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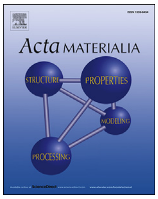

Full length article

Response of ytterbium disilicate–silicon environmental barrier coatings to thermal cycling in water vapor

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ABSTRACT

A preliminary study of a promising bi-layer environmental barrier coating (EBC) designed to reduce the susceptibility of SiC composites to hot water vapor erosion is reported. The EBC system consisted of a silicon bond coat and a pore-free ytterbium disilicate (YbDS; $Yb_2Si_2O_7$) topcoat. Both layers were deposited on α-SiC substrates using a recently optimized air plasma spray method. The two layers of the coating system had coefficients of thermal expansion (CTE) that were well matched to that of the substrate, while the YbDS has been reported to have a moderate resistance to silicon hydroxide vapor forming reactions in water vapor rich environments. Thermal cycling experiments were conducted between 110 °C and 1316 °C in a flowing 90% H_2O/10% O_2 atmospheric pressure environment, and resulted in the formation of a thermally grown (silica) oxide (TGO) at the silicon-ytterbium disilicate interface. The TGO layer exhibited linear oxidation kinetics consistent with oxidizer diffusion through the ytterbium silicate layer controlling its thickening rate. The effective diffusion coefficient of the oxidizing species in the YbDS layer was estimated to be 2×10^{-12} m^2s^{-1} at 1316 °C. Slow steam volatilization of the YbDS topcoat resulted in the formation of a thin, partially protective, high CTE ytterbium monosilicate layer on the outside of the YbDS coating. Progressive edge delamination of the coating system was observed with steam exposure time, consistent with water vapor volatilization of the TGO edges that were directly exposed to the environment. This was aided by outward bending of the delaminated region to relax TGO and YbMS surface layer stresses developed during the cooling phase of each thermal cycle.

1. Introduction

Environmental barrier coatings (EBCs) will be needed to protect SiC-based ceramic components in future gas turbine engines [1]. Their primary purpose is to eliminate the rapid volatilization of SiC in water vapor rich environments [1–6], while also inhibiting oxidation of SiC-based ceramic matrix composites (CMCs) [7–12]. The design of these EBCs is therefore driven by a different combination of objectives to those of the thermal barrier coating (TBC) systems that are widely used to extend the life of superalloy airfoils [13]. The objective of TBC designs is to reduce a metal component's surface temperature while delaying delamination failure from oxidation [14,15], reactions with molten silicate deposits [16,17] and various forms of hot corrosion [18–23]. However, unlike TBC systems where coating spallation results in damage, but usually not loss of the airfoil, the premature failure of an EBC could have consequences that are much more serious, since the life of an unprotected SiC component in a combustion environment is likely to be short [2,3,24–26]. Since no sufficiently durable EBC system for use in thermo-cyclic, water vapor rich environments has been reported to date, the development of such a coating system is of considerable importance for the implementation of SiC-based CMCs in advanced gas turbine engines.

For non-rotating applications involving several thousand hours of environmental exposure at temperatures up to 1316 °C (2400 °F), research has focused on multilayered EBC systems that use a Si "bond coat" applied to the SiC. The design objective for the bond coat is similar to that of the aluminum-rich metallic layer used in TBC systems [27]. It serves as a sacrificial layer that forms a protective thermally grown oxide (SiO_2) upon exposure to oxidizing species, thereby inhibiting their access to the SiC-based component

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surface. However, volatilization of the protective oxide occurs by its reaction with water vapor to form gaseous silicon hydroxides such as $Si(OH)_4$. The bond coat must therefore be covered (environmentally protected) by materials that impede the diffusion of oxygen and water vapor to the Si surface, while being highly resistant to silicon hydroxide forming reactions with water to avoid steam volatilization during engine operation [1,25,28–38].

These design objectives must be achieved subject to numerous constraints. For example, the various layers of the EBC system must have a coefficient of thermal expansion (CTE) similar to SiC to avoid cracking or delamination during heating and cooling over the wide temperature range experienced during operation of a gas turbine engine [32,33]. The EBC materials must be stable and thermochemically compatible so that deleterious reaction products are not formed between them or with the substrate. In addition, they must meet other demands of the engine environment which include resistance to erosion by fine (dust) particles [39–42], impact by larger foreign objects [41–43] and survival of reactions with molten calcium-magnesium-aluminum-silicates (CMAS) [44–48]. Finally, when used on rotated components, EBCs will be required to sustain significant static and cyclic loads that could cause creep deformation and fracture [22,35–37,49–52]. This use environment is therefore one of the most severe ever envisioned for an advanced material system.

