NASA/TM—2003-212318 ARL–TR– 2969

Surface Cracking and Interface Reaction Associated Delamination Failure of Thermal and Environmental Barrier Coatings

Dongming Zhu
U.S. Army Research Laboratory, Glenn Research Center, Cleveland, Ohio

Sung R. Choi
Ohio Aerospace Institute, Brook Park, Ohio

Jeffrey I. Eldridge
Glenn Research Center, Cleveland, Ohio

Kang N. Lee
Cleveland State University, Cleveland, Ohio

Robert A. Miller
Glenn Research Center, Cleveland, Ohio

April 2003

The NASA STI Program Office . . . in Profile

Since its founding, NASA has been dedicated to the advancement of aeronautics and space science. The NASA Scientific and Technical Information (STI) Program Office plays a key part in helping NASA maintain this important role.

The NASA STI Program Office is operated by Langley Research Center, the Lead Center for NASA's scientific and technical information. The NASA STI Program Office provides access to the NASA STI Database, the largest collection of aeronautical and space science STI in the world. The Program Office is also NASA's institutional mechanism for disseminating the results of its research and development activities. These results are published by NASA in the NASA STI Report Series, which includes the following report types:

- TECHNICAL PUBLICATION. Reports of completed research or a major significant phase of research that present the results of NASA programs and include extensive data or theoretical analysis. Includes compilations of significant scientific and technical data and information deemed to be of continuing reference value. NASA's counterpart of peer-reviewed formal professional papers but has less stringent limitations on manuscript length and extent of graphic presentations.

- TECHNICAL MEMORANDUM. Scientific and technical findings that are preliminary or of specialized interest, e.g., quick release reports, working papers, and bibliographies that contain minimal annotation. Does not contain extensive analysis.

- CONTRACTOR REPORT. Scientific and technical findings by NASA-sponsored contractors and grantees.

- CONFERENCE PUBLICATION. Collected papers from scientific and technical conferences, symposia, seminars, or other meetings sponsored or cosponsored by NASA.

- SPECIAL PUBLICATION. Scientific, technical, or historical information from NASA programs, projects, and missions, often concerned with subjects having substantial public interest.

- TECHNICAL TRANSLATION. English-language translations of foreign scientific and technical material pertinent to NASA's mission.

Specialized services that complement the STI Program Office's diverse offerings include creating custom thesauri, building customized databases, organizing and publishing research results . . . even providing videos.

For more information about the NASA STI Program Office, see the following:

- Access the NASA STI Program Home Page at ***http://www.sti.nasa.gov***

- E-mail your question via the Internet to help@sti.nasa.gov

- Fax your question to the NASA Access Help Desk at 301–621–0134

- Telephone the NASA Access Help Desk at 301–621–0390

- Write to:
  NASA Access Help Desk
  NASA Center for AeroSpace Information
  7121 Standard Drive
  Hanover, MD 21076

NASA/TM—2003-212318 ARL–TR–2969

Surface Cracking and Interface Reaction Associated Delamination Failure of Thermal and Environmental Barrier Coatings

Dongming Zhu
U.S. Army Research Laboratory, Glenn Research Center, Cleveland, Ohio

Sung R. Choi
Ohio Aerospace Institute, Brook Park, Ohio

Jeffrey I. Eldridge
Glenn Research Center, Cleveland, Ohio

Kang N. Lee
Cleveland State University, Cleveland, Ohio

Robert A. Miller
Glenn Research Center, Cleveland, Ohio

National Aeronautics and
Space Administration

Glenn Research Center

April 2003

Acknowledgments

This work was supported by the NASA Ultra-Efficient Engine Technology (UEET) Program.
The authors are grateful to George W. Leissler and Terry R. McCue, QSS, Inc., at NASA Glenn Research Center
for their assistance in the preparation of plasma-sprayed TBC/EBC coatings and the SEM analysis, respectively.

