High-Temperature Thermal Properties Using Reflected Sample Generated Radiation

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ABSTRACT

A method for determining high-temperature thermal properties of thick solid materials involves short-term radiation pulses (20–200 s) on the front surface of a sample via a light guide, such as a quartz or sapphire rod. The magnitude of the flux absorbed must be accurately known for thermal conductivity, but not for thermal diffusivity. Power sources may include a lamp or laser but each has difficulties. For example, a lamp often has too little power and needs complex optics, and a laser beam may have excessive power that also lacks control. A heated sample transmits considerable radiation that varies with temperature and distance from the end of the light guide. This article explores the use of reflected radiation from a sample above about 700 K. Variables include geometry, type of light guide, and sample emissivity. Work with molybdenum and alumina samples has demonstrated the technique.

Keywords: thermal conductivity, diffusivity, front surface heating, light guide, reflection, radiation.

1. INTRODUCTION

Thermal diffusivity and conductivity can be measured by subjecting one surface to a radiation pulse and measuring the sample response, such as an induced thermal gradient. This is similar to the conventional laser flash method (ASTM 1461, 1992) and is adoptable to simultaneous use with a dilatometer (Wolff, 2013). The applied radiation pulse must be at least partially absorbed and of sufficient magnitude to induce a measurable front surface temperature change and/or a substantial gradient in the sample. Sources of such radiation pulses can include flash lamps, pulsed lasers, and many powerful light sources. At high temperatures (viz, above 500°C), the sample radiation may easily exceed the pulsed radiation level. If the reflected and absorbed radiation fluxes can be quantified, they can be used to analyze the sample response and lead to a value of its thermal conductivity. Requirements can be readily met by using the reflected radiation from the sample itself.

The thermal gradients can be measured with thermocouples such as type K (Ni based) or type C (W-Re based). The power levels of the radiation source must be variable and controllable to accommodate a wide range of materials and their emissivities.

2. THEORY

In the laser-flash method, the rear face temperature is measured when the front face is subjected to a radiation pulse. The front surface ($x = 0$)

emissivity/absorptivity is made near unity (via a thin carbonaceous coating) to maximize the S/N ratio. Similarly, in the PMIC XDC instrument (Wolff, 2013), both front and rear temperatures are measured, and the results interpreted from theoretical models are derived by (Carslaw & Jaeger, 1959; Martin & Blanco Muriel, 1989; Plummer et al., 1962):

Laser Flash

$$\chi = 1.37 \, (L/\pi)^2/t_{1/2} \tag{1}$$

$$\lambda = \chi \rho C_p \tag{2}$$

Semi-infinite model for diffusivity χ-

$$\Delta T(x = L)/\Delta T(x = 0) = \sqrt{\pi}\, \mathrm{ierfc}(L/(4\chi t))^{0.5} \tag{3}$$

$$\lambda = 2\varepsilon F \sqrt{(\chi \Delta t/\pi)}/\Delta T_{\mathrm{front}} \tag{4}$$

Finite length model

$$\chi = L^2/(6t_r) \text{ at } x = 0 \tag{5}$$

$$\lambda = \varepsilon FL/2(\Delta T_{\mathrm{front}} - \Delta T_{\mathrm{rear}}) \tag{6}$$

χ is the diffusivity (cm²/s), λ is the thermal conductivity (W/cm K), ε is the emissivity, t_r is the relaxation time, or time when rear surface heating rate extrapolates to $T = T_0$; $t_{1/2}$ is the time for rear surface to reach half its maximum value, T is the temperature (K), L is the sample length, F is the flux (W/cm²), C_p is the specific

heat, and ρ is the density. Note that for diffusivity data, it is not necessary to know the absorbed flux level.

A parametric analysis to determine the required flux levels may assume that we wish to measure a front temperature change of ≥5°C in ≤200 s (to minimize possible sources of error such as system temperature drifts, vibrations, instabilities, etc.). We can deduce that for metals a relatively high flux is needed. Assuming a surface emissivity of 0.8, a typical high temperature metal such as Mo will require a flux on the order of 5–10 W/cm². Plastics need only about 0.02 W/cm², and for ceramics, the flux can be intermediate, about 1 W/cm². Clearly, the surface emissivity should be known and as high as possible (e.g., C-coating).

