Development and Evaluation of Environmental Barrier Coatings for Silicon Nitride

Tania Bhatia, Harry Eaton, John Holowczak, Ellen Sun, and Venkat Vedula

United Technologies Research Center East Hartford, Connecticut

Presented at DOE EBC Workshop in Nashville on November 18, 2003





Acknowledgements

Funding Support

DOE – Debbie Haught and Steve Waslo

ONR – Steve Fishman

NASA – Dave Brewer, Bob Draper, and Joe Shaw

Collaborations

Solar – Jeff Price, Josh Kimmel, Mark Van Roode, and Oscar Jimenez NDE – Bill Ellingson at ANL

Mechanical Testing – Matt Ferber and H.T. Lin at ORNL

Keiser Rig – Karren More and Peter Tortorelli at ORNL

Team Members

Gary Linsey, Xia Tang, Bill Tredway, Tom Lawton, Bob Barth, Mark Hermann, and Shanti Nair (U Mass)





Outline

- Highlights of EBC developed for silicon carbide (EBC_{SiC})
- Adapting EBC_{SiC} to silicon nitride: Important learnings for EBC_{SiN} development
- EBC for silicon nitride development
- Summary and future challenges





EBC for Silicon Carbide (EBC_{SiC})





Current state of the art EBC_{SiC}

- Development was initiated under NASA HSCT (High Speed Civil Transport) EPM (Enabling Propulsion Materials) program in mid 1990's for SiC/SiC CMC.
- Scaled-up and improved under DOE/Solar Turbines CSGT (Ceramic Stationary Gas Turbine) program since late 1990's.



Barium Strontium Aluminum Silicate (BSAS)

Intermediate layer (Mullite+BSAS)

Silicon bond coat





DOE EBC Workshop, Nashville, TN

BSAS-based EBC_{SiC} demonstrated for long-term 2200°F applications

- ~50,000 hours field tests in Solar Turbines Centaur 50S engines
 - Bakersfield, CA : 13,937 hours
 - Malden Mills II, MA : 15,144 hours



CMC well protected in areas where EBC remained intact



DOE EBC Workshop, Nashville, TN

November 18, 2003



UTRC burner rig demonstration: BSAS-based coating with mullite thermal barrier survived 100 thermal cycles





P.7

- Achieved 2700°F surface temperature with 300°F EBC thermal gradient
- EBC coated monolithic SiC survived 100 atmospheric burner rig thermal cycles





SAS as "new" steam barrier layer in EBC_{SiC}

- SAS believed to provide better EBC life than BSAS
- SAS-based EBC has been applied to Solar CMC liners
- Plasma spray parameters for SAS system optimized





DOE EBC Workshop, Nashville, TN

November 18, 2003



Applying EBC_{SiC} to Si_3N_4





EBC_{SiC} applied to cooled FT8 Si_3N_4 vanes and survived two trip shutdowns



- SN282 vanes uncoated
- AS800 vane coated with EBC_{SiC}
- 30 hours at 70% power (1230°C at coated vane), 2 trip shutdowns from ~60% power, 1 hour at 80% power (1260°C at coated vane)



Suction Side





Adapting EBC_{SiC} to silicon nitride: Effective protection in ORNL Keiser Rig



Uncoated surface after 1500-hr steam exposure at 1200°C





- Coating remained intact except through thickness cracks due to CTE mismatch
- No oxidation observed in the AS800 substrate underneath crack-free regions after 2000-hr steam exposure at 1200°C indicating effective barrier function

Results provided by Karren More, ORNL



DOE EBC Workshop, Nashville, TN

November 18, 2003



- EBC_{SiC} on Solar's CSGT Si₃N₄ nozzles: FEA undertaken to model the effect of EBC on component
 - Background
 - Majority of SN282 nozzles survived Solar's proof test (>90%)
 - ➢ Nozzles with EBC_{SiC} failed the proof test during transient heating
 - Motivation
 - Determine the effect of EBC on thermal gradients and stresses in nozzle during proof test







DOE EBC Workshop, Nashville, TN



FEA of the SN282 nozzles (Uncoated)

- Solid model and thermal boundary conditions for proof test obtained from Solar Turbines, Inc.
- Predicted steady state temperature gradients and stresses match Solar results
- Maximum stress at vane trailing edge









