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Microstructural evolution at interfaces of thermal barrier coatings during isothermal oxidation

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ABSTRACT

Using transmission electron microscopy, we investigate microstructural evolution at interfaces of a plasma sprayed thermal barrier coating system with an yttria–stabilized zirconia top coat and a NiCrAlY bond coat after isothermal oxidation at 1100 °C for up to 50 h. Interestingly, thermally grown oxide (TGO) layers split into two sub-layers. In principle, one of the two sub-layers comprises a mixture of α-Al_2O_3, Cr_2O_3 and $Ni(Al, Cr)_2O_4$, while the other one consists of dense α-Al_2O_3 and Cr_2O_3. Main microstructural evolution of the TGO layers follows a similar sequence that: original phases $\rightarrow$ α-$Al_2O_3 + Cr_2O_3 \rightarrow \alpha$-$Al_2O_3 + Cr_2O_3 + Ni(Al, Cr)_2O_4$. Particularly, after 50 h oxidation, γ-Ni and NiO form in the upper TGO sub-layer at the top coat–bond coat interface, and γ-Ni and α-Cr form in the upper TGO sub-layer at the bond coat-substrate interface. Moreover, γ-Al and β-Ti precipitate at the TGO-substrate interface after 50 h oxidation.

1. Introduction

Thermal barrier coatings (TBCs) fabricated via electron beam physical vapor deposition (EB-PVD) [1,2] or air/vacuum plasma spray (A/VPS) [3] are widely used as surface protection systems in jet engines and gas turbines, because they can enhance the overall engine efficiency by increasing the operating temperature up to 200 °C [4,5]. Usually, the TBCs consist of a ceramic top coat, primarily acting as a thermal insulator to reduce the heat transfer to metallic substrates [6], and a metallic bond coat, which protects the substrates from high temperature oxidation and/or corrosion and joins the top coat and the substrates firmly. For over 30 years yttria partially stabilized zirconia (YSZ) has been the prime choice for ceramic top coat owing to the fact that its thermal conductivity is over one order of magnitude lower than that of Ni-based superalloys. In regard to bond coats, both MCrAlY-type (M = Fe, Co and/or Ni) overlays [7] and Pt modified aluminide coatings [8] are prevailing. Additionally, when the TBCs are in service, thermally grown oxide (TGO) layers often form as a result of oxidation of bond coats.

Although different mechanisms that control TBCs failure have been reported, generally accepted key factors are mismatch of thermal-expansion coefficient between ceramic top coats and bond coats, oxidation, and continuous change in compositions, microstructures, interfacial morphologies derived by the element interdiffusion between coats and superalloy substrates [1]. The above key factors are shown simultaneously at the interfaces due to dramatic composition change, the highest thermal-expansion mismatch and severe stress concentration at the interfaces, thus making the interfaces the weakest sites of TBCs. For example, the TGO formed at the top coat–bond coat interface has been identified as the driving force for coating degradation when a TBC is thermally exposed at high temperatures [9]. Using micro-mechanical test the TBC failures have been investigated intensively (e.g. [10,11]), however, interfacial behaviors, such as element segregation and phase evolution which are important sources of mechanical stress, have not been paid enough attention.

Despite the fact that a few studies have investigated the interfacial behaviors of TBCs systems, they only focused on either top coat–bond coat interfaces [12–14] or bond coat-substrate interfaces [11,15]. As a complete TBC system, interaction among top coats, bond coats and substrates always occur. For instance, element interdiffusion usually exhibits a continuous gradient across top coat–bond coat-substrate interfaces, and the oxidation process relies on penetrating of O from the environment through top coat and bond coat to the substrate, thus performance of the top coat–bond coat interface will influence that of the bond coat–substrate interface. Hence it is necessary to study both top coat–bond coat and bond coat–substrate interfaces simultaneously

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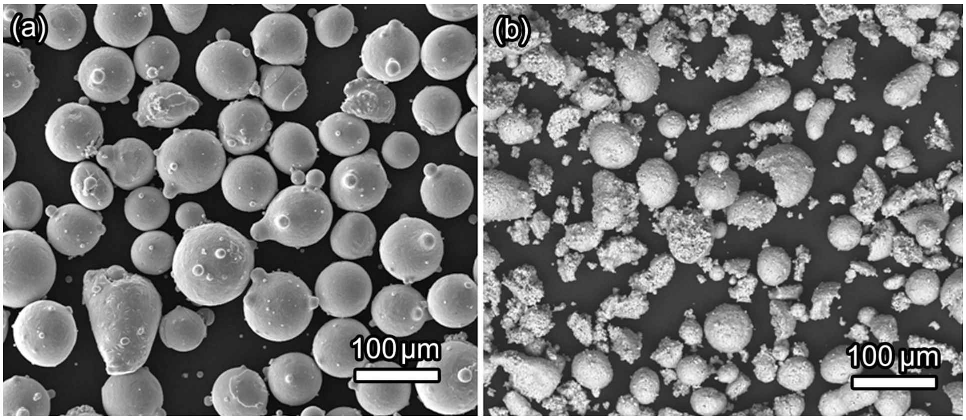

Fig. 1. SEM images showing the morphologies of (a) Metco Amdry 962 (Ni-22Cr-10Al-1Y,wt%) and (b) AI-1075 (ZrO_2–$8Y_2O_3$,wt%) powders, used for fabrication of the bond coat and the top coat, respectively.

in a complete TBC system. To our best knowledge, the only work covering both top coat–bond coat and bond coat–substrate interfaces of a complete TBC system was conducted by Wu and Reed [16], which implies that TBC compatibility depends upon substrate composition. However, they [16] did not provide any information about microstructural evolution.

