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Influence of vacuum heat treatment parameters on the surface composition of MCrAlY coatings

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ABSTRACT

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Keywords: Thermally grown oxide MCrAlY bondcoat Chromium evaporation Heat treatment Yttrium content In the present study the synergistic effect of vacuum heat treatment pressure and coating Y and O content on the MCrAlY surface oxide is investigated. For this purpose, free standing MCrAlY coatings with two different Y contents were exposed at 1100 °C for times between 1 and 5 h in vacuum at pressures of $<10^{-3}$ Pa and 10^{-2} Pa, and in Argon gas at 10^{-1} Pa. The surface scale composition and morphology were analysed with a variety of analysis techniques. It is shown that the composition at the MCrAlY coating surface is mainly governed by two competing processes: Cr evaporation (β -NiAl formation) and Y oxidation (internal and external). The latter reaction has been observed to depend strongly on the Y reservoir in the coating. It can be observed that the chromium evaporation enhances for reducing pressure. More Y and less 0 in the coating result in enhanced Y rich oxide formation (Y₂O₃ and YAP; Yttrium Aluminium Perovskite) and decreased chromium evaporation. On rough surfaces β -NiAl enrichment in hills and yttria formation in valleys can be found. Additionally no alumina formation can be observed under all studied vacuum heat treatment conditions.

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1. Introduction

Increasing the efficiency of natural gas fired power plants requires longer lifetimes at higher turbine inlet temperatures of components in gas turbines. Handling of higher operating temperatures and the protection of turbine components against high temperature corrosion and oxidation are achieved by a cooling system and a two-layered Thermal Barrier Coating (TBC) [1–3]. In industrial gas turbines the TBC system typically consists of a MCrAlY (M = Ni, Co) bond coat (BC) and an vttria stabilized zirconia (YSZ) topcoat on a Ni-base During service a thermally grown oxide (TGO) laver, which is typically based on α -alumina, forms at the TBC/BC interface. This TGO plays a major role in the failure of TBC systems [1,4,5]. Therefore growth rate, composition and adherence of the TGO are important factors determining the lifetime of TBCs [6]. Several investigations illustrate that the TGO formation is affected by the vacuum heat treatment commonly applied prior to TBC deposition [7,8]. Variation of the heat treatment parameters (e.g.: vacuum quality) may result in formation of different oxides on BCs with nominally the same composition [7,9]. As a result, TBC systems with nominally the same composition may exhibit a wide distribution of lifetimes [5,10]. In particular vacuum heat treatment leads to Y oxidation and Cr evaporation from the BC-surface. At temperatures above 1000 °C, the MCrAlY bond coats typically consist of two phases: γ -Ni solid solution and β -NiAl

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[11,12]. Aluminium rich phases (e.g.: β -NiAl) promote the formation of a protective alpha alumina based oxide scale. This β -NiAl phase may form at the MCrAlY surface due to the evaporation of volatile chromium species during annealing in vacuum [7,13].

The protective properties of alumina base TGOs, in particular adherence to the bond coat, are improved by addition of the reactive element yttrium. The addition of yttrium leads to gettering sulphur and for changing the alumina scale growth mechanism [9,11,14]. Yttrium forms during vacuum heat treatment of yttrium containing oxides. Normally three types of oxides can be found: garnet (Y₃Al₅O₁₂, YAG), perovskite (YAlO₃, YAP), and yttria (Y₂O₃). Such oxides can be found locally on the bond coat surface after vacuum annealing [8] and affect morphology, growth rate, and mechanical properties of the alumina scale formed during oxidation [15].

Most of the previous studies [1,7,9,12,14] have dealt with chromium evaporation and yttrium oxidation as separately occurring processes. It can be expected that, especially at high yttrium contents, both phenomena can interact with each other depending on the heat treatment conditions. Therefore in the present study the influence of vacuum annealing conditions and the yttrium content in the MCrAIY bond coat on the oxide formation and phase changes near the surface during annealing in vacuum are investigated.

2. Experimental procedures

Three types of free standing MCrAlY coatings were used for the vacuum heat treatment experiments. The coatings had the same main composition (Ni-25Co-17Cr-10Al; (wt.%)) but differed in

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Table 1

Yttrium and oxygen content (wt.%) in the free-standing MCrAIY coatings, as measured by ICP-OES.

