Influence of Interface Roughness, Substrate and Oxide-Creep on Damage Evolution and Lifetime of Plasma Sprayed Zirconia-based Thermal Barrier Coatings

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Abstract

The stress state in thermal barrier coating (TBC) systems for gas turbine blades during thermal cycling is mainly governed by (i) the thermal and mechanical mismatch between the ZrO$_2$-base topcoat and the Ni-base substrate alloy, (ii) the roughness profile at the interface between MCrAlY-bondcoat (BC) and TBC, and (iii) plastic deformation and creep in the BC and in the oxide scale growing at the interface between TBC and BC. In the present study, the influence of (ii) and (iii) on the thermal cycling lifetime was studied using a simplified model system consisting of (a) FeCrAlY substrates with different creep strength, i.e. a conventional and an oxide dispersion strengthened alloy, (b) oxide layers with different grain size, i.e. a coarse grained version applied by oxidation and a nanocrystalline version applied by sputtering and (c) a plasma sprayed ZrO$_2$-TBC stabilized by 8% Y$_2$O$_3$. A trend of decreasing lifetime with increasing creep strength of the FeCrAlY was observed. Infrared pulse thermography analysis of delamination crack growth during thermal cycling tests showed that in case of low creep strength substrates, small delaminations grow continuously, link to each other and finally lead to spallation of the TBC, whereas in the case of high creep strength substrates fast crack growth and spallation occurs as soon as one delamination exceeds a diameter of approx. 2 mm. A fracture mechanics model for delamination crack growth is able to roughly describe this behavior. The influence of oxide creep seems to be less pronounced than that of substrate creep. However, the highest lifetimes were observed for the combination of Fecralloy substrate and nanocrystalline oxide layer. The influence of roughness on lifetime remains unclear due to a wide lifetime scatter.

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1. Introduction

Thermal barrier coatings (TBCs) are used to protect gas turbine components made of Ni-superalloy, especially blades and vanes in the first stages, from heat and hot corrosion to enable higher gas temperatures and therefore increased turbine efficiency. They consist of Y$_2$O$_3$-partially-stabilised ZrO$_2$ (P-YSZ) produced by
air plasma spraying (APS) or electron beam physical vapour deposition (EB-PVD). A Ni(Co)CrAlY bond coat (BC) is applied before the deposition of the TBC to provide mechanical bonding of the TBC to the substrate. The BC is in most cases produced by vacuum plasma spraying (VPS). In addition to improving the adherence of the TBC, it protects the substrate from oxidation, by forming a dense alumina scale (thermally grown oxide – TGO) at high temperature [1-3].

In service stresses arise in the coating system caused on the one hand by thermal mismatch of the coating components during start up and shut down, and on the other hand by lateral growth and thickening of the TGO at high temperature. This leads to crack initiation and growth, mostly in the TBC-BC interface. Coalescence and growth of these cracks causes delamination of the TBC and finally large-area spallation. Moreover, the TBC spallation can cause subsequent damage of other turbine parts. Therefore, in order to avoid in-service TBC failure, the coatings of turbine blades and vanes are refurbished in certain intervals [1-3]. To maximise the inspection intervals the TBC lifetime should be as long as possible, since shut down of the turbine for inspection is very cost-intensive. Therefore the influence of the key mechanical properties of the coating components as well as of the TBC-BC interface morphology on lifetime has to be understood and optimized.

The inspection intervals for gas turbines are determined by estimation of the TBC lifetime by calculating, for example, crack initiation and growth using fracture mechanics approaches [4, 5]. To improve these models they have to be validated by experimentally measured crack growth data.

Rösler et al. [6] and Bednarz [7] performed finite element method (FEM) simulations and investigated the influence of mechanical properties and interface roughness on the stresses in TBC-systems. To decrease calculation time, the system was reduced to 2 dimensions (2-D) and a periodic interface roughness profile. They showed that faster creep of the BC and TGO causes significantly lower stresses in the system and therefore a delayed crack formation and an enlarged lifetime. They also simulated variable amplitude and wavelength $\lambda$ of the periodic 2-D BC roughness: Increasing $a$ and/or decreasing $\lambda$ cause higher tensile stress in the TBC near the roughness peaks.

