

Editorial

Special Issue: Environmental Barrier Coatings

Kang N. Lee

NASA Glenn Research Center

Cleveland, OH 44135 USA

Abstract: The global increase in air travel will require commercial vehicles to be more efficient than ever before. Advanced turbine hot section materials are a key technology required to keep fuel consumption and emission to a minimum. Ceramic matrix composites (CMCs) are the most promising material to revolutionize turbine hot section materials because of their excellent high-temperature properties. Rapid surface recession due to volatilization by water vapor is the Achilles heel of CMCs. Environmental barrier coatings (EBCs), which protect CMCs from water vapor, is an enabling technology for CMCs. The first CMC component entered into service in 2016 in a commercial engine, and more CMC components are scheduled to follow within the next few years. One of the most difficult challenges to CMC components is EBC durability because failure of EBC leads to a rapid reduction in CMC component life. Novel EBC chemistries, creative EBC designs, and robust processes are required to meet EBC durability challenges. Engine-relevant testing, characterization, and lifing methods need to be developed to improve EBC reliability. The aim of this Special Issue is to present recent advances in EBC technology to address current EBC challenges.

Keywords: EBC, CMC, Oxidation, Volatility, CMAS, Thermomechanical, Modeling

1. Introduction

Ceramic matrix composites (CMCs) are considered a game changer for gas turbine hot section materials because of their excellent high-temperature mechanical properties, oxidation resistance, and light weight – one-third of nickel-based superalloys [1]. The introduction of CMCs enables a fuel burn reduction up to two percent – few other technology in today’s pipeline have this much capability for fuel burn reduction [2]. Additionally, the light weight enables over 50 percent reduction in the turbine component weight [2].

SiC/SiC CMCs derive their oxidation resistance from dense slow-growing silica (SiO_2) scale [3]. In combustion environments, however, the protective silica scale volatilizes by reacting with water vapor (H_2O), leading to rapid CMC surface recession [4-9]. Alumina (Al_2O_3) in

oxide/oxide CMCs, although not as severe as silica, also suffers surface recession due to water vapor-induced volatilization [10]. External barrier coatings, known as environmental barrier coatings (EBCs), have been developed to protect CMCs from surface recession [11,12]. Over two decades of intense research and development led to the first EBC-coated CMC component - a high-pressure turbine CMC shroud - to enter service in LEAP engine by CFM International (Cincinnati, Ohio) [2]. More CMC components, including combustor liners and high-pressure turbine vanes, are scheduled to follow in the near future [2].

One of the most difficult challenges to CMC components is EBC durability because failure of EBC leads to a rapid reduction in CMC component life. Key contributors to EBC failure include recession, oxidation, degradation by calcium-aluminum-magnesium silicates (CMAS) deposits, thermal and thermo-mechanical strains, particle erosion, and foreign object damage (FOD) [12,13]. Novel EBC chemistries, creative EBC designs, and robust processes as well as engine-relevant testing, characterization, and lifing methods are required to meet EBC durability and reliability challenges.

2. This Special Issue

The nine research articles in this special issue present recent advances in EBC technology in the following areas: Recession (Ref [14]); EBC chemistry & processes (Refs [15, 16]); Oxidation & recession (Ref [17]); Oxygen permeability & EBC design (Ref [18]); CMAS thermal & mechanical properties (Ref [19]); CMAS infiltration modeling (Ref [20]); Mechanical behavior in thermal shock (Ref [21]); Thermomechanical reliability: modeling and validation (Ref [22]).

Smialek et al. [14] investigated the volatility of transient TiO_2 and steady-state Al_2O_3 scales formed on Ti_2AlC MAX phase ceramic in 1300°C high velocity (Mach 0.3, 100 m/s) and high pressure (6 atm, 25 m/s) burner rig tests (BRT). Unlike metals, the ceramic was stable at 1300°C . Unlike SiC and Si_3N_4 , neither burner test produced a weight loss, unless pre-oxidized. Volatility of TiO_2 was indicated by removal of Ti-rich oxide grains after BRT, while Al_2O_3 loss, not as pronounced as TiO_2 , was indicated by grain boundary etching and porosity. Al_2O_3 net weight loss was observed only on pre-oxidized samples, whose linear loss rate was about one-fifth of SiO_2 . 7YSZ TBC on Ti_2AlC survived for 500 h in the Mach 0.3 burner test with no indication of volatility or spalling.

