Effective Medium Theories for Artificial Materials Composed of Multiple Sizes of Spherical Inclusions in a Host Continuum

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***Abstract*— This paper presents the application of nonempirical effective medium theories to describe composite mixtures of spherical inclusions within a host continuum. It is shown that the most common effective medium theories collapse into Bruggeman's asymmetric formula when they are implemented in an iterative scheme to extend their validity to higher volume fractions. Comparisons of dc and 4-GHz data show that of all the formulas Bruggeman's asymmetric formula corresponds best with experiment for large differences between the complex permittivities of the host and inclusion materials. Permeability values are also formulated and compared with experiment and a simple scheme is considered to extend the effective medium theories herein to a description of the diamagnetic effect of induced current in metal spherical inclusions.**

***Index Terms*—Effective medium, spherical inclusions, synthetic medium.**

I. Introduction

EFFECTIVE medium theories (EMT's) describe a composite mixture in terms of a spatially homogeneous electromagnetic response [1]–[18], [21]–[25]. EMT's describe mixtures in the dual limits of a static analysis and a low-volume fraction of inclusions within a host, neglecting inclusion clustering. In this paper, comparisons are made between common nonempirical EMT's with experimental data from the literature and with measurements of carbonyl-iron powder (CIP) and rubber mixtures at 4 GHz. Specifically, mixtures with multiple-sized spherical inclusions mixed in a host continuum with a process that suppresses stochastic percolation are considered.

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Existing EMT's differ in one of two ways: 1) in the way they model the averaging effects (such as the formulas of Clausius–Mossotti and Maxwell–Garnett (MG or C-M), the formula of Maxwell, the symmetric Bruggeman (S-BG) or Böttcher formula, the asymmetric Bruggeman (A-BG) formula, and the formula of Looyenga [1], [5]–[10], [12], [13]) or 2) in the way they model polarizability effects (ordered lattice numerical calculations [2], percolation theory [16]–[18], [21]–[26], and the Doyle and Jacobs effective cluster model [4]). In general, set (1) requires that the individual inclusions be far from each other (neglecting higher order multipole interactions in the low-volume fraction or the "dilute limit") while the theories of set (2) rigorously model the approach of the mixture to the limit in which the inclusions touch each other. This paper concentrates on the first classification of theories (1) above, as these tend to provide simple nonempirical formulas for the effective parameters.

The requirement of the theories of class (1) above to stay in the dilute limit is a serious hindrance to their utility. The artificial dielectrics obtained by adding spherical metal inclusions to an insulating host matrix attain useful high dielectric constants only for high-volume fractions. Even for needle like inclusions, the required volume fraction may appear to be reduced, but the absolute inclusion density may still be large enough to invalidate the dilute limit. A simple model that can explain the properties of the spherical loaded artificial dielectric over its full range of usable volume fractions with a minimum of empirical correction factors would be very useful to electromagnetic material designers.

In this paper, the most common models for mixtures of spherical inclusions in a host continuum are explored. It can be shown that the low-volume fraction limit of the most common effective medium theories can be extended iteratively to converge to one model valid for all volume fractions up to 100% (this is shown analytically for the Clausius–Mossotti formula in the Appendix). However, Bruggeman introduced a more elegant approach in his asymmetric model to which our iterative technique turns out to be equivalent [1], [14]. In this paper, it is shown that this A-BG theory gives the best fit when compared to collected historical data for the dielectric constant of solid mixtures of metal spheres in an insulating host continuum. The use of these EMT's to describe the diamagnetic effect of induced current on nonmagnetic conducting spheres is also briefly explored. All the EMT's presented in this paper, except that of Looyenga, theoretically should hold for any inclusion or host complex permittivity values.

