



Modelling and experimental study on β -phase depletion behaviour of HVOF sprayed free-standing CoNiCrAlY coatings during oxidation



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ARTICLE INFO

Article history:

Received 16 November 2015

Revised 1 February 2016

Accepted in revised form 2 February 2016

Available online 3 February 2016

Keywords:

HVOF spraying

MCrAlY

Oxidation

β Depletion

Diffusion

ABSTRACT

This paper investigates the β -phase depletion behaviour during oxidation of free-standing CoNiCrAlY (Co–31.7%Ni–20.8%Cr–8.1%Al–0.5%Y, all in wt.%) bond coats prepared by high velocity oxy-fuel (HVOF) thermal spraying. The microstructure of the coatings was characterised using scanning electron microscopy with energy dispersive X-ray (EDX) analysis and electron backscatter diffraction (EBSD). It comprises a two phase structure of fcc γ -Ni and bcc β -NiAl, with grain sizes varying largely from 0.5 to 2 μ m for both phases. Isothermal oxidation tests of the free-standing coatings were carried out at 1100 °C for times up to 250 h. The β phase depletion behaviour at the surface was measured and was also simulated using Thermo-Calc and DICTRA software. An Al flux function derived from an oxide growth model was employed as the boundary condition in the diffusion model. The diffusion calculations were performed using the TTNi7 thermodynamic database together with the MOB2 mobility database. Reasonable agreement was achieved between the measured and the predicted element concentration and phase fraction profiles after various time periods. Grain boundary diffusion is likely to be important to element diffusion in this HVOF sprayed CoNiCrAlY coating due to the sub-micron grain size.

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1. Introduction

Thermal barrier coatings (TBCs) are widely used to protect high temperature components in turbine engines from harsh operating environments [1,2]. TBC systems consist of a ceramic top coat, a metallic bond coat and a superalloy substrate [3–5]. Additionally, a thermally grown oxide (TGO) forms at the interface between the top coat and the bond coat during service at elevated temperature due to the fact that oxygen permeates through the ceramic top coat and oxidises the bond coat. The durability of the overall TBC system is largely determined by the microstructural, chemical and mechanical characteristics of the bond coat [6–8]. Among different types of bond coats, MCrAlY overlay coatings, where M=Co, Ni or a combination of the two, are of great interest for their excellent oxidation resistance and good adhesion between the top coat and the superalloy substrate [9–16]. Depending on the compositions, MCrAlYs generally exhibit a multi-phase structure; e.g. a fcc γ -Ni matrix and a bcc β -NiAl secondary phase. The β -NiAl acts as an aluminium source and promotes the stable and continuous formation of aluminium oxides, predominantly Al₂O₃, during oxidation. This Al₂O₃ scale, also referred to as the TGO, forms between the top coat and the MCrAlY bond coat [10,17–19]. It is widely recognised that the growth of the TGO during service causes progressive build-up of stress in the system, leading to the spallation of the ceramic top coat and eventually

causing detrimental effects to the turbine engine components [20–22]. The continuous growth of the TGO due to aluminium diffusion from the MCrAlY bond coat results in subsequent β phase depletion at the oxide/bond coat interface [23–28], causing mechanical and chemical degradation of the coating. Degradation of the bond coat also occurs due to interdiffusion of element between the MCrAlY and the superalloy substrate. This is not considered in the present paper but has been studied by others, e.g. [29–41]. In these research works, the emphasis has been on substrate/bond coat interdiffusion and specific comparisons between measured and predicted β -phase depletion at the oxide/bond coat interface have not been considered. On the other hand, several analytical models concerned with the oxidation of two phase systems in which the secondary phase dissolves during oxidation have been reported, e.g. [42–48]. In these models, the secondary phase depletion behaviour during oxidation can generally be represented by the parabolic diffusion law, Eq. (1),

$$x_d \sim \sqrt{t} \quad (1)$$

where x_d is the width of the second phase depletion zone and t is the oxidation time.