Available from

NASA Center for Aerospace Information
7121 Standard Drive
Hanover, MD 21076

National Technical Information Service
5285 Port Royal Road
Springfield, VA 22100

Available electronically at http://gltrs.grc.nasa.gov

Surface Cracking and Interface Reaction Associated Delamination Failure of Thermal and Environmental Barrier Coatings

Dongming Zhu
U.S. Army Research Laboratory
Glenn Research Center
Cleveland, Ohio 44135

Sung R. Choi
Ohio Aerospace Institute
Brook Park, Ohio 44142

Jeffrey I. Eldridge
National Aeronautics and Space Administration
Glenn Research Center
Cleveland, Ohio 44135

Kang N. Lee
Cleveland State University
Cleveland, Ohio 44115

Robert A. Miller
National Aeronautics and Space Administration
Glenn Research Center
Cleveland, Ohio 44135

Abstract

In this paper, surface cracking and interface reactions of a barium-strontium-aluminosilicate (BSAS) coating and a multi-layer ZrO_2–8wt%Y_2O_3 and mullite/BSAS/Si thermal and environmental barrier coating system on SiC/SiC ceramic matrix composites were characterized after long-term combined laser thermal gradient and furnace cyclic tests in a water vapor containing environment. The surface cracking was analyzed based on the coating thermal gradient sintering behavior and thermal expansion mismatch stress characteristics under the thermal cyclic conditions. The interface reactions, which were largely enhanced by the coating surface cracking in the water vapor environment, were investigated in detail, and the reaction phases were identified for the coating system after the long-term exposure. The accelerated coating delamination failure was attributed to the increased delamination driving force under the thermal gradient cyclic loading and the reduced interface adhesion due to the detrimental interface reactions.

Introduction

Thermal and environmental barrier coatings (T/EBCs) will play a critical role in future advanced gas turbine engines to protect Si-based engine components in the harsh combustion environments. In order to develop high performance ceramic coating systems, advanced laser based high-heat-flux approaches have been established to test coating systems under the simulated engine heat flux condition (refs. 1, 2). The laser thermal gradient test rigs can establish a required thermal gradient across the coating system, so the coating can be evaluated at very high surface temperatures while the coating/substrate interface and the substrate can still be maintained below a safe temperature limit. The laser thermal gradient approaches have been demonstrated in evaluating cyclic and sintering behavior of thermal/environmental barrier coatings on SiC/SiC substrates (refs. 3, 4).

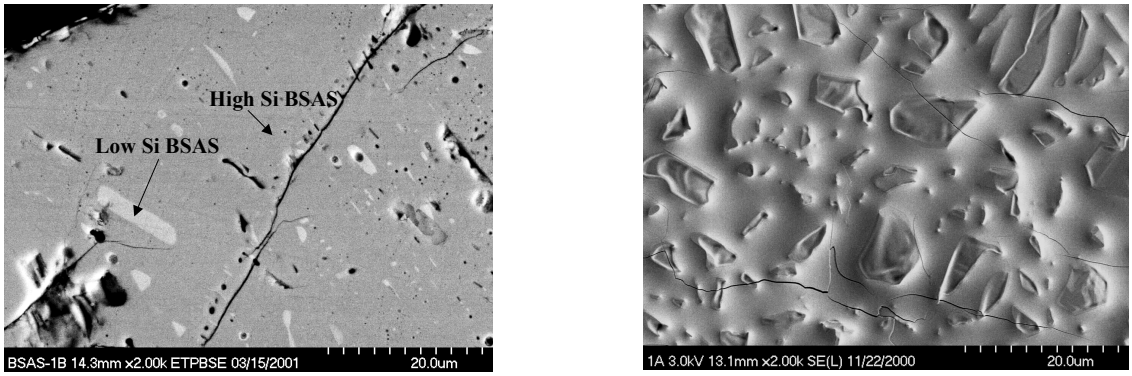
The purpose of this study is to investigate coating cracking and interface chemical reactions induced failure of typical thermal and environmental barrier coating system on SiC/SiC ceramic matrix composite (CMC) substrates under laser thermal gradient cyclic test conditions. The coating surface and delamination cracking was analyzed based on the coating thermal gradient sintering behavior and thermal expansion mismatch stress characteristics under the thermal cyclic conditions. The thermal strain/stress characteristics under large thermal gradients were also described. The accelerated coating failure was discussed based on the delamination driving force under the thermal gradient cyclic loading and the interface degradation due to the BSAS phase chemical stability and reactions in high temperature water vapor environment.

