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Interaction of molten silicates with thermal barrier coatings under temperature gradients

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Abstract—This paper examines the effect of temperature gradients on the interaction between silicate deposits and thermal barrier coating (TBC) systems. A dedicated test facility, in which a CO_2 laser is employed to impose a controllable thermal gradient through the coating and underlying substrate, is used to investigate the interaction between two silicate compositions with state-of-the-art 7YSZ EB-PVD TBCs. The experimental results are then used to guide the development of expressions that describe the nature of silicate infiltration in the TBCs, the evolution of coating elastic modulus, and the generation and release of stresses.

Keywords: Thermal barrier coating; Delamination; CMAS; Thermal gradient; Infiltration

1. Introduction

The rising operating temperature of aero-engines has increased the prevalence of molten deposits of calcium magnesium alumino-silicates (CMAS) on the hardware surfaces surrounding the hot gas path [1–4]. The combustor and airfoils in the high-pressure turbine, where CMAS deposition is most prevalent, are typically protected by thermal barrier coatings (TBCs). These coatings have microstructures with tailored porosity designed to impart high in-plane compliance to minimize thermal stresses, and low thermal conductivity, to maximize the temperature drop across the coating. Both of these properties are generally degraded by CMAS infiltration [1,5].

Capillarity drives the penetration of CMAS into the coating as molten silicates readily wet oxide ceramics [6,7]. The depth to which CMAS penetrates depends upon the temperature gradient across the coating as well as the composition of the melt through its effects on viscosity, and the chemical interaction with the TBC. As the temperature at the TBC-bond coat interface is typically greater than the glass transition temperature, T_g, infiltration into the coating is either controlled by viscous flow and therefore the time at which the TBC is above T_g, or by the crystallization kinetics. Both of these factors depend on the composition of the glass, with the Si:O ratio [8,9] having a controlling effect on the viscosity and the concentration of minor elements, particularly, Fe [8], and Ti [10], playing a strong role on the crystallization kinetics.

As the molten silicate penetrates into the cooler interior of the coating, crystallization may occur even in the absence of any interaction with the TBC as long as the undercooling is sufficient to activate the crystallization kinetics. Typical crystallization products for CMAS include anorthite, diopside, one of the wollastonite variants, and either tridymite or gehlenite depending on the silica concentration [1,11,12]. More often, however, melt penetration leads to dissolution of the TBC at the interface with the melt [7], and subsequent reprecipitation of one or more crystalline phases. For state-of-the-art TBCs based on 7 wt.% yttria partially-stabilized zirconia (7YSZ), the reprecipitated phases may be Y-lean tetragonal zirconia, transformable to monoclinic on cooling, Y-rich cubic zirconia [7] and zircon, calcium zirconate or garnet [12,13], depending on the composition of the melt.

Once infiltrated, the modulus of the TBC is expected to markedly increase due to the presence of crystalline silicates and/or residual solid glass in the original pores. The rise in modulus increases the magnitude of the stresses in the TBC that are generated from the inherent CTE mismatch between the ceramic topcoat and the superalloy component [4]. It is therefore of interest to analyze how the CMAS induced variation in stiffness through the coating couples

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with the temperature gradient to generate thermal stresses in the TBC and how these stresses drive delamination [14,15].

This work is part of a broader investigation to elucidate the effect CMAS has on the durability of TBC systems and to identify possible mitigation approaches. A novel, laser based, thermal gradient testing facility is used to thermally cycle 7YSZ TBCs with prescribed temperature gradients. The microstuctural changes observed in these experiments are then paired with analytical expressions that capture key features of the TBC–CMAS interaction, namely: the rate of CMAS infiltration in a thermal gradient, the evolution of CMAS induced TBC stiffening, and the development of thermal stresses.

2. Experimental approach

2.1. Thermal gradient test

The TBCs examined in this work were thermally cycled in a facility specially designed to impose a tunable temperature gradient. This experimental set-up is capable of modulating both surface temperatures by using a 2 kW, 10.6 μm, CO_2 laser to heat the surface of the TBC, and an air jet to cool the uncoated backside of the substrate, Fig. 1. To homogenize the incident energy, the laser beam was passed through a ZnSe facetted lens that was rotated at 300 rpm with the center of the lens translated 3 mm off of the beam axis. Additionally, the TBC was raised 6 mm above the focal plane to further enhance the uniformity of the heating profile, Fig. 1(b) [16].

