



Effect of internal oxidation on the interfacial morphology and residual stress in air plasma sprayed thermal barrier coatings



Jishen Jiang^{a,b}, Zhonghua Zou^c, Weizhe Wang^{a,b,*}, Xiaofeng Zhao^c, Yingzheng Liu^{a,b}, Zhaomin Cao^{a,b}

^a Key Laboratory of Power Machinery and Engineering, School of Mechanical and Power Engineering, Shanghai Jiao Tong University, Shanghai 200240, China

^b Gas Turbine Research Institute, Shanghai Jiao Tong University, Shanghai 200240, China

^c Shanghai Key Laboratory of Advanced High-temperature Materials and Precision Forming, Shanghai Jiao Tong University, Shanghai 200230, China

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ABSTRACT

Significant internal oxidation was found in air plasma sprayed (APS) thermal barrier coating (TBC) samples with double-layered high velocity oxy-fuel (HVOF) NiCoCrAlY bond coats. The volumetric fraction of internal oxides within the bond coat increased from 5% to 38% during 799 h isothermal exposure at 1150 °C, indicating significant swelling of the internal oxides. This swelling behavior not only significantly increases the thickness of the bond coat layer from 175 μm to 260 μm, but also probably elevates the risk of interfacial failure in the TBCs, due to the roughness change at the interface and the huge residual stress generated. Thus, different finite element (FE) models were established to understand the role of the internal oxides in interfacial morphology and residual stress. The internal oxidation behavior was modeled on experimental results. The FE results showed that although the internal oxidation only slightly increased the interfacial roughness, significant changes were found in the patterns of the stress component S22 and the interfacial normal stress along the interface, increasing the risk of micro-cracks and changing the crack locations.

1. Introduction

Thermal barrier coatings (TBCs) have been widely applied in high temperature components of gas turbines and aero engines, due to their superior performance in terms of oxidation, corrosion, and heat resistance [1]. In a TBC system, the metallic bond coat (BC), usually MCrAlY (M = Ni and/or Co) or Pt and Ni diffusion aluminide, serves to bond a ceramic top coat (TC) with low thermal conductivity to a super alloy substrate [1,2]. However, the penetration of oxygen into the BC layer inevitably results in oxidation during processing and operating at high temperatures. Precipitation of oxides leads to the formation of a thin thermally grown oxide (TGO) layer at the TC/BC interface, and the dynamic and uneven growth of this layer due to continuous precipitation significantly changes the interfacial morphology of TC/BC. Internal oxides in the BC layer then transform the properties and microstructure of the BC layer [3–5]. Previous studies have focused on TGO layers with huge stress concentrations, which play an important role in the premature failure of TBCs [1–9]. However, the swelling behavior in the BC layer due to internal oxidation significantly influences the morphology and residual stress of TBCs, which will likely change the failure

behavior of the TBC system. Thus, identifying the extent of the influence of internal oxides on the interfacial morphology and stress state of TBCs can help further understand the failure mechanisms of TBC systems.

Internal oxidation is widely found in the BC layer, particularly if it is manufactured by air plasma spraying (APS) [3–5]. During high temperature exposure, the internal oxides continuously grow within the BC, accompanied by precipitation of the TGO layer, accelerating the depletion of Al. Patterson et al. [3] found that the proportion of internal oxides could reach up to 50% (in area) in APS CoNiCrAlYSi BC after 120 thermal cycles at 1121 °C. They concluded that the APS TBC samples had long lifetimes during thermal cycling, though the fraction of internal oxides was much larger than in the conventional APS TBC samples. The extension of TBC lifetimes was attributed to the alleviation of the thermal mismatch between the BC and TC, as internal oxides decrease the overall thermal expansion coefficient (CTE) of BC to a range similar to that of ceramic TC [3]. Our previous study [10] researched that internal oxides with high volume fraction might also have detrimental effect on TBCs lifetime, since their swelling in a double-layered bond coat during thermal exposure slightly increased the interfacial

* Corresponding author at: Key Laboratory of Power Machinery and Engineering, School of Mechanical and Power Engineering, Shanghai Jiao Tong University, Shanghai 200240, China.

E-mail address: wangwz0214@sjtu.edu.cn (W. Wang).

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roughness of TBCs. However, after detailed observation the influence of internal oxides on TBCs failure is found to be more complicated. The microstructure of internal-oxidizing BC can resemble a metal-matrix composite. Once oxidized, the swelling of internal oxides will significantly alter the interfacial morphology and redistribute the residual stress in TBCs, which are both directly related to the failure [3,5–7]. Ahmadian and Jordan [11] suggested that internal oxidation can change the surface shape of BC and may also be partially responsible for the observed rumpling in BC. Wang et al. [5] found from numerical calculations that the “island-like” oxides (namely internal oxides) in BC decreased the stress level in TGO and reduced the crack propagation velocity. The island-like oxides they applied in their finite element (FE) model were assumed to be formed by static oxidation at high temperature. However, as internal oxides, like the TGO layer, dynamically grow during high temperature exposure [3], the changes of interfacial morphology and residual stress in TBC caused by the swelling of internal oxides should be carefully considered. Through experimental tests a new FE model was thus obtained in our study, which included the evolution of internal oxides.

The objective of the present study was to investigate the influence of internal oxides on the interfacial morphology and residual stress redistribution. Samples of APS TBCs with double-layered high velocity oxy-fuel (HVOF) BCs were subjected to long isothermal exposure at 1150 °C. The double layers of BC were produced with different HVOF spraying parameters. The porous upper layer generated internal oxides, while the lower layer provided sufficient Al for oxidation in the upper layer. Finite element models were established to further understand the effect of the internal oxidation on the interfacial morphology and residual stress of TBCs. The growth of TGO and swelling of internal oxides based on experimental measurements were implemented in ABAQUS by a CREEP user subroutine. The effects of volumetric fraction, location, and distribution of internal oxides on the interface morphology and residual stress were then examined.