Experimental Method and Materials

The laser high-heat-flux based technique was employed to test a barium-strontium-aluminosilicate (BSAS) and a multi-layered ZrO_2–8wt%Y_2O_3 and BSAS+mullite mixture/Si TBC/EBC system on SiC/SiC CMC substrates under thermal gradient cyclic conditions. The thermal gradient cyclic testing of the ceramic coatings was carried out using a 3.0 kW CO_2 laser (wavelength 10.6 μm) high-heat flux rig. The general test approaches have been described elsewhere (refs. 1–4). The surface test temperature was approximately at 1482 °C, and interface temperature was at 1300 °C. The coatings specimens were also subjected to alternating the laser thermal gradient cycling test in air and the furnace thermal cycling tests in a 90% water vapor environment at 1300 °C (every 50 hours). The thermal cyclic tests were conducted using 1 hour hot time temperature cycles, with 3 min cooling between each cycle to ensure that the test specimens were cooled below 100 °C. The coatings were tested up to total 400, 1 hour cycles. The coating surface cracking and interface reactions of the coating systems were characterized after the testing using a scanning electron microscope (SEM) equipped with an energy dispersive spectroscopy (EDS) system.

Results and Discussion

Figure 1 shows SEM microstructures of the BSAS/Si environmental barrier coating after laser thermal gradient cyclic testing. It can be seen from Figure 1 (a) that the BSAS coating was first decomposed and precipitated a lower silicon (Si) containing phase, with the matrix being a high Si containing BSAS. As the coating was further cyclically tested at a surface temperature of 1482 °C, the BSAS matrix was melted because of the formation of the higher Si and lower melting point BSAS. The surface cracking was also observed after coating melting and glass phase formation.



(a) (b)

Figure 1.—(a) The BSAS coating was decomposed into low Si (bright second phase) and high Si (dark matrix) phases during initial laser thermal cyclic testing. (b) The higher Si phase became the glass phase and surface cracks were formed after 100 hour cyclic testing at the surface temperature of 1482 °C.

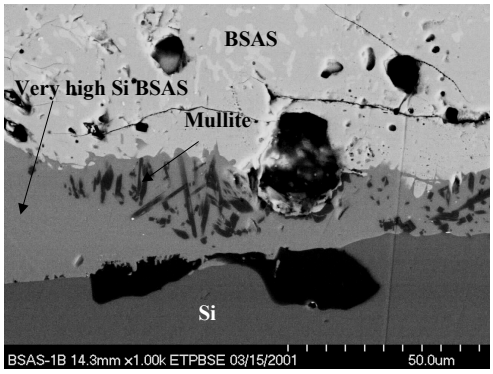


Figure 2.—SEM cross-section micrograph of the BSAS coating after 100 hour laser thermal gradient cycling test. The interface reactions resulted in a dark reaction BSAS layer with high Si concentrations. The mullite phase (very dark needle-shaped phase) precipitation was also found within the high Si reaction BSAS region.

Figure 2 shows a SEM cross-section micrograph of the BSAS coating after 100 hour laser thermal gradient cycling test. The interface reactions between the Si bond coat and BSAS coating resulted in a dark reaction layer with the layer thickness of 30 to 40 microns. The EDS analysis has shown that this BSAS region has very high Si concentrations due to the Si diffusion from the Si bond coat into the BSAS. The high Si concentration BSAS phase was also decomposed into mullite phase (very dark needle-shaped phase) within the high Si BSAS reaction region. Although the coating interface melting was not observed under the 1300 °C interface temperature of the thermal gradient testing for 100 hours, the high Si concentration BSAS reaction layer has been reported to facilitate the coating melting and the glass phase formation under the uniform temperature furnace testing in a water vapor environment at higher temperatures (ref. 5).

