

Characterisation of air plasma sprayed TBC coating during isothermal oxidation at 1100°C

L. Swadźba ^a, G. Moskal ^{a,*}, B. Mendala ^a, T. Gancarczyk ^b

^a The Silesian University of Technology, ul. Krasińskiego 8, 40-019 Katowice, Poland

^b WSK "PZL Rzeszów", ul. Hetmańska 120, Rzeszów, Poland

* Corresponding author: E-mail address: grzegorz.moskal@polsl.pl

Received 23.10.2006; accepted in revised form 15.11.2006

Manufacturing and processing

ABSTRACT

Purpose: The purpose of the research was characterization of failure modes of air plasma sprayed thermal barrier coatings system in isothermal oxidation test conditions at 1100°C.

Design/methodology/approach: The research allowed the identification of microstructural changes that take place during oxidation test of TBC system in area of ceramic top-layer, bond coat and superalloy substrate. The examinations were conducted on specimens after 50, 120, 500, 1000, 1500 and 2000h of exposition. The scope of the research encompassed a microstructural analysis with the use of macro and micro investigation - LM, SEM microscopy.

Findings: It was found that the lifetime of commercially quality TBC system is related to progress of degradation in area of bond coat and ceramic top layer. During isothermal oxidation, the bond coat in the TBCs was oxidized to form an alumina and a mixed oxide layer between the top coat of yttria stabilised zirconia (YSZ) and bond coat of NiCoCrAlY alloy. At the moment of oxidation acceleration, the cracks initiation and propagation was observed in the mixed oxide layer near the YSZ. The crack propagation induced the delamination and spallation of top coat after 1000h oxidation.

Research limitations/implications: The discussed research proves that main reason of degradation in TBC system is related to progressive development of cracks at the interface of the thermally grown oxide with bond coat, within the top coat or at its interface with TGO.

Practical implications: The results obtained allow the determination of the degree of life-time lost of the investigated TBC system and specification of the time of safety operation.

Originality/value: The results obtained are valuable contribution to durability characterization of TBC systems. They enable the identification of the degradation mechanisms, which enhances the durability and safety of high temperature operation.

Keywords: Thin & thick coatings; TBC; Degradation; Oxidation

1. Introduction

During the last decade, research efforts were devoted to the development and manufacturing of ceramic TBCs on turbine parts because the traditional turbine materials have reached the limits of their temperature capabilities. Thermal barrier coatings (TBCs) are used to sustain the highest temperature at the surface in high temperature superalloy substrates. TBCs have been wider used in hot-section metal components in gas turbines either to increase

the inlet temperature with a consequent improvement of the efficiency or to reduce the requirements for the cooling air [1–10]. The typical TBC used in gas turbines consists of a bond coat produced by the vacuum or low pressure plasma-sprayed MCrAlY (M = Ni, Co) and a top coat of yttria partially stabilized zirconia made by the atmospheric plasma spraying or electron beam-physical vapor deposition (EB-PVD) [11,12]. Plasma-sprayed zirconia-yttria ceramic layer with a nickel-chromium-aluminum-yttrium bond coat on a substrate made of nickel-based superalloy a common superalloy/TBC system. Application of

these superalloy/TBC systems can be found in both aerospace and land-based gas turbine engines. In automotive applications, the piston head for diesel engine is coated to achieve longer life time and higher performance in terms of fuel reduction and power. However, these coatings have durability problems, due to the material and thermal mismatch between the coating and the metallic substrate. Thermal residual stresses develop during cool down from processing temperatures in TBC/metallic substrate. Environmental effects, specially oxidation, create additional residual stresses due to the growth of an oxide layer causing additional material mismatch between the oxide surface and the TBC. These residual stresses may initiate microcracks such as debonding and radial cracks and can have profound effects on the response of the TBC and interfacial damage accumulation and failure. Their understanding is essential to predict the behavior of the coatings and their performance [13]. The bond coat is arguably the most crucial component of the TBC system [14]. Its chemistry and microstructure influence durability through the structure and morphology of TGO created as it oxidized. Moreover, system performance is linked to its creep and yield characteristics. BCs are in two categories: one is based on the NiCoCrAlY system and the second category consists of a Pt-modified diffusion aluminide. The interface between BC and TGO, which morphology changes in service, is another critical element: generally, high stresses are present in the interfacial zone, due to oxide thickening, to thermal expansion misfit and to applied loads. As a result, crack can initiate and propagate, causing the ceramic layer to spall off, leading to the system degradation [15].

