A Sensitivity Analysis of a Thin Film Conductivity Estimation Method

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ABSTRACT

An analysis method was developed for determining the thermal conductivity of a thin film on a substrate of known thermal properties using the flash diffusivity method. In order to determine the thermal conductivity of the film using this method, the volumetric heat capacity of the film must be known, as determined in a separate experiment. Additionally, the thermal properties of the substrate must be known, including conductivity and volumetric heat capacity. The ideal conditions for the experiment are a low conductivity film adhered to a higher conductivity substrate. As the film becomes thinner with respect to the substrate or, as the conductivity of the film approaches that of the substrate, the determination of the thermal conductivity of the film becomes more difficult.

The present research examines the effect of inaccuracies in the known parameters on the estimation of the parameter of interest, the thermal conductivity of the film. As such, perturbations are introduced into the other parameters, which are assumed to be known, in a synthetic experiment, to find the effect on the estimated thermal conductivity of the film. A baseline case is established and each of the baseline parameters is varied from its baseline value in succession. For each variation of each baseline parameter, a synthetic experimental data file is generated. Each of these data files is individually analyzed by the program to determine the effect of the perturbations on the estimated film conductivity. Thus, quantitative values are obtained for the vulnerability of the method to various laboratory measurement errors.

NOMENCLATURE

a	=	thickness of substrate (m)
b	=	thickness of film (m)
l_c	=	total sample thickness (m)

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c_{p1} = specific heat of the substrate ($kJ \cdot kg^{-1} \cdot K^{-1}$)
c_{p2} = specific heat of the film ($kJ \cdot kg^{-1} \cdot K^{-1}$)
$A_n\ B_n\ C_n\ D_n$ = constants in the series solution (K)
Bi_1 = Biot number for the substrate
Bi_2 = Biot number for the film
h = convection coefficient ($W \cdot m^{-2} \cdot K^{-1}$)
k_1 = thermal conductivity of the substrate ($W \cdot m^{-1} \cdot K^{-1}$)
k_2 = thermal conductivity of the film ($W \cdot m^{-1} \cdot K^{-1}$)
N_n = norm for the series solution
q_o = heat pulse magnitude ($J \cdot m^{-2}$)
t = time (s)
T_1 = temperature of the substrate (K)
T_2 = temperature of the film (K)
T = ambient temperature (K)
V = volume (m^3)
x = spatial variable in direction of heat transfer (m)
γ = eigenvalue for substrate (m^{-1})
δ = Dirac delta function (s^{-1})
η = eigenvalue for film (m^{-1})
ρ_1 = density of substrate ($kg \cdot m^{-3}$)
ρ_2 = density of film ($kg \cdot m^{-3}$)

INTRODUCTION

The measurement of thermal diffusivity by the flash method has been in use since the early 1960s, with instruments which perform these experiments now manufactured by several companies. This method involves the testing of a small disc-shaped sample of material, usually 1-2 cm in diameter with a thickness of 1-2 mm. The sample is placed in the flash diffusivity instrument and is subjected to a brief but intense light flash with a duration of several milliseconds and an intensity several kilowatts per square millimeter. A temperature history is then recorded from the non-heated side of the sample with an optical measurement system. This temperature history is then analyzed so as to calculate the thermal diffusivity of the material. The flash diffusivity method has several advantages. These include the small sample size and the short duration of the experiments, usually on the order of seconds. An additional advantage of the non-contact temperature measurement system is that samples can be tested at very high temperatures.

The seminal paper associated with flash diffusivity experiments was Parker et al. [1] which dealt with a single-layer solid sample. The amount of time required to raise the sample temperature to ½ of the final equilibrium temperature, assuming no heat loss subsequent to the flash, was the only information utilized in this analysis. In subsequent years, more sophisticated analysis methods have been used in analyzing flash diffusivity experiments. Cowan [2] developed tabulated heat-loss correction factors to refine the method used by Parker et al.[1]. These charts could be entered with properties such as surface emissivity, ambient temperature and peak sample temperature. Clark and Taylor [3] expanded upon this work so as to include heat losses from the sample circumference, accounting for some two-dimensionality of the sample. More precise work performed subsequently involved the principle of nonlinear regression using least squares to adapt a heat transfer model to the experimental measurements in these types of experiments. Koski [4] performed some of the initial work in this area followed by Taylor [5]. The latter also attempted to account for heat losses, as well as various laser pulse shapes. Raynaud et al. [6] investigated the adequacy of various competing mathematical models using least squares. This has also been accomplished by Beck and Dinwiddie [7]. Cape and Lehman [8] have also made contributions to the field of diffusivity measurement by the flash method. More specifically, they have addressed the effects of flash duration on the outcome of the experimental analysis.

