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THERMOPHYSICAL PROPERTY MEASUREMENT AT THE MICRO- TO NANO-SCALE OF CONDUCTIVE WIRES—A COMPARISON OF THE ELECTROTHERMAL TECHNIQUE AND 3 OMEGA METHOD

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ABSTRACT

Accurate measurement of thermophysical properties (thermal conductivity and diffusivity and specific heat capacity) of micro- to nano-scale thin wires or films is a very difficult process; consequently, there are very few methods available to do so. Besides the optical setups in which thermal diffusivity is possible to be measured, the determination of thermal properties of fine fibres is limited to two major methods: the 3-omega method or the Transient/Generalized Electrothermal Technique. A comparative analysis of the two techniques using conductive platinum wires has taken place to determine the benefits and drawbacks of both. Variables such as accuracy, measurement theory, time to measure, and difficulty of measurement are all taken into account. The results for both methods were compared to theoretical and literature values. Trends and values indicate that both methods can yield reliable results with respect to diffusivity and conductivity and for specific heat capacity with 3-omega. The measurement process and results indicate that the ideal method is application specific.

INTRODUCTION

Important thermophysical parameters of a material include the values of thermal conductivity, thermal diffusivity, and specific heat capacity. The measurement of these properties at the micro- to nano-scale of thin wires/fibers and films, such as carbon nanotubes [1] polyacrylonitrile wires [2], and natural [3] and synthetic spider silks, has many different methods associated with it. Some photothermal methods are available to determine thermal property for fine fibers [4-5], but these have many drawbacks. Consequently, methods using electric currents and metallic resistive thermometers were developed. Two specific methods have been derived and used extensively due to the accuracy and ease of measurement, the transient/generalized electrothermal technique and the 3-omega method.

As an extension of a reference [5] the transient/generalized electrothermal technique (TET/GET) was used for the carbon nanotube [6] and spider silk [3] measurement. A typical setup for this technique is the suspended conductive or coated non-conductive sample, acting as both an electrical heater and thermometer on substrates. The measurement time is typically very short (milliseconds to seconds range depending on properties) with a good signal to noise ratio. The thermal diffusivity can be extracted by fitting the transient temperature response of the sample and the thermal conductivity determined by the overall resistance change being related to a change in temperature.

As one of the more reliable methods for thermal property determination the 3-omega method has been employed in multiple cases [8-12]. The system used for this method is the same as that for the TET, with the heater on substrate or resistive thermometer. This method, however, differs in that a modulated current is passed through the sample at a specific frequency and the temperature change propagates into the sample at the second harmonic. The voltage response due to the change in resistance can be monitored at the third harmonic to determine diffusivity, conductivity, and also the product of density and specific heat capacity. This measurement has a longer time to completion (minutes to hours) and can have differing signal to noise ratios depending on the technique used.

The effort of this research is to comparatively analyze the pros and cons of these two methods for thermal property determination and determine conditions under which each method would be suitable. Therefore, several fine platinum wires have been measured using both techniques at a range of temperatures. The results have been recorded and a comparison has been completed.

NOMENCLATURE

D	[m]	diameter of sample
I	[A]	current passing through sample
I_0	[A]	initial or reference current
k	[W/mK]	thermal conductivity
L	[m]	length of sample
C_p	[J/kgK]	specific heat capacity
q_0	[W]	volumetric heat generation
$R_{0,1}$	[Ω]	initial and steady state resistance, repsectively
R'	[Ω/K]	temperature coefficient of resistance times the resistance at T=0 degrees C

T	[K]	temperature
t	[s]	time
V_s	[m^3]	volume of the sample
x	[m]	cartesian axis direction

Special Characters

α	[m^2/s]	diffusivity of sample
α_T	[K^{-1}]	temperature coefficient of resistivity
ΔT	[K]	change in temperature
$\Delta\overline{T}$	[K]	average change in temperature
ω	[rad/s]	modulated frequency
ρ	[kg/m^3]	density

METHODS

The two methods have the same sample setup and preparation. Figure 1 below, presents a schematic of both methods.