2. Description of experiments, methodology and materials

Ni-base superalloy In 738 was the substrate material for the TBC system used to this investigations. As a bond coat a Amdry 365-2 powder was first applied by plasma spraying to use the bonding and adherence of TBC on the substrate and to improve the corrosion resistance of the TBC as well. As a ceramic top-coat a Metco 204B-NS zirconia powder (stabilized with 8 wt.% yttria) was plasma sprayed on the bond coat. Samples (rolls with diameter 1,5 cm and height 3 cm) were prepared by APS method using typical plasma-spray parameters. All bond coatings were identically deposited to a nominal thickness of 120µm as well as ceramic top layer (ca.500µm thick). The composition of Amdry 365-2 bond coat powder was Ni23%Co17%Cr12,5%Al0,45%Y (wt.%). The samples were oxidized isothermally in air at 1150°C for 50, 120, 500, 1000, 1500 and 2000h, respectively.

3. Description of achieved results of own researches

3.1. Characterization of amdry 365-2 and metco 204B-NS powder

Results of XRD investigations of bond coat powder's showed that NiAl phase modified by Co - Al(Ni,Co) and phase on the

base Ni - Ni(Cr,Y) type were predominant in initial condition. The quantitative analysis proves that contribution of aluminide phase is ca. 60%. SEM investigations showed that powder shape is generally nearing to sphere with small effect of conglomeration. X-ray diffraction of ceramic powder in initial conditions as well proved the presents of zirconia in three crystallographic form: monoclinic, tetragonal and cubic. The quantitative investigations give the possibility of estimation of capacity of monoclinic zirconia and other phases. In the case of Metco 204B-NS powder the contribution of monoclinic ZrO₂ was 7,5%, cubic – 37% and tetragonal 55%. Metco 204B-NS ceramic powder exhibit in generally nearing to sphere shape of powder with small effect of conglomeration.

3.2. Characterization of as-sprayed TBC system

X-ray diffraction of as-sprayed ceramic top layer detected the presents of primarily of a tetragonal ZrO₂ phase, with minor amounts of cubic and monoclinic phases. The thickness of ceramic layer was ca. 550 µm and was very similar on the all surface of the specimen. The thickness of bond coat was ca. 130 mm. The typical microstructure of the plasma sprayed YSZ could be seen with pores, lamellae boundaries and microcracks.

The metallography of the as-sprayed NiCoCrAlY bond revealed a dense, non-homogeneous coating structure containing visible splat boundaries and very small interlamellar porosity, isolated spherical internal porosity, and non-uniform dispersion of Y₂O₃ and Al₂O₃. Total porosity of the ceramic top layer and bond coat were evaluated by image analysis from the polished cross section. In initial condition of TBCs total porosity was ca. 0,9%. The details of ceramic top coat showed pores and probably vertical segmentation crack.