A method for determining the thermal conductivity of a thin film on a substrate of known thermal properties using the flash method was developed by McMasters et al [9]. This method used an exact solution for the two-layer solid and included convective heat losses in the direct solution. This two-layer program forms the basis of the present research, as further investigation is made into the effects of measurement errors on the performance of the two-layer program. In the present work, synthetic data files are generated for various cases of substrate conductivity, substrate volumetric heat capacity, film volumetric heat capacity, film thickness and substrate thickness. Utilizing the two-layer program to analyze these files generated results for the thermal conductivity of the film that could be compared with the known conductivity. From this comparison, insight could be obtained into the effects of measurement errors in the known parameters on the estimation of the parameter of interest, the film thermal conductivity.

EXPERIMENT DESCRIPTION

The experiments analyzed in this research were synthetically generated using an exact solution of the two-layer solid subjected to an instantaneous flash, with convective boundary conditions otherwise. The heat transfer through the sample was assumed to be one-dimensional. The geometry for the experiment is shown in Figure 1. The objective of the experiment is to determine the thermal conductivity of the film. In order to calculate this thermal conductivity, five parameters must be known regarding the two-layer material. These are: film thickness, substrate thickness, film volumetric heat capacity, substrate volumetric heat capacity, and substrate thermal conductivity. As part of the parameter estimation process, three parameters are simultaneously estimated. Only one of these parameters, film thermal conductivity, is desired. The other two parameters, which are convection coefficient and the magnitude of the flash, must be simultaneously estimated along with film thermal conductivity. However, these additional parameters are not of interest and are not part of the experimental objective. Two differential equations are required for the direct solution in this analysis. One differential equation is required for each material layer as follows.

$$k_1 \frac{\partial^2 T_1}{\partial x^2} = \rho_1 c_{p1} \frac{\partial T_1}{\partial t} \quad \text{and} \quad k_2 \frac{\partial^2 T_2}{\partial x^2} = \rho_2 c_{p2} \frac{\partial T_2}{\partial t} \tag{1}$$

In these equations, the subscripts 1 and 2 designate the layers of the material, specifically, the substrate and the film, respectively. The symbol k designates thermal conductivity and ρc_p is volumetric heat capacity. The boundary conditions for this problem are

$$-k_1 \frac{\partial T_1}{\partial x}\bigg|_{x=0} = h(T_\infty - T_1) + q_o \delta(t) \quad \text{and} \quad -k_2 \frac{\partial T_2}{\partial x}\bigg|_{x=c} = h(T_2 - T_\infty) \tag{2}$$

where h designates the convection coefficient, q_o is the magnitude of the heat pulse and $\delta(t)$ is the Dirac delta function. The compatibility conditions at $x = a$ are

$$T_1 = T_2 \quad \text{and} \quad k_1 \frac{\partial T_1}{\partial x}\bigg|_{x=a} = k_2 \frac{\partial T_2}{\partial x}\bigg|_{x=a} \tag{3}$$

