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Reliability of Laser Flash Thermal Diffusivity Measurements of the Thermal Barrier Coatings

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The thermal diffusivity of free standing thermal barrier coatings (TBCs) was measured by the laser flash technique. The combination of low thermal conductivity (1 to 2 W/m K) and small TBC thickness (300 to 600 μm thick) can cause errors in the measurements. Back surface (opposite the laser) temperatures of free standing plasma-sprayed TBCs were measured as a function of time and laser power. The front surface temperatures were calculated using thermal transport equations. In the high power region, thermal diffusivity decreased significantly with increasing laser power. In the moderate power region, thermal diffusivity remained constant. In the low power region, measurement became unreliable because of noise. The detector nonlinearity was believed to be a possible cause of deviation in the high power region. Measurements at different laser power levels should be conducted in order to obtain reliable thermal diffusivity values for TBCs.

Keywords TBC, thermal diffusivity, thermal conductivity, laser flash

1. Introduction

Thermal conductivity data of thermal barrier coatings (TBCs) are used by engine designers to calculate the thicknesses needed to protect the metallic components and ultimately to determine the maximum temperature to which the TBC coated superalloy can be exposed. Thus, obtaining reliable and accurate thermal conductivity data for the TBCs is important to optimize design and to assure the safe operation of turbine engines. The laser flash technique[1,2,3] is often used to measure thermal diffusivity of TBCs because of its rapid measurement and high-temperature capability. Using the measured thermal diffusivity and knowing the specific heat and the density of the coating, thermal conductivity is calculated as the product of these three parameters. However, thermal diffusivity of TBCs presents more problems than specimens such as superalloys or silicon nitride. From the material point of view, sintering of the ceramic will occur at the application temperature. The hysteresis behavior of thermal diffusivity upon heating (aging) has been shown to be very significant for as-sprayed coatings.[4] Thermal diffusivity will also change as a function of gas pressure and gas content of the pores in the TBCs. In addition, the unique features of TBCs such as small thickness, low thermal conductivity, and translucency in the infrared region also cause problems in thermal diffusivity measurements. This paper addresses the accuracy and reliability of thermal diffusivity measurements of TBCs.

Regular thermal diffusivity measurement requires the specimen to be opaque to both wavelengths of the laser pulse (1.06 μm) and the infrared detector (3 to 5 μm). The basic assumptions for the thermal diffusivity calculation are (1) all the laser energy is absorbed at the front surface and (2) the IR (infrared) detector only measures thermal radiation of the rear surface. The TBC materials are typically translucent; therefore, the conventional treatment is to put opaque coatings on both sides (graphite or metallic films such as Au or Au/Pd). The Au or Au/Pd coatings are only good up to about 900 °C; however, typical TBC application requires data up to 1400 °C. The high-temperature measurement requires some special sample preparation such as thick graphite coatings (four to five passes) or Pt coatings.

Even with opaque coatings, TBCs have other characteristics that make the diffusivity measurements difficult. Free-standing coatings are frequently used for thermal diffusivity measurement. Air plasma sprayed (APS) and electron beam-physical vapor deposited (EB-PVD) TBCs are typically 300 μm thick or thinner. The TBCs also have very low thermal conductivities.

Under these conditions, the front and rear sample surface temperatures will increase much more than regular specimens after the laser pulse than a thicker sample with moderate thermal conductivity. The excessive temperature increase could drive the IR detector into a nonlinear temperature-voltage response and thereby cause significant errors in the measured diffusivity values. Laser power levels should be large enough to obtain good signal-to-noise ratios, yet not result in excessive rear surface temperature rises (*i.e.*, $\Delta T > 5$ °C).

Parameters such as laser power, detector linearity, and stability and coating opacity are discussed in this paper. The purpose of this work is to point out potential sources of error during laser flash thermal diffusivity measurement and to recommend ways to overcome these errors.

2. Experimental

A laser flash system, Flashline 5000™, manufactured by the Anter Co. (Pittsburgh, PA), was used in this study. A description of this system can be found in Refs. 5. A room-temperature measurement station was added to the system, which permits use of the same IR detector and laser. To measure the rear surface temperature of the TBC, a K-type thermocouple was attached to the surface. The maximum temperature was recorded after the laser pulse. The thermocouple wire was thin and flexible and did not

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affect the sample holder being pushed down to the test position. The “ΔT” mode in the thermometer was used, the laser was fired manually, and the peak temperatures were recorded.

