Transport involving conducting fibers in a non-conducting matrix

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Abstract

Several transport models are investigated in the context of two-dimensional fiber composites. We compare the thermal and electrical transport through the fiber composites as a function of the conductivity ratio and the fiber density. Three models will be considered and compared: 1) discretized solutions, 2) equivalent resistance, and 3) effective medium approximation. In the case of electrical transport, where the conductivity of the fiber is presumably many orders of magnitude larger than the matrix, the second model provides a fast and reliable way to predict conductance of the combined system. However, if the two materials are similar in conductivity, the second model fails to accurately capture the conductivity. Thermal transport is predicted using the discretized model because the conductivity ratio is non-negligible. The third model is an analytic approximation based on Maxwell's equation and is used to predict both types of transport through a compound with inclusions of ellipsoidal geometry. The analytic model works well for lower conductivity ratios and all area densities but under-predicts conductivity for high conductivity ratios.

Key words: percolation threshold, conducting fibers, thin-film devices

1. Introduction

Thermal and electrical transport through a low-conductivity matrix containing high-conductivity fibers is important to several applications including flexible thin-film transistors (TFT) [1], proton exchange membranes (PEM) [2], and direct-energy conversion devices [3].

For flexible TFTs, low-temperature processes are required to prevent destruction of the substrate, but most semiconductors with good electrical performance require high-temperature processing[4]. Most research has been directed toward finding compatible high-temperature substrates [5] or high-performance electronic materials with low processing temperatures [6, 7]. Another approach is to combine the good electrical performance of nanofibers into flexible substrates. In fact, Biercuk et al. [8] have shown that nanotube/epoxy composites percolate at 0.1–0.2 wt% loading. This feature suggests that composite materials may retain flexibility while providing good electrical performance with low processing temperatures. In direct energy conversion devices—particularly Peltier devices—high electrical conductivity and low thermal conductivity are preferred for superior performance [9]. However, most materials do not exhibit both of these properties simultaneously and strategies for tuning the material properties are being sought [10]. For example, composite structures involving high conductivity fibers and a low conductivity matrix could improve the performance of direct energy conversion devices. Nanofibers can limit the thermal transport through

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phonon confinement and boundary scattering while maintaining high electrical conductivity [3, 11]. The substrate, while not contributing to the electrical performance, will also limit thermal transport [12]. The net result is a material with high electrical conductivity and low thermal conductivity.

If we consider the fiber composite more carefully, we also notice that the conductivity ratio for thermal and electrical performance is different by orders of magnitude. Table 1 illustrates this feature for a composite composed of fibers (calcined needle coke Fl08) embedded in a thermoplastic copolymer matrix (Vectra A950RX) [13]. Because the transport is a function of the conductivity ratio, we expect that different concentrations of fibers in the matrix will affect the thermal and electrical transport in different ways. In fact, Zimmerman [14] has shown that as the aspect ratio of fibers increases, the transport properties of the compound become more sensitive to the conductivity ratio.

The purpose of the present work is to investigate the effectiveness of several transport models in the context of two-dimensional fiber composites and to compare the thermal and electrical transport through a given material as a function of the conductivity ratio, fiber aspect ratio and the fiber density. Because the conductivity ratio of the fiber to the matrix is different when considering thermal transport compared to electrical transport, the rate of conductivity enhancement with fiber density should also be different. If the thermal and electrical properties can be decoupled, then the opportunity for designing improved TFTs and energy conversion devices can be identified.