II. Reality Factors

The models presented herein apply to the typical composite materials obtained by dispersing spherical inclusions into a host matrix. Two factors of reality must be recognized in the mathematical modeling of this material. First, the spherical inclusions are never truly monodisperse. Clusters of inclusions, multiple sizes of inclusions, and the breakup of inclusions due to grinding between inclusions during mixing all combine to give a variety of particle sizes [27]. Therefore, the particles cannot be assumed to have only one typical radius. This recognition led Doyle and Jacobs [4] to propose their effective cluster model. In their model it is recognized that in the resulting mixture there will be both isolated particles and large semi–spherical clusters of agglomerated particles, each contributing their own unique polarizability to the final composite. If the polarizability of the clusters is that of a body centered cubic (BCC) array of spheres, their model fits historical data up to volume fractions as high as 40%, much better than the classical Maxwell, Clausius–Mossotti, and MG, or the S-BG–Böttcher models.

However, the assumption that the polarizability of the clusters corresponds to that of a BCC array violates the reality of the behavior of a nonmonodisperse mixture. A randomly filled BCC array of spheres of a single size reaches a maximum filling at 63% [4]. Yet, if there are smaller spheres to fill the interstitial spaces, a nonmonodisperse mixture could be packed well beyond the 63% limit. In fact, with an appropriate distribution of particle sizes, the 100% volume fraction limit can be approached arbitrarily close [27].

The second reality factor is closely related to the last comment above. Composite materials are typically manufactured through a dynamic mixing process (for instance, three roll milling). Therefore, the inclusions cannot be assumed to be located randomly throughout the composite's volume. The mixing forces tend to break apart large clusters. For those clusters that survive, only spherical configurations are likely. These forces also absolutely prevent the early formation of stochastic percolation paths as assumed in percolation theory [16], [17], [21]–[26]. In addition, grinding between particles as the composite is mixed tends to break up the individual inclusions (at least at high-volume fractions with the degree of breakage dependent on the material) creating smaller inclusions to fill the interstitial spaces [27] and contributing to the justification for the first of our reality factors.

These two reality factors suggest that unless our inclusions can be guaranteed to be monodisperse and unless they can be forced to arrange themselves without any preferential groupings, the theoretical limit for the percolation threshold of a typically manufactured composite mixture approaches 100%.

III. Collected Effective Medium Theoreis

EMT's describe composite mixtures in terms of ϵ_{eff} and μ_{eff}, which characterize a quasi-statically equivalent material.

TABLE I
Various EMTS and Their DC Limit for Metallic Inclusions

	Formula	$\lim_{\lvert\epsilon_i\rvert\to\infty}$
Maxwell	$\epsilon_{eff} = \epsilon_h + 3p\frac{\epsilon_i-\epsilon_h}{\epsilon_i+2\epsilon_h}\epsilon_h$	$\epsilon_{eff} = \epsilon_h(1+3p)$
MG or C-M	$\epsilon_{eff} = \epsilon_h \left[\frac{1+2p\left(\frac{\epsilon_i-\epsilon_h}{\epsilon_i+2\epsilon_h}\right)}{1-p\left(\frac{\epsilon_i-\epsilon_h}{\epsilon_i+2\epsilon_h}\right)}\right]$	$\epsilon_{eff} = \epsilon_h\frac{1+2p}{1-p}$
S-BG	$\epsilon_{eff} = \frac{1}{4}\left[3p(\epsilon_i-\epsilon_h)+2\epsilon_h-\epsilon_i + \sqrt{(1-3p)^2\epsilon_i^2+2(2+9p-9p^2)\epsilon_i\epsilon_h+(3p-2)^2\epsilon_h^2}\right]$	$\epsilon_{eff} = \frac{\epsilon_h}{1-3p}$
A-BG	$\frac{\epsilon_i-\epsilon_{eff}}{\epsilon_i-\epsilon_h} = (1-p)\left(\frac{\epsilon_{eff}}{\epsilon_h}\right)^{\frac{1}{3}}$	$\epsilon_{eff} = \frac{\epsilon_h}{(1-p)^3}$
Looyenga	$\epsilon_{eff} = \left[(\epsilon_i^{\frac{1}{3}}-\epsilon_h^{\frac{1}{3}})p+\epsilon_h^{\frac{1}{3}}\right]^3$	