Figure 3 shows cross-section micrographs and compositional maps of the ZrO_2–8wt%Y_2O_3/BSAS+mullite mixture/Si on a SiC/SiC ceramic matrix composite substrate, after 100 hours testing in a combined alternating laser thermal gradient cyclic and furnace water vapor exposure cyclic

testing. It can be seen that substantial thermal gradient induced coating surface wedge-shape cracking has occurred and the resulting delamination was observed for the coating specimen. As shown in Figure 3, significant interface reactions within the mullite+BSAS EBC layer occurred. The reaction affected zone can be as thick as 30 to 40 microns, especially near the interface and the BSAS phase (bright phase) regions. The reactions between mullite and BSAS phases were also noticed with a typical reaction layer thickness of 2 to 5 microns. However, the inter-diffusion of Ba and Al between the mullite and BSAS can occur at a much greater distance, as indicated by the EDS composition maps in Figure 3 (d) and (e). The reactions were further enhanced by the surface crack penetrations into the EBC layer which allowed the water vapor fast access to the coating at the EBC/Si interface. The accelerated coating damage under the thermal gradient cyclic testing combined with the furnace water vapor testing was attributed to the increased delamination driving force under the thermal gradient cyclic loading and the reduced interface adhesion due to the detrimental interface reactions (refs. 4, 6).

