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Full-field characterization of thermal diffusivity in continuous-fiber ceramic
composite materials and components

by

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ABSTRACT

Continuous-fiber ceramic matrix composites (CFCCs) are currently being developed for various high-temperature applications, including use in advanced heat engines. Among the material classes of interest for such applications are silicon carbide (SiC)-fiber-reinforced SiC ($\text{SiC}_{(f)}/\text{SiC}$), SiC-fiber-reinforced silicon nitride ($\text{SiC}_{(f)}/\text{Si}_3\text{N}_4$), aluminum oxide (Al_2O_3)-fiber-reinforced Al_2O_3 ($\text{Al}_2\text{O}_{3(f)}/\text{Al}_2\text{O}_3$), and others. In such composites, the condition of the interfaces (between the fibers and matrix) are critical to the mechanical and thermal behavior of the component (as are conventional mechanical defects such as cracks, porosity, etc.). For example, oxidation of this interface (especially on carbon coated fibers) can seriously degrade both mechanical and thermal properties. Furthermore, thermal shock damage can degrade the matrix through extensive crack generation. A nondestructive evaluation method that could be used to assess interface condition, thermal shock damage, and to detect other "defects" would thus be very beneficial, especially if applicable to full-scale components. One method under development uses infrared thermal imaging to provide "single-shot" full-field assessment of the distribution of thermal properties in large components by measuring thermal diffusivity. By applying digital image filtering, interpolation, and least-squares-estimation techniques for noise reduction, we can achieve acquisition and analysis times of minutes or less with submillimeter spatial resolution. The system developed at Argonne National Laboratory has been used to examine the effects of thermal shock, oxidation treatment, density variations, and variations in oxidation resistant coatings in a full array of test specimens. Subscale CFCC components with nonplanar geometries have also been studied for manufacturing-induced variations in thermal properties.

Keywords: ceramics, continuous fiber ceramic matrix composites, CFCCs, thermal diffusivity, NDE, IR imaging

2. INTRODUCTION

Continuous fiber ceramic matrix composites are currently being considered as replacements for traditional metallic materials in some gas turbine applications. Because of their relatively high strength and toughness at high temperatures ($>1250^\circ\text{C}$), as well as their inherent corrosion resistance and substantially lower density, such components would contribute significantly toward improved efficiency, through their lower mass and their ability to withstand higher operating temperatures (requiring no air-cooling), and improved performance lifetime. Among the specific materials systems under

consideration are $\text{SiC}_{(f)}/\text{SiC}$ and $\text{Al}_2\text{O}_{3(f)}/\text{Al}_2\text{O}_3$. Because these materials are desired specifically for their thermal properties (e.g., thermal diffusivity, thermal conductivity), any variations in those properties will significantly affect their ability to transfer heat properly, thereby reducing or eliminating the very advantages for which these materials were chosen. Such variations can be caused by processing defects (e.g., improper infiltration, poor fiber distribution, etc.), service induced damage such as thermal shock, or thermally induced degradation of the fiber/matrix interface.

A nondestructive method which can quantitatively measure thermal properties is therefore critical to component reliability. While several such methods exist and have been used extensively in the study of both ceramic and nonceramic materials systems, they are used primarily for materials characterization in which the thermal properties are assumed uniform across a small test specimen¹⁻⁵. The critical issue in most CFCC applications, however, is the distribution of thermal properties within a given component. In these situations, thermal imaging methods are of greater utility. Because thermographic techniques are potentially full-field, they offer the ability to simultaneously and independently measure thermal properties over a two-dimensional distribution of locations. As a result, large-area specimens of nonplanar geometries can be readily inspected. Furthermore, because this detection and the small thermal excitation required can be accomplished radiometrically, requiring only "visual" access to the specimen, the method is inherently noncontact and can be performed at elevated temperatures. Although conventional thermographic imaging (i.e., surface temperature imaging) can often detect variations in thermal properties through a local variation in thermal response to a given excitation, it cannot quantitatively relate these variations to some thermal property and is often insensitive to small variations, particularly between components.

The work described here therefore applies a modified thermographic imaging technique to the quantitative measurement of thermal diffusivity of CFCC materials. While previous researchers have applied thermal diffusivity measurement to the inspection of small test specimens requiring high spatial resolution and long acquisition times⁶, the current work requires that measurements be made in the shortest time possible, while retaining at least 1 mm resolution. A complete thermal history is recorded for the area of interest from the time of thermal excitation of the specimen's front surface until the back-surface temperature reaches a maximum. From this temperature-time relationship, thermal diffusivity is calculated. By imaging at a higher resolution than required and then spatially averaging the resulting images before diffusivity computation, sufficient noise reduction is achieved in a single heating cycle while maintaining the required spatial resolution.

