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Weighted effective medium approximations for conductivity of random composites

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Abstract

Effective medium approximations are developed to estimate the conductivity of composite media that are random mixtures of inhomogeneities of various shapes. The approximations use dilute solution results for inclusions of certain shapes embedded in the effective medium matrix, and incorporate weight parameters to reflect the proportions of the inhomogeneities of various shapes from different component materials in a mixture. Since the approximations are based on the differential scheme, which is realizable by a certain hierarchical model, they never violate the mathematical requirements imposed upon a homogenized property including the bounds. As illustrations, the approximations are used to interpret empirical data on the electrical conductivity of water-saturated sedimentary rocks, and the thermal conductivity of some multiphase dispersions of metallic particles in silicone rubber matrix.

Keywords: Random composite; Conductivity; Effective medium approximation; Weight parameter

1. Introduction

Macroscopic (effective) properties of randomly inhomogeneous materials, which appear statistically homogeneous and isotropic, depend upon the properties, volume proportions, and particular geometry of the constituents. The micro geometry of a random mixture is usually irregular, which makes the problem of direct calculation of an effective property intractable. With the only available information about the properties and volume proportions of the component materials, the best estimates for the effective conductivity (thermal, electrical,...) k_e of an isotropic composite are those of Hashin and Shtrikman [1–4]

$$P_k(2k_{max}) \geqslant k_e \geqslant P_k(2k_{min}), \tag{1}$$

where

$$P_k(k_*) = \left(\sum_{i=1}^{N} \frac{v_i}{k_i + k_*}\right)^{-1} - k_*, \tag{2}$$

$$k_{max} = \max\{k_1, \ldots, k_N\}, \quad k_{min} = \min\{k_1, \ldots, k_N\}, \tag{3}$$

v_i, k_i $(i = 1, \ldots, N)$ are the volume proportions and conductivities of the constituents of the N-component composite.

To get more-definite values of the effective properties, one should incorporate specific information about the particular geometry of a composite into some idealistic geometric model in various effective medium approximations [5–20]. To determine the fields within the inhomogeneities, one often considers each of them as an inclusion of spherical (or ellipsoidal) shape embedded in the matrix of effective medium, and uses the respective exact dilute solution result. However, the inhomogeneities within a practical random mixture generally would not have a single specific shape, but various ones, and this fact should be accounted for in a weighted effective medium scheme presented in this study.

2. Weighted effective medium approximation

To find an effective property one should determine the respective fields within the inhomogeneities of a mixture. The inhomogeneities from a particular composite may

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Nomenclature

k_e the effective conductivity of a composite
k_i $(i = 1,\ldots,N)$ the conductivities of the constituents of the N-component composite
k_I, k_M the conductivities of the inclusion and matrix components
k_w the electrical dc conductivity of the water filling pores
v_i $(i = 1,\ldots,N)$ the volume proportions of the composite's components
v_I the volume proportion of the inclusion component
v_w the porosity

have some relatively specific shapes. However, without any definite information about the specific arrangements of the inhomogeneities within a composite (as it is usually the case for a completely random mixture, consult e.g. [6,7,21]), the best one can do, in calculating the fields within an inhomogeneity of certain shape, is to consider the inhomogeneity as embedded in the effective medium. Using the dilute solution result for an inclusion (of conductivity k_I) embedded in the matrix (of conductivity k_M) one has the formal expression for the effective conductivity k_e of the dilute mixture:

$$k_e/k_M = 1 + v_I D(k_I, k_M) + O(v_I^2), \quad v_I \ll 1, \tag{4}$$

where v_I is the volume fraction of the inclusions, $D(k_I, k_M)$ some inclusion-shape-dependent function. If all the inhomogeneities of the composite have approximately the same shape, the effective medium approximation equation for the effective conductivity k_e of the N-component material reads (see more details in Appendix A)

$$\sum_{i=1}^{N} v_i D(k_i, k_e) = 0 \quad \left(\sum_{i=1}^{N} v_i = 1\right). \tag{5}$$

