Modeling Tensorial Conductivity of Particle Suspension Networks

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Significant microstructural anisotropy is known to develop during shearing flow of attractive particle suspensions. These suspensions, and their capacity to form conductive networks, play a key role in flow-battery technology, among other applications. Herein, we present and test an analytical model for the tensorial conductivity of attractive particle suspensions. The model utilizes the mean fabric of the network to characterize the structure, and the relationship to the conductivity is inspired by a lattice argument. We test the accuracy of our model against a large number of computer-generated suspension networks, based on multiple in-house generation protocols, giving rise to particle networks that emulate the physical system. The model is shown to adequately capture the tensorial conductivity, both in terms of its invariants and its mean directionality.

Introduction

The electrical conductivity of heterogeneous materials has been extensively studied by many different researchers over the years\textsuperscript{3,4,12,19,20}. The literature primarily focuses on heterogeneous materials which are mixtures of two materials that each have different, isotropic electrical conductivities. The most well-known result is that of Maxwell, which is based on an effective-medium approximation for dilute suspensions\textsuperscript{14}. Hashin and Shtrikman approached the problem in a different way. Rather than attempt to solve for an exact expression for the effective conductivity of a randomly structured material, they applied a variational method to derive upper and lower bounds on the effective conductivity\textsuperscript{9}. They chose to use a variational approach to derive bounds on the conductivity because solving the exact problem for an arbitrarily structured heterogeneous material was analytically intractable. Torquato\textsuperscript{19–21} has studied the effective conductivity problem in great depth. He has improved the bounds laid out by Hashin and Shtrikman, has solved for effective conductivity of a number of different lattice types, and has expressed the exact tensorial effective conductivity in terms of an infinite series of N-point probability functions, which can be used to describe the microstructure of a heterogeneous material. The particular case of a suspension consisting of a conductive particle network within an insulating medium has been considered theoretically, to our knowledge, in one existing study\textsuperscript{12}. The approach they take assumes a spatially homogeneous potential gradient field imposed upon the structure, leading to a model for the conductivity that can be proven to be an upper bound.

Much of the aforementioned work is concerned with the isotropic conductivity of heterogeneous materials. In this work, we aim to model the full tensorial conductivity, with a focus on suspended networks of conductive particles. These particle networks are of practical importance, especially in flowable battery technology currently under development by the Joint Center for Energy Storage Research (JCESR)\textsuperscript{5}. In these batteries, a conductive, flowing suspension of carbon black forms an integral component of the system, see Figure 1(a). It has been shown in related systems\textsuperscript{13} that shear flows induce anisotropy in a contact network of suspended particles, as pictured in Figure 1(b). In instances where suspension conductivity arises from particle-particle contacts, this structure anisotropy should give rise to conductivity anisotropy. It is this behavior that we seek to describe. It has been shown experimentally that the electrical conductivity of a suspension is highly sensitive to shear rate\textsuperscript{1}, dropping by several orders of magnitude as shear rate increases. From this observation and the evidence of particle microstructure changing in shearing flow, we deduce that a suitably chosen description of the particle network should be sufficient to predict the electrical conductivity of a suspension.

In the granular media literature, a great deal of attention has been given to describing the structure of the contact network between particles. Perhaps the simplest structural measure for such a network that includes anisotropy is the fabric tensor\textsuperscript{4,5,6,18}. While more complex structural measures exist, such as pair- and higher-order particle correlation functions\textsuperscript{19}, whose use could enable greater accuracy in constructing a conductivity model, we shall show that a suitable model can be achieved solely in terms of the fabric. Key to our model development is the solution of a simple case, based on a network conforming to a lattice structure. The results instruct the form for a new conductivity model, whose accuracy is then tested.
against many thousands of random particle networks. To explore a range of particle networks, we describe two distinct algorithms for creating random packings — one for denser packings, and one for more dilute packings that closely resemble those formed by carbon-black — and demonstrate the model’s predictive capability against thousands of packings generated from both algorithms.