2.1 Objectives

The theory (Equations (3) and (4)) suggests that a continuous known product of εF from 0.1 to at least 20 W/cm² be available, which can be imposed on the sample at any temperature. Theoretically, a powerful lamp with appropriate optics could be adequate but a simpler solution is to reflect the sample radiation on to itself. Reflected radiation will depend primarily on the system geometry. We note that radiation absorption depends on the emissivity (Martin, 1989) of the sample, the light guide, and the window to contain a vacuum or inert atmosphere. These are dependent of the source temperature. The objective here is to calibrate the received flux in order to utilize Equations (3) and (4).

3.   EXPERIMENTAL

3.1 How is the flux applied?

Figure 1 shows the system used to calibrate the flux from reflected radiation. X represents the total distance from the end of the light guide (LG) to either (a) a small flux meter (SFM), (b) a lamp or laser (L), or (c) the front of a first surface mirror (M). A constant flux F is desired for 20–200 s during Δt term in Equation (4). QW represents a quartz window sealed by a silicone high temperature rubber (R) at the end of a quartz tube (QT) serving as a vacuum
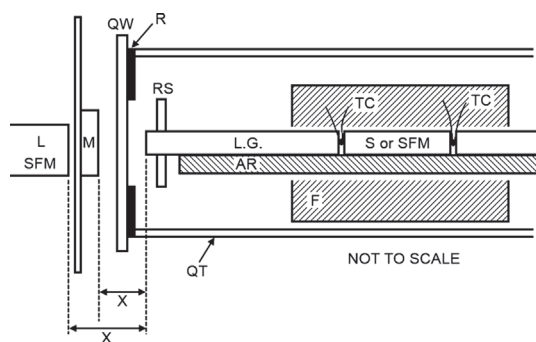
Figure 1. Schematic of system to calibrate flux from reflected sample radiation.
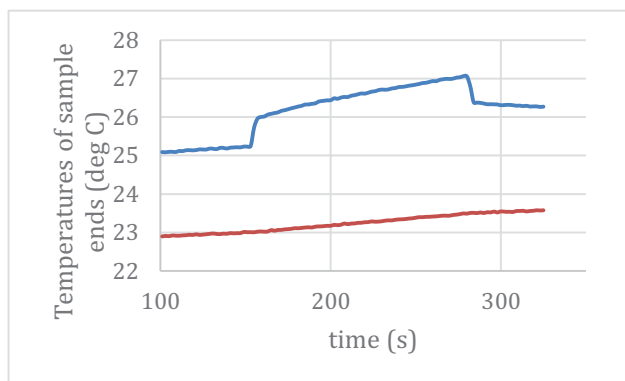
Figure 2. Molybdenum sample subjected to 120 s lamp illumination ($F = 0.6$ W/cm²) via a sapphire light rod.

container. S is the test sample, TC is a thermocouple junction, and F is a Nichrome wire tube furnace. A larger furnace may also be used to surround the QT vacuum chamber. A graphite tube RF induction heater susceptor surrounded by graphite wool may also be used above 1200°C. The light guide is a 10 mm diameter by 150 mm long quartz or sapphire rod. It should withstand the high temperatures required and transmit the flux with minimal absorption. One end of the LG is placed about 10 mm behind the QW. The other end is held against the 10 mm diameter sample separated only by a small thermocouple and possibly a thin graphite coating to adjust the sample emissivity. The sample and the light guide rod are supported by alumina tubes (AR). The separation (X) is preferred to the use of filters to control the reflected flux since the latter get hot and thus add to the flux recorded.

3.2 What is the response of the sample when a lamp is used?

Figure 2 shows the temperatures of the two thermocouples as a function of time for a 21-mm long Mo sample when a lamp is turned on. After the initial front surface temperature jump, it rose about 1.1°C in 120 s. Using Equation (4) and taking the diffusivity of Mo as 0.55 cm²/s, the emissivity as 0.3, and the flux as 0.55 W/cm², the conductivity calculates to the expected 1.4 W/cm-K value at about 300 K. The rear surface temperature is shown in the lower curve.