This effective medium equation in the classical sense, by construction, should be restricted to the class of quasisymmetric mixtures because it treats all the inclusions geometrically in the same fashion. More generally, the inhomogeneities from different component materials would have different shapes represented by the different shape functions $D_i(k_I, k_M)$, hence the respective effective medium equation should have the form

$$\sum_{i=1}^{N} v_i D_i(k_i, k_e) = 0. \tag{6}$$

Generally, the inhomogeneities of every i-component material may also have different shapes described by the shape functions $D_{i\alpha_i}(k_I, k_M)$ and respective volume proportions $v_{i\alpha_i}(\alpha_i = 1,\ldots,N_i)$ of the inclusions having the same shapes. Then we have

$$\sum_{i=1}^{N} \sum_{\alpha_i=1}^{N_i} v_{i\alpha_i} D_{i\alpha_i}(k_i, k_e) = 0 \quad \left(\sum_{\alpha_i=1}^{N_i} v_{i\alpha_i} = v_i;\ i = 1,\ldots,N\right). \tag{7}$$

Eq. (7) is the general weighted effective medium equation for N-component mixture, which incorporates the weight parameters $v_{i\alpha_i}$ and the respective shape functions $D_{i\alpha_i}$ reflecting distribution of various shapes within a practical random mixture.

Using the differential scheme [5–7,11,13,15–17] one can show that the effective conductivities determined by the Eqs. (5), (6), or (7), correspond, at least, to some geometric hierarchical models constructed incrementally, which adds at each step an infinitesimal amount of well separated inhomogeneities of all shapes and from all the constituents (with the relative proportions corresponding to the final composition of the composite) into the mixture of the previous step (constructed from the geometrically similar but smaller-scale inhomogeneities!) until the final composition of the mixture is reached. This fact secures the effective medium approximations (5)–(7) certain mathematical justification: they never violate the bounds (1)–(3) or any exact mathematical restrictions imposed upon an effective property.

It is difficult to find the explicit expression of the shape function $D(k_I, k_M)$ for an inclusion of arbitrary shape. From (1)–(4) one can find that

$$(k_I - k_M)\left(\frac{1}{3}k_I + \frac{2}{3}k_M\right)^{-1} \leqslant D(k_I, k_M) \leqslant (k_I - k_M)\left(\frac{1}{3}k_I^{-1} + \frac{2}{3}k_M^{-1}\right), \tag{8}$$

when $k_I > k_M$, and the order is reversed when $k_I < k_M$.

For an ellipsoidal inclusion with the aspect ratio a:b:c, one has [13]

$$D(k_I, k_M) = \frac{k_I - k_M}{3}\left[\frac{1}{k_I A + k_M(1-A)} + \frac{1}{k_I B + k_M(1-B)} + \frac{1}{k_I C + k_M(1-C)}\right], \tag{9}$$

where

$$A = \frac{abc}{2}\int_0^\infty \frac{dt}{(a^2+t)\Delta(t)}, \quad B = \frac{abc}{2}\int_0^\infty \frac{dt}{(b^2+t)\Delta(t)},$$
$$C = \frac{abc}{2}\int_0^\infty \frac{dt}{(c^2+t)\Delta(t)}, \quad \Delta(t) = \sqrt{(a^2+t)(b^2+t)(c^2+t)}. \tag{10}$$

For a spheroidal inclusion with the aspect ratio a:b:b, (9) and (10) reduce to

$$D(k_{\rm I},k_{\rm M}) = \frac{k_{\rm I}-k_{\rm M}}{3}\left[\frac{1}{k_{\rm I}A+k_{\rm M}(1-A)}+\frac{4}{k_{\rm I}(1-A)+k_{\rm M}(1+A)}\right], \tag{11}$$

where

$$A = \frac{ab^2}{2}\int_0^\infty \frac{{\rm d}t}{(a^2+t)^{3/2}(b^2+t)}. \tag{12}$$

The three limiting cases of a spheroidal inclusion are: sphere $A = 1/3$; circular cylinder (needle) $A = 0$; circular disk (platelet) $A = 1$. According to (7), (11), (12), the weighted effective medium approximation for the N-component mixture having just these extreme shapes is determined by the equation

$$\sum_{i=1}^{N}(k_i-k_{\rm e})\left[\frac{9v_{is}}{k_i+2k_{\rm e}}+v_{in}\left(\frac{1}{k_{\rm e}}+\frac{4}{k_i+k_{\rm e}}\right)+v_{ip}\left(\frac{1}{k_i}+\frac{2}{k_{\rm e}}\right)\right]=0, \tag{13}$$

where v_{is}, v_{in}, v_{ip} $(i = 1,\ldots,N)$ are the weights (volume fractions) of the inhomogeneities from the i-component that have the spherical, needle, and platelet shapes, respectively.

