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Effect of aging on the onset of cracks due to redistribution of residual stresses in functionally graded environmental barrier coatings of mullite/ZrO_2

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ABSTRACT

Environmental barrier coatings (EBCs) are proposed as an option to reduce the high temperature water vapour corrosion in gas turbines ceramic components made of Si_3N_4 or $SiC/SiC_{f,}$ which are projected to achieve further energy efficient gas turbines. These coating are commonly designed as multilayer systems firmly attached to the ceramic substrate with the aim of retarding or avoiding its degradation after exposure to environmental conditions close to those in gas turbines. Therefore, to fulfil this function crack formation/propagation in the coatings must be controlled. In present work, three types of environmental barrier coatings fabricated by air plasma spray and containing a Si layer attached to SiC substrate plus 2 to 5 layers of different mullite/Y_2O_3 stabilized–ZrO_2 mixtures are examined. To determine the level of residual stresses in the as-sprayed coating/substrate systems a three dimensional finite element model is developed and also tested for same coatings but aged under, high temperature and rich water vapour atmosphere. The model calculates the zones of maximum tensile stresses in the coatings which agree with experimental observation identifying the type, number and location of cracks. This model could be extended to similar EBC systems, and more importantly, could be use as a powerful designing tool for these complex structures.

1. Introduction

The seek for higher efficiency and energy saving in the future gas turbine engines, in addition to the requirements of environmental friendly systems, has greatly focused on using high performance non-oxide ceramics (SiC/SiC_f and in situ toughened Si_3N_4) for the hot-sections [1]. During turbine operation the extreme temperature and water vapour pressure conditions can corrode these materials [1], and this effect is retarded or minimized by depositing the so-called environmental barrier coatings (EBCs). In particular, different multilayer coatings have been designed to serve as operative barrier against diffusion of hot gases towards the substrate and to suppress the deleterious crack formation as well [2–4]. Several studies have evidenced that the layer arrangement in a multilayer system has a crucial influence over the crack growth resistance and the mechanical reliability of the whole system [5,6]. Therefore, determining the crucial parameters for the design of these complex multilayer EBCs presents a clear interest and furthermore, can help to assess the behaviour of tentative systems after subjected to aging conditions.

In a recent work by some of the authors, graded mullite/YSZ (Y_2O_3 stabilized ZrO_2) EBC systems consisting of a YSZ top layer, a Si coated SiC substrate and diverse mullite + YSZ intermediate functional layers aimed for reducing thermal expansion mismatch were investigated under hot water vapour environments simulating engine conditions [7]. In general, the mullite/YSZ layered systems showed good water vapour corrosion under cyclic and static conditions, where some layer combinations seemed more successful for reducing crack formation/propagation. Hence, it became clear that the build-up of residual stresses in these complex systems is decisive, and find out the distribution of residual stresses in the coatings can help not only to the design of more promising systems but also to understand the behaviour of these coatings after being subjected to corrosion conditions. Presently,

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some of the more successful EBC systems in terms of crack suppression consist on multilayer coatings of the type Si/Mullite/BSAS ($BaO–SrO–Al_2O_3–SiO_2$ compound), which clearly benefit from the low elastic modulus of BSAS phases [8]. Our previous work [7] also raised the question on the beneficial effect of a confined compliant mullite porous layer for reducing the number of cracks. Consequently, porosity was induced as a way to decrease the layer stiffness without compromising much the diffusion hindrance of hot gases towards the substrate.

In the present work, the distribution and the effect of residual stresses are studied using a three-dimensional finite element (3D FE) model on three mullite/YSZ multilayer coatings over SiC substrates. The coatings, deposited by air plasma spraying, are formed by different number of layers of variable composition and two of the systems enclose a relatively porous compliant mullite layer, to quantify the effects of these modifications in the overall residuals stress distributions. The changes in elastic modulus caused by long term thermal aging in rich water atmosphere, simulating combustion conditions, are also used to model redistribution of stresses in the coatings after aging. Location of maximum tensile stresses predicted by the model is supported by onset observation of "tunnelling" and "edge" cracks in the different coatings. The present 3D FE model can serve as a powerful tool to tailor architectures beforehand not only for present coatings but in alike complex multilayer systems as well.

2. Characteristics and properties of the multilayer EBC systems

Coatings were created by sequentially plasma spraying mullite/YSZ blends, always ending with a bimodal ZrO_2 topmost layer and having a Si layer attached to the SiC substrate of 25 × 25 mm size. The formation of amorphous mullite that could induce extensive cracking upon crystallization was essentially prevented as stated in a previous paper [9]. The composition of each layer was: Si, mullite (M100), 75 vol.% mullite +25 vol.% YSZ (M75), 50 vol.% mullite +50 vol.% YSZ (M50) and YSZ. Coatings were aged under conditions plainly described in Ref. [7], which basically consisted in long time exposure (up to 200 h) at thermal cycles of 2 h at 1300 °C under a water vapour rich atmosphere [7].

