Investigation of BSAS Environmental Barrier Coatings for Silicon Carbide Turbine Blades

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Rolls-Royce would like to increase the operating temperature of its gas turbine engines by replacing the current nickel-based superalloy blades with Ceramic Matrix Composite (CMC) blades. The blades allow for a greater amount of power per fuel consumption and are lighter weight. However, due to the tendency of silicon carbide (SiC) to oxidize at higher temperatures, an Environmental Barrier Coating (EBC) is needed to protect it from the humid operating environment. This project investigated several EBC slurry coatings for SiC substrates. The slurries had different quantities and types of sintering aid. The liquid-based coating was intended as an alternative to spray coatings, which have difficulty achieving an even coating on parts of complex geometries. The most suitable slurry was found to be the 0.5 vol% MgO polysilazane slurry and was sintered for 6 hours at 1200°C.

This project was sponsored by Rolls-Royce North America in Indianapolis, Indiana.

Project Background

- Ceramic Matrix Composites (CMCs), are currently being considered as replacement materials for the turbine blades in gas turbine engines currently made from nickel-based superalloys.
- Silicon carbide is the current favorite for its low density and coefficient of thermal expansion (CTE).
- SiC has a disadvantage as it oxidizes in high humidity environments through the following reaction[1]

$$SiC(s) + 3\,H_2O(g) = SiO_2(s) + CO(g) + 3\,H_2(g)$$
$$SiO_2(s) + 2\,H_2O(g) = Si(OH)_4(g)$$

- The SiC can be protected from the environment through the use of an environmental barrier coating (EBC). For our purposes, it was decided the coating should be Barium Strontium Aluminosilicate (BSAS), due to its similar CTE to SiC.
- Design limitations: the total production time should be less than 48 hours and the coating should be non-line of sight. This means complex parts can be coated using a method other than the spray method currently used. Dip-based ceramic slurries were used instead.

[1] Opila, Elizabeth. ECS Transactions, Vol. 3 Issue 14 (2007).

Experimental Procedure

Slurry Production

1. The BSAS and sintering aid powders were attrition milled to sub-micron size in an ethanol solution for 4 hours.
2. The ethanol was evaporated and the powder sifted using a mortar and pestle and sieve.
3. The desired amounts of material would be added in the following order: BSAS, PVB or polysilazane, ethanol, the sintering aid, and Darvan C. The total mass of the mixture was 100g.

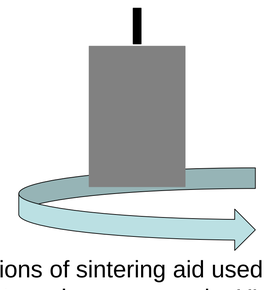
Substrate Preparation

- The SiC substrates were cut, cleaned in acetone, and had their edges ground to remove sharp edges where thermal stresses could be concentrated.

Dipping Process

- The substrates were dipped 3 times, each time having the excess slurry spun off and left to dry using the method depicted below.

(Right) the method used to give each substrate multiple coatings.



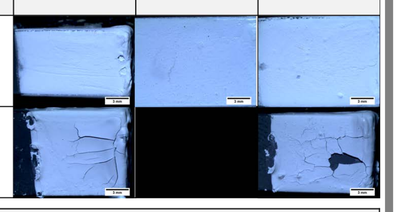
(Below) The different concentrations of sintering aid used for each slurry tested at 1300°C. O's indicate a slurry was made, X's were not.

