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A prominent driving force for the spallation of thermal barrier coatings: Chemistry dependent phase transformation of the bond coat

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ABSTRACT

The influence of substrate and bond coat chemistry on the degradation mechanism leading to the early spallation of thermal barrier coatings (TBCs) has not been well understood despite years of research effort. This is largely due to the sheer number of factors (i.e. interfacial rumpling and oxide growth kinetics) that all seem to contribute to the degradation of TBCs. To clarify the chemical effect, extensive characterizations and in-depth analysis near the oxide-bond coat interface, were carried out on the isothermally exposed TBC specimens. It is evident that the formation of γ' along the grain boundaries can significantly enhance rumpling, while martensitic transformation during cooling creates out-of-plane stresses and causes crack nucleation at the oxide-bond coat interface. These partial phase transformations in the β bond coat system were determined to be a prominent driving force for the TBC spallation. To prevent the early spallation of TBCs, it is necessary to minimize the formation rate of γ' and martensitic phases, which can be achieved by tailoring the inherent substrate/bond coat composition as elucidated in this paper.

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1. Introduction

Diffusion coating has been widely adopted as an intermediate bonding material in state-of-the-art thermal barrier coatings. This type of bond coat has tailored compositions to ensure a stable formation of an adhesive alumina scale upon oxidation [1–4]. Despite being oxidation resistant, these TBCs are prone to spallation failure near the TGO/bond coat interface, driven primarily by the interfacial rumpling of the bond coat during the course of high temperature oxidation and thermal cycling [5–8]. Rumpling is generally believed to be caused by the mismatch in the coefficient of thermal expansion between the multilayered structures of TBCs and the lateral growth strain of oxide [7–9].

Although previous modeling work based on the micro-mechanics of the coating systems provided valuable insight into the TBC degradation behavior, the predicted results (i.e. stress state and rumpling amplitude) often deviate from those obtained from real experimental specimens. This is largely due to the lack of consideration of the influence of bond coat and substrate chemistry on the interfacial evolution (i.e. material properties) of TBC system

[7,8,10]. These modeling works, calculated based on simplified geometric and material properties, are only capable of evaluating the relative influence of a particular parameter (i.e. bond coat creep strength) on the overall failure process. In order to make accurate prediction of TBC life, it is necessary to account for the exact material properties, and their evolution with thermal cycling [11].

Many recent literature [12–14] suggested that the TGO stress evolution and failure mechanism near the TGO/bond coat interface are significantly influenced by both bond coat and superalloy compositions. The lifetime of TBCs were shown to vary more than 3 folds with slight differences in the inherent Pt, Al, Re, Ru, and Ti contents of the bond coat and superalloy. In a recent work by Zhao et al. [15], the inward Pt diffusion and the subsequent release of Ti into the TGO layer accompanied by γ' to γ transformation, were found to degrade the lattice boundary coherency at the TGO/bond coat interface, which may lead to the degradation of interfacial adhesion.

The effect of phase transformation on the TBC degradation and failure has also been studied previously. In the modeling works by both Chen [16] and Balint [17], the volume change associated with the β to martensitic transformation was shown to have a significant influence on the stress distribution and interfacial rumpling in platinum modified nickel aluminide bond coat. On the contrary,

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Tolpygo [18] maintained that the volumetric shrinkage associated with both β to martensitic and γ' has rather small contribution to the interfacial rumpling of TBCs.

In a previous work by the authors [19], the oxide-bond coat interfacial adhesion of several commercial TBCs was measured semi-quantitatively using a cross-sectional indentation technique. The crack rates (i.e. linear slopes of crack lengths vs isothermal exposure time plot) were then plotted against various common TBC degradation parameters (i.e. linear slopes of TGO growth kinetics vs isothermal exposure time plot), in order to identify the key parameters influencing the interfacial adhesion. β to γ' phase transformation was determined to be one of the most important factors affecting the oxide-bond coat interfacial adhesion. The SRR99 LT Pt-Al and the TMS-82+ HT Pt-Al TBC systems, both of which had relatively faster γ' transformation rate, were found to have the worst interfacial adhesion and spallation lifetime (See Fig. 1). Nonetheless, it remained unclear as to how the phase transformation degrades the interfacial adhesion.

Preliminary analysis by electron probe microscope analysis (EPMA) indicated that while traces of detrimental TiO_2 were present in the TGO layer of the SRR99 LT Pt-Al specimen, the same oxide formation was not present on the TMS-82+ HT Pt-Al specimen with similar Ti enriched γ' formation near the TGO/bond coat interface (see Fig. 2). This implies that the formation of less adhesive oxide cannot be the sole reason leading to the interfacial degradation of TBCs.