3.3 What happens if we raise the average temperature and use a lamp?

Figure 3 shows the sample cooling curve with the lamp ($F = 0.7$ W/cm²) turned on at $t = 1150$ s and off at $t = 1192$ s. Linear extrapolation of the cooling curve during this time interval shows the curve has been raised about 5° (ΔT). For alumina, $\chi = 0.015$ cm²/s at 1300 K, $\varepsilon = 0.25$ at 1200 K (Martin, 1989), $L = 2.5$ cm, and Equation (4) predicts $\lambda = 0.055$ W/cm² assuming the rear temperature went up a degree (based on other
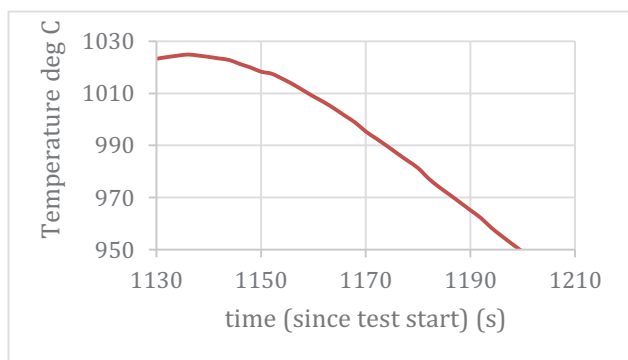
Figure 3. Front surface of a cooling alumina sample temperature vs. time. Lamp flux via sapphire rod on at $t = 1150$ s, off at 1192 s.

Figure 4. Theoretical flux available from a hot sample.

Figure 5. Measurement of flux at 40 mm (X) in front of the light guide end as a function of temperature of an uncoated alumina sample.

measurements). This is compared to Thermophysical Properties Research Center value (Touloukian et al., 1970) of about 0.06 W/cm². Conclusions from similar tests lead to a demand for more applied flux when a conductivity measurement is needed. This would solve several problems;

(a)   a generally higher S/N ratio

(b)   A clearer signal when sample is rapidly cooling or heating

(c)   A continuously variable source of flux is needed, e.g. 0.5–10 W/cm²

3.4 How much flux is available to be reflected?

The total flux available to be reflected from a hot body starts with integrating the Planck spectral emissive power over the spectrum:

$$F = C\varepsilon\sigma\left(T^4 - T_0^4\right) \tag{7}$$

where σ is the Stefan–Boltzmann constant, 5.6704×10^{-12} W/cm² T⁴. C is a view factor here restricted by the fluxmeter (Heat Flow Sensors) to a function of X, the distance from the end of the light guide and through the quartz window. It also depends on the material of the light guide, whether quartz or sapphire. Figure 4 shows the theoretical flux available as a function of sample temperature. Since the sample end has the same area as the light guide cross section, this would be emitted through the light guide end close to the quartz window. Measurements of this flux with the flux sensor showed slight (<0.2 W/cm²) absorption by the light guide and the window. For example, Figure 5 shows flux sensor output at 40 mm in front of the light guide end. The closer the sensor is to the window (smaller X), the greater the flux recorded. Replacing the heat flux sensor with a first surface mirror causes reflection of the flux. Its absorption by the sample is again reduced by its emissivity, the light guide material and geometry. Note that the emissivity of the alumina sample changes from about 0.2 to over 0.6 as it cools (Martin, 1989).
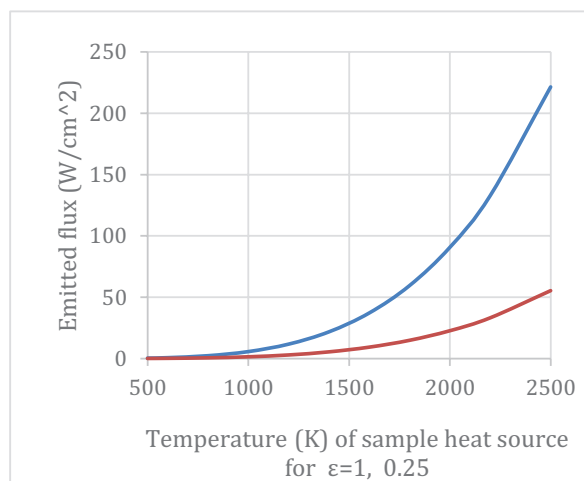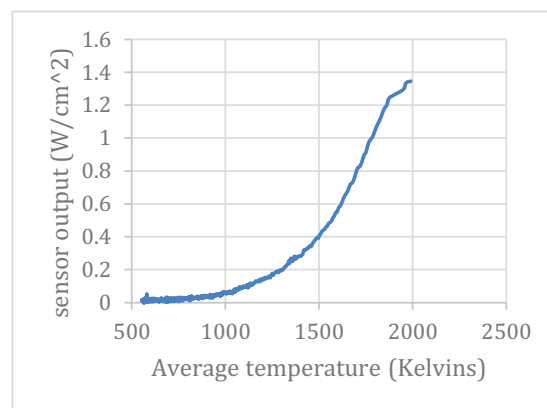
3.5 How do we reflect?

