Simulation and Modeling of Laser Flash Analysis of a Polymer Composite

Tom Mulholland
University of Wisconsin–Madison, Madison, WI

Abstract

Thermally conductive polymers can be easily produced by compounding conductive fillers into the polymer. This can be done to produce filament for an additive manufacturing system. However, conventional methods are insufficient for measuring the thermal conductivity of the filament without altering the fiber orientation within the polymer. This article explores methods to determine the thermal diffusivity of a conductive polymer embedded in an epoxy resin.

Introduction

A thermally conductive polymer composite of polyamide 6 (PA6) and copper fibers was developed for use in fused filament fabrication (FFF), an additive manufacturing technique. The copper fibers have a diameter of 30 µm and a length up to 3 mm, giving them an aspect ratio as high as 100. The fillers become highly oriented during compounding and filament production, leading to a high thermal conductivity in the extrusion direction, and a lower conductivity perpendicular to that.

One of the most common techniques for measuring the thermal diffusivity of a material is Laser Flash Analysis (LFA), which uses a flash lamp to deliver an energy pulse to one side of a sample, while simultaneously measuring the temperature rise on the back side of the sample with an infrared camera. The thermal conductivity can then be calculated from the product of the thermal diffusivity, the specific heat capacity, and the density.

In a single material system, the thermal diffusivity can be calculated from the well-known Parker model [1], along with other models, using LFA. For curve fitting of the LFA signal, the response on the back side of the sample can be taken as $V(t) = T(t) / T_M$, where T_M is the maximum temperature achieved, then

$$\omega = \frac{\pi^2 \alpha t}{L^2} \tag{1}$$

$$V = 1 + 2\sum_{n=1}^{\infty}(-1)^n e^{-n^2\omega} \tag{2}$$

where α is the thermal diffusivity and L is the sample thickness. Then, it can be shown that

$$\boldsymbol{\alpha = \frac{1.38L^2}{\pi^2 t_{1/2}}} \tag{3}$$

where $t_{1/2}$ is the time for the back surface of the sample to reach half of the maximum temperature.

A typical sample size for LFA of polymer systems is 10 mm x 10 mm x 1 mm, which is required to completely cover the measurement window. This is several times wider than standard FFF filament, which is typically 1.75 mm or 2.85 mm in diameter, so sample preparation is required to measure the thermal properties of the compounded filament. Although samples were successfully prepared by compression molding, and the thermal diffusivity could be measured parallel and perpendicular to the extrusion direction, compression molding alters the copper fiber orientation within the filament.

In order to measure the thermal diffusivity of the filament without altering the copper fiber orientation, several filament strands were aligned together and impregnated with an epoxy resin. Details on the sample preparation method are available in [2]. However, the typical models used for calculating the thermal diffusivity of the LFA samples could not properly fit the resulting data for the composite system (epoxy and filament).

This article probes several methods for estimating the thermal diffusivity of the composite conductive filament, PA6-copper fiber in this case, embedded in an insulating matrix, epoxy in this case. In all cases, measurements and simulations were done for the extrusion direction, parallel to the orientation of the fibers.

Simulation

ANSYS was used to simulate a 1/8 scale model of a filament-epoxy LFA sample. The filament was placed within the epoxy at a volume fraction of 0.7, with the copper fiber orientation in the direction of measurement. A model of a unit cell for hexagonal packing of the composite, which is discussed later in this article, was also analyzed for comparison, which contained a single filament strand surrounded by a cylinder of epoxy. The models are shown below in Figure 1. The parameters for the models are given in Tables 1 and 2.

The models were meshed uniformly, using the maximum number of elements through the thickness that was permitted by the software. Radiation and convective loads were applied to the top and bottom surfaces, with a convection coefficient of 5 W/m^2-K. The energy pulse was applied by raising the temperature of the bottom nodes by an amount equal to

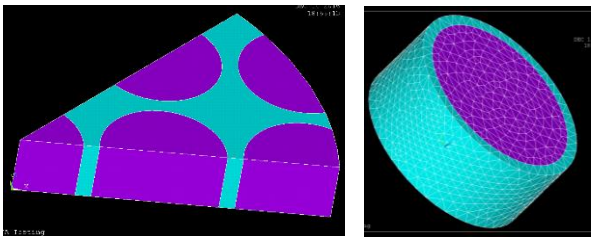

Figure 1. ANSYS models of filament (purple) embedded in epoxy (teal). Left: 1/8 model. Right: Unit cell.

