Recent work [30] has shown that a dense and continuous Al_2O_3 overlay on the surface of TBC deposited by EB-PVD reduced the permeability to gas and salt, and subsequently improved the hot-corrosion resistance of the TBC and suppresses the oxidation rate of the bond coat.

Previous works of this project have shown that Al_2O_3 overlay can significantly reduce the hot corrosion rate when exposure to molten Na_2SO_4 salt containing V_2O_5 at high temperature. As mentioned above, bond coat oxidation and the growth of the thermally grown oxide (TGO) are another major failure mechanisms that cause TBC spallation in gas turbine application. The present work will focus on the effect of Al_2O_3 overlay on the bond coat oxidation and the growth of the thermally grown oxide (TGO) when exposure to air and to molten Na_2SO_4 salt containing V_2O_5 at high temperature.

2. EXECUTIVE SUMMARY

Without Al_2O_3 overlay on the surface, cracking occurred within YSZ TBCs near the bond coat due to extensive growth of TGO. Al_2O_3 overlay deposited on the YSZ TBCs surface can not only reduce the hot corrosion rate, but also significantly prevents the bond coat from oxidation either exposure to air or exposure to the molten salt.

3. EXPERIMENTAL

The TBC system used in this study consisted of 6061 nickel-based superalloy substrate, CoNiCrAlY alloy bond coat as well as zirconia-8%yttria (YSZ) ceramic top coating. The substrate was grit-blasted with alumina particles and then deposited with a $100\ \mu m$ thick CoNiCrAlY alloy (weight percent: 32%Ni, 21%Cr, 8%Al, 0.5%Y and 38.5%Co) bond coat by low-pressure plasma spray (LPPS) process. The LPPS spraying was carried out under the spraying voltage of 68 V and the current of 630 A with a primary gas Ar flow of 60 l/min, a secondary gas H_2 flow of 8.5 l/min and a carrier gas Ar of 8.5 l/min. The substrate with the CoNiCrAlY bond-coat was sprayed with a $200\ \mu m$ thick ZrO_2 -8wt% Y_2O_3 top coat by an air plasma-spray (APS) process under the spraying current of 550 A and the spraying voltage of 68 V with a primary gas Ar of 41 l/min, a secondary gas H_2 flow of 10 l/min and a carrier gas Ar flow of 3 l/min. Al_2O_3 overlay of 25 thick was deposited by HVOF thermal spray on the surface

of bond coat, using the Praxair HV-2000 gun with propylene as fuel. On the other hand, 2 μm thick Al_2O_3 overlay was prepared using sol-gel method according to a previous report.

High temperature oxidation, and hot corrosion test was performed on the TBCs with and without Al_2O_3 coating. For hot corrosion test, the TBC plates coated with salt mixture were placed into a still air furnace, and isothermally held at 950 °C up to 100 hours. Approximately 50 mg/cm^2 salt mixture was sprayed on the surface of TBC using an aqueous solution (1000 g/l 95wt% Na_2SO_4 + 5wt% V_2O_5). After exposure, the samples were cooled down to room temperature in the furnace. The exposed samples were cleaned in de-ionized water. The cross-section was examined using the PHILIPS XL30 scanning electron microscope (SEM) with which an energy-dispersive spectrometer (EDS) was equipped.

4. RESULTS AND DISCUSSION

4.1 Oxidation of TBC

Figure 1 shows the SEM images of YSZ TBC without Al_2O_3 overlay sample after exposure to air at 950 °C up for 100 hours.

