**INFLUENCE OF INTERFACIAL SURFACE CONDUCTION
ON THE DC ELECTRICAL CONDUCTIVITY OF
PARTICLE REINFORCED COMPOSITES**

By

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Influence of interfacial surface conduction on the DC electrical conductivity of particle reinforced composites

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Abstract

We describe new effects associated with electrical conduction along phase interfaces for particle reinforced conductors. For particles of general shape we introduce a new quantity β_1 called the "surface to volume dissipation" of a particle. This quantity is a measure of the particle's ability to dissipate energy on its surface relative to the energy dissipated in its interior. It is described mathematically as the minimum value of a suitably defined Rayleigh quotient and is related to an eigenvalue problem posed on the particle surface. We consider the overall conductivity of a particle reinforced conductor when the particle conductivities are less than that of the matrix. It is shown that the overall conductivity will be increased by the presence of a specific particle when the particle's "surface to volume dissipation" lies above a critical value. We calculate the surface to volume dissipation for a sphere and for starlike particles we provide a lower bound in terms of particle dimensions. These estimates allow for the prediction of new particle size effects. Secondly we present a new criterion on the particle size distribution for which the overall conductivity lies below the matrix conductivity.

1 Introduction

We consider particle reinforced composite materials. The case when electrical conduction occurs along phase interfaces as well as inside the particle and matrix phases is addressed. The interfacial surface conductivity has dimensions of "conductivity $\times$ length" and is denoted by α . The conductivity of the particle phase is given by $\mathbf{c}_p$ and that of the matrix by $\mathbf{c}_m$. Both matrix and particles are assumed to have anisotropic conducting properties and $\mathbf{c}_p$ and $\mathbf{c}_m$ are given by 3×3 symmetric, positive definite matrices. We suppose that the conductivity of the particle is less than the matrix, ie., $\mathbf{c}_p < \mathbf{c}_m$. This inequality holds in the sense of quadratic forms. The results reported here describe new effects related to electrical conduction along phase interfaces. For a particle of general shape we identify a quantity called the "surface to volume dissipation". It is shown that the overall conductivity is increased by the presence of the particle when the particle's surface to volume dissipation lies above a critical value. The second result presented here is a new criterion on the suspension geometry for which the overall conductivity lies below the conductivity of the matrix material.

The composite domain is denoted by Ω and its volume is given by $|\Omega|$. The conductivity inside the composite is described by $\mathbf{c}(\mathbf{x})$ taking the values $\mathbf{c}_p$ in the particles and $\mathbf{c}_m$ in the matrix. The electric potential in the composite is denoted by φ . On the boundary of Ω we suppose that the potential is given by the linear function $\mathbf{E} \cdot \mathbf{x}$, where $\mathbf{E}$ is a constant vector. Inside the particle and matrix phases the current is denoted by $\mathbf{j}$. The current is related to the potential by the constitutive law $\mathbf{j} = \mathbf{c}(\mathbf{x})\nabla\varphi$ and in each phase we have:

$$\operatorname{div}(\mathbf{j}) = 0, \quad (1.1)$$

The potential is continuous across phase interfaces and on the matrix-particle interface we have:

$$\mathbf{j}_m \cdot \mathbf{n} - \mathbf{j}_p \cdot \mathbf{n} = -\alpha \Delta_s \varphi. \quad (1.2)$$

Here Δ_s denotes the surface Laplacian on the interface, the subscripts indicate on which side of the interface the normal component of the current is evaluated, and $\mathbf{n}$ is the unit normal pointing into the matrix phase.

We observe from (1.2) that the jump in the normal component of the current produces an interfacial surface charge density that is coupled to the electric potential through a Poisson equation on the interface. The interface jump condition given by (1.2) models, to first approximation, the effects of a thin highly conducting interphase layer between the matrix and particle phases. Indeed one can show rigorously that (1.2) is obtained in the distinguished limit of vanishing layer thickness and increasing interphase conductivity: see, Ref.1.

The effective conductivity $\mathbf{c}_e$ is given by:

$$\mathbf{c}_e \mathbf{E} = |\Omega|^{-1} \int_{\partial\Omega} c(\mathbf{x}) \nabla \varphi \cdot \mathbf{n} \mathbf{x} ds. \quad (1.3)$$

Here ds is the element of surface area, and the vector $\mathbf{n}$ is the exterior unit normal.