More general, according to (7), (11), (12), the weighted spheroidal effective medium approximation for the N-component mixture having the range of shapes with geometric parameters $A_{i\alpha_i}$ and respective weights (volume proportions) $v_{i\alpha_i}$ would be

$$\sum_{i=1}^{N}(k_i-k_{\rm e})\sum_{\alpha_i=1}^{N_i}v_{i\alpha_i}\left[\frac{1}{k_iA_{i\alpha_i}+k_{\rm e}(1-A_{i\alpha_i})}+\frac{4}{k_i(1-A_{i\alpha_i})+k_{\rm e}(1+A_{i\alpha_i})}\right]=0. \tag{14}$$

Eq. (14) contains a large amount of possible shapes, including the extreme one-dimensional needle, two-dimensional platelet, and three-dimensional spherical ones of (13) as well as intermediate shapes between. For an example we consider two-component spheroidal mixtures with $N = 2$, $N_1 = N_2 = 1$, and respective geometric parameters $A_1, A_2, k_1 = 1$, $k_2 = 20$, $v_2 = 0 \to 1$. The mixtures with the geometric parameters varying over all the range $0 \leqslant A_1$, $A_2 \leqslant 1$, cover the major part between the bounds (1)–(3) and enveloped by the two extreme sphere-1-platelet-2 (spheres from phase 1 mixed with platelets from phase 2) and platelet-1-sphere-2 (platelets from phase 1 mixed with spheres from phase 2) models, as reported in Fig. 1. This observation indicates that the spheroidal models in our particular scheme here might be representative enough to approximate most random mixtures (in the Maxwell–Fricke model for two-phase mixtures used in [12], the spheroidal inclusions are shown to be sufficient to cover all at the interval between the Hashin–Shtrikman bounds).

On a higher level of generality, according to (7), (9), (10), we have the weighted ellipsoidal effective medium approximation with the respective weights $v_{i\alpha_i}$ and geometric parameters $A_{i\alpha_i}, B_{i\alpha_i}, C_{i\alpha_i}$:

$$\sum_{i=1}^{N}(k_i-k_{\rm e})\sum_{\alpha_i=1}^{N_i}v_{i\alpha_i}\left[\frac{1}{k_iA_{i\alpha_i}+k_{\rm e}(1-A_{i\alpha_i})}+\frac{1}{k_iB_{i\alpha_i}+k_{\rm e}(1-B_{i\alpha_i})}+\frac{1}{k_iC_{i\alpha_i}+k_{\rm e}(1-C_{i\alpha_i})}\right]=0. \tag{15}$$

Eq. (15) has unique solution (see Appendix B).

To use the most general weighted effective medium approximation (7) one should define the shape function D from the dilute solution result (4) for an inclusion of arbitrary shape explicitly. Clearly there is no general analytical solution. One possible way to approximate the function is to compare the numerical result (4) for some dilute solution of the inclusion of the given shape with that of spheroidal one from (11) and some fitting free parameter A, or with that of the ellipsoidal one from (9), (10) and more fitting parameters A, B, C, or broader with any multi-parameter function satisfying restriction (8). However, in this work, we would not elaborate further in this direction. The free weight parameters $A_{i\alpha_i}$ of (14), or those of (15) may also be chosen just to fit empirical data for the macroscopic conductivity of a practical mixture. Then the parameters have the physical sense as reflecting the distribution of possible shapes within the mixture. An illustrative example of the application of the weighted effective medium approximation will be given in the next section.

3. Water-saturated porous rocks

Experimental data for the electrical dc conductivity of a large variety of water-saturated sedimentary rocks leads to an empirical law for the aggregate conductivity [22,9]

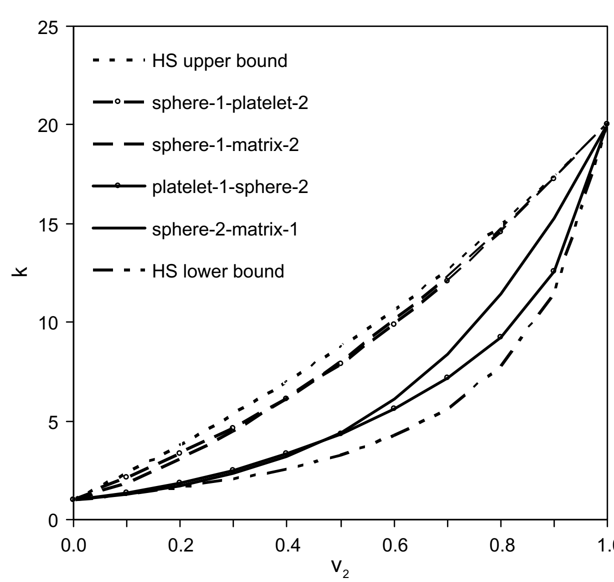

Fig. 1. Hashin–Shtrikman bounds, extreme spheroidal models (sphere-1-platelet-2 and platelet-1-sphere-2), and differential models (sphere-1-matrix-2 and sphere-2-matrix-1) for the effective conductivity of two-phase mixtures with $k_1 = 1$, $k_2 = 20$, $v_2 = 0 \to 1$.

