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Accounting for Finite Flash Duration in Diffusivity Experiments

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The laser flash method, as a means of measuring thermal diffusivity, is well established, and several manufacturers produce equipment for performing these types of experiments. Over time, mathematical models of increasing sophistication employing nonlinear regression have been used in the analysis of flash diffusivity experiments. These models have historically assumed an instantaneous flash and have been highly accurate for most samples of moderate diffusivity and sample thickness. As samples become thinner and more highly conductive, the duration of the experiments becomes very short. Since the duration of the flash is typically on the order of several milliseconds, the assumption that this period of time is instantaneous becomes less valid for very short experiments. In the present research, three models accounting for the duration of the flash are applied to three samples of stainless steel of varying thicknesses and compared with a model that assumes an instantaneous flash. The models accounting for the finite flash duration generate results that are much more consistent between samples than the model assuming an instantaneous flash. Moreover, the conformance of the mathematical models accounting for flash duration is much closer to the measured data than the model that assumes an instantaneous flash.

Nomenclature

α	=	thermal diffusivity, m^2/s
c	=	specific heat, J/kg-K
k	=	thermal conductivity, W/mK
L	=	thickness of sample, m
m	=	counting integer for infinite series solution
n	=	number of temperature measurements
p	=	number of parameters
q	=	magnitude of heat flux, W/m^2
T	=	temperature in sample above ambient, K
T_o	=	Ambient temperature, K
t_1	=	flash start time, ms
t_2	=	flash end time, ms
ρ	=	density, kg/m^3

Introduction

THE flash heating method for determining thermal diffusivity in materials has been used since the 1960s [1]. Subsequent to that time, the method has become widespread and is now the most common method of diffusivity measurement. One of the primary advantages of the method is the small sample size with which diffusivity measurements can be made. The experiments are also very short in duration, typically lasting on the order of seconds. In cases of thin materials with high diffusivities, the experiments may only last several tenths of a second. Once the experimental setup work is complete, these features of thermal diffusivity measurement allow multiple experiments to be conducted and repeated in a short amount of time.

The duration of the flash heating phase of the experiment is typically several milliseconds. Over this brief segment of time, the heating of the sample is very intense, with the rate of heating being as high as several kilowatts per square millimeter. A rise in temperature is observed on the unheated side of the sample in response to the flash heating. A temperature rise of several degrees may be observed, although a measurement of the magnitude of the temperature rise is not needed for the determination of thermal diffusivity. Measurements recorded in terms of detector voltage, or some other property proportional to the temperature of the samples, is all that are required. From the shape of the curve of temperature vs time, the thermal diffusivity can be determined.

The analysis method used in determining thermal diffusivity from the measured data can have a significant impact on the results. Early analysis methods used for flash diffusivity experiments were quite simple and assumed no convective heat losses. Additionally, an instantaneous flash duration was assumed [1]. For many types of flash diffusivity experiments, these methods are still perfectly valid and no substantial improvements come about through the use of more sophisticated analysis methods. This is particularly true at moderate experiment temperatures (where convective losses are small), in cases with solid nonporous materials (where flash penetration is minimal), and in materials with low-to-moderate diffusivity. In the analysis used by Parker et al. [1], the half-rise time was the only parameter required for determining the thermal diffusivity of the sample. The American Society for Testing and Materials has established standard methods for performing the flash diffusivity experiment [2], which follow the Parker method [1] very closely. Subsequent methods, such as Cowan [3], applied correction factors to the Parker method [1], which accounted for heat losses. The development of charts for this purpose allowed the user to modify the results obtained using the half-rise time. Clark and Taylor [4] further added correction factors for the outer edge heat losses, providing some accommodation for two-dimensional convection. Koski [5] moved the analysis methods to a new level by fitting mathematical models to the experimental data using least-squares. This method provided a more reliable result, since it used data from the entire time domain of the experiment. This method eliminated the susceptibility to measurement errors near the point of half-rise time. The method of least-squares modeling was expanded to include heat losses by Taylor [6]. Additionally, errors due to various shapes of the laser pulse were addressed in this research. As a summary paper, Raynaud et al. [7] makes a comparison of the adequacy of some of these models. Similar comparisons are also made by Beck and Dinwiddie [8]. This paper evaluates the necessity for making distinctions between the two convection coefficients estimated for opposite sides of the sample. It was shown that the results of the experiment are

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largely unaffected by estimating only one convection coefficient for both sides. McMasters et al. [9] explored a procedure for finding the thermal conductivity of a thin film on a known substrate using the flash diffusivity method. As part of this work, the volumetric heat capacity of the film and the substrate were both known. Research that bears a similarity to the work examined presently, although not involving flash diffusivity testing, is described by Koo et al. [10] and Hender [11]. In both of these papers, refinements were made in the mathematical models in order to achieve a closer conformance to experimental measurements. McMasters [12] examined two-dimensional models including flash penetration beyond the surface of the material. Various models were allowed to compete to find the fit that was most appropriate.