We denote the region occupied by the particle phase by A_p and the region occupied by the matrix by A_m . The interface between the particle and matrix phase is assumed sufficiently regular in order for the jump condition (1.2) to hold and is denoted by Γ . The energy dissipated inside the composite is given by the variational principle:

$$\mathbf{c}_e \mathbf{E} \cdot \mathbf{E} = \min_{\phi \in V} \{C(A_p, \phi)\} \quad (1.4)$$

with

$$C(A_p, \phi) = |\Omega|^{-1} \left\{ \int_{\Omega} \mathbf{c}(\mathbf{x}) \nabla \phi \cdot \nabla \phi dx + \alpha \int_{\Gamma} |\nabla_s \phi|^2 ds \right\}, \quad (1.5)$$

and

$$V = \left\{ \phi : \int_{\Omega} |\nabla \phi|^2 + |\phi|^2 dx < \infty, \phi = \mathbf{E} \cdot \mathbf{x} \text{ on } \partial\Omega \right\}. \quad (1.6)$$

Here ∇_s is the gradient operator on the interfacial surface and the minimizer of (1.4) is precisely the electric potential φ .

We provide the geometric criterion that determines the length scale at which interfacial conduction compensates for the lower conductivity of the particle. This criterion is general and applies to any particle shape. In order to give the criterion, we introduce the matrix $\mathbf{P}_{cr}$ given by:

$$\mathbf{P}_{cr} = \alpha(\mathbf{c}_m - \mathbf{c}_p)^{-1} \quad (1.7)$$

Here each element of $\mathbf{P}_{cr}$ has dimensions of length. This quantity provides a measure of the relative magnitude of the interfacial conductance with respect to the mismatch between the conductivity tensors of the matrix and reinforcent. For a given particle domain denoted by " Σ ", we introduce a geometric quantity called the "surface to volume dissipation" of the particle. The "surface to volume dissipation" is denoted by " $\beta_1(\mathbf{c}_p)$ " and has dimensions of $(\text{conductivity} \times \text{length})^{-1}$. It is given by the following quotient:

$$\beta_1(\mathbf{c}_p) = \min_{\phi \in \mathcal{U}} \frac{\int_{\partial\Sigma} |\nabla_s \phi|^2 ds}{\int_{\Sigma} \mathbf{c}_p \nabla \phi \cdot \nabla \phi dx}, \quad (1.8)$$

Here the class of admissable functions is given by, $\mathcal{U} = \{\phi | \text{div}(\mathbf{c}_p \nabla \phi) = 0, \int_{\partial\Sigma} \phi ds = 0, \text{ and } \phi \neq 0.\}$ The minimizer of (1.8) is the "surface to volume dissipation potential" denoted by φ_{β_1} . Taking the first variation of the quotient (1.8) shows that β_1 is the first eigenvalue of the eigenvalue problem on the particle surface given by:

$$-\Delta_s \varphi_{\beta} = \beta \mathbf{n} \cdot \mathbf{c}_p \nabla \varphi_{\beta} \text{ on } \partial\Sigma \quad (1.9)$$

where $\text{div}(\mathbf{c}_p \nabla \varphi_{\beta}) = 0$ inside the particle. Since $\text{div}(\mathbf{c}_p \nabla \varphi_{\beta_1}) = 0$ inside the particle, we see that equation (1.9) shows that the surface Laplacian of φ_{β_1} is proportional to the Dirichlet to Neumann map of the potential φ_{β_1} .

The "surface to volume dissipation" is a measure of the particles ability to dissipate energy on its surface relative to the energy dissipated in its interior. For spherical particles filled with isotropic material of conductivity c_p we show in Section III that this ratio is proportional to the reciprical of the sphere radius " a " and is given by $\beta_1 = \frac{2}{c_p a}$. For more general shapes we provide lower bounds on the "surface to volume dissipation" in terms of the physical dimensions of the particle: see Section III.