The static solution of the potential around one inclusion of permittivity ϵ_i utilized in the EMT's requires the assumption that the scale of the inclusions is much smaller than the electromagnetic wavelength in the host material (ϵ_h). In addition, the mixture must be homogeneous on a macroscopic scale. Any mixing must distribute the particles well, albeit in a somewhat random fashion, so that for any region of a scale on the order of the wavelength in the host, there are a constant volume fraction p of inclusions. All permittivities or permeabilities considered herein are relative complex values, even though the conventions associated with the meter kilogram second (MKS) system of units are used.

Collected in Table I are the Maxwell EMT as attributed in Lord Rayleigh's work [2] and derived in [13], MG EMT, which, for dc conductor dielectric mixtures, corresponds to the Claussius–Mossotti formulation and is also known as the Lorentz–Lorentz or Poisson's formula [4]–[9], [21], the S-BG formula [1], [3], [8], [11], [15], [16], [21], [23], which corresponds to the Böttcher formula [14], the A-BG formula [10], [14], and the Looyenga formula [10]. Also shown in Table I are the EMT's predictions in the metallic inclusion limit $|\epsilon_i| \rightarrow \infty$. The formulation of Looyenga is only valid for low contrast between inclusion ϵ_i and host ϵ_h permittivity and, thus, is not appropriate in the metallic limit. The S-BG formula is so named since it is symmetric in the sense that if the host and inclusion parameters are interchanged the resultant effective permittivity is unchanged. For a nonsymmetric morphology such as a host continuum surrounding spherical inclusions the S-BG EMT can be only expected to hold for low-volume fractions of inclusions.

These EMT's may lead to a unified model which overcomes the low-volume fraction restriction if an iterative procedure is applied as described in the Appendix. The iterative technique is illustrated in Fig. 1 and generalizes any of the EMT's beyond their low-volume fraction limit. This iterative technique, however, is inherent to the derivation of the A-BG model and like that model is appropriate specifically for discrete noncontacting inclusions within a host continuum.

Of all the theories presented, the A-BG theory appears to have the best agreement with published data for spherical

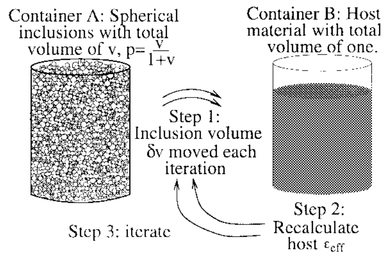

Fig. 1. The iterative technique to extend EMT's to high-volume fractions.

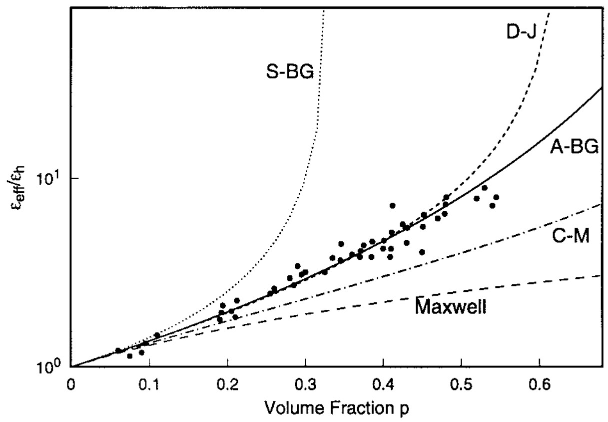

Fig. 2. Comparison of various EMT's and expermental data from Doyle and Jacobs.