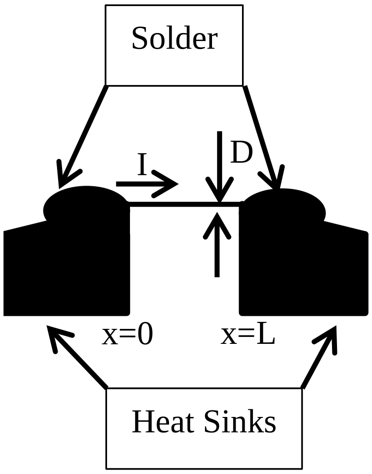

Figure 1 Schematic sample arrangement for both samples

As seen above the sample is suspended between two constant temperature heat sinks and connected by solder. For the TET a constant current is then passed through the sample to induce Joule heating, thereby elevating the temperature. Axial conduction is assumed due to the large aspect ratio (L/D). Using a reduced model that neglects radiation due to the small sample length, the non-dimensional temperature profile for thermal diffusivity estimation and explicit determination of thermal conductivity can be expressed as Eq. 1 and Eq. 2, respectively, with the heat generation specified by Eq. 3[13].

$$\frac{\Delta T}{\Delta T_S} = 1 - \frac{96}{\pi^4}\sum_{m=1}^{\infty}\frac{e^{-(2m-1)^2\pi^2\alpha t/L^2}}{(2m-1)^4} \tag{1}$$

$$k = \frac{q_0''' R' L^2}{12(R_1 - R_0)} \tag{2}$$

$$q_0''' = I^2 R_0 / V_s \tag{3}$$

To solve the first expression for diffusivity a curve fit method based on Levenberg-Marquardt [14] regression is used. To obtain thermal conductivity, a calibration needs to be performed to determine the slope of resistance with respect to temperature, *R'*, and then the conductivity can be determined explicitly. The sample resistances were taken as the optimized initiation and the steady-state plateau value on the Joule heating curve.

There are several methods available for the 3-omega determination. The measurement can be performed with a voltage source in combination with a Wheatstone bridge, a current source, or a current source with cancellation of the fundamental frequency signal. This paper will use the current source with cancellation technique. Each method was attempted. Without cancellation the product of density and specific heat capacity could not be determined, and the voltage source demonstrated large instabilities, most likely due to fluctuations in resistance and current being large in magnitude to the resistance of the sample. Typically the voltage measurement is the most stable when measuring a sample that is larger in resistance.

The three omega method also uses Joule heating to induce a temperature rise in the sample. This temperature rise is due to a modulated current that can be expressed by Eq. 4, and a heat generation expressed as Eq. 5.

$$I = I_0\cos\omega t \tag{4}$$

$$q''' = \frac{I^2 R(T)}{V_s} = \frac{I_0^2(1+\cos 2\varpi t)R(T)}{2V_s} \tag{5}$$

This temperature change will induce a resistance change that is correlated to the temperature coefficient of resistivity, as presented in Eq. 6.

$$R(T) = R_0 + R'\Delta T = R_0(1+\alpha_T\Delta T) \tag{6}$$

Since the temperature rise is reflected in the resistance change, the resistance needs to be monitored. For the three omega method this is typically done by monitoring the voltage across the sample as a function of resistance, as shown in Eq. 7.

$$V_c = IR(\mathrm{T}) = I_0R_0\cos\omega t + I_0R'\cos\omega t\bullet\Delta T(2n\omega t),\ n=1,2,... \tag{7}$$

Modelling the temperature change as the classical heat diffusion equation with a volumetric heat generation can solve for the periodic temperature change as a function of given parameters. Equation 8 shows this solution.

$$\overline{\Delta T_p} = \frac{I_0^2 R_0[2-2\cosh(mL)+mL\sinh(mL)]}{m^3L^2k\pi D^2\sinh(mL)} \tag{8}$$

where $m^2 = \frac{i2\varpi}{\alpha}$ when radiation influence (for the platinum samples) is negligible.

Substituting into Eq. 7 and cancelling the first harmonic can determine the voltage at the third harmonic. The voltage from the third harmonic can be expressed as Eq. 9.

$$V_3 = \frac{I_0^3 R_0 R' L[2-2\cosh(mL)+Lm\sinh(mL)]}{k\pi D^2(mL)^3\sinh(mL)} \tag{9}$$

After simplification at a low frequency limit of zero, using a first harmonic cancellation technique this voltage is expressed as Eq. 10, and at a high frequency limit of negative ninety, the voltage can be expressed as Eq. 11.

$$V_3 = \frac{I_0^3 R_0 R' L}{12 k \pi D^2} \tag{10}$$

$$V_3 = \frac{I_0^3 R_0 R'}{i 2 \varpi \rho C_p \pi D^2 L} \tag{11}$$

The low frequency limit is independent of the heat capacity and therefore is suitable for thermal conductivity determination, whereas the high frequency limit is independent of thermal conductivity and consequently suitable for heat capacity determination. To determine thermal diffusivity a fit must be done with the portion of the phase response at different frequencies between the low and high frequency limit. This approach avoids propagation of error from different thermal properties by determining each of them separately.