Our analytic model is an effective medium approximation for a compound with inclusions of ellipsoidal geometries. This approximation works well to predict the total conductivity of a compound material with a low density of inclusions. However, the approximation underestimates the conductivity at higher fiber densities because the model does not account for fiber overlap. Numerical models are well suited for iteratively solving for transport and better approximate for fiber-fiber overlap. Thermal transport is determined using a percolation model. Fibers are placed randomly on the surface of the device. When a direct path of fibers is established between the two contacts, the compound is said to percolate. This model works well to determine thermal transport because the conductivity ratio of fiber/matrix is $O(10^3)$. Thus, thermal transport is predicted by a discretization model, which superimposes a small square mesh on the device. Conductivity is determined by solving a system of linear equations which obey Kirchoff's law at each node of the network. Electrical transport is predicted by assuming that the matrix does not contribute to transport because the conductivity ratio of fiber/matrix is $O(10^{16})$. A resistor is placed between any two fiber intersections and Ohm's law is used to relate the current through the resistor to the potential across the resistor.

Table 1: Conductivity ratios of the fiber to matrix. Data obtained from King et al. [13]

	Thermal (W/mK)	Electrical (1/Ωm)
matrix	0.2	1×10^{-14}
fiber	600	1×10^{2}
ratio	3×10^{3}	1×10^{16}

2. Transport Models

2.1. *Effective Medium Approximation*

Most effective medium approximation (EMA) models predict the effective conductance of a composite material where severe restrictions are placed on the inclusion geometry and material properties. For example, Maxwell's model is valid for circular inclusions that are randomly distributed and non-overlapping. This approach has been shown to be valid only for low concentrations [15]. This model however has been extended to ellipsoidal geometries where the thermal conductivity of the compound k can be found as

$$\frac{k}{k_m} = \frac{1-\beta c}{1+\beta c}, \tag{1}$$

where c is the area fraction of fibers to matrix material, and

$$\beta = \frac{(1-r^2)(1+\alpha)^2}{4(1+\alpha r)(\alpha+r)}. \tag{2}$$

In the foregoing expression, the matrix material thermal conductivity is k_m, and the ellipse is described by the major axis radius, and α is the aspect ratio defined as the fiber width (w) divided by the fiber length (L_f). The thermal conductivity ratio ($r = k_f/k_m > 1$) is the conductivity of the fiber divided by the thermal conductivity of the matrix. This method can be used to approximate an inclusion of arbitrary aspect ratio including fibers ($\alpha \to 0$). In the limit of extremely narrow fibers compared to their length, the fiber-density parameter na^2 becomes a more meaningful independent variable than the area fraction [16], where a is the fiber length normalized by the device size. Note that a non-trivial limit can only be obtained for a conductivity ratio of $r \to \infty$. In this case, the conductivity of the compound becomes

$$\frac{k}{k_m} = \frac{1+n\pi a^2/4}{1-n\pi a^2/4}. \tag{3}$$

This form may be appropriate for the electrical transport, but equation 1 should be used for thermal transport where the conductivity ratio is smaller. Zimmerman [14] further provides a comparison between the extended Maxwell's model and the differential method, which gives an implicit form (stated without derivation).

$$\frac{1}{1-c} = \left(\frac{k}{k_m}\right)^{\frac{2\alpha}{(1+\alpha)^2}} \left(\frac{k_m-k_f}{k-k_f}\right)\left(\frac{k+k_f}{k_m+k_f}\right)^{\left(\frac{1-\alpha}{1+\alpha}\right)^2} \tag{4}$$

This implicit formulation will be used to compare to the numerical solutions.

Note that Maxwell's model does not account for overlap of the fibers, but the differential method does statistically. Therefore, Maxwell's method is strictly only valid in the dilute limit, but conceivably the differential method applies for a wider range of fiber densities. In addition, Maxwell's model is for inclusions with ellipsoidal geometry and not for fibers. Because the fibers have a larger area for the same aspect ratio compared to ellipsoids, the fiber density will not be accurate using Maxwell's model. For comparison, we have used two numerical models which will increase the accuracy of the simulation for conducting fibers in a polymer matrix.