Vaßen et al. [15] investigated various oxides as an EBC for both oxide/oxide and SiC/SiC CMCs. The goal was to obtain dense and crystalline coatings. APS (atmospheric plasma spraying) Y_2O_3 , $\text{Gd}_2\text{Zr}_2\text{O}_7$, $\text{Y}_3\text{Al}_5\text{O}_{12}$ and $\text{Yb}_2\text{Si}_2\text{O}_7$ were investigated for oxide/oxide CMCs, while Y_2SiO_5 and $\text{Yb}_2\text{Si}_2\text{O}_7$ were investigated for SiC/SiC CMCs. Y_2O_3 and $\text{Gd}_2\text{Zr}_2\text{O}_7$ showed promising results for oxide/oxide CMCs: Y_2O_3 coating showed excellent adhesion due to the formation of chemical bond, while $\text{Gd}_2\text{Zr}_2\text{O}_7$ coating required laser structuring of the CMC surface for good adhesion. APS Y_2SiO_5 required a heat treatment to obtain a desirable microstructure for SiC/SiC CMCs. $\text{Yb}_2\text{Si}_2\text{O}_7$ on SiC/SiC CMCs was investigated using various thermal spray process (APS, HVOF (high velocity oxygen fuel), VLPPS (very low pressure plasma spraying), SPS (suspension plasma spraying)). APS coating was dense, but with a high

degree of amorphous phase content. HVOF gave higher crystalline phase content than APS, but with some porosity. VLPPS coating gave the highest crystallinity and density. SPS gave a high degree of crystallinity, however segmental cracks could not be avoided. The study demonstrated the feasibility of various thermal spray methods besides APS for EBC processing.

Gatzen et al. [16] investigated $YAlO_3$ as an EBC for Al_2O_3/Al_2O_3 CMCs using APS and VLPPS processes. APS resulted in a poor quality coating not suitable as an EBC. VLPPS, on the other hand, resulted in a coating with high crystallinity, high purity, and strong adhesion. The strong adhesion was attributed to the formation of chemical bonding due to the formation of $Y_3Al_5O_{12}$ interfacial phase. The coating exhibited excellent thermal cyclic durability and CMAS resistance in laboratory testing, demonstrating its promise as an EBC for oxide/oxide CMCs.

Klemm et al. [17] investigated the oxidation and corrosion (water vapor-induced SiO_2 volatilization) of two plasma-sprayed EBC systems (Al_2O_3/YAG and $Si/Yb_2Si_2O_7+SiC/Yb_2SiO_5$) on SiC/SiC CMCs. Two EBC failure mechanisms were observed: spallation due to TGO-induced stresses and corrosion-induced gap at the TGO/EBC interface. Al_2O_3/YAG EBC, after burner rig test, suffered corrosion at the TGO/EBC interface and inside the Al_2O_3 bond coat. In $Si/Yb_2Si_2O_7+SiC/Yb_2SiO_5$, TGO-induced failure was delayed because SiC particles in the intermediate layer oxidized first, forming a shell surrounding SiC , and thereby served as a getter to reduce the permeation of oxidants (O_2 , H_2O) through EBC. In burner rig test, small pores developed in the intermediate layer due to SiO_2 corrosion. The beneficial gettering function of SiC particles is temporary, disappearing when the SiC particles are fully consumed.

Kitaoka et al. [18] proposed an EBC design based on oxygen permeability calculations to improve oxygen shielding capability and phase stability of mullite coating in $Si/mullite$ EBC for SiC/SiC CMCs. Grain boundary (GB) diffusion of O ions in mullite, from high PO_2 mullite surface to low PO_2 mullite/Si interface, and simultaneous GB diffusion of Al ions in the opposite direction were calculated. Calculations projected: (i) Mullite would decompose at the mullite/Si interface due to the outward Al diffusion; (ii) Replacing Si bond coat with β' - $SiAlON$ bond coat would increase oxygen shielding capability of mullite – this is because the higher equilibrium PO_2 for the oxidation of β' - $SiAlON$ to form SiO_2 compared to the oxidation of Si to form SiO_2 suppresses the inward O ion diffusion. Experiments confirmed the mullite decomposition at the mullite/Si interface and the improvement of mullite stability when Si bond coat was replaced by β' - $SiAlON$. The latter is because β' - $SiAlON$ serves as a source for Al.