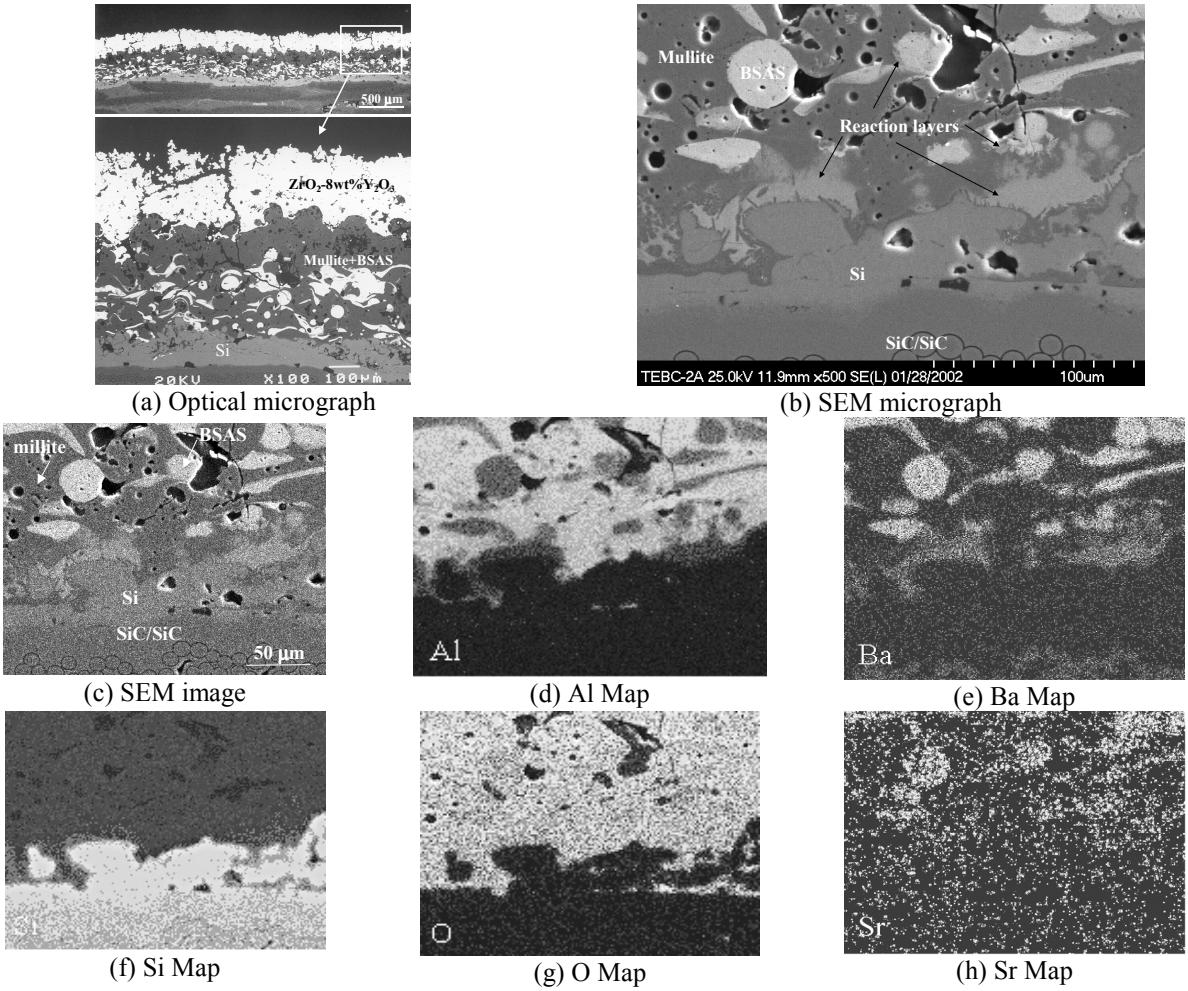
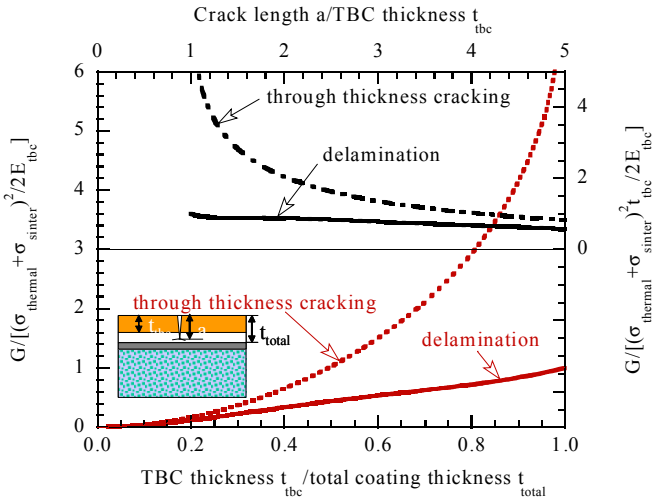
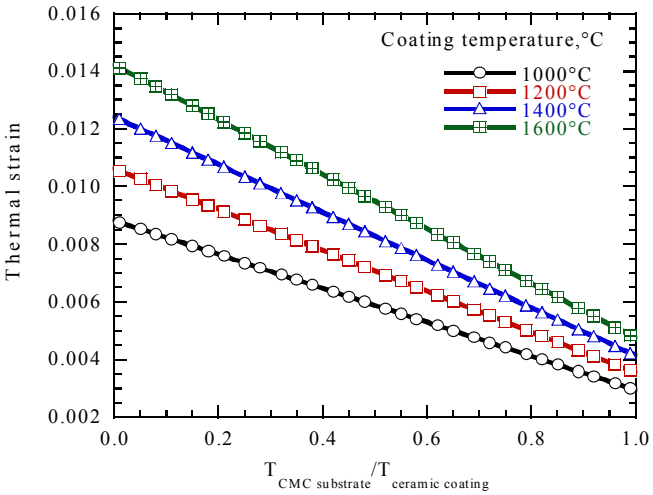


Figure 3.—SEM images and EDS element composition maps of the ZrO_2–8wt%Y_2O_3/BSAS+mullite mixture/Si on SiC/SiC ceramic matrix composite substrates after laser testing, showing significant interface reactions between BSAS and bond coat Si layer, and the BSAS and mullite. (a) and (b) Optical and SEM micrographs of the coating after 100 hours testing, showing the surface cracking and interface reactions; and (c) to (h) SEM cross-section micrograph and the corresponding EDS elemental maps for the coating systems near the interface.