**Homogenization**

The tensorial form of Ohm’s law relates the electric field vector $\mathbf{E}$ to the current density vector $\mathbf{J}$ through a second-order conductivity tensor $\mathbf{K}$, i.e.

$$\mathbf{J} = \mathbf{K} \mathbf{E} \quad (1)$$

The conductivity tensor is a symmetric, positive-definite tensor. An effective conductivity for a representative volume $\Omega$ of a heterogeneous material must be defined prior to any analytical or numerical work. The effective conductivity of an ergodic medium is defined by

$$\langle \mathbf{J} \rangle = \mathbf{K} \langle \mathbf{E} \rangle \quad (2)$$

where $\langle \mathbf{E} \rangle$ and $\langle \mathbf{J} \rangle$ are, respectively, the spatially-averaged electric and current density fields over $\Omega$. To avoid a possibly over-reaching assumption of ergodicity — our tests will be conducted on finite domains — we specify that $\langle \mathbf{E} \rangle$ is imposed by prescribing a linear boundary potential $\varphi(\mathbf{x} \in \partial \Omega) = -\langle \mathbf{E} \rangle \cdot \mathbf{x}$, and that $\langle \mathbf{J} \rangle$ is redefined as the flux that is power-conjugate to $\langle \mathbf{E} \rangle$. That is,

$$\langle \mathbf{E} \rangle \cdot \langle \mathbf{J} \rangle = \frac{1}{V} \int_{\Omega} -\nabla \varphi \cdot \mathbf{j} \, dV \quad (3)$$

where $\mathbf{j}$ is the local current density field. In the ergodic limit of the ensuing analysis, $\langle \mathbf{J} \rangle$ reduces to a standard spatial average.

Assuming that the current density obeys Kirchoff’s current law and Ohm’s law — respectively, $\nabla \cdot \mathbf{j} = 0$ and $\mathbf{j} = -\sigma \nabla \varphi$ for some non-negative conductivity field $\sigma(\mathbf{x})$ — a symmetric, positive-definite conductivity tensor $\mathbf{K}$ must exist that obeys (2). By using a calculus identity, Eq (3) can be transformed into

$$\langle \mathbf{E} \rangle \cdot \mathbf{K} \langle \mathbf{E} \rangle = \frac{1}{V} \int_{\partial \Omega} -\varphi \mathbf{j} \cdot \mathbf{n} \, dA \quad (4)$$

where $\mathbf{n}$ is the outward-pointing normal vector.

We model the particles as perfect conductors, the fluid as a perfect insulator, and we suppose electrical resistance arises only at the contacts between particles. Likewise, the field $\varphi$ is approximated as a constant within each particle but possibly varying from particle to particle. The above integral can now be broken into a sum of integrals over the boundary. In the locations where the boundary passes through free space (i.e., not a particle), then we know that $\mathbf{j}$ is exactly $\mathbf{0}$. This leaves only the parts of the boundary that pass through particles, which allows us to write the integral over the set of boundary particles $B$, i.e.

$$\langle \mathbf{E} \rangle \cdot \mathbf{K} \langle \mathbf{E} \rangle = \frac{1}{V} \sum_{i \in B} -\varphi_i \int_{\partial \Omega_i} \mathbf{j} \cdot \mathbf{n} \, dA. \quad (5)$$

where $\Omega_i$ is the intersection of the $i$th boundary particle with $\partial \Omega$, and the potential within particle $i$, denoted $\varphi_i$ above, can be brought outside the integral since it is constant within a particle. Although the precise nature of $\mathbf{j}$ is unknown within the particle, the value of the integral $\int_{\partial \Omega_i} \mathbf{j} \cdot \mathbf{n} \, dA$ is the current that is flowing out of $\Omega_i$. Denoting this current as $I_i^{\text{out}}$ we can write the final expression for the right-hand-side of (4),

$$\langle \mathbf{E} \rangle \cdot \mathbf{K} \langle \mathbf{E} \rangle = \frac{1}{V} \sum_{i \in B} -\varphi_i I_i^{\text{out}}. \quad (6)$$
can be defined for particle \( p \) by the relation 

\[ A^p = \sum_i n_i \otimes n_i \]  