2.2. *Thermal Transport*

Thermal transport was determined by creating a numerical model where the device is discretized into an equivalent resistor network of uniformly spaced resistors. Fibers are randomly-oriented on the surface and a square mesh is superimposed on the composite device. Figure 1 shows how the thermal resistances are defined relative to the discretized material description. The mesh size is defined smaller than the width of the fibers so that several nodes lie within each fiber in both directions. The thermal resistance is defined as $R_i = L/k_i A$, where the conductivity is designated by the material of the cell. For all resistances, the length of the resistor L and the cross sectional area of the cell A are identical. Therefore the ratio of resistances is related to the conductivity ratio ($R_m/R_f = r$). A sample discretized device is shown in Figure 2. Note that to include thermal contact resistance between materials, the overall resistance between any two adjacent nodes is

$$R = \begin{cases} 2R_m & \text{both matrix} \\ 2R_f & \text{both fiber} \\ R_m + R_f + R_c & \text{otherwise} \end{cases} \tag{5}$$

where R_c represents the contact resistance. The value of R_c is largely unknown and can depend on a wide number of variables including materials and processing parameters. Therefore, a representative value is difficult to define. Consequently, we have assumed that the contact resistance is negligible and left this study for another time.

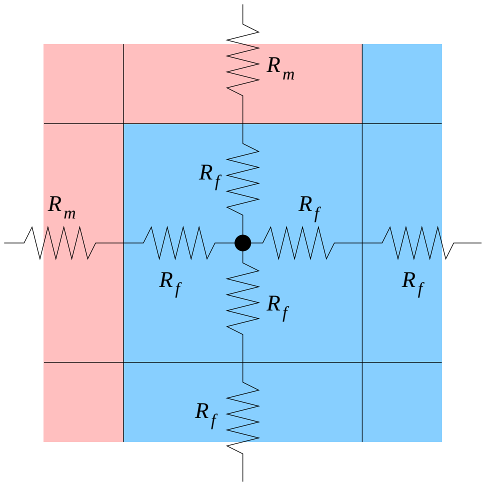

Figure 1: One example cell with surrounding cells is shown. The different colors represent different material types for each cell, and the corresponding resistances are indicated R_f for fiber resistance and R_m for matrix resistance. The temperature is fixed at the center of the cell.

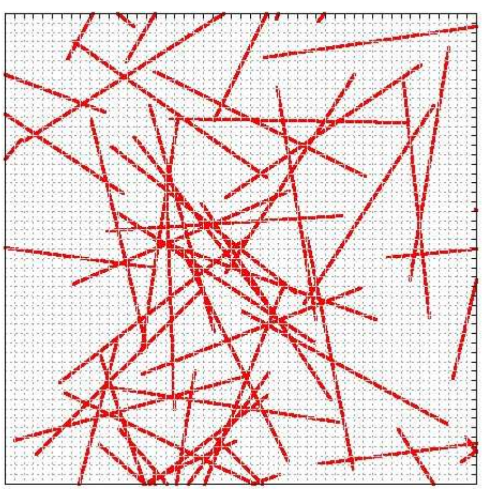

Figure 2: Discretization of equivalent resistance for $n = 50$ and $L_f = 0.5$.

A system of linear equations obeying Kirchoff's law at each node of the network is then generated. These equations can be gathered using matrix notation

$$\mathbf{Av} = \mathbf{0}, \tag{6}$$

where $\mathbf{A}$ is a square matrix that has the size of the number of sites and the number of resistors placed in each node with equations for Ohm's law and Kirchoff's law, $\mathbf{v}$ is the vector containing the potential and current at each node.

Finally, the system is solved iteratively. In this way, the value of the potential can be calculated across the network for a known applied field. This leads to the total conductance of the lattice. In both numerical models, the system is treated as being periodic in terms of fiber placement. If a fiber extends beyond the boundary, it is allowed to reenter at the opposite side. Consequently, the fiber density remains constant for each simulation.