The calibration to determine an exact F value for Equations (3), (4), (6), etc., tends to greater accuracy at higher temperatures. For example, although the window heats up as the sample does, the transmitted radiation increases more rapidly than the window temperature. Further calibrations were made to quantify radiation losses due to the mirror, and quartz window compared to sapphire rod transmission.

Insertion of a small thermocouple junction between the light guide and the sample end also causes small variations of the flux emitted by the sample. Studies with different mirrors showed that first surface aluminized glass surfaces are best; others, such as polished Si wafers, have lower reflectivity. Curved mirrors show promise for reflectivity improvements.

3.6 Results with reflected flux to calculate diffusivity and conductivity

Figure 6 shows the effect of reflected radiation on an alumina sample. The mirror was placed at $X = 11$ mm
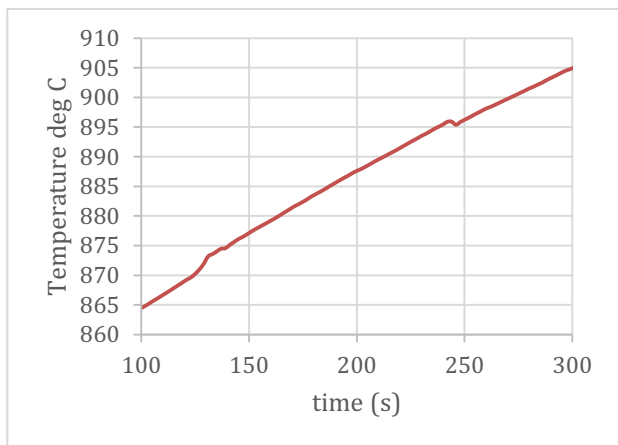
Figure 6. Front surface of alumina sample when exposed to reflected radiation at $t = 130$ s for 111 s.

in front of the light guide end (1 mm in front of quartz window). The flux was previously measured here at 0.3 W/cm² when the sample was 850°C. Thus, with $\varepsilon = 0.22$ and the indicated $\Delta T = 1.5$°C, Equation (4) predicts $\lambda = 0.06$ W/cm² K, as expected. An effective way to designate the start and finish of a flux pulse with either a lamp or a mirror reflection is to plot the instantaneous change in temperature dT/dt against the actual sample temperature. Pyroceram based standard reference materials were tested to about 1200 K, while Mo and alumina samples were tested to over 2000 K.

4.   CONCLUSIONS

Reflected radiation from a hot sample was shown to be a useful flux source for measurements of thermal diffusivity and conductivity. First surface mirrors can be calibrated with flux sensors to give accurate flux pulses on sample surfaces. An advantage of high flux pulses is to keep Δt short, thus minimizing effects of chemical reactions leading to surface contamination and possible unwanted changes in sample emissivity (Foster et al., 1956). The shorter the pulse, the more accurate is the average temperature for the property of interest. Figure 6 is typical of most materials and

can be done either on heating or on cooling. The rear temperature is measured similarly for diffusivity data using Equation (3).

The product εF must be accurately known for thermal conductivity. The emissivity will vary for each material and with temperature, although many materials have "gray surfaces." The flux received by the sample through the light guide from a lamp can be measured at ambient to the temperature limit of the flux meter. The flux to be reflected is normally measured at distance X, again to the capability limit of the flux meter. Higher fluxes reflected by a mirror derive from higher sample temperatures and an extrapolated F–T curve for that light guide.

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