Figure 1 shows the failure progress of TBCs during isothermal conditions. The failure nature is different in different time of test. There is not observed macro-degradation effect after first 500h of exposure. The most remarkable differences is observed from 1000 to 2000h of oxidation test. After 1000h the final failure appears by macro-cracking starting from edge of the ceramic top coat but still larger areas of the TBC system are adherent. After longer time of exposure during in this conditions the failure occurred is related to partially (1500h) and nearing totally delamination and in consequence spallation of the ceramic top-coat. Cross-sectional metallurgical investigations after 50h of oxidation showed any important effect of cracks nucleation and delamination processes. The dark layer of TGO between ceramic layer and bond coat is dense with small amount of gray areas of oxides attached to the YSZ. After 50h of exposition the thickness of oxides in TGO is ca. 6µm. After 120h of exposition dark layer of oxides in TGO became more bigger then after 50h (Fig.2). The microanalysis of chemical composition of oxides areas in TGO showed that dense dark areas are rich in Al (the main component is Al₂O₃, but in the gray oxides EDS showed presents of Al, Cr, Co and Ni. In this area exist probably mixed oxides such as Al₂O₃, Cr₂O₃ and probably spinel (Ni,Co)(Cr,Al)₂O₄. Prior the final failure, damages in the form of micro-cracks were observed in the area of TGO. All this cracks are oriented parallel to the interfaces BC-TGO-YSZ.

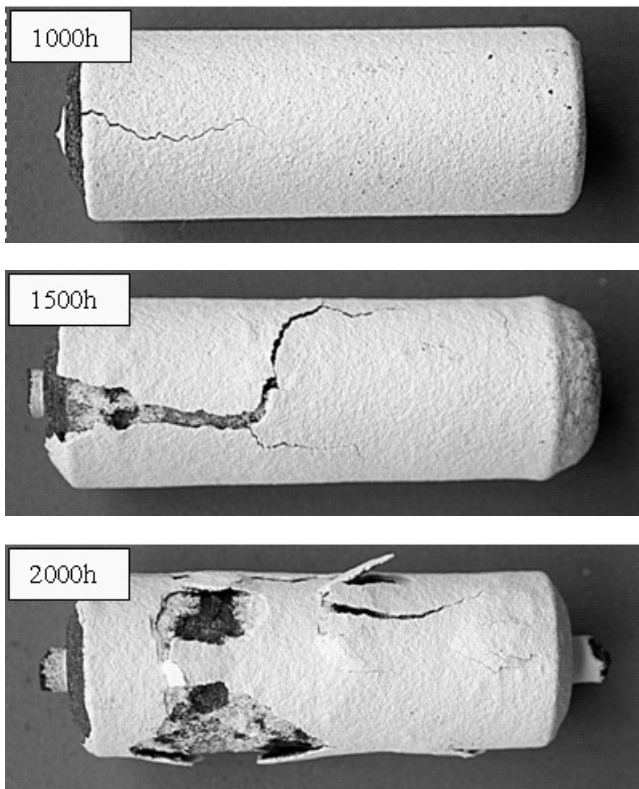


Fig. 1. General view of specimens with TBCs after each stage of isothermal oxidation at 1100°C

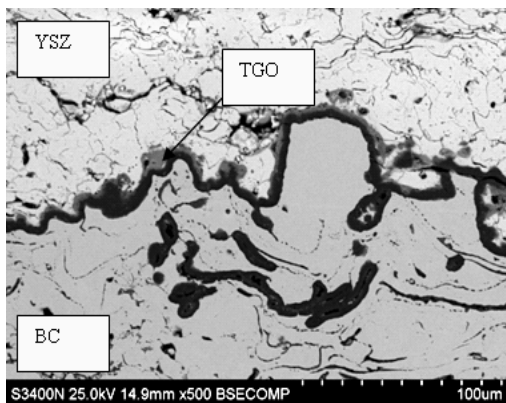


Fig. 2. Cross section of sample oxidized at 1100°C for 120h

The presents of micro-cracks and cracks in TGO led to oxygen penetration through the TGO and in consequence internal oxidation of in the bond coat were some areas of oxides were detected. The investigations of TBC system after 500h of oxidations in air showed initial stages of partially delamination effects in area of TGO and YSZ. In additions, the horizontal cracks are observed from top surface of ceramic to bond coat. The micro-cracks were either located within TGO or start in the TGO and subsequently penetrate into the zirconia top-coat (Fig.3). The main crack is formed in the TGO in the area of porous gray mixed

oxides. This type of oxides dominated after 500h of exposition. For longer exposure time of $t > 1000h$ the longer cracks in TGO area were observed. Cracks were parallel to the BC-TGO interface and analogical for earlier observations, their penetrate into zirconia top-coat.

