On the Electrical Conductivity of Particulate Composites

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ABSTRACT: Two new equations are developed for effective electrical conductivity of concentrated particulate composites using a differential scheme along with the solution of an infinitely dilute dispersion of particles in a continuous matrix. The proposed equations are evaluated using 16 sets of experimental data on the electrical conductivity of two-phase particulate systems. The following model developed in the paper describes the experimental data very well: $(\sigma/\sigma_m)^{1/3}(\sigma_d - \sigma_m)/(\sigma_d - \sigma) = (1 - \phi/\phi_m)^{-\alpha\phi_m}$ where σ, σ_m and σ_d are electrical conductivities of composite, matrix, and dispersed phase (filler) respectively, ϕ is volume fraction of filler, ϕ_m is the maximum packing volume fraction of filler, and α is a constant of the order of unity. In the special case of $\alpha = 1$ and $\phi_m = 1$, this model reduces to the well-known Bruggeman equation for the electrical conductivity of two-phase particulate systems. The predictions of the proposed model are significantly different from the predictions of the existing general effective media (GEM) model.

KEY WORDS: composite, two-phase, electrical conductivity, properties.

INTRODUCTION

PARTICULATE COMPOSITES CONSISTING of nearly isometric (same dimensions in all directions) particles dispersed in a continuous matrix phase are widely used in industrial applications. To make efficient use of composite materials the variations of physical properties such as electrical conductivity with the kind and concentration of filler particles should be known. For example, electrically non-conducting particles are often added to a metal matrix to enhance the mechanical properties. But the addition of non-conducting particles can decrease the electrical conductivity of the metal by a significant amount. From a practical point of view, it is important to be able to predict this decrease in electrical conductivity with the increase in volume fraction of insulating filler. Likewise, it is important to be able to predict the increase in electrical conductivity of an insulating matrix with the increase in electrically conducting filler content; many practical

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applications in electronics and electrical industries require electrically conductive polymer composites (composites of non-conducting polymer matrix and electrically conducting filler particles).

Over the past several decades, a number of experimental and theoretical studies have been published on the electrical properties of particulate composites [1–21]. However, no single relationship is yet found to describe the experimental electrical conductivity data of a wide variety of particulate composite systems.

BRIEF BACKGROUND

For an infinitely dilute composite of spherical filler particles, the exact expression for the effective electrical conductivity is given by:

$$\frac{\sigma}{\sigma_m} = 1 + 3\left(\frac{\sigma_d - \sigma_m}{\sigma_d + 2\sigma_m}\right)\phi \tag{1}$$

where σ, σ_m, and σ_d are electrical conductivities of composite, matrix, and filler (dispersed-phase), respectively, and ϕ is the volume fraction of filler. Equation (1) was originally derived by Maxwell [1] and is referred to as the Maxwell equation. The Maxwell equation can be re-written as:

$$\sigma_r = 1 + 3\left(\frac{\lambda - 1}{\lambda + 2}\right)\phi \tag{2}$$

where σ_r is the relative electrical conductivity defined as σ/σ_m and λ is the electrical conductivity ratio defined as σ_d/σ_m.

For composites with electrically non-conducting filler particles and conducting matrix, $\lambda \to 0$ and the Maxwell equation reduces to:

$$\sigma_r = 1 - \frac{5}{2}\phi. \tag{3}$$

In the case of composites with conductive filler particles and non-conductive matrix, $\lambda \to \infty$ and the Maxwell equation becomes:

$$\sigma_r = 1 + 3\phi. \tag{4}$$

Wagnar [2] extended the Maxwell model [Equation (1)] to higher filler concentrations. The Maxwell–Wagnar equation (also known as Wiener's rule [3]) is given by:

$$\left(\frac{\sigma - \sigma_m}{\sigma + 2\sigma_m}\right) = \left(\frac{\sigma_d - \sigma_m}{\sigma_d + 2\sigma_m}\right)\phi. \tag{5}$$

Another expression which is widely discussed in the literature is that of Bruggeman [4]:

$$\left(\frac{\sigma_d - \sigma}{\sigma_d - \sigma_m}\right)\left(\frac{\sigma_m}{\sigma}\right)^{1/3} = 1 - \phi. \tag{6}$$

When $\lambda \to 0$, the Bruggeman equation reduces to:

$$\sigma_r = \frac{\sigma}{\sigma_m} = (1 - \phi)^{3/2}. \tag{7}$$

For situations where $\lambda \to \infty$, the Bruggeman equation gives:

$$\sigma_r = (1 - \phi)^{-3}. \tag{8}$$

Equations for the electrical conductivity of particulate composites have been proposed by other authors as well [5–10]. Unfortunately the equations proposed by these authors have not found widespread applicability.