Teixeira et al. [8] determined the stress in an EB-PVD TBC on Ni-base alloy substrates experimentally. The average BC roughness depth was varied by grinding with abrasive paper with different grain sizes (P220 and P1200). The stress was measured by X-ray diffraction, micro-Raman techniques as well as curvature measurement using a laser transducer. They showed that the average compressive stress after deposition of the TBC decreases with increasing roughness depth of the BC-TBC interface. Gil et al. [9] determined the stress in a TGO on a free standing oxidised NiCoCrAlY layer after cooling to RT. For these investigations fluorescence spectroscopy was used with a spatial resolution of 3 $\mu$m, giving evidence of compressive stress in the TGO. The average compression was lower for roughened substrates than for polished ones. Nusair et al. [10] determined the BC-stress in a system with air plasma sprayed TBC and VPS-BC on flat stainless steel substrate by the hole drill method, where a hole is brought step by step into the surface The resulting strains are measured from which the residual stresses are calculated. The depth steps were 0.05-0.1 mm; the spatial resolution was not reported. They measured a lower maximum BC-stress for higher roughness.

Several authors also investigated the influence of roughness depth on the thermal cycling lifetime and crack formation of TBCs experimentally: Vaßen et al. [11] showed that the average crack lengths in an APS-TBC after a certain cycle number decreased with increasing roughness depth. Further Nusair, Herzog and Traeger found higher lifetimes for higher roughness depth [10, 4, 12]. Nusair and Vaßen assume that increasing roughness causes better mechanical bonding between TBC and BC [10, 11] and that the stress conversion from compression to tension in the valley regions of the TBC-BC interface roughness profile is delayed [11]. In contrast to that, Lau and Zhang showed that lifetime is lower for higher interface roughness depth [13, 14]. Zhang assumes that increased roughness causes a stronger “shadowing” effect during APS of the TBC, and therefore a lower adhesion [13].

The aim of the present study is to investigate the influence of BC and TGO creep strength and of interface roughness on TBC lifetime and damage evolution experimentally and to compare the results with the above mentioned experimental findings and with fracture mechanics based simulations of delamination crack growth. Therefore, cylindrical FeCrAlY-substrates (simulating the BC, see section 2) with different creep strength were coated with $\text{Al}_2\text{O}_3$-layers of different grain size to vary the oxide creep strength and, subsequently, a P-YSZ TBC was applied by APS. These specimens were thermally cycled and lifetime was determined. The damage
evolution during thermal cycling was documented by infrared pulse thermography. Furthermore, the microstructure was studied at different cycle numbers.

2. Test material and experimental procedure

Thermal cycling tests were performed to investigate the influence of the BC and TGO creep strength as well as of roughness depth on the lifetime of a TBC. A simplified model TBC system was produced: the Ni-base substrate was not used in order to exclude the influence of interdiffusion and thermal mismatch between Ni-superalloy and bond coat. Instead, the TBC and the TGO were directly applied on a solid FeCrAlY-substrate simulating the BC, enabling systematic variation of creep strength, which is not possible in plasma sprayed bondcoats, because, in contrast to Ni(Co)CrAlY-alloys, Fecalloy is available with sufficiently big variety of creep strength and has similar oxidation properties and thermal expansion coefficient as the NiCoCrAlY alloys usually applied as BC material. To vary the substrate creep strength, the oxide dispersoid strengthened (ODS) alloy “Incoloy MA956” and the conventional (non-ODS) “Fecalloy Eisen/Chrom” were used. MA956 was fabricated by SpecialMetals Wiggin as a rod with 20 mm diameter and has a chemical composition of 74.3 wt.% Fe, 4.39 wt.% Al, 19.6 wt.% Cr and 0.34 wt.% Y as \( \text{Y}_2\text{O}_3 \)-dispersoids. Fecalloy was delivered by GoodFellow as a rod with 16 mm diameter and contains 74.0 wt.% Fe, 4.84 wt.% Al, 21.8 wt.% Cr and < 0.005 wt.% Y. Cylindrical specimens were produced with a length of 30 mm and a diameter of 9 mm by electro discharge machining (MA956) and drawing (Fecalloy). Afterwards, the samples were sandblasted at Technische Universität Braunschweig, IfW. Grain size variation of the abrasive material resulted in roughness depths \( R_z \) classes of 3.6-4.2; 7.7; 9.6-13.1 and 17.9-18.8 \( \mu \)m measured by line profilometry. The \( \text{Al}_2\text{O}_3 \) creep strength was varied as follows: Before the TBC was deposited, one set of samples was pre-oxidised for 220 h in air to obtain a coarse crystalline \( \text{Al}_2\text{O}_3 \) scale with high creep strength. The resulting average \( \text{Al}_2\text{O}_3 \)-layer thickness was 2.4 \( \mu \)m and the grain size was 0.46 \( \mu \)m for Fecalloy. For MA956 an oxide layer thickness of 3.6 \( \mu \)m and a grain size of 0.45 \( \mu \)m was obtained. Another set of samples was coated by PVD to obtain a nano-crystalline \( \text{Al}_2\text{O}_3 \) scale with low creep strength. The resulting average \( \text{Al}_2\text{O}_3 \)-layer thickness was 2.1 \( \mu \)m for Fecalloy and 2.9 \( \mu \)m for MA956. The average grain size of the PVD-\( \text{Al}_2\text{O}_3 \) was < 0.1 \( \mu \)m for both substrates. Finally, an 8 % \( \text{Y}_2\text{O}_3 \) partially stabilised APS-\( \text{ZrO}_2 \)-TBC was deposited at Technische Universität Braunschweig, IfW, with a coating thickness of 300 \( \mu \)m and tapered edges. A maximum of two samples with MA956 substrate were produced in each roughness class, because of limited availability of MA956 raw material. A completely coated cylindrical sample is shown in Fig. 1 a).