Webster et al. [19] investigated the intrinsic thermal and mechanical properties of a volcanic ash glass obtained from the Eyjafjallajökull eruption of 2010. The properties studied include crystallization, melting temperature, CTE (coefficient of thermal expansion), modulus, hardness, and viscosity. The glass had a low propensity to crystallize in bulk form compared to synthetic sand glasses. Compared to sand glasses, the melting temperature and viscosity were higher, while the CTE and modulus were lower. The higher viscosity was attributed partly to a higher SiO_2 and lower CaO content in the volcanic ash. Hardness was similar, but the indentation fracture toughness was about twice that of sand glasses, which was attributed to the presence of Fe_3O_4 crystallites in the volcanic ash. Understanding the variation of CMAS properties with composition can provide insight into CMAS/coating interactions, which can be highly valuable for the development of CMAS-resistant coatings.

Kabir et al. [20] developed a numerical approach for studying the infiltration kinetics of molten CMAS in EB-PVD TBCs. Detailed analyses of the infiltration kinetics were performed, and the results were verified by experimental infiltration depths of the feathery and coarse microstructures. The study identified key morphological features that are important for infiltration. The rate of longitudinal and lateral infiltration could be minimized by reducing the gap between columns and/or increasing the length of the feather arms. Long feather arms having a lower lateral inclination decreased the infiltration rate and, therefore, reduced the infiltration depth. A key takeaway is that the infiltration kinetics can be altered by careful tailoring of coating microstructure such as feather arm lengths and inter-columnar gaps.

Seo et al. [21] investigated crack formation, crack stability, and crack healing of Si/mullite, Si/(mullite + Yb_2SiO_5), and Si/ Y_2SiO_5 EBCs on SiC/SiC CMCs under thermal shock cycling between room temperature and 1350°C in air. Mechanical behavior was determined before and after test via indentation tests. EBCs developed mud cracks on the surface and unidirectional vertical cracks in the top coat after several thermal shock cycles. Mullite-based EBCs exhibited crack healing phenomena after 4000-5000 cycles, where cracks were covered and partially filled with a new phase. Post-test XRD showed only crystalline Al_2O_3 in addition to the EBC component phases, indicating the new phase is likely amorphous. The nature of the new phase responsible for cracking healing was not fully understood yet. Si/ Y_2SiO_5 EBC delaminated after 4000 cycles with no crack healing phenomena. After 5000 cycles, mullite-based EBCs showed no significant changes in modulus and hardness due to cracking healing, whereas Y_2SiO_5 EBC showed a change from elastic to plastic behavior due to delamination.

Kawai et al. [22] investigated the thermomechanical stability of an as-processed EBC by comparing the ERR (energy release rate) for interfacial cracks calculated by FEM analysis and the interfacial fracture toughness determined experimentally. The EBC studied was (SiC/SiAlON/mullite/Yb disilicate-Yb monosilicate graded layer/Yb monosilicate with porous segmented structure). FEM showed thinner SiALON/thicker mullite combinations lower the initial ERR. Fracture in interface fracture test occurred at the SiC/SiALON and SiALON/mullite interfaces, indicating these interfaces are the weak link. ERR by FEM was sufficiently higher than the experimentally determined interfacial fracture toughness, indicating that as-processed EBC possesses sufficient thermomechanical reliability. As-processed EBC with $5\ \mu\text{m}$ to $25\ \mu\text{m}$ SiALON and mullite showed no interfacial delamination, confirming the thermomechanical reliability. This study considered only as-processed stresses. Further work is needed to assess the thermomechanical reliability in service environments by incorporating oxidation, CMAS, thermal cycling in temperature gradient, and mechanical stresses.