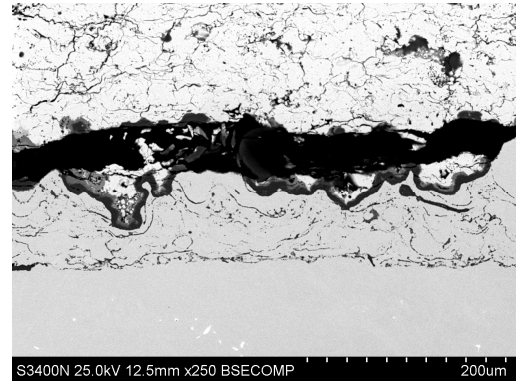


Fig. 3. Cross section of sample oxidized at 1100°C for 500h

In macro-scale the effect of delamination was much stronger than in 500h of exposition case (macro-cracks on the top-coat ceramic surface). After 1500h of isothermal oxidation there was many of microcracks in TGO and longest cracks in ceramic sublayer. In additions, the deep horizontal cracks are observed from top surface of ceramic to bond coat to 1500h of exposition. After 2000h exposure the main mode of degradation was related to totally delamination and spallation of ceramic layer. Metallographic examinations of cross section showed presents not only horizontal cracks but also long vertical macro-cracks especially in area of ceramic top-coat. Process of delamination was located generally in zirconia near TGO interfaces. The oxidation kinetics of TBC was determined by measuring the thickness of the TGO as a function of square root of oxidation time (Fig.4). The oxidation rate followed the parabolic law for up to 500h. This stable state of oxidation is due to protective influence of TGO, because its growth rate was quite low. At longer time of exposure oxidation led to a substantial increase in the TGO growth rate and in consequence in thickness, but it appeared that the oxidation rate still followed the parabolic law with high oxidation rate constant.

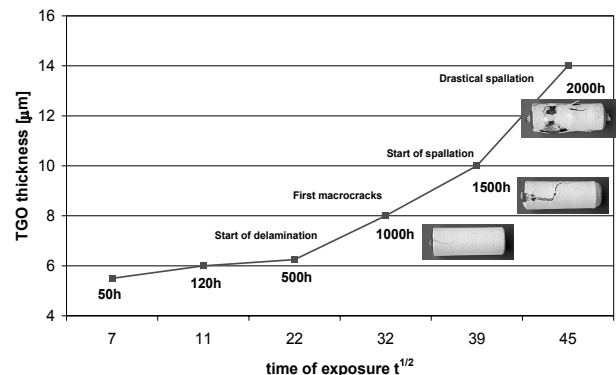


Fig. 4. Scale thickness vs. oxidation time for TBC

3.3. Summary

The high temperature degradation of air plasma sprayed thermal barrier coating at 1100°C was evaluated. The results obtained by isothermal oxidation test to 2000h of exposure in air showed that the initial significant degradation effects of TBC were observed after 500h of exposition. This degradation is related to micro-cracking in TGO area and in consequence with local delamination on interface between TGO and ceramic top-layer. In this stage alumina was formed initially net to the NiCoCrAlY sublayer. A mixed gray oxides layer of probably chromia and $(\text{Ni,Co})(\text{Cr,Al})_2\text{O}_4$ spinel were formed near to zirconia during further oxidation. The thickness of this oxides sublayer increase continuously to the time of oxidation progress. The examination after 1000h of exposition showed significant effect of macro-cracks and in next step local and full effect of spallation of ceramic top-coat. This effect is due to increasing rate of oxidation, primary controlled by diffusion through the alumina, then controlled by the diffusion through the mixed oxide. The substantial increase in the oxidation rate lead to the formation of micro-cracks in the TGO area, their growth to the form of vertical and horizontal cracks and in consequence their transformation to macro-cracks, delamination and finally spallation of top-layer.