An InSb IR detector nonlinearity test was carried out on the room-temperature station of the Anter system. As shown in Fig, 1, an aluminum can filled with water was used as the specimen. The bottom of the can was sprayed with graphite to make the surface emissivity similar to that of most specimens. The can was half filled with hot water. The water temperature cooled down slowly to room temperature. For the room temperature detector stability test, the aluminum can was left overnight to achieve equilibrium. Thermal insulation was placed between the can and the base to avoid temperature shift due to thermal transport into the base. The IR detector received the thermal signal of the target *via* a 45° Au-coated mirror. The detector linearity tests were performed as a function of water temperature. The detector stability tests were performed as a function of time.

The TBC samples were provided by GE Aircraft Engine (Cincinnati, OH) and Siemens Westinghouse Power Generation (Orlando, FL). The GE specimen was a plasma-sprayed ytrria-stabilized zirconia free-standing coating. The Siemens Westinghouse specimen was also a plasma-sprayed TBC, which had been thermally aged. All the samples were spray coated with graphite on both sides. The thermal history of each sample was carefully recorded so the effect of aging could be taken into account.

3. Results and Discussion

3.1 Surface Temperatures on a TBC

According to Parker *et al.*[1] assumptions for laser flash measurements, the material (at least the surface) should be opaque, and only a small depth t' should absorb the energy. His requirement for the temperature as a function of depth x at time zero and heat pulse Q is

$$T(x,0) = Q/\rho c t' \qquad \text{for} \qquad 0 < x < t' \qquad \text{(Eq 1)}$$

$$T(x,0) = 0 \qquad \text{for} \qquad t' < x < L \qquad \text{(Eq 2)}$$

where ρ is the density in g/cm^3, L is thickness in cm, and c is heat capacity in J/g °C. The front surface temperature should increase instantaneously and reach a much higher value than the rear surface. After the heat pulse, heat is conducted away from the front surface toward the rear surface. After the heat pulse, heat is conducted away from the front surface toward the rear surface. The increase in front surface temperature, ΔT_f, must decrease until it reaches $\Delta T_{L\max}$. Parker *et al.*[1] gave an approximation of the change of front surface temperature:

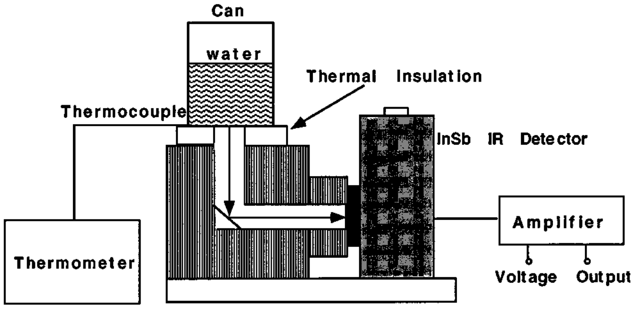

Fig. 1 Experimental setup for IR detector nonlinearity and stability tests

$$\Delta T_f = 38 L \Delta T_{L\max}\, \alpha^{-0.5} \qquad \text{(Eq 3)}$$

where α is thermal diffusivity and the maximum temperature rise at the rear surface is

$$\Delta T_{L\max} = Q/\rho c L = \Delta T \qquad \text{(Eq 4)}$$

The TBCs have very low thermal conductivities and can be an extreme case for surface temperature estimation. Assuming the $\Delta T_{L\max}$ (ΔT) is 2 °C, the thickness is 0.06 cm, and a = 0.008 cm^2/s, then, by using Eq 3, the front surface temperature, ΔT_f, of the TBC is calculated to be 51 °C. For a different rear surface temperature, ΔT, the front surface temperature of this particular TBC can be calculated as

$$\Delta T_f = 25.5\, \Delta T \qquad \text{(Eq 5)}$$

for $\Delta T = 3$ °C, $\Delta T_f = 76$ °C; for $\Delta T = 4$°C, $\Delta T_f = 102$ °C; and for $\Delta T = 5$ °C, $\Delta T_f = 128$ °C. Thus, the front surface can get extremely hot. One way to measure this effect is to attach a thermocouple to the rear surface of the sample and measure the maximum temperature rise. It should be pointed out that the very high front surface temperature only exists for a very short period of time. Since the heat pulse diffuses into the material very quickly, the rear surface temperature increase is only a small fraction of this instantaneous front surface temperature. Thus, varying laser power is a better way to see the temperature effect.

