Interaction of yttrium disilicate environmental barrier coatings with calcium-magnesium-iron alumino-silicate melts


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Abstract

Reactions between molten calcium-magnesium-iron alumino-silicate (CMFAS) deposits and yttrium disilicate (Y$_2$Si$_2$O$_7$, YDS) based environmental barrier coatings (EBC) on SiC/SiC ceramic matrix composites (CMCs) were investigated at 1300°C. The coating readily dissolves into the melt from which an apatite phase, nominally Ca$_2$Y$_8$(SiO$_4$)$_6$O$_2$, precipitates. These reactions are sufficiently fast to consume the majority of the approximately 275 μm thick coating in 24 h. Liquid phase separation, producing an essentially pure SiO$_2$ second phase, occurs near the reaction front suggesting dissimilar rates of CaO and SiO$_2$ exchange with the overlaying deposit. The rise of large bubbles through the melt above the coatings appears to disrupt the reaction layer and distributes apatite throughout the residual deposit. Channel cracks were found in the deposits and the reaction layers; after longer exposures, the cracks branch and extend laterally through the Si bond coat and into the underlying CMC. Complementary experiments performed on monolithic YDS pellets yielded long-term recession rates similar to those of the coatings, although some differences were evident in recession rates and reaction layer morphologies in the early stages. Thermodynamic calculations were used to understand the evolving driving force for the YDS-to-apatite conversion. The agreement between the simulated and experimentally observed behaviors suggests that such calculations could be used to predict the influence of temperature and deposit composition on EBC degradation.

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1. Introduction

Environmental barrier coatings (EBCs) are essential to the successful implementation of SiC-based ceramic matrix composites (CMCs) in the hot section of advanced gas turbines [1]. The need arises because of the propensity for moisture-induced volatilization of the protective thermally grown SiO$_2$ scale (TGO) on SiC under the high temperature/pressure/velocity conditions characteristic of the combustion gases in the engine [2,3]. Because the TGO/EBC assemblage must act as a barrier to the permeation of O$_2$ and H$_2$O, the layers must be dense. The need to minimize thermal stresses that could induce cracking requires minimization of the mismatch in coefficient of thermal expansion (CTE) with the substrate. In current technology the TGO is generated by a Si “bond coat” that is both thermo-chemically [4] and nearly thermo-mechanically [5] compatible with SiC. The CTE matching restriction limits the choices for EBCs largely to silicate systems, all of which are susceptible to SiO$_2$ volatilization to an extent depending on their SiO$_2$ activity [6].

Currently favored EBC systems are based on rare earth (RE) silicates [7]. The disilicates are thermo-chemically compatible with the underlying SiO$_2$ they are designed to protect, and those of the other smaller REs (e.g., Lu, Yb, and Y) exhibit polymorphs that are also reasonably CTE-matched to the SiC substrate [8]. In contrast, the monosilicates would react with SiO$_2$ to form disilicates, and generally have higher CTEs than the substrate, but have substantially lower SiO$_2$ activity and thus improved resistance to volatilization in flowing water vapor [7,9,10]. This suggests that a favorable architecture may consist of a disilicate layer next to the bond coat, and a thinner monosilicate layer near the surface [11].

It is now established, however, that all viable EBCs are susceptible to substantial degradation by molten silicate deposits generally known as CMAS (calcium-magnesium alumino-silicates) [12–14]. These are generated by mineral debris ingested with the intake air, which deposits on the hot section components forming a melt if the surface temperature exceeds their incipient melting temperature.
point [15]. Coating recession proceeds by dissolution of the EBC material into the silicate melt and, in the case of RE silicates, precipitation of alternate crystalline phases interspersed with residual melt. This modified layer may have a different resistance to volatilization than the original coating and, due to changes in the thermo-mechanical properties, is more likely to crack upon thermal cycling [15]. Understanding the factors influencing the rate of coating recession under silicate deposits and the ultimate failure mechanism(s) is critical to developing EBC life models and to identifying coating materials and architectures offering improved performance.

The present article compares the behavior of yttrium disilicate (Y₂Si₂O₇, YDS) atmospheric plasma spray (APS) coatings with that of dense pellets upon exposure to a model silicate deposit. Attention is focused on elucidating the micro- and meso-scale evolution of the reaction layer as the recession proceeds, as well as identifying failure mechanisms relevant to in-service components. This work complements earlier studies on yttrium monosilicate (Y₂SiO₅, YMS) [13] as well as ytterbium mono- (Yb₂SiO₅, YbMS) and di-silicate (Yb₂Si₂O₇, YbDS) [14,16]. Additionally, whereas the past studies were heated in a box furnace in covered alumina crucibles at 1050 °C for the prescribed time, and cooled at 10 °C/min. The YDS pellets were placed on Pt foil (Alfa Aesar, Haverhill, MA) to prevent reaction with the crucible. The 10 min exposures were performed in a tube furnace with a sliding alumina stage. Specimens on the stage were first moved to a position in the furnace corresponding to 1100 °C for 5 min to thermally equilibrate, then moved to a position corresponding to 1300 °C. After the 10 min dwell, the specimens were cooled rapidly by sliding the stage out of the furnace.

