THERMAL DIFFUSIVITY IMAGING OF CERAMIC COMPOSITES

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ABSTRACT

Strong, tough, high temperature ceramic matrix composites are currently being developed for application in advanced heat engines. One of the most promising of these new materials is a SiC fiber-reinforced silicon nitride ceramic matrix composite (SiCf/Si3N4). The interfacial shear strength in such composites is dependant on the integrity of the fiber's carbon coating at the fiber-matrix interface. The integrity of the carbon rich interface can be significantly reduced if the carbon is oxidized. Since the thermal diffusivity of the fiber is greater than that of the matrix material, the removal of carbon increases the contact resistance at the interface reducing the thermal diffusivity of the composite. Therefore thermal diffusivity images can be used to characterize the progression of carbon depletion and degradation of the composite. A new thermal imaging technique has been developed to provide rapid large area measurements of the thermal diffusivity perpendicular to the fiber direction in these composites. Results of diffusivity measurements will be presented for a series of SiCf/Si3N4 (reaction bonded silicon nitride) composite samples heat-treated under various conditions. Additionally, the ability of this technique to characterize damage in both ceramic and other high temperature composites will be shown.

INTRODUCTION

Ceramic composites are currently being developed for applications in advanced high temperature engines. These applications require a high integrity in the thermal properties for proper heat transfer as well as the mechanical properties. New nondestructive techniques are required for both thermal and mechanical characterization of these structures. These techniques are applicable both in the development stage of these composites as well as being appropriate for insuring the safety and integrity of the final system.

Possible candidates for inspection of these composites include thermographic techniques. These techniques give data which can be reduced to a direct measurement of the thermal properties of the structures. In addition, recent studies on SiC fiber-reinforced silicon nitride ceramic matrix composite (SiCf/Si3N4) have shown that a degradation of the fracture toughness correlates to a change...
in the thermal properties of the composite. It is suggested that this is a result of a degradation in the fiber matrix interface which is reflected in a reduction in the thermal diffusivity of the sample.

Also, thermographic techniques have some inherent characteristics which make them attractive for material characterization. The thermal excitation and temperature measurement can both be performed radiometrically, therefore no contact is required for the measurement. Procedures can also be developed for measurements at the operational temperatures of the composite. Since the measurements can be performed radiometrically, large area measurement are possible for non flat and parallel structures. Additionally, the relative stability of the solution to the heat flow equations simplifies the reduction and inversion of thermographic data.

Presented here is a thermal imaging technique developed to provide rapid large area measurements of the thermal diffusivity perpendicular to the fiber direction in these composites. A method is presented for reducing the thermal images to a diffusivity measurement. The method uses the complete time history of the surface temperature to calculate the diffusivity at each point on the sample. This technique produces diffusivity images with significantly improved signal to noise ratios compared to conventional data reduction schemes. Results are presented for a series of SiC/Si$_3$N$_4$ (reaction bonded silicon nitride) composite samples heat-treated under various conditions. These results are compared with previous experimental and analytical work in this field.

MEASUREMENT SYSTEM FOR DIFFUSIVITY IMAGING

A simple method for the determination of thermal diffusivity was first described in 1960 by Parker et al.[1]. This method uses a flash of thermal energy supplied to the front surface of the sample of a time duration which is short compared with the thermal transient time for the particular sample of interest. The thermal transient time is given by:

$$t_t = \frac{L^2}{4\alpha \pi^2}$$

where $L$ and $\alpha$ are the thickness and the thermal diffusivity of the material respectively.

In Parker's method the temperature as a function of time on the back surface of the sample was then measured with a point infrared radiometer.

The measurement system developed here also uses flash heating as the thermal excitation. Rather than measuring the temperature evolution at a single point the temperature is measured over the entire surface of the composite using a scanning radiometer. A block diagram of the experimental setup is shown in Figure 1.

This allows for a measurement of the through-the-thickness diffusivity of the sample with a single measurement. The results can be displayed as a grayscale or color diffusivity image where spatial variations in the measured diffusivity may indicate microstructural variations.

The scanning radiometer is a commercial infrared (IR) radiometer which uses a scanned HgCdTe (Mercury - Cadmium - Telluride) liquid nitrogen cooled detector. The radiometer's minimum detectable temperature difference, cited by the manufacturer, is 0.15°C when operating the detector in the 8 to 12 micrometer wavelength range. The radiometer produces images at approximately 30 per second (video frame rates) in an RS170 format compatible with standard video equipment. The field-of-view and the resolution element size were reduced by a factor of 3 with external optics, consisting of a 3x telescopic lens with four germanium optical elements.