References

1. Anson D, Richerson DW. Progress in Ceramic Gas Turbine Development Vol. 2. Edited by Roode M van, Ferber M, Richerson DW. ASME PRESS, New York, NY. 2003: 1-10.
2. Steibel J. Ceramic matrix composites taking flight at GE Aviation. American Ceramic Society Bulletin. 2019; 98 (3): 30-33.
3. Jacobson NS. Corrosion of silicon-based ceramics in combustion environments. J. Am. Ceram. Soc. 1993; 76(1): 3-28.
4. Opila EJ, Hann R. Paralineer oxidation of CVD SiC in water vapor. J. Am. Ceram. Soc. 1997; 80(1): 197-205.
5. Smialek JL, Robinson RC, Opila EJ, Fox DS, Jacobson NS. SiC and Si₃N₄ scale volatility under combustor conditions. Adv. Composite Mater. 1999; 8(1): 33-45.
6. Robinson RC, Smialek JL. SiC recession caused by SiO₂ scale volatility under combustion conditions: I. Experimental results and empirical model. J. Am. Ceram. Soc. 1999; 82(7): 1817-25.
7. Opila EJ, Smialek JL, Robinson RC, Fox DJ, Jacobson NS. SiC recession caused by SiO₂ scale volatility under combustion conditions: II. Thermodynamics and gaseous-diffusion model. J. Am. Ceram. Soc. 1999; 82(7): 1826-1834.
8. More KL, Tortorelli PF, Ferber MK, Walker LR, Kiser JR, Miriyala N, et al. ASME paper 99-GT-292, ASME TURBOEXPO 1999. Indianapolis, IN. 1999 (June).
9. Ferber MK, Lin HT, Parthasarathy V, Brentnall WD. ASME paper 99-GT-265, ASME TURBOEXPO 1999. Indianapolis, IN. 1999 (June).
10. Opila EJ, Myers, D.L. Alumina volatility in water vapor at elevated temperatures. J. Am. Ceram. Soc. 2004; 87: 1701-1705.
11. Lee KN, Fritze H, Ogura Y. Progress in Ceramic Gas Turbine Development Vol. 2. Edited by Roode M van, Ferber M, Richerson DW. ASME PRESS, New York, NY. 2003: 641-664.
12. Lee KN. Environmental barrier coatings for CMCs; in Ceramic Matrix Composites. Edited by Bansal NP, Lamon J. Wiley, New York, NY. 2015: 430-451.
13. Lee KN. Yb₂Si₂O₇ environmental barrier coatings with reduced bond coat oxidation rates via chemical modifications for long life. J. Am. Ceram. Soc. 2019; 102 (3): 1507-1521.

14. Smialek JL. Relative Ti_2AlC scale volatility under 1300°C combustion conditions. [EBC special issue]
15. Vaßen R, Bakan E, Gatzel C, Kim S, Mack DE, Guillon O. Environmental barrier coatings made by different thermal spray technologies. [EBC special issue]
16. Gatzel C, Mack DE, Guillon O, Vaßen R. $YAlO_3$ - A novel environmental barrier coating for Al_2O_3/Al_2O_3 -ceramic matrix composites. [EBC special issue]
17. Klemm H, Schönfeld K, Kunz W. Delayed formation of thermally grown oxide in environmental barrier coatings for non-oxide ceramic matrix composites. [EBC special issue]
18. Kitaoka S, Matsudaira T, Kawashima N, Yokoe D, Kato T, Takata M. Structural stabilization of mullite films exposed to oxygen potential gradients at high temperatures. [EBC special issue]
19. Webster RU, Bansal NP, Salem JA, Opila EJ, Wiesner VL. Characterization of thermochemical and thermomechanical properties of Eyjafjallajökull volcanic ash glass. [EBC special issue]
20. Kabir MR, Sirigiri AK, Naraparaju R, Schulz U. Flow kinetics of molten silicates through thermal barrier coating: A numerical study. [EBC special issue]
21. Seo H-I, Kim D, Lee KS. Crack healing in mullite-based EBC during thermal shock cycle. 1. [EBC special issue]
22. Kawai E, Kakisawa H, Kubo A, Yamaguchi N, Yokoi T, Akatsu T, Kitaoka S, Umeno Y. Crack initiation criteria in EBC under thermal stress. [EBC special issue]