To study the entire process of the failures of TBCs, long time-intervals of thermal oxidations have often been used [16,17]. For example, Wu and Reed [16] studied the compatibility of superalloy with a TBC system after 100 h, 250 h and 500 h oxidation. Schilbe [17] investigated element diffusion of substrate alloy in a TBC system after cyclic oxidation at 1408 K for 400, 600 and 1050 cycles. However, structural changes at the initial stage which is crucial to understand the failure mechanisms could be missed since structural evolution can happen in just a few hours. Therefore, it is necessary to investigate the interfacial behaviors with a short time-interval, especially in the early oxidation stage.

As for analyzing microstructural evolution, transmission electron microscopy (TEM) can not only provide basic morphology and phase information, but also supply elemental mapping over a large area with a high spatial resolution when working in the model of scanning transmission electron microscopy combining energy-dispersive X-ray spectroscopy (STEM-EDS). Although TEM has such a big advantage on studying microstructures, previous researches [18–20] only implemented TEM on top coat–bond coat interfaces at one stage of oxidation process as a result of difficulty in preparation of cross-sectional TEM specimens [21]. Consequently, it is meaningful to technically elucidate structural evolution at all interfaces of a complete TBC system and at as many early stages as possible.

The aim of this work is to analyze in depth the interfacial microstructure evolution of a plasma sprayed TBC system by a combination of scanning electron microscopy (SEM), X-ray diffraction (XRD), TEM and STEM-EDS. The initial interfaces among top coat, bond coat, and substrate, as well as TGO layers formed at the initial interfaces after a series of isothermal oxidation at 1100 °C, all are named as interfaces here and investigated systematically. The results of this study will be helpful to understand the failure mechanism of TBC systems.

2. Experiments

2.1. Sample fabrication

A Ni-based single crystal superalloy with a composition of 8Co–8Cr–8W–6Ta–5Al–1Ti–0.001P–0.0005S (wt%) was used as the substrate, which has a diameter of 40 mm and a thickness of 3 mm. Metco Amdry962 (Ni–22Cr–10Al–1Y, wt%; Sulzer Metco Inc., New York, USA) and AI-1075 (ZrO_2–$8Y_2O_3$, wt%; Praxair Surface Technologies Inc., Indiana, USA) powders were used for fabrication of the bond coat and the top coat, respectively. The nominal diameters of the Metco Amdry962 and AI-1075 powders are 25–75 μm as shown in the typical SEM images of Fig. 1.

Table 1
Spray parameters for bond coat (NiCrAlY) and top coat (YSZ).

Spray parameters	NiCrAlY (VPS)	YSZ (APS)
Current (A)	500	650
Voltage (V)	125	160
Pressure of primary gas Ar (MPa)	0.52	0.48
Pressure of second gas H_2 (MPa)	0.45	0.41
Flow rate of primary gas Ar (m^3/h)	2.83	2.55
Flow rate of second gas H_2 (m^3/h)	0.42	0.57
Powder feeding rate (g/min)	30	25
Spray distance (mm)	150	100
Pressure of the spraying chamber (Pa)	10^3	

Before coating fabrication, surfaces of the substrates were grit-blasted with an 8070P-B sandblasting machine (sand specifications: 0.3 mm white corundum (α-Al_2O_3); blasting strength: 0.7 MPa) so as to facilitate bonding of the coatings, then cleaned via ethanol and acetone mixture in a KQ-100B type ultrasonic cleaning machine, finally dried by a DHG-9031 type electric thermostatic drying oven.

Afterwards, the grit-blasted substrates were interposed into a special chucking appliance and preheated to the spraying temperature. The TBC system was deposited by a Metco 9 M PS machine equipped with a vacuum plasma spray (VPS) system. First, using a VPS process, a NiCrAlY bond coat with a thickness of 50–70 μm was fabricated via the Metco Amdry962 powder. Then, by air plasma spraying (APS), a YSZ top coat with a thickness of 100–150 μm was sprayed via the Metco 204B-NS powder. The spray parameters are listed in Table 1.

2.2. Isothermal oxidation

Using an improved SNOL 4/1100LSC01 furnace equipped with temperature and time control instrument, isothermal oxidation experiments were performed at 1100 °C in static air. Specimens were respectively mounted in crucibles and put into the furnace at room temperature (22 ± 0.5 °C), then heated to 1100 °C in 40 min, at last, removed from the furnace and cooled down to room temperature with a fan after each holding time.

Nineteen holding times, 0.5 h, 1–10 h ($\Delta t = 1$ h), 15–50 h ($\Delta t = 5$ h) were set for the oxidation. For each holding time 5 samples were oxidized simultaneously. Although all samples were

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