The initial conditions for this problem are

$$T_1(x,0) = T_2(x,0) = T_\infty \tag{4}$$

where T_∞ is the ambient temperature. The solution for the differential equations and boundary conditions here are solved in detail in Reference [9]. The final solution given there is

$$T_2(c,t) = q_o \sum_{n=1}^{\infty} \frac{A_n(C_n \cos \eta c + D_n \sin \eta c)}{N_n} e^{-k\eta^2 t/\rho c} \tag{5}$$

where c is the distance along the horizontal axis corresponding to the overall thickness of the two-layer sample, as shown in Figure [1]. This equation therefore gives the temperature of the film surface corresponding to the measured surface temperature. The other variables in the solution include the norm, which is

$$\begin{aligned} N_n = \rho_1 c_{p1} &\left[A_n^2 \left(\frac{a}{2} + \frac{1}{4\gamma} \sin 2\gamma a \right) + \frac{A_n}{\gamma} \sin^2 \gamma a + \left(\frac{a}{2} - \frac{1}{4\gamma} \sin 2\gamma a \right) \right] \\ &+ \rho_2 c_{p2} C_n^2 \left(\frac{b}{2} + \frac{1}{4\eta} (\sin 2\eta c - \sin 2\eta a) \right) \\ &+ \rho_2 c_{p2} \frac{C_n D_n}{\eta} (\sin^2 \eta c - \sin^2 \eta a) + D_n^2 \left(\frac{b}{2} - \frac{1}{4\eta} (\sin 2\eta c - \sin 2\eta a) \right) \end{aligned} \tag{6}$$

This, in turn, is composed of the other constants which are

$$C_n = \left[\frac{k_1 \gamma}{h} \cos(\gamma a) \cos(\eta a) + \sin(\gamma a) \cos(\eta a) \right]$$
$$+ \left(\frac{\gamma k_1}{\eta k_2} \right) \left[\frac{k_1 \gamma}{h} \sin(\gamma a) \sin(\eta a) - \cos(\gamma a) \sin(\eta a) \right] \tag{7}$$

and

$$D_n = \left[\frac{k_1 \gamma}{h} \cos(\gamma a) \sin(\eta a) + \sin(\gamma a) \sin(\eta a) \right]$$
$$- \left(\frac{\gamma k_1}{\eta k_2} \right) \left[\frac{k_1 \gamma}{h} \sin(\gamma a) \cos(\eta a) - \cos(\gamma a) \cos(\eta a) \right] \tag{8}$$

and

$$A_n = \frac{k_1 B_n \gamma}{h} \tag{9}$$

Finally, the eigenvalue η is determined by the eigencondition

$$\frac{\eta b \tan(\eta b) - Bi_2}{Bi_2 \tan(\eta b) + \eta b} = -\left(\frac{\gamma k_1}{\eta k_2} \right) \frac{\gamma a \tan(\gamma a) - Bi_1}{Bi_1 \tan(\gamma a) + \gamma a} \tag{10}$$

where $Bi_1 = ha/k_1$ and $Bi_2 = hb/k_2$. In this equation, the eigenvalues γ and η must be computed simultaneously, where γ is the eigenvalue corresponding to the substrate and η corresponds to the film. These are found by locating asymptotes which occur when the denominator of each side zero, which is how one equation can be solved to find an infinite number of unknown values. Table 1 below shows the values of the parameters used in this experiment to generate the data files.

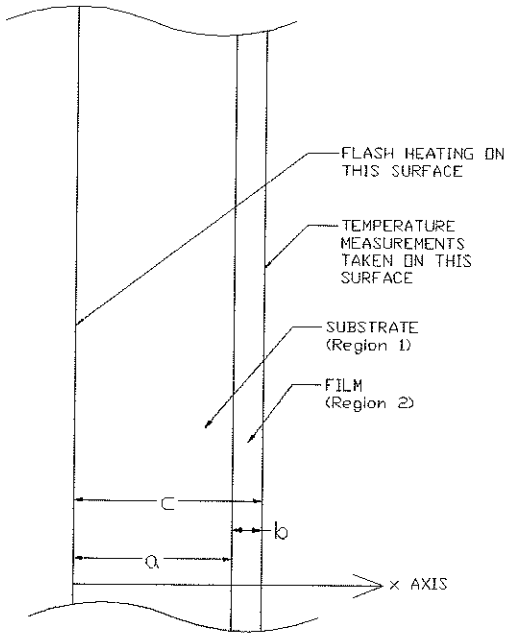

Figure 1. Section view of the flash diffusivity sample showing the two layers of material.