One serious drawback of the existing equations is that they fail to predict the right behavior when $\phi \to \phi_m$ where ϕ_m is the maximum packing volume fraction of particles where the particles touch each other. When $\lambda(=\sigma_d/\sigma_m) \to \infty$, the electrical conductivity of a composite is expected to approach infinity at $\phi \to \phi_m$. For random close packing of uniform spheres, ϕ_m is 0.637. Furthermore, no single existing equation is found to adequately describe the available experimental data on the electrical conductivity of particulate composites.

In this paper, new models are developed for the electrical conductivity of particulate composites. The models are evaluated using 16 sets of experimental electrical conductivity data on particulate composites.

NEW MODELS FOR THE ELECTRICAL CONDUCTIVITY OF PARTICULATE COMPOSITES

For infinitely dilute composites of filler particles, the Maxwell expression for electrical conductivity [Equation (2)] can be generalized to:

$$\frac{\sigma}{\sigma_m} = 1 + 3\alpha\left(\frac{\sigma_d - \sigma_m}{\sigma_d + 2\sigma_m}\right)\phi \tag{9}$$

where α is a 'correction-factor' of the order unity to account for the deviations from the assumptions made by Maxwell in the derivation of Equation (1). For example, most particulate composites of practical interest consist of non-spherical irregular-shaped fillers. Even the equiaxed particle fillers used in particulate composites are non-spherical. Furthermore, more often than not, some interfacial additive is present at the interface of particle-matrix.

Equation (9) cannot be applied at finite concentration of particles as the interaction between the particles is not considered in its derivation. To extend the applicability of Equation (9) to concentrated systems, a differential effective medium approach is used.

According to this approach, a concentrated composite is considered to be obtained from an initial matrix phase by successively adding infinitesimally small quantities of particles to the system until the final volume fraction of filler is reached. The incremental change in electrical conductivity upon the addition of an infinitesimally small quantity of particles to the system can be determined from the generalized Maxwell equation [Equation (9)]. Therefore,

$$d\sigma = 3\alpha\sigma\left[\frac{\sigma_d - \sigma}{\sigma_d + 2\sigma}\right]d\phi. \tag{10}$$

This equation can be re-written as:

$$\frac{1}{3\alpha}\left[\frac{1}{\sigma} + \frac{3}{\sigma_d - \sigma}\right]d\sigma = d\phi. \tag{11}$$

Upon integration with the limit $\sigma \to \sigma_m$ at $\phi \to 0$, Equation (11) gives

$$\left[\frac{\sigma}{\sigma_m}\right]^{1/3}\left(\frac{\sigma_d - \sigma_m}{\sigma_d - \sigma}\right) = \exp(\alpha\phi) \tag{12a}$$

or:

$$\sigma_r^{1/3}\left(\frac{\lambda - 1}{\lambda - \sigma_r}\right) = \exp(\alpha\phi). \tag{12b}$$

Equation (12) (a or b), referred to as Model 1 in the remainder of the paper, is expected to describe the electrical conductivity of particulate composites at low to moderate values of ϕ (volume fraction of particles). This is because in the derivation of the differential equation [Equation (10)] leading to Model 1 [Equation (12)], it is assumed that all the volume of the existing composite, before a differential quantity of new particles are added to the existing composite, is available as free volume to the new particles. In reality, the free volume available to disperse the new particles is significantly less, due to the volume pre-empted by the particles already present. This means that when a differential quantity of new particles are added to the existing composite the increase in the actual volume fraction of the dispersed phase is larger than $d\phi$. The increase in the volume fraction of the dispersed phase is $d\phi/(1 - k_0\phi)$ where k_0 accounts for the so-called 'crowding effect' caused by packing difficulties of particles. k_0 is equal to $1/\phi_m$, where ϕ_m is the maximum packing volume fraction of particles. A similar argument was made by Krieger and Dougherty [22] in the derivation of their celebrated equation for the viscosity of concentrated suspensions. Thus, Equation (11) should be re-written as:

$$\frac{1}{3\alpha}\left[\frac{1}{\sigma} + \frac{3}{\sigma_d - \sigma}\right]d\sigma = \frac{d\phi}{1 - (\phi/\phi_m)}. \tag{13}$$