2.3. Electrical Transport

Because the fibers are long compared to their width, a dilute random array of fibers in a matrix could conceivably be arranged such that the fibers provide a direct conduction path between two contacts (either electrical or thermal). When a direct path is established, the network of fibers is said to conduct. As a result, the effective conductivity of the compound may appear more like that of the fiber material despite having low fiber densities. This argument suggests that the fiber laden compound could conduct at significantly lower densities than compounds with circular inclusions. Therefore, we have developed a simple conduction model based on the fiber connectivity. In this model, we assume the matrix does not contribute to the transport. Instead, only when the fibers create a conduction path will the transport be non-zero.

To compute the effective conductivity using the fiber network model, the fibers are placed randomly in the device. Between any two intersections that share a fiber, we create a resistor. A sample device

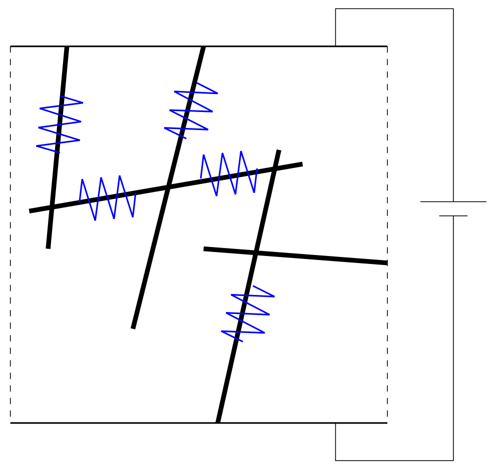

Figure 3: Fiber network (5 fibers) showing resistors in a sample device. The resistance is given by $R = \rho L/A_f$ where L is the length between fiber crossings and A_f is the cross sectional area of the fiber.

with 5 fibers is shown in Figure 3. The electrical resistance (R_e) depends on the material resistivity (ρ), length and width (diameter) of the fiber and reduces to

$$R_e = \frac{2\rho}{\pi a \alpha}, \tag{7}$$

where the resistivity is the reciprocal of conductivity. Ohm's law ($V = IR$) is used to relate the current through the resistor to the potential across the resistor. Furthermore, at each intersection, we balance the currents to produce a linear system with the same number of unknown potentials and currents as equations. The applied voltage divided by the total current through the device gives the effective conductivity of the compound. The resistance network approach is valid when the conductivity ratio is large. This model should approach the percolation model for large fiber density but will predict zero conductivity depending on the network formed by the random placement of fibers. If there is no conductive path, then the conductivity is zero. This model can be used to identify the percolation threshold as well as predict the effective conductivity of the network.

3. Results

Using the properties in Table 1, we have constructed a study on the electrical and thermal transport of a two-terminal planar device that contains a compound material. The compound consists of a matrix with random placed fibers. The number and length of fibers are varied to determine the effects of the fiber-density parameter na^2 on transport, where n is the number of fibers in the device. Note that the area density c for the analytic solutions (equation 1) can be deduced from the density parameter as

$$c = \frac{nA_{ellipse}}{A_{device}} = \frac{n\pi(L_f/2)(w/2)}{L^2} = \frac{n\pi a^2\alpha}{4} \tag{8}$$

where $a = L_f/L$ is the fiber length normalized by the square root of the device area ($L = \sqrt{A}$), and w is the fiber width. In the present case, L is simply the length of a side of the square device. Because the fibers in the computational model are rectangular, the area density c is expressed as

$$c = \frac{nA_{fiber}}{A_{device}} = \frac{nL_f w}{L^2} = na^2\alpha \tag{9}$$

where A_{fiber} is the projected area of the fiber on the device. Note that neither equation 8 or 9 are exact because they do not consider overlap. Therefore, the approximation becomes worse for large numbers of fibers. Furthermore, the accuracy also depends on the aspect ratio $\alpha = w/L_f$; for long thin fibers the overlap area between two intersecting fibers decreases as w^2, but the fiber area decreases as w.

Because fibers are placed randomly, several simulations were performed, results were averaged to obtain a representative conductivity. In the case of a compound whose conductivity ratio is large (fiber conductance is much larger than the matrix), the variation in predicted compound conductance can be orders of magnitude. A simple average will prefer values at the high end of the range and mask the fact that many orientations will result in nearly zero conductance.

