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Optimized sol–gel thermal barrier coatings for long-term cyclic oxidation life

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Abstract

New promising thermal barrier coatings (TBCs) processed by the sol–gel route are deposited onto NiPtAl bond coated superalloy substrates using the dip and/or spray coating technique. In this study, the optimization of the process, including an appropriate heat treatment prone to densify the yttria-stabilized-zirconia (YSZ) top-coat and leading to the sintering and the development of a resulting crack network, is investigated. In particular, relevant information on internal strain evolution during the heat treatment are obtained using in situ synchrotron X-rays diffraction and confirm a stabilization of the TBC through the occurrence of the micro-cracks that beneficially releases the in-plane sintering stress. Such TBCs are subsequently reinforced using additional material brought within the cracks using sol–gel spray coating. The effect of various process parameters, such as the pre-oxidation of the bond-coat, on the sol gel TBCs consolidation and their cyclic oxidation resistance enhancement, is presented. Reinforced sol–gel TBCs are successfully oxidized up to more than one thousand 1 h-cycles at 1100 °C, without any detrimental spallation.

Keywords: Thermal-barrier-coating; Cyclic-oxidation; XRD-synchrotron; Sol–gel-processing; Spallation

1. Introduction

Thermal barrier coatings (TBCs) are widely used for various applications in turbojet engine gas turbines and combustion chambers in relation with their excellent thermal protection properties allowing drastic improvement of component durability and efficiency.[1,2]

Typically, the overall thermal protection system includes: (i) the TBC itself, a ceramic top coat (TC) made of yttria-stabilized-zirconia (YSZ) acting as thermal insulator, (ii) the superalloy substrate that supports mechanical loading, and (iii) an aluminium rich Bond Coat which enhances the cohesion between the substrate and the TBC and develops by oxidation a fine alumina film referred to as the thermally grown oxide (TGO). Within this multi-materials structure, each layer, characterized by specific physical, thermal and mechanical properties, shows – upon processing and "in-service" thermal exposures – distinct thermomechanical behaviour, thereby resulting in the establishment of internal thermal stresses. In addition, during high temperature exposure, the alumina (Al_2O_3) TGO, acting as a diffusion barrier, continuously grows at the interface BC/TBC which likely induces local increases of the mismatch between the BC and TBC layers.

As a consequence, in highly complex TBC systems, failure mechanisms upon temperature cycling are very intricate and lots of theoretical as well as experimental research investigating mechanistic behaviours and microstructural mechanisms are dedicated to understanding the various processes of crack initiation and propagation, delamination and spallation.[2,3]

Up to now, two main coating processes are used to deposit TBCs for industrial applications, namely the electron beam physical vapor deposition (EB-PVD) and the air plasma spray (APS), each generating specific layer morphology, deposit microstructure and thermo-physical properties.

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EB-PVD results in a columnar structure with grain boundaries roughly normal to the substrate surface allowing satisfactory accommodation of in-service lateral thermomechanical stresses and related strains. However, the thermal conductivity of EB-PVD coatings is not fully optimized as the perpendicular growth of the columns favours extensive heat transfer between the outer surface of the multi-material system and the superalloy substrate. As a comparison, the typical microstructure for APS coating is highly lamellar, resulting in both lower thermal conductivity as the porosity is distributed perpendicular to the heat flux, and lower capability to satisfactorily sustain lateral constraints. Various alternative ways to synthesize TBCs have been proposed in the literature, based on soft chemical processing.[4–6]

In this paper, a new promising method for depositing and reinforcing TBCs, is investigated. Indeed, recently the synthesis and deposition of TBCs using a new attractive sol–gel route has been successfully developed.[6–9] This versatile technique, promoting, on contrary to EBPVD and APS, non-directional deposition, allows to produce either thin or thick coatings by using dip or spray technique or combined method of both techniques depending on the required result. Sol–gel TBCs show isotropic microstructure with randomly distributed porosities, which straightforwardly results in an interesting compromise between thermal conductivity and mechanical strength. In previous papers,[8,9] the optimization of the manufacturing process of sol–gel TBCs, the means to enhance their cyclic oxidation resistance using structural reinforcement as well as the mechanisms responsible for their possible damages due to long term cyclic exposure at high temperature have been discussed in detail.

