DEVELOPMENT OF NONDESTRUCTIVE EVALUATION METHODS FOR CERAMIC COATINGS

J. G. Sun
Argonne National Laboratory, Nuclear Engineering Division
9700 South Cass Avenue, Argonne, IL 60439
E-mail: sun@anl.gov; Telephone (630)252-5169; FAX (630)252-2785

ABSTRACT

Nondestructive evaluation (NDE) methods are being developed for ceramic thermal barrier coatings (TBCs) applied to components in the hot-gas path of advanced high-efficiency and low-emission gas turbines, including syn-gas fired turbines. The objectives of the NDE development are to assess TBC condition for quality control as well as to monitor TBC degradation and predict TBC life during service. These objectives can be achieved based on accurate measurement of TBC properties and high-resolution detection of material flaws by NDE methods. Current NDE development is focused on two thermal imaging methods: multilayer thermal modeling and thermal tomography. The multilayer-modeling method may determine the thermal property distribution of the TBC layer over the entire component surface, and the thermal tomography method can image the TBC and substrate structure in 3D. Considerable progress was made in developing rigorous multilayer-model algorithms to determine TBC thermal properties with accuracies comparable to standard measurement methods. On the other hand, thermal tomography has been used to detect delamination as well as micro-cracking in TBC samples. In addition for TBC characterization, both methods were also used for other material systems including metallic coatings. This paper describes recent developments and experimental results from these NDE methods.

INTRODUCTION

Ceramic thermal barrier coatings (TBCs) are extensively used on hot gas-path components in advanced high-efficiency and low-emission gas turbines, including syn-gas fired turbines.[1-2] In this application, a thermally insulating ceramic topcoat (the TBC) is bonded to a thin oxidation-resistant metal coating (the bond coat) on a metal substrate. TBC coated components can therefore be operated at higher temperatures, with improved performance and extended lifetime. TBCs are typically applied by electron beam–physical vapor deposition (EB-PVD) and air plasma spraying (APS). As TBCs become "prime reliant," it becomes important to know their conditions nondestructively to assure the reliability of these components. NDE methods can be used to assess the quality of new coatings, identify defective components that could cause unscheduled outages, monitor degradation rates during engine service, and provide data for reaching rational decisions on replace/repair/re-use of components.

Work at Argonne National Laboratory (ANL) is underway to develop advanced NDE methods for TBCs. TBC failure normally starts from initiation of small cracks at the TBC/bond coat interface. These cracks then grow and link together to form delaminations which eventually cause TBC spallation. Conventional NDE methods are mostly based on optical spectroscopy or imaging.[3-5] Although these methods are successful for EB-PVD TBCs, they are generally not suitable for APS TBCs because these coatings are thicker and less transparent. NDE development at ANL is therefore focused on thermal imaging methods that can be used for all TBCs, as well as other coatings, during their entire life cycle. Two thermal imaging methods are being developed: multilayer thermal modeling[6] and thermal tomography.[7] The multilayer-modeling method may determine the thermal property distribution of the TBC layer over the entire component surface, and the thermal tomography method can image the TBC and substrate structure in 3D.

Because the primary function of a TBC is for thermal insulation, the most important TBC parameters are thermal properties, particularly the thermal conductivity. TBC conductivity can be measured by several methods. The most reliable and commonly used method is laser flash method.[8] This method however is a destructive method and requires two-sided access of the specimen, so it cannot be used to analyze TBCs coated on real components. To establish thermal imaging for not only NDE but also fundamental studies for TBCs, development for the multilayer-modeling method has been focused on developing rigorous algorithms so that its measurement accuracy is comparable to the laser-flash method. As a result, the multilayer-modeling NDE method may be used to monitor TBC degradation and predict TBC lifetime based on the evolution of TBC thermal conductivity.[9] On the other hand, thermal tomography has been used to detect delamination as well as micro-cracking within TBC layer. In addition for TBC characterization, both thermal imaging methods were also evaluated for other coating systems including metallic coatings. This paper presents recent developments and experimental results to demonstrate the applications of these NDE methods for measurement of coating properties and detection of material flaws/damages.