With the exception of [6], all of the above mentioned methods assume that the laser flash is instantaneous. In the case of that paper, the effect on the accuracy of the results were examined for a range of noninstantaneous flash durations. Similarly, Azumi and Takahashi [13] looked at triangular- and rectangular-shaped flash time domains and found effective time-shift values by evaluating a time-weighted integral of the pulse. This time-shift term was used for evaluating the experiment as if the flash had happened at a time other than zero. This paper underscores the fact that thin samples of high thermal conductivity are the most sensitive to noninstantaneous flash durations. Cape and Lehman [14] developed estimates for correction factors on the half-rise time in evaluating experiments. Although actual experimental data was not used in this research, the errors associated with noninstantaneous flash experiments were estimated analytically. Larson and Komaya [15] measured the shape of various flash lamps using an oscilloscope and used these measurements to find characteristic times. Results were generated from copper and iron samples with corrections to the half-rise time method for the particular pulse shapes encountered with the oscilloscope measurements. Likewise, Taylor and Clark [16] also proposed adjustments to the half-rise time analysis method for accounting for noninstantaneous flash duration.

In contrast to previous methods used, the present research examines an analysis method that makes use of various noninstantaneous flash heating mathematical models, with each model fitted to the experimental measurements by nonlinear regression. These models not only accommodate different heating rates, distinguished by the shape of the flash as a function of time, but allow for variation in the start time of the flash, to account for any inaccuracies in the triggering mechanism used in the flash diffusivity instrument. A significant reduction in the standard deviation of the residuals was achieved using this method. Moreover, the residual signature was greatly reduced when compared with a model that did not account for various flash shapes and start times of the flash.

Description of Experiment

The material selected for this research was National Institute of Standards and Technology standard reference material SRM-1461 stainless steel. Three samples were cut from and the same rod to insure uniformity of properties. The thicknesses of the three samples were 0.683, 1.295, and 2.703 mm. The rationale behind testing multiple-thickness samples of the same material was that all of the samples should result in the same thermal diffusivity in each experiment, independent of sample thickness. Experiments were performed at the Thermo-Physical Properties User Center at the High Temperature Materials Laboratory at the Oak Ridge National Laboratory. The flash diffusivity instrument used was manufactured by Anter Corporation. The instrument used a neodymium glass laser with a wavelength of 1.053 μm. The duration of the flash was thought to be nominally 3 ms, which is normally varied by the instrument automatically, along with the intensity of the flash. The flash duration and intensity were selected by software provided with the Anter instrument. This feature is intended to accommodate differences in sample thickness and volumetric heat capacity so that the temperature rise of the nonheated side is within a consistent range. The instrument normally performs several preliminary shots with the laser for each sample before performing the final recorded experiment. The preliminary data is used by the instrument to optimize the duration and intensity of the flash for each sample tested. Three tests were performed on each sample and the results of the tests showed to be very consistent.

The experiments were performed at 300°C with the samples in an oven, provided as part of the flash diffusivity instrument. The instrument automatically computes the thermal diffusivity of each sample using the method of Parker et al. [1] at the completion of each experiment. Although there is no published thermal diffusivity for this material, using the published thermal conductivity and dividing by volumetric heat capacity measured in the laboratory, a diffusivity of 4.62 ± 0.42 mm^2/s was obtained for this material.

