Transient thermal conductivity measurements: comparison of destructive and nondestructive techniques

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Abstract. Transient thermal conductivity methods have benefited from the rapid nature of the testing, but at the same time these methods are destructive because a sample must be prepared. Laser flash diffusivity is an example of a well-established transient method of measuring thermal conductivity, which requires samples to be prepared to a diameter of 12.4 mm.

The newly introduced modified hot wire (MHW) and transient plane source (TPS) techniques are transient as well, but are nondestructive in nature. This is possible because both of these new techniques are reflectance methods that operate by applying and measuring heat at the same surface. Essentially any flat surface can be measured, with no maximum size constraints. These transient techniques have been used to evaluate materials of differing size, structure, and homogeneity. While results compare well for homogeneous materials from foams to ceramic, composite materials differ considerably. This is because the two methods penetrate the samples to different degrees, and the results are a reflection of the testing depth. As test times are varied in the reflectance techniques, information was gathered that has been related to the thermal conductivity as a function of position normal to the sample.

Techniques are discussed and results on homogeneous materials from two different transient techniques are compared.

Introduction

Thermal management is necessary for all areas of material processing and also in a growing number of end user applications. Products such as electronic assemblies, injection molding molds, automotive 'under the hood' components must transfer heat efficiently. Such products encounter cyclic loading and their ability to conduct heat must not deteriorate over the desired product life. To transfer heat rapidly, the materials used in these applications must have high thermal conductivity.

A second family of products are designed with the opposite intention—to prevent heat transfer. Home insulation, firemen's protective clothing, boiler liners, and appliances (such as refrigerators, freezers, stoves, washers, and dryers) represent a cross-section of products that utilize materials designed for low thermal conductivity. New materials are developed for these markets in order to meet increasingly stringent energy or safety regulations. Foams and ceramics dominate these applications, with ceramics having the advantage for high temperature use. Whether the goal is to expedite or retard heat transfer, the response and behavior of these products must be modeled and then measured. There are three modes of heat transfer: radiation, convection, and conduction, and all require a driving force in the form of a temperature difference. All modes of transfer occur in a given situation, but often one or two will have negligible contribution. The focus of this work will be the conduction mode.

Conduction occurs primarily through a solid material. Conduction is related to the temperature (T), the thermal conductivity (k), the cross-sectional area (A) for heat (Q) to flow through, and the distance of travel (L) from the hot to cold side of the solid material:

$$Q = \frac{kA}{L}(T_{\text{hot}} - T_{\text{cold}}). \tag{1}$$

When a material is being developed or produced for a thermal management application, it is the material properties that must be measured. Thermal conductivity, heat capacity, and density are the primary properties of interest.

Thermal conductivity is a consideration at all levels when thermal engineers design for optimum heat dissipation, bonding, and homogeneity. Characterization of these new materials is crucial in the development cycle in order to optimize the production process. The same characterization is required on-line, to monitor the quality control during production.

There are two main categories of techniques available to measure thermal conductivity: transient and steady state. Both categories of measurement technique provide a temperature gradient and then monitor the response of the material to the gradient. The techniques differ in areas such as sample size, testing time, and range. Steady state techniques, such as the guarded hot plate described in ASTM C518-85 (American Society for Testing Materials 1985) or the technique of ASTM D5470-93 (American Society for Testing Materials 1993), involve placing a solid sample of fixed dimension between two temperature-controlled plates. One plate is heated while the other is cooled and the temperatures of the plates are monitored until they are constant. The steady state temperatures, the thickness of the sample, and the heat input per unit time to the hot plate are used to calculate the thermal conductivity from equation (1) (see figure 1). The difficulties with steady state techniques are that the measurement requires access to both sides of the sample, the sample must conform to specified dimensions, and the test time is lengthy in order to allow the steady state to be reached.
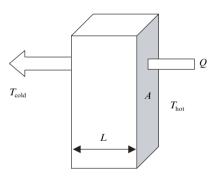

Figure 1. Experimental variables for steady state thermal conductivity tests.

2 Experimental

Transient techniques have gained popularity because of their speed. Measurements are conducted in seconds rather than minutes or hours. Two transient techniques have been established for a long enough period to have ASTM standards associated with them. They are described below.

2.1 Hot wire

The hot wire method (ASTM C1113, American Society for Testing Materials 1990) is a transient technique that involves inserting an electrically heated wire into a material. This intrusive method is possible for foams and fluids and melted plastics. In some cases a hole can be drilled into the sample to allow insertion of the wire. The heat flows out radially from the wire into the sample and the temperature of the wire is measured (see figure 2). The plot of the wire temperature versus the logarithm of time is used to calculate thermal conductivity. Testing can be conducted at a wide range of temperatures.
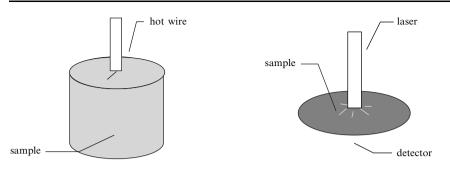
Figure 2. Experimental setup for hot wire experiments.