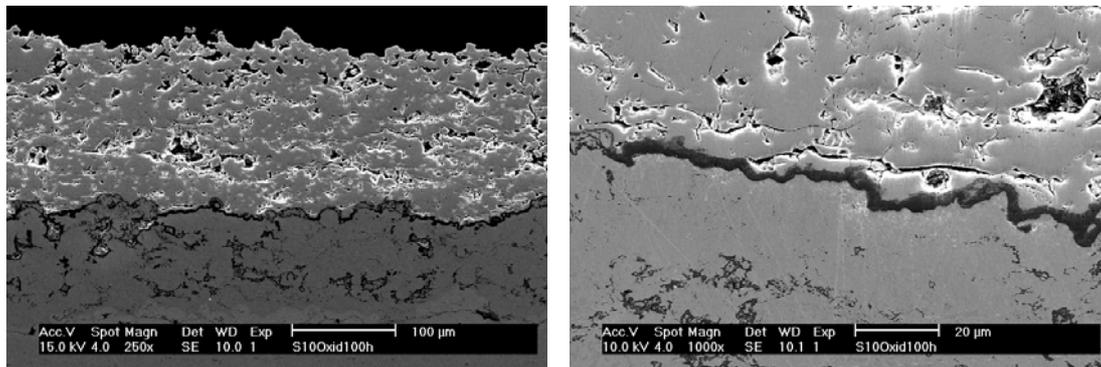


Figure 1 SEM cross-section images of TBC without Al_2O_3 overlay sample after exposure to air at 950 °C for 100 hours.

It can be clearly seen that the bond coat has been extensively oxidized. The thickness of thermally grown oxide (TGO) was 5.5-6.0 μm . In addition, many cracks were found in YSZ coating adjacent to the TGO layer, due to the thermally grown stress. EDX analyses demonstrated that the TGO was mainly consisted of Al_2O_3 , which is consistent with the previous results.

On the contrary, the thickness of thermally grown oxide (TGO) was only 1.4-2.1 μm when TBC with Al_2O_3 overlay exposure to air at 950 °C up for 100 hours, as shown in Figure 2. Moreover, no cracks could be found in YSZ coating near the TGO layer, due to much smaller thermally grown stress.

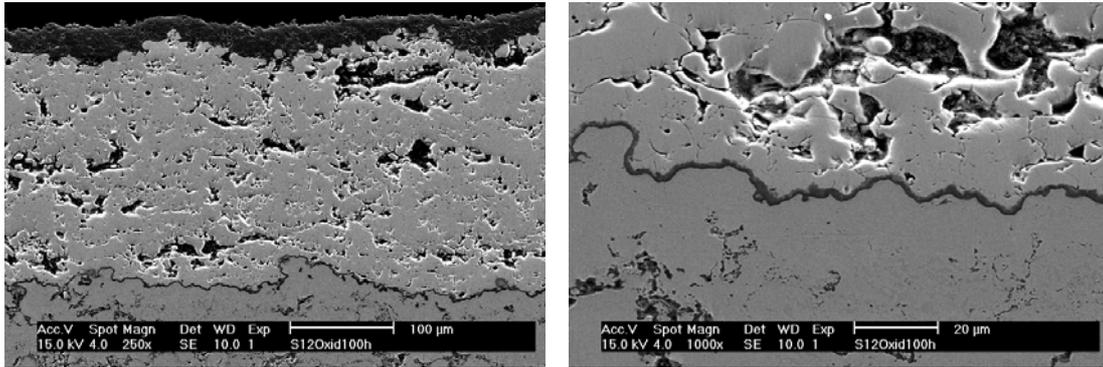


Figure 2 SEM cross-section images of TBC with Al_2O_3 overlay sample after exposure to air at $950\text{ }^\circ\text{C}$ for 100 hours.

4.2 Hot corrosion tests of TBCs

During hot corrosion, in addition to the penetration of molten salts into YSZ, oxygen could also diffuse into YSZ and react with bond coat to form thermally grown oxide (TGO). Figure 3 and Figure 4 show the SEM cross-section images of YSZ TBC without and with Al_2O_3 overlay sample after exposure to molten salt at $950\text{ }^\circ\text{C}$ for 100 hours.