Analysis Method

In contrast to the analysis methods described in [1–4], the present method uses a temperature calculation extending over the entire duration of the experiment. Employing the method of least-squares, the parameters are adjusted until a best fit is found, using the principles of [17]. The method used for minimizing the sum of squares of the errors was the generalized reduced gradient nonlinear optimization method developed by Lasdon et al. [18]. Using this tool, three of the parameters were iteratively adjusted to arrive at a fit producing a minimum sum of squares of errors. The direct solution was a one-dimensional configuration solved using Green's functions to find $T(L, t)$, which is the transient temperature at $x = L$, the nonheated side of the sample. In this expression, L is the sample thickness (m). The transient conduction equation solved is

$$\frac{1}{\alpha}\frac{\partial T}{\partial t} = \frac{\partial^2 T}{\partial x^2} \tag{1}$$

where α represents thermal diffusivity (m^2/s). For convenience, the diffusivity resulting from the experiments is displayed in terms of mm^2/s. The boundary conditions for this experiment are

$$q(0, t) = f(t) \tag{2}$$

and

$$q(L, t) = 0 \tag{3}$$

where $f(t)$ is an arbitrary function of time. Although a small amount of cooling takes place in the experiment following the flash, the time scale of the experiment is a maximum of 0.5 s, during which time a negligible amount of cooling takes place. Previous analyses of metal samples have shown no advantage in reduction of the residuals by the addition of the heat loss parameter. Moreover, in examining the raw data, the cool down rate linear fit is 0.0402 deg/s after the peak temperature of 0.704 deg is reached. With the period of analysis lasting only 0.5 s, the maximum possible effect of cooling is less than a 3% change in measured temperature, which is comparable to the measurement noise. The initial conditions in the formulation of the model are

$$T(x, 0) = 0 \tag{4}$$

Since the measurement instrument records only temperature rise above ambient, the ambient temperature is set to zero for convenience.

To obtain the boundary condition given by Eq. (2), the principle of superposition was used from a generic solution generated combining multiples of

$$q(0, t) = 1 \tag{5}$$

This is not to say that the heat flux is constant by any means, but that this heat flux is a fundamental building block employed in superposition. The solution generated from this differential equation and these boundary conditions, using the method of [19], is

$$T(L, t) = \frac{1}{\rho c L}\left[t - 2\sum_{m=1}^{\infty}\frac{(-1)^m L^2}{\pi^2 m^2 \alpha}\left(1 - e^{-\pi^2 m^2 \alpha t / L^2}\right)\right] \tag{6}$$

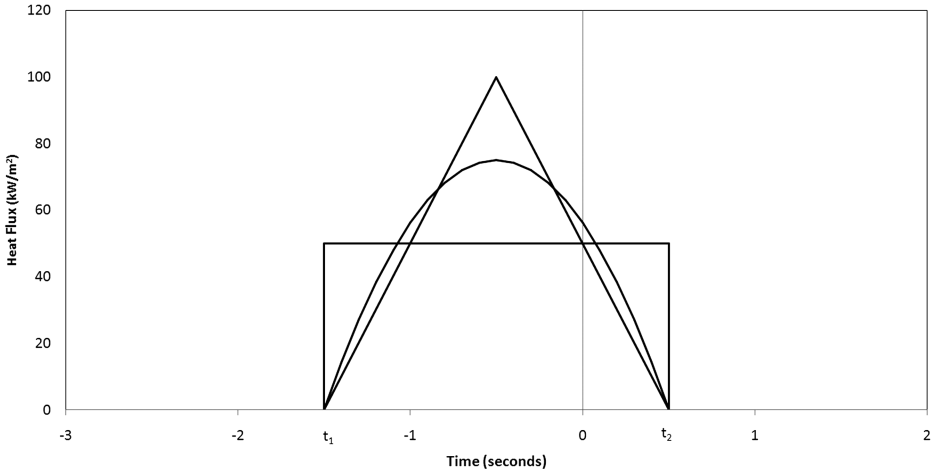

Fig. 1 Plot of the three pulse shapes tested. Shapes shown are of duration 2 ms depositing 100 $\mathbf{J/m^2}$ of energy on the sample face.

In this equation, L is the sample thickness (m), ρ is density ($\mathrm{kg/m^3}$), and c is specific heat ($\mathrm{kJ/kg\text{-}K}$). The volumetric heat capacity, ρc, is unknown and unneeded in this formulation, since the thermal diffusivity is the only parameter of interest. The shape of the temperature vs time curve is the only information needed in order to find thermal diffusivity. The magnitude of the heat flux is estimated but is kept in an arbitrary group of parameters as $q/\rho cL$ and is estimated as part of the analysis.