(9)

where \( \otimes \) denotes the dyadic product, and \( n_i \) is the unit normal vector connecting particle centroids of the \( i \)'th contact on the particle. This is illustrated in Figure 3. To homogenize over the entire particle network, or at least meso-sized region of it, the average fabric tensor is defined as the system average of the particle fabric tensors.

\[ A = \frac{1}{N_{\text{particles}}} \sum_{p=1}^{N_{\text{particles}}} A^p \]  

(10)

The definition of the fabric tensor has some attractive features. It is symmetric and positive-semidefinite, guaranteeing that the eigenvalues are non-negative and that the eigenvectors are orthogonal. These properties are shared by the conductivity tensor \( \mathbf{K} \), suggesting the fabric tensor could be an appropriate independent variable in the conductivity’s functional form.

**Lattice-Reduced Model**

We propose an analytical model to elucidate the connection between electrical conductivity and the fabric tensor based on a simplified lattice structure. We will test this model’s applicability to random packings in the later sections.

The particles are imagined to live on an idealized infinite, periodic lattice. The lattice is parameterized by a set of numbers that describe the particle size and spacing. These parameters are (1) particle diameter \( D_p \), (2) distance in x-direction between chains \( d_x \), (3) distance in y-direction between chains \( d_y \), (4) distance in z-direction between chains \( d_z \). In 2D, only the first three parameters are used. An illustration of a 2D lattice characterized by these parameters is shown in figure 4(a), with its fundamental unit cell shown in figure 4(b).

Both the average fabric tensor and effective conductivity can be computed analytically. The average fabric tensor is defined as the spatial average of the fabric tensor for all of the particles in the unit cell and ultimately results in the formula

\[ A = \frac{2}{N_x + N_y - 1} \begin{bmatrix} N_x & 0 \\ 0 & N_y \end{bmatrix} \]  

(11)

In this expression, the key quantities to recognize are the number of particles in the x-oriented chain, \( N_x = d_x/D_p \), and the number of particles in the y-oriented chain, \( N_y = d_y/D_p \).

Next, the effective conductivity was derived for the unit cell. To do this, imagine applying an arbitrary voltage difference
across the x-oriented and y-oriented chains separately. These voltages are \( \Delta \phi_x \) and \( \Delta \phi_y \), respectively. By applying Ohm’s law through the corresponding chains, we can recover the components of the vector form of Ohm’s Law shown in (12). For example, for the x-oriented chain
\[
j_x = \left( \frac{1}{N_y R_c} \right) (\nabla \phi)_x
\]
with \( (\nabla \phi)_x = \Delta \phi_x / d_x \). Due to the geometry of the problem, we know that the off-diagonal components of the conductivity tensor \( K \) are exactly zero. Therefore, we can say
\[
K_{11} = \frac{1}{N_y R_c}.
\]
Similarly analysis yields
\[
K_{22} = \frac{1}{N_x R_c}.
\]
Finally, the parameters \( N_x \) and \( N_y \) can be algebraically eliminated to give the components of \( K \) in terms of the components of \( A \), yielding the tensorial relationship
\[
K = \frac{1}{R_c} \frac{\text{tr} A - 2}{\det A} A.
\]
We refer to the formula in (15) as the “lattice model”. A similar analysis can be carried out for a three-dimensional unit cell, which will yield the following expression for the conductivity tensor,
\[
K = \frac{1}{4D_p R_c} \frac{(\text{tr} A - 2)^2}{\det A} A.
\]
The formulae above apply when \( \text{tr} A - 2 \) is non-negative. Otherwise the solution is \( K = 0 \).

Despite its inspiration from the lattice structure, there are several reasons to consider the applicability of the lattice model to more general particle networks. For one, the formula purports codirectionality of the fabric and conductivity, i.e. the deviators of the two tensors are aligned, implying that the direction of anisotropy of one tensor gives the anisotropy direction of the other, which to a first approximation ought to match the behavior of general particle networks. Second, the results imply that conductivity should vanish when \( \text{tr} A < 2 \), which is sensible more generally (though not strictly) because particles in a percolating chain, as needed to conduct current across the sample, must have coordination number at least two. Above this threshold, conductivity increases with \( \text{tr} A \) in line with one’s basic intuition for more highly coordinated networks.