PULSED THERMAL IMAGING FOR TBC MATERIALS

Pulsed thermal imaging is based on monitoring the temperature decay on a specimen surface after it is applied with a pulsed thermal energy that is gradually transferred inside the specimen. A schematic one-sided pulsed-thermal-imaging setup for testing a 3-layer material system is illustrated in Fig. 1. The premise is that the heat transfer from the surface (or surface temperature/time response) is affected by internal material structures and properties and the presence of flaws such as cracks.[10] By analyzing the surface temperature/time response, the material property and depth of various subsurface layers under the surface can be determined.

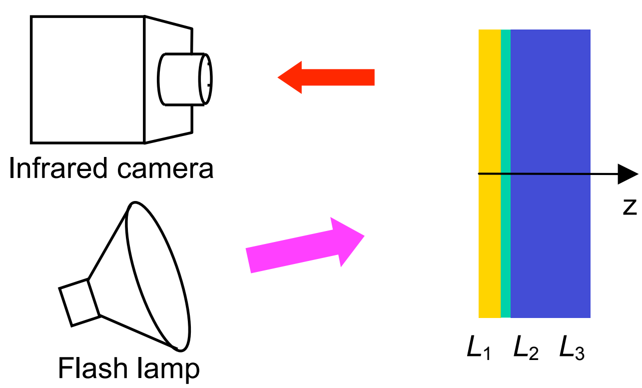


Fig. 1. Schematics of pulsed thermal imaging of a 3-layer material system.

Thermal imaging data are sensitive to several important TBC parameters, including the thickness, thermal conductivity and heat capacity (the product of density and specific heat) of the top ceramic TBC layer. The thermal imaging data can be used as input to the multilayer thermal modeling method and the thermal tomography method. The multilayer-modeling method may determine the thermal property distribution of the TBC layer over the entire component surface, and the thermal tomography method can image the TBC and substrate structure in 3D.

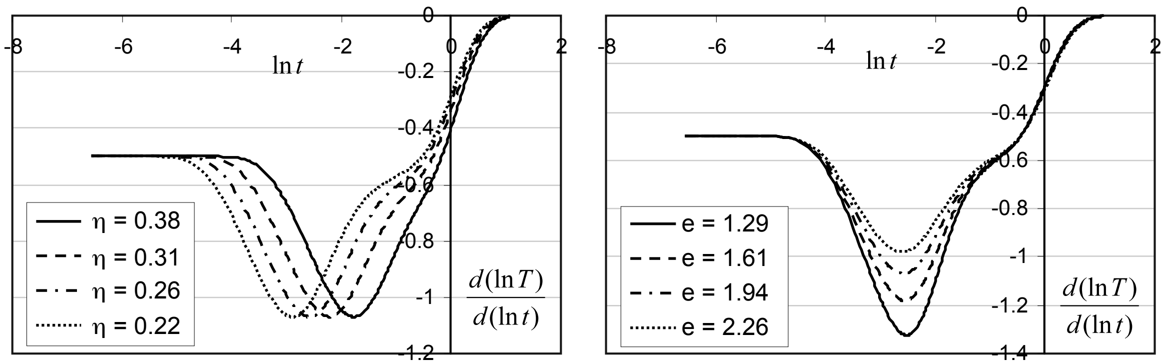
MULTILAYER THERMAL MODELING METHOD

In the multilayer thermal modeling method,[6] a TBC is modeled by a multilayer material system and the 1D heat-transfer equation governing the pulsed thermal-imaging process is solved by numerical simulation. The numerical formulation may also incorporate other factors related to experiment or sample conditions; e.g., the finite flash duration of the flash lamps and the TBC translucency which causes volume heat absorption. The numerical solutions (of surface temperature decay) are then fitted with the experimental data at each pixel by least-square minimization to determine unknown parameters in the multilayer material system. Multiple parameters in one or several layers can be determined simultaneously. This data fitting process is automated for all pixels within the thermal images and the final results are presented as images of the predicted TBC parameters.

The important TBC parameters to be determined by thermal imaging include the thickness L, thermal conductivity k, and heat capacity ρc (where ρ is density and c is specific heat) of the top ceramic TBC layer. These three TBC parameters, however, may not be independent so may not be determined individually from the thermal imaging test. This problem is inherent to many thermal measurement methods. For example, the well-known laser flash method[8] can only determine one single parameter from the same set of three parameters in a single-layer material.