EXPERIMENTAL SETUP

An experiment was conducted on several 25.4 µm diameter, 99.95% platinum wires (SurePure Chemetals). Thermal properties were determined using both the transient electrothermal and 3-omega techniques. Samples were mounted on a copper substrate, with copper mounting blocks doubling as heat sinks, attached by two-part epoxy. One side of the copper substrate had a common copper heat sink, while the other had several individual bars to electrically isolate. Heat sinks were sloped slightly outward to ensure good contact at the inner edge. Two electrical wires were soldered to each heat sink along with a platinum wire sample for each. A type K thermocouple was soldered to the common heat sink to monitor temperature during the measurement. A pictorial representation of a sample mount can be found in Figure 2.

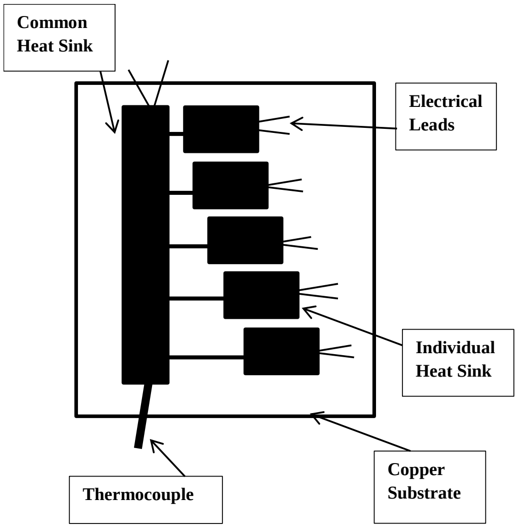

Figure 2 Pictorial representation of a sample mount

The samples were placed in an isothermal copper enclosure and tubing setup, temperature controlled by a Cole-Parmer Polystat circulating water bath. This was placed in vacuum and brought down to less than 10e-7 of barometric pressure. The samples were tested in a range of temperatures from 10 to 30 degrees Celsius. The setup for each method is pictured in Figure 3.

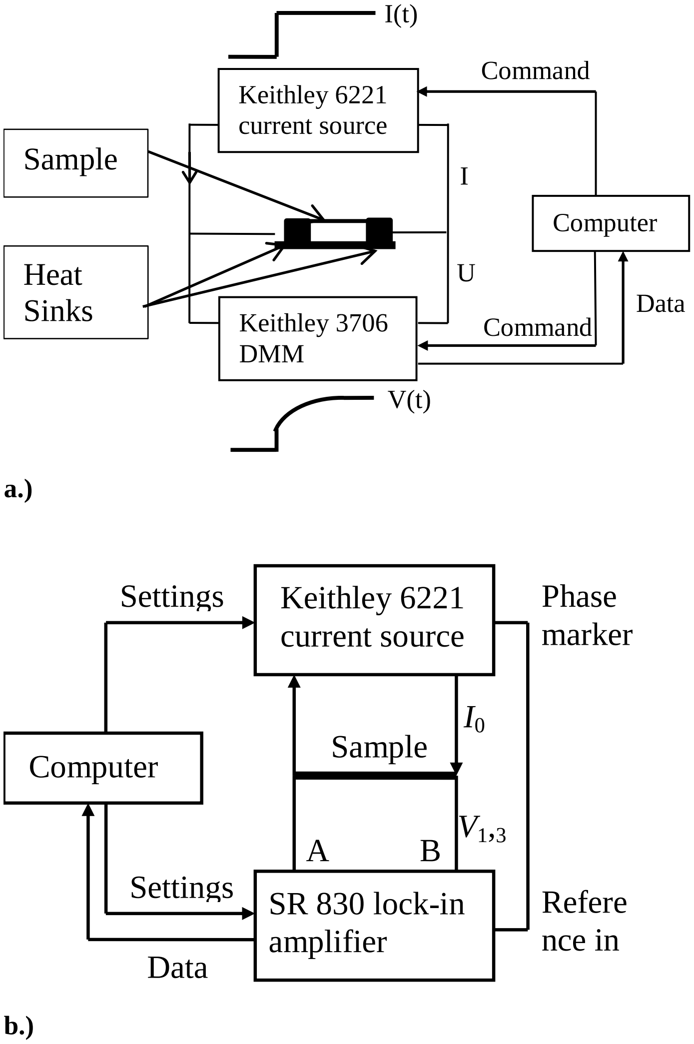

Figure 3 a.) Schematic of the experimental setup for the transient electrothermal technique b.)Schematic of the experimental setup for the 3-omega technique

TRENDS AND RESULTS

Reference values for platinum thermal properties are readily available. The results at room temperature for a 4.56 mm platinum wire are detailed in Table 1 for both measurement methods, as well as values found in literature.