For a fixed configuration of particles occupying the region A_p we add a new particle Σ of conductivity $\mathbf{c}_p$ to the suspension. We assume that the particle Σ is compactly contained in the matrix, (ie., $\Sigma \subset A_m$ and $\partial\Sigma \cap \partial A_m = \emptyset$). The region occupied by the particle phase is now given by $A_p \cup \Sigma$ and the associated effective conductivity tensor is denoted by $\tilde{\mathbf{c}}_e$. The principal result given in this paper is the following criterion on the particle geometry:

1.1 Energy Dissipation Inequality

Given a reinforcement particle " Σ ", if its "surface to volume dissipation" β_1 satisfies:

$$\mathbf{P}_{cr}^{-1} \leq \mathbf{c}_p \beta_1(\mathbf{c}_p), \quad (1.10)$$

then

$$\tilde{\mathbf{c}}_e \geq \mathbf{c}_e. \quad (1.11)$$

Here (1.10) and (1.11) hold in the sense of quadratic forms. No assumptions on the topological nature of the particle domain Σ is made. Indeed it can be a disjoint union of multiply connected components. This inequality is established in Section II. The "surface to volume dissipation" of a sphere is calculated in Section III. For particles that are starlike we provide lower bounds on β_1 in Section III. These observations together with the energy dissipation inequality are applied in Section IV to obtain energy dissipation inequalities in terms of the physical dimensions of the particle. Such size effect inequalities predict the existence of a critical particle dimension below which the particle will always increase the

overall conductivity of the composite. These predictions apply to a wide class of particle shapes.

Earlier size effects have been found in the context of isotropic, monodisperse suspensions of isotropically conducting spheres in an isotropically conducting matrix. The conductivities of the matrix and particles are given by the scalars c_p and c_m and the quantity $\mathbf{P}_{cr}$ reduces to the scalar $P_{cr} = \alpha(c_m - c_p)^{-1}$. For this case the results have focused on critical radii for a monodisperse suspension of spheres. Here the critical radius is precisely $2P_{cr}$ and is that for which the overall conductivity of the composite equals that of the matrix conductivity. The critical radius was found by Lipton [2] and by Torquato and Rintoul [3] using different methods. In [2], this effect is shown to persist even if the suspension is not isotropic. Moreover, the electric field is shown to be uniform throughout the composite and is precisely $\mathbf{E}$: see Lipton [2]. For isotropic monodisperse suspensions of isotropically conducting spheres Torquato and Rintoul [3] have shown that the overall conductivity lies above the matrix value when the common sphere radii are below $2P_{cr}$ and that the overall conductivity is decreased when the radii lie above $2P_{cr}$. More recently, results involving the statistics of the size distribution of spheres have been found in the context of isotropic polydisperse suspensions of spheres: see Lipton [4]. There it is shown that if the arithmetic mean of the sphere radii lie below $2P_{cr}$ then the effective conductivity lies above the matrix value. It is also shown that if the harmonic mean of the sphere radii lie above $2P_{cr}$ then the effective conductivity lies below that of the matrix. In the final Section we show that this phenomenon persists even when the suspension is anisotropic.

2 Energy dissipation inequality

We establish the energy dissipation inequality given in Section I. For any constant vector $\mathbf{E}$ in R^3 , we can write the difference $\delta = \tilde{\mathbf{c}}_e \mathbf{E} \cdot \mathbf{E} - \mathbf{c}_e \mathbf{E} \cdot \mathbf{E}$ as:

$$\delta = C(A_p, \tilde{\varphi}) - C(A_p, \varphi) + R(\Sigma, \tilde{\varphi}), \quad (2.1)$$

where $\tilde{\varphi}$ is the potential associated with the configuration of particles given by $A_p \cup \Sigma$ and the remainder $R(\Sigma, \tilde{\varphi})$ is given by:

$$R(\Sigma, \tilde{\varphi}) = \alpha \int_{\partial\Sigma} |\nabla_s \tilde{\varphi}|^2 ds - \int_{\Sigma} (\mathbf{c}_m - \mathbf{c}_p) \nabla \tilde{\varphi} \cdot \nabla \tilde{\varphi} dx. \quad (2.2)$$