A theoretical analysis[11] was conducted to identify the number of independent TBC parameters that can be determined from a one-sided thermal imaging test. For a two-layer TBC system where the bond coat is considered as part of the substrate and the TBC top coat is opaque (TBC surface is covered by a black paint), the analytical solution for the surface temperature as a function of time, which is measured in thermal imaging, contains only two independent parameters for each layer: the thermal effusivity e (= $(k\rho c)^{1/2}$) and the parameter $\eta = L/\alpha^{1/2}$ where α (= $k/\rho c$) is the thermal diffusivity. The dependency of the solution to the two TBC parameters becomes clear when examining the surface temperature decay slope, $d(\ln T)/d(\ln t)$, as function of time. Figure 2 shows the front surface temperature slope $d(\ln T)/d(\ln t)$ for TBCs with a varying parameter η (Fig. 2a) or effusivity e (Fig. 2b) while keeping the other parameter constant. The most visible characteristic of the temperature slope data is the large negative peak. This negative slope peak is the result of an increased heat conduction rate when the absorbed surface heat from the flash lamp is transferred from the topcoat into the substrate that has a higher thermal effusivity than the topcoat. Both the position (or transition time) and the magnitude of the peak change with the TBC thermal properties. In Fig. 2a, it is seen that the transition time for the slope change from -0.5 to -1.07 (the negative peak) is related only to the parameter η. Therefore, it is concluded that the parameter η is an independent parameter that determines the transition time for temperature-slope change, as predicted from theoretical analysis. On the other hand, the TBC effusivity e affects only the maximum slope value (see Fig. 2b). This result further confirms that among the three TBC parameters, thickness L and two thermal properties, only two of them can be determined uniquely from thermal imaging data.

The multilayer modeling method can therefore predict two TBC properties: the thermal effusivity e and the parameter η. With a known TBC thickness L, TBC thermal conductivity k and heat capacity ρc are determined from: $k = Le/\eta$, $\rho c = e\eta/L$. Alternatively, if the TBC thermal properties are know, the TBC thickness can be determined.



(a) (b)

Fig. 2. Surface temperature decay slope as function of time for two-layer TBC systems with variation of (a) parameter η ($s^{1/2}$) and (b) effusivity e (kJ/m^2-K-s$^{1/2}$).

Factors Affecting Measurement Accuracy

Although the fundamental theory for thermal imaging of multilayer TBC materials is rigorous and simple, the prediction accuracy can be affected by many factors from experimental and sample condition variations.[12] The experimental factors may come from various sources, including the characteristics of the flash (duration and intensity curve), nonlinear relationship between camera signal and temperature, emissivity variation of TBC surface, and thermal reflection from surrounding heat sources. To address these issues, a dynamic calibration procedure was developed to determine the correct surface temperature and an infrared filter was used to completely eliminate the flash-lamp infrared radiation.[13]

At least two factors from TBC samples may affect the measurement accuracy: the black paint normally applied on TBC surface and the TBC surface roughness. The condition of the paint may significantly affect the measurement results for thin TBC coatings, as demonstrated below. The roughness of the TBC surface has complex effect to the surface temperature transient; it is not studied at present.

Black paints have been used in most of the thermal imaging and laser flash tests for TBCs. These paints consist of micron-sized graphite particles and some binder materials. Depending on the composition, they may have different thermal properties (conductivity and heat capacity). When applied on TBC surface, a sufficient thickness of the paint layer is required to prevent the light penetration through the paint. As a result, the black paint may have to be considered as an additional layer of material on the TBC surface when its thermal effect cannot be neglected. This effect comes from the combination of the thickness and the thermal properties of the paint, as well as a possible infiltration of the paint inside the pores of the TBC coating. To investigate the effect of the paint, two graphite-based paints, both are normally used as dry-film lubricant, were tested on TBC samples of various thicknesses. Paint #1 is a coarse heavy-duty lubricant and Paint #2 is a long-wearing lubricant with fine graphite particles. Because they are normally sprayed manually on TBC surfaces, their thicknesses cannot be controlled and are therefore unknown. Intuitively, it is believed that the film thickness of Paint #1 is usually thicker than that of Paint #2.