![Fig. 1. a) Completely coated cylindrical sample and b) disc-shaped sample placed on a glass sample holder.](image)

Additionally, disk-shaped specimens were cut from TBC coated (not pre-oxidised) samples with substrates made of Fecalloy and PM2000 (an ODS-alloy with similar chemical composition as MA956) to observe the damage evolution at the TBC-substrate-interface during thermal cycling after cooling to room temperature by light microscopy. The cross sections of these specimens were polished to obtain sufficient quality of the micrographs. A completely coated disc-shaped sample is shown in Fig. 1 b).
All samples were thermally cycled by moving them automatically into and out of a tube furnace which was kept at constant temperature. The minimum and maximum temperature of the thermal cycles was 60 °C and 1050 °C, respectively. The dwell time at maximum temperature was 2 h and the heating and cooling time 13.3 minutes. Lifetime was defined as the cycle number after which the first macroscopic segmentation crack occurred in the TBC. To investigate the damage evolution by thermography, the samples were removed from the furnace after cooling in intervals of approximately 9 cycles. An infrared camera (FLIR, ThermaCam2000) was used. Further details of the thermography procedure are reported in [15]. In the grey-scale images given in Fig. 4 of the present paper, white or light grey thermography areas represent adherent TBC and dark grey zones indicate delaminations. The detection limit for delaminations was determined by comparing the thermography images with cross-sections of the corresponding samples. The diameters visible in the thermography images correlate well with the size of the delaminations measured at metallographic cross sections. The detection limit for delaminations is a lift-off of 1-2 μm [15]. The delaminated area was measured from the thermographies using the phase measuring tool of the image analysis software analysis.

3. Results and discussion

3.1. Life time

Figure 2 shows the lifetimes of the samples. The lifetimes of the TBCs on the low creep strength Fecralloy substrate are in most cases higher than those observed for MA956 substrate which has significantly higher creep resistance. It should be noted that the highest lifetimes are obtained for Fecralloy substrates combined with the nano-crystalline PVD-Al₂O₃-layer. However, the influence of oxide creep strength seems to be much less pronounced than that of substrate creep and scatter of lifetime is larger in case of the PVD-Al₂O₃ than in case of the naturally grown coarse crystalline alumina interlayer. Because of the large scatter in lifetime a systematic influence of roughness depth on lifetime could not be determined for all specimen variants.

![Fig. 2. Lifetimes of the thermally cycled cylindrical samples with all variations of substrate and oxide creep strength as well as of interface roughness.](image-url)
3.2. Microstructure at end of life

The examination of cross sections of the cylindrical specimens at end of life shows that in case of Fecralloy substrate the roughness depth at the TBC-substrate interface was substantially increased in the delaminated zones compared to the initial state (Fig. 3 a). This indicates that the growth stress of the Al$_2$O$_3$-layer and the thermal mismatch stress during cooling were relaxed by wrinkling of the TBC-substrate interface due to creep in the near-surface regions of the substrate. In contrast, MA956 substrates showed no roughness increase in the delaminated zones (Fig. 3 b). It should also be noted that both, enhanced oxidation and a pronounced increase in interface roughness in most of the investigated samples with Fecralloy substrate took place in front of segmentation cracks of the TBC (Fig. 3 c). In contrast, the samples with MA956 substrate exhibited only very few or even no segmentation cracks at end of life and no enhanced substrate oxidation or local increase of interface roughness under the unaffected TBC and in front of segmentation cracks.