Noting that $\tilde{\varphi}$ is an admissible trial for the variational principle (1.4) we have:

$$C(A_p, \tilde{\varphi}) - C(A_p, \varphi) \geq 0, \quad (2.3)$$

thus

$$\delta \geq R(\Sigma, \tilde{\varphi}). \quad (2.4)$$

From (1.8) it follows that:

$$\int_{\partial\Sigma} |\nabla_s \phi|^2 ds - \beta_1 \int_{\Sigma} \mathbf{c}_p \nabla \phi \cdot \nabla \phi dx \geq 0, \quad (2.5)$$

for every function ϕ such that $\text{div}(\mathbf{c}_p \nabla \phi) = 0$. Comparing the righthand side of (2.2) with (2.5), we discover that:

$$\delta \geq 0 \quad (2.6)$$

for

$$\alpha^{-1}(\mathbf{c}_m - \mathbf{c}_p) \leq \mathbf{c}_p \beta_1(\mathbf{c}_p) \quad (2.7)$$

and the energy dissipation inequality follows. We observe that strict inequality in (2.6) follows from strict inequality in (2.7) provided $\tilde{\varphi}$ is not identically equal to a constant in Σ .

3 The surface to volume dissipation of a sphere and lower bounds on β_1 for starlike particles

We calculate the surface to volume dissipation for a spherical particle of radius a filled with material of unit conductivity. Separation of variables in the eigenvalue problem (1.9) shows that the surface to volume dissipation is given by:

$$\beta_1 = \frac{2}{a} \quad (3.1)$$

The associated eigenspace is given by the span of the coordinate functions x_1, x_2, x_3 restricted to the surface of the sphere. We remark that separation of variables shows that the set of eigenvalues for the eigenvalue problem (1.9) is given by $n(n+1)/a$ for $n = 1, 2, \dots$. The associated eigenspace for the n^{th} eigenvalue is the span of the spherical harmonics of order " n " given by: $a^n e^{im\omega} P_n^m(\cos(\theta))$, $m = 1, \dots, n$ restricted to the surface of the sphere. Here $P_n^m(x)$ are the Legendre functions and ω, θ are polar coordinates on the unit sphere. For a sphere filled with an isotropic conductor of conductivity c_p we easily obtain that $\beta_1 = \frac{2}{c_p a}$.

Next we consider particles Σ that are starlike with respect to a point $\check{x}$ inside Σ . We introduce the quantity $h_m(\check{x}) \equiv \min_{\mathbf{x} \in \partial\Omega} \{(\mathbf{x} - \check{x}) \cdot \mathbf{n}\}$. Here $\mathbf{n}$ is the unit outward normal to the boundary of Σ . The minimum distance from the point $\check{x}$ to a tangent plane on the boundary is given by $h_m(\check{x})$. Since the domain is starlike with respect to $\check{x}$ we have $h_m(\check{x}) > 0$. The maximum distance from $\check{x}$ to a point on the boundary is given by $r_M(\check{x})$. We denote the largest eigenvalue of the particle conductivity tensor $\mathbf{c}_p$ by γ and state the following inequality:

3.1 A lower bound on the surface to volume dissipation for starlike particles

If the particle Σ is starlike with respect to the point $\check{x}$ then,

$$\beta_1(\mathbf{c}_p) \geq \frac{1}{r_M(\check{x})} \left(\frac{1/\gamma}{3 + r_M(\check{x})/h_m(\check{x})} \right). \quad (3.2)$$

To establish the lower bound we may assume that the particle Σ is starlike with respect to the origin and appeal to the identity:

$$\partial_k \{ (x_k(\mathbf{c}_p)_{ij} - x_i(\mathbf{c}_p)_{kj} - x_j(\mathbf{c}_p)_{ik}) \partial_i u \partial_j u \} = -2(\mathbf{x} \cdot \nabla u)(\text{div}(\mathbf{c}_p \nabla u)) + \mathbf{c}_p \nabla u \cdot \nabla u. \quad (3.3)$$

Here repeated indices are to be summed. Integration over the particle Σ and application the Gauss-Green theorem gives:

$$\int_{\partial\Sigma} (\mathbf{n} \cdot \mathbf{x}) \mathbf{c}_p \nabla u \cdot \nabla u - 2(\mathbf{x} \cdot \nabla u)(\mathbf{n} \cdot \mathbf{c}_p \nabla u) ds = -2 \int_{\Sigma} (\mathbf{x} \cdot \nabla u)(\text{div}(\mathbf{c}_p \nabla u)) dx + \int_{\Sigma} \mathbf{c}_p \nabla u \cdot \nabla u dx. \quad (3.4)$$