There were three pulse shapes considered as part of this research for the whole-domain least-squares fit models. These three pulse shapes are shown in Fig. 1 as rectangular, triangular, and parabolic. Notice that each of these pulses integrates to the same area which, in this case, is 100 J, just for the sake of example. The pulse energy of the model must integrate to the same value, regardless of the pulse shape, for any given experiment. This is because each theoretical pulse must generate the same net temperature rise in the sample for a given experiment. In each of the example cases shown, t_1 represents the time of the start of the pulse and t_2 represents the time of the end of the pulse. Each of these values was assumed as an independent parameter, along with thermal diffusivity. The overall magnitude of the heat pulse was determined separately and subsequently to these other three parameters. This is because the heat pulse magnitude is a linear parameter and nonlinear iteration is not required. Estimating the heat pulse magnitude in this fashion increases the speed and stability of the nonlinear regression analysis, since fewer nonlinear parameters must be found. This gives a total of four parameters in the model that were determined: α, t_1, and t_2 simultaneously by nonlinear least-squares fit and $q/\rho cL$ by linear least-squares fit. During each iteration involving changes in one of the first three parameters, the heat flux magnitude was calculated.

Results

The finite-pulse-width model was tested on each of the three stainless steel samples and the residuals were compared from these curve fits to determine model adequacy. Figure 2 shows a typical plot of the raw data from the experiment alongside the curve generated by the mathematical model using the Parker et al. [1] method. The data in this curve happens to be from the 2.703-mm-thick sample. As can be seen in this figure, the measured temperature exhibits a rise above ambient much sooner than is predicted by the model. As would be expected from physical insight, a rise in temperature would be anticipated after some time has elapsed from the moment of the flash. This would allow time for the diffusion of the heat to take place, bringing about a temperature rise on the unheated side. As shown in Fig. 2, however, heating has started much earlier than the initial time designated as time zero by the flash diffusivity instrument.

As a test to investigate whether the ambient temperature was not being properly zeroed by the instrument, an analysis was run with ambient temperature as a parameter to be estimated. In other words, instead of only estimating four parameters in the model (namely, thermal diffusivity α heat pulse magnitude, $q/\rho cL$ flash start time t_1,

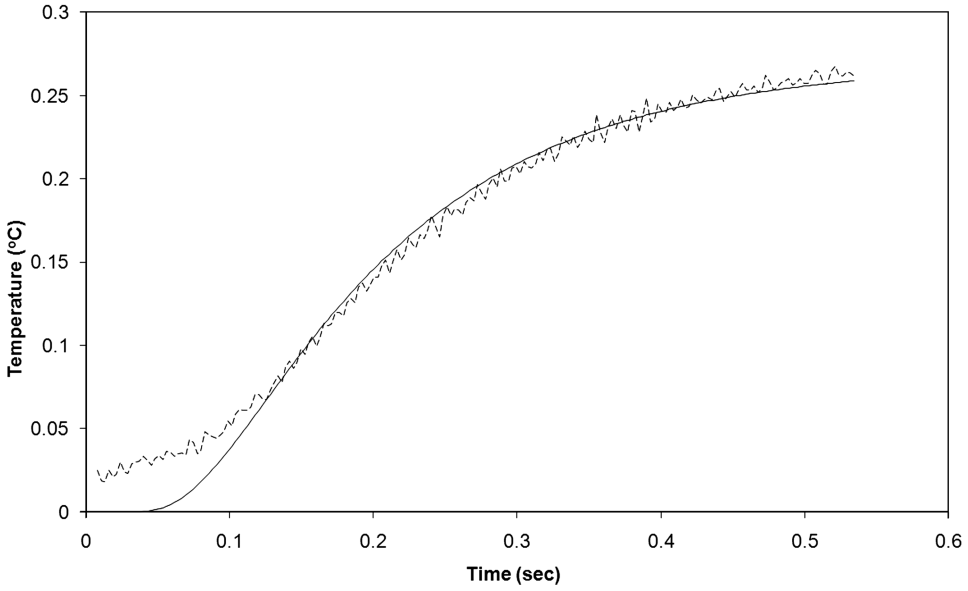

Fig. 2 Temperature as a function of time on the nonheated side of the sample. The experimentally measured temperature is shown as the jagged line and the mathematical model generated using [1] is the smooth line. Heating before time zero is evident in the measured data.