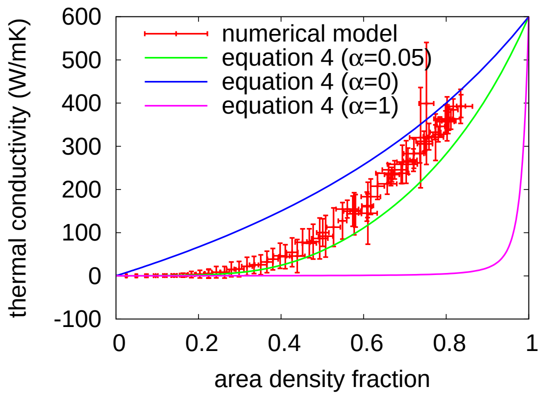

Figure 4: Thermal conductivity for $L_f = 0.5$ ($\alpha = 0.05$). The infinite aspect ratio case ($\alpha = 0$) is an upper bound, and the circular inclusion case ($\alpha = 1$) is a lower bound.

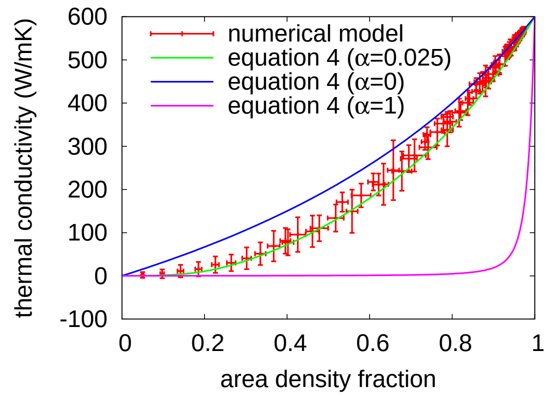

Figure 5: Thermal conductivity for $L_f = 1.0$ ($\alpha = 0.025$). The infinite aspect ratio case ($\alpha = 0$) is an upper bound, and the circular inclusion case ($\alpha = 1$) is a lower bound.

3.1. Thermal results

The effective thermal conductivity as predicted by the discretized model is shown in Figure 4 and 5. The results are shown for $L_f = 1.0$ and 0.5. In all simulations, the normalized width of the fibers is 0.025 or 2.5% of the device length. The horizontal error bars represent the variance in the fiber density which results from the amount of overlap due to the random placement of the fibers and the vertical error bars represent the variance in the calculated thermal conductivity. The blue line represents equation 4, which accounts for the difference in area between fibers, with fiber overlap, and inclusions with ellipsoidal geometry. These results represent a limiting case of negligible contact resistance. As the contact resistance becomes comparable to the resistance of the matrix material, the effect of the fibers will be reduced and the conductivity of the matrix is eventually recovered for all area densities.

Both the numerical and analytic model are in very good agreement for both fiber lengths. However, the analytic model (equation 4) predicts lower conductivity at higher area densities for the shorter fibers. The reason for the discrepancy is unknown but is likely due to the difference between ellipse and rectangular geometry of the analytic fibers versus the numerical fibers. As the fibers become shorter this discrepancy has a bigger impact. Nevertheless, these results correspond well with observed low percolation thresholds observed in CNT/epoxy compounds [8].

As expected the conduction for the shorter fibers is lower than that of the longer fibers for the same area density. In other words, we can have twice as many short fibers, but the conductivity will still be lower. This is because the possibility of overlap between the short fibers is reduced until very high loadings are achieved. This effect will be important to device designers.

3.2. Electrical results

The calculated electrical conductivity is shown in Figure 6 and 7 for the $L_f = 0.5$ and the $L_f = 1.0$ size fibers respectively. As suggested by Zimmerman [14], the large conductivity ratio makes the solution much more sensitive to the aspect ratio compared to the thermal solution. Consequently, the approximation of the geometry of the fiber in the analytic approach (ellipse) may reduce the accuracy of the analytic solution compared to the numerical solution.