The signal from the radiometer is digitized in 8-bits and stored,
in the lower byte of 16-bit buffers, at video frame rates in a commercially available real-time image processor with a personal computer as a controller. The image processor has 8 megabytes of image memory available for storage making it possible to acquire 16 images, each of which contains 512 x 512, 16-bit deep pixels. The image processor can also subsample the incoming video signal producing 64 images of 256 x 256 16-bit deep pixels each. The computer is also configured with an input/output card which permits acquisition and production of analog and digital signals under program control.

The data collection and analysis techniques were selected to take advantage of the ability of the image processor to perform 16-bit integer mathematics on the incoming video signal, at video frame rates. The 16-bit storage capacity allows for a number of 8-bit digitized images to be added together without exceeding the dynamic range of the image processor (note: No stretching of the data takes place during the data collection). An accumulation of the incoming video signal produces a "cyclic sum," which enhances the system's signal-to-noise ratio. A "cyclic sum" can be used when the incoming video signal can be repeated at some fixed frequency over the entire measurement time. In forming a "cyclic sum," data is collected for some fixed number of images (typically 32, approximately 1 second of data). The incoming data can then be repeated in the next collection cycle, the heat pulse is reapplied, and each newly digitized image is added to its corresponding image from the previous collection cycle. If sufficient time is allowed between each cycle there will be no significant long-time rise in the sample's temperature during the measurement.

The short duration pulse of heat required for the diffusivity measurement is supplied by commercially available photographic-type xenon flash lamps. The lamp output is 6.4 kilojoules and the flash duration is approximately 10 milliseconds which is much faster than the thermal transient time in the sample considered in this study. (For a sample of thickness 0.239 cm. and thermal diffusivity 0.026 cm²/s the thermal transient time as given by equation [1] equals 0.233 s.) Either one or more lamps can be used depending on sample size. The time of the flash is synchronized with the beginning of each data collection cycle via the input/output card of the computer controller.

FIGURE 3: THERMAL DIFFUSIVITY IMAGES OF REACTION BONDED SILICON NITRIDE SAMPLES. (a) BEFORE HEAT TREATING AND (b) FOLLOWING 100 HOURS AT 600°C IN AN O₂ FLOW.

0.05 cm²/s

Diffusivity

0.00 cm²/s

(a)

(b)
The resulting set of cyclicly averaged temperature images are stored first in image memory on the image processor then to hard disk on the personal computer controller. In the current configuration the data is transferred to another computer system for analysis. This enables the system to continue to acquire data from other samples. The reduction of the thermal data to diffusivity images is described in the next section.

REDUCTION OF THERMAL DATA TO DIFFUSIVITY IMAGES

Flash techniques typically involve a determination of the diffusivity of the sample from the time required for the temperature at the back surface to increase to a fraction of the maximum temperature obtained by the back surface of the sample. For example, Parker, et al. calculated the diffusivity from the time required, as measured beginning with the time of the input of the heat pulse, for the temperature of the back surface to achieve 1/2 of its maximum value ($T_{1/2}$). Therefore the noise in the measured temperature at or near $T_{1/2}$ can be a significant contribution to the variance in the measurement. For single point measure, where the detector is focus on a single point, the signal to noise ratio in the measurements is high and the noise level is not typically of concern. When the detector is scanned to perform an area measurement, the signal to noise ratio is often on the order of 10 to 1 and is of concern in thermal measurements.

An reduction in the variance can be achieved by incorporating the time history of the surface temperature in the determination of the diffusivity. A common technique for doing this is a nonlinear least squares estimation of the diffusivity based on the time history of the back surface temperature. This results in a significant decrease in the variance of the technique, since all of the data is incorporated in the calculation of the diffusivity. The temperature of the back surface for a sample of thickness $L$ is given by [1]:

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

for long times or

$$T(L, t) = \frac{2Q}{\rho c \sqrt{\pi \alpha t}} \left[ \sum_{n=0}^{\infty} \exp \left( -\frac{(n+1)L^2}{4\alpha t} \right) \right]$$

for short times, where $\rho$, $c$, $t$ and $\alpha$ are density, specific heat, time and diffusivity respectively. $Q$ is the amount of heat per unit area that is deposited on the front surface during the flash and $n$ is the summation index for the series expansion. This assumes uniform heating over the entire sample and no significant lateral heat flow during the measurement. This assumption is appropriate if the samples are thin relative to the lateral dimensions of interest.