7YSZ TBCs are semi-transparent in the visible and near infrared range ($\lambda = 0.3$–5 μm), but the absorptivity increases with expanding wavelength and above $\lambda > 7$ μm TBCs are highly adsorptive [17,18]. Eldridge and co-workers measured the transmittance of 10.6 μm radiation through a 60 μm thick coating to be 0.2% at 1170 °C and attributed the low transmittance to the large absorption coefficient, which is in excess of 1000 cm^{-1} at temperatures greater than 1000 °C [18,19] which corresponds to an optical penetration depth of less than 10 μm. Further, unlike the scattering coefficient, which is markedly decreased by CMAS infiltration, the absorption coefficient is not expected to be influenced by the presence of CMAS [5]. The highly absorptive nature of the TBC minimizes the need to consider radiative heat transfer through the coating in the analysis.

The surface temperature of the TBC was monitored by a far-infrared, (7–10 μm) imaging pyrometer. A single surface temperature was calculated by averaging the temperature in a region of interest covering the majority of the surface of TBC specimen. The long wavelength pyrometer was used to minimize the collection of radiation from the interior of the coating, which, in the thermal gradient, can skew temperature readings to lower values. Thin, 250 μm diameter, k-type thermocouples were used to measure the temperature on the back surface of the substrate and, notably,

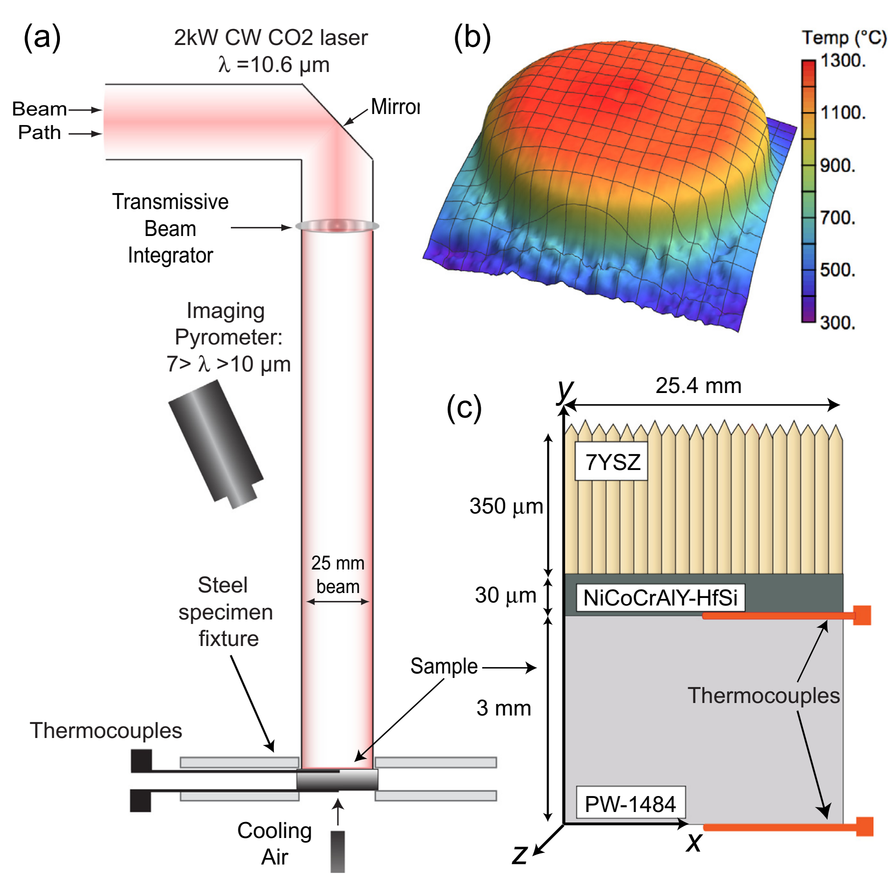

Fig. 1. (a) Schematic illustration of the laser gradient testing apparatus. (b) Thermal image of the TBC surface demonstrating the uniformity of the homogenized laser beam. (c) Schematic illustration of the TBC sample with the coordinate system used in the mechanical analysis is overlaid.