We are aware of one other fabric-based analytical model for conductive particle networks, which was developed by Jagota and Hui. In their work, a uniformity hypothesis is made with regard to the potential gradient, which results in a conductivity model that is fully linear in the fabric tensor,
\[
K = \frac{N_V D_p^2}{4R_c} A.
\]
The above, which can be proven to be an upper-bound on the real conductivity, is for a two-dimensional system and \( N_V \) is the particle number fraction (per area in 2D). This model differs from ours most notably in that the conductivity is not thresholded by the coordination number, the formula depends explicitly on the particle area fraction as well fabric, and it does not depend on the fabric determinant.

**Numerical Simulation**

In order to perform numerical experiments and determine the generality of the lattice model, a large number of random particle networks (packings) must be created. There are a number of methods to do this already in the granular and particulate matter literature. See the references for a broad summary of the currently available granular packing algorithms. Attractive suspensions have been modeled with the Diffusion-Limited Aggregation (DLA) model of Witten and Sander. A common feature of many of the granular statics methods is that they solve force equilibrium equations for a system of particles. This was not a feature that was required for this study,
so these types of methods were not used, in the interest of saving computational time. Instead, we developed two methods for creating two-dimensional random contact networks of particles, and we tested our model against numerous packings generated by each method. Both methods allow us to influence the resulting anisotropic structure of the packings.

Algorithm 1: Our first packing algorithm was designed to create a dense random contact networks of particles. This is in contrast to a later algorithm, to be described below, which created packings that resulted in much lower-density packings. The dense packings were created by perturbing a 2D hexagonal close-packing of particles. This was achieved by placing points into a triangular lattice, adding random noise to the position of each point, and finally growing each particle as large as possible such that no particles overlapped. Anisotropy can be influenced by shearing the points with an affine transformation $x' = Fx$ before growing the radii. This process is described in pseudocode below (Algorithm 1). An example of the resulting packing overlaid by its analogous resistor network is shown in figure 5.

Algorithm 1

Seed $L \times L$ box with close-packed points
Perturb points with random noise
Move each point to new location $x'$ by $x' = Fx$
while Not all radii frozen do
  Find smallest distance that any particle can grow
  Grow all particles by this amount
  Freeze radii of particles that come into contact
end while

Algorithm 2: This procedure was motivated by a need to better understand the conductivity of carbon black suspensions in an insulating medium. The self-attraction carbon black particles leads to fractal particle networks that are electrically percolating at low volume fraction (below 1 vol%)\(^5\).

To produce structures that more closely resemble carbon black suspensions, we developed our second packing algorithm, which is inspired by the “hit-and-stick” behavior of the carbon particles. In addition, the new algorithm is able to include the effects of particle Brownian motion but this is not essential to the algorithm.

First, clusters (single particles at this stage) are seeded randomly into a $L^d$ box, where $d$ is the number of spatial dimensions. Next, a linear velocity field is imposed directly on each cluster’s centroid according to

$$v = -B(x - O)$$  \hspace{1cm} (18)$$

where $O$ is a point in the middle of the original box. This imposed velocity field serves to pull all of the clusters together. The matrix $B$ is a $d \times d$ matrix that allows us to impose an anisotropic velocity field. This allows us to influence (but not completely impose) the fabric tensor that results from this packing method. After the velocity field is imposed, the particle positions are updated by assuming a time step $dt$ (computed at runtime). Then, the clusters are checked to determine whether any contacts have been made with other clusters. If so, the clusters are cohered into a single cluster for all future steps. This process of imposing velocity, updating positions, and handling contacts is repeated until only a single cluster remains. The process is outlined in pseudocode in Algorithm 2.

An example of a packing resulting from this process is shown in figure 6 and a larger example is displayed in figure 7.

The box-counting fractal dimension\(^6\) of the resulting packings was computed in order to determine if they resembled real-life packings found in experiments. The fractal dimension of packings produced by this method is approximately $d = 1.75$. This was compared against the particle network image in figure 1. This network has a fractal dimension of approximately $d = 1.7 \pm 0.1$. Uncertainty in the measurement is due to the image processing techniques used to identify particles. Based on these measurements, we are satisfied that this algorithm produces realistic packings, although more detailed correlation function measurements would be needed for a firmer conclusion.