Upon integration with the limit $\sigma \to \sigma_m$ at $\phi \to \phi_m$, Equation (13) gives:

$$\left(\frac{\sigma}{\sigma_m}\right)^{1/3}\left(\frac{\sigma_d - \sigma_m}{\sigma_d - \sigma}\right) = \left(1 - \frac{\phi}{\phi_m}\right)^{-\alpha\phi_m} \tag{14a}$$

or:

$$(\sigma_r)^{1/3}\left(\frac{\lambda-1}{\lambda-\sigma_r}\right)=\left(1-\frac{\phi}{\phi_m}\right)^{-\alpha\phi_m}. \tag{14b}$$

Equation (14) (a or b) is referred to as Model 2 in the remainder of the paper. For conducting particles dispersed in non-conducting matrix such that $\lambda\to\infty$, Model 2 gives:

$$\sigma_r=\left(1-\frac{\phi}{\phi_m}\right)^{-3\alpha\phi_m}. \tag{15}$$

For insulating particles dispersed in conducting matrix such that $\lambda\to 0$, Model 2 reduces to:

$$\sigma_r=\left(1-\frac{\phi}{\phi_m}\right)^{3\alpha\phi_m/2}. \tag{16}$$

It should be noted that the Bruggeman equation [Equation (6)] is a special case of Model 2 when $\alpha=1$ and $\phi_m=1$. Interestingly, Model 2 predicts the relative electrical conductivity of particulate composite to be infinite when $\lambda\to\infty$ and $\phi\to\phi_m$, as expected.

PREDICTIONS OF NEW MODELS

Figure 1 compares relative electrical conductivity (σ/σ_m) predicted from different models. In the figure, 'M' refers to Maxwell model [Equation (1)], 'M–W' refers to Maxwell–Wagnar model [Equation (5)], '1' refers to Model 1 [Equation (12)], and '2' refers to Model 2 [Equation (14)]. For Model 2 calculations, ϕ_m is taken to be 0.637 corresponding to random close packing of uniform spheres. When the conductivity ratio λ is 10, σ/σ_m is greater than unity for all values of ϕ (volume fraction of particles); at low values of ϕ ($\phi<0.2$), different models predict nearly the same value of σ/σ_m whereas at high values of λ, the predictions are quite different; at high values of ϕ, the predictions of σ/σ_m from different models are in the following order: Model 2 > Maxwell–Wagnar (M–W) model > Model 1 > Maxwell (M) model. When the conductivity ratio λ is 0, σ/σ_m is less than unity for all values of ϕ; at low values of $\phi(\phi<0.15)$, different models predict nearly the same value of σ/σ_m whereas at high values of ϕ, the predictions from different models are different, with the following order: Model 1 > Maxwell–Wagnar (M–W) model > Maxwell (M) model > Model 2.

Figure 2 shows the effect of filler content ϕ on the relative electrical conductivity of particulate composites. The plots are generated from Model 2 [Equation (14)] with $\phi_m=0.637$ and $\alpha=1.0$. When the electrical conductivity ratio is unity, the relative conductivity σ/σ_m is unity for all values of ϕ. For $\lambda<1.0$, σ/σ_m decreases with the increase in ϕ. For $\lambda>1.0$, σ/σ_m increases with the increase in ϕ.

