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Improved cyclic oxidation resistance of electron beam physical vapor deposited nano-oxide dispersed β-NiAl coatings for Hf-containing superalloy

Hongbo Guo *, Yongjing Cui, Hui Peng, Shengkai Gong

School of Materials Science and Engineering, Beihang University, No. 37, Xueyuan Road, Beijing 100191, China
Beijing Key Laboratory for Advanced Functional Materials and Thin Film Technology, Beihang University, No. 37, Xueyuan Road, Beijing 100191, China

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ABSTRACT

Oxide dispersed (OD) β-NiAl coatings and OD-free β-NiAl coatings were deposited onto a Hf-containing Ni-based superalloy by electron beam physical vapor deposition (EB-PVD). Excessive enrichment of Hf was found in the TGO on the OD-free coating due to outward diffusion of Hf from the superalloy, causing accelerated TGO thickening and spalling. The OD-coating effectively prevented Hf from outward diffusion. Only small amount of Hf diffused to the coating surface and improved the TGO adherence by virtue of the reactive element effect. The OD-coating exhibited an improved oxidation resistance as compared to the OD-free coating.

1. Introduction

Aluminide coatings are widely used for high-temperature protection of Fe-, Co-, Ni- and Ti-based alloys, as they have excellent resistance to high-temperature oxidation [1–5]. The aluminide coating can be also used as a bond coat beneath ceramic thermal barrier coating (TBC) [6,7] to protect the superalloy substrate against oxidation and hot-corrosion, as well as to reduce thermal stresses resulting from large thermal expansion difference between the TBC and superalloy substrate. The adhesion of oxide scale grown on the aluminide coating is of particular importance, because scale spallation would lead to failure of the TBC [8,9].

Extensive efforts have been made to improve the oxide scale adherence. Pt and reactive elements (REs) such as Hf, Y and Zr as well as their oxides dispersions have been doped in NiAl [6,10–15]. The results indicate that the addition of Pt significantly improves the scale adherence, while a small amount of reactive elements (RE) improve not only the adherence but also decrease the growth rate of the scale. Also, the addition of minor Dy to β-NiAl slowed down the growth of alumina scale and enhanced scale adherence [16]. Cyclic oxidation resistance of the NiAl–0.05Dy coating produced by electron beam physical vapor deposition (EB-PVD) at 1473 K was improved by more than ten times as compared to the undoped NiAl coating.

Another significant issue is the inter-diffusion between the aluminide coating and superalloy substrate at high temperatures. Diffusion of Al from the coating to the underlying substrate usually leads to depletion of Al in the coating, degrading the oxidation resistance of the latter [17–19]. Also, the presence of Mo and W in the coating resulting from outward diffusion of the alloying elements in the substrate could weaken the adhesion of scale to coating and cause premature spallation failure of thermal barrier coatings [12,20]. Characteristics of the substrate including the contents of carbon and refractory elements in the alloy also play a very important role in the oxidation resistance and cyclic lifetimes of TBCs [21–24]. It has been shown that the addition of Hf, Y, or Zr is helpful for enhancing alumina scale adherence. Too high a content of reactive elements such as hafnium in the bond coat, however, could result in a detrimental over-doping effect [25]. It is also reported by Goedjen and Wagner et al. [26] that a short lifetime of a PtAl bond coat on MAR M002 was caused by excessive enrichment of hafnia in the TGO. On the other hand, it has been found that the inter-diffusion of elements between the coating and superalloy substrate could lead to precipitation of topologically close-packed (TCP) phase in superalloy [27,28]. Recently, a new type of microstructural instability, termed as secondary reaction zone (SRZ), has been observed in advanced single crystal superalloys containing high levels of refractory elements such as Re and Ru. It has been shown that the SRZ leads to a significant reduction in the high temperature stress-rupture creep resistance of the single crystals [27–29].

Various techniques have been employed to constrain the inter-diffusion between the metallic coating or bond coat in TBC and superalloy substrate. One method is to introduce a diffusion barrier between the coat and substrate [30,31]. Some barrier compounds

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* Corresponding author. Tel.: +86 10 8231 7117; fax: +86 10 8233 8200.
E-mail address: Guo.hongbo@buaa.edu.cn (H. Guo).

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such as TiN, AlON, Ir and Re, have been investigated [31–35]. However, the diffusion barrier layers could lose their barrier effects after short-term exposure because of their destabilization [34]. In contrast to other metastable materials, oxide dispersed (OD) coatings seem to be more promising, because they have a close-packed crystal structure and as a stable modification, they have few phase transition between room temperature and service temperatures [2,36–39]. The OD coatings effectively hindered the diffusion of various elements from substrate to coating during oxidation [38]. Although some methods such as chemical vapor deposition (CVD) and arc ion-plating [30], have been used for manufacturing diffusion barrier coatings, little attention has been paid to oxide dispersed (OD) diffusion barrier coatings with graded structure produced by electron beam physical vapor deposition (EB-PVD).

In the present work, oxide dispersed (OD) β-NiAl coatings and OD-free β-NiAl coatings were deposited onto a Hf-containing Ni-based superalloy using EB-PVD, respectively. Inter-diffusion process between the NiAl coatings and the superalloy substrates was investigated. Also, the effects of inter-diffusion of elements on the cyclic oxidation behavior of the coatings were studied.