inclusions within a host continuum. It extends the low-volume fraction limit of the EMT's up to a high-volume fraction validity range. In fact, since in the low-volume fraction limit all the theories give about the same results it can be shown that when they are iterated or considered in the differential limit of the iterative process, they all (except that of Looyenga) converge to the A-BG equation. The A-BG or iterative model appears to be a fundamental model for mixtures of multiple sized spheres in which mixing suppresses stochastic percolation allowing a maximum packing up to a volume fraction of one. The formula of Looyenga has been reported to give a slightly better correspondence with the published experiments for dielectric–dielectric mixtures in [9] and in [14], however, it breaks down as the magnitude of the host and inclusion permittivities become very different as a result of its somewhat arbitrary assumption that the small amount of inclusion material added has a permittivity only slightly different from the effective permittivity for that iteration.

Fig. 2 summarizes how the EMT's of Table I model the dc limit of metallic spheres in an insulating host. This figure shows a comparison between the EMT models for the metallic limit $|\epsilon_i| \rightarrow \infty$ and experimental results taken from Fig. 5 of Doyle and Jacobs [4]. The simplicity of the A-BG result for the metallic limit (Table I) is especially pleasing considering how well it matches the trend of published data. It clearly fits the data better than the other EMT expressions considered herein, and even better than the Doyle–Jacobs effective cluster model of [4] from $p = 0$ to $p = 0.6$. Since $p = 0.6$ is close to the practical limit for the filling of a host medium, that still

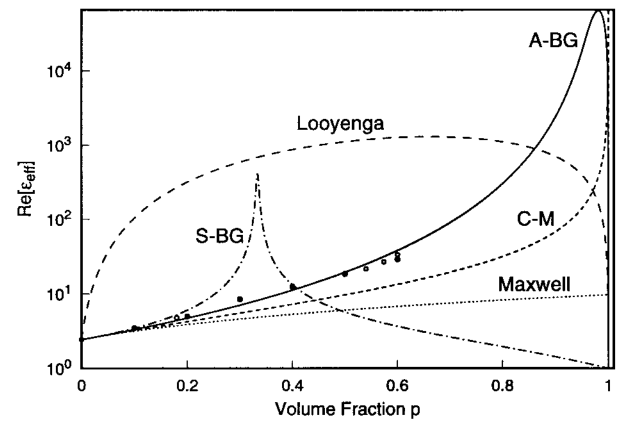

Fig. 3. $\mathrm{Re}\{\epsilon_{\text{eff}}\}$ from various EMT's at 4 GHz for $\epsilon_h = 2.43 - j0.029$ and $\epsilon_i = 1.0 - j\sigma/(\omega\epsilon_0)$ with $\sigma = 10^4$. Also shown are measurements made on mixtures of CIP in a rubber host (○ ○ ○) and measurements from the paper by Olmedo *et al.* (● ● ●).

guarantees physical integrity of these composites, we have a theory that spans their entire useful design range.

The successful comparison of the A-BG model with experiment suggests that the mathematical iteration procedure correctly models the dynamics involved in mixing particle filled composites. In fact, when the mixing mechanisms that invalidate the monodisperse assumption [27] are removed such as in the vibrating bed Aetna oil and mercury experiments of Guillen (presented in [4]), the measured data agrees with the Doyle–Jacobs [4] rather than the A-BG model. Therefore, we assume that in those cases the mixture behaves as an ordered array of equal sized spheres with a maximum packing and complete percolation at the $p = 0.63$ randomly filled BCC limit.

IV. Dispersive Properties of Particle Filled Composites

Comparisons with measurement at 4 GHz of a CIP rubber mixture's complex ϵ_{eff} and μ_{eff} and the various EMT's predictions are considered in this section.