2. Experiment observation

2.1. Experiment procedure

In the present study, the TBC samples consisted of a 400 µm-thick APS ZrO₂-8 wt% Y₂O₃ (8YSZ) top coat, a 405 µm-thick double-layered HVOF NiCoCrAlY bond coat, and a 3 mm-thick grit-blasted Hastelloy-X substrate, as shown in Fig. 1a. The double layers of BC were produced using the same NiCoCrAlY alloy powders, Amdry 365-2 (Oerlikon Metco), but with different HVOF spraying parameters, following the procedure described in Ref. [12]. The two BC layers were sprayed at constant manipulator speed of 500 mm/s, spray distance of 200 mm, shroud N₂ of 390 SLPM (SLPM = standard liters per minute) and 5 spray passes. For BC I layer, the O₂/H₂ flow was 195/630 SLPM and the powder flow was 15 g/min; while for BC II layer, the O₂/H₂ flow was 155/630 SLPM and the powder flow was 40 g/min. The porosity level of BC layer increased as the O₂/H₂ flow was decreased and the powder flow was increased [12]. The 230 µm-thick lower layer (BC I) was expected to be fully dense and provide sufficient Al for oxidation, while the 175 µm-thick upper layer (BC II) was porous and would produce internal oxides. All bond coated specimens were annealed in a vacuum at 1120 °C for 24 h before the deposition of the top coats. The TC was sprayed at the OEM facility using their process parameters, which were kept proprietary. Isothermal oxidation at 1150 °C was conducted on a batch of samples in a furnace after 10 min heat-up. The samples were exposed at 1150 °C with different time intervals to oxidize, followed by 20 min air-cooling to room temperature for cross-sectional microstructural examination.

Using a scanning electron microscope (SEM, FEI F50) coupled with an energy dispersive spectrometer (EDS, Oxford), the polished cross-sections of the TBC specimens were observed and the microstructural evolution of TBCs as a function of isothermal exposure time was

documented. All images were taken in backscattered electron mode with a 1500 µm × 1300 µm field of view and a numerical resolution of ~0.68 µm/pixel, and the internal oxidation zone was cropped for the final image. Image analysis of 6 randomly selected areas from the cross section was carried out, with a sampling area of > 1.2 mm², using Image J software to quantify the volume fraction of internal oxides within BC II [10]. The internal oxides were recognized and summed up by grey scale analysis of the experimental images, and the interfacial TGO layer was not included in the measurement.

2.2. Experimental results

Fig. 1 shows the SEM images of cross-sectional microstructures of TBC samples before thermal exposure (Fig. 1a, b, and c) and after 799 h isothermal exposure at 1150 °C (Fig. 1d). Fig. 1a shows that two layers of BC integrate together with no clear boundary, due to the same deposition techniques and post-deposition vacuum treatments. Both layers of BC consist of ~41.5% vol. β-NiAl (grey color) phase and ~58.5% vol. γ-Ni phase (grey color), shown in Fig. 1b and c. Due to the poor molten state of NiCoCrAlY particles in BC II, the amounts of micro-pores and inter-splat gaps appearing in the BC II layer (with approximate 5% porosity) are more than those in the BC I layer (with approximate 1.2% porosity), which provide sufficient place for internal oxidation in fully dense BC II. In addition, the poor molten state also leads to large surface roughness of BC II. The original roughness, R_a, reaches ~20 µm, which is larger than that of conventional APS TBC samples. During the long time exposure, oxidation occurs both at the TC/BC interface and within the BC II layer. The TGO layer grows in thickness from 1 µm to 10.5 µm at the rough interface, and internal oxides also form and grow on the surfaces of pores and gaps in BC II with an increasing volumetric fraction. As a result, after 799 h oxidation, the fraction of internal oxides in BCII was up to 38%, while the oxides in BC I is negligible (see Fig. 1d). Some of the internal oxides adjacent to the interface are connected to the TGO layer and most sprawl irregularly within BC II, resulting in an intricate BC microstructure. The presence of internal oxides expands the volume and increases the thickness of the whole BC layer.

After long isothermal oxidation, interfacial cracks appear in some samples, as shown in Fig. 1d. These cracks initiate at the TC/TGO/BC interfaces or penetrate through the whole TGO layer. Finally, macro interfacial delamination appears due to the coalescence of micro-cracks initiated around the interface. Internal oxidation is expected to be non-negligible in the initiation and propagation of interfacial crack in TBCs, as the high internal oxide content greatly changes the microstructure of BC so it resembles a metal-matrix composite. The interfacial properties and the stress distribution of the TBCs vary because of internal oxidation. Finite element models are thus developed for detailed stress analysis to examine the role of internal oxidation, which is directly related to the failure of TBCs.

3. Numerical modeling

3.1. Oxidation behavior

Oxidation at the surface of and within the BC layer occurs simultaneously during high temperature exposure. The growth of TGO is predominantly governed by the inward diffusion of oxygen along the TGO grain boundaries. Once the alumina scale forms, it can be regarded as a diffusion barrier to retard further oxidation of the metallic BC. Fig. 2 shows the dependency of the average TGO thickness on isothermal exposure time at 1150 °C. The isothermal TGO growth can be described by a phenomenological equation, followed by [13]:

$$h_{TGO} = k_p \cdot (t_{Tmax})^n \quad (1)$$

where h_{TGO} is the average TGO thickness, t_{Tmax} is the dwell time at the maximum temperature, k_p is the oxidation coefficient, and n is the

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