Figure 3. Experimental diagram for laser flash diffusivity testing.

2.2 Laser flash diffusivity

The laser flash diffusivity technique (ASTM E1461-92, American Society for Testing Materials 1992b) is a transient method that involves applying a short pulse of heat to the front face of a specimen by means of a laser flash, and measuring the temperature change of the rear face with an infrared (IR) detector (see figure 3). The resulting temperature rise of this face of the test specimen is monitored as a function of time and the half time to full temperature rise is used to determine the thermal diffusivity:

$$\alpha = \frac{k}{\rho c_p}, \tag{2a}$$

where α is thermal diffusivity, k is thermal conductivity, ρ is density, and c_p is the heat capacity. This must be combined with density and heat capacity data to calculate thermal conductivity.

Both of these methods are considered destructive if the samples to be studied are solids. In the case of the hot wire, the sample must either be melted or have a hole drilled into it. It is primarily a method for measuring liquids. Laser flash samples must be prepared to dimensions of 12.4 mm in diameter, thus destroying the solid product from which the sample was extracted. While the testing time was orders of magnitude faster than for steady state methods, the sample preparation time was just as rigorous.

New techniques have been developed recently in order to address the needs of both research and quality control facilities that required the ability to test various sizes of samples nondestructively. ASTM standards are not available for these techniques at the date of this publication.

2.3 Modified hot wire

The modified hot wire (MHW) technique is a transient heat reflectance technique similar to the hot wire (Mathis and Picot 1998). The modification is that the heating element is supported on a backing, which provides a rectangular one-dimensional heat flow (see figure 4). This eliminates the intrusive nature and allows for solids to be tested without being melted. The heating element relies on interfacial contact with a solid flat sample having minimum dimensions of 5 mm by 25 mm. The heating element is operated under constant current conditions and temperature of the heating element can be monitored during sample testing to measure temperature rise over time. The temperature rise of the element is inversely proportional to the ability of the sample to transfer the heat. From this relationship and calibration with certified standard materials, the value of effusivity, b (sometimes referred to as thermal inertia), is measured:

$$b = (k\rho c_p)^{1/2}. \tag{2b}$$

From the effusivity, thermal conductivity can be calculated by dividing by density and heat capacity. If density and heat capacity are not available, alternative analysis methods
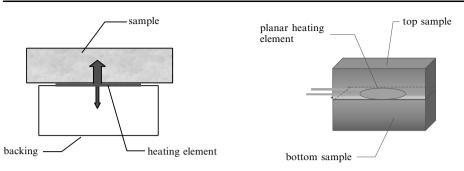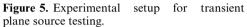
Figure 4. Experimental setup for the modified hot wire technique.

Figure 5. Experimental setup for transient plane source testing.

are available to obtain thermal conductivity and/or thermal conductivity ratios from the MHW technique (Mathis 1999).

2.4 Transient plane source

The transient plane source (TPS) technique is another transient heat reflectance technique similar to the hot wire (Gustafsson et al 1991; Log and Gustafsson 1995). The heating element is encapsulated in a protective coating but is not supported on a backing. A three-dimensional heat flow emanates from the circular heating element (see figure 5). The heating element relies on interfacial contact with two identical solid flat samples having minimum dimensions of 3 mm in diameter. As with MHW, the heating element is operated under constant current conditions and the temperature of the heating element has to be monitored during sample testing to measure temperature rise over time. The resultant data are curve fitted and iterated to determine thermal conductivity, diffusivity, and volumetric heat capacity without the need for calibration. This technique can be used for solids, liquids, and powders.

Both hot wire and TPS techniques are interfacial; heat is applied and detected on the same side of the material. This facet of operation allowed for the system to be brought on-line for testing thermal properties during production. Most other instrumentation for thermal conductivity measurement operates under principles that require heat input at one side of a material to be detected on the opposite side.

The drawback to interfacial/reflectance type testing is that the sample cannot be completely penetrated during a test. If penetration does occur, the medium on the other

Table 1. Comparison of benefits and features.

Benefits	Hot wire	Laser flash	MHW	TPS
No maximum sample size	✓		✓	✓
Small sample size (<250 mm^2)		✓	✓	✓
Minimal sample preparation	✓		✓	✓
Rapid testing (<1 min)	✓	✓	✓	✓
Homogeneity testing method			✓	✓
Finished product testing possible			✓	✓
On-line quality control			✓	✓
Thermal conductivity (k) directly	✓			✓
High k testing (>10 W m^{-1} K^{-1})	✓	✓		✓
Low k testing (<0.05 W m^{-1} K^{-1})	✓		✓	✓
Range of testing temperatures	✓	✓		✓
Liquid testing capability	✓	✓	✓	✓
ASTM compliant	✓	✓		
No sample contact required		✓		
No calibration required	✓	✓		✓
Complete unit $\leqslant$ \$25 000	✓		✓	

side of the sample would be factored into the test results produced by the instrument, thus providing a false representation of the thermal property value. To avoid this occurrence during testing, the techniques have methods and procedures to test films under 0.5 mm thick (Samuels and Mathis 1999).