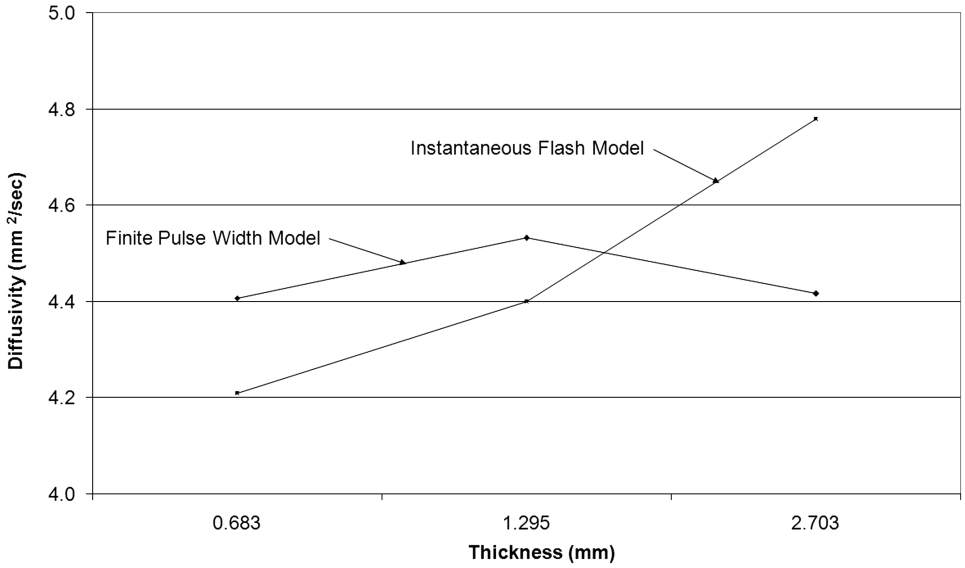

Fig. 3 Comparing estimated thermal diffusivity using two methods. The diffusivity based on the published value for thermal conductivity of this material is 4.62 ± 0.42 mm^2/s.

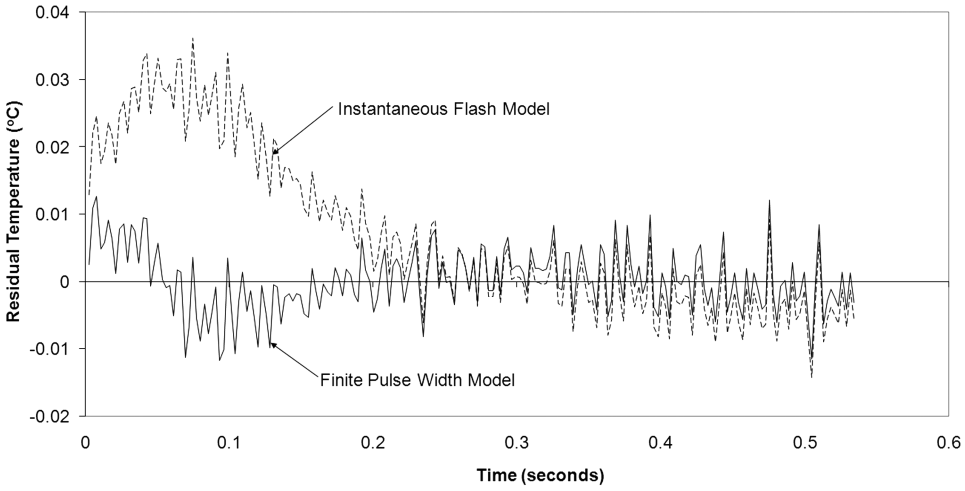

Fig. 4 Comparing residuals from the two methods.

and flash end time t_2), a fifth parameter was also simultaneously estimated, which was ambient temperature T_o. The results from applying this model to the experimental data provided no additional gain in conformance to the data and the estimated ambient temperature was extremely close to the same zero point measured by the instrument. This test showed that heating had definitely started before the time $t = 0$ designated by the instrument.

