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The effect of preoxidation atmosphere on oxidation behavior and thermal cycle life of thermal barrier coatings

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Abstract

The effects of oxygen partial pressure (pO_2) of the preoxidation atmosphere on the growth of thermally grown oxide (TGO) and thermal cycle life of plasma-sprayed thermal barrier coatings (TBCs) were investigated. The pO_2 of the preoxidation atmosphere was controlled by using a solid-state electrochemical oxygen pump system. The purity and microstructure of continuous Al_2O_3 layer formed on the bond coat during preoxidation at 1050 °C were highly influenced by the pO_2 of the atmosphere. The specimen preoxidized at 1050 °C under a pO_2 of 10^{-14} to 10^{-15} atm, which is around the dissolution pressure of (Co, Ni)(Al, Cr)$_2$O$_4$ spinel, showed the lowest growth rate of TGO and the longest thermal cycle life.

Keywords: Thermal barrier coating; Thermally grown oxide; Preoxidation; Oxygen partial pressure

1. Introduction

Development of advanced thermal barrier coatings (TBCs) is the most promising way to increase the efficiency of gas turbines. Current state-of-the-art TBCs typically consist of an Y_2O_3 stabilized ZrO_2 (YSZ) top coat and a metallic bond coat (MCrAlY, M = Co, Ni). The top coat acts as a thermal insulator, while the bond coat provides oxidation protection for the underlying superalloy by forming protective oxide scale.

It is generally accepted that bond coat oxidation is a critical factor controlling the life of TBCs. When TBCs are used at high temperatures, a thermally grown oxide (TGO) forms at the top coat/bond coat interface. The TGO growth leads to increase residual stress, which accelerates the spallation of TBCs. Recent research has shown that failure of TBCs occurred when the TGO attained a critical thickness in the range of 3–10 μm [1].

The oxidation resistance of a bond coat relies on the ability of the alloy to produce a stable, continuous, slow growing and adherent TGO on its surface. Formation of pure α-Al_2O_3 as a protective layer on the bond coat surface is preferred due to the low diffusion rate of oxygen and metal ions through it, as well as its high chemical and thermal stability [2]. However, the oxidation of MCrAlY usually accompanies fast growing, non-protective oxide phases such as (Co, Ni)(Al, Cr)$_2$O$_4$ and (Co, Ni)O, which are believed to promote the spalling of TBCs [3–5].

Much effort has been directed toward improving oxidation resistance of bond coats to prolong the lives of TBCs. The incorporation of thin protective α-Al_2O_3 between a top coat and a bond coat before use is one of the most promising ways to reduce the rate of TGO growth. A protective interlayer can be formed uniformly by applying heat treatments before or after the deposition of a top coat [6–9] or by using another method such as CVD [10,11].

There are patents which propose that the preoxidation of a bond coat under a low oxygen partial pressure (pO_2) before the deposition of a top coat is effective in suppressing the oxidation due to protective α-Al_2O_3 formation [8,9]. However, no experimental results showing the optimum pO_2 for the preoxidation atmosphere are available in the open literature.

The purpose of this study is to clarify the effect of pO_2 during the preoxidation of bond coats on the growth of TGO at high

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temperature. Thermal cycle lives of the TBCs under controlled preoxidation condition are also determined.

2. Experimental

Co–Ni–Cr–Al–Y alloy (in mass% of Co–32%Ni–16%Cr–8%Al–0.5%Y) was vacuum plasma sprayed to a thickness of 150 μm on grit-blasted substrates (Inconel 738LC) with dimensions of 20 mm × 20 mm × 3 mm. Preoxidation heat treatments of the substrates before the deposition of top coats were performed at 1050 °C for 4 h in an Ar flow with a controlled pO_2 by using a furnace equipped with a solid-state oxygen pump system. Oxygen in the Ar flow was pumped out in a CaO stabilized ZrO_2 tube with an imposed voltage of up to 1.3 V at 800 °C, and then the Ar flow with a controlled pO_2 was introduced into the furnace. The pO_2 in the furnace during the heat treatment was monitored using an oxygen sensor located at the outlet of the furnace. The sensor was calibrated under various pO_2 values by using another oxygen sensor which was inserted directly into the furnace operated at 1050 °C.

Samples fabricated in this study are listed in Table 1. Sample A was preoxidized in air, while sample B was preoxidized in an Ar flow without controlling the pO_2. The flow rate of Ar was 2×10^{-4} m^3/min. The purity of Ar was >99.9999% with a pO_2 of about 10^{-4} atm. The pO_2 of the atmosphere during preoxidation of sample B was measured to 10^{-12} to 10^{-13} atm. Samples C and D were annealed in an Ar flow with a reduced pO_2 of 10^{-14} to 10^{-17} atm. A sample without preoxidation (sample E) was also prepared for comparison.