Several instrument options are summarized in table 1, along with some key benefits. Computer data acquisition, manipulation, and display are standard.

3 Results

Testing was conducted on a series of homogeneous solid samples with different amounts of conductive alumina filler in a continuous matrix. The University of New Mexico supplied these samples and prepared them as 12.4 mm disks appropriate for laser flash diffusivity testing. After the samples were evaluated by laser flash, the same samples were evaluated by the TPS technique.

The results of testing are given below in figure 6. Both techniques illustrated that the thermal conductivity increased with the level of filler. The sample with 30% fill gave a lower level of thermal conductivity than expected by both methods, showing that the two techniques are linearly related. This trend is more clearly indicated in figure 7 where the results of thermal conductivity testing are plotted as a function of method and not fill level. A regression of the results plotted in figure 7 gives a regression coefficient of 0.99 when the intercept is set at zero. The slope of 0.8085 indicates that there is a bias between the two techniques, with laser flash producing thermal conductivities consistently 20% higher than TPS.
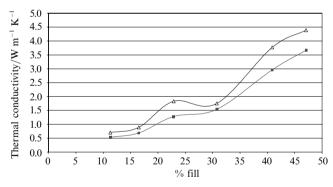
Figure 6. Thermal conductivity increased with percent fill of alumina: △, laser flash method; ■, TPS method.

regression equation:
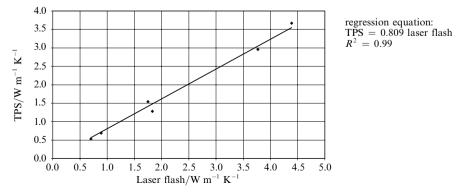TPS = 0.809 laser flash
$R^2 = 0.99$

Figure 7. Thermal conductivity results from the two techniques show a linear relationship but with a 20% bias.

4 Discussion

The bias between the two techniques may be related to one or more of the following potential causes: sample surface, use of external data, outer boundary effects, or sample nonhomogeneity. All precautions were taken to avoid these errors, but they will be mentioned below to provide an understanding of what to take into account when testing with these techniques.

4.1 Sample surface

In order to prevent light scattering from the sample surface during laser flash testing, gold was sputtered onto the sample. The gold film is thick enough not to allow any penetration of the laser radiation into the material. Even a slight penetration into the ceramic material would be 'pushing up' the thermal diffusivities of the material quite considerably. The gold layer theoretically provides a constant temperature distribution on the face of the sample. If the temperature distribution is Gaussian within the gold layer, lateral heat losses will occur that will result in a lower overall heat transmission detected at the back face of the sample. The lower overall temperature rise would shift the half time to total rise to a lower value as shown in figure 8. A lower time (or faster penetration) would be interpreted by the method as a higher thermal diffusivity.

During the TPS measurement surface contact is required where it is not in laser flash measurements. If the surface contact were questionable, a low result would result from TPS measurements. Any possible poor thermal contact between the sensor and the sample did not show up in the primary data of TPS. Both of these surface issues are consistent with the trend in this work.
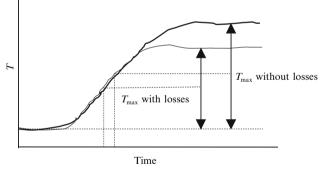

Figure 8. Heat losses at the surfaces shifts the penetration time to a lower value, indicating a higher diffusivity.

4.2 External data

A second reason may come from the fact that laser flash diffusivity relies on density and heat capacity measurements in order to calculate thermal conductivity from diffusivity. Because the sample thickness was predetermined by the laser flash requirements, the TPS method had insufficient thermal residence time to gather enough data to measure thermal conductivity directly. It found diffusivity as well and relied on a second measurement of volumetric heat capacity by the same instrument to determine thermal conductivity. If either of these measurements had systematic errors that reproduced through the entire series, it could produce the type of bias that was found. The raw diffusivity values will be obtained and compared in future work.

4.3 Boundary effects

A third reason for bias is that the samples may have experienced surface effects during TPS testing. If the heat wave penetrated to the far side or edges during testing, the

thermal conductivity would have had a low bias as found in this study. In examining the data, however, no influence from the far side of the sample pieces could be seen.

4.4 *Sample nonhomogeneity*
If the samples being tested had a higher concentration of fillers in the center as compared to the edges, then laser flash results would have been higher because the method evaluated the entire thickness. TPS only evaluates a certain depth into the sample that is related to the test time and the sample properties.

5 Conclusion
Transient techniques are a rapid method of evaluating thermal conductivity. Several options exist for instrument configurations that address sample specifications and testing environment requirements. A bias of 20% was found between TPS and laser flash.

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