4. Conclusions

The main mode of air plasma spraying TBCs degradation is related to thermal expansion mismatch strains and interfacial oxide scale growth, which are the dominant mechanism leading to failure of TBC system by spallation of the YSZ top coat.

It was found that that crack initiation and propagation in the TGO layer also caused the failure of thermal barrier coating system.

The significant effect of degradation was observed after 500h of exposure – starting of delamination; 1000h- macrocracking of top surface layer; 1500h – partially spallation of zirconia, 2000h – drastically effects of spallation of top-coat.

References

- [1] F. Cernusci, P. Bianchi, M. Leoni, P. Scardi, *Journal of Thermal Spray Technology* 8 (1) (1999) 102.
- [2] J.T. DeMasi-Marcin, D.K. Gupta, *Surface and Coating Technology*, 68/69 (1994) 1.
- [3] J. Wigren, L. Pejryd, in: C. Coddet (Ed.), *Proceedings of the 15th International, Thermal Spray Conference on Thermal Spray Meeting the Challenges of the 21st Century*, France, SMInternational, Materials Park, OH, USA, 1998, 1531.
- [4] K.A. Khor, S. Jana, *Pulsem laser processing of plasma sprayed thermal barrier coating*, *Journal of Materials Processing Technology* 66 (1996) 4-8.
- [5] B. Siebert, C. Funke, R. Vaben, D. Stover, *Changes in porosity and Young's Modulus due to sintering of plasma sprayed thermal barrier coatings*, *Journal of Materials Processing Technology* 92-93 (1999) 217-223.
- [6] M. Konter, M. Thumann, *Materials and manufacturing of advanced industrial gas turbine components*, *Journal of Materials Processing Technology* 92-117 (2001) 386-390.
- [7] J. Kamalua, P. Byrdb, A. Pitman, *Variable angle laser drilling of thermal barrier coated nimonic*, J. Kamalua, *Journal of Materials Processing Technology* 122 (2002) 355-362.
- [8] V. Teixeira, M. Andritschky, W. Fischer, H.P. Buchkremer, D. Stover, *Analysis of residual stresses in thermal barrier coatings*, *Journal of Materials Processing Technology* 92-93 (1999) 209-216.
- [9] J.F. Li, H.L. Liao, C.X. Ding, C. Coddet, *Optimizing the plasma spray process parameters of yttria stabilized zirconia coatings using a uniform design of experiments*, *Journal of Materials Processing Technology* 160 (2005) 34-42.
- [10] A.K. Ray, *Characterization of bond coat in a thermal barrier coated superalloy used in combustor liners of aero engines*, *Materials Characterization* 57 (2006) 199-209.
- [11] W.A. Nelson, R.M. Orenstein, *Journal of Thermal Spray Technology* 6 (2).
- [12] D. Stover, C. Funke, *Directions of the development of thermal barrier coatings in energy applications*, *Journal of Materials Processing Technology* 92-93 (1999) 195-202.
- [13] S.Q. Nusier, G.M. Newaz, *Growth of interfacial cracks in a TBC/superalloy system due to oxide volume induced internal pressure and thermal loading*, *International Journal of Solids and Structures* 37 (2000) 2151-2166.
- [14] A.G. Evans, D.R. Mumm, J.W. Hutchinson, G.H. Meier, Pettit FS., *Mechanisms controlling the durability of thermal barrier coatings*, *Progress in Materials Science* 46 (2001) 505-53.
- [15] M. Martena, D. Botto, P. Fino, S. Sabbadini, M.M. Gola, C. Badini, *Modelling of TBC system failure: Stress distribution as a function of TGO thickness and thermal expansion mismatch*, *Engineering Failure Analysis* 13 (2006) 409-426.