The coating surface cracking is largely due to ceramic top coat sintering and thermal expansion mismatch, especially under the large thermal gradient conditions. As can be seen in Figure 4, with a relatively thick, higher thermal expansion ZrO_2–Y_2O_3 TBC layer on mullite-BSAS EBC and CMC substrates, the coating surface cracking and delamination driving force can increase significantly, based on a thermal gradient testing delamination model proposed in the literature (ref. 7). The significantly larger thermal stresses generated under thermal gradients will also accelerate the coating delamination under the thermal cyclic conditions.



(a)



(b)

Figure 4.—(a) ZrO_2–Y_2O_3 TBC layer on mullite-BSAS EBC and CMC substrates, the coating surface cracking and delamination energy release rates can increase significantly with a relatively thick, and larger thermal expansion ZrO_2–Y_2O_3 TBC layer on mullite-BSAS EBC on CMC substrates. (b) The significantly larger thermal stresses/strains will be generated with increasing the thermal gradients (the temperature difference between the ceramic coating and the substrates).

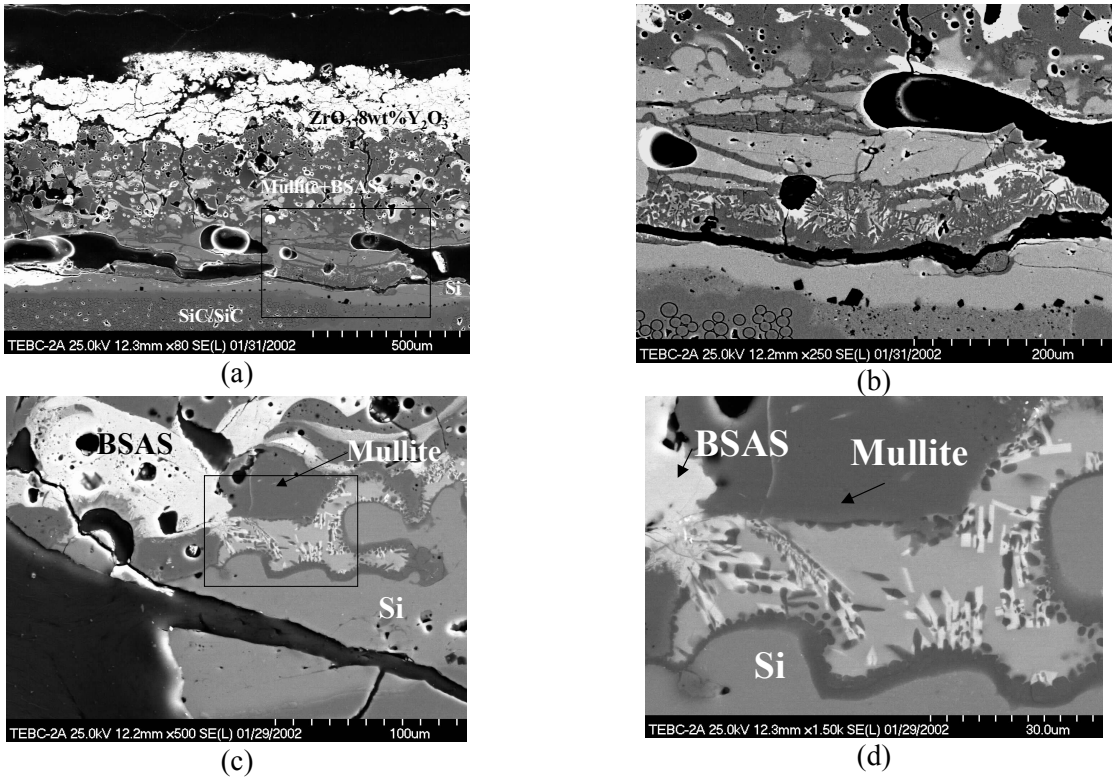


Figure 5.—SEM cross-section micrographs of the ZrO_2–8wt%Y_2O_3/BSAS+mullite mixture/Si on a SiC/SiC ceramic matrix composite substrate, after total 400 hours testing in the combined laser thermal gradient and furnace water vapor cyclic testing. (a) Significant interfacial pore formation due to the water vapor attack through the vertical cracks under the thermal gradient cycling conditions. (b) Low melting eutectic phase was also observed near the BSAS-mullite-Si interfaces after the testing.

Figure 5 shows the SEM cross-section micrographs of the ZrO_2–8wt%Y_2O_3/BSAS+mullite mixture/Si on a SiC/SiC ceramic matrix composite substrate, after 400 hours of testing in the combined laser thermal gradient and furnace water vapor cyclic testing. Extensive coating delamination and some spallation were observed during the testing. From Figure 5 (a) and (b), it can be seen that, in addition to the more significant reactions between the EBC mullite, BSAS and Si bond coat, large interfacial pores developed due to the water vapor attack through the vertical cracks in the EBC coatings. From Figure 5 (c) and (d), it can also be seen that a low melting point eutectic phase around the BSAS, Si, and mullite interfaces formed. For the top ZrO_2–Y_2O_3 coating side, no obvious reactions were found between mullite and ZrO_2–8wt%Y_2O_3 coatings after the 400 hours combined laser and furnace water vapor thermal cycling test.

Conclusions