Applying boundary conditions: In order to apply the solution method described above to an arbitrary packing of particles, appropriate boundary conditions must be applied. In these simulations, a prescribed voltage was applied to particles all around the boundary. This process consists of two steps: first, the boundary must be identified, and second, the linear system must be updated to reflect the known voltages.
Algorithm 2

Seed N clusters (particles) in \( L^d \) square

while \( N_{\text{Clusters}} > 1 \) do

Move clusters according to \( \mathbf{v} = -B(\mathbf{x} - \mathbf{O}) \)
Locate collisions between clusters
Combine clusters in contact and recompute centroids

end while

Fig. 6 Example (using a small number of particles) of a packing resulting from Algorithm 2 using a small number of particles.

For the first packing algorithm, identifying the boundary is a trivial process, since the particle locations are known a priori. For algorithm 2, however, the particle positions are not known. A boundary can be located visually quite easily at the end of the simulation process, but performing this step manually would be prohibitively slow. In order to expedite and automate the simulation process, the following method was devised to locate the boundary.

First, histograms of the particle \( x \) and \( y \) positions were separately created. To find the “left” and “right” boundaries, denoted \( x^- \) and \( x^+ \) respectively, the histogram of \( x \) positions was thresholded. The value \( x^- \) is defined as the smallest \( x \) value where the histogram reaches 50% of its maximum value. The value \( x^+ \) is defined as the largest \( x \) value that meets the same criterion. The top and bottom boundaries, \( y^+ \) and \( y^- \), are found in the same manner using the histogram of particle \( y \) coordinates. The threshold value 50% was determined empirically to locate the same boundary that one would identify visually. An example packing and its associated \( x \)-position histogram is shown below in figure 7 to demonstrate the efficacy of the method.

The expression in (6) can be computed easily from the solution of the particle network, so by judiciously choosing \( \langle \mathbf{E} \rangle \), the components of \( \mathbf{K}_e \) can be extracted. In two dimensions, the effective conductivity tensor has three independent components, so three simulations are sufficient to extract all of the components. The \( K_{11} \) component can be extracted by setting \( \langle \mathbf{E} \rangle = e_x \). This corresponds to evaluating the integral for an applied boundary voltage of \( \varphi = -x \). The remaining tensor components may be similarly extracted by applying specific potential fields at the boundary and evaluating the summation given in (6).

Tests

The previously described packing algorithms and solution procedures for the current/potential have been implemented in Matlab. Algorithm 1 was used to create 50,000 separate packings.
400-particle packings. In all of these packings, the $F_{11}$ and $F_{22}$ components of the affine transformation $F$ equaled 1.0. The $F_{12}$ component that controlled the shearing of the packing ranged between 0 and 0.5 in increments of 0.01. Any particles that were sheared out of the original bounding rectangle were reflected to the other side of the box to return the packing to a rectangular geometry. Algorithm 2 was used to create 10,000 separate 5,000-particle packings. In the $B$ matrix, the $B_{11}$ component remained 1.0, and the $B_{22}$ component was varied in $[1.0, 1.9]$ in increments of 0.1 to influence the level of anisotropy of the resulting packings. In all simulations, the contact resistance $R_c$ was assigned to be 1, so it did not have any affect on the following analyses. After applying the previously described procedure to each packing to obtain the effective conductivity tensor and average fabric tensor for each packing, the data were analyzed to determine the extent that the results agree with the model’s prediction for the isotropic behavior. These tests are described next, and thereafter we shall proceed to show how well the lattice model performs compared to the existing model, equation (17).

The isotropic behavior of the conductivity can be investigated by taking the trace of both sides of (15). The average coordination number is the most natural independent variable when examining the isotropic behavior, so in addition to taking the trace of both sides of (15), both sides were multiplied by $\det A$ in order to make the right-hand side a single-valued function of $\text{tr}A$. This results in (19).

$$R_c \text{tr}K \det A = (\text{tr}A - 2) \text{tr}A$$

where a subscript 0 denotes the deviator of the tensor, and the term $\frac{A_0}{|A_0|}$ is commonly referred to as the direction or sign of the tensor $A_0$. The left hand side of this was plotted against $|A_0|$ to test the predictive power of the model. It can be seen in figure 9 that, although there is a large amount of noise in the measurements, the model captures the mean behavior very closely.