Table 1. Material properties for the simulation.

Property	Units	Filament	Epoxy
Specific Heat Capacity	J/kg-K	1336	2383
Density	kg/m^3	2690	1083
Conductivity, Parallel	W/m-K	7.48	0.20
Conductivity, Perpendicular	W/m-K	1.94	0.2

Table 2. Geometry of the ANSYS models.

Model	Units	1/8	Unit Cell
Sample Diameter	mm	10	1.14
Filament Diameter	mm	2.00	1
Volume Fraction of Filament	-	0.7	0.7
Sample Thickness	mm	1	1
Energy Pulse Duration	ms	0.31	0.31
Number of Nodes	-	19424	28986

the volume of the bottom elements multiplied with the density, specific heat capacity, and energy of the pulse, E, for the duration of the pulse, as shown in Equation (4) below.

$$\Delta T = V_{elem}\rho cE \quad (4)$$

Because of the physics of the problem, the simulation was also attempted by applying a constant heat flux to the bottom surface for the duration of the energy pulse, but this caused problems with the FEM solvers, probably due to the very large heat fluxes during the very short energy pulse.

The average temperature over time on the top surface was recorded, and Equation (3) was used to compute the thermal diffusivity. In order to check the accuracy of the model, simulations were run with 100% epoxy and with 100% filament, and the resulting diffusivities were within 5% of the input values. For the composite, the filament diffusivity was adjusted until the resulting composite diffusivity matched the LFA measurement.

Figure 2 shows the resulting plots of dimensionless temperature and dimensionless time for the two models

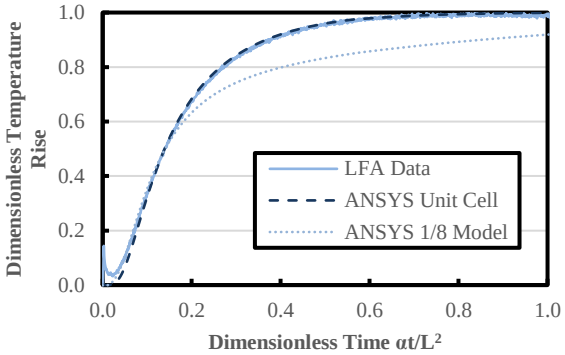

Figure 2. Dimensionless temperature vs. dimensionless time plots for the LFA data and two ANSYS models. The 1/8 model does not capture the shape of the curve.

Table 3. Predicted diffusivity in LFA tests and in the ANSYS simulations. Filament diffusivity was adjusted until the simulated composite diffusivity matched the LFA tests.

Data Source	Diffusivity [mm^2/s]
LFA of Filament (Compression Molded)	2.08
LFA of Composite	1.18
Filament (Unit Cell)	1.4
Simulated Composite (Unit Cell)	1.13
Filament (1/8 Model)	1.8
Simulated Composite (1/8 Model)	1.16

and measured LFA data, and Table 3 summarizes the results.

The 1/8 model is not able to predict the shape of the response curve very well, but it predicts a higher thermal diffusivity of the filament. It's worth noting that it does predict virtually the same time of the half temperature rise, $t_{1/2}$, which per Equation (3) is the variable that matters for calculating the diffusivity.

The unit cell predicts the LFA data curve shape very well, although the predicted filament diffusivity is lower than expected.

Modeling

A number of models exist for predicting the thermal conductivity of composites. The most basic models consider only the thermal conductivities of the matrix and filler, k_m and k_f, respectively, along with the filler volume fraction, f. The series model, also known as the rule of mixtures, as well as the parallel model and geometric model are given in Equations (5-7) [3].

$$k_{eq} = (1-f)k_m + fk_f \quad (5)$$

$$\frac{1}{k_{eq}} = \frac{1-f}{k_m} + \frac{f}{k_f} \quad (6)$$

$$k_{eq} = k_f^f k_m^{(1-f)} \quad (7)$$

The Lewis-Nielsen model [4] is also frequently employed because of its relatively simple application.

$$\frac{k_{eq}}{k_m} = \frac{1+ABf}{1-B\psi f} \quad (8)$$

$$B = \left(\frac{\lambda-1}{\lambda+A}\right) \quad (9)$$

$$\psi = 1 + \left(\frac{1-\phi_m}{\phi_m^2}\right) f \quad (10)$$

$$\lambda = \frac{k_f}{k_m} \quad (11)$$

$$A = k_e - 1 = 2\frac{L}{D} - 1 \quad (12)$$

ϕ_m is the maximum volume fraction for the packing configuration, which is 0.907 for hexagonal packing, k_e is the Einstein coefficient, given by [5] as 2L/D for fibers parallel to the heat flow. Table 4 gives the thermal conductivity measured in compression molded samples, along with the conductivities predicted by the models above.