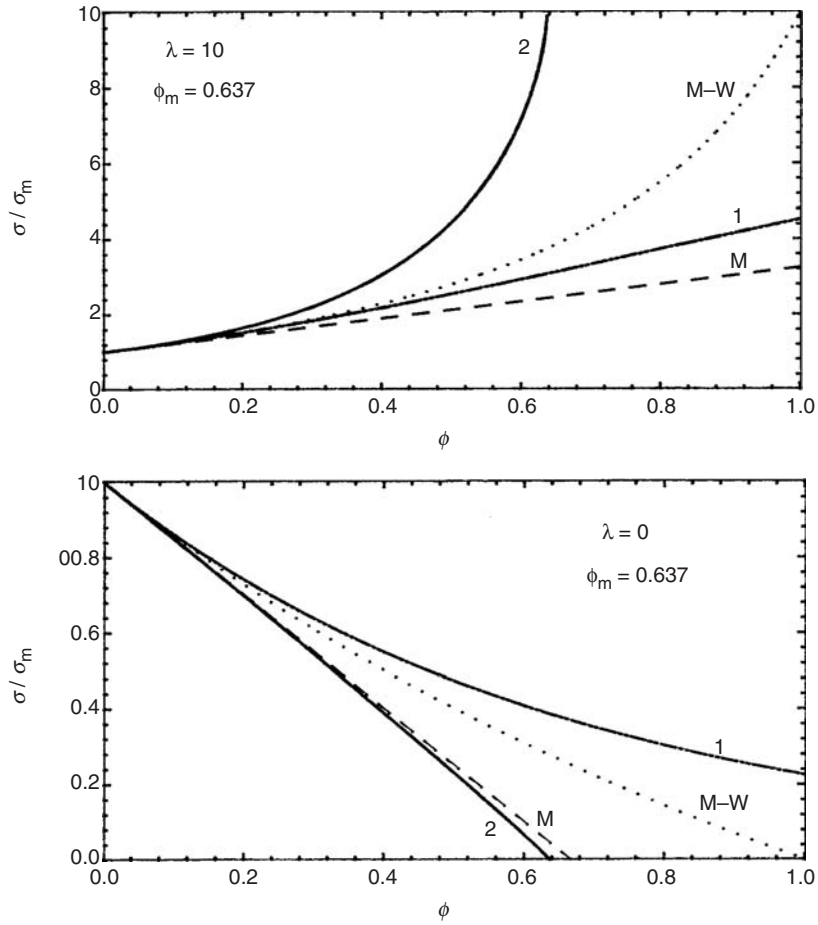

Figure 1. *Comparison of relative electrical conductivity σ/σ_m predicted from different models. 'M' refers to Maxwell model [Equation (1)], 'M–W' refers to Maxwell–Wagnar model [Equation (5)], '1' refers to Model 1 [Equation (12)], '2' refers to Model 2 [Equation (14)]. For Models 1 and 2, α is taken to be unity. The value of ϕ_m in Model 2 is taken to be 0.637.*

COMPARISON OF NEW MODELS WITH EXPERIMENTAL DATA

Sixteen sets of experimental electrical conductivity data on particulate composites covering broad ranges of electrical conductivity ratio λ and volume fraction of filler particles ϕ are considered to evaluate the models. Table 1 gives a summary of the various particulate composites considered in the present work.

Figures 3–5 show comparisons between the experimental data and predictions of Models 1 and 2 [Equations (12) and (14), respectively]. When $\lambda \to 0$ (12 sets in total: sets 1A–1E, set 2, sets 3A–3B, sets 4A–4B, sets 7 and 8), Model 1 [Equation (12)] overpredicts the values of σ/σ_m especially when $\phi > 0.20$. When $\lambda \to \infty$ (4 sets in total: sets 5 and 6, sets 9 and 10), Model 1 underpredicts the values of σ/σ_m especially when $\phi > 0.15$. However, all the 16 sets of experimental data can be described very well by Model 2 [Equation (14)] using the ϕ_m and α values summarized in Table 2.

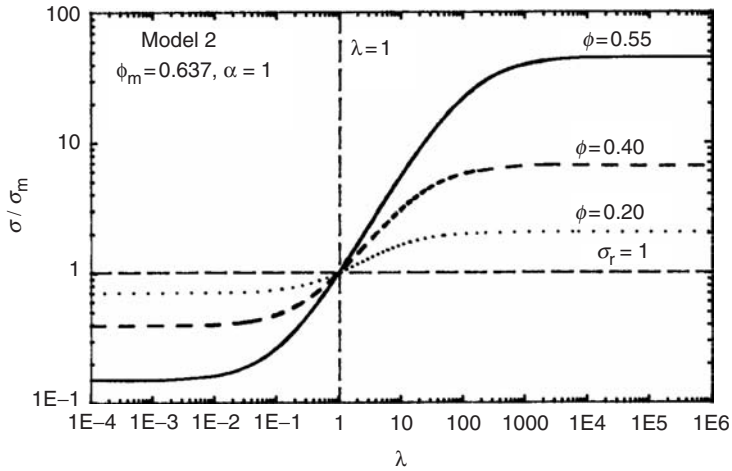

Figure 2. *Effect of volume fraction of filler particles (ϕ) on the relative electrical conductivity σ/σ_m. The plots are generated from Model 2 [Equation (14)] using $\alpha = 1$ and $\phi_m = 0.637$.*

Table 1. Summary of various particulate systems considered in the present work.