As can be seen in Table 1, there are eight parameters specified. These include the five "known" parameters as well as the three unknown parameters. As part of the investigation in the present research, the five "known" parameters were allowed to vary, one at a time, from the values stated in Table 1 to values as low as 50 percent less than these stated values. The intentionally-introduced errors constituted a means by which the effect of measurement errors on the known properties would affect the thermal conductivity of the film found by the parameter estimation program.

TABLE I. PARAMETERS USED IN PREPARING THE DATA FILES FOR THE SYNTHETIC EXPERIMENTS.

Substrate thermal conductivity	$1.0\ W{\cdot}m^{-1}{\cdot}K^{-1}$
Substrate volumetric heat capacity	$10^6\ J{\cdot}m^{-3}{\cdot}K^{-1}$
Substrate thickness	0.8 mm
Film thickness	0.2 mm
Film volumetric heat capacity	$10^6\ J{\cdot}m^{-3}{\cdot}K^{-1}$
Film thermal conductivity	$0.1\ W{\cdot}m^{-1}{\cdot}K^{-1}$
Convection coefficient	$20\ W{\cdot}m^{-2}{\cdot}K^{-1}$
Magnitude of heat absorbed during the flash	$1000\ J{\cdot}m^{-2}$

RESULTS

Applying Equation (5) in a parameter estimation scheme, as described by Reference [10], the data files can be analyzed to find the thermal conductivity of the film. This is accomplished through iterative nonlinear regression, fitting the mathematical model to the experimental data. Figure 2 shows the best fit obtained between the mathematical model and the experimental data file when the substrate thickness was actually 0.4 mm when the mathematical model was using a substrate thickness of 0.8 mm as given in Table 1. The two curves appear to be very close to one another but the error between the two can be examined more closely by plotting the residuals. Figure 3 shows a plot of the residuals, which is simply a plot of the error in temperature between the measured data and the mathematical model. As can be seen in this plot, there is a systematic difference between the two. Ideally, the error should oscillate across the zero temperature axis, reflecting the measurement noise in the experiment. In this case, there are many consecutive data points above and then many consecutive data points below the axis, indicating a systematic error between the experimental measurements and the mathematical model. Indeed, this difference in this experiment is known to be from the error in the measurement of the substrate thickness.

Figure 4 gives a composite presentation of the results from varying the "known" parameters by introducing errors of as much as 50 percent. The effects of these introduced errors can be seen in the parameter of interest, the thermal conductivity of the film. As can be seen in this plot, the thickness of the film is the

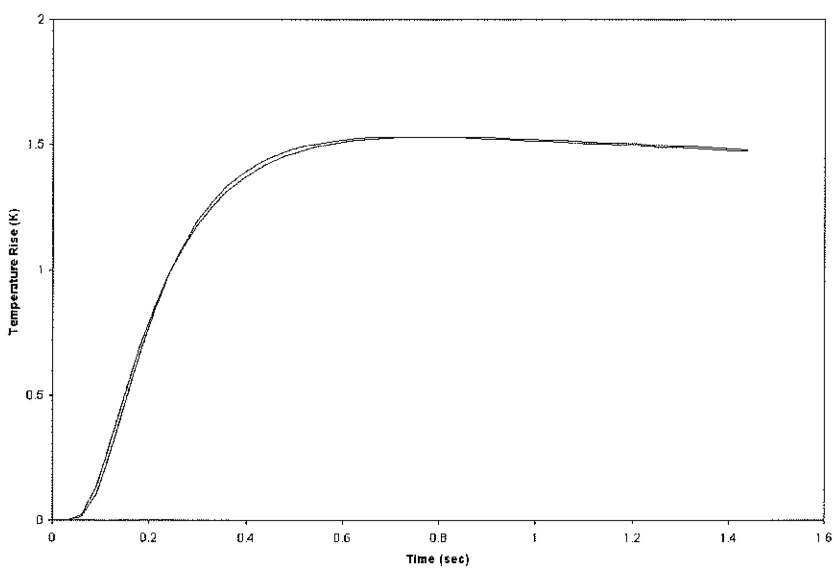

Figure 2. A plot of calculated and measured temperature for the converged solution in a case wherean error of 50 percent is introduced in the substrate conductivity.