3. THERMAL IMAGING SYSTEM

The thermal imaging system used for this work is illustrated in Fig. 1. This system uses the method of Parker et al.⁷ to calculate thermal diffusivity. This method requires that a thermal pulse of short duration be incident on the front surface of a specimen and that the temperature of the back surface be monitored as a function of time. This is accomplished in the current method by heating the front surface with a 6 kJ photographic flash lamp (with a pulse duration of less than 8 ms) and monitoring the back-surface temperature with a commercially available scanning radiometer infrared camera. The camera was equipped with a 3-12 μm optical band-pass lens system and a liquid-nitrogen-cooled HgCdTe detector. Images were acquired by a Macintosh II with an on-board frame grabber capable of storing 128 images in real time at 512 x 512 pixel 8-bit resolution. Image acquisition (at 30 Hz) and analysis were performed with locally written software. To minimize lateral

heat flow, test specimens were mounted in diatomaceous earth. However, in order to determine the exact time of the thermal pulse, a small amount of the "pulse" was allowed to ambiently reach the back surface. This resulted in a photometric excitation of the detector at the moment of the flash. By determining the position of the scanning mirrors at the time of the flash (i.e., by determining which of the 512 image rows were excited by the flash) the exact flash time could be determined with submillisecond resolution.

Figure 1. Diagram of experimental arrangement used for thermal diffusivity imaging.

Figure 2 shows the theoretically predicted back-surface temperature T as a function of time t and specimen thickness L according to Parker et al.⁷ from the relationship

$$T(L, t) = \frac{Q}{\rho CL} \left[1 + 2 \sum_{n=1}^{\infty} (-1)^n \exp \left(\frac{-n^2 \pi^2}{L^2} \alpha t \right) \right], \quad (1)$$

where Q is the radiant energy incident on the front surface, ρ is density, C is specific heat, and α is the thermal diffusivity. Because the axes of Fig. 2 have been normalized (where T_M is the maximum back-surface temperature), the relationship is universal to all specimens. Two methods of determining α from the graph become evident. First, the more common method notes that when $V = 0.5$ (the back-surface temperature rise has reached half of its maximum), $\omega = 1.37$. Thus, using the "half-rise-time" method,

$$\alpha = \left(\frac{1.37 L^2}{\pi^2 t_{\frac{1}{2}}} \right). \quad (2)$$

As is evident from the plot of Fig. 2, any noise in the temperature signal can significantly alter the computed thermal diffusivity value. Because the noise level in scanning radiometers is notoriously high (as much as 10% for temperature variations of a few degrees Celsius in the current system), some method of noise reduction must be used. Previous researchers have performed a "cyclic averaging" procedure in which the specimen is heated and allowed to cool a number of times, and the time/temperature history for all such cycles is then averaged⁶. They have also employed a nonlinear least-squares estimation of the entire time/temperature relationship, which requires significant computation time.

Figure 2. Theoretical plot of normalized back-surface temperature as a function of normalized time.

To minimize the time needed to measure thermal diffusivity, and because the curve is nearly linear in the immediate vicinity of $V = 0.5$, a linear least-squares fit in that vicinity was used to interpolate the value of $t_{1/2}$ for the current work. Furthermore, by imaging the components at higher resolution than required, spatial averaging (at a size that could be specified for the particular application) was permissible without loss of sensitivity to the minimum defect size. For the results presented here, a 10 x 10 pixel block was used. Thus a 512 x 413 pixel digitized image was transformed into a 51 x 41 pixel reduced image with a substantially better signal-to-noise ratio. As an additional means of noise reduction, the quarter-rise time ($t_{1/4}$) and three-quarter-rise time ($t_{3/4}$) were also determined for each block, where $\omega(t_{1/4}) = 0.92$ and $\omega(t_{3/4}) = 2.08$, according to the extension of the Parker et al. procedure suggested in ASTM specification E 1461-92⁸. The thermal diffusivity values obtained at these locations were averaged with that of the $t_{1/2}$ value to determine the effective thermal diffusivity. This permitted determination of thermal diffusivity from a single thermal cycle, thereby reducing the total acquisition time.

The resulting thermal diffusivity values for all 10 x 10 pixel subsets were assembled into diffusivity images were stored with 16-bit resolution. This avoided the need to scale each image independently (for better comparison between images) while retaining flexibility for image enhancement in display (which was performed at 8-bit resolution).

Figure 3. Typical time-temperature plots obtained for graphite using the half-rise-time thermal diffusivity system.