We consider trial functions u such that $\text{div}(\mathbf{c}_p \nabla) = 0$ inside Σ and we decompose the gradient ∇u on the boundary $\partial\Sigma$ into its normal and tangential parts to obtain:

$$\begin{aligned} \int_{\partial\Sigma} (\mathbf{n} \cdot \mathbf{x}) \mathbf{c}_p \nabla_s u \cdot \nabla_s u ds - \int_{\partial\Sigma} (\mathbf{c}_p \mathbf{n} \cdot \mathbf{n}) (\mathbf{n} \cdot \mathbf{x}) (\partial_n u)^2 ds - 2 \int_{\partial\Sigma} (\mathbf{c}_p \mathbf{n} \cdot \mathbf{n}) (\mathbf{x} \cdot \nabla_s u) (\partial_n u) ds \\ - 2 \int_{\partial\Sigma} (\mathbf{x} \cdot \nabla_s u) (\mathbf{c}_p \mathbf{n} \cdot \nabla_s u) ds = \int_{\Sigma} \mathbf{c}_p \nabla u \cdot \nabla u dx. \end{aligned} \quad (3.5)$$

Here we have written the normal derivative $\mathbf{n} \cdot \nabla u$ as $\partial_n u$. Next we apply Cauchy's inequality to find:

$$\begin{aligned} -2(\mathbf{c}_p \mathbf{n} \cdot \mathbf{n}) (\mathbf{x} \cdot \nabla_s u) (\partial_n u) &\leq (\mathbf{c}_p \mathbf{n} \cdot \mathbf{n}) (\zeta (\mathbf{x} \cdot \nabla_s u)^2 + \zeta^{-1} (\partial_n u)^2), \\ -2(\mathbf{x} \cdot \nabla_s u) (\mathbf{c}_p \mathbf{n} \cdot \nabla_s u) &\leq 2|\mathbf{c}_p \mathbf{n}| |\mathbf{x}| |\nabla_s u|^2, \end{aligned} \quad (3.6)$$

and

$$(\mathbf{x} \cdot \nabla_s u)^2 \leq |\mathbf{x}|^2 |\nabla_s u|^2, \quad (3.7)$$

Here ζ is any positive real number. Applying (3.6) and (3.7), noting that $\mathbf{c}_p \leq \gamma I$ in the sense of quadratic forms, and $|\mathbf{n}| = 1$ we obtain:

$$\begin{aligned} \int_{\Sigma} \mathbf{c}_p \nabla u \cdot \nabla u dx &\leq \\ \int_{\partial\Sigma} \gamma (2|\mathbf{x}| + \mathbf{n} \cdot \mathbf{x} + \zeta |\mathbf{x}|^2) |\nabla_s u|^2 ds - \int_{\partial\Sigma} \gamma (\mathbf{n} \cdot \mathbf{x} - \zeta^{-1}) (\partial_n u)^2 ds. \end{aligned} \quad (3.8)$$

We have the inequality $h_m(0) \leq \mathbf{n} \cdot \mathbf{x}$ for $\mathbf{x}$ on the boundary of Σ . Thus we set $\zeta^{-1} = h_m(0)$ to find:

$$\int_{\Sigma} \mathbf{c}_p \nabla u \cdot \nabla u dx \leq \int_{\partial\Sigma} \gamma (2|\mathbf{x}| + \mathbf{n} \cdot \mathbf{x} + h_m(0)^{-1} |\mathbf{x}|^2) |\nabla_s u|^2 ds. \quad (3.9)$$

Lastly we note that $\mathbf{n} \cdot \mathbf{x} \leq |\mathbf{x}|$, to obtain:

$$\int_{\Sigma} \mathbf{c}_p \nabla u \cdot \nabla u dx \leq \gamma \{3r_M(0) + r_M(0)^2/h_m(0)\} \int_{\partial\Sigma} |\nabla_s u|^2 ds. \quad (3.10)$$

The lower bound on β_1 follows noting that (3.10) holds for all functions u such that $\text{div}(\mathbf{c}_p \nabla u) = 0$ on Σ .