The environmental stability, CTE, and elastic modulus of many candidate EBC materials have been reviewed [1,30,34,53]. The ytterbium monosilicate (Yb_2SiO_5)/mullite ($Al_6Si_2O_{13}$)/Si tri-layer system has received significant recent attention due to the thermochemical compatibility between its component materials and the very low steam volatility of Yb_2SiO_5 (YbMS) [53]. However, the CTE of YbMS has been found to be substantially higher than that of SiC ($7.5 \times 10^{-6}\ ^\circ C^{-1}$ for YbMS versus $4.7 \times 10^{-6}\ ^\circ C^{-1}$ for SiC) [1,34]. During cooling from 1316 °C, this was found to result in the development of a high (biaxial) tension in the YbMS layer, which was relieved by channel (mud) cracking [32,33]. The presence of cracks through the YbMS and mullite (which also has a higher CTE than that of SiC) layers then provided oxidizing species a gas phase transport path to the Si bond coat, resulting in the rapid growth of a β-cristobalite (SiO_2) phase on the outer silicon surface. The thermally grown oxide (TGO) also underwent a large reduction in volume (~4.5%) during its cubic to tetragonal (inversion type) $\beta \rightarrow \alpha$ phase transformation upon cooling through ~220 °C, resulting in severe TGO microcracking, loss of its oxidation protection qualities, and premature spallation of the EBC. These observations indicate that high CTE topcoat candidates (including YbMS, other rare earth silicates with high CTE, and aluminum-silicates such as mullite), are likely to be unsatisfactory for the diffusion-impeding layers of EBC systems.

Ytterbium disilicate (YbDS), a line compound in the Yb_2O_3–SiO_2 binary phase diagram with composition $Yb_2Si_2O_7$, is a promising alternate material for environmental barrier layer applications. Its recently measured temperature dependent CTE is about $4.1 \times 10^{-6}\ ^\circ C^{-1}$ [53] (compared to $4.7 \times 10^{-6}\ ^\circ C^{-1}$ for SiC) [47]. YbDS therefore satisfies one of the primary design requirements of an EBC application. However, its resistance to volatilization by reactions with water vapor is significantly less than that of its monosilicate counterpart [53], and its thermochemical stability in contact with a Si bond coat has not been reported. A recent study has led to the identification of an optimized atmospheric plasma spray (APS) deposition method for applying YbDS coatings to Si bond coat protected SiC substrates [54].

The objective of the preliminary study reported here is to begin an investigation of the thermomechanical and environmental response of an APS deposited YbDS/Si EBC system on a SiC substrate as it is subjected to thermal cycling between 110 °C and 1316 °C in a flowing steam/oxygen environment. The study reports and characterizes the growth of a TGO layer on the silicon bond coat during steam cycling for up to 2000 h, and estimates the diffusion coefficient of the oxidizing species through the YbDS coating. It reports and investigates the process by which the topcoat is partially volatilized during steam cycling, and explores the mechanism by which edge coating failure eventually occurs during thermal cycling in a water vapor rich environment. This EBC system is the first to be reported with sufficient resistance to volatilization and thermal cycling for an engine application.

2. Experimental

2.1. Coating deposition

Bi-layer YbDS/Si coatings were deposited onto surface roughened 25.4 mm × 12.7 mm x 4.8 mm thick α-SiC Hexoloy™ substrates (Saint Gobain Ceramics, Niagara Falls, NY), Fig. 1. The substrate edges were first ground forming a 45° chamfer to facilitate over-coating the edges of the sample where delamination most often initiates [32,32]. The target thickness of the silicon layer was 50 μm while that of YbDS layer was 125 μm. The plasma spray deposition parameters used for each layer are summarized in Table 1. The structure of as deposited and stabilization annealed YbDS layers deposited using this spray parameter combination has been recently reported by Richards et al. [54] These spray parameters resulted in a dense YbDS coating with as deposited grain size of several hundred nanometers. During thermal cycling at 1316 °C, this gradually increased to ~5 μm. These spray conditions were chosen as a compromise to ensure deposition of low void content coatings while limiting the loss of SiO from the liquid droplets during transit through the plasma plume. This SiO loss is a result of its higher vapor pressure (compared to ytterbium containing vapor species) at the droplet temperatures encountered in plasma spray deposition [34]. It results in the formation of a two phase YbDS + YbMS coating observable by a difference in gray scale contrast when imaged in the scanning electron microscope using backscattered electrons (BSE mode imaging), Fig. 2(a). In the coatings studied here, the YbMS volume fraction was ~15%, and can

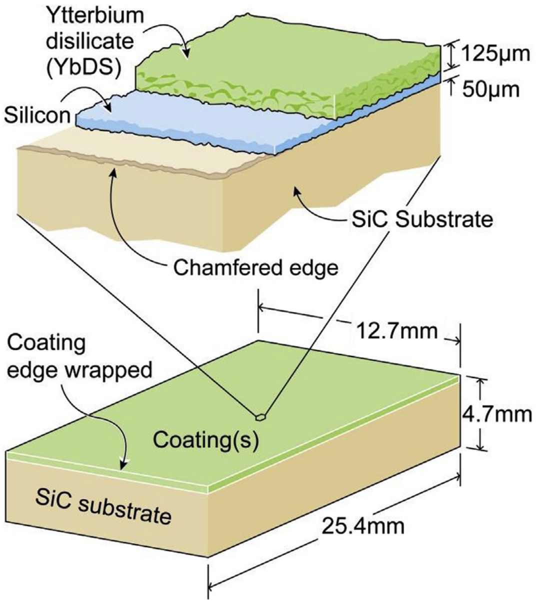

Fig. 1. Schematic illustration of a bi-layer $Yb_2Si_2O_7$/Si (YbDS/Si) coated α-SiC substrate.