A. Complex Permittivities

The CIP was represented with a bulk permittivity of $\epsilon_i = 1.0 - (j\sigma/\epsilon_0\omega)$ with $\sigma = 10^4\ [\Omega m]^{-1}$. The host dielectric was measured at 4 GHz as $\epsilon_h = \epsilon_h' - j\epsilon_h'' = 2.43 - j0.029$ for the rubber material used in mixtures we manufactured and measured at GEC Marconi Materials Corporation, San Diego, CA. For this measurement, ϵ_h'' fluctuated between small positive and negative values in measurements from 2 to 18 GHz, indicating that the imaginary part of the permittivity was below the measurement error threshold. Results for the real part of the effective permittivity are shown in Fig. 3 and results for the imaginary part are shown in Fig. 4. These two figures also include data taken from Fig. 3 of Olmedo *et al.* [3] in which the real part of permittivity of the host material was $\epsilon_h' \approx 2.5$ and the imaginary part was too small to read off the scale of the figure (i.e. $|\epsilon_h''| < 0.2$). To match the data in [3] ϵ_h'' was increased slightly to $\epsilon_h = 2.43 - j0.129$

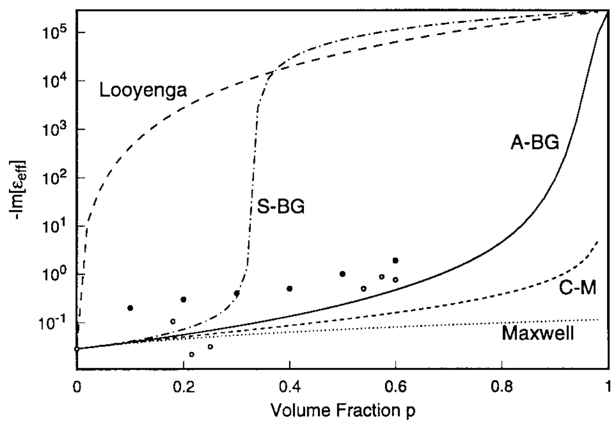

Fig. 4. $-\mathrm{Im}\{\epsilon_{\mathrm{eff}}\}$ for various EMT's at 4 GHz for $\epsilon_h = 2.43 - j0.029$ and $\epsilon_i = 1.0 - j\sigma/(\omega\epsilon_0)$ with $\sigma = 10^4$. Also shown are measurements made on mixtures of CIP in a rubber host (○ ○ ○) and measurements from the paper by Olmedo *et al.* (● ● ●).

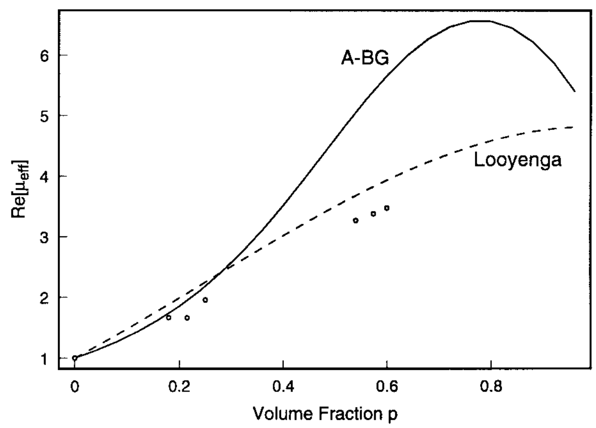

Fig. 6. $\mathrm{Re}\{\mu_{\mathrm{eff}}\}$ for the A-BG and the Looyenga formulas compared with data measured for the CIP rubber mixture at 4 GHz.

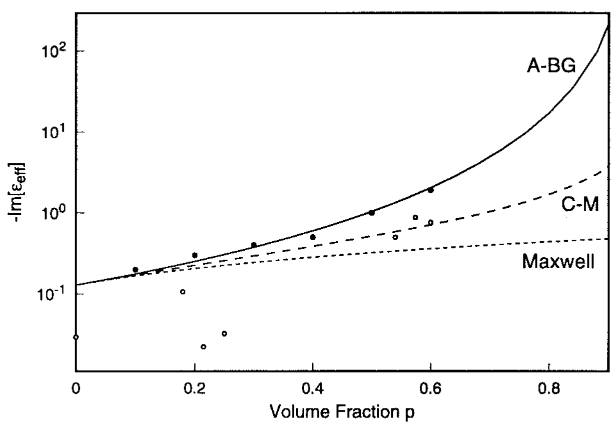

Fig. 5. $-\mathrm{Im}\{\epsilon_{\mathrm{eff}}\}$ when $\epsilon_h = 2.43 - j0.129$ compared with data from Olmedo *et al.* (● ● ●) and for the CIP rubber mixture (○ ○ ○).