MCrAlY coating	Y	0
High Y, VPS	0.56	0.027
Low Y, VPS	0.32	0.034
Low Y, HVOF	0.22	0.241

composition by the Y and O content. One coating was designed to contain 0.6 wt.% yttrium, whereas the other two coatings were designed to contain 0.3 wt.% yttrium. One of the coatings with low yttrium content was deposited by High Velocity Oxygen Fuel (HVOF), whereas the other two coatings were applied by Vacuum Plasma Spraying (VPS) onto a steel substrate. The three coatings are further designated as 1.) high Y, VPS, 2.) low Y, VPS, and 3.) low Y, HVOF, respectively.

The deposition method had a strong influence on the oxygen content of the coating as shown in Table 1, the VPS coatings had a much low oxygen content (<0.05 wt.%), than the HVOF coating (0.25 wt.%). After the spraying processes the coatings were separated from the steel substrate by means of spark erosion. For all coatings, one side of the samples (i.e. the side adjacent to the steel substrate) was ground to a 4000 grit surface finish. The other side was intentionally kept rough. The specimens, with one flat and one rough, as sprayed surface were exposed at 1100 °C for times between 1 and 5 h in a vacuum tube furnace with variable atmospheres and pressures. The coatings were exposed at two different atmospheres in vacuum at pressures of <10⁻³ Pa and 10⁻² Pa, and in Argon gas at 10^{-1} Pa.

After vacuum annealing, the oxidized coatings were analysed with a variety of techniques. The surface scale composition and morphology were analysed by sputtered neutrals mass spectrometry (SNMS), Raman spectroscopy, and X-ray diffraction (XRD). Optical metallography and scanning electron microscopes (SEM) with field emission gun (FEG), a LEO 1530-Gemini and a Zeiss Supra50 equipped with a cathodoluminescence detector and an energy dispersive X-ray spectroscopy (EDX) system, were used for imaging the cross sections and for chemical microanalysis, respectively.

3. Results

The X-ray diffraction patterns in Fig. 1 show the phase composition near the surface of the VPS MCrAIY coating with low yttrium content. For all three investigated pressures the coating consists of two major phases after vacuum annealing: an aluminium rich β-NiAl phase and an aluminium poor γ -Ni phase. The intensity of the diffraction peaks from the γ -Ni phase at $2\theta = 52^{\circ}$ increases with increasing pressure and in all three coatings diffraction peaks of the B-NiAl phase can be found. In addition, diffraction peaks of yttrium aluminium perovskite (YAP, YAlO₃) are detectable at all pressures. Cross sections of these low Y, VPS MCrAlY coatings (see Fig. 2) reveal a β-NiAl subsurface enrichment and the formation of minor amounts of oxide particles. The enrichment of β -NiAl is stronger during annealing at lower pressures. Comparison of the images after annealing at $< 10^{-3}$ and 10^{-2} Pa shows, that decreasing oxygen partial pressure promotes the formation of β-NiAl. This increased formation of β-NiAl with decreasing oxygen partial pressure was found for all three studied coating compositions.

Oxidation products were detected on the surface of all three studied coatings. X-ray diffraction (Fig. 1) and Raman analysis (Fig. 3) revealed that no alumina is formed on the surface. Rather the VPS coating with high yttrium content forms primarily Y_2O_3 and the VPS and HVOF coatings with low yttrium content forms primarily YAIO₃ (YAP).

The cross section micrographs of the three studied coatings (Fig. 4) show clear differences between the amounts of Y rich oxides and amount of β -NiAl formed after vacuum annealing at 10^{-2} Pa. On the coating with high yttrium content (Fig. 4a), a large amount of yttrium rich oxides was detected. On this coating with 0.6 wt.% Y a locally thin, continuous yttrium oxide scale with pegs and internal yttrium rich oxide particles formed. In contrast to the low Y coatings (Fig. 4b and c) the high Y substrate shows a relatively minor enrichment of β -NiAl. The coatings with low yttrium content exhibit a stronger enrichment of β -NiAl and a smaller amount of oxide particles on the surface (Fig. 4b and c). This indicates that the surface yttrium rich oxides suppress the enrichment of β -NiAl. Of the two low Y coatings the β -NiAl enrichment is stronger for the HVOF than for the VPS coating. Both coatings have nominally the same yttrium content but differ in oxygen content (Table 1).



Fig 1. XRD diffractograms of the low Y, VPS MCrAIY coating after vacuum annealing at 1100 °C for 5 h at pressures of <10⁻³ Pa, 10⁻² Pa, and 10⁻¹ Pa.

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