Figure 4. Thermal diffusivity image of 19 mm thick NIST graphite specimen (25.4 mm diameter), with absolute diffusivity scale given at right.

4. EXPERIMENTAL RESULTS

4.1. Calibration specimens

The first specimens examined in this work were several cylinders of a NIST thermal properties calibration standard (8426 graphite) with a stated thermal diffusivity value of $72 \times 10^{-6} \text{ m}^2/\text{s}$. Three cylinders were cut from the 25.4-mm-diameter specimen with a diamond wafering saw, in nominal thicknesses of 4, 11, and 19 mm. Figure 3 shows typical time-temperature plots for the 11 and 19 mm specimens, from which thermal diffusivity values of 74 and $76 \times 10^{-6} \text{ m}^2/\text{s}$, respectively, were measured by the half-rise-time method and averaging over the center 1 cm^2 . The resulting thermal diffusivity image for the 19 mm specimen is shown in Fig. 4. Despite the noise in the region surrounding the graphite disk, thermal diffusivity clearly shows little variation across the face of the disk, indicating that the insulating mount was sufficient to ensure that lateral diffusion is indeed negligible over the time period used for these measurements. Furthermore, the standard deviation of thermal diffusivities measured in the center 1 cm^2 was $\pm 2 \times 10^{-6} \text{ m}^2/\text{s}$. Thus, the absolute value of the thermal diffusivity measured with this system deviated from the calibration value by $\approx 5\%$ or less (because of thermal losses and approximations in the determination of $t_{1/2}$), and the variation of that measured diffusivity was $<3\%$.

4.2. Thermally quenched specimens

To determine susceptibility to thermal shock damage, a Nextel fiber chemical vapor infiltration (CVI) SiC CFCC composite material was thermally analyzed. The composite was made using a 2-D plain weave, with a carbon interlayer between the fibers and the matrix, and was fabricated into flexure bars measuring $4 \times 6 \times 10 \text{ mm}$. As with the graphite calibration specimen, average thermal diffusivity was determined for the central region of each specimen.

Figure 5 shows the results of thermal diffusivity measurements as a function of both quench-temperature difference and number of quench cycles. Thermal diffusivity remains relatively constant up to a ΔT of $\approx 400^\circ\text{C}$. However, at a ΔT of 1000°C , a $\approx 40\%$ drop in diffusivity is evident. For comparison, an additional specimen was heated to the same temperature but allowed to air-cool; this specimen showed a drop in diffusivity of only 25% . Thus, the 40% drop in diffusivity seen in the 1000°C ΔT quenched specimen is only partially attributable to thermal shock damage, while the balance is likely caused by high-temperature degradation of the material itself (e.g., oxidation of the carbon interlayer, debonding of the fiber/matrix interface). Furthermore, Fig. 5b shows that there is also a 25% drop in diffusivity even at a lower ΔT of 600°C after four quench cycles.

Figure 5. Thermal diffusivity as a function of (a) quench ΔT and (b) number of quench cycles in thermal shock testing.

4.3. Seeded-defect specimens

The aforementioned applications of this thermal diffusivity imaging system are primarily those of materials characterization. While these applications use the full-field nature of the technique, they only require a single measurement from a given specimen and therefore do not take full advantage of the system's capability to determine the distribution of thermal diffusivities. In the examination actual components, the distribution of properties becomes critical, because what constitutes a potentially dangerous defect in one location might be an acceptable variation in another less-hazardous location. To examine the system's capability to distinguish defects of various sizes in an $\text{SiC}_{(f)}/\text{SiC}$ CFCC, several "seeded-defect" $\text{SiC}_{(f)}/\text{SiC}$ composite panels were made; a diagram of one of these panels is shown in Fig. 6. The panels were made from 12 layers of 2-D plain weave Nicalon fabric. The simulated defects were created by cutting out small sections of two inner plies and replacing the fabric with Graphoil sections. The weave was then infiltrated with SiC by CVI.

Figure 6. Schematic diagram of "seeded-defect" $\text{SiC}_{(n)}/\text{SiC}$ composite panels (all dimensions are in millimeters).

(a)

(b)

Figure 7. X-ray radiographs of "seeded-defect" $\text{SiC}_{(n)}/\text{SiC}$ composite panels: (a) 10% porosity and (b) 25% porosity.

A number of panels were created by varying several parameters (including Graphoil section thickness, defect size, fiber coatings, defect depth, and infiltration density), for a total of 32 panels; for brevity, only two such panels are presented here. Because the most likely defect type to be seen in the actual components evaluated for this work (and presented below) was poor infiltration, the two panels presented here were identical in every parameter except infiltration density. One panel was well-infiltrated to a remaining porosity of 10%, while the other was less infiltrated to a remaining porosity of 25%.