FEA of the SN282 nozzles with EBC_{SiC}

- UTRC generated solid model for the nozzle with15 mil EBC
- SN282 with EBC leads to a nozzle temperature decrease of 24°F and principal stress increase by ~7%
- Assumptions
 - Hot-pressed BSAS properties used for EBC
 - Effect of silicon layer not included









Residual stress effects on steady-state thermal stress

Nozzle Analysis	Analysis Step	Temperatures (F)		Location of Max.	Max. Principal
		Nozzle	EBC	Stress	Stress (ksi)
No EBC	RT to operation	2376/1185	-	Nozzle TE	35.5
With EBC: Residual Stress	APS to RT	RT	RT	Coating	23 - 25
	RT to operation	2352/1191	2413/1255	Nozzle TE	37.8
With EBC: No Residual Stress	RT to operation	2352/1191	2413/1255	Nozzle TE	52.8

- Nozzle thermal stress: Between 37.8 and 52.8 ksi
- Strong function of residual stress relaxation in EBC and silicon nitride
- Thermoelastic analysis includes effects of thermal gradients and CTE/Elastic mismatch only





November 18, 2003



Transient analysis (Start-up) of Solar nozzles



- Transient stress during heating is ~35% higher than steady state stress
- The time at which transient stress peaks (~15 secs) matches the experimental failure of nozzle during heat-up



November 18, 2003



Strength debit in EBC_{SiC} coated Si₃N₄ discovered







Failure probability of EBC_{SiC} coated SN282 solar nozzle determined to be close to 100%

	Material/ Property		As-processed SN282 (source: Kyocera)	SN282 with EBC (source: ORNL)
	Strength (ksi)	RT	92	10
CARES/LIFE		2552 F	68	Not Available
Analysis	Weibull Modulus (estimate)		10	10
	Over-all Vane Failure Probability	RT	3.10E-04	1.00E+00
		2552 F	6.30E-03	

Reasons for nozzle failure:
(a) Strength debit in SN282 with EBC

(b) Higher thermal stresses in coated nozzles

- Thermal stress strongly dependent on CTE mismatch between EBC and substrate and weakly dependent on modulus mismatch
- Thermal stress increases by 6 to 48% for coated nozzles, depending on the extent of residual stress relaxation



November 18, 2003



EBC_{SiC} debits silicon nitride strength

- Coating and substrate are not compatible in thermal expansion behavior.
 - ➤ CTE of EBC_{SiC}: ~5.0 ppm/°C

ightarrow CTE of Si₃N₄ : ~ 3.2 ppm/°C

- EBC debits the strength of silicon nitride :
 - > 50% debit at room temperature
 - ➤ ~15% debit at high temperature





P.19

 EBC strength debit observed for different grades of silicon nitride: AS800, SN282 and NT154 (Room Temperature)





Mechanisms causing RT strength knockdown identified

- Limited effect of surface preparation
- Thermal expansion mismatch between coating and substrate induces cracks in the coating. The cracks in the coating may act as strength controlling flaws and reduce substrate strength.
- Further investigation of chemical interaction between coating and substrate
- Coating system under development to mitigate substrate strength knockdown





P.20



DOE EBC Workshop, Nashville, TN



Current EBC approaches for Si₃N₄

Bond Coat

- Eliminate cracking in coating
- Reducing CTE mismatch

Top Coat

• CTE matched moisture and oxidation barrier layers





DOE EBC Workshop, Nashville, TN

November 18, 2003



Barrier layer with CTE matching: Plasma spray parameters for crystalline yttrium silicate developed



- Under ordinary conditions, plasma spraying results in amorphous phase
- Achieving the "correct" phase assemblage critical to developing crack free, adherent coatings



November 18, 2003



EBC for silicon nitride – Summary and challenges

- The key limitation in the applicability of EBC for silicon carbide to silicon nitride materials is the CTE mismatch between the coating and the silicon nitride.
- Promising approaches to develop EBC systems for silicon nitride have been identified (bond coat and barrier layer)
- Coating system needs optimization:
 - bond layer adhesion to substrate
 - chemical compatibility and stability
 - ➤ thermal cycling
 - effect on substrate properties
- Understanding and predicting recession mechanisms in complex silicates in high pressure and high velocity environments required for life prediction



November 18, 2003