A laser heat-flux thermal gradient test approach has been used to investigate the interface reactions and failure of plasma-sprayed BSAS and ZrO_2–8wt%Y_2O_3/BSAS+mullite/Si coatings on SiC/SiC ceramic matrix composites under thermal gradient cyclic conditions. The failure of the coating system can be characterized as wedge-shaped surface coating cracking, surface cracking-enhanced coating

delamination and interface debonding and spallation under the thermal cyclic conditions. Significant BSAS phase related chemical reactions were observed due to its low temperature stability and high diffusion activity. The BSAS coatings were found to melt at even relatively low temperatures (1300 to 1482 °C) because of the formation of higher Si content, and low melting-point phases, either at the surface due to its high temperature decomposition into high Si containing matrix phase with low Si BSAS precipitates, or near the interface when reacted with Si bond coat. The accelerated coating delamination failure was attributed to the increased delamination driving force under the thermal gradient cyclic loading and the reduced interface adhesion due to the detrimental interface reactions.

References

1. D. Zhu and R. A. Miller, "Thermal Conductivity and Elastic Modulus Evolution of Thermal Barrier Coatings Under High Heat Flux Conditions," NASA Glenn Research Center, Cleveland, Ohio, NASA/TM-209069, April 1999. Journal of Thermal Spray Technology, vol. 9, pp. 175–180, 2000.
2. D. Zhu, N. P. Bansal, K. N. Lee, and R. A. Miller, "Thermal Conductivity of Ceramic Thermal Barrier and Environmental Barrier Coating Materials," NASA Glenn Research Center, Cleveland NASA/TM-211122, September 2001.
3. D. Zhu, K. N. Lee, and R. A. Miller, "Thermal Conductivity and Thermal Gradient Cyclic Behavior of Refractory Silicate Coatings on SiC/SiC Ceramic Matrix Composites," Ceram. Eng. Sci. Proc., vol. 22, pp. 443–452, 2001.
4. D. Zhu, K. N. Lee, and R. A. Miller, "Sintering and Cyclic Failure Mechanisms of Multi-Layered Thermal and Environmental Barrier Coating Systems under High Thermal Gradient Test Conditions," Ceram. Eng. Sci. Proc., vol. 23, pp. 505–516, 2002.
5. K.N. Lee, D.S. Fox, J.I. Eldridge, D. Zhu, R.C. Robinson, N.P. Bansal, and R.A. Miller, "Upper Temperature Limit of Environmental Barrier Coatings Based on Mullite and BSAS," NASA/TM—2002-211372.
6. Dongming Zhu, Kang N. Lee and Robert A. Miller, "Thermal Gradient Cyclic Behavior of Thermal/Environmental Barrier Coating System on SiC/SiC Ceramic Matrix Composites," in Proceedings of ASME Turbo Expo 2002, June 3–6, 2002, ASME Paper GT-2002-30632, Amsterdam, The Netherlands.
7. J. H. Hutchinson and A. G. Evens, "On the delamination of thermal barrier coatings in a thermal gradient," Surface and Coatings Technology, vol. 149, pp. 179–184, 2002.