$$k_e = k_w v_w^m, \quad 1.3 \leqslant m \leqslant 4, \tag{16}$$

where k_w and v_w are, respectively, the water conductivity and volume fraction of water-filled pores (the porosity); m is called the cementation index.

The considered sedimentary rocks can be considered as two-phase media composed of solid rock grains of zero conductivity ($k_r = 0$) and water filling the intergranular pore spaces. Geometric rock models, that can explain the empirical law (16) have been the subject of many studies (e.g. [9,10,13,15]). The highly consolidated rock having interconnected pore space filled with water can be modeled as a random mixture of the components: the solid rock granules of approximately spherical form (volume fraction v_{rs}); the platelet water films (volume fraction v_{wp}) positioned between the neighboring grains; the cylindrical (needle) water tunnels (volume fraction v_{wn}) positioned at the intersections of the grain boundaries; the spherical water pockets (volume fraction v_{ws}) positioned at the intersection corners of the rock grains (else the intersections of the water tunnels). This is the picture of a two-phase mixture having the geometrically complicated and interconnected water phase with volume fraction $v_w = v_{ws} + v_{wp} + v_{wn} = 1 - v_{rs}$. Application of the weighted effective medium approximation (13) would yield the equation determining the effective conductivity k_e of the rock aggregate (keeping in mind $k_r = 0$)

$$(k_w - k_e)\left[\frac{9v_{ws}}{k_w + 2k_e} + v_{wn}\left(\frac{1}{k_e} + \frac{4}{k_w + k_e}\right) + v_{wp}\left(\frac{1}{k_w} + \frac{2}{k_e}\right)\right] - \frac{9}{2}v_{rs} = 0. \tag{17}$$

Let the geometric parameter $g_p = v_{wp}/v_w$ denote the proportion of the conductivity-effective water film within the water phase filling pore space, while $g_s = v_{ws}/v_w$ that of the water pockets, which contribute least to the aggregate conductivity (the remaining water tunnels yield intermediate contributions). Then Eq. (17) can be rewritten as

$$(k_w - k_e)\left[\frac{9g_s}{k_w + 2k_e} + (1 - g_s - g_p)\left(\frac{1}{k_e} + \frac{4}{k_w + k_e}\right) + g_p\left(\frac{1}{k_w} + \frac{2}{k_e}\right)\right] - \frac{9(1 - v_w)}{2v_w} = 0. \tag{18}$$

In Fig. 2a we plot some empirical curves (16) corresponding to $m = 1.4$; $m = 1.8$; $m = 3$, and some models from (18) closed to them with geometric characteristics $g_p = 0.9$, $g_s = 0.1$; $g_p = 0.3$, $g_s = 0.6$; $g_p = 0.1$, $g_s = 0.9$; respectively. In Fig. 2b we report the Hashin–Shtrikman upper bound (from (1), the lower bound in this case is the trivial zero conductivity), the extreme curves of the empirical law (16) at $m = 1.3$ and $m = 4$, and some models of (18): $g_p = 1$, $g_s = 0$ (pure platelet water films); $g_p = 0$, $g_s = 0$ (pure water tunnels); $g_p = 0.2$, $g_s = 0.7$ (the water-filled pore space includes all three extreme shapes: films, pockets, tunnels). The model $g_p = 1$, $g_s = 0$ represents the highest values of the aggregate conductivity the weighted spheroidal approximation can give.