The layered arrangement of the three different coatings with the composition and thickness of each layer is shown in Table 1. The M100FG label stand for the mullite layer but with some porosity (~20%), induced by the freeze granulation method [10] used for conditioning the powders (see Ref. [7] for further details), and the n-YSZ refers to a nanostructured zirconia layer that also presents some inherent porosity [7,11]. SiC-substrate has 3000 μm thickness for all coatings. System EBC1 is the less functionally graded, composition wise, followed by the EBC2 system and EBC3 as the most graded one, besides EBC1 and EBC2 systems contain a porous mullite layer.

General views of a polished cross-section of these coatings obtained with the scanning electron microscope (SEM, Tabletop Microscope TM-1000, Hitachi, Japan) are displayed in Fig. 1. The presence of regularly spaced vertical cracks is mainly perceived in the denser layers (see arrows). To avoid the introduction of accidental cracks in the coatings during the machining process the 2.5 × 2.5 cm billets were vacuum impregnated with epoxy resin and sliced in three pieces of about 2.5 cm × 0.6 cm. Polishing was done with an automatic polishing machine (Phoenix 4000, Buehler, Germany) using diamond suspensions of 6, 3 and 1 μm.

Table 2 presents, for the different layers, the values of the elastic modulus (E), Poisson coefficient (ν) and thermal expansion coefficient (α) that will be used in the FEM model. E was measured by Vickers micro-indentation tests carried out on the polished cross sections of as sprayed an aged specimens. At least 5 measurements with a load of 2.9 N for 15 s were performed for each layer using an instrumented hardness testing machine (ZHU 2.5; Zwick GmbH & Co., KG, Germany) that simultaneously records load and displacement. The reduced modulus is calculated from the load displacement curve during the unloading cycle by the equipment software, based on indentation contact analysis [12]. The used indentation load produces typical imprint diagonals of ~30 μm in length, which scale with characteristic features of the layers microstructure and, at the same time, are small enough to avoid any influence of the adjacent layers because the minimum layer thickness is about 60 μm. The possible effect of the epoxy resin on the indentation characteristics by penetrating into the pores in the case porous layers (n-YSZ, M100FG) may be estimated considering typical elastic modulus of epoxy resins (~3 GPa) and by using a simple mixing rule calculation; accordingly, an overestimation of the elastic modulus of about 25% is reckon (for 17% porosity), which was disregarded in the FE modelling as did not introduce sensible changes. For the denser layer the influence over the elastic modulus is null. The values of the coefficient of thermal expansion (α) for M100, M100FG, M75 and M50 layers were measured in bulk specimens of each compositions between room temperature and 900 °C with a dilatometer (Netch Gerätebau 402 EP, Germany) equipped with silica support and rod and at heating rate of 5 $°C\ min^{-1}$; for the rest, reference values were used. Values for ν were either taken from the literature or estimated by the mixing rule (mr). Data for the SiC substrate are also displayed in Table 2. We assume that ν and α essentially remain unchanged after aging as both Poisson ratio and thermal expansion coefficient of ceramics do not significantly change up to porosity levels of 50% [13,14], limit that is well above the porosity estimated for the aged porous n-YSZ layer (20%) [7].

The cracks observed in the coatings were counted on SEM micrographs of polished cross-sections. They were classified according to their morphology and nucleation zone (Table 2) as: *Surface* cracks, vertical cracks that initiate at the top n-YSZ layer, *interior* or vertical cracks that nucleate at middle layers without reaching the top coat and *branched* cracks, formed by the coalescence of *surface* cracks. When two or more *surface* cracks are linked by a horizontal crack that propagates through one specific layer they were classified as *horizontal*. Finally, *continuous* cracks were defined as *through thickness* cracks also propagating along the interface between the Si bond coat and the layer immediately above. Table 3 summarizes the different crack types referred to the total number of cracks measured in the particular coating.

It should be mentioned that semicircular crack-like defects observed in the n-YSZ layer, and greatly magnified after aging due to the increased densification, are typical of this nanostructured coating and were not considered for the quantification of the number of cracks [7,11].

Table 1
Composition and thickness of each layer for the three EBC systems. Full coating thickness is also shown.

Reference	Total	Layers					
		Si	M100FG	M100	M75	M50	n-YSZ
EBC1	336 ± 30 μm	86 ± 21 μm	104 ± 24 μm				159 ± 25 μm
EBC2	400 ± 30 μm	74 ± 14 μm	96 ± 19 μm			83 ± 25 μm	155 ± 27 μm
EBC3	369 ± 30 μm	74 ± 14 μm		97 ± 20 μm	58 ± 16 μm	49 ± 14 μm	92 ± 34 μm