Essentially, the degradation of sol–gel TBCs is initiated by the formation of a regular crack network occurring either during the post-deposition thermal treatment required to sinter the deposit or during the very first cycles of oxidation. It is worth to notice that, in both cases, this regular surface crack network is a result of the in-plane stress release due to the sinter-induced shrinkage of the zirconia scale. Subsequently, under cumulative oxidation cycles, enlargement and coalescence of the cracks occur, promoting the detachment of individual TBC cells and further the complete spallation of the TBC.

To improve the cyclic oxidation resistance of the TBCs, two refinements were proposed both related to the overall processing, namely (i) to enhance the efficiency of the sintering thermal treatment carried outright after TBC deposition and (ii) to stabilize the crack network by filling crack grooves using supplementary dip or spray coating passes. It was shown that the heat treatment parameters such as the heating/cooling rates and the holding time at dwell temperature, dramatically impact the geometrical characteristics of the crack network and consequently their response to cyclic oxidation. After adjustments, the "optimal" thermal treatment parameters, resulting in a significant extent of the TBC life, correspond to an exposure at 1100 °C during two hours with heating and cooling rates of 50 °C/h.[8]

In addition, the feasibility of consolidating sol–gel TBCs by additional fillings of zirconia into the sinter-induced cracks was investigated by adjusting different process parameters such as the choice of either dip-coating or spray-coating and the modification of the slurry viscosity.[9] It subsequently turned out that spray-coating technique leads to a more efficient and a more homogeneous filling of crack as well as the selection of a specific slurry viscosity for each individual pass depending on the depth and width of crack to fill (by modifying the weight percent of powder). This filling optimization allows salient improvement of the cyclic oxidation behaviour of the spray-coat reinforced TBCs.[9]

Note that the failure mechanisms of the optimized and reinforced sol–gel TBCs are more complex than that of non reinforced TBCs. This results from the more connected microstructure – though more uniform in thickness – of the reinforced TBC showing a composite-like morphology including a skeleton or frame, corresponding to the partially filled cracks, and a matrix, namely the initial sintered YSZ.

The degradation of such TBCs results from the initiation and propagation of cracks, mainly located at the interface between the TBC and the TGO. As a matter of fact, as reinforcement of crack prevails, cracks can extend much more than in non-reinforced TBC before generating spallation, greatly limiting the detachment of individual spalls. Spallation develops following the complete propagation of the crack throughout the whole specimen, producing – when occurring subsequently to a high number of cumulated oxidation cycles – large-scale degradation as observed in EB PVD TBC. Basically, the optimization of both the sintering heat treatment and the procedure for filling the initial crack network, allows a significant improvement of the sol–gel TBC durability during cyclic oxidation at 1100 °C. Typically, sol–gel TBC properly sintered and adequately reinforced can be cycled for 1 h at 1100 °C one thousand and five hundred times without spalling which is roughly equivalent to the performance of EB-PVD TBCs.

The present paper proposes to investigate thoroughly (i) the crack network formation due to the initial sintering as well as the effect of the very first oxidation cycles in sol–gel TBCs using synchrotron radiation to monitor "in-situ" the evolution of the thermal strain throughout the TBC and (ii) the 1100 °C cyclic oxidation durability of sol gel TBCs for which optimized processing and filling as well as a preliminary oxidation of the bond coat is applied. The overall performances of reinforced sol gel TBCs are compared to previous results and EBPVD TBCs.

2. Materials and experimental techniques

2.1. Processing of the sol–gel TBC

The various operations conducted to synthesize thermal barrier coatings by the so-called sol–gel route, are presented in details in previous works developed in the laboratory.[6,8,9] Main steps can be summarized as follows:

i) First step consists in the production of YSZ powders by hydrolysis/condensation (to obtain a gel), supercritic drying and heat treatment at 700 °C of a precursor YSZ sol (9.7% mol $YO_{1.5}$). YSZ aerogel powders crystallize in the tetragonal form. N_2 adsorption/desorption analysis of such