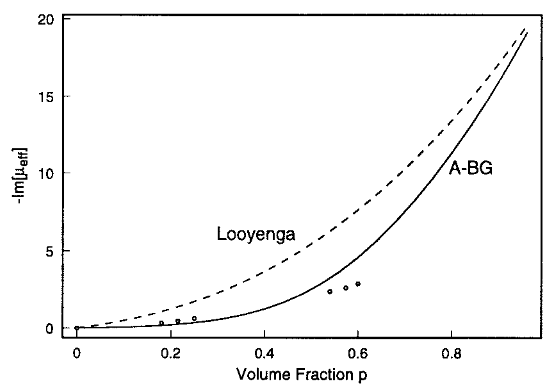

Fig. 7. $-\mathrm{Im}\{\mu_{\mathrm{eff}}\}$ for the A-BG and the Looyenga formulas compared with data measured for the CIP rubber mixture at 4 GHz.

resulting in a much better match of the imaginary part of effective permittivity shown in Fig. 5 and resulting in virtually no change in the real part of the permittivity on the scale of Fig. 3. For the CIP rubber mixture, the quasi-static limit should hold as the wavelength in the host material is $\lambda_h = c/(4\ \mathrm{GHz}\ \sqrt{\epsilon_h}) = 4.8$ cm, much larger than the inclusion size of $1 - 10$ μm. In Fig. 3, the real part of the permittivity approaches one as $p \rightarrow 1$, $(\epsilon'_{\mathrm{eff}}(p = 1) = \epsilon'_i = 1.0)$ for all the formulas except Maxwell's.

B. Complex Permeabilities

The A-BG theory was also used to compare with the permeability measured for the same CIP rubber mixtures. For the EMT predictions the host was considered nonmagnetic $\mu_h = 1.0 - 0j$, however, values were not available for the bulk properties of the carbonyl iron. As a result a single Debye relaxation was used to match the dispersive behavior predicted for Kittel's model of a single-domain iron particle [28] with a dc relative permeability of 100, a high-frequency limiting permeability of zero (due to the diamagnetic effect of conducting currents) and a single relaxation frequency of 0.9 GHz [18]. Using this single-domain iron model for the CIP, the inclusions bulk permeability was calculated to be $\mu_i = 4.819 - j21.416$ at 4 GHz. The comparison between measured and calculated effective permeability is shown in Fig. 6 for the real part and in Fig. 7 for the imaginary part versus the volume fraction of CIP.

C. Diamagnetic Effects Predicted with EMT's

Finally, the permeability predictions of the EMT's of Table I and an extension of Lord Rayleigh's analysis of arrays of metal spheres from Lam [2] are considered to describe the diamagnetic effects of nonmagnetic metal spheres in an insulating host material. The magnetic polarizability of a metal sphere (in the limit that the skin depth in the sphere goes to zero) becomes $-1/2$ that of its corresponding electric polarizability. Since the polarizabilities of spherical inclusions are proportional to volume fraction in the EMT's, the EMT formulas that incorporate the diamagnetic effect of current in metal spheres can be written by setting p to $-p/2$ for the inclusion volume fractions or, to be more precise, by setting $\epsilon_i = 0$ in any of the EMT theories and replacing the epsilons with μs. For the Maxwell, Claussius–Mossotti or MG, and A-BG formulas the diamagnetic effect yields a renormalized permeability of,