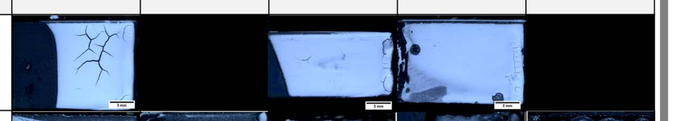
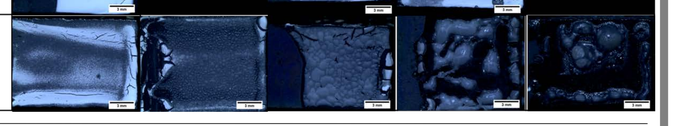
Sintering Aid and dispersant present in BSAS slurry	Volume % Sintering Aid					Weight % Sintering Aid		
	0.25	0.5	1	5	10	1	5	10
MoO_3 + PVB	X	X	O	O	O	O	O	O
YSZ + PVB	X	X	O	X	O	O	O	O
MgO + PVB	O	O	O	O	O	X	X	X
MoO_3 + polysilazane	O	X	O	O	X	X	X	X
YSZ + polysilazane	X	X	X	X	X	X	X	X
MgO + polysilazane	O	O	O	O	O	X	X	X

Resulting Coatings

- Below are thermal maps of the tested slurries. Four different series (A-D) were run, but only 2 (C & D) resulted in acceptable coatings.
- Empty cells in the test matrix were left untested due to lack of time and materials, requiring nearly 3 weeks for some materials to be received.
- Three different sintering aids and 2 dispersants were compared.

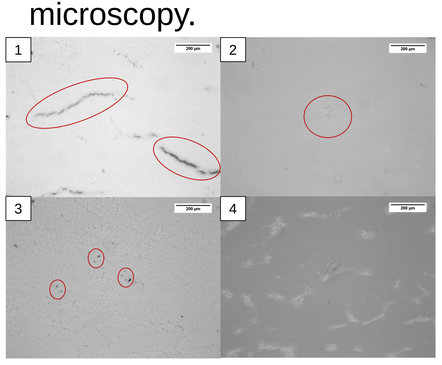
The 3 sintering aids are molybdenum trioxide, yttrium stabilized zirconia (YSZ), and magnesium oxide. The dispersants are polyvinyl butyral (PVB) and polysilazane. In series A and B, only MoO_3 and YSZ were tested with PVB.

Series C	Coatings by Volume Percent Sintering Aid (PVB, 1300°C)		
	1 vol%	5 vol%	10 vol%
MoO_3			
YSZ			

Series D	Coatings by Volume Percent Sintering Aid (polysilazane, 1300°C)				
	0.25 vol%	0.5 vol%	1 vol%	5 vol%	10 vol%
MoO_3					
MgO					

Surface Flaw Analysis

- In a method used by last year's team, 4 different types of flaws were searched for under optical microscopy.



Possible Flaws:
1) Through thickness cracks
2) Entrapped air pockets
3) Pinholes (~10 μm)
4) Mud Cracking

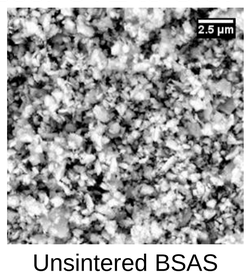
- All usable coatings were in the range of 100-200 μm.

The table below lists the defects (1-4) in each coating sintered at 1300°C. Any containing through thickness cracks automatically fail (-). X's were not tested.

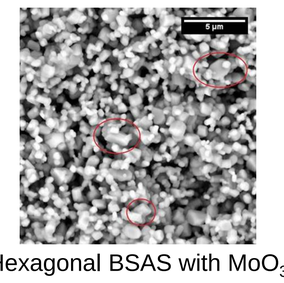
Sintering Aid and dispersant present in BSAS slurry	Volume % Sintering Aid					Weight % Sintering Aid		
	0.25	0.5	1	5	10	1	5	10
MoO_3 + PVB	X	X	2,3,4	2,3,4	-	-	-	-
YSZ + PVB	X	X	-	X	-	-	-	-
MgO + PVB	-	-	-	-	-	X	X	X
MoO_3 + polysilazane	-	X	3,4	3,4	X	X	X	X
YSZ + polysilazane	X	X	X	X	X	X	X	X
MgO + polysilazane	-	-	-	-	-	X	X	X

Phase Changes of BSAS

- Research[2] indicates that upon sintering at elevated temperature, MoO_3 may cause the BSAS to transition from monoclinic to hexagonal, which has a closer CTE to SiC.