4 Novel size effects for particle reinforced composites with interfacial surface conduction

It follows from the energy dissipation inequality and (3.1) that if Σ is a sphere of radius a filled with isotropic conductor c_p , that:

4.1 Size effect for spheres

$$\tilde{\mathbf{c}}_e \geq \mathbf{c}_e \quad (4.1)$$

if

$$aI \leq 2\alpha(c_p \mathbf{I} - \mathbf{c}_m)^{-1}. \quad (4.2)$$

This inequality holds in the sense of quadratic forms. (ie., (4.1) holds if a is less than or equal to the least eigenvalue of the matrix $2\alpha(c_p\mathbf{I} - \mathbf{c}_m)^{-1}$.)

When both matrix and particles have isotropic conductivities specified by c_m and c_p respectively then:

4.2 Size effect for an isotropically conducting spherical particle in an isotropic matrix

$$\tilde{\mathbf{c}}_e \geq \mathbf{c}_e \quad (4.3)$$

if

$$a \leq 2P_{cr} = 2\alpha(c_m - c_p)^{-1}. \quad (4.4)$$

More generally, we consider starlike inclusions Σ . We apply the inequality (3.2) together with the energy dissipation inequality to obtain:

4.3 Size effect for starlike particles

Given that Σ is starlike with respect to a point $\check{x}$ inside Σ , then:

$$\tilde{\mathbf{c}}_e \geq \mathbf{c}_e \quad (4.5)$$

if

$$\{r_M(\check{x})(3 + r_M(\check{x})/h_m(\check{x}))\}\gamma\mathbf{c}_p^{-1} \leq \mathbf{P}_{cr}. \quad (4.6)$$

We consider an ellipsoidal particle. Here we suppose that the half lengths of the major and minor axes are specified by a and c respectively. For this case we choose " $\check{x}$ " to be the center of mass for the ellipse and it follows that $r_M = a$, $h_M = c$, and we have:

4.4 Size effect for ellipsoidal reinforcement

Given an ellipsoidal reinforcement Σ with major and minor axes specified by a and c respectively, then:

$$\tilde{\mathbf{c}}_e \geq \mathbf{c}_e \quad (4.7)$$

if

$$\{a(3 + a/c)\}\gamma\mathbf{c}_p^{-1} \leq \mathbf{P}_{cr} \quad (4.8)$$

Next we consider cylindrical inclusions of length ℓ and radius R and choose $\check{x}$ to be the center of mass of the cylinder. If $\ell/2 \geq R$ then $r_M = ((\ell/2)^2 + R^2)^{1/2}$ and $h_m = R$. On the other hand if $\ell/2 \leq R$, then: $h_m = \ell/2$. Such inclusions can be used to model chopped fiber suspensions. We have:

4.5 Size effect for a cylindrical inclusion with $\ell/2 \geq R$:

$$\tilde{\mathbf{c}}_e \geq \mathbf{c}_e, \quad (4.9)$$

if

$$\{(3R + \sqrt{(\ell/2)^2 + R^2})\sqrt{(\frac{\ell}{2R})^2 + 1}\}\gamma\mathbf{c}_p^{-1} \leq \mathbf{P}_{cr}. \quad (4.10)$$

4.6 Size effect for a cylindrical inclusion with $\ell/2 \leq R$:

$$\tilde{c}_e \geq c_e, \quad (4.11)$$

if

$$\{(3\ell/2 + \sqrt{(\ell/2)^2 + R^2})\sqrt{(\frac{2R}{\ell})^2 + 1}\} \gamma c_p^{-1} \leq P_{cr}, \quad (4.12)$$

We remark that these inequalities may be used to select particle dimensions for the design of composites with optimal DC conductivity.

5 Size effects based on the statistics of the particle size distribution for anisotropic suspensions of anisotropic conductors

We provide a new criteria on the suspension geometry for which the overall conductivity lies below the conductivity of the matrix material.