4. Differential scheme equations and some silicone rubber matrix mixtures

Consider a matrix mixture as a $(N + 1)$-component composite with the inclusions from N components dispersed in a continuous phase (the $(N + 1)$th component). Each inclusion component is supposed to be composed of the inclusions made of the same material and having the same shape characterized by the same shape function $D(k_I, k_M)$ stated in (4). To construct the $(N + 1)$-component composite by the differential scheme, one starts with the base matrix, and adds incrementally well separated inclusions from the remaining N components in their relative volume proportions until the final composition is reached [5–7,11,13,15,16]. The resulting differential equation corresponding to the weighted effective medium approximation procedure proposed above has the form (see Appendix A)

$$\frac{dk}{dt} = \frac{k}{1 - v_I t}\sum_{i=1}^{N}\sum_{\alpha_i=1}^{N_i} v_{i\alpha_i} D_{i\alpha_i}(k_i, k), \quad v_I = \sum_{i=1}^{N}\sum_{\alpha_i=1}^{N_i} v_{i\alpha_i}, \quad k(0) = k_{N+1}, \quad 0 \leqslant t \leqslant 1, \tag{19}$$

and $k_e = k(1)$. In the limit of vanishing matrix phase $v_{N+1} = 0$, $v_I = 1$, from (19) we obtain the weighted effective medium approximation for the N-phase mixture stated in (7). While being realizable-at least-by a nonrealistic hierarchical model, the scheme aims to approximate behavior of usual nonhierarchical mixtures. The differential scheme can take into account the shapes of the inclusions, but not that

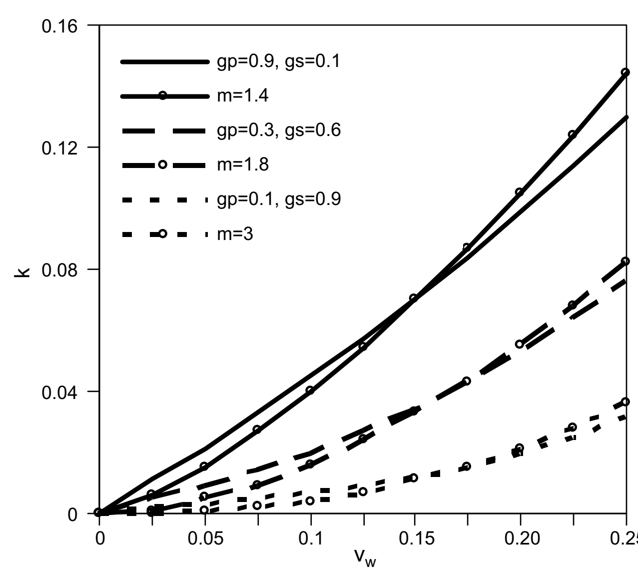

Fig. 2a. Some conductivity against porosity curves from Archie's law (with $m = 1.4, 1.8, 3$), and models containing proportions of the water-saturated pores in the forms of water films (g_p), pockets (g_s), and tunnels (the remaining).

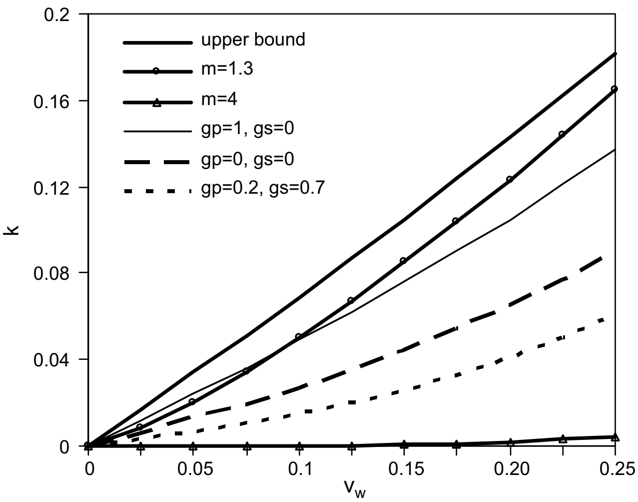

Fig. 2b. Some extreme conductivity against porosity curves for sedimentary rocks: The Hashin–Shtrikman upper bound; Archie's law at ($m = 1.3, 4$); Pure water film model ($g_p = 1,\ g_s = 0$); Pure water tunnel model ($g_p = 0,\ g_s = 0$); A mixed model ($g_p = 0.2,\ g_s = 0.7$).