The current manuscript, therefore, seeks to clarify previously unanswered questions concerning the phase transformation induced interfacial degradation mechanism of the bond coat by focusing on the comparison between the evolution of interfacial adhesion [19] and phase distribution with time, and correlating the role of bond coat and substrate chemistry with those degradation phenomena. A systematic analysis on the basis of Electron backscatter diffraction (EBSD) and EPMA chemical mapping was carried out by examining the same set of systematically prepared TBC samples as in the previous study [19]. The results clearly indicated that the γ' formation at the grain boundary of the bond coat and

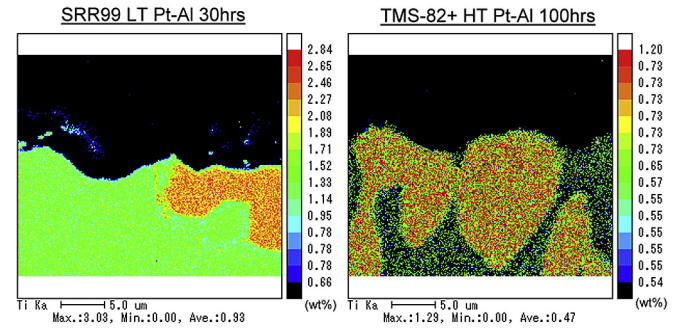


Fig. 2. EPMA mapping showing the formation of Ti concentrated γ' in the bond coat region near the oxide-bond interface of the SRR99 LT Pt-Al (containing traces of TiO_2 in the TGO) and TMS-82+ HT Pt-Al after 30 and 100 h of isothermal exposure, respectively.

martensitic transformation near the interface act as the prominent driving force for TBC spallation. The extent of these phase transformation phenomena was found to depend on the inherent Al, Ni, and Ti contents of the as-fabricated bond coat and substrate. A thermodynamic analysis indicated that Cr and Ta contents, if given high enough amount in the bond coat, could alleviate these detrimental phase transformation by stabilizing the β and γ' phases.

2. Experimental details

The same set of TBCs, consisting of three industrial standard single-crystal nickel based superalloys: CMSX-4, SRR99, and TMS-82+, used in the previous study [19], was utilized in this current

Table 1
Nominal compositions of the three superalloys used in this work (wt%).

Substrate	Co	Cr	Mo	W	Al	Ti	Ta	Hf	Re	Ru	C	Ni
SRR99	5.0	8.0	—	9.5	5.5	2.2	2.8	—	—	—	—	Bal.
TMS-82+	7.8	4.9	1.9	8.7	5.3	0.5	6.0	0.1	2.4	—	—	Bal.
CMSX-4	9.6	6.5	0.6	6.4	5.6	1.0	6.5	0.1	3.0	—	—	Bal.

Interfacial adhesion vs. γ' formation

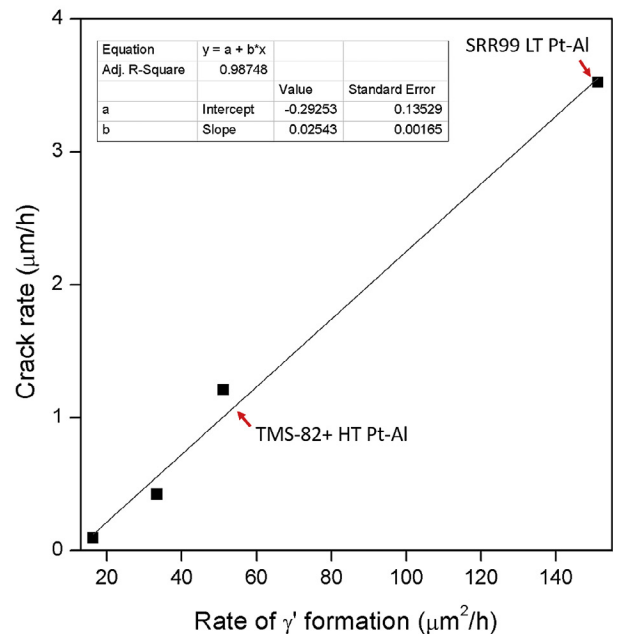
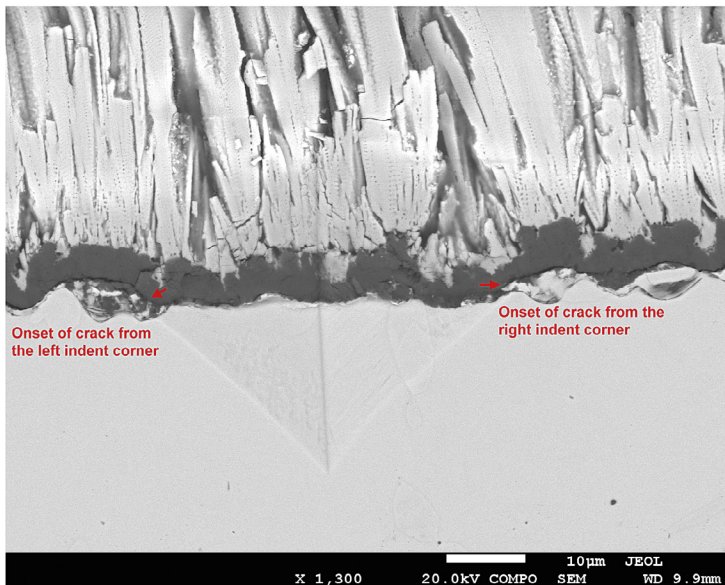


Fig. 1. Previous results suggesting that a linear relationship exists between the interfacial adhesion and the rate of gamma prime formation [19].

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