Impermeable thin Al_2O_3 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 *mm* thick Al_2O_3 overlay were deposited by HVOF thermal spray, respectively, onto to the surface of YSZ coating. In the next reporting period, we will measure or calculate the residue stress within Al_2O_3 overlay and YSZ coating to study the mechanism of effect of Al_2O_3 overlay on spalling of YSZ coating. However, due to the thermal expansion mismatch between YSZ coating and Al_2O_3 overlay, such surface modification using Al_2O_3 overlay might deteriorate strain tolerance of the TBC. In the present work, in order to investigate the effect of Al_2O_3 overlay on residual stress developed in the samples during cooling after hot corrosion at high temperature, Finite Element method (FEM) was employed to determine the detailed stress states in the test specimens after cooling.

The results showed that there is no high stress concentration at the interface between the YSZ and the bond coat for TBCs system without Al_2O_3 overlay. On the other hand, the maximum compressive stress with a value of approximately, -330 MPa occurred within the Al_2O_3 overlay. The maximum tensile stress in YSZ coat near the Al_2O_3 overlay is in the range of 10-133 MPa. The maximum compressive stress of approximately -160 MPa occurred near the YSZ-bond coat interface. X axis stress play a dominant role in influencing the coating failure and sappling.

In the next reporting period, we will study the thickness of Al_2O_3 overlay on hot corrosion resistance and spalling of YSZ coating.

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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 vane. The most common TBC materials is Y_2O_3 (8wt%)-stabilized ZrO_2 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 V_2O_5 or NaVO_3 to form YVO_4 , causing structural destabilization of ZrO_2 (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 (In_2O_3) [11, 12], scandia (Sc_2O_3) [13] and ceria (CeO_2) [8,14] as well as Ta_2O_5 [6,15] and YTbO_4 [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°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.

However, due to the thermal expansion mismatch between YSZ coating and Al_2O_3 overlay, such surface modification using Al_2O_3 overlay might deteriorate strain tolerance of the TBC. In the present work, in order to investigate the effect of Al_2O_3 overlay on residual stress developed in the samples during cooling after hot corrosion at high temperature, Finite Element method (FEM) was employed to determine the detailed stress states in the test specimens after cooling.

2. EXECUTIVE SUMMARY

There is no high stress concentration at the interface between the YSZ and the bond coat for TBCs system without Al_2O_3 overlay. On the other hand, the maximum compressive stress with a value of approximately, -330 MPa occurred within the Al_2O_3 overlay. The maximum tensile stress in YSZ coat near the Al_2O_3 overlay is in the range of 10-133 MPa. The maximum compressive stress of approximately -160 MPa occurred near the YSZ-bond coat interface. X axis stress play a dominant role in influencing the coating failure and saplling.

3. EXPERIMENTAL

3.1 Fabrication of TBCs and hot corrosion test

The TBC system used in this study consisted of 601 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 $100\text{ }\mu\text{m}$ 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 $200\text{ }\mu\text{m}$ 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.

Hot corrosion test was performed on the TBCs with and without Al_2O_3 coating. 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.

3.2 Finite element analysis of TBCs

Finite element method was employed to determine detailed stress states in the TBCs coated with and without Al_2O_3 overlay as cooling to 25 °C from a stress-free state at 950 °C (hot corrosion temperature). The actual modeling is done using a general computer program known as ANSYS.

3.2.1. Specimen geometry and computational mesh

A thin slice of the test sample, as shown in Fig. 1, was selected as the geometry of the models. The specimen structure is composed of three layers: The bond coat thickness, 0.10 mm; YSZ coat thickness, 0.25 mm; and Al_2O_3 overlay thickness, 0.025 mm.

3.2.2. Material behaviors

For the TBCs system, the materials of each part are assumed to be homogeneous, isotropic and elastic. The material parameters were not considered to be temperature-dependent, in order to make simulation simply. The material data sets are listed in Table 1.

3.2.3. Boundary conditions

Due to the different CTE (coefficient of thermal expansion) values for the materials of TBCs system, both X axis and Z axis expansion (or contraction) occurs as heating (or cooling). The left side of the model shown in Fig. 1 is fixed, and the right side is assigned nodal constraint so that it can move in both X and Z axis.

(a)

(b)

Fig.1 TBCs finite element model. (a) two dimension and (b) three dimension.

Table 1 Materials properties (at room temperature) for indentation test

material	Thickness (mm)	E (GPa)	n	$\alpha (\times 10^{-6} / ^\circ C)$
TBC(t-phase)	250	50	0.1	11
Bond coat	100	200	0.3	14
Alumina overlay	25	375	0.25	8.3

4. RESULTS AND DISCUSSION

Detailed stress distribution in the TBCs system were obtained to reflect the thermal expansion mismatch when the coatings experiencing a temperature decrease of 925 °C from a stress-free state at 950 °C.

The computed X axis stresses (SX) within the TBCs system without Al₂O₃ overlay is shown in Fig. 2. It can be found that the stress in the bond coat is tensile in nature, whereas that in the YSZ coat is compressive. However, there is no high stress concentration at the interface between the YSZ and the bond coat.

Fig.2 Stress in X axis direction in the TBC system without Al₂O₃ overlay.

The computed X axis stresses (SX) within the TBCs system with Al₂O₃ overlay is shown in Fig. 3. It can be seen in Fig. 3, the stress in the X axis direction in the Al₂O₃ overlay is compressive in nature, and whereas the stress in the YSZ coat is tensile adjacent to Al₂O₃ overlay and compressive near the bond coat. The maximum compressive stress with a value of approximately, -330 MPa occurred within the Al₂O₃ overlay. The maximum tensile stress in YSZ coat near the Al₂O₃ overlay is in the range of 10-133 MPa. The maximum compressive stress of approximately -160 MPa occurred near the YSZ-bond coat interface.

Fig.3 Stress in X axis direction in the TBC system with Al_2O_3 overlay

On the other hand, the computed Z axis stress is somewhat lower than X axis stress, and Y axis stress is much smaller. This implies that the X axis stress play a dominant role in influencing the coating failure and sappling.

These results clearly showed that the Al_2O_3 overlay significantly increased the compressive stress developed in YSZ coat near the bond coat upon cooling. This increased compressive stress caused the spalling of TBCs after hot corrosion much easy than that without Al_2O_3 overlay, especially for a thicker Al_2O_3 overlay, as discussed in the previous report. Resent FEM analysis result is consistence with that we studied in the previous report.

5. PLANS FOR THE NEXT REPORTING PERIOD

In the next reporting period, we will study the thickness of Al_2O_3 overlay on hot corrosion resistance and spalling of YSZ coating.

6. CONCLUSION

- (1) The maximum compressive stress with a value of approximately, -330 MPa occurred within the Al_2O_3 overlay.
- (2) The stress in the YSZ coat is tensile adjacent to Al_2O_3 overlay and compressive near the bond coat. The maximum compressive stress of approximately -160 MPa occurred near the YSZ-bond coat interface.
- (3) There is no high stress concentration at the interface between the YSZ and the bond coat for TBCs without Al_2O_3 overlay.

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