of the matrix phase, except the fact that the phase remains continuous throughout the whole process. Alternatively, one could also take the matrix phase as composed of inclusions of extreme platelet form (see (11) with $A = 1$), and then apply the weighted effective medium approximation (7) directly to the composite. The inclusions of extreme platelet form (approaching infinitesimal thickness and infinite extensions) disorderly oriented in the material space also form interconnected phase as a matrix one. There is no reason to foretell generally which approaches ((19) or (7)) approximates reality better, while numerical results (as we shall see) indicate that both approaches yield results close to each other in many cases. In the mean time, the second one (that of (7)) appears simpler. As a first example we consider again the two-phase mixtures of Fig. 1. The figure has presented, among others, the graphics of the sphere-1-platelet-2 curve, which is the solution of the respective Eq. (7)

$$1 - v_1 = \left(\frac{k_2}{k_e}\right)^{1/3} \frac{k_1 - k_e}{k_1 - k_2}, \tag{20}$$

and is near the Hashin–Shtrikman upper bound. Also given in the figure is the platelet-1-sphere-2 mixture curve, which is near the Hashin–Shtrikman lower bound. On the other hand, the solution k_e of the Eq. (19) for the sphere-1-matrix-2 mixture (spherical inclusions from phase 1 dispersed in the matrix from phase 2) is

$$v_1 = \left[1 - 9\frac{(k_1 - k_e)k_e k_2}{(k_2 - k_e)(k_e + 2k_2)(k_1 + 2k_e)}\right]^{-1}, \tag{21}$$

which is close to the sphere-1-platelet-2 curve given in (20) over a range of parameters. The sphere-2-matrix-1 curve is close to that of the platelet-1-sphere-2 mixture, as can be seen in Fig. 1.

Direct application of the weighted effective medium approximation (7) to the water-saturated porous rocks as given in Section 3 also yields the results close to those from the differential matrix model (19) given in [13,15].

Next, we apply the weighted effective medium approximation (7) as well as the differential matrix one (19) to some composite mixtures made from RTV-60 silicone rubber as the continuous phase and aluminum, lead, nickel, and bismuth as the discontinuous phases, experimental measurements on the thermal conductivity of which are reported in [23]. The thermal conductivity of the silicone rubber is 0.384, while those of aluminum, lead, nickel, and bismuth are 204.14, 34.6, 89.96, and 8.04, respectively (all in W m^{-1} K^{-1}). The results of measurements as well as calculations from the model (20) and (21) for some two-phase mixtures (phase 2 is the silicone rubber matrix, phase 1 is composed of spherical inclusions from the respective metal) in some volume proportions of the phases are compared in Table 1: k_{sp} is the sphere-1-platelet-2 model (20), k_{sm} is the sphere-1-matrix-2 model (21), k_{exp} is the experimental value. Hashin–Shtrikman upper (k^{u}_{HS}) and lower (k^{l}_{HS}) bounds from (1) to (3) are also included in the Table. We see that both approximations are close to the experimental values.

Further, consider three-phase mixtures with spherical inclusions from two metals phases (phases 1 and 2) dispersed in the silicone rubber matrix (phase 3). The weighted effective medium approximation (7) applied to the mixture (sphere-1-sphere-2-platelet-3, $k_e = k_{ssp}$) is

$$v_1(k_1 - k_e)\frac{9}{k_1 + 2k_e} + v_2(k_2 - k_e)\frac{9}{k_2 + 2k_e} + v_3(k_3 - k_e)\left(\frac{1}{k_3} + \frac{2}{k_e}\right) = 0. \tag{22}$$

The differential scheme (19) applied to the model (sphere-1-sphere-2-matrix-3) $k_e = k_{ssm}$ has particular form

Table 1
Approximations and experimental values for some two-phase matrix mixtures: k_{exp} – experimental value; k^{u}_{HS}, k^{l}_{HS} – Hashin–Shtrikman bounds; k_{sp} – sphere-1-platelet-2 approximation; k_{sm} – sphere-1-matrix-2 approximation; k_1 and v_1 are the thermal conductivity and volume fraction of the dispersed phase; the conductivity of the matrix phase is $k_2 = 0.384$ (all in W m^{-1} K^{-1})

k_1	v_1	k^{u}_{HS}	k_{sp}	k_{exp}	k_{sm}	k^{l}_{HS}
204.14	0.05	7.297	0.449	0.474	0.447	0.444
	0.16	23.36	0.692	0.765	0.645	0.602
	0.28	42.36	1.185	1.038	1.018	0.829
34.6	0.05	1.550	0.448	0.462	0.445	0.443
	0.16	4.258	0.674	0.663	0.634	0.595
	0.24	6.362	0.937	0.860	0.840	0.732
89.96	0.05	3.427	0.450	0.458	0.446	0.444
	0.16	10.50	0.687	0.656	0.641	0.600
	0.24	15.99	0.975	0.815	0.860	0.742
8.04	0.05	0.650	0.440	0.433	0.438	0.436
	0.16	1.265	0.619	0.590	0.596	0.570
	0.24	1.742	0.802	0.732	0.754	0.688