3.2 Rear Surface Temperature after the Laser Pulse

The Anter laser flash system uses five capacitors in parallel to deliver the power. Each capacitor can be charged from 1200 to 2800 V. Laser power was changed by charging the capacitors to different voltages and using an increasing number of capacitors (from one to five). The measured rear surface temperature as a function of laser power is shown in Fig. 2. Assuming $\Delta T = 5$ °C is a limit for a TBC, using maximum capacitor voltage (2800 V) is safe for one and two capacitors. When three and more capacitors were used, with $V > 1800$ V an excessive temperature rise was observed, up to 19 °C for five capacitors and 2800 V. When five capacitors are used, the charging voltage has to be less than 1800 V in order to keep $\Delta T < 5$ °C. It should be noted that each laser flash system is different in design and independent tests should be carried out for each system.

To test the detector linearity, the aluminum can was filled with hot water at about 50 °C. The temperature of the can and output voltage of the IR detector were recorded while the water was cooling. Data were recorded every 0.5 °C down to a few degrees above room temperature. Ice water was then added for lower temperature measurements. The results, shown in Fig. 3, indicated that over the 22 to 48 °C temperature range, the detector output versus temperature was nonlinear. Three small linear regions can be found in the curve with two obvious changes of

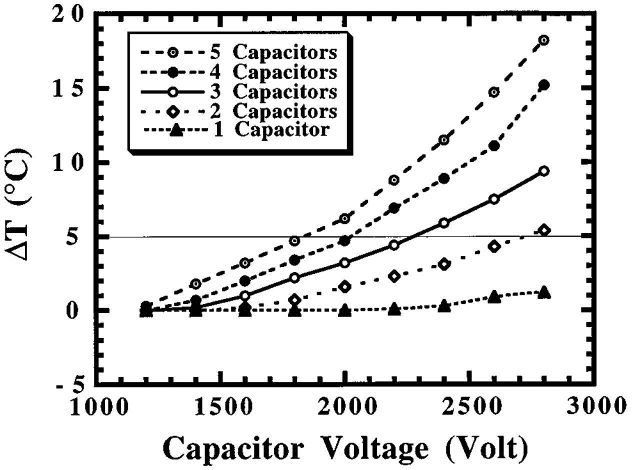

Fig. 2 Rear surface temperature rise as a function of laser power (laser power is a function of capacitor voltage and number of capacitors)

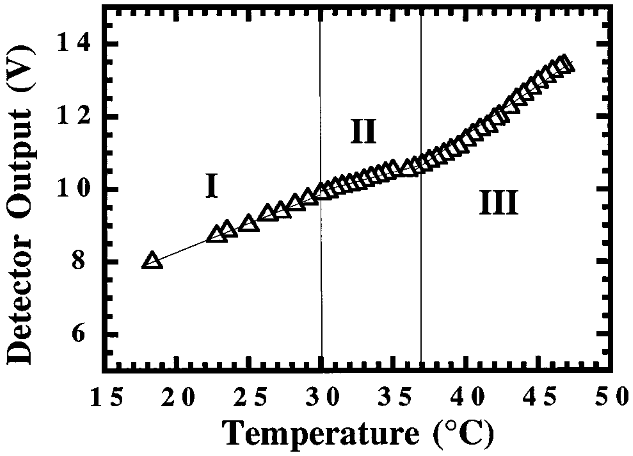

Fig. 3 IR detector output as a function of temperature. Three linear regions can be found with obvious changes of slope at 30 and 37 °C.

slope occurring at 30 and 37 °C. This problem has been reported in the literature[6] and can result in an overestimation of the half rise time for thermal diffusivity measurement and, therefore, give a lower diffusivity value. A correction for detector nonlinearity should be used if the temperature change at the rear surface is too large. However, if the temperature change is kept within 5 °C, the detector output can be regarded as a linear function of temperature. Another reason for keeping the temperature change below 5 °C is to assure the sample temperature stays close to the assigned measurement temperature. The thermal diffusivity obtained under this condition should be considered accurate.