For the electrical transport studies, percolation is determined by the number of fibers needed to create a conduction path across the device. Because the electrical transport depends on creating a conduction path across the device, simulations for the lowest loadings can be unreliable. At small loadings,

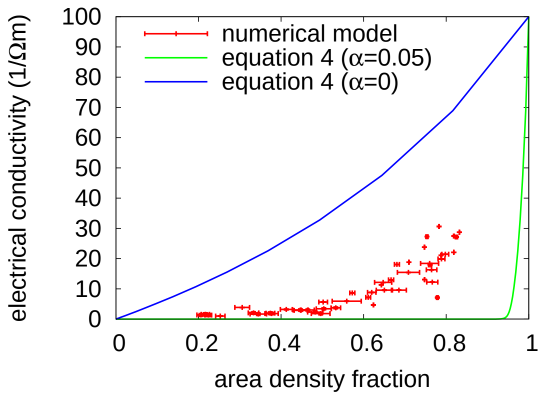

Figure 6: electrical conductivity for $L_f = 0.5$ ($\alpha = 0.05$). The infinite aspect ratio case ($\alpha = 0$) is an upper bound.

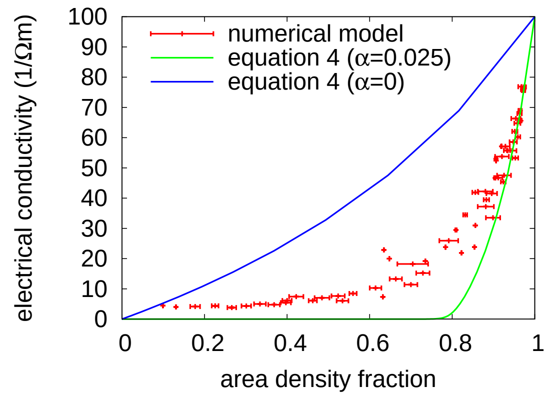

Figure 7: Electrical conductivity for $L_f = 1.0$ ($\alpha = 0.025$). The infinite aspect ratio case ($\alpha = 0$) is an upper bound.

a conduction path may not be formed since the fibers are laid randomly. In such cases, the conductivity is exactly zero. Therefore, only loadings larger than about 10% projected area density were able to create a conduction path and exhibit non-negligible conductivities. (This equates to about 4 fibers of length $L_f = 0.5$ or two fibers of length $L_f = 1.0$. Even here, the probability of not forming a conduction path is non-negligible.) This agrees well with Kymakis et al. [17] who found that a 2D mixture of CNTs and P3OT percolates at 11% weight fraction, which suggests that in compounds where the two materials have dramatically different conductivities, a conduction path is required to achieve percolation. This is in contrast to the thermal results where transport is achieved even at very low loadings. These calculated results also agree well with experimental results from Thonruang et al. [18], who showed conductivities for 0.25 mm fibers (corresponding to our $L_f = 1.0$ case) of 2 $(\Omega\text{m})^{-1}$ at 20% loading. Our results over-predict their measurements by 20-30%, which may be due to the difference in the conductivity of the two matrix materials, the contact resistance that is not considered in our model, or uncertainty in the measurements. Nevertheless, these experimental data validate at least a portion of our analysis.

According to the numerical results, the composite did not percolate until $c \approx 0.18$ for $L_f = 0.5$ (Figure 6). This is further supported by [18], which suggests that high aspect ratio fibers will percolate at lower concentrations. In general, the numerical models agree well with equation 4 but only in trend. As the conductivity ratio becomes larger, the percolation threshold increases. But because we can create a conduction path across the device, we see a low percolation threshold that can not be predicted by the analytic model. Nevertheless, we would have expected better agreement between the numerical and analytic model at higher area densities. Interestingly, we see better agreement between the models for longer fibers. Yet, there is nothing in the derivation that would suggest that the analytic model is deficient for shorter fibers (See Zimmerman [14]).