Considering the significance of β depletion on the degradation of TBCs, therefore, the aims of the work reported in this paper were to investigate specifically the kinetics of β depletion at the oxide surface in free-standing MCrAlY coatings during isothermal oxidation, to develop a numerical model for phase evolution and element diffusion using

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the available thermodynamic and kinetic data of the MCrAlY system, and to study the grain size effect in these thermally sprayed MCrAlY coatings.

2. Materials and experimental procedure

2.1. Material preparation, heat treatment and isothermal oxidation

The coatings used for isothermal oxidation experiments were prepared by high velocity oxy-fuel (HVOF) thermal spraying using powder with a nominal composition of Co–31.7%Ni–20.8%Cr–8.1Al–0.5Y (all in wt.%). The powder was obtained from Praxair (commercially available as CO-210-24) and had a size range $-45 + 20 \mu\text{m}$. The coatings were deposited onto 800-grit ground mild steel substrates with dimensions $60 \times 25 \times 1.8 \text{ mm}$ using a Met Jet III liquid fuel HVOF gun. The details of the spraying process are given in reference [49]. Coatings were sprayed to a thickness of approximately 0.5 mm and were then debonded from the ground substrate by bending around a mandrel. Two surface conditions were used in the experiments on the debonded free-standing CoNiCrAlY coatings: (I) an as-sprayed rough surface (surface roughness, $R_a = 6.1 \pm 0.5 \mu\text{m}$) and (II) a polished surface ($R_a = 0.2 \pm 0.1 \mu\text{m}$).

An initial vacuum heat treatment (HT) was given to the debonded coatings prior to oxidation. Coatings were heat treated in a vacuum with a nominal pressure of $6.0 \times 10^{-3} \text{ mbar}$ at $1100 \text{ }^\circ\text{C}$ for 2 h followed by air cooling to replicate the initial heat treatment given to the bond coat material in conventional TBC manufacture. Previous work has demonstrated that the initial heat treatment has the effect of reducing any coating porosity to a minimal level [49] and allows the β phase to coarsen and precipitate [50]. For isothermal oxidation experiments, heat treated samples were exposed to laboratory air in a Carbolite furnace at $1100 \text{ }^\circ\text{C}$ for 50 h, 75 h, 100 h and 250 h followed by natural air cooling.

2.2. Material characterisation

For microstructural observations, samples were mounted in a conductive resin and successively ground and polished to a $1 \mu\text{m}$ surface finish. The microstructures of coating cross-sections before and after isothermal oxidation were examined in a FEI XL 30 scanning electron microscope (SEM) operated at 20 kV. Backscattered electron (BSE) imaging was used to form images and semi-quantitative energy dispersive X-ray (EDX) analysis was also utilised to obtain element profile measurements and phase identification through chemical analysis. Image analysis software, ImageJ [51], was used to measure the width of the β depletion zone and β -phase volume fractions. SEM-based electron backscatter diffraction (EBSD) was carried out to investigate the grain morphology of coating cross-sections using a Zeiss 1530 VP field emission gun scanning electron microscope (Carl Zeiss, Inc., Mape, Grove, MN) with an EDAX Pegasus combined electron backscatter diffraction system (EDAX, Mahwah, NJ, USA). For EBSD analysis, a further stage of chemical/mechanical polishing using colloidal silica was required after the standard grinding and polishing procedure to achieve reduced surface deformation. The EBSD patterns were recorded at an acceleration voltage of 20 kV with a beam current of 26 nA at a specimen tilt angle of 70° . EBSD maps were collected with a step size of $0.1 \mu\text{m}$ and areas of $50 \mu\text{m} \times 50 \mu\text{m}$. The EBSD data acquisition and processing were performed using the EDAX OIM5 software suite.