Set No.	Range of ϕ	Dispersed-phase	Matrix	λ	Ref. No.
1A–1E (5 sets)	0–0.75	Oil droplets	Water with and without surfactant and salt (NaCl)	0	11
2	0–1.0	Gas bubbles	Different aqueous solutions	0	9
3A–3B (2 sets)	0–0.86	CO_2 bubbles	Different aqueous solutions	0	20, 21
4A–4B (2 sets)	0–0.415	Glass particles, steel spheres coated with oxide film	Ordinary Portland cement	0	18
5	0–0.516	Brass spheres	Tap water	∞	5
6	0–0.415	Steel spheres	Ordinary Portland cement	∞	18
7	0–0.226	Alumina powder	Aluminum	0	16
8	0.543–0.675	Alumina particles	Aluminum	0	19
9	0–0.17	Graphite particles	Polyethylene	∞	14
10	0–0.20	Zinc powder	Nylon 6	∞	17

It is interesting to note from Table 2 that except for data sets 7 and 9–10, all other data sets can be described very well using a ϕ_m value of unity and α values close to unity in the range of 0.867–1.03. Composite systems with ϕ_m value of unity are clearly non-percolating type. Particulate composites with data sets 7 and 9–10 are percolating systems with percolation threshold values of ϕ (volume fraction of filler) equal to 0.22 and 0.18, respectively.

COMPARISON OF NEW MODEL WITH EXISTING GENERAL EFFECTIVE MEDIA (GEM) MODEL

As observed in the preceding section, model 2 (Equation (14)) describes the experimental data very well. It is worthwhile to compare the predictions