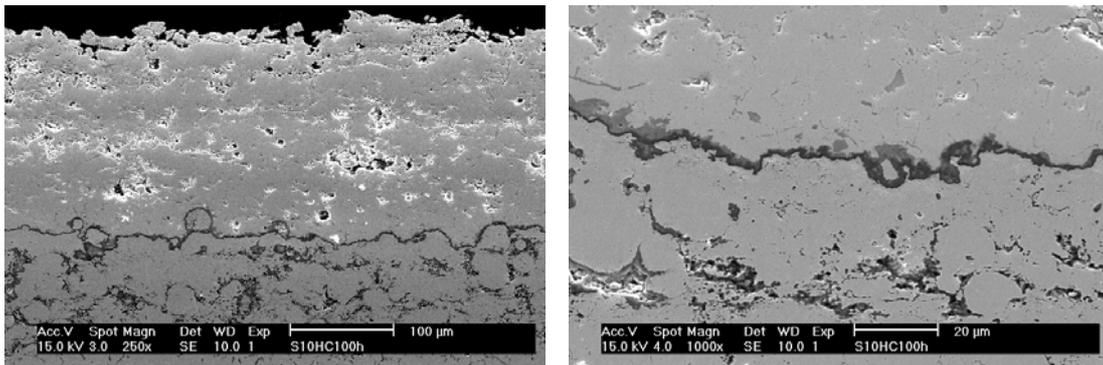


Figure 3 SEM cross-section images of TBC without Al_2O_3 overlay sample after exposure to the molten salt at $950\text{ }^\circ\text{C}$ for 100 hours.

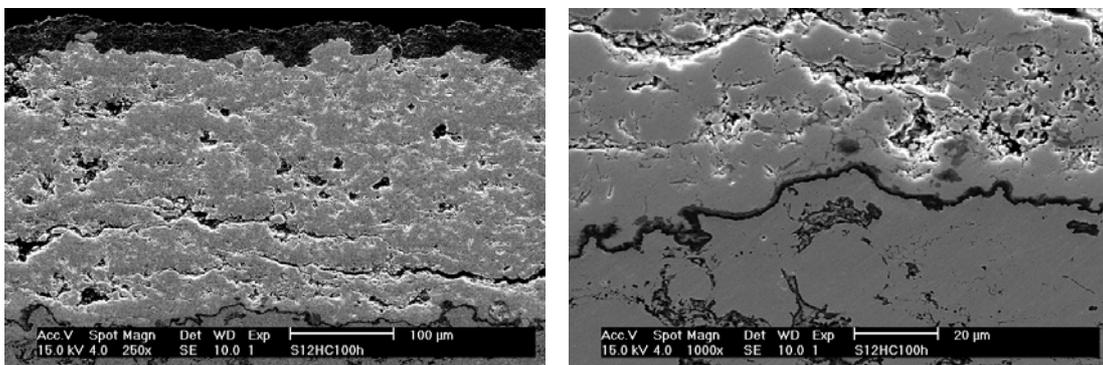


Figure 4 SEM cross-section images of TBC with Al_2O_3 overlay sample after exposure to the molten salt at $950\text{ }^\circ\text{C}$ for 100 hours

It can be seen that the thickness of thermally grown oxide (TGO) in TBC sample without Al₂O₃ overlay was reduced to 2.6-4.0 μm , due to the molten salt deposited on the surface which acted as a barrier layer for oxygen diffusion into the YSZ coating. For TBC sample with Al₂O₃ overlay, however, the thickness of thermally grown oxide (TGO) was similar to that when exposure to air, indicating the less influence of molten salt covering on the TBC surface, because the Al₂O₃ overlay has already acted as a better barrier layer for oxygen diffusion into the YSZ coating than molten salt covering.

5. PLANS FOR THE NEXT REPORTING PERIOD

A final report will be carried out.

6. CONCLUSION

Al₂O₃ overlay deposited on the YSZ TBCs surface can not only reduce the hot corrosion rate, but also significantly prevents the bond coat from oxidation either exposure to air or exposure to the molten salt.

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