The IR detector used in the thermal diffusivity test usually operates at liquid nitrogen temperature. Detector output after filling in liquid nitrogen was monitored over a long period of time. The liquid nitrogen (0.15 L) can last at least 12 h. Significant noise can be observed when the liquid nitrogen is used up and the detector is warming up. It was also noticed during several tests that the detector response is also dependent on when the test was carried out. The detector response curves obtained for experiments started a few minutes after filling with liquid nitrogen, in the middle of the day, and at the end of the day exhibited quite different curves. This indicates that the detector is not stable over time, and especially so when following filling with liquid nitrogen.

An IR detector response test was carried out immediately after filling in liquid nitrogen into a warm detector. The sample was kept at a constant temperature of 20.7 °C for 4 h before the test to eliminate possible temperature drift caused by the environment. As shown in Fig. 4, the detector output dropped very fast in the first 25 min and then started to recover exponentially. It took the detector nearly 60 min to reach a steady state. Small voltage drifts were still detectable after 100 min. If thermal diffusivity measurements are carried out too soon after filling a warm detector, significant error will occur because the IR detector has not reached equilibrium. The same effect was observed when a cold detector was being refilled with liquid nitrogen. For a cold detector, *i.e.*, an InSb detector manufactured by the Electro-Optical System Inc. (Phoenixville, PA), it will take close to 60 min to reach steady state.

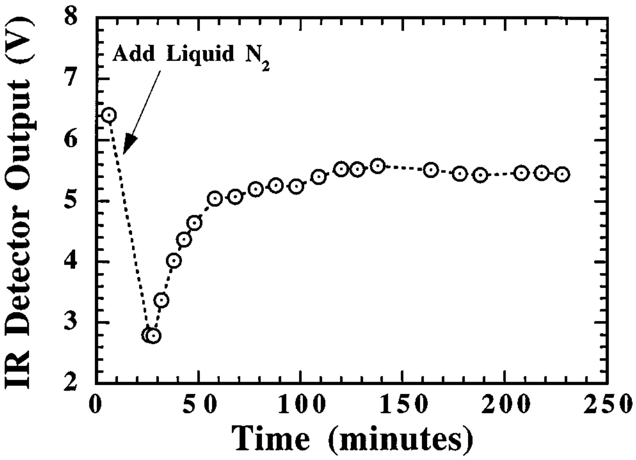

Fig. 4 Detector output as a function of time after adding liquid nitrogen. The sample was at constant temperature.

3.3 Thermal Diffusivity as a Function of Laser Power

Thermal diffusivity measurements of an APS specimen (Al_2O_3 based) were conducted at 110 °C using 12 different power levels. The results are shown in Fig. 5. Thermal diffusivity values were about 10% lower when full power was used. The diffusivity values increased when the laser power decreased. Diffusivity reached a stable value of 0.0174 cm^2/s when the laser power was close to 1800 V (capacitor voltage) with five capacitors. This result agreed well with the discussions of detector nonlinearity and rear surface temperature limit. Similar thermal diffusivity versus laser power curves were observed by Hasselman on graphite and AIN.[6] Ideally, the lowest laser power should be used. However, at low power, the signal becomes very noisy and this makes the measured values unreliable. Therefore, only a few data points were recorded in the stable region. Our recommendation is to use the lowest power possible while keeping the signal-to-noise ratio high. This often requires a scan over the available laser power range. This practice is very important to determine what is a correct power level

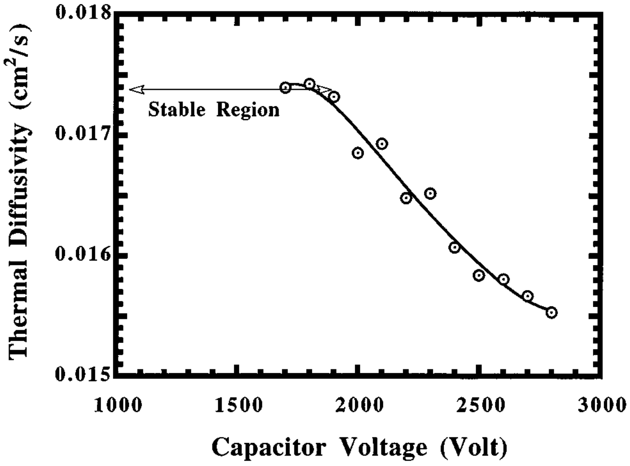

Fig. 5 Thermal diffusivity as a function of laser power (capacitor voltage) at 110 °C