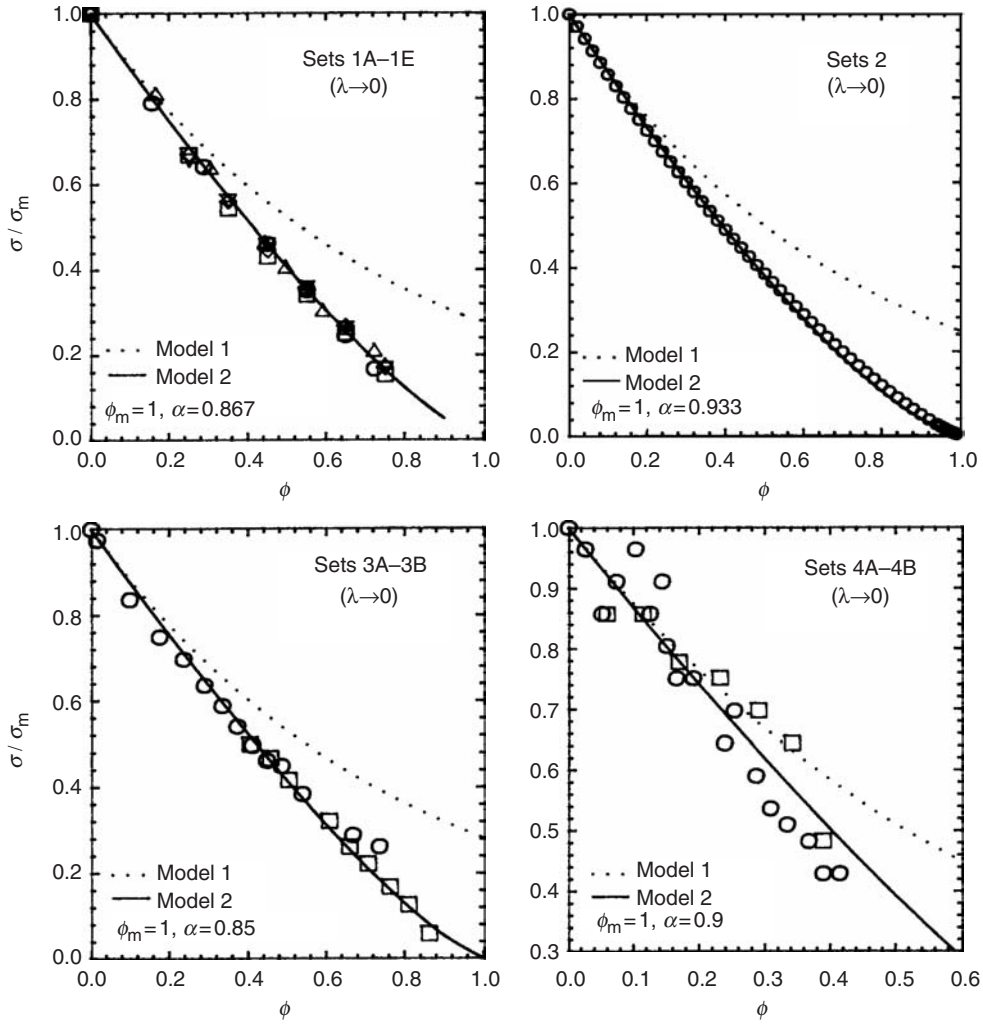

Figure 3. *Comparison between experimental σ/σ_m data of particulate composites and proposed Models 1 and 2. The values of α and ϕ_m used in the models are indicated in the figure. Note that the same value of α is used in Models 1 and 2.*

of model 2 with the existing general effective media (GEM) model of McLachlan [15,23–26]:

$$\frac{f\left[\sigma_l^{1/t}-\sigma^{1/t}\right]}{\left\{\sigma_l^{1/t}+[f_c/(1-f_c)]\sigma^{1/t}\right\}}+\frac{(1-f)\left[\sigma_h^{1/t}-\sigma^{1/t}\right]}{\left\{\sigma_h^{1/t}+[f_c/(1-f_c)]\sigma^{1/t}\right\}}=0 \tag{17}$$

where f is the volume fraction of low-conductivity component, f_c is the critical volume fraction (percolation threshold) of low-conductivity component, σ_l is the conductivity of low-conductivity component, σ_h is the conductivity of high-conductivity component, σ is the conductivity of the composite, and t is an exponent.

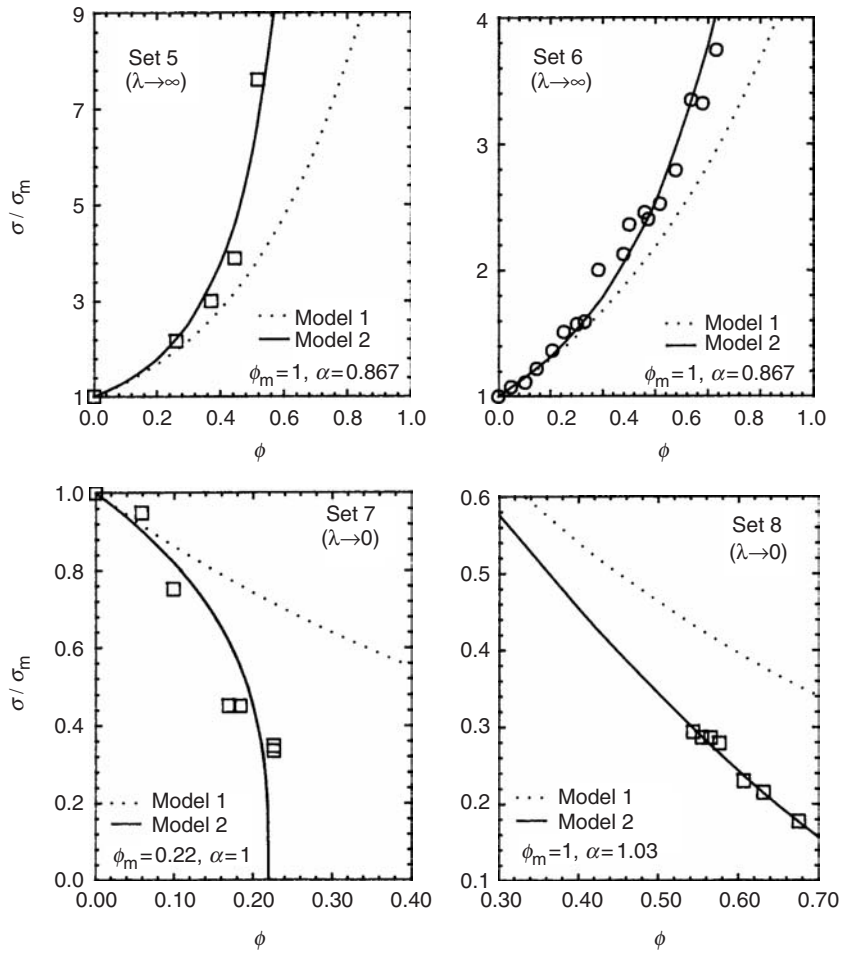

Figure 4. *Comparison between experimental σ/σ_m data of particulate composites and proposed Models 1 and 2. The values of α and ϕ_m used in the models are indicated in the figure. Note that the same value of α is used in Models 1 and 2.*

