Bond coatings with high rumpling resistance: Design and characterization

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ABSTRACT

The behavior of two new high-strength γ'-Ni$_3$Al-base bond coats have been compared to a baseline β-(Pt,Ni)Al bond coat via thermal cycling to a maximum temperature of 1204 °C. The rumpling amplitude of the γ' coating is significantly lower than the (Pt,Ni)Al as characterized by a nondestructive optical profilometry technique. The γ' coatings resist rumpling and spallation of the thermally grown oxide in spite of faster oxidation compared to Pt-modified NiAl. This behavior is analyzed using an analytical rumpling model to explain that the improved bond coat strength is the major contributing factor to rumpling inhibition.

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1. Introduction

Bond coats are an integral component of the thermal barrier coating (TBC) system in turbine engines and must satisfy two main requirements. First and foremost, the coating acts as a primer layer for the ceramic topcoat and provides compatibility with the underlying metallic superalloy [1]. Maintaining topcoat adhesion is paramount for protecting the superalloy from the high-temperature combustion environment. Second, a bond coat is required to protect the superalloy from the oxidizing atmosphere by serving as an aluminum reservoir from which the coating may grow and maintain an α-Al$_2$O$_3$ thermally grown oxide (TGO) [1]. Industry-standard coatings, MCrAlY and β-phase platinum-modified nickel aluminide ((Pt,Ni)Al, henceforth PtAl), have been optimized for this latter requirement with a dense and slow-growing Al$_2$O$_3$ scale [1]. While the PtAl provides good oxidation and corrosion protection, it has modest high-temperature strength and is vulnerable to deformation in service [2]. As the surface temperature of TBCs is increased in pursuit of higher operating efficiency, rumpling is accelerated, leading to TBC failure [3] wherein the bond coat forms undulations of a characteristic wavelength due to a ratcheting creep mechanism during thermal cycling [4,5]. The ceramic topcoat, which has a low out-of-plane compliance, cannot deform with the bond coat and therefore detaches from the bond coat, leading to buckling, and subsequent spallation, resulting in loss of thermal protection. Bond coat creep is driven by the thermal expansion mismatch

with the superalloy and thermally grown oxide during thermal cycling [5–9]. Analytical models indicate that the primary rumpling wavelength is a function of the system properties (elastic moduli, thermal expansion coefficients (CTE), thickness) [4] and has been commonly modeled as a 2D sinusoidal surface undulation that grows in amplitude [10]. Theoretical analysis indicates that increasing the yield and creep strengths of the bond coat will mitigate rumpling [7–9].

Another failure mechanism observed during thermomechanical cycling is crack initiation and growth through oxidation-assisted fatigue. Large through-thickness thermal gradients develop on internally cooled turbine blades during takeoff and landing cycles that put the outer surface into a state of compression at high temperature. Creep relaxation during the compressive hold results in a tensile stress upon release of the compressive strain, which can open surface cracks and promote crack face oxidization. The oxidized crack tips push into the coating and substrate upon the next compression cycle, as has been shown in sustained peak low-cycle fatigue (SPLCF) experiments [11, 12]. Crack growth in this scenario is dependent on the superalloy and coating properties [12]. Modeling of the SPLCF experiments shows that increasing the creep strength of the bond coat will decrease the crack extension rate [13] even after the crack has penetrated completely into the substrate [14].

Modeling indicates that improving the yield strength and creep properties of the bond coat will mitigate *both* rumpling and oxidation assisted fatigue failure mechanisms of TBC systems on turbine components. To this end, a number of experimental coatings with compositions in the γ'-Ni$_3$Al phase field have been designed in order to investigate their high-temperature performance in comparison to β-

NiAl coatings. The γ' phase has the $L1_2$ crystal structure, which is an ordered FCC. The principal advantage of γ' over β, which has the BCC-based B2 structure, is that the tighter atomic packing in Ni_3Al results in an inherently slower diffusion rate, which should improve creep properties at high temperature [15–18]. Further, it has been shown that the yield strength of the γ' phase is significantly higher than that of the β phase [19–22]. The focus of this manuscript will be on a comparison of the rumpling behavior of γ' and β coatings.

The oxidation behavior of Ni_3Al is less ideal than that of NiAl due to the formation of $NiAl_2O_4$-type (spinel) oxides upon thermal cycling of the former [23]. However, it is known that small additions of Cr improve the oxidation characteristics of Ni_3Al by selectively forming and maintaining an Al_2O_3 scale [24,25].

The objective of the present study was to design coatings that resist the high-temperature deformation that leads to rumpling and oxidation-assisted fatigue while maintaining adequate oxidation properties. The service life of a thermal barrier coating is ultimately determined by the weakest link in the interconnecting chain of mechanical properties, oxidation properties, and interlayer compatibility. We presently show that a γ' coating can resist rumpling during high temperature cyclic oxidation testing. The presence and absence of rumpling in PtAl and γ' coatings is characterized nondestructively throughout the test.

2. Coating design

The design of bond coats for TBCs, by necessity, requires a systems approach that considers the properties of each constituent of the multi-layered system. It is essential that the bond coat maintains adhesion to the overlying ceramic thermal barrier layer because the presence of the TBC drops the superalloy temperature as much as 300 °C [26]. The creep and oxidation rates of a superalloy will increase drastically after a TBC spall. Further, the temperature and oxidation increase will result in rapid disintegration of the superalloy substrate. Therefore, the design of bond coats for use in such systems must emphasize TBC adhesion. TBC adhesion stems from a combination of rumpling resistance and oxidation properties. The bond coat must have high temperature strength to resist out-of-plane deformation that the TBC cannot accommodate. Also, the bond coat must serve as an adequate Al reservoir from which to grow a TGO that is compatible with the TBC (Al_2O_3). With this in mind, new bond coats that sacrifice an acceptable level of oxidation properties for the benefit of high-temperature strength were sought in order to improve the overall performance of the bond coat/TBC system.