Table 4. Thermal conductivity of filament predicted by different models, and measured on a compression molded sample.

	Thermal Conductivity [W/m-K]
Composite Conductivity - LFA	3.88
Series Model - Filament	5.45
Parallel Model - Filament	-0.58
Geometric Mean - Filament	13.69
Lewis-Nielsen - Filament	∞
Compression Molded Filament	7.48

The composite conductivity is calculated from the LFA data, using a well-known correction for the density and specific heat capacity.

$$(\rho c)_{eq} = f(\rho c)_f + (1-f)(\rho c)_m \quad (13)$$

$$k_{eq} = \alpha_{eq}\rho c_{eq} \quad (14)$$

The Lewis-Nielsen model predicts an equivalent composite conductivity around 1 W/m-K, but the filament conductivity tends to infinity when solving the equivalent conductivity to match the measured value.

The standard thermal conductivity models fail to predict filament thermal conductivity in the composite system, probably because the heat transfer in LFA is transient and two-dimensional. Therefore, a new model is proposed for fitting the LFA data for the composite and predicting the thermal diffusivity of the filament.

The new model is based around the idea of thermal "streamlines" along which the transient heat flows. Each streamline has a different equivalent thermal resistance and path length, so the LFA response can be modeled as a spectrum of thermal diffusivities. By relating the spectrum diffusivities only to the thermal properties of the filament and epoxy, the unknown diffusivity of the filament can be fit and predicted.

For hexagonal packing of cylinders, Figure 3 shows the filament radius, *R*, and equivalent coated radius, *r*, which is given by [6]:

$$r = R\sqrt{\frac{\pi}{2\sqrt{3}f}} \quad (15)$$

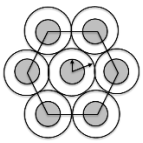

Figure 3. Depiction of hexagonal packing of cylinders.

From this geometry, the theoretical spectrum of streamlines can be sketched. *L* is the sample and filament thickness, L_j is a series of total path lengths, and theta is the angle the heat flows through the epoxy. This is taken as 45°, since the epoxy is isotropic, The resistance to heat flow through the filament is 20 to 40 times lower than through the epoxy, so most of the heat should tend to flow through the filament.

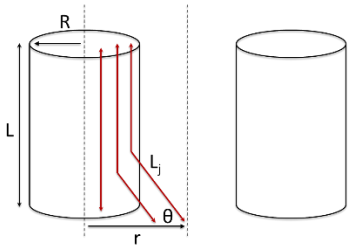

Figure 4. Theoretical heat flow paths in the filament-epoxy composite.

$$l_j = \frac{\Delta rj}{\cos(\pi/4)} + (L - \Delta rj) \quad (16)$$

where l_j is the path length for the jth term and *L* is the sample thickness. Then,

$$\Delta r = \frac{r-R}{J} \quad (17)$$

where the series runs from j=0 to J.

A weighting factor spectrum can be defined, similar to the volume fraction, based on the path length l_m, such that

$$w_j = \frac{L-\Delta rj}{l_j} \tag{18}$$

For conduction through composite walls [7],

$$k_{avg} = \left(\frac{f}{k_f} + \frac{1-f}{k_m}\right)^{-1} \tag{19}$$

If we use the length fraction along the streamline in each material, instead of the total volume fraction, then the thermal resistance to conduction along each streamline is

$$R_j = \frac{w_j l_j}{k_f} + \frac{(1-w_j)l_j}{k_m} \tag{21}$$

where R_n is the thermal resistance. Similarly, based on Equation (13),

$$(\rho c)_j = w_j\,(\rho c)_f + (1-w_j)(\rho c)_m \tag{22}$$

The equivalent thermal conductivity along the streamline can be calculated from the definition of thermal resistance to conduction as

$$k_j = \frac{l_j}{R_j} \tag{23}$$

Then,

$$\alpha_j = k_j(\rho c)_j \tag{24}$$

As a check, when j = 0, $R_j = L/k_f$ and $\alpha_j = k_f(\rho c)_f$, which is expected.