2. Methods

2.1. Materials

Recession experiments were performed on two types of specimens. The first was a multilayer EBC provided by Pratt & Whitney (East Hartford, CT). The coatings, deposited by plasma spray onto a 2 mm thick SiC/SiC CMC panel, include a 250 to 300 μm Si bond coat, a <1 μm thick SiO₂ TGO, and a 250 to 300 μm thick YDS-based top coat. As-received panels were sectioned into coupons measuring 17 mm square using a low-speed diamond saw lubricated with deionized water. The coupons were then heat treated at 1325 °C in air for 4 h to stabilize the microstructure prior to exposure to the model silicate deposit.

Comparative experiments were performed on nominally dense YDS pellets approximately 9.5 mm in diameter, 2 mm thick, and weighing ~0.7 g. The pellets were fabricated from the same YDS powder used to produce the coatings, which was also provided by Pratt & Whitney. The pellets were densified using a field-assisted sintering unit (FAST, FCT Systeme GmbH, Frankenblick, Germany). The procedure involved filling the powder in the FAST die, heating at 100 °C/min, holding for 9 min at 1450 °C to 1500 °C under 90 to 100 MPa pressure, and cooling at 100 °C/min. Prior to the silicate deposit exposure, the pellets were annealed in air for 24 h at 1400 °C and then ground flat using 240 grit SiC paper. Due to the presence of the Si bond coat, it was not possible to anneal the coatings at the higher temperatures used to process the pellet specimens.

2.2. Molten silicate exposure

The synthetic silicate used in this investigation had a nominal composition CsM₂F₂A₁₂S₄S₅, hereinafter CMFAS. Its synthesis, melting, and crystallization behavior were described previously [17]. This deposit is based on the average composition (excluding NiO) of melts infiltrated into TBCs on turbine shrouds following service in a desert environment [18]. Except for the proportional addition of Fe, it is equivalent to the more widely used Cs₃F₃A₁₂S₄S₅ chemistry [12,13,19]. The pre-reacted crystalline deposit has an incipient melting temperature of 1191 °C, a glass transition temperature of 734 °C, and is fully molten at 1300 °C. The CMFAS powder was consolidated into 50 mg, 6.25 mm-diameter pellets, sintered in air at 1100 °C for 12 h, and mechanically thinned to achieve a loading (λ) of 18–19 mg/cm² over the initial contact area.

The pellets and EBC coupons were ultrasonically cleaned in deionized water and dried overnight under vacuum (10⁻³ torr). A CMFAS disk was placed on top of each specimen and allowed to react for 10 min, 4 h, and 24 h at 1300 °C in air; additionally, a 100 h pellet exposure was performed. For exposures ≥4 h, the specimens were heated in a box furnace in covered alumina crucibles at 1050 °C/min, held at 1300 °C for the prescribed time, and cooled at 10 °C/min. The YDS pellets were placed on Pt foil (Alfa Aesar, Haverhill, MA) to prevent reaction with the crucible. The 10 min exposures were performed in a tube furnace with a sliding alumina stage. Specimens on the stage were first moved to a position in the furnace corresponding to 1100 °C for 5 min to thermally equilibrate, then moved to a position corresponding to 1300 °C. After the 10 min dwell, the specimens were cooled rapidly by sliding the stage out of the furnace.

2.3. Characterization

X-ray diffractometry (XRD, Empyrean, PANalytical, Almelo, The Netherlands) was used to ascertain the phase constitution of the starting YDS powder, the specimens at various processing stages, and following CMFAS exposure. As-processed and CMFAS-exposed specimens were mounted in air-curing epoxy, sectioned using a low-speed diamond saw lubricated with DI water, re-mounted in epoxy, and polished to a 1 μm diamond finish. The polished sections were imaged using scanning electron microscopy (SEM, FEI Sirion XL30, Hillsboro, OR) in secondary (SE) and back-scattered (BSE) imaging modes. The volume fraction of porosity was measured in all cases by image segmentation. Lamellae suitable for transmission electron microscopy (TEM) were extracted from regions of interest using a focused ion beam microscope (FIB, FEI Helios). Bright field and annular dark field TEM imaging was performed using FEI Tecnai G2 Sphera and Titan microscopes. Compositional analysis was performed by energy-dispersive X-ray spectroscopy (EDS) in both the SEM and TEM.

3. Results

3.1. Characteristics of the pristine materials

The starting powder used in the production of the yttrium silicate coating and the pellets was single-phase Y₂Si₂O₇ (Powder) based on XRD analysis (Fig. 1(a)). Following a 24 h, 1400 °C heat treatment the sintered pellets were predominantly Y₂Si₂O₇ (P2_1/n), although a small fraction of Y₂SiO₅ (below the XRD detection limit) was observed in SEM micrographs of the pellet cross sections. The pellets are relatively dense; the majority of the ~4 vol% porosity

1 The compositions are designated by the first letter of the oxide cation and the subscripts denote the concentration in mole percent of single-cation formula unit, i.e. C=CaO, M=MgO, Fe=Fe₂O₃, A=Al₂O₃, S=SiO₂. (It is assumed that all Fe is present as Fe²⁺ [17]).
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