		TET	3-omega	Literature	
α	[m^2/s]	2.49e-5	2.43e-5	2.51e-5	[18]
k	[W/mK]	71.65	71.94	71.6	[19]
C_p	[J/kgK]	-	134.3	133	[20]

Table 1 Summary of values determined using both methods for 25.4 µm platinum wires

The measured values follow those reported in literature very closely. The thermal conductivity has also been determined for a range of temperatures with a 2.83 mm long wire. These results are presented in Figure 4.

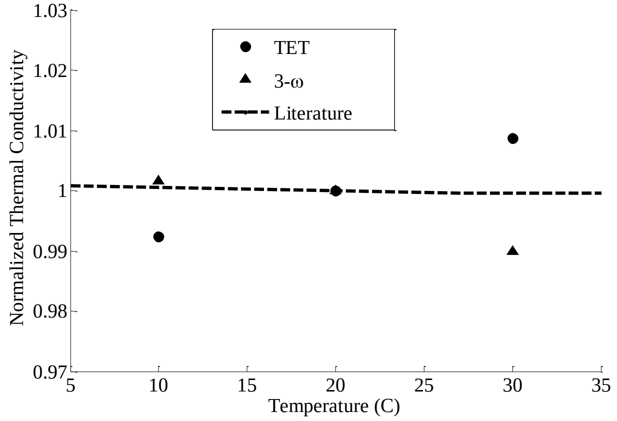

Figure 4 Normalized values of conductivity at different temperatures. Note how the 3-omega method follows the trend.

The 3-omega method appears to be following the recommended trend more closely than the TET method, with a decrease in conductivity with increasing temperature.

DISCUSSION

For the room temperature measurements, both methods appear to be arriving at accurate results when compared with literature values. However, as temperature is varied, the 3-omega method more closely parallels the trend found in literature. In fact, the TET has results that are contrary to the widely accepted trend. This discrepancy could fall within the uncertainty band.

A large benefit to using the 3-omega method is that it allows for thermal conductivity and diffusivity and specific heat capacity to all be determined separately, unlike the TET. This methodology prevents large propagation of uncertainty when calculating the value of a third property.

The derivation/theory behind each method was performed prior to measuring samples. Comparatively speaking the theory behind the 3-omega method is more complicated and in depth than that for the TET, both from a mathematical and engineering standpoint when it comes to derivation. However, at the implementation stage, the mathematics and engineering associated with the 3-omega method are far simpler, because the implementation of the Levenberg-Marquardt regression when using the TET to determine diffusivity is very complicated.

The TET has an extremely short time to measure (several milliseconds to seconds range depending on α). This particular feature makes it ideal for implementation at temperatures largely deviated from room. The short time to measure only requires environmental and heat sink temperature stability for a few seconds. The 3-omega method is much different. The time to measure can range from several minutes to hours at a time. For certain applications this can be difficult. The temperature controller used in this experiment was extremely stable at the measurement temperatures; however, experience with tools such as cryo-coolers dictates fluctuations in temperature over time, making the 3-omega measurement non-ideal for large deviations from room temperature without a very stable isothermal environment.

The difficulty of the 3-omega method implementation is slightly higher than that of the TET. This is due primarily to the electronics setup. Application of a lock-in amplifier, operational amplifier, resistive filter, and fundamental harmonic cancellation make up the extra electrical device knowledge that is not present in the TET.

CONCLUSION

The thermoelectric technique and 3-omega method are widely used measurement methods for thermal property determination. Determining which to use is application specific. To better understand the methods, several 25.4 μm platinum wires were measured for thermal diffusivity and conductivity and specific heat capacity at a range of temperatures from 10 to 30 degrees Celsius and then comparisons between the two were made. The comparison was based on accuracy, measurement/data processing theory, time to measure, and difficulty of measurement. At room temperature, both methods yield results that do not deviate significantly from those reported in literature. However, when compared to literature trends in the range of temperatures measured, the 3-omega followed more closely. The theory behind the two methods is about equally weighted with difficulties in derivation and data processing presenting themselves in the 3-omega method and TET, respectively. With the implementation of more electronics the 3-omega poses a problem with difficulty of implementation. Due to the extended time to measure with the 3-omega, it is recommended to use the TET for applications where the thermal environment is unstable.

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