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CRACKING BEHAVIOUR OF AN AIR-PLASMA-SPRAYED THERMAL BARRIER COATING

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ABSTRACT

The degradation of a thermal barrier coating (TBC) system is dominated by the formation and growth of a thermally grown oxide (TGO) layer between the ceramic topcoat and metallic bond coat, leading to a separation near the ceramic/bond coat interface during service. Crack propagation in a thermally sprayed TBC normally proceeds via the opening and growth of pre-existing discontinuities in the ceramic layer near the ceramic/bond coat interface region assisted by cracking associated with the evolution of the TGO. The combined effect of these degradation processes results in premature TBC failure. In the present study, TGO growth and cracking behaviours were investigated under cyclic oxidation conditions with different cycle frequencies. The results showed a likely relationship between the maximum crack length and TGO thickness, suggesting that the establishment of an empirical formula may be possible to serve as the basis for a TBC life prediction model.

INTRODUCTION

The metallic bond coat (BC) is an important constituent of thermal-sprayed thermal barrier coating (TBC) systems. It enhances the adhesion of the ceramic thermal barrier layer (the topcoat or TC) to the substrate and also provides oxidation and corrosion protection to the substrate metal. At elevated temperatures, however, oxidation of the bond coat results in the formation of a thermally grown oxide (TGO) layer at the ceramic/bond

coat interface. Formation of the TGO, preferably composed of a continuous scale of α -Al₂O₃, creates a diffusion barrier during extended thermal exposure in service, thus helping to protect the substrate from further oxidation and improving durability of the system under service conditions. However, oxidation of the bond coat has been recognized as the cause for separation of the ceramic layer from the substrate [1-6].

It has been suggested [7] that the durability of TBC systems is governed by a sequence of crack nucleation, propagation and coalescence events that accumulate prior to final failure by large scale buckling and spalling. An analytical model [7] suggested that TGO thickness in a TBC could be expressed as

$$\delta = \frac{2\sqrt{\pi}(1-\nu^2)mK}{(m-1)RE} a^{3/2} \quad (1)$$

- δ TGO thickness
- a half crack length
- R radius of the imperfection
- m ratio of volume change
- E Young's modulus of the TBC
- ν Poisson's ratio
- K stress intensity factor

Recent work [8] showed a likely power-law relationship between the maximum crack length ($a_{\max}$) in the coating and equivalent TGO thickness (δ_{eq}), i.e.,

$$a_{\max} = k\delta_{eq}^n \quad (2)$$

and an n value of 0.597 was determined in an air-plasma-sprayed TBC system under thermal cycling condition, which is close to that derived in the analytical model [7]. However, a much higher n value was observed in a TBC with high-velocity oxy-fuel (HVOF) CoNiCrAlY bond coat under low-frequency thermal cycling condition [9].

It was reported that longer holding times resulted in a slower mass gain rate during thermal cycling [10]; however, the relationship between holding time and crack propagation has not yet been reported in literature. This work will present the influence of holding time on both TGO growth and crack propagation in an air-plasma-sprayed TBC system. The relationship among the maximum crack length in the coating, TGO thickness and cooling history will also be discussed, to support the establishment of an empirical model for TBC life prediction.

EXPERIMENTAL

The TBC samples consisted of a CoNiCrAlY bond coat and a ZrO_2 -8wt% Y_2O_3 topcoat. The bond coat was deposited with a thickness of 140-180 μm by the air plasma spray (APS) technique (F4-MB, Sulzer Metco, Westbury, NY, USA), with powders of Co-32Ni-21Cr-8Al-0.5Y (wt.%) (Amdry 9951, Sulzer Metco, Westbury, NY, USA), onto $\phi 16 \text{ mm} \times 10 \text{ mm}$ Inconel 625 disks. On top of the CoNiCrAlY, the topcoat was deposited to a thickness of 250-280 μm also by the APS technique, employing the same plasma spray torch, with powders of ZrO_2 -8wt.% Y_2O_3 (Metco 204NS, Sulzer Metco, Westbury, NY, USA). The nominal particle size distributions for the BC and TC powders were 5-37 μm and 11-125 μm , respectively. The BC and TC were produced based on the spray parameters recommended by the manufacturer of the torch and feedstock (Sulzer Metco).

Some of the as-sprayed specimens were heat-treated in a vacuum furnace at $<10^{-5}$ torr ($P_{\text{O}_2} < 0.00028 \text{ Pa}$) and 1080°C for 4 hours (vacuum heat treatment or VHT). Both as-sprayed and VHT TBCs were subsequently subjected to thermal cycling consisting of 8~12-minute ramping, 45-minute holding at 1050°C, and 30~40-minute cooling to the ambient temperature (25°C) in air. Some VHT TBC specimens underwent thermal cycling in air, which consisted of 10~15-minute ramping, 100-hour holding at 1050°C, and >40-minute cooling to the ambient temperature (25°C). The oxidized samples were sectioned after completion of a predetermined time of thermal exposure between 45 minutes and 4500 hours, mounted using epoxy, and mechanically polished. The specimens were then examined using a Philips XL30S FEG scanning electron microscope (SEM) with an energy-dispersive spectrometer (EDS). The area and length of TGO, as well as the crack length in the TBC, were measured using ImageTool software, based on the micrographs taken from the cross sections of the tested samples. The TGO area was measured by drawing a polygon along the ceramic/TGO and TGO/bond coat

interfaces, while the TGO length was determined by drawing a line as short as possible along the TGO. From each specimen, 20-70 micrographs were taken for the TGO measurement. The various oxides were identified by SEM-EDS.