Thermo-Calc Software with a TCNI5 Ni-based superalloys database [27] was used with a CalPhaD-based approach to explore γ' composition space and identify candidate coating compositions with a high likelihood of success at mitigating the rumpling and oxidation-assisted fatigue mechanisms outlined in Section 1. Pt and Pd were not considered for this investigation to demonstrate the feasibility of improving overall coating performance without the use of precious metals. The outline of the design process is as follows. First, elements known to stabilize the γ' phase (Ta, Mo, W, Ti, Hf), strengthen the γ' phase (Si, Hf, Ta, Ti) [28,29], and have beneficial effects on the cyclic oxidation behavior of NiAl or Ni_3Al (Cr, Si, Zr, Hf, Y) [30–33] were considered as potential alloying additions. Second, because bond coats are continuously depleted of Al during service by oxidation and interdiffusion with the substrate, the candidate alloying elements were evaluated for their relative γ' stabilization efficacy as measured by the total width of the γ' field as a function of Al (in an isopleth). Stabilizing the γ' phase ensures the coatings maintain high strength for a greater fraction of service life. Third, the change in the solidus temperature as a function of alloying composition was evaluated to avoid incipient melting during post-coating annealing heat treatments or service. In all, thousands of compositions were screened for interdiffusion compatibility with the superalloy René N5 before selecting a base composition (γ') and a second composition having additional solid solution strengtheners

($\gamma'_{+Si,Ti}$) whose experimentally measured chemistry is listed in Table 1. The amount of each alloying element was an attempt to optimize a multi-parameter space-solidus, high-temperature strengthening, oxidation, and substrate interdiffusion (via matching the chemical activity at 1150 °C).

3. Experimental

The coatings were deposited onto (001) single crystal René N5 disk substrates ($\varnothing = 19.1$ mm $h = 2.1$ mm) using ion plasma deposition [34]. The samples were vacuum heat treated for four hours at 1080 °C. A standard PtAl coating ($\varnothing = 25.4$ mm, $h = 3.2$ mm) made by electroplating Pt onto René N5 followed by vapor phase aluminization was used as a standard for comparison. All coatings were grit blasted with a standard grit blasting procedure using 220 grit alumina particles prior to testing. The specimens were ultrasonically cleaned for twenty minutes in methanol before being thermally cycled 120 times at 1204 °C in an air atmosphere. The hour-long thermal cycle consisted of a ten-minute ramp from room temperature to 1204 °C followed by a forty-four minute hold and six minutes of cooling by forced ambient air, reaching a minimum inter-cycle temperature of 70 °C. Cyclic oxidation testing was carried out in a bottom-loading Rapid Temp furnace (CM Furnaces). An interrupted test was conducted to 60 cycles and showed good repeatability.

The samples were periodically removed for analysis and the mass, surface profile, oxide coverage, and surface phase constitution were measured using a balance, Alicona InfiniteFocus optical profilometer, SEM, and XRD. The profilometer has a vertical resolution of tens of nanometers and a lateral resolution of hundreds of nanometers.

Cross sections showing the microstructures of the two experimental coatings in the *as*-heat treated condition are shown in Fig. 1. Both of the nominally γ' coatings were slightly Al-rich and therefore contain a small amount of secondary β-phase. Some of the surplus Al in these coatings diffused into the substrate during the heat treatment and no β phase was detected by the end of the first thermal cycle.

3.1. Characterization of surface topology

Characterization of the progressive rumpling was investigated using a custom-coded *Mathematica* program to calculate Fourier transforms (FTs) of the surface profiles. An area of approximately 5×1 mm near the center of each sample was measured with the optical profilometer and converted to a 2D matrix of height values having dimension 5893×1238. Fig. 2a is an example surface profile from the PtAl coating taken after 120 cycles. The matrix of height values was zero-padded around the perimeter to dimensions of 6000×6000 points to increase the frequency resolution upon transformation and ensure that the frequency sampling was the same in all directions after the FT. A discrete Fourier transform of each matrix was computed, shifted to DC-center the data (Fig. 2b), and then radially averaged to collect the information from all directions along the surface (Fig. 2c). The radially averaged FTs of the surface profiles of subsequent cycles were

Table 1
Composition (in atomic %) of the experimental coatings designed using the CalPhaD approach and subsequently synthesized experimentally. Compositions were measured by X-Ray fluorescence (XRF) and microprobe (EPMA) in the as-coated condition. The sulfur and carbon concentrations were measured by glow discharge mass spectroscopy (GDMS) at Evans Analytical Group and are listed in ppmw.

	(at%)	Ni	Al	Cr	Ta	Hf	Ti	Si	C (ppmw)	S (ppmw)
γ'	EPMA	69.4	24.0	3.3	2.0	0.63	–	–	720	2.1
	XRF	70.8	24.1	1.8	2.6	0.73	–	–		
$\gamma'_{+Si,Ti}$	EPMA	67.9	24.0	3.3	2.4	0.67	0.90	0.60	1060	4.1
	XRF	68.9	24.9	1.8	2.6	0.75	0.80	0.40		