Table 2
Approximations and experimental values for some three-phase matrix mixtures: k_{exp} – experimental value; $k^{\mathrm{u}}_{\mathrm{HS}}, k^{\mathrm{l}}_{\mathrm{HS}}$ – Hashin–Shtrikman bounds; k_{ssp} – sphere-1-sphere-2-platelet-3 approximation; k_{ssm} – sphere-1-sphere-2-matrix-3 approximation; k_1, k_2 and v_1, v_2 are the thermal conductivities and volume fractions of the dispersed phases; the conductivity of the matrix phase is $k_3 = 0.384$ (all in W m^{-1} K^{-1})

k_1	k_2	v_1	v_2	$k^{\mathrm{u}}_{\mathrm{HS}}$	k_{ssp}	k_{exp}	k_{ssm}	$k^{\mathrm{l}}_{\mathrm{HS}}$
204.14	89.96	0.06	0.04	11.79	0.541	0.559	0.525	0.511
		0.13	0.09	26.21	0.899	0.804	0.802	0.705
204.14	8.04	0.04	0.08	6.514	0.558	0.637	0.541	0.525
		0.15	0.03	22.10	0.783	0.770	0.682	0.629
8.04	34.6	0.04	0.08	1.885	0.555	0.592	0.539	0.524
		0.10	0.08	3.351	0.698	0.692	0.657	0.614

$$\frac{dk}{dt} = \frac{k}{1 - v_{\mathrm{I}}t}\left(3v_1\frac{k_1 - k}{k_1 + 2k} + 3v_2\frac{k_2 - k}{k_2 + 2k}\right),$$
$$v_{\mathrm{I}} = v_1 + v_2, \quad k(0) = k_3, \quad 0 \leqslant t \leqslant 1, \tag{23}$$

and $k(1) = k_{\mathrm{e}}$. The solutions $k_{\mathrm{e}} = k_{\mathrm{ssp}}$ of (22), and $k_{\mathrm{e}} = k_{\mathrm{ssm}}$ of (23) are reported in Table 2 together with experimental data k_{exp}, and Hashin–Shtrikman bounds ($k^{\mathrm{u}}_{\mathrm{HS}}, k^{\mathrm{l}}_{\mathrm{HS}}$). Both approximation are close to the experimental values.

The simple weighted effective medium approximation (7) as well as the more general differential matrix model (19), by construction, is always realizable, at least, by certain hierarchical models. Hence, both approximations never violate strict mathematical requirements for an effective property, including the bounds. The schemes are flexible enough that they could include not only the properties and volume fractions of the components, but also the range of shapes for the inhomogeneities. They can also take into account the continuity of the matrix phase in a matrix composite, by starting the differential scheme (19) from the matrix phase, or treating it as a disordered network of extreme platelet inhomogeneities in (7). Still, the schemes are just simple convenient engineering approximations, which could not incorporate specific information about arrangements of the inhomogeneities in the material space. However, such arrangements for many practical random composites are often irregular and can hardly be given through some simple mathematical description to be used in engineering applications. The schemes cannot incorporate also the relative size distributions of the inhomogeneities, except for the trivial case: the inclusions are widely separated in sizes. In that case, the set of those inhomogeneities of smaller size (together with the matrix) should be treated firstly as an effective medium before the next construction, which involves bigger inclusions, according to the differential scheme, as well as common sense.

5. Conclusion

To determine theoretically the macroscopic conductivity of a randomly inhomogeneous continuum, firstly one should find the respective fields within the constituent inhomogeneities. To calculate the fields within every separated inhomogeneity from a completely random mixture, as often in many cases, we have to consider the inhomogeneity as embedded in the effective medium. The effective medium approximation is started from this dilute solution result. The weighted effective medium approximation could incorporate the distribution of various shapes of inhomogeneities with respective weights, hence might be flexible enough to approximate practical mixtures. The simple weighted spheroidal approximation, which includes all the extreme three-dimensional spherical, two-dimensional platelet, and one-dimensional needle shapes, appears especially useful. The weighted effective medium approximation is not only simple for application, but also has the mathematical justification that the scheme corresponds, at least, to some hierarchical model, therefore it never violates exact mathematical relations imposed upon the effective properties including the bounds.

