Integrated TBC/EBC for SiC Fiber Reinforced SiC Matrix Composites for Next Generation Gas Turbines

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Advanced Ceramic Research

Overall Goals

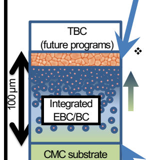
- Develop an integrated and graded EBC/BC that is:
  - Good bonding with CMC;
  - Graded compositions without sharp interfaces to mitigate thermal stresses from CTE mismatch;
  - Low oxygen transport rate, low oxidation rate and low volatility in high temperature, high velocity steam environment;
  - Tolerant to certain degree of oxidation thereby preventing catastrophic failure;
  - Chemically stable and compatible with CMC and TBC
- Create a strong collaborative team with complementary expertise and state-of-the-art facilities
  - The Clemson University team of Drs. Bordia and Peng.
  - The GE team, led by John Delvaux

Objectives

Objectives:

- Investigate the effect of composite stoichiometry (*i.e.* Si/B/C/N ratio in the precursor and the ratio of the Si-based precursor to yttrium oxide (Y_2O_3) (or ytterbium oxide (Yb_2O_3)) particle filler and processing conditions on the resultant phases and nanostructure of the composite ceramics.
- Investigate the effect of the composition and nanostructure on the thermal properties and oxidation and volatilization behavior in oxidizing and high velocity steam environments.
- Process the graded Y_2O_3 (or Yb_2O_3) particulate /silicon boron carbon nitride (SiBCN) matrix composite coating and investigate the phase and microstructure stability during high velocity steam exposure at temperatures up to 1500ºC.
- Develop a method to create Y_2O_3 (or Yb_2O_3) and SiBCN powders suitable for atmospheric plasma spraying (APS). The powders will be provided to GE Power for the fabrication of integrated environmental barrier coating/bond coating (EBC/BC) using APS.
- Evaluate the performance of integrated BC/EBCs from APS under high velocity steam environments at temperatures up to 1500ºC.

The concept of integrated TBC/EBC/BC



- Dense Y_2O_3 or Yb_2O_3 top layer ensures low oxygen diffusion and volatilization rates under high temperature high velocity steam environment.
- Graded composition allows smooth transition from SiC to SiBCNO with increasing amount of embedded Y_2O_3 particles.
  - Graded structure avoids sharp CTE mismatch
  - Tolerant to oxidation; further increase TBC lifetime
- PDC SiC bond coat/SiC substrate interface provides excellent adhesion

Research Progress

Task 1: Project management and planning

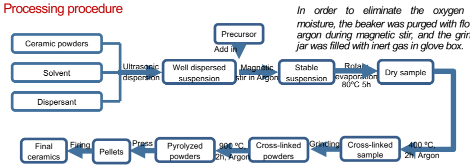
- Kickoff meeting 10/27/17
- A poster at the 2017 UTSR review meeting
- Recruitment of UG students and post doc to work on the project
- Coordination with GE Team including scheduling review meetings
- Regular review meetings with DoE Program Manager and GE team
- A talk and a poster at the 2018 UTSR review meeting

Task 2: Processing and stability of Y_2O_3-Si-B-C-N and Yb_2O_3-Si-B-C-N composites

Raw materials	Specifications	Supplier	Function in the system
Y_2O_3	Purity: 99.9%; APS: 0.5~1 μm	American Elements (USA)	Ceramic filler
Yb_2O_3	Purity: 99.9%; APS: 3~5 μm	American Elements (USA)	Ceramic filler
Durazane 1800	Polysilazane	Merck KGaA (Germany)	Precursor for SiCN ceramic
SMP10	Polycarbosilane	Stafire (USA)	Precursor for SiC ceramic
Decaborane	Purity: 98%	Alfa Aesar (USA)	Boron source
Disperbyk-2070		BYK (Germany)	Dispersant
Dicumyl peroxide (DCP)	Purity: 99%	Acros Organics (USA)	Cross-link agent
Cyclohexane	Purity: 99%	Alfa Aesar (USA)	Solvent for SMP10
Di-n-butyl ether (DNB)	Purity: 99%	Acros Organics (USA)	Solvent for Durazane 1800

- Processing procedure

In order to eliminate the oxygen and moisture, the beaker was purged with flowing argon during magnetic stir, and the grinding jar was filled with inert gas in glove box.