For TBC coatings with thickness ≥0.3 mm, it was found that measured thermal properties are essentially the same for TBC samples coated with both paints. However, when an additional layer of the same paint was applied, predicted thermal properties for the TBC sample coated with Paint #1 showed considerable increases, while those with Paint #2 did not change. Similarly, the predicted thermal properties for thinner TBCs were higher in Paint #1 coated samples. To illustrate the effect of the paints, a 152-µm-thick APS TBC sample was coated with each paint on half of its surface, and a thermal imaging test was conducted for this sample. The average temperature-slope curves in the two painted surface areas are shown in Fig. 3. A significant difference is observed for both position and magnitude of the negative peak: it occurs at an earlier time and has a low magnitude in Paint #1 area. Consequently, the predicted thermal properties within the Paint #1 surface are much higher, k = 0.93 W/m-K and ρc = 3.27 J/cm^3-K, compared with those in Paint #2 surface, k = 0.80 W/m-K and ρc = 3.06 J/cm^3-K. This result demonstrates the importance in selection and application of the black paint for thermal imaging and likely laser flash tests for TBC samples, and further studies should be considered to understand the mechanisms.

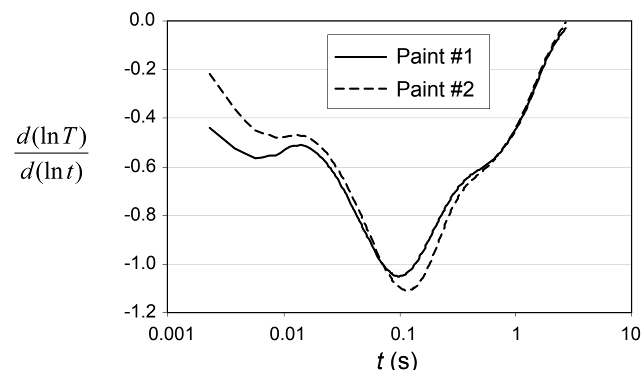


Fig. 3. Temperature-derivative data on surface areas coated with Paint #1 and #2.

Thermal Property Measurement for TBCs

The multilayer modeling method was used to measure TBC thermal properties, conductivity and heat capacity, with known TBC thickness for APS and EBPVD TBC specimens. Measurement results for a thick and a thin TBC specimen are presented. Figure 4 shows the measured data for the thick APS TBC specimen (sample curtsey of Dr. Y. Tan of Stony Brook University, NY); these data are compared with those typical for such materials obtained from laser flash test. The top ceramic coating is 0.86 mm thick with considerable surface roughness, and the substrate is a stainless steel material with a thickness of 10 mm. The predicted average TBC conductivity was 0.93 W/m-K, which is consistent with the measured values of 1.0±0.2 W/m-K from laser flash tests. The predicted average TBC heat capacity was 2.19 J/cm^3-K, which is also in good agreement with typical value of 2.0 J/cm^3-K for this TBC. The minor differences are probably due to the material variation in individual samples and small variations in the TBC thickness.

Thermal property measurement was also performed for a very-thin as-processed EBPVD TBC specimen with a surface area of 25.4 mm in diameter (sample curtsey of Dr. A.M. Limarga and Dr. D. Clarke of Harvard University). The top ceramic coating is only 50-μm thick, and the substrate has a thickness of 3.1 mm. The predicted TBC conductivity and heat capacity distributions are shown in Fig. 5. It is seen that the predicted thermal conductivity and heat capacity images are uniform. The predicted average TBC conductivity is 0.87 W/m-K, and the predicted average TBC heat capacity is 2.90 J/cm^3-K, both are within a few percent of reference values.

The results in Figs. 4 and 5 show that the multilayer modeling method can be used to predict TBC conductivity and heat capacity with accuracies comparable to standard testing methods. A significant advantage of this method is that it can simultaneously determine two thermal properties, k and ρc, while each would require a separate test by conventional methods.

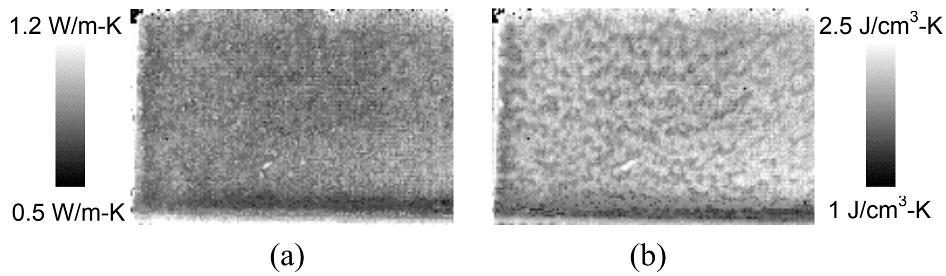


Fig. 4. Predicted (a) conductivity and (b) heat capacity images of a 0.86mm-thick TBC sample.