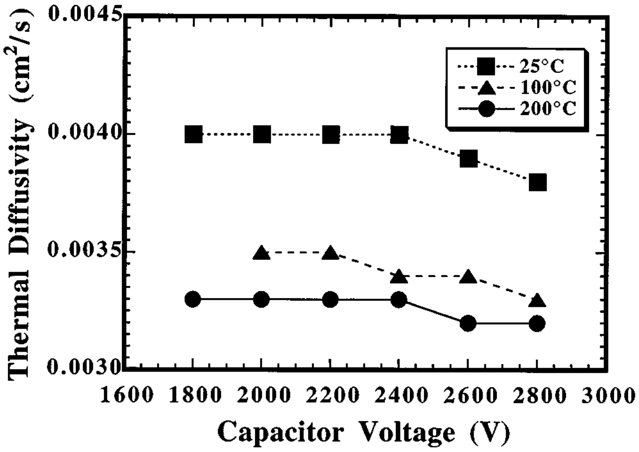

Fig. 6 Thermal diffusivity of a TBC as a function of laser power (capacitor voltage) at different temperatures

to use.

Similar tests were repeated on a plasma-sprayed YSZ TBC. Figure 6 shows the thermal diffusivity as a function of laser power and temperature of test environment. From room temperature to 200 °C, laser power levels below 2200 V (capacitor voltage) were found to yield consistent thermal diffusivity values, *i.e.*, variations less than 2%. It was also found that the detector nonlinearity problem became less serious at higher specimen temperatures.

3.4 Thermal Diffusivity of an APS TBC

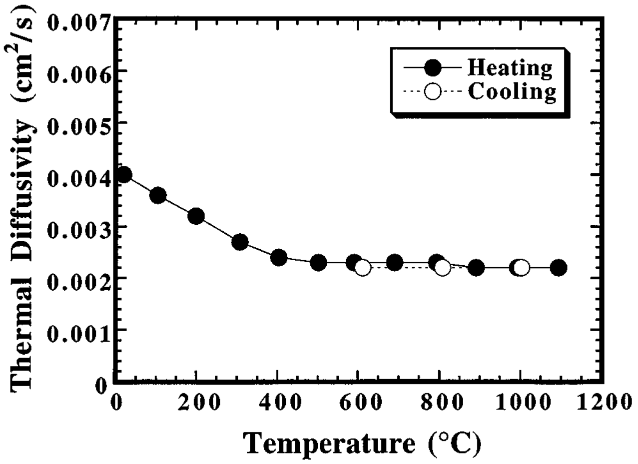

Fig. 7 Thermal diffusivity of an APS TBC during heating and cooling

An APS TBC specimen was tested from room temperature up to 1100 °C in nitrogen under slightly positive pressure. It was found that thick graphite coatings (*i.e.*, five passes of spraying from an aerosol can about 1 foot away) on both surfaces were sufficient to prevent light penetration up to 1100 °C. Typical thickness of graphite was on the order of a few microns. The specimen was placed in front of an intense light source to check for possible light penetration. The effect of laser power was tested on this specimen and adequate laser power levels at each temperature were used in the thermal diffusivity test. The thermal diffusivity versus temperature curve is shown in Fig. 7. This particular sample had been exposed to high temperatures for an aging test; therefore, no further increase of thermal diffusivity (hysteresis effect) was observed during the test. Thermal diffusivity measurements on this specimen showed very good reproducibility. The thermal diffusivity values are consistent with results in the literature.[4,7–11] This test result brought us back to the focus of this paper: *i.e.*, the reliability of laser flash thermal diffusivity tests. Although there are difficulties presented by TBCs, reliable measurements can be made when factors such as laser power level, detector linearity, opacity, and thermal history of the specimen are taken into account.

4. Conclusions

This study has shown that the rear and front surface temperatures of a TBC specimen during a laser flash thermal diffusivity test can be very high because of the combination of small thickness and very low thermal conductivity. The IR detector has shown nonlinearity over a temperature range of RT to 48 °C. It can cause the measured thermal diffusivity to be 10% lower than the real value. To stay within the linear range of the IR detector and to avoid overheating the front surface, care must be taken to keep the rear surface temperature below 5 °C. It was also shown that a cold IR detector needs about 60 min to reach equilibrium after adding liquid nitrogen and even longer time if the detector is warm. However, reliable thermal diffusivity values of TBCs can be obtained when cautions are exercised during the laser flash tests.

Acknowledgments

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