After the preoxidation, ZrO_2–8 wt.% Y_2O_3 top coats were air-plasma sprayed on the substrates to a thickness of about 300 μm. Samples without top coats were also produced for analysis of the TGO.

Samples were subjected to an oxidation test at 1200 °C in air. The thickness of the TGO formed by oxidation was measured by cross-sectional images. A scanning electron microscope (SEM, Hitachi S-4500) equipped with an energy dispersive X-ray spectrometer (EDS) was employed to investigate the microstructure of the TGO. TGO phases were analyzed by X-ray diffraction (XRD, Philips, PW1877) using Cu Kα radiation. Photostimulated Cr^{3+} luminescence spectroscopy (PSLS) [12] was also used for phase identification in the initial TGO.

The element distribution as a function of depth below the initial TGO surface was determined by secondary ion mass spectrometry (SIMS, Physical Electrons ADEPT1010). The depth profiling was performed using a 5 keV Cs^+ beam over an area of about 88 μm × 144 μm up to a depth of 3 μm. The results were quantified using a profile of an Al–Cr–Ni–Co–O sintered compact as a standard.

Table 1
Samples used in this study

Sample	pO_2 during preoxidation at 1050 °C
A	0.2 atm (in air)
B	10^{-12} to 10^{-13} atm
C	10^{-14} to 10^{-15} atm
D	10^{-16} to 10^{-17} atm
E	Without preoxidation

Thermal cycle tests of the samples were performed using a vertical furnace. The heating time was 10 min at 1150 °C, then the samples were moved into the water at 25 °C for 2 min. Before the testing, samples were oxidized in air at 1200 °C for 50 h. The lives of the coatings were determined by the number of cycles at which the coating failure occurred.

3. Results and discussion

3.1. Analysis of initial TGO formed by preoxidation

Samples that had not been overlaid with top coats were analyzed by XRD, SEM and SIMS in order to investigate the phases in the initial TGO. Fig. 1 presents XRD patterns of the bond coat surface of samples after preoxidation. Besides Co–Ni–Cr–Al–Y peaks (γ/γ′ and β), α-Al_2O_3 and (Co, Ni)(Al, Cr)$_2$O$_4$ spinel are identified in sample A, showing that the initial TGO consists mainly of these two phases. In contrast, only α-Al_2O_3 is identified as a phase of the initial TGO in samples B–D.

Fig. 2 shows the surface morphologies of the initial TGO of samples A–D. Blade-like crystals are observed on the surfaces of samples A and B, whereas such crystals are not seen in samples C and D. PLPS measurements showed evidence of both θ- and α-Al_2O_3 on the surfaces of samples A and B, showing that the blade-like crystals are transient Al_2O_3 formed during the transformation from metastable γ- and/or θ-Al_2O_3 to α-Al_2O_3 as reported previously [13,14]. The transient Al_2O_3 is believed to be deleterious because the growth rate of θ-Al_2O_3 as a TGO is an order of magnitude higher than that of α-Al_2O_3 [15]. The large decrease in volume that accompanies transformation from γ- and/or θ-Al_2O_3 to α-Al_2O_3 at high temperature will also be detrimental to the TBC life [2,13,16]. On the other hand, only α-Al_2O_3 was identified by PLPS for samples C and D. It is noteworthy that the formation of transient Al_2O_3 shows dependence on the pO_2 of the preoxidation atmosphere. The fundamental

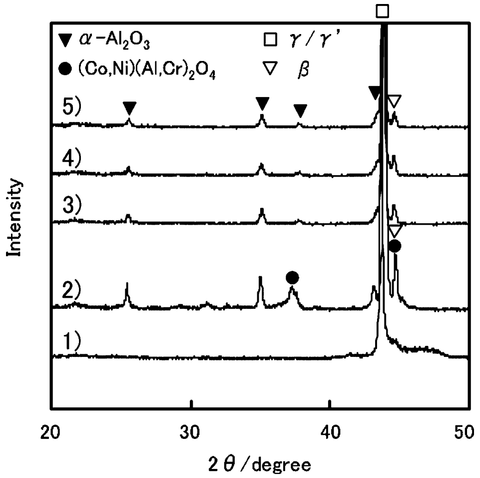

Fig. 1. XRD patterns of the bond coat surfaces of samples before overlaying with top coat: (1) sample E, (2) sample A, (3) sample B, (4) sample C and (5) sample D.