Next, we determine the extent the analytical lattice model predicts the anisotropy of the conductivity. To remove the influence of the isotropic behavior, we take the deviator of both sides of (15). In this case, the most natural independent variable is the magnitude of the fabric deviator, so the resulting equation was manipulated to be a single-valued function of this quantity. After manipulation, (15) can be written as (20).

$$R_c K_0 : \frac{A_0}{|A_0|} \left( \frac{\det A}{\text{tr}A - 2} \right) = |A_0|$$

where $\text{tr}$ is the trace of the matrix and $\det$ is the determinant. The right hand side of this was plotted against $|A_0|$ to test the predictive power of the model. It can be seen in figure 10 that, although there is a large amount of noise in the measurements, the model captures the mean behavior very closely.

The final prediction that must be examined is the notion of codirectionality. The analytical model in (15) predicts that the fabric and conductivity tensors have the same eigenvectors. To examine this, the angle difference between the fabric and conductivity deviators was calculated, which is equivalent to the (signed) angle between the eigenvectors corresponding to the largest eigenvalues of the two tensors, denoted $e_K$ and $e_A$. The deviators were chosen because, in 2D, the eigenvector corresponding to the positive eigenvalue can be unambiguously chosen. The probability density function of the angle difference as a function of $\Delta \theta$ is plotted in figure 10. It can be seen that this distribution is symmetrically centered around zero, indicating that the fabric and conductivity are strongly codirectional.

![Fig. 8 Predicted relationship between the (modified) trace of the conductivity and the fabric trace, compared to numerical results of 50,000 packings generated by Algorithm 1 and 10,000 generated under Algorithm 2. Inset is a zoom-in of the vicinity of $\text{tr}A = 2$.](image-url)
Finally, we also compared the lattice model [15], to the existing model by Jagota & Hui [18] shown in [17]. For the same 60,000 packings generated using both packing algorithms, we computed the relative error of the prediction of the trace and the determinant of the conductivity using each model and plotted the results in figures 11 and 12. In every case, we found that the new lattice model predictions were closer to the true values from the numerical experiments than the previous model by Jagota & Hui. On the other hand, the Jagota & Hui model maintains a strong upper bound on both invariants of the conductivity tensor, whereas the lattice model is not strictly an upper bound, as previously discussed.

**Discussion and Conclusions**

In this paper we have derived and tested a new model relating the structure of a packing of particles to its tensorial electrical conductivity. The assumptions implicit in the model are that the suspending medium is a perfect insulator and that electrical resistance arises only at particle contacts. The structural measurement used was the fabric tensor, and the model arises from a straightforward analysis of a representative problem involving a lattice structure. The resulting model takes a non-linear functional form, and was tested multiple ways against numerical simulations of many thousands of random particle packings. The agreement in its predictions of the various scalar properties and tensorial orientation is significant, especially in light of the simplistic nature of the fabric tensor being the sole independent variable for the model. In our tests, the lattice model’s accuracy was shown to be higher than an existing conductivity model, a model which requires more structural input data than the lattice model. While it is definitely possible to write a more accurate model by including dependences on more structural variables — some of our data spread is due to the finite nature of the datasets, but some is surely due to modeling error — the current simplicity of the lattice model is an advantage for its usage in engineering applications involving flowing suspension networks. Modeling frameworks for the evolution of anisotropy tensors in flowing media have
been developed over the last decades\textsuperscript{7,8,17}, keeping our model in terms of fabric, then, suggests a path to the simulation of simultaneous flow and current transfer fields in nontrivial systems by coupling a fabric evolution rule and a rheology with our conductivity model. Such a capability would be key in the targeted application of modeling flow battery systems, which rely on a flowing conductive suspension that closely resembles the idealized system that we considered.

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