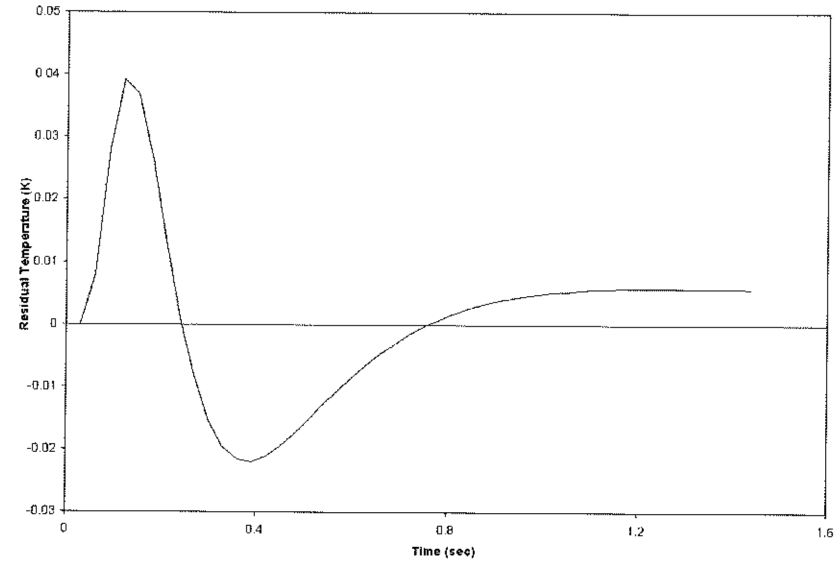

Figure 3. A magnified view of the error between the two curves shown in Figure 3.

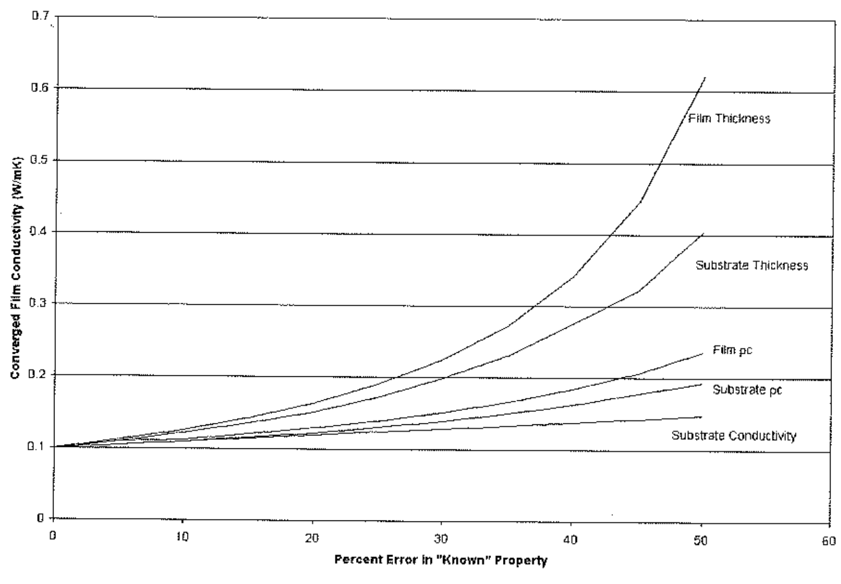

Figure 4. A plot of the calculated film thermal conductivity as functions of errors introduced into the known parameter values.

parameter which causes the greatest sensitivity in the estimation of the thermal conductivity of the film. If the measurement of the thickness of the film is in error by 50 percent, and is reported at 0.2 mm instead of the actual 0.1 mm value in this case, the calculated value of the thermal conductivity will be in error by over a factor of 6. The measurement of the substrate thickness does not cause the same sensitivity in the results of film thermal conductivity, but will still cause an error of as much as a factor of 4. This error would be caused, in this example, by reporting a substrate thickness of 0.8 mm when the actual thickness was 0.4 mm. Errors of 50 percent in the volumetric heat capacity of either the film or the substrate can cause errors in the estimated conductivity by as much as a factor of 2. Finally, errors in the value used for substrate thermal conductivity generate errors of approximately the same magnitude in film thermal conductivity.