We write the overall electric energy dissipation due to a prescribed potential $\phi = \mathbf{E} \cdot \mathbf{x}$ on the boundary $\partial\Omega$ as:

$$c_e \mathbf{E} \cdot \mathbf{E} = \min_{\phi \in V} \{C(A_p, \phi)\} \quad (5.1)$$

where

$$C(A_p, \phi) = \int_{\Omega} c_m \nabla \phi \cdot \nabla \phi dx - \int_{A_p} (c_m - c_p) \nabla \phi \cdot \nabla \phi dx + \alpha \int_{\Gamma} |\nabla_s \phi|^2 ds. \quad (5.2)$$

The energy dissipated in the region Ω when filled with pure matrix conductor is simply $c_m \mathbf{E} \cdot \mathbf{E}$. Thus substitution of $\phi = \mathbf{E} \cdot \mathbf{x}$ into (5.1) gives the estimate:

$$c_e \mathbf{E} \cdot \mathbf{E} \leq c_m \mathbf{E} \cdot \mathbf{E} + T(A_p, \mathbf{E}), \quad (5.3)$$

where $T(A_p, \mathbf{E}) \equiv - \int_{A_p} (c_m - c_p) \mathbf{E} \cdot \mathbf{E} dx + \alpha \int_{\Gamma} |\nabla_s (\mathbf{E} \cdot \mathbf{x})|^2 ds$. A straight forward computation shows that:

$$T(A_p, \mathbf{E}) = \{\alpha \mathbf{M} - |A_p| (c_m - c_p)\} \mathbf{E} \cdot \mathbf{E}, \quad (5.4)$$

where

$$\mathbf{M} = \int_{\Gamma} \{\mathbf{I} - \mathbf{n} \otimes \mathbf{n}\} ds = -3 \int_{\Gamma} \mathbf{x} \otimes \mathbf{n} \mathcal{H} ds, \quad (5.5)$$

Here $\mathcal{H}$ is the mean curvature of the interface. It is evident that the inequality (5.3) delivers the following criterion:

5.1 Energy dissipation inequality

$$\text{If } |A_p|^{-1} \mathbf{M} \mathbf{E} \cdot \mathbf{E} \leq P_{cr}^{-1} \mathbf{E} \cdot \mathbf{E} \quad \text{then} \quad c_e \mathbf{E} \cdot \mathbf{E} \leq c_m \mathbf{E} \cdot \mathbf{E}.$$

For a polydisperse suspension of N spheres of radii $a_1, a_2, \dots, a_N$, calculation shows that:

$$|A_p|^{-1} \mathbf{M} = 2 \langle a^{-1} \rangle \mathbf{I}, \quad (5.6)$$

where $\langle a^{-1} \rangle$ is the volume average of the inverse radii given by:

$$\langle a^{-1} \rangle = |A_p|^{-1} \sum_{i=1}^N a_i^{-1} |B_i|. \quad (5.7)$$

Here $|B_i|$ is the volume of the i^{th} sphere and $|A_p| = \sum_{i=1}^N |B_i|$. Thus for polydisperse suspensions of spheres we have:

5.2 Energy dissipation inequality based upon the size distribution of spheres

$$\text{If } \langle a^{-1} \rangle \leq \frac{(2\mathbf{P}_{cr})^{-1} \mathbf{E} \cdot \mathbf{E}}{|\mathbf{E}|^2} \text{ then } \mathbf{c}_e \mathbf{E} \cdot \mathbf{E} \leq \mathbf{c}_m \mathbf{E} \cdot \mathbf{E}.$$

We denote the smallest eigenvalue of the matrix $(2\mathbf{P}_{cr})^{-1}$ by λ_c . It follows immediately from the previous inequality that:

$$\text{If } \langle a^{-1} \rangle \leq \lambda_c \text{ then } \mathbf{c}_e \leq \mathbf{c}_m \text{ in the sense of quadratic forms.}$$

We conclude noting that when both matrix and particle conductors are isotropic that $\lambda_c = \frac{c_m - c_p}{2\alpha}$ and we have the following:

5.3 Energy dissipation inequality for isotropically conducting spheres in an isotropic matrix.

$$\mathbf{c}_e \leq \mathbf{c}_m \text{ in the sense of quadratic forms when } \langle a^{-1} \rangle \geq \frac{2\alpha}{c_m - c_p}.$$

We stress that these results hold for anisotropic suspensions.

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