The examination of the polished, disk-shaped specimens during thermal cycling showed that for both substrates segmentation cracks already formed after the first cycle. The average segmentation crack distance for Fecralloy substrate was 777-900 $\mu$m. Furthermore, plastic deformation of Fecralloy substrates in front of TBC segmentation cracks was observed after 71 cycles (Fig. 3 d), i.e. around 1/12 of the highest observed total thermal cycling life of samples with Fecralloy substrate and coarse grained Al$_2$O$_3$ interlayer (see Fig. 2). In contrast, the PM2000 substrate exhibited no enhanced plastic deformation in front of TBC segmentation cracks after a similar cycle number of 77.

**Fig. 3.** (a-c) View on the cross section of a cylindrical specimen and (d) on the free surface of a TBC-coated disk-shaped specimen after thermal cycling.

3.3. Damage evolution during thermal cycling

The total delaminated area was measured from the thermographies as a function of cycle number. The influence of substrate creep strength on the delamination area increase is shown in Fig. 4 (experimental data are
given by diamond- and square-shaped data points, respectively). Note that for Fecralloy substrate the total delaminated area increases continuously with increasing cycle number until end of life. In contrast, for MA956 substrates, an abrupt acceleration of delamination area growth is observed, as soon as the first delamination with a diameter of > 2 mm appears. (Further details about the thermography are given in [15].) The slower delamination crack propagation correlates with the higher lifetimes of the TBC on Fecralloy substrates. This general substrate influence is independent from the roughness depth (see Fig. 2). The following hypothesis could explain the substrate influence on crack growth and lifetime: In case of MA956 substrate the stress intensity at the delamination crack tip increases with increasing delamination area without any significant reduction of the crack tip stresses by creep processes or increase of roughness near the crack tip. At a certain delamination size, a critical stress is reached, resulting in strongly accelerated growth of the delaminated area. In case of the Fecralloy substrate, the stress intensity at the delamination crack tip is reduced by wrinkling of the substrate surface due to creep processes (see Fig. 3), reducing the increase in stress intensity with increasing delaminated area, and resulting in a slow, continuous delamination crack growth with increasing cycle number and therefore higher TBC lifetimes than at MA956 substrates.

| Experimental data | Fracture mechanics model |
|-------------------|--------------------------|
| delamination crack area | delamination crack area |
| lifetime | substrate creep strength |
| --- | --- | --- |
| DBTT = 400 °C low (Fecralloy) | DBTT = 1000 °C high (MA956) |

Fig. 4. Influence of substrate creep strength on the evolution of total delamination crack area as a function of cycle number: Experimentally measured (square and diamond shaped data points) and modelled (thick lines) curves. In addition, thermography images of the samples are included (the sample is framed, the borders represent the background, dark grey areas on the sample are delaminations).
The thick lines additionally plotted in Fig. 4 show the crack growth curves predicted by a TBC lifing model developed at FZ Jülich - IEK-2 which is described in detail in [5]. This model calculates delamination crack growth using a two-step approach: In the first step, the growth of microcracks is assumed to be proportional to the TGO growth kinetics which is modelled using a power-law for time- and an Arrhenius approach for temperature-dependence of TGO thickness and is fitted to experimentally obtained TGO growth curves. As soon as the TGO thickness reaches ¼ of the peak-to-peak roughness of the TGO-TBC interface, a fracture mechanics approach considering thermal mismatch and TGO growth induced stresses is used to calculate crack propagation. The time dependence of crack length is calculated by a power law and the crack growth coefficient and exponent are fitted to experimental data. In the present case, the model was fitted to the crack growth curve for MA956 substrate, for which a ductile-brittle transition temperature (DBTT) of 1000 °C was assumed (i.e. the material exhibits sufficiently high strength to establish significant thermal mismatch stress without complete relaxation up to 1000 °C). For calculation of the behaviour of the TBC system based on a Fecralloy substrate, the DBTT was reduced to 400 °C without any further changes in the model parameters. The assumptions for DBTT of both substrates are based on tensile tests reported in [16]. The longer lifetime compared to the TBC on MA956 substrate is predicted with fair accuracy. However, the shape of the simulated crack growth curve shows significant differences to the experimentally measured area growth. This is attributed to the fact that the model calculates initiation and propagation of one single delamination crack, whereas delamination growth in the Fecralloy based TBC system is more governed by formation and coalescence of multiple cracks.