For composites of non-conducting particles and conducting matrix ($\lambda = \sigma_d/\sigma_m < 1.0$), the GEM model [Equation (17)] reduces to:

$$\frac{\phi\left[\lambda^{1/t} - \sigma_r^{1/t}\right]}{\left\{\lambda^{1/t} + [\phi_c/(1-\phi_c)]\sigma_r^{1/t}\right\}} + \frac{(1-\phi)\left[1 - \sigma_r^{1/t}\right]}{\left\{1 + [\phi_c/(1-\phi_c)]\sigma_r^{1/t}\right\}} = 0 \tag{18}$$

where ϕ is the volume fraction of the dispersed phase (non-conducting particles).

For composites of conducting particles and non-conducting matrix ($\lambda = \sigma_d/\sigma_m > 1.0$), the GEM model [Equation (17)] reduces to:

$$\frac{(1-\phi)\left[1 - \sigma_r^{1/t}\right]}{\left\{1 + [(1-\phi_c)/\phi_c]\sigma_r^{1/t}\right\}} + \frac{\phi\left[\lambda^{1/t} - \sigma_r^{1/t}\right]}{\left\{\lambda^{1/t} + [(1-\phi_c)/\phi_c]\sigma_r^{1/t}\right\}} = 0 \tag{19}$$

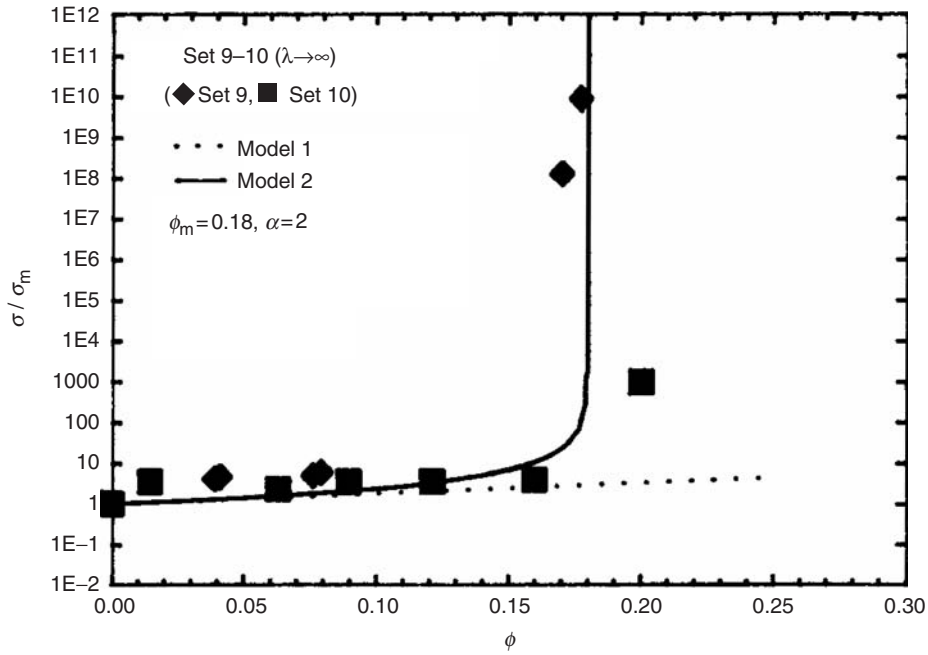

Figure 5. *Comparison between experimental σ/σ_m data of particulate composites and proposed Models 1 and 2. The values of α and ϕ_m used in the models are indicated in the figure. Note that the same value of α is used in Models 1 and 2.*

Table 2. Summary of ϕ_m and α values used in Model 2.

Set No.	ϕ_m	α
1A–1E	1	0.867
2	1	0.933
3A–3B	1	0.85
4A–4B	1	0.90
5	1	0.867
6	1	0.867
7	0.22	1
8	1	1.03
9–10	0.18	2

where ϕ is the volume fraction of the dispersed phase (conducting particles in the present case).