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Appendix A

The differential scheme construction process for Eqs. (4)–(7), and more generally Eq. (19), starts with the base matrix phase $N+1$. At each step of the procedure, we add proportionally infinitesimal volume amounts $v_{i\alpha_i}\Delta t$ ($\Delta t \ll 1,\ \alpha_i = 1, \ldots, N_i,\ i = 1, \ldots, N$) of randomly oriented inclusions into already constructed composite of the previous step, which contains volume fractions $v_{i\alpha_i}t$ of the inclusion phases (the parameter t increases from 0 to 1, as the differential scheme proceeds). The newly added particles will see an effective continuum, owing to their relative sizes, and the new composite can be considered as a dilute suspension of particles from phases α_i, of volume fractions

$$\frac{v_{i\alpha_i}\Delta t}{1 + \sum_{i=1}^{N}\sum_{\alpha_i=1}^{N_i} v_{i\alpha_i}\Delta t} = \frac{v_{i\alpha_i}\Delta t}{1 + v_{\mathrm{I}}\Delta t}$$

in a matrix of conductivity k (v_{I} is the total volume fractions of the included phases). The effective conductivity of the new composite is

$$k + \mathrm{d}k = k + k \sum_{i=1}^{N} \sum_{\alpha_i=1}^{N_i} \frac{v_{i\alpha_i} \Delta t}{1 + v_{\mathrm{I}} \Delta t} D_{i\alpha_i}(k_i, k).$$

Since the volume fraction of the included phase α_i increases by

$$v_{i\alpha_i} \mathrm{d}t = \frac{v_{i\alpha_i} t + v_{i\alpha_i} \Delta t}{1 + v_{\mathrm{I}} \Delta t} - v_{i\alpha_i} t = \frac{v_{i\alpha_i} \Delta t}{1 + v_{i\alpha_i} \Delta t} (1 - v_{\mathrm{I}} t),$$

we obtain the following differential equations for the effective conductivity of the composite (Eq. (19))

$$\frac{\mathrm{d}k}{\mathrm{d}t} = \frac{k}{1 - v_{\mathrm{I}} t} \sum_{i=1}^{N} \sum_{\alpha_i=1}^{N_i} v_{i\alpha_i} D_{i\alpha_i}(k_i, k),$$
$$k(0) = k_{N+1}, \quad 0 \leqslant t \leqslant 1.$$

If in the equation above, we eliminate the matrix phase $N+1$, then $v_{\mathrm{I}} = 1$, and at the end of the process $t \to 1$, the multiplier $k/(1 - v_{\mathrm{I}} t) = k/(1-t) \to \infty$, hence the sum

$$\sum_{i=1}^{N} \sum_{\alpha_i=1}^{N_i} v_{i\alpha_i} D_{i\alpha_i}(k_i, k) \to 0,$$

as k should be finite. Thus we obtain the effective medium equations (5)–(7) as specific cases.

Appendix B

Eq. (15) can also be rewritten as

$$\sum_{i=1}^{N} k_i \sum_{\alpha_i=1}^{N_i} v_{i\alpha_i} \left[\frac{1}{k_i A_{i\alpha_i} + k_{\mathrm{e}}(1 - A_{i\alpha_i})} + \frac{1}{k_i B_{i\alpha_i} + k_{\mathrm{e}}(1 - B_{i\alpha_i})} + \frac{1}{k_i C_{i\alpha_i} + k_{\mathrm{e}}(1 - C_{i\alpha_i})} \right]$$
$$= k_{\mathrm{e}} \sum_{i=1}^{N} \sum_{\alpha_i=1}^{N_i} v_{i\alpha_i} \left[\frac{1}{k_i A_{i\alpha_i} + k_{\mathrm{e}}(1 - A_{i\alpha_i})} + \frac{1}{k_i B_{i\alpha_i} + k_{\mathrm{e}}(1 - B_{i\alpha_i})} + \frac{1}{k_i C_{i\alpha_i} + k_{\mathrm{e}}(1 - C_{i\alpha_i})} \right].$$

As $0 \leqslant A_{i\alpha_i}, B_{i\alpha_i}, C_{i\alpha_i} \leqslant 1$, the left-hand-side expression of the above equation is a decreasing function of the positive variable k_{e}, while the right-hand-side one is an increasing function, hence the equation does not have more than one solution. On the other hand, the left-hand-side of (15) is positive if $k_{\mathrm{e}} \leqslant k_{\min} = \min\{k_i,\ i = 1, \ldots, N\}$, and negative if $k_{\mathrm{e}} \geqslant k_{\max} = \max\{k_i,\ i = 1, \ldots, N\}$, hence the unique solution k_{e} of (15), indeed, exists within the interval $[k_{\min}, k_{\max}]$.

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