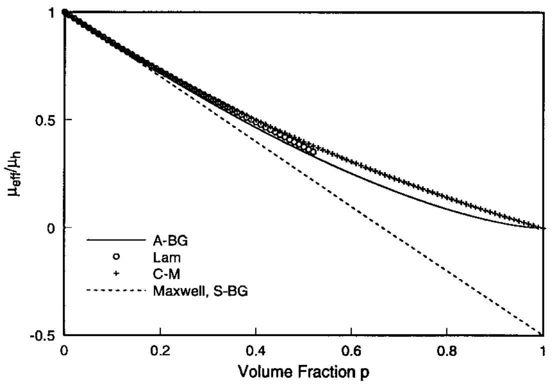

Fig. 8. The diamagnetic effect of nonmagnetic metal spheres in a nonmagnetic host dielectric. The effective permeability normalized to the host permeability is plotted versus volume fraction of metal spheres.

respectively,

$$\mu_{\text{eff-M}} = \mu_{\text{eff-S-BG}} = \mu_h\left(1 - \frac{3p}{2}\right) \tag{1}$$

$$\mu_{\text{eff-MG}} = \frac{\mu_h(1-p)}{1 + \frac{p}{2}} \tag{2}$$

$$\mu_{\text{eff-ABG}} = \mu_h(1-p)^{3/2}. \tag{3}$$

The S-BG or Böttcher formula in this diamagnetic limit that $\epsilon_i = 0$ gives the same result as the Maxwell EMT of (1). To compare these predictions, of the diamagnetic effects of metallic spheres, the formula derived in Lam [2] was considered in the same limit. In Fig. 8 a comparison is made between Lam's prediction and the EMT's of equations (1)–(3). The formula from Lam is based on an expansion in powers of p and is calculated up to terms with p^6. Since the formula of Lam was formulated for an infinite, simple cubic lattice of identical spheres, it is only valid up to $p_{\max} = \pi/3 \approx 0.5236$. As can be seen from Fig. 8 both the Claussius–Mossotti formula and the A-BG formula converge to this expected limit of complete field exclusion $(\mu_{\text{eff}} = 0)$ at $p = 1$. The A-BG formula and the Claussius–Mossotti formula both agree well with Lam's formula from [2] for $p < 0.53$. This diamagnetic case displays the versatility of the A-BG formula to again provides an accurate description of mixtures of nonpercolating spherical inclusions in a host continuum.

V. Conclusions

The A-BG formula was applied to describe a mixture of multiple sizes of metal spheres in a dielectric host, both to formulate the dc effective dielectric constant of this mixture versus volume fraction of metal inclusions and to describe the effective permittivity and permeability of a mixture of CIP and rubber at 4 GHz with a powder dispersion of sizes from 1–10 μm before mixing. For both the DC and 4 GHz cases the A-BG formula permittivity predictions, coincided qualitatively as well as quantitatively with experiment and corresponded more accurately with experiment than the other EMT's considered. Since the bulk permeability of CIP was not available, the permeability comparisons only demonstrate that this EMT results in values that are physically reasonable. When single-domain iron was used to model the inclusion the A-BG theory appeared to match the trend in the experimental data at least in describing the imaginary permeability, while the theory of Looyenga seemed to match the real permeability trend. Each of these theories predicted reasonable values, however, for a more concrete comparison intrinsic permeability values for carbonyl-iron are required. A more complete model of the CIP incorporating multiple relaxation frequencies as in [16], [19], and [20] could provide a more definitive statement about the accuracy of each EMT for the permeability. In addition, an extension of the EMT's considered was presented to describe diamagnetic effects in conductor dielectric mixtures.

