

Upper Temperature Limit of Environmental Barrier Coatings for Enabling Propulsion Materials Established

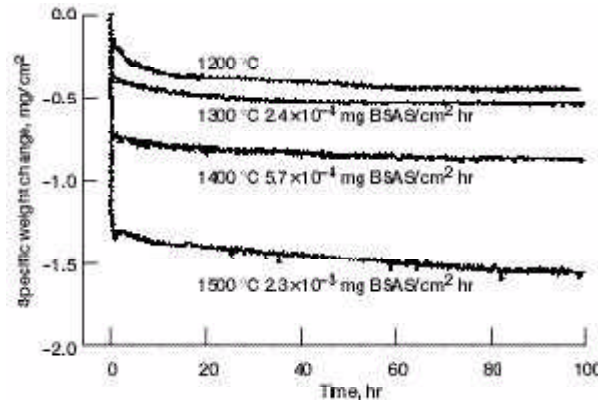
Silicon-based ceramics, such as SiC/SiC composites and Si_3N_4 , are the prime candidates for hot section structural components of next-generation gas turbines. A key barrier to such an application is the rapid recession of silicon-based ceramics in combustion environments because of the volatilization of silica scale by water vapor (refs. 1 and 2). Environmental barrier coatings (EBC's) were developed to prevent recession in the High Speed Research--Enabling Propulsion Materials (HSR-EPM) Program (refs. 3 and 4). An investigation under the Ultra-Efficient Engine Technology Program was undertaken at the NASA Glenn Research Center to establish the upper temperature limit of the EPM EBC.

The EPM EBC consists of three layers: a silicon bond coat, a mullite ($3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$) or mullite-based bond coat, and a barium-strontium-aluminum silicate (BSAS: $\text{BaO}_x \cdot \text{SrO}_{1-x} \cdot \text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$) topcoat. Volatility and environmental/chemical stability of the EBC in water vapor were the key criteria in establishing the upper temperature limit in this study.

TABLE I.--RECESSION OF BSAS
[1000 hr, 6 atm; gas velocity, 25 m/sec; $p_{\text{H}_2\text{O}}$, 0.6 atm]

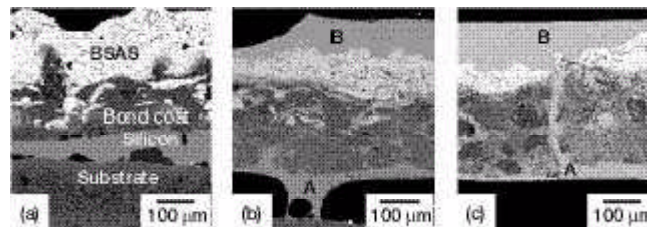
Temperature, °C	1300	1400	1500
Recession, μm	7.5	17.8	72

The volatility was investigated by exposing monolithic hot-pressed BSAS coupons in 50 vol % H_2O /balance O_2 at 1200 to 1500 °C while continuously monitoring the weight change using a microbalance. The following graph plots weight change versus time for hot-pressed BSAS in water vapor. The rapid initial weight loss is an artifact of the experimental procedure. A linear weight loss was observed, with the rate increasing with temperature. In the table, the weight loss rate is converted to a recession rate in a simulated combustion environment (pressure, 6 atm; gas velocity, 25 m/sec; $p_{\text{H}_2\text{O}}$, 0.6 atm). The recession rate of BSAS determined in a high-pressure burner rig at 1300 °C agreed with the converted recession rate in the table, within a factor of two, supporting the validity of the conversion. The high recession rate at $T > 1400$ °C can become an issue, especially for thin EBC's (<100 μm).



Weight change versus time for hot-pressed BSAS in 50 vol % H₂O/balance O₂.

For our environmental/chemical stability study, EBC-coated silicon-based ceramic coupons were exposed to 90 vol % H₂O/balance O₂ at 1300 to 1480 °C under thermal cycling. Each cycle consists of 1 hr at high temperature and 20 min at room temperature. The exposed coupons were subsequently prepared metallographically and the environmental/chemical degradation was characterized using scanning electron microscopy and energy dispersive spectroscopy. The following images show the cross sections of EBC-coated silicon-based ceramics after 100 hr at 1400, 1440, and 1480 °C, respectively. Silicon was not applied on coupons exposed to 1440 and 1480 °C to avoid melting it (melt temperature, 1410 °C). The EBC cross section at 1400 °C remained similar to the as-processed state, except for some limited oxidation at the silicon/mullite-based bond coat interface. In contrast, a severe reaction developed at 1440 and 1480 °C: a thick scale formed at the silicon/mullite-based bond coat interface (area A in parts (b) and (c)), a new layer formed on top of the BSAS (area B), and an extensive reaction occurred between mullite and BSAS within the bond coat. X-ray diffraction indicates that the new top layer is a glass, which energy dispersive spectroscopy analysis shows has the same composition as the interface scale. According to the BaO-SiO₂-Al₂O₃ phase diagram, BSAS and silica form a low melting (1296 °C) silicate phase. It is therefore suggested that the glass phase is a reaction product between the silica (grown on SiC as a result of oxidation) and the BSAS. The glass moved to the EBC surface, presumably due to capillary forces or the low surface energy of the glass phase. The significant thinning of the BSAS topcoat at $T > 1440$ °C and the channel that developed between the interface scale and the top glass layer (part (c)), supports this suggestion. The presence, and therefore the formation, of a low melting glass must be avoided because it leads to the spallation of the EBC.



Cross section of EBC-coated silicon-based ceramics after 100 hr in 90 vol % H₂O/balance O₂, showing the progress of the environmental/chemical reaction with

temperature. (a) 1400 °C. (b) 1440 °C. (c) 1480 °C.

Based on the foregoing results, one may conclude that the upper temperature limit of EPM EBC should be set at 1400 °C for a targeted 1000-hr life.

References

1. Robinson, R.C.; and Smialek, J.L.: SiC Recession Caused by SiO₂ Scale Volatility Under Combustion Conditions: I, Experimental Results and Empirical Model. J. Am. Ceram. Soc., vol. 82, no. 7, 1999, pp. 1817-1825.
2. Opila, E.J., et al.: SiC Recession Caused by SiO₂ Scale Volatility Under Combustion Conditions. Part II Thermodynamics and Gaseous Diffusion Model. J. Am. Ceram. Soc., vol. 82, no. 7, 1999, pp. 1826-1834.
3. Lee, K.N.: Current Status of Environmental Barrier Coatings for Si-Based Ceramics. Surface and Coatings Technology, vol. 133-134, nos. 1-7, 2000.
4. Lee, K.: Effect of Impurities and Coating Design on the Durability of EBC for Silicon-Based Ceramics. Proceedings of the Per Kofstad Memorial Symposium on High Temperature Corrosion and Materials Chemistry, Michael McNallan, ed., Electrochemical Society, Pennington, NJ, 2000, pp. 417-428.

Cleveland State University contact: Dr. Kang N. Lee, 216-433-5634,
Kang.N.Lee@grc.nasa.gov

Glenn contact: Dr. Dennis S. Fox, 216-433-3295, Dennis.S.Fox@grc.nasa.gov

Dynacs Engineering Corporation, Inc., contact: R. Craig Robinson, 216-433-5547,
Raymond.C.Robinson@grc.nasa.gov

Authors: Dr. Kang N. Lee, Dr. Dennis S. Fox, and R. Craig Robinson

Headquarters program office: OAT

Programs/Projects: UEET