3. Modelling procedure

3.1. Model description

The initial composition is taken to be Co–31.7%Ni–20.8%Cr–8.1Al (all in wt.%). The small amount ($\sim 0.5 \text{ wt.}\%$) of Y actually present in the alloy is neglected and remaining elements are scaled accordingly. The alloy is assumed to be at thermodynamic equilibrium initially and the

temperature is set to be $1100 \text{ }^\circ\text{C}$. The free-standing coating is held at this temperature for different periods of time and Al_2O_3 growth, taken to be planar, occurs on both surfaces according to a kinetic law (described later in Section 3.2). At any time, t , the microstructure during oxidation is shown by the schematic cross-section of Fig. 1. In this system, the Al_2O_3 scale grows at the oxide/coating interface and is in contact with a single γ -Ni phase region known as the β depletion zone whilst the interior of the sample comprises a two phase ($\gamma + \beta$) region as predicted by thermodynamic equilibrium for the alloy composition. During oxidation, the β phase volume fraction is expected to vary as shown schematically in Fig. 2, where the β phase depletes as the Al_2O_3 forms and eventually the fraction of β in the $\gamma + \beta$ region decreases.

3.2. Oxidation model

The β depletion model assumes that growth of Al_2O_3 scale requires a flux of aluminium from the coating. Owing to the approximately planar nature of this free-standing coating system, the whole system can be approached by a one-dimensional model. In this study, the oxidation model reported by Meier et al. [34] for a Ni–Co–Cr based bond coat was employed, in which they proposed a simplified power law oxide growth model as expressed in Eq. (2),

$$\delta_{(\mu\text{m})} = \left\{ \exp \left[Q \left(\frac{1}{T_0} - \frac{1}{T} \right) \right] t \right\}^n \quad (2)$$

where $Q = H/R$, H is the activation energy in J/mol, R is the gas constant in $\text{J}/(\text{mol} \cdot \text{K})$, T_0 is a temperature constant in K, T is the oxidation temperature in K, t is the oxidation time in seconds and n is a constant. By the combination of the above parameters, oxide growth thickness can be obtained in μm . Q , T_0 and n were found by Meier et al. to be 27,777.4 K, 2423.7 K and 0.332 respectively by fitting this oxide growth behaviour between 1038–1149 $^\circ\text{C}$ to experimental data [34]. Taking a differential of Eq. (2) with respect to time, the oxide growth rate can be obtained as Eq. (3).

$$\dot{\delta}_{(\mu\text{m/s})} = n \left\{ \exp \left[Q \left(\frac{1}{T_0} - \frac{1}{T} \right) \right] t \right\}^{n-1} \quad (3)$$

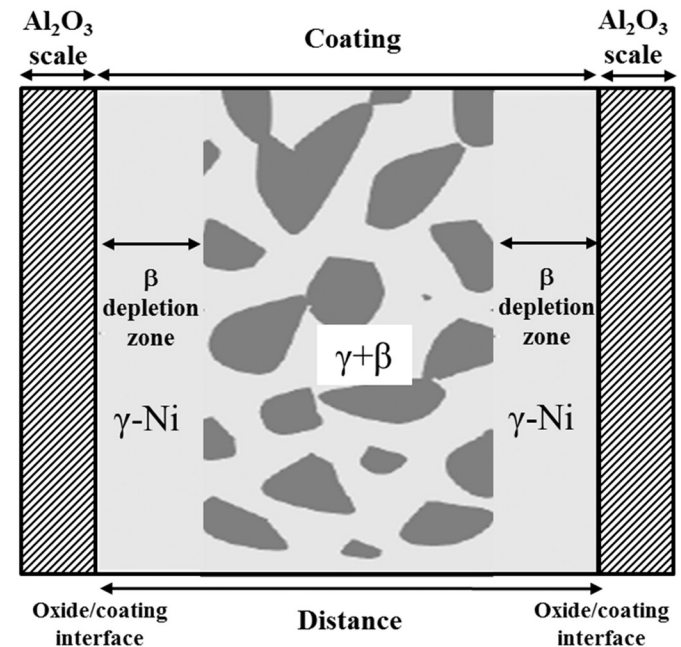


Fig. 1. A schematic cross section of a free-standing MCrAlY coating showing the development of β depletion zone during oxidation.

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