Now, the dimensionless temperature rise, V, can be predicted using a summation of the responses to the spectrum of diffusivities by modifying Parker's model.

$$\omega_j = \frac{\pi^2 \alpha_j t}{{l_j}^2}$$

$$V = 1 + 2\left[f\left(\sum_{n=1}^{\infty}(-1)^n e^{-n^2\omega_0}\right) + \frac{(1-f)}{J-1}\sum_{j=1}^{J}\sum_{n=1}^{\infty}(-1)^n e^{-n^2\omega_j}\right]$$

The first term, corresponding to the diffusion through the filament, is weighted by the volume fraction of the filament. The rest of the terms are weighted equally. The model can then be fit to experimental data. Since the only unknown is the diffusivity of the filament, it can be found from the best fit model.

The initial diffusivity predicted by the model was higher than 20 mm^2/s, while the diffusivity of the compression molded filament is 2.08 mm^2/s. However, the model was being fit to data that includes an initial spike, which is caused by radiation passing completely through the translucent epoxy sample during the LFA test. When this spike is removed, the results are more reasonable, with a predicted diffusivity of 1.46 mm^2/s. The Parker model predicts a diffusivity of 1.12, which was the reported value for the composite, but does not predict the diffusivity of the filament. Figures 5-7 show the effect of adding more terms, the model error, and the fit of LFA data for the composite.

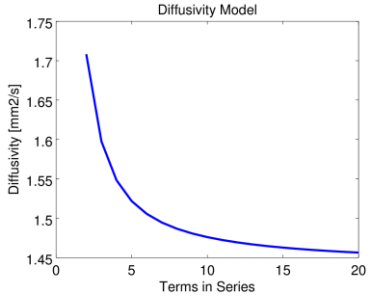

Figure 5. The predicted diffusivity decreases asymptotically with increasing terms.

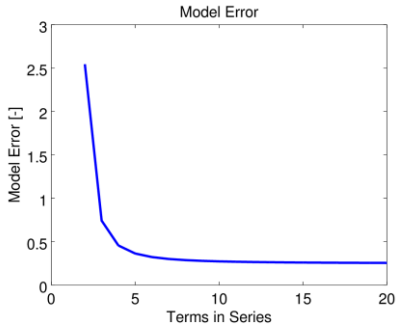

Figure 6. The model error decreases asymptotically with increasing terms.

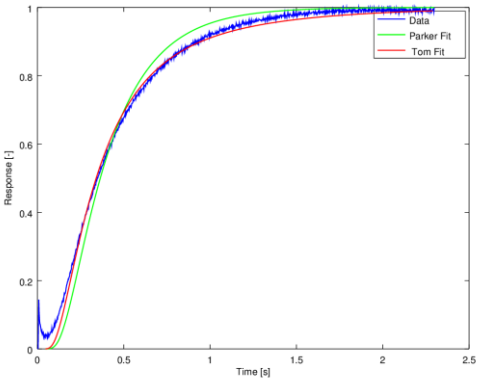

Figure 7. The model fits the data better than the standard Parker model when the initial spike is ignored.

Conclusions

An FEA simulation developed in ANSYS was able to predict the thermal diffusivity of single-material systems. When applied to the composite system, the 1/8 model and the unit cell model provided different results, but they were similar to the diffusivity measured in LFA tests. By adjusting the filament diffusivity until the apparent diffusivity matched the measurements, the filament diffusivity could be estimated between 1.4 and 1.8 mm^2/s, depending on the model.

Theoretical models for the thermal conductivity of composites were unable to predict reasonable filament conductivities for the composite system, probably due to the transient, 2D nature of the LFA test. The model developed in this work, based on the Parker model, can predict the thermal diffusivity as well as the FE model. Because the model is derived entirely from the geometry of the problem and the thermal diffusivity of the filament, the weighting factors may possibly be adjusted to give more accurate results, but this is left for future work.

The LFA data for the composite show that the epoxy is relatively translucent, which allows some of the energy pulse to be absorbed initially into upper layers of the sample. To increase the accuracy of the analytical model, it could be adjusted to the standard LFA fitting model that accounts for radiation.

However, without an experimental method to evaluate the thermal diffusivity of the filament without altering the internal copper fiber orientation, it is difficult to evaluate the absolute accuracy of the model.

References

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