RESULTS AND DISCUSSION

Coating microstructures:

The ceramic topcoat contained porosity and some crack-like discontinuities (Fig. 1a). The maximum dimension of the crack-like discontinuities in the ceramic was in the range of 70-100 μm . The CoNiCrAlY bond coat contained segmented Al_2O_3 veins within the bond coat but also at the interface between the ceramic topcoat and the bond coat, as a result of partial oxidation of the bond coat during coating deposition. Some porosity also appeared at the topcoat/bond coat interface. SEM/EDS semi-quantitative analysis showed that the aluminum concentration was highly non-uniform in the as-sprayed bond coat, ranging from 9.6 to 17.9 at.% Al. This non-uniformity of Al distribution in the bond coat can probably be attributed to the preferential oxidation and/or vaporization in the plasma, because the particle surface temperature was much higher than the melting point of the CoNiCrAlY (~1300°C).

Figure 1: Microstructures of (a) as-sprayed (i.e., no vacuum heat treatment applied) and (b) VHT TBCs with APS-CoNiCrAlY bond coat.

After the VHT, although the Al_2O_3 layer remained discontinuous at the ceramic/bond coat interface, the amount of Al_2O_3 apparently increased (Fig. 1b), compared to that in the as-sprayed condition. The aluminum concentration became rather uniform and was <11 at.% in the bond coat, lower than that of the feedstock powder where the Al level was about 16 at.%. Therefore, to a certain degree the vacuum heat treatment can homogenize the highly non-uniform microstructures resulting from thermal spraying of CoNiCrAlY powders in air.

Oxidation of TBCs in thermal cycling with short holding time:

Upon thermal exposure in air, an oxide layer of predominantly $(\text{Cr},\text{Al})_2\text{O}_3 + (\text{Co},\text{Ni})(\text{Cr},\text{Al})_2\text{O}_4$ (CS) formed along the interface between the ceramic topcoat and the bond coat (Fig. 2a) in the as-sprayed CoNiCrAlY. The mixed oxide scale was found to consist of 5-16 at.% Ni+Co and 28-43 at.% Al+Cr. The Al_2O_3 portion in this oxide layer was apparently less than 20%. At the same time, mixed oxide clusters of chromia, spinel and nickel oxide, abbreviated as CSN, also formed at the ceramic/bond coat interface. As such, the TGO, formed in the as-sprayed TBC, was composed of a CS layer and a number of CSN clusters.

Figure 2: As-sprayed APS-CoNiCrAlY shows (a) a TGO layer of chromia+spinel with CSNs after one 45-min. cycle, (b) discontinuity opening after two hundred 45-min. cycles, and (c) crack propagation and coalescence associated with the TGO after five hundred 45-min. cycles.

As thermal cycling continued, the TGO layer grew thicker (Fig. 2b), and in the meantime, pre-existing crack-like discontinuities in the ceramic developed into fully open cracks. Extensive crack propagation in the ceramic topcoat and associated with the CSNs was also observed after 500 cycles (Fig. 2c). The as-sprayed TBC samples failed at around 500 cycles.

The TGO, formed in the VHT TBC, was composed of a layer of Al_2O_3 and $(\text{Cr},\text{Al})_2\text{O}_3 + (\text{Co},\text{Ni})(\text{Cr},\text{Al})_2\text{O}_4$, as well as some oxide clusters of alumina, spinel and nickel oxide (ASN) and CSNs (Fig. 3a). The Al_2O_3 portion in this TGO was apparently >70%. After several thermal cycles, some chromium-rich oxide, with $\text{Cr}+\text{Al}>35$ at.% and $\text{Ni}+\text{Co}<7$ at.%, started to form underneath the previously formed Al_2O_3 scale, which grew into Al_2O_3 during subsequent thermal exposure (Fig. 3b), leading to the complete transformation from Al_2O_3 to CS (Fig. 3c). The formation of Cr-rich oxide underneath the Al_2O_3 layer in thermal-sprayed TBC systems has been reported in previous studies [11,12], and it could be attributed to aluminum depletion in the bond coat such that a composition was reached at which other oxide(s) could form [13]. As demonstrated in the as-sprayed TBC, the opening of pre-existing discontinuities also occurred in the VHT TBC (Fig. 3b), which led to crack propagation and coalescence, forming longer cracks in the ceramic after extensive thermal cycling (Fig. 3c). One of the VHT specimens lasted only 1470 cycles, but another did not fail until after 2100 cycles.