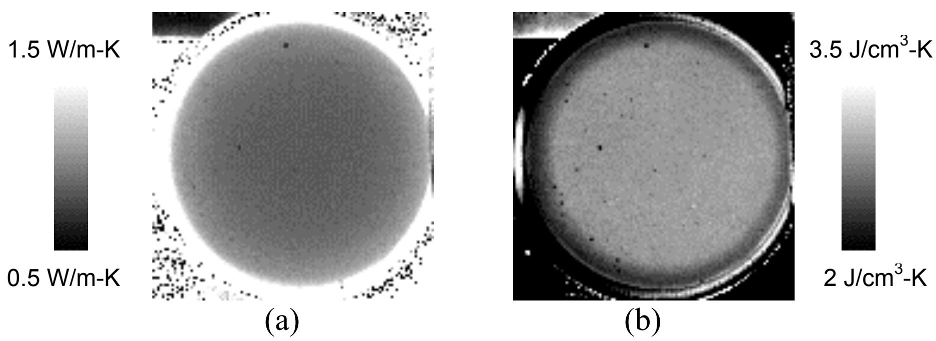


Fig. 5. Predicted (a) conductivity and (b) heat capacity images of a 50μm-thick TBC specimen.

THERMAL TOMOGRAPHY METHOD

Thermal tomography is the first 3D thermal imaging method developed at ANL.[8] This method directly converts the pulsed thermal-imaging data into a 3D thermal effusivity data that can be viewed/sliced for analysis in any plane direction (similar to 3D data from x-ray CT). Because thermal effusivity is an intrinsic material property, thermal tomography data can be used to evaluate the properties of TBCs as well as to detect damages/flaws in the TBC material. It has been shown for detection of small cracks at the TBC/bond coat interface that would lead to delaminations and eventual spallation of the coating.[14] By detecting the small cracks early in the TBC degradation process, this NDE technology may be used to monitor TBC degradation and predict TBC life. In addition to TBCs, thermal tomography may also be used for NDE of metallic coatings. Figure 6 shows four plane thermal effusivity images for an 1-in.-diameter sample with a standard metallic bond coat (~125µm thick) on a superalloy substrate (sample curtsey of Dr. A. Kulkarni of Siemens). These four planes are at depths of ~60, 110, 180, and 330µm, respectively, below the surface. Correspondingly, they are at mid bond-coat depth, bottom of bond coat (just above interface), 60µm and 210µm below the interface. It is apparent that there are some defects at and below the interface, few further extend and become larger in deeper depths in the substrate. It is recognized that defects within substrate are generally not considered possible, so these results need to be further confirmed by destructive evaluation. Nevertheless, thermal tomography is capable to detect abnormal features and resolve their depths in thin-layered materials; this has not been achieved by any conventional NDE methods.

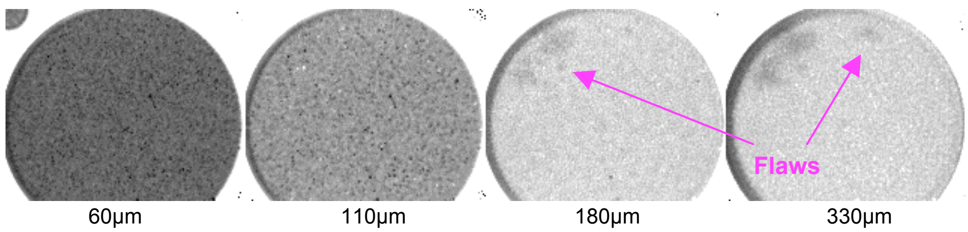


Fig. 6. Plane thermal effusivity images at depths ~60 (mid bond-coat layer), 110 (just above interface), 180 (60µm below interface), and 330µm (210µm below interface) for a 1-inch-diameter sample with a standard bond coat on a superalloy substrate.