The standard deviation of the residuals is a useful measure of the adequacy of the fit between a mathematical model and a set of experimental measurements. Ideally, the standard deviation of the residuals should compare very closely with the standard deviation of the measurement errors. Errors substantially larger than these usually signify a disparity between the mathematical model and the physics of the experiment. Examining a plot of the residuals, such as shown in Figure 3, can give greater insight into the nature of the disparity between the mathematical model and the experiment. Figure 5 shows a summary of the standard deviation of the residuals generated in each of the test cases in this research, as a function of the error introduced in the various known parameters. With no errors introduced, the converged mathematical model used in the parameter estimation scheme would be expected to match exactly with the experimental data, since the experiments in this research were synthetic, as generated by the mathematical model. As the errors in the known parameters were gradually increased, the standard deviation of the residuals would also be expected to increase, since the best converged temperature values generated by the mathematical model would not be expected to match the experimental data exactly. This gives the shape of the curves shown in Figure 5.

It is interesting to note, in looking at Figure 5, that the cases with the greatest errors in the estimation of the film thermal conductivity do not necessarily correspond to the cases with the largest standard deviation of residuals. In comparing Figures 4 and 5, the order of sensitivity of the estimated film conductivity to the errors in the known parameters is: film thickness, substrate thickness, film volumetric heat capacity, substrate volumetric heat capacity and substrate thermal conductivity. However, the order of the standard deviation of the residuals to the errors in the known parameters is: substrate thickness, substrate volumetric heat capacity, film thickness, substrate thermal conductivity and film volumetric heat capacity. This shows that the errors in estimated values of film thermal conductivity cannot always be predicted, simply by looking at the standard deviation of the residuals. In some cases, the error in film thermal conductivity can be quite significant and yet the mathematical model can choose a combination of parameters for film thermal conductivity, convection coefficient and heat pulse magnitude, that allow somewhat close conformance of the calculated and measured temperatures. The analyst must be mindful of these factors when comparing the results generated from various experiments.

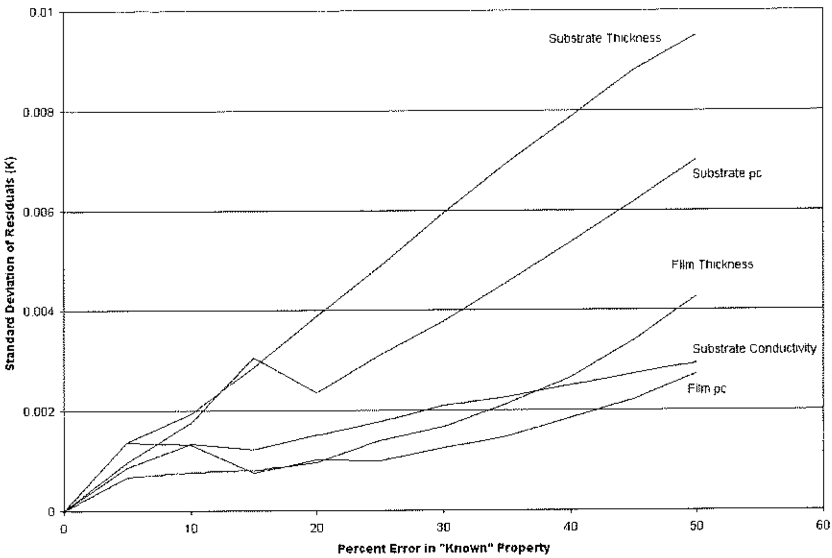

Figure 5. A plot of the standard deviation of the residuals which arise when estimating film thermal conductivity as functions of errors introduced into the known parameter values.

CONCLUSIONS

A sensitivity analysis was performed on a method for determining the thermal conductivity of a thin film on a substrate of known thermal properties. It was found that the known property to which the estimation of film thermal conductivity is most sensitive is the film thickness. The other known properties producing the greatest sensitivity in the thermal conductivity estimation are substrate thickness, film volumetric heat capacity, substrate volumetric heat capacity and substrate thermal conductivity, in order of decreasing sensitivity. For this reason, the measurement of the film thickness is of critical importance in obtaining accurate values of film thermal conductivity.

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