2. Experimental

A directionally solidified Ni-based superalloy was used as substrate materials. The nominal composition of the alloy is listed in Table 1. Hf of 1.5 wt.% was used for the strengthening of the superalloy. Rectangular specimens (20 mm × 15 mm × 3 mm) were fabricated and polished with SiC papers, followed by slight sand blasting. Then, the specimens were ultrasonically washed in acetone for 30 min and then fixed in vacuum chamber of EB-PVD facility with a pressure level of 10^{-3} Pa.

NiAl ingots (51Ni49Al, in at.%) with a diameter of 65 mm were produced by arc-melting for preparation of NiAl coatings by EB-PVD. The EB-PVD facility was equipped with four EB guns and four crucibles by which multi-components evaporation can be accomplished. An EB gun with a power up to 30 kW was applied for heating the NiAl ingot, while another EB gun used for pre-heating the superalloy specimens. An oxygen jet was linked to the EB-PVD facility for providing oxygen input during deposition of NiAl coating. During evaporation stage, the EB current for heating the ingot was 0.6 A. Oxygen was transported to the specimen surfaces by gradually increasing the flow rate of oxygen from 0 to 200 ml/min within 30 min. Then, the oxygen jet was closed while the evaporation and deposition of NiAl coatings went on. The EB current for heating the NiAl ingot was kept at 0.8 A and the substrate temperature kept at around 1073 K during the deposition of NiAl coating. For comparison, OD-free NiAl coatings were also produced by EB-PVD under the same condition except that no oxygen input was introduced.

The as-deposited coatings were annealed in vacuum at 1323 K for 4 h, cyclic oxidation testing of the NiAl coated specimens was performed in air with a furnace equipped with mechanical drive system that allows the specimens moving in and out automatically. The coatings were deposited onto all sides of the specimens, including their edges. The coated specimens were oxidized in air in alumina crucibles at 1423 K in the furnace for 1 h, then moved out of the furnace and cooled by compressed air for 3 min. The lids in the crucibles provided that spalled oxides were collected during the experiment. The weights of the crucibles with the specimens were recorded by an electronic balance (Sartorious CPA 225D, Germany) with a precision of 0.01 mg. The weight gain for each type of coating was determined based on an average value of three specimens' weight gains.

Table 1
Nominal composition of directionally solidified superalloy (in wt.%).

Ni	Ti	Al	Co	Cr	Ta	W	Mo	Hf	S	C	P
Bal.	0.9	5.2	9.9	9.2	3.8	7.1	2.2	1.5	⩽0.01	0.1	⩽0.01

The morphologies of the oxidized specimens were examined to observe the oxide and the effect related to the inter-diffusion of elements between the coatings and superalloy substrates. The microstructures of the coated specimens were characterized by a QUANTA 600 scanning electron microscope (SEM, FEI, Holland) and transmission electron microscopy (TEM, JEM-2100F). The chemical compositions were investigated by electron probe micro-analyzer (EPMA, JXA-8100). The phase constituents of the thermally grown oxides (TGO) on the coatings were identified by X-ray diffraction (XRD, D/max2200PC) using Cu K_α radiation.

3. Results and discussion

3.1. Microstructure of the OD-NiAl coating

The OD-NiAl coating and OD-free NiAl coating were deposited onto superalloy substrates and then annealed in vacuum at 1323 K for 4 h. According to XRD results as shown in Fig. 1, the NiAl coatings basically consist of β-NiAl phase. The composition of the coating was analyzed by EPMA to be 52.5 Ni and 47.5 Al (in at.%), which is closed to the composition of the NiAl ingot.

The cross-section morphologies of the two coatings were compared. Both of the coatings were deposited to nearly 50 μm thick, as shown in Fig. 2. For the OD-NiAl coating, a dark zone of ~20 μm in thickness can be observed, indicating the presence of the OD zone, as shown in Fig. 2b. The color of the OD zone gradually became darker towards the coating surface, implying increased concentration of oxides in the zone. It can be supposed that a gradient structure is formed in the OD zone. Inter-diffusion zones of ~20 μm in thickness were formed in both of the coatings. In the inter-diffusion zone, granular precipitates appeared in the matrix of β phase (white particles in Fig. 2). In our previous work, the precipitate was identified to be σ-TCP phase [39].

A micrograph of the OD zone in higher magnification is shown in Fig. 3. Numerous black particles with submicron size were found to be dispersed in the OD zone. The composition of the black spot in Fig. 3 (marked by the arrow) was analyzed by EPMA and the result is given in Table 2. The spot contains a larger amount of Al and O. Besides, a certain amount of Ni was also detected in the spot. It

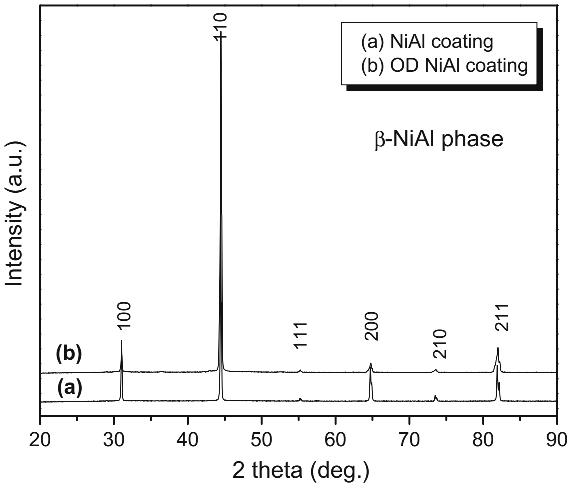

Fig. 1. XRD patterns of the NiAl coatings after 4 h annealing at 1323 K in vacuum (Cu K_α, λ = 1.5418 nm).