CONCLUSION

Quantitative NDE methods are being developed to determine the physical and geometrical parameters of TBC materials, including the TBC thickness and thermal properties. These TBC parameters are representative of the TBC quality, so they can be used to evaluate as-processed TBCs and monitor TBC degradation. For both thin and thick TBCs, two thermal-imaging methods are being developed: multilayer thermal modeling and thermal tomography. The multilayer-modeling method can determine thermal property distributions of the TBC layer over the entire component surface. One significant advantage of this method is that it can simultaneously determine two thermal properties, k and ρc, while each would require a separate test by conventional methods. The multilayer thermal modeling method was used to measure the thermal properties of thick and thin TBC samples, and both predicted conductivity and heat capacity are in good agreement with known data for these materials. On the other hand, the thermal tomography method was used to image 3D thermal effusivity distributions in TBC and bond-coat samples. It was demonstrated to be capable of detecting abnormal features and resolving their depths; this cannot be achieved by other conventional NDE methods. With further development, thermal imaging will be established for accurate property measurement as well as NDE characterization for TBCs.

ACKNOWLEDGMENT

The author thanks Dr. Y. Tan of Stony Brook University, Dr. A.M. Limarga and Dr. D. Clarke of Harvard University, and Dr. A. Kulkarni of Siemens for providing TBC specimens used in this study.

REFERENCES

1. North Atlantic Treaty Organization, "Thermal Barrier Coatings," Advisory Group for Aerospace Research and Development Report, AGARD-R-823, Neuilly-Sur-Seine, France, April 1998.
2. US National Research Council, National Materials Advisory Board, "Coatings for High Temperature Structural Materials," National Academy Press, Washington, DC, 1996.
3. W.A. Ellingson, S. Naday, R. Visher, R. Lipanovich, L. Gast and C. Deemer, "Development of Nondestructive Evaluation Technology for Ceramic Coatings," Proc. 20th Annual Conference on Fossil Energy Materials, Knoxville, TN, June 12-14, 2006.
4. J.I. Eldridge, C.M. Spuckler, and R.E. Martin, "Monitoring Delamination Progression in Thermal Barrier Coatings by Mid-Infrared Reflectance Imaging," *Int. J. Appl. Ceram. Technol.*, Vol. 3, pp. 94-104, 2006.
5. B. Heeg and D.R. Clarke, "Non-destructive thermal barrier coating (TBC) damage assessment using laser-induced luminescence and infrared radiometry," Surface & Coatings Technology, Vol. 200, pp. 1298 – 1302, 2005.
6. J.G. Sun, "Method for Analyzing Multi-Layer Materials from One-Sided Pulsed Thermal Imaging," Argonne National Laboratory Invention Report, US patent pending, 2006.
7. J.G. Sun, "Method for Thermal Tomography of Thermal Effusivity from Pulsed Thermal Imaging," U.S. Patent No. 7,365,330, issued April 29, 2008.
8. ASTM E1461-07, Standard Test Method for Thermal Diffusivity by the Flash Method.
9. J.G. Sun, "Development of Nondestructive Evaluation Method for Ceramic Coatings and Membranes," Proc. 23rd Annual Conference on Fossil Energy Materials, Pittsburgh, PA, May 12-14, 2009.
10. J. G. Sun, "Analysis of Pulsed Thermography Methods for Defect Depth Prediction," *J. Heat Transfer*, Vol. 128, pp. 329-338, 2006.
11. J.G. Sun, "Measurement of Thermal Barrier Coating Conductivity by Thermal Imaging Method," in Ceramic Eng. Sci. Proc., eds. D. Singh and J. Salem, Vol. 30, no. 3, pp. 97-104, 2009.
12. J.G. Sun, "Thermal Property Measurement for Thermal Barrier Coatings by Thermal Imaging Method," paper presentation at the American Ceramic Society's 34th International Conference and Exposition on Advanced Ceramics and Composites, Daytona Beach, FL, January 24-29, 2010.
13. J.G. Sun, "Optical Filter for Flash Lamps in Pulsed Thermal Imaging," U.S. Patent No. 7,538,938 issued May 26, 2009.
14. J.G. Sun, "Development of Nondestructive Evaluation Method for Thermal Barrier Coatings," Proc. 22nd Annual Conference on Fossil Energy Materials, Pittsburgh, PA, July 8-10, 2008.