The GEM model [Equation (17)] in its asymptotic form reduces to the percolation expressions [15,27]:

$$\sigma_r = \left(1 - \frac{\phi}{\phi_c}\right)^{-t} \quad (\text{when } \lambda \to \infty) \tag{20}$$

$$\sigma_r = \left(1 - \frac{\phi}{\phi_c}\right)^{t} \quad (\text{when } \lambda \to 0). \tag{21}$$

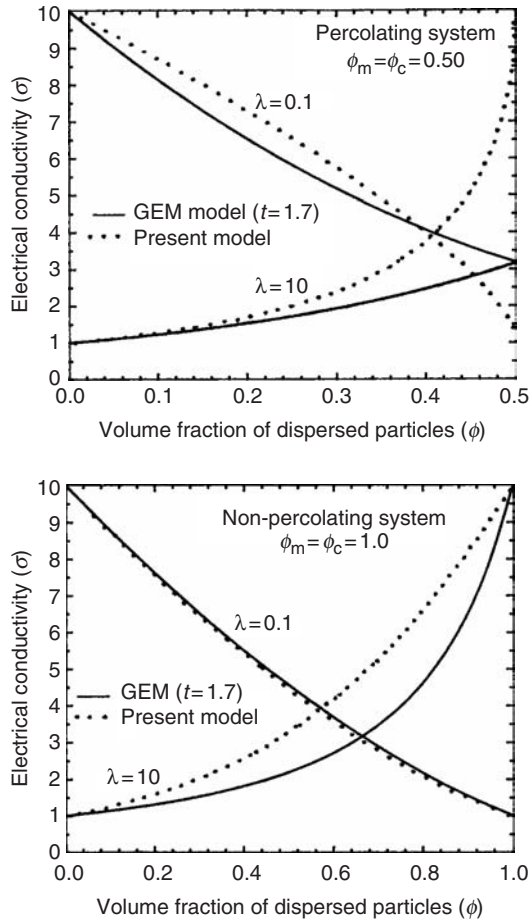

Figure 6. *Comparisons between the proposed model (Model 2) and the literature GEM model.*

The model developed in this work [Equation (14)] gives similar expressions in the limiting cases of $\lambda \to \infty$ and $\lambda \to 0$ [see Equations (15) and (16)]. However, the exponent in the proposed model is not the same in two cases of $\lambda \to \infty$ and $\lambda \to 0$. When $\lambda \to \infty$, the proposed model predicts $t = 3\alpha\,\phi_m$ and when $\lambda \to 0$, the proposed model gives $t = 1.5\,\alpha\,\phi_m$ The GEM model assumes the same value of the exponent 't' in the two cases [15,26].

For the non-percolating systems ($\phi_c = 1$), the GEM model gives the following expressions for the relative conductivity (σ_r) :

$$\sigma_r = [1 - \phi(1 - \lambda^{-1/t})]^{-t} \quad (\text{when } \lambda > 1) \tag{22}$$

$$\sigma_r = [1 - \phi(1 - \lambda^{1/t})]^{t} \quad (\text{when } \lambda < 1). \tag{23}$$

Figure 6 compares the predictions of the proposed model [Equation (14)] with the GEM model [Equation (17)] for percolating ($\phi_c = \phi_m < 1$) as well as non-percolating ($\phi_c = \phi_m = 1$) composite systems. The value of α in Equation (14) is taken to be 1.0.

The percolation-threshold concentration in the case of percolating system is taken to be 0.50 as an example. The ratio σ_h/σ_l is taken to be 10. The exponent 't' in the GEM model is taken to be 1.7; this value of 't' is considered to be the typical value [13,15]. Clearly there are significant differences between the predictions of the proposed model and the GEM model, both in percolating and non-percolating systems.

CONCLUSIONS

Two new equations for the effective electrical conductivity of concentrated particulate composites are developed. The proposed equations are evaluated using 16 sets of experimental data on the electrical conductivity of particulate composites covering broad ranges of conductivity ratio λ (ratio of dispersed phase electrical conductivity to matrix conductivity) and volume fraction of filler ϕ. All the experimental data can be described adequately by the following equation developed in the paper:

$$(\sigma/\sigma_m)^{1/3}\left(\frac{\sigma_d-\sigma_m}{\sigma_d-\sigma}\right)=\left(1-\frac{\phi}{\phi_m}\right)^{-\alpha\phi_m}$$

where σ, σ_m, and σ_d are electrical conductivities of composite, matrix and filler (dispersed-phase) respectively, ϕ is volume fraction of filler, ϕ_m is maximum packing volume fraction of filler, and α is a constant of the order of unity. This equation reduces to the well-known Bruggeman equation in the special case of $\alpha=1$ and $\phi_m=1.0$. The predictions of the proposed model are also compared with the existing GEM model; there are significant differences between the predictions of the two models.

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