Figure 7 shows X-ray radiographs of both specimens, with defect locations clearly evident.

Figure 8 shows images generated for these two panels. Figures 8a and 8d are the raw thermal images taken at the point of maximum thermal contrast between the defect and nondefect regions, and Figs. 8b and 8e are the same images after

substantial image processing. While the defects are clearly evident in the processed images, the relative extent of "damage" is still unknown, and the image processing has required a trained user. Furthermore, direct quantitative comparison of the two images is difficult at best. Figures 8c and 8f are the thermal diffusivity images of the same specimens; not only is the contrast as good as or better than that in the processed thermal images, but the spatial resolution is slightly better and no user interaction was required. Best of all, because the thermal diffusivity images are quantitative, direct comparison is possible not only between the defect and nondefect regions (in which the defect region has a higher diffusivity), but between the two porosities as well.

Figure 8. Images of two $\text{SiC}_{(r)}/\text{SiC}$ composite panels. Images (a-c) are of a panel with 10% porosity, while images (d-f) are of a panel with 25% porosity. Images (a) and (d) are raw thermal images, (b) and (e) are processed thermal images, and (c) and (f) are thermal diffusivity images (with absolute diffusivity scales given at right).

Figure 9. Photograph of subscale 20 cm diameter SiC_f/SiC combustor liner.

4.4. Combustor liners

The final components inspected in this work were three subscale SiC_f/SiC CFCC turbine combustor liners produced by CVI of an angle interlock weave of Nicalon fibers. The cylindrical liner was approximately 20 cm in diameter, 20 cm long, and 3 mm thick. Figure 9 is a photograph of one such liner (although the sample pictured is actually twice as long as those inspected). For this component, the experimental configuration of Fig. 1 was slightly modified in that the flash lamp was placed inside the liner while the IR camera monitored the temperature of the outside surface. To reduce the effects of angular emissivity variation from the cylindrical surface, the outer surface was divided into 3 rows of 11 circumferential segments each, for a total of 33 segments, each of which was approximately 57 x 46 mm (30 mm on each end of the liner was not inspected in order to eliminate edge effects). By using aluminum foil inside the liner, light from the flash lamp was concentrated in an area (≈ 100 mm square) centered on but slightly larger than the segment of interest to prevent excessive lateral thermal diffusion from the region of interest. By containing the area exposed to the flash, the entire liner was not heated during each flash cycle. Thus, consecutive acquisitions of time/temperature profiles for nonadjacent segments were possible with no delay required for component cooling, thereby minimizing overall acquisition time.

Figure 10. (a) Conformally mapped thermal diffusivity image of subscale SiC_f/SiC combustor liner (where white = high diffusivity, black = low diffusivity) and (b) X-ray CT cross-section of same liner (where white = high density, black = low density) from location noted in (a).

Thermal diffusivity images for the individual segments were then assembled into a single conformally mapped composite image of the entire combustor liner. One such image is shown in Fig. 10a. The lines between segments are caused by tape used to mark the segment locations. One large area of low thermal diffusivity is seen on the left side of the image, while other areas of higher diffusivity are seen as vertical bands across the image. The bands of higher diffusivity correlated well to the locations of seams in the weave lay-up. However, the large region of low diffusivity was not related to any known geometrical condition of the liner. A density cross section was therefore obtained for that region (as indicated in the figure) with a microfocus X-ray CT system, and this image is shown in Fig. 10b. Clearly, the region of low thermal diffusivity correlates directly to a region of low density (high porosity) in the liner, the result of poor matrix infiltration.

5. CONCLUSIONS

A new thermographic technique provides "single-shot" measurement of the distribution of thermal diffusivity and is applicable to large components. The method is a rapid, noncontact, full-field technique that provides a quantitative measure of thermal properties. By applying digital image filtering, interpolation, and least-squares-estimation techniques for noise reduction, acquisition and analysis times of minutes or less can be achieved with submillimeter spatial resolution. Examination of calibration specimens has shown an absolute accuracy within 5% of tabulated values. The system has been used to examine the effects of thermal shock, oxidation treatment, density, and variations in fiber coating in a full array of test specimens. In addition, subscale SiC_f/SiC CFCC cylindrical gas turbine combustor liners have been inspected for manufacturing-induced defects (e.g., poor matrix infiltration, fiber ply overlap), and show strong correlation with established techniques in detecting these defects.

6. ACKNOWLEDGMENTS

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