Unsintered BSAS

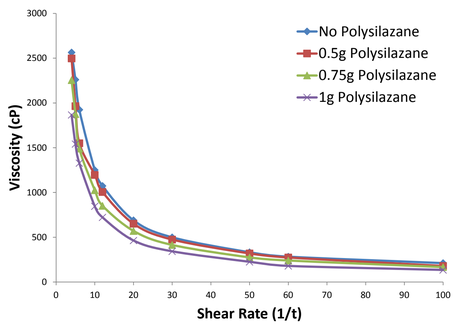


Hexagonal BSAS with MoO_3

[2] Lee, Kang; Et al. "Key Durability Issues With Mullite-Based Environmental Barrier Coatings for Si-Based Ceramics": ASME-10.1115/1.1287584. May, 2000.

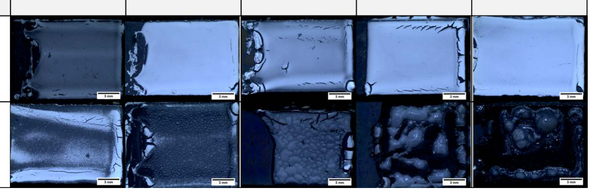
Slurry Viscosity

- Viscosity testing on a series D slurry was conducted to show the relationship between Polysilazane and viscosity.
- Viscosity of the slurry is inversely proportional to the amount of polysilazane added to the slurry.
- Increased slurry viscosity increases coating thickness and cracking.

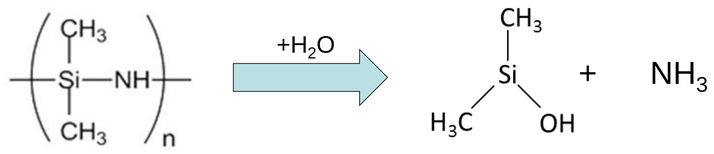


Possible Failure Mechanisms

- MgO with polysilazane sintered at 1200°C but at 1300°C the coating melted and formed a glassy phase.

Sintering Temperature	MgO Coatings by Volume Percent				
	0.25 vol%	0.5 vol%	1 vol%	5 vol%	10 vol%
1200°C					
1300°C					

- Polysilazane reacts with water to form ammonia and silicic acid which becomes silica. Even 200 proof ethanol may contain up to 0.5% water.

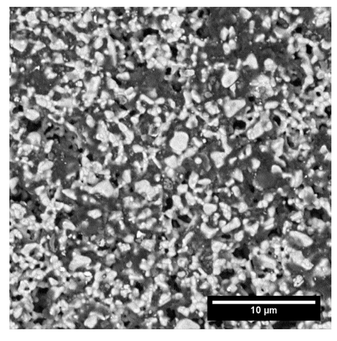


- The largest concern is the formation of an inter-particle glassy phase, which has a significantly different CTE than either the SiC or BSAS alone[3].

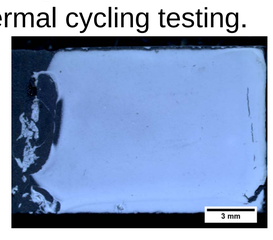
[3] Lee, Kang. (2003). "Upper Temperature Limit of Environmental Barrier Coatings Based on Mullite and BSAS". Cleveland, Ohio: J. Am. Ceram. Soc, 86 [8] 1299–306.

Recommendations

- The 0.5 vol% MgO polysilazane coating had only pinhole flaws under cross-sectional analysis.
- It's exact formulation is 19.15 vol% BSAS, 0.1 vol% MgO, 80 vol% Ethanol, 0.2 vol5 Darvan-C, and 0.55 vol% Polysilazane and was sintered at 1200°C for 6 hrs.
- Anhydrous Tetrahydrofuran (THF) may be used as a solvent, having little to no water, future testing permitting.



- As suggested by the MgO comparison table, this coating may not withstand thermal cycling testing.



Future Work

- XRD analysis should be performed before and after sintering to quantify the formation of new phases.
- Thermal Testing of the MgO samples at engine operating temperatures.
- Environmental Testing of the sintered samples.