Overall the A-BG formula matches well with experimental results and provides the best fit for predicting the effective permittivity of artificial dielectric mixtures of multiple sized spheres within a host material in which the percolation threshold of the spheres is suppressed. In addition most (all those presented except Looyenga's) of the other EMT's collapse into the A-BG theory when their low-volume fraction behavior is extended in an iterative approach. Only quasi-static EMT's are presented in this paper, with the only account of stochastic effects being phenomenological. Future work will consider extending the iterative technique of the A-BG EMT to describe ellipsoidal inclusions as well as consideration of this iterative extension to describe full-wave theories.

Appendix

In this Appendix an iterative technique is presented to show how the EMT of Clausius–Mossotti converges to the Bruggeman asymmetric formula when describing the dielectric constant of metal spheres in an insulating host. This iterative technique was first considered by the authors to extend the Claussius–Mossotti EMT before its correspondence with the A-BG formula was determined.

To apply the iterative technique for the Claussius–Mossotti description of metallic spheres in an insulating host, first consider a continuous host material. Then incrementally add a volume fraction δv of the inclusive material as illustrated in Fig. 1. Next, use the Clausius–Mossotti relation to calculate the effective permittivity of the mixture with volume $1 + \delta v$. This new effective permittivity then represents a homogeneous material to which another incremental volume of inclusions is added. Through this iterative procedure the Clausius–Mossotti equation's high accuracy at low-volume fractions should be extended to higher volume fractions as long as the inclusions effects can be well represented with effective parameters.

For a volume fraction p of inclusive (metal) material with total volume v considering an initial host material with volume one, then

$$p = \frac{v}{1+v}; \quad v = \frac{p}{1-p}; \quad p_n = \frac{\delta v}{1 + n\delta v} \tag{4}$$

p_n is the constant volume fraction of inclusions considered in the nth iteration.

After N iterations of the Claussius–Mossotti formula of Table I

$$\epsilon_{\rm eff} = \epsilon_h \prod_{n=1}^{N} \frac{1 + \dfrac{2\delta v}{1+n\delta v}}{1 - \dfrac{\delta v}{1+n\delta v}}. \tag{5}$$

Equation (5) can be rewritten in the limit $N \rightarrow \infty$

$$\lim_{N\to\infty} \ln(\epsilon_{\rm eff}) = \ln(\epsilon_h) + \lim_{N\to\infty} \sum_{n=1}^{N} \cdot \left[\frac{3\delta v}{1+n\delta v} - \frac{\delta v^2}{(1+n\delta v)^2} + \cdots\right]. \tag{6}$$

Each term in the sum in (6) is then considered. With $\delta v = v/N$, the first term is

$$\lim_{N\to\infty} \frac{v}{N} \sum_{n=1}^{N} \frac{1}{1+\dfrac{nv}{N}} = \ln(1+v). \tag{7}$$

The higher order terms in the sum of (6) all converge to zero in their summation under the limit that $N \rightarrow \infty$ as can be seen by examining the second term

$$\lim_{N\to\infty} \sum_{n=1}^{N} \frac{9\delta v^2}{2(1+n\delta v)^2} = \left(1 - \left(\frac{2v}{2} + \frac{v^2}{3}\right) + \cdots\right) \lim_{N\to\infty} \frac{v^2}{N} = 0. \tag{8}$$

The same limit holds for the sums of all the higher order terms in (6). As a result, the effective permittivity can be written as the following for the A-BG theory in Table I:

$$\epsilon_{\rm eff} = \epsilon_h (1+v)^3 = \frac{\epsilon_h}{(1-p)^3}. \tag{9}$$

Any of the EMT's except that of Looyenga can be evaluated numerically for complex inclusion and host permittivities in this iterative procedure to give the same result as the A-BG EMT. This was done numerically for 5000 and 100 000 iterations; all of the theories (except that of Looyenga) converged to the A-BG EMT, the theory of Looyenga converged to itself as a result of its own iterative derivation.

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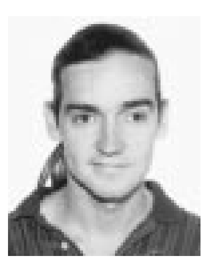

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