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

Public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302, and to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503.

1. AGENCY USE ONLY (*Leave blank*)	2. REPORT DATE	3. REPORT TYPE AND DATES COVERED
	April 2003	Technical Memorandum

4. TITLE AND SUBTITLE

Surface Cracking and Interface Reaction Associated Delamination Failure of Thermal and Environmental Barrier Coatings

5. FUNDING NUMBERS

WBS–22–714–04–05
1L161102AF20

6. AUTHOR(S)

Dongming Zhu, Sung R. Choi, Jeffrey I. Eldridge, Kang N. Lee, and Robert A. Miller

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)

National Aeronautics and Space Administration
John H. Glenn Research Center at Lewis Field
Cleveland, Ohio 44135–3191

8. PERFORMING ORGANIZATION REPORT NUMBER

E–13831–1

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)

National Aeronautics and Space Administration
Washington, DC 20546–0001
and
U.S. Army Research Laboratory
Adelphi, Maryland 20783–1145

10. SPONSORING/MONITORING AGENCY REPORT NUMBER

NASA TM—2003-212318
ARL–TR–2969

11. SUPPLEMENTARY NOTES

Dongming Zhu, U.S. Army Research Laboratory, NASA Glenn Research Center; Sung R. Choi, Ohio Aerospace Institute, Brook Park, Ohio 44142; Jeffrey I. Eldridge and Robert A. Miller, NASA Glenn Research Center; Kang N. Lee, Cleveland State University, Cleveland, Ohio 44115. Responsible person, Dongming Zhu, organization code 5160, 216–433–5422.

12a. DISTRIBUTION/AVAILABILITY STATEMENT

Unclassified - Unlimited
Subject Categories: 24, 27, and 23 Distribution: Nonstandard

Available electronically at http://gltrs.grc.nasa.gov

This publication is available from the NASA Center for AeroSpace Information, 301–621–0390.

12b. DISTRIBUTION CODE

13. ABSTRACT ***(Maximum 200 words)***

In this paper, surface cracking and interface reactions of a BSAS coating and a multi-layer ZrO_2-8wt%Y_2O_3 and mullite/BSAS/Si thermal and environmental barrier coating system on SiC/SiC ceramic matrix composites were characterized after long-term combined laser thermal gradient and furnace cyclic tests in a water vapor containing environment. The surface cracking was analyzed based on the coating thermal gradient sintering behavior and thermal expansion mismatch stress characteristics under the thermal cyclic conditions. The interface reactions, which were largely enhanced by the coating surface cracking in the water vapor environment, were investigated in detail, and the reaction phases were identified for the coating system after the long-term exposure. The accelerated coating delamination failure was attributed to the increased delamination driving force under the thermal gradient cyclic loading and the reduced interface adhesion due to the detrimental interface reactions.

14. SUBJECT TERMS

Thermal barrier coatings; Environmental barrier coatings; Thermal gradient heat flux testing; Failure and delamination mechanisms; Interface diffusion

15. NUMBER OF PAGES

13

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
Unclassified	Unclassified	Unclassified	