The results from the equivalent resistance numerical model should be questioned because of inherent issues with the solution approach. Fibers with shorter length and systems with low fiber density are often problematic due to the fact low density systems often yield non-invertible singular matrices. Consequently, solutions are difficult to obtain for dilute systems. The discretized model used for the thermal transport study could not be used to predict electrical transport because the fiber/matrix conductivity ratio is too large and stiff matrices resulted from the formulation.

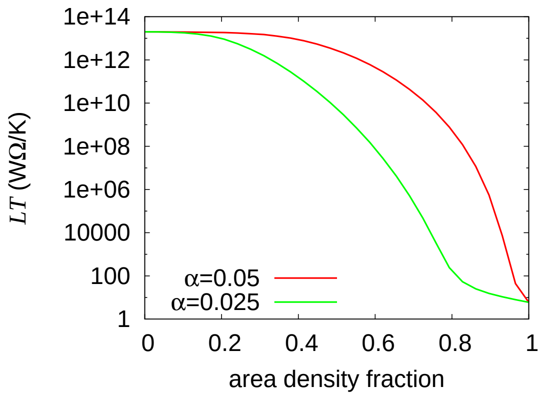

Figure 8: Lorenz number times temperature as a function of fraction of area covered by fibers.

3.3. *Combined transport*

The Lorenz number (L) is the ratio of thermal to electrical conductivity and a plot of $LT = k/\sigma$ versus area density is shown in Figure 8. Since LT is not constant, the models presented in this work show that thermal and electrical properties can be predicted and decoupled for the design of electronic devices. In the design of thermoelectric materials for example, a designer wants low thermal conductivity and high electrical conductivity (smaller Lorenz number). One then would select a high loading of longer (high aspect ratio) fibers.

4. Conclusion

Effective thermal and electrical transport through randomly-oriented conducting fibers in a non-conducting matrix was predicted using analytical and numerical methods. Analytical solutions were obtained by calculating the effective conductivity of a matrix with small ellipsoidal inclusions using an effective medium approximation. This model works well for dilute systems and for systems with low conductivity ratios. However, the analytic model does not account for fiber-fiber overlap exactly and therefore under-predicts the conductivity for systems with high fiber density. Numerical methods must be used in order to determine conductivity for non-dilute systems and systems with large conductivity ratio.

Electrical transport was predicted by assuming that the matrix does not contribute to transport and that the system will conduct when a direct path of fibers is established between contacts. The conductivity is calculated by placing a resistor between two fiber-fiber intersections and using a system of linear equation which obey Ohm's law to predict the resistance of the system. Although this approach is grounded in engineering principles, the results leave some doubt whether the approximation is valid.

On the other hand, the discretized model used to calculate thermal transport agrees very well with the analytic model suggesting that the discretization approach would be suitable for multi-material systems and complex geometries.

None of the models presented here consider the contact resistance between fibers, which can affect or even dominate the resistance in percolation networks. However, the analysis uses "effective" material properties, which could include a contact component. For dense networks where the contact dominates, the resulting conductance may be lower than predicted here. In addition, we have not considered three dimensional percolation networks [19]. Presumably, neither effect will change the conclusion concerning the relative change of thermal and electrical conductances in films. The numerical models, however, can be extended to three dimensions; in general, the analytic solution can not.

In metals, the Lorenz number is a constant. In non-metals it can vary significantly. If the thermal and electrical conductivities can be controlled separately, the effects are decoupled. Usually, though, the addition of a filler in a composite that improves one conductivity, will also improve the other conductivity. We are investigating how these two conductivities (electrical and thermal) change relative to each other with the addition of fibers to a polymer. The results indicate that certain loadings and fiber lengths can be used to tune the relative properties, which can be important, for example, for the design of thermoelectric devices, which require low thermal conductivity and high electrical conductivity. In addition, these findings could be used to predict an optimum thermal and electrical transport without sacrificing the flexibility of the TFT.

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