4. Conclusions

The influence of bond coat roughness and of creep strength of bond coat and oxide layer on the thermal cycling damage evolution and lifetime of a plasma-sprayed ZrO$_2$-based thermal barrier coating system was investigated. A model system was used, where FeCrAlY substrates simulated the bond coat. Substrate creep was varied by using the oxide dispersoid strengthened alloy MA956 and the conventional material Fecralloy Eisen-Chrom. Oxide creep was varied by depositing a coarse-crystalline, and a nanocrystalline oxide layer by oxidation and PVD, respectively, before the TBC was plasma sprayed. Roughness depth was varied by sand blasting with different grain sizes. A clear trend to higher lifetimes for the low creep strength substrate was observed. Thermography showed that in case of substrates with low creep strength the delamination area increases continuously until end of life, whereas in case of high creep strength substrates a relatively abrupt acceleration occurs as soon as a delamination exceeds a diameter of approx. 2 mm. The influence of oxide creep seems to be less pronounced than that of substrate creep. However, the highest lifetime was observed for combination of low creep strength substrate and low creep strength oxide layer. The influence of roughness depth remains unclear due to relatively large lifetime scatter. A fracture mechanics model previously published by the same working group proved to roughly cover the influence of substrate creep on thermal cycling life.

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References

[1] N.P.Padture, M.Gell, E.H.Jordan, Thermal Barrier Coatings for Gas-Turbine Engine Applications, Science, 296(2002)280–284.
[2] N.S.Cheruvu, K.S.Chan, R.Viswanathan, Evaluation, degradation and life assessment of coatings for land based combustion turbines. Mater. Sci. Eng. for Energy Systems, 1(2006)33–47.
[3] J.DeMasi–Marcin, D.K.Gupta. Protective coatings in the gas turbine engine. Surf. Coat. Technol., 1994; 68/69:1–9.
[4] R.Herzog, F.Schubert, L.Singheiser, Contribution to life prediction of thermal barrier coatings using the concept of critical strain. *J. Mater. Process. Technol.*, 117(2001)3.

[5] T.Beck, R.Herzog, O.Trunova, M.Offermann, R.W.Steinbrech, L.Singheiser. Damage mechanisms and lifetime behavior of plasma-sprayed thermal barrier coating systems for gas turbines – Part II: Modeling. *Surf. Coat. Technol.*, 202(2008)5901–5908.

[6] J.Rösler, M.Bäker, K.Aufzug. A parametric study of the stress state of thermal barrier coatings Part I: creep relaxation. *Acta Mater.*, 52(2004)4809–4817.

[7] P.Bednarz. Finite element simulation of stress evolution in thermal barrier coating systems. Jülich: *Forschungszentrum Jülich GmbH*, 2006.

[8] V.Teixeira, M.Andritschky, W.Fischer, H.P.Buchkremer, D.Stöver. Analysis of residual stresses in thermal barrier coatings. *J. Mater. Process. Technol.*, 92-93(1999)209–216.

[9] A.Gil, V.Shemet, R.Vassen, M.Subanovic, J.Toscano, D.Naumenko, L.Singheiser, W.J.Quadakkers. Effect of surface condition on the oxidation behaviour of MCrAlY coatings. *Surf. Coat. Technol.*, 201(2006)3824–3828.

[10] A.Nusair Khan, J.Lu, Thermal cyclic behavior of air plasma sprayed thermal barrier coatings sprayed on stainless steel substrates. *Surf. Coat. Technol.*, 201(2007)4653–4658.

[11] R.Vaßen, G.Kerkhoff, D.Stöver. Development of a micromechanical life prediction model for plasma sprayed thermal barrier coatings. *Mater. Sci. Eng. A*, 303(2001)100–109.

[12] F.Traeger, M.Ahrens, R.Vaßen, D.Stöver. A life time model for ceramic thermal barrier coatings. *Mater. Sci. Eng. A*, 358(2003)255–265.

[13] D.Zhang, S.Gong, H.Xu, Z.Wu, Effect of bond coat surface roughness on the thermal cyclic behavior of thermal barrier coatings. *Surf. Coat. Technol.*, 201(2006)649–653.

[14] H.Lau, C.Leyens, U.Schulz, C.Friedrich. Influence of bondcoat pre-treatment and surface topology on the lifetime of EB-PVD TBCs. *Surf. Coat. Technol.*, 165(2003)217–223.

[15] M.Schweda, T.Beck, L.Singheiser, Thermal damage evolution of a thermal barrier coating and the influence of substrate creep, interface roughness and pre-oxidation. *Int. Journ. Mat. Res.*, (formerly Z. Metallkd.) 103(2012)40–49.

[16] M.Schweda, Optimierung von APS-ZrO2-Wärmedämschichten durch Variation der Kriechfestigkeit und der Grenzflächenrauhigkeit. Jülich: *Forschungszentrum Jülich GmbH*, 2011.