Oxidation of TBC in thermal cycling with long holding time:

Only the VHT APS-CoNiCrAlY specimens were tested in the thermal cycling with long holding time, which consisted of 10-15-minute ramping, 100-hour holding at 1050°C, and >40-minute cooling to the ambient temperature (25°C). The oxidation performance

was nearly the same as that in 45-min./cycle thermal cycling: the TGO was initially Al_2O_3 with some CSNs (Fig. 4a), followed by the formation of Cr-rich oxides (Fig. 4b) and a complete transformation from Al_2O_3 to chromia+spinel (Fig. 4c). It was noticeable that crack propagation in the coating was reduced in the 100-hour/cycle thermal cycling condition (Fig. 4a), compared to the 45-min./cycle thermal cycling environment (Fig. 3b), although the former experienced a longer time of thermal exposure.

Figure 3: VHT APS-CoNiCrAlY shows (a) a TGO layer with >70% Al_2O_3 after ten 45-min. cycles, (b) the growth of the Cr-rich oxide into previously formed Al_2O_3 scale and discontinuity opening in the ceramic layer after five hundred 45-min. cycles, and (c) crack propagation and coalescence within the ceramic layer near the TGO as a result of discontinuity opening and associated with the TGO growth after twenty-one hundred 45-min. cycles.

TGO growth and crack propagation in the TBCs:

To take into account the entire TGO, an equivalent TGO thickness, δ_{eq} , is defined as [14]:

$$\delta_{eq} = \frac{\Sigma (\text{cross sectional TGO area})}{\Sigma (\text{cross sectional length of TC/BC interface})} \quad (3)$$

Figure 4: VHT APS-CoNiCrAlY shows (a) a TGO layer with predominantly Al_2O_3 after five 100-hour cycles, (b) the growth of the Cr-rich oxide into previously formed Al_2O_3 scale and discontinuity opening in the ceramic layer after ten 100-hour cycles, and (c) crack propagation and coalescence within the ceramic layer near the TGO as a result of discontinuity opening and associated with the TGO growth after forty-five 100-hour cycles.

Use of this approach was due to the rough TC/BC interface, as well as heterogeneous TGO growth especially when CSN clusters were formed between the ceramic topcoat and the bond coat (Figs. 2-4). The relationship between δ_{eq}^2 and exposure time (Fig. 5) shows a parabolic growth of TGO, which is consistent with previous observations [11,15-17]. It can also be seen that VHT can reduce the growth rate of the TGO layer; this can be attributed to the reduced formation of mixed oxides during thermal exposure. Moreover, slower TGO growth can be noticed when tested in 100-hour/cycle thermal cycling condition, which is also consistent with previous observation [10].

Figure 5: (a) The growth of TGOs in APS-TBC during thermal cycling, and (b) the early stage of TGO growth.

The longest crack found in viewing all areas of the cross-section of the coating was used to study the cracking behaviour in the TBCs, which was assumed to be the crack of maximum length in the coating. In general, the maximum crack length in the TBCs increased as exposure time increased (Fig. 6). It may be noticed that cracks propagated faster in the as-sprayed APS-CoNiCrAlY specimens than in the VHT TBCs, and that the cracks propagated even slower in the VHT TBCs under 100-hour/cycle thermal cycling condition. When plotting the maximum crack length as a function of TGO thickness (Fig. 7), it appears that crack length increased with TGO thickness. Regression analysis shows that the

relationship between the maximum crack length in the TBC and TGO thickness can be expressed by a power law, i.e. Equation (2). It is interesting to note that the data determined from the 45-min./cycle and 100-hour/cycle thermal cycling tests fall on nearly the same curve, where the value of n is determined to be 0.7074, although some scatter can be observed.

Figure 6: The maximum crack length in the TBC as a function of the time of thermal exposure.

Figure 7: The maximum crack length as a function of TGO thickness in TBCs with APS-CoNiCrAlY bond coat.

Even though it has been reported that longer holding times resulted in a slower mass gain rate during thermal cycling [10], a thorough understanding of the effect of holding/cycle time on crack development has not been achieved in the literature.

It has been shown that the critical crack size for initiating TBC spallation is somehow related to the topcoat thickness [18]. It is possible that TBCs with different topcoat thickness values would fail at different TGO thickness levels. As such, understanding of the relationship between crack length and TGO thickness, as well as the influence of spraying process on such a relationship, may be helpful in TBC life prediction. This suggests that the nature of such a relationship merits further investigation in both numerical and experimental studies.

CONCLUSIONS

The present work investigated the cracking behaviour in an air-plasma-sprayed TBC system under different thermal cycling conditions. Cracking appeared to proceed via crack propagation in the ceramic layer along with crack nucleation and propagation associated with the TGO, leading to premature TBC failure.

The results also showed a likely power-law relationship between the maximum crack length and the TGO thickness in APS-TBC, with holding time having little influence on such relationship.

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