



Full length article

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

David L. Poerschke*, John H. Shaw, Nisha Verma, Frank W. Zok, Carlos G. Levi

Materials Department, University of California, Santa Barbara, CA 93106-5050, United States

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ABSTRACT

Reactions between molten calcium-magnesium-iron alumino-silicate (CMFAS) deposits and yttrium disilicate ($Y_2Si_2O_7$, 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 $Ca_2Y_8(SiO_4)_6O_2$, 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_2O , 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 generically 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

* Corresponding author. Present address: Department of Chemical Engineering and Materials Science, University of Minnesota, United States.

E-mail address: poerschke@umn.edu (D.L. Poerschke).

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 ($Y_2Si_2O_7$, 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 (Y_2SiO_5 , YMS) [13] as well as ytterbium mono- (Yb_2SiO_5 , YbMS) and di-silicate ($Yb_2Si_2O_7$, YbDS) [14,16]. Additionally, whereas the past studies examined either dense pellets [13] or APS coatings [14], the present comparison between coatings and pellets facilitates understanding how well pellet-based experiments capture the behavior of the coating system. The use of pellets rather than coatings would simplify future efforts to more broadly assess the sensitivity of the reaction process to changes in temperature and deposit composition. The experimental analysis is complemented with thermodynamic calculations modeling the evolving equilibrium between the silicate melt and YDS as recession occurs. These calculations are useful for further accelerating the process of screening candidate EBC materials under a range of conditions. The results also provide insight into the relative behavior of mono- and di-silicate EBCs.

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 SiO_2 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 $C_{31}M_9F_5A_{12}S_{43}$ ¹ 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 $C_{33}M_9A_{13}S_{45}$ 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^{-5} 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 10 °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 δ - $Y_2Si_2O_7$ (*Pnam*) based on XRD analysis (Fig. 1(a)). Following a 24 h, 1400 °C heat treatment the sintered pellets were predominantly γ - $Y_2Si_2O_7$ (*P2₁/n*), although a small fraction of Y_2SiO_5 (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=FeO_{1.5}, A=AlO_{1.5}, S=SiO₂ (It is assumed that all Fe is present as Fe³⁺ [17].)

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