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A combinatorial investigation of palladium and platinum additions to β-NiAl overlay coatings

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Abstract

A combinatorial approach was used to investigate the effects of higher-order additions to NiAl on the oxidation, phase stability and interdiffusion behavior of the coatings. Overlay coatings with compositions (in at.%) in the range Ni–38Al–5Cr–(0.1–0.5)Hf–(2–8)Pd and Ni–36Al–5Cr–(0.1–0.5)Hf–(2–8)Pt were deposited on René N5 single crystal substrates via ion plasma deposition (IPD). Cyclic oxidation experiments on ten different Pd + Hf- or Pt + Hf-modified NiAlCr coatings at 1100 °C revealed similar weight gain and oxide scale thickness. While the thickness of alumina scales formed on Pd- and Pt-modified coatings were comparable, a higher density of oxide spalls were observed in the former. High-temperature transformations from $\beta \rightarrow \gamma' \rightarrow \gamma + \gamma'$ occurred during extended exposure in both Pd- and Pt-modified coatings owing to Al depletion to substrate and surface oxide. On cooling, Al-depleted β-NiAl transformed to two martensite phases, 3R and fine-twinned 7R. Rumpling or surface roughening occurred for some coating compositions. Microprobe measurements showed that the difference in penetration depths of Pd into the substrate was not as large as expected from the higher interdiffusion coefficient of Pd in β-NiAl and γ-Ni.

Keywords: Coating; NiAl; PGM; Platinum; Palladium

1. Introduction

The continuing demand for increased efficiency in gas turbine engines has, in turn, driven the development of more reliable thermal barrier coating (TBC) systems that enable the turbine components to operate at increasingly high temperatures [1–4]. The intermetallic interlayer between the ceramic topcoat and the superalloy substrate is key to the durability and performance of the TBC systems [5–7]. The role of the bond coat is to retain the TBC while promoting the formation of a slow-growing α-Al_2O_3 thermally grown oxide (TGO) for the protection of the underlying substrate from oxidation and corrosion [8–10]. However, bond-coat phase instability, thermal expansion mismatch with the substrate, and rumpling contribute to the reduction in service life [8,11–20].

Platinum group metals (PGMs) exhibit high solubility in NiAl and have the potential to beneficially alter properties such as high temperature strength, thermal expansion and oxidation resistance of the bond coat layer. The effect of PGMs, especially of platinum, on improving TBC lifetime has been widely reported [9,16,21–32]. Diffusion platinum aluminides are typically coated by electroplating a

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5–10 μm Pt layer, followed by aluminizing via pack cementation or vapor deposition methods [21,22,25,30,33–36]. However, the high cost of Pt has given rise to the desire to reduce or eliminate it via replacement with lower-cost elements. Palladium, having many chemical similarities to platinum but at a significantly lower price, is a natural candidate for Pt replacement. While some reports on Pd-modified coatings indicated that they are inferior to Pt-modified coatings [37–41], others have suggested the opposite [40–43]. This necessitates a more systematic approach to evaluating the role of PGM addition on the overall bond coat stability and oxidation behavior.

The overarching goal of this study was to develop a fundamental basis for the design of B2-based overlay coatings with chromium, PGM and reactive element (RE) additions. The PGM elements Pd and Pt are compared directly. The beneficial effects of RE include improved oxide adherence and reduced oxide scale growth [9,13,15,22,24,32,44–47]. The RE Hf is used in the present study. While Cr is known to improve the corrosion resistance and promote α-Al_2O_3 growth [4], its effect on bond coat performance is not investigated, and the level is maintained constant. The effect of composition variation in (Ni, PGM)CrAlHf coatings on their (a) oxidation kinetics, (b) coating phase stability, (c) oxide rumpling and spallation and (d) interdiffusion is investigated through long-term cyclic oxidation experiments.

2. Experimental

2.1. Specimen preparation

René N5 single crystals (Ni–7.5Co–7Cr–6.5Ta–6.2Al–5W–3Re–1.5Mo–0.15Hf–0.05C in wt.%) were used as the substrates. These were machined into small cylindrical buttons, the dimensions of which are listed in Table 1. The coupons were grit blasted on the back face with 220-grit alumina and coated with a baseline NiAlCrZr coating [48] using the ion plasma deposition (IPD) process described in the next section. The as-deposited composition was measured using X-ray fluorescence (XRF) spectroscopy, followed by a vacuum anneal at 1079 °C for 4 h to densify the coating and establish a metallurgical bond via interdiffusion with the substrate. The front face of the coupon was subsequently grit blasted with alumina, followed by deposition of the PGM-modified coating. The front-face coating compositions were measured using XRF, followed by vacuum annealing at 1079 °C for 4 h. As a result, each coupon contained a baseline NiAlCrZr coating on the back side and a PGM-modified coating on the front side and edges. It should be noted that the NiAl + Cr + Zr overlay bond coat was chosen as the baseline coating because of its improved durability and better protection of the underlying substrate in comparison with the current state of the art diffusion platinum aluminide bond coat, as concluded in an earlier study [48].

Table 1
Button-substrate dimensions.

	Pd-modified	Pt-modified	Baseline
Diameter, d (mm)	12.79 ± 0.02	15.72 ± 0.05	15.66 ± 0.05
Thickness, t (mm)	1.14 ± 0.02	2.10 ± 0.02	2.12 ± 0.02

2.2. IPD

An IPD coating process was used to prepare the coatings [49,50]. In the IPD process, a consumable cathode is placed in a vacuum chamber. A vacuum arc is established at the cathode surface and is maintained by a current-controlled power supply. The cathodic arcs travel randomly across the face of the cathode, creating high-pressure metal plasma and ejecting a mixture of energetic ions, atom clusters and macroparticles into the coating chamber and onto the substrates. IPD provides an effective means of depositing dense, multicomponent coatings at rates ranging from 10 to 100 $\mu m\ h^{-1}$. To produce compositional gradients, the substrate buttons were placed at uniform intervals in a plane below the three cathode sources (composition listed in Table 2) to achieve controlled variations in composition of the deposited coatings (Fig. 1a). The specimen arrangement as well as the elemental distribution in the combinatorial experiments is shown in Fig. 1. The first set of combinatorial experiments used Pd and Hf additions (cathodes 1, 2 and 3), while the second combinatorial experiment used Pt-containing cathodes (cathodes 1, 2 and 4). The resulting compositions, including trace elements, were analyzed using XRF, EMPA and GDMS.

2.3. Oxidation and post-test characterization

Coated specimens with compositions as listed in Table 3 were cleaned with acetone before being placed in the cyclic oxidation furnace. Henceforth, coatings will be identified by their PGM and Hf additions; for example, 0.11Hf–7.95Pd for Pd-43. The specimens were supported on curved alumina platens in a conventional bottom-loading cyclic oxidation furnace (CM Inc. Rapid Temperature Furnace). The cyclic oxidation was conducted in air at 1100 °C using a cycle consisting of (1) ramping from 50 °C to 1100 °C in 13 min, (2) isothermal exposure at 1100 °C for 60 min and (3) fan-cooling to 50 °C for 13 min. In addition to the PGM-modified alloys, coupons coated on both sides with the baseline NiAlCrZr composition were also tested. The

Table 2
Cathode compositions (in at.%) used in IPD combinatorial deposition process.

Cathode	Ni	Al	Cr	X
1	Balance	46	4	0.5 Hf
2	Balance	46	4	–
3	Balance	46	4	12 Pd
4	Balance	46	4	12 Pt