- Processing protocol for SiCN/Y_2O_3 composite
- *Same protocol for SiCN/Yb_2O_3, SiC(B)/ Y_2O_3 and SiC(B)/ Yb_2O_3 composite*

Composition effect

Sample Id	Composition: Y_2O_3 (vol.%)	Composition: Durazane 1800 (vol.%)	Processing temperature (ºC)	Dwell time (h)	Processing atmosphere	Processing method
D10:90-1500	10	90	1500	2	Argon	Pressureless
D30:70-1500	30	70	1500	2	Argon	Pressureless
D50:50-1500	50	50	1500	2	Argon	Pressureless
D70:30-1500	70	30	1500	2	Argon	Pressureless
Dd90:10-1500	90	10	1500	2	Argon	Pressureless

Processing temperature effect

Sample Id	Composition: Y_2O_3 (vol.%)	Composition: Durazane 1800 (vol.%)	Processing temperature (ºC)	Dwell time (h)	Processing atmosphere	Processing method
D30:70-1300	30	70	1300	2	Argon	Pressureless
D30:70-1400	30	70	1400	2	Argon	Pressureless
D30:70-1500	30	70	1500	2	Argon	Pressureless
D30:70-1600	30	70	1600	2	Argon	Pressureless
D30:70-1650	30	70	1650	2	Argon	Pressureless

Processing atmosphere effect

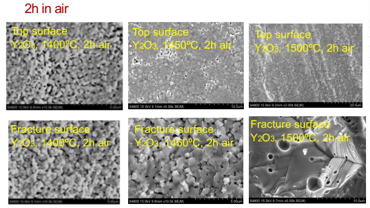
Sample Id	Composition: Y_2O_3 (vol.%)	Composition: Durazane 1800 (vol.%)	Processing temperature (ºC)	Dwell time (h)	Processing atmosphere	Processing method
D30:70-1500-a	10	90	1500	2	Air, Nitrogen	Pressureless
D30:70-1500-a	30	70	1500	2	Air, Nitrogen	Pressureless
D30:70-1500-a	50	50	1500	2	Air, Nitrogen	Pressureless
D30:70-1500-a	70	30	1500	2	Air, Nitrogen	Pressureless
D30:70-1500-a	90	10	1500	2	Air, Nitrogen	Pressureless

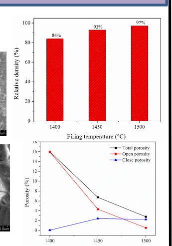
Processing method effect

Sample Id	Composition: Y_2O_3 (vol.%)	Composition: Durazane 1800 (vol.%)	Processing temperature (ºC)	Dwell time (h)	Processing atmosphere	Processing method
D10:90-1500-h	10	90	1500	2	Argon	Hot press
D30:70-1500-h	30	70	1500	2	Argon	Hot press
D50:50-1500-h	50	50	1500	2	Argon	Hot press
D70:30-1500-h	70	30	1500	2	Argon	Hot press
D90:10-1500-h	90	10	1500	2	Argon	Hot press

Sintering of Y_2O_3 top coat

- Y_2O_3 ceramic pellets were fired at 1400, 1450, 1500ºC for 2h in air





- Density increases and porosity decreases with increasing processing temperature
- At 1500ºC, the relative density reaches 97% with only 0.5% of open porosity
- Some closed pores in the inside of sample. Increase firing time may reduce more pores
- Y_2O_3 fabricated at 1500ºC can be used at top EBC coating

Composite Ceramic Powders

SiCN/Y_2O_3 composite

Sample Id	Composition: Y_2O_3 (vol.%)	Composition: Durazane 1800 (vol.%)	Synthesis temperature (ºC)	Grinding Time (h)	Average particle size (μm)
D10:90	10	90	900	36	0.84
D30:70	30	70	900	36	0.75
D50:50	50	50	900	36	0.84
D70:30	70	30	900	36	0.78
D90:10	90	10	900	36	0.57

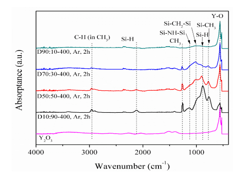
SiC(B)/Y_2O_3 composite

Sample Id	Composition: Y_2O_3 (vol.%)	Composition: Durazane 1800 (vol.%)	Synthesis temperature (ºC)	Grinding Time (h)	Average particle size (μm)
S10:90	10	90	900	60	0.60
S30:70	30	70	900	60	0.77
S50:50	50	50	900	60	0.36
S70:30	70	30	900	60	0.55
S90:10	90	10	900	60	0.53

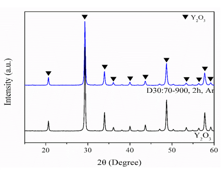
- All the powders can be ground to an average particle size smaller than 1 μm.
- For Y_2O_3/SiCN ceramic powders, all the powders have been ground for 60h. The particle size will be characterized.