Next, the finite-flash-duration models were applied to the data. All of the finite-pulse-width models gave better results than the instantaneous flash model in terms of a reduction in the standard deviation of the residuals, and in terms of a reduction in residual signature. No single model, of the pulse shapes chosen, stood out from the other in terms of the residuals when applied to the experimental data. The standard deviation of the residuals ranged from 0.00327 to 0.00977°C for the 0.683-mm-thick samples, 0.00485 to 0.00801°C for the 1.295-mm-thick samples and 0.00385 to 0.00556°C for the 2.703-mm-thick samples. Since the three pulse shapes employed in the models gave similar results and, since the rectangular pulse was a slightly simpler and faster than the triangular- or parabolic-pulse-shaped models, it was used as the preferred model. Figure 3 shows a summary of the thermal diffusivity results for the three thicknesses of the stainless steel samples generated by the two primary mathematical models used in this analysis. As can be seen in this figure, the mathematical model accounting for the duration of the flash produced results that were much more consistent than those produced using the method of [1]. The same thermal diffusivity would be expected for each of the samples, since each sample was cut from the same bar of stainless steel material. The primary means of comparing the adequacy of the models is by comparing the root mean square of the residuals for each of the results. The residuals are the individual differences between the measured data points and the calculated data points for each of the measurements in the experiment. As this root-mean-square deviation of the residuals becomes smaller, the model conforms more closely to the experimental data. If the model does not conform well to the experimental data, a characteristic signature will appear in the plot of

Table 1 Results using the square-pulse model

Thickness, mm	Diffusivity, mm^2/s	Start time, ms	End time, ms	$q/\rho cL$, °C/s	RMS dev, °C
0.683	4.407	−1.649	2.904	194.5	0.006042
1.295	4.532	−2.562	5.768	89.6	0.004975
2.703	4.416	−147.4	80.61	3.135	0.005183

Table 2 Results using the instantaneous flash model as given in Parker et al. [1]

Thickness, mm	Diffusivity, mm^2/s	RMS dev, °C	Max measured temp, °C
0.683	4.21	0.009225	1.407
1.295	4.4	0.005398	0.603
2.703	4.78	0.013997	0.263

the residuals. Figure 4 shows a plot of the residuals for the 2.703-mm-thick test case, comparing the residuals generated using the method of [1] and the residuals generated from the square-pulse model. This figure shows much better conformance to the experimental measurements obtained using the finite-pulse-width model. In this figure, a dip can be seen in the very early time steps. This could be due to voltage fluctuations in the measurement system at the time of the flash or possibly some other physical effect unaccounted for by the model. However, these fluctuations are quite small in comparison to the residuals generated from the model that does not account for the time duration of the flash.

Table 1 shows the results from the finite-pulse-width model in tabular form. The consistency of the results for the estimated thermal diffusivity can be seen in this table, consistent with Fig. 3. The flash duration tends to increase with the thickness of the material, which is a function of the automatic controls of the flash diffusivity instrument. The magnitude of the flash likewise decreases as sample thickness increases. However, the total amount of energy deposited on the sample remains nominally the same for all of the sample thicknesses. The overall temperature rise encountered in each experiment, as a result, can be seen to drop as sample thickness increases. The overall temperature rises are nominally 1.4, 0.6, and 0.26 deg for the sample thicknesses of 0.683, 1.295, and 2.703 mm, respectively. The experiments for the thicker samples, therefore, come with a lower signal-to-noise ratio and can sometimes be slightly more difficult to analyze.

Table 2 provides the results for the model that assumes an instantaneous flash. The most significant means of comparison between the models is the root-mean-square deviation of the residuals, which quantifies the degree to which the mathematical model matches the experimental measurements. A reduction in the residuals for each of the three thicknesses tested can be seen in these results.

Conclusions

Three stainless steel samples of varying thicknesses were tested in flash diffusivity experiments. Four different mathematical models were applied to the data, pursuant to finding the effects of pulse duration and pulse shape on the analysis results. One of the mathematical models assumed the flash was instantaneous and occurred at the zero point on the time scale of the experiment. The other three models assumed that the flash was of some noninstantaneous duration, and this duration was a parameter that was estimated as part of the analysis of the experimental data. Additionally, the start time of the flash was estimated as a parameter, so that four parameters were estimated simultaneously: thermal diffusivity, pulse magnitude, pulse start time, and pulse end time. The three pulse shapes tested were rectangular, triangular, and parabolic.

The analysis showed that the thermal diffusivity determined for the three samples was more consistent using the finite-pulse-width models. Although accounting for finite pulse width is unnecessary for samples with low thermal diffusivity, due to the longer duration of the experiments, the pulse duration can become a more significant effect in thin highly conductive materials.

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