A determination of the diffusivity at a given point is performed first by finding its time history from the time series of images of the back surface temperature collected after the input heat pulse. Two parameters ($a$ and $b$) given by:

$$a = \frac{Q}{\rho c \sqrt{\pi \alpha}}$$

and

$$b = \frac{\alpha}{L^2}$$

are varied to minimize the difference between the acquired data and equation [1] or [2]. For this work equation [2] was found to be adequate using only the first 10 terms in the summation. If the sample thickness ($L$) is known then equation [4] can be solved to yield the diffusivity perpendicular to the sample surface. This is
done point by point for the sample to produce a thermal diffusivity image for the sample.

EXPERIMENTAL RESULTS

A typical thermal diffusivity image is shown in figure 2. Shown in the figure are the thermal diffusivity images for two SiC/RBSN ceramic matrix composites, one which is as fabricated and the second has been thermal shocked. The advantage of the diffusivity imaging is clearly seen in this figure, where the damage caused by the thermal shock appears as variations in effective diffusivity. The image of thermal diffusivity not only shows the change in thermal properties but also gives an indication of the extent of the damage. The decrease in thermal diffusivity due to the thermal shock is typical of previously observed thermal damage in these samples. As a result of the thermal shock the thickness of the composite increased in the shock region, this effect was corrected for when determining the diffusivity.

To determine the sensitivity of the technique to variations in the thermal diffusivity of the material, the diffusivity of a sample was changed in a systematic way by heating the sample in flowing oxygen at 600°C for 100 hours. The difference in the diffusivity images for before and after heat treatment is shown for two reaction bonded silicon nitride samples (Si3N4 / SCS-6). The first sample (Figure 3a) is without heat treatment and has an interfacial shear strength(σ), determined from fiber push out tests, of 18.0 MPa. The second sample (Figure 3b) was heat treated in an O2 flow at 600°C for 100 hours which resulted in a reduction of the interfacial shear strength to 0.8 MPa. The interfacial shear strength in such composites is reduced due to the degradation of the fiber-matrix interface[3]. The oxidation of the carbon at the interface during heat treatment also results in a greater thermal contact resistance between the fiber and the matrix thus reducing the thermal diffusivity of the material[2]. This change in diffusivity can be seen in the quantitative results produced by this measurement technique.
The variability of the measurement was determined with a series of 30 measurements made on a single ceramic composite sample over the course of several days. The average diffusivity was 0.025 cm$^2$/s with a standard deviation in the measurements of 0.0007 cm$^2$/s or approximately 3%. This indicates the high degree of measurement reproducibility for this technique.

The sample to sample variation was investigated with a series of 20 samples from 3 batches of material. Physical dimensions of the samples were 7.5cm in length, 1.2cm. in width and 0.21cm. in thickness. Materials from each of the batches were then heat treated at 3 different temperatures (600°C, 1000°C and 1400°C) for either 1 hour or 10 hours duration. Two of the samples were left as fabricated, with no heat treatment. Figure 4 is a representative collection of thermal diffusivity images from these samples, variations in the thermal diffusivity due to the different heat treatment parameters can be seen in the images. Figure 5 indicates the dependence of the average diffusivity over the entire sample with the heat treatment temperature for both 1 hour (Figure 5a) and 10 hour (Figure 5b) treatment times.

As can be seen from Figure 5, there is little variability in the initial values measured for the diffusivity of the sample. This indicates the techniques reproducibility as well as the fabrication reproducibility. Further the reduction in thermal diffusivity is in agreement with previous measurements[2]. The sample to sample variation following oxidation therefore is real and it is expected to be a function of the ability of the oxygen to penetrate into the individual samples.

CONCLUSIONS

A thermographic technique has been presented which show potential of characterization of the thermal and mechanical properties of ceramic composites. The technique presented provides a noncontacting, rapid, large area, quantitative measurement of the thermal diffusivity for ceramic composites as well a host of other materials. The technique uses the entire time history of the temperature at the back surface of a sample to estimate the thermal diffusivity of the sample at a given point. The result gives a signif-
significantly increase in the accuracy of the thermal diffusivity over conventional techniques. While it is presented here as a two-sided measurement, it can potentially be performed from one side of the sample.

The technique has also been shown to be effective in characterizing changes in material properties due to heat treatment, thus displaying the potential for a nondestructive technique to measure the interfacial shear strength in ceramic composites. Also the ability to characterize diffusivity changes due to thermal shocking is of great interest in applications of these composites such as heat engines.

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REFERENCES


[3]. R. T. Bhatt, 1990, "Influence of Interfacial Shear Strength on
FIGURE 5: CHANGES IN THERMAL DIFFUSIVITY WITH HEAT TREATING TEMPERATURES CAN BE SEEN FOR BOTH (a) 1 HOUR AND (b) 10 HOUR HEAT TREATMENT TIMES.