Processing of Y_2O_3/SiCN Composite Ceramic

- Y_2O_3/SiCN ceramic pellets (900ºC) characterization-Phase composition



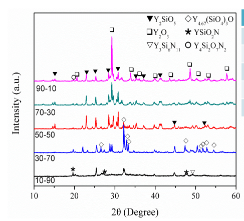
FTIR of cross-linked Y_2O_3/SiCN sample



XRD of Y_2O_3 and sample after pyrolysis at 900ºC

- For Y_2O_3-Durazane system, Si-H, Si-CH_3, Si-NH-Si, Si-CH_2-Si belongs to cross-linked Durazane, while Y-O belongs to Y_2O_3.
- For Y_2O_3-SMP10 system, Si-H, Si-CH_3, and Si-CH_2-Si belongs to the cross-linked SMP10, while Y-O belongs to Y_2O_3.
- After pyrolysis at 900ºC, only Y_2O_3 phases are shown in the XRD curves. This is because SiCN ceramics are amorphous after pyrolysis

- Y_2O_3/SiCN ceramic pellets (1400ºC) characterization-Phase composition

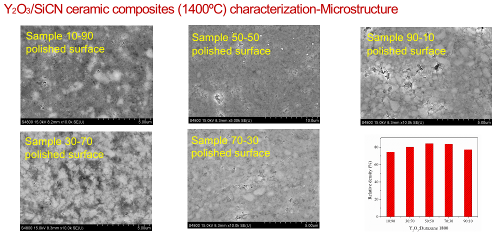


Sample	Phase compositions
10-90	$YSiO_2N$, Y_2SiO_5, $Y_3Si_6N_{11}$, $Y_{4.67}(SiO_4)_3O$
30-70	$Y_{4.67}(SiO_4)_3O$, Y_2SiO_5 Y_2O_3
50-50	$Y_{4.67}(SiO_4)_3O$, Y_2SiO_5 Y_2O_3
70-30	Y_2O_3, Y_2SiO_5,
90-10	Y_2O_3, Y_2SiO_5

$$Y_2O_3(s) + SiO_2(s) = Y_2SiO_5(s)$$
$$Y_2O_3(s) + Si_2N_2O(s) = 2YSiO_2N\ (s)$$
$$4Y_2O_3(s) + Si_3N_4(s) + SiO_2(s) = 2Y_4Si_2O_7N_2$$
$$2.335Y_2O_3(s) + 3SiO_2(s) = Y_{4.67}(SiO_4)_3O(s)$$
$$2/3Y_{4.67}(SiO_4)_3O(s) + 4/9Y_2O_3 = 2Y_2SiO_5$$

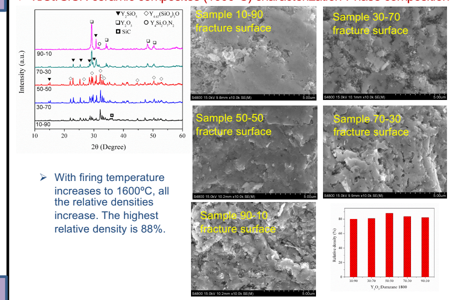
- No Si_3N_4 or SiC phases are detected in the samples, mainly because the amorphous SiCN are stable up to 1500 ºC

- Y_2O_3/SiCN ceramic composites (1400ºC) characterization-Microstructure



- The green densities, bulk densities and skeletal densities of samples show increasing trend with increasing Y_2O_3 volume. Density of Y_2O_3 (5.01 g/cm³) > Density of PDC-SiCN
- The highest relative density is 84%.

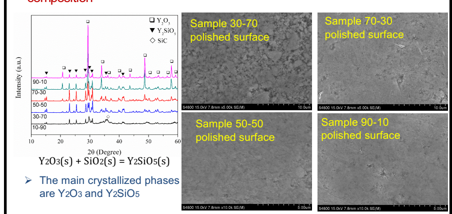
- Y_2O_3/SiCN ceramic composites (1600ºC) characterization-Phase composition



- With firing temperature increases to 1600ºC, all the relative densities increase. The highest relative density is 88%.

Processing of Y_2O_3/SiC(B) Composite Ceramic

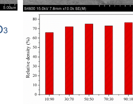
- Y_2O_3/SiC(B) ceramic composites (1500ºC) characterization-Phase composition



$$Y_2O_3(s) + SiO_2(s) = Y_2SiO_5(s)$$

- The main crystallized phases are Y_2O_3 and Y_2SiO_5

- The densities of samples increase with increasing Y_2O_3 volume.
- The highest relative density is 76%.



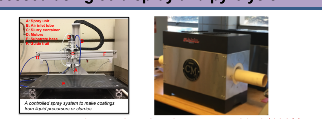
Summary of Task 2

- High density top coat processed
- Composite powders synthesized over a broad range of compositions
- Milling procedure optimized
- Effect of processing temperature on the density and microstructure in some systems
- The densities of the intermediate layers are in the acceptable range.

Ongoing work

- Complete the effect of processing temperature on the density and microstructure
- Effect of processing atmosphere

Task 4: Processing and performance of graded coatings processed using cold spray and pyrolysis



- The cold spray system to make coatings has been assembled (100% completion)
- Purchased the tube furnace for the investigation of oxidation response of the composites (100% completion)
- Designing and setting up the steam jet oxidation furnace (60% completion)

Acknolegement

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