Journal of Magnetism and Magnetic Materials 324 (2012) 1063–1066

Contents lists available at SciVerse ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

A mixing rule for predicting frequency dependence of material parameters in magnetic composites

Konstantin N. Rozanov [a,*], Marina Y. Koledintseva [b], James L. Drewniak [b]

[a] Institute for Theoretical and Applied Electromagnetics of Russian Academy of Sciences, Moscow, Russia
[b] Center for Electromagnetic Compatibility, Missouri University of Science and Technology, Rolla, MO 65401, USA

ARTICLE INFO

Article history:
Received 28 July 2011
Received in revised form
13 October 2011
Available online 20 October 2011

Keywords:
Composite material
Microwave magnetic
Mixing rule
Permeability

ABSTRACT

A number of mixing rules are proposed in the literature to predict the dependence of effective material parameters (permittivity and permeability) of composites on frequency and concentration. However, the existing mixing rules for frequency dependence of permeability in magnetic composites typically do not provide satisfactory agreement with measured data. Herein, a simple mixing rule is proposed. Its derivation is based on the Bergman–Milton spectral theory. Both the Bruggeman effective medium theory and the Maxwell Garnett approximation are included as particular cases of the proposed mixing rule. The derived mixing rule is shown to predict accurately the frequency dependence of permeability in magnetic composites, which contain nearly spherical inclusions.

1. Introduction

Many composite magneto-dielectric materials have been developed recently for various applications in radio frequency and microwave electronic devices [1–7]. Prior to engineering composites with desirable electromagnetic properties, it is important to predict wideband frequency responses of their effective material parameters, specifically, permittivity ε_e and permeability μ_e, as functions of the concentration p, permittivity ε_i and permeability μ_i of inclusions, and permittivity ε_h and permeability μ_h of the host matrix.

The effective permittivity and permeability of a composite are conventionally assumed to be governed by the same mixing rule. A number of mixing rules have been proposed in literature, e.g., [8]. The most commonly used mixing rules are the Maxwell Garnett approximation (MGA)

$$\frac{\beta-1}{1+n_0(\beta-1)}=p\frac{\alpha-1}{1+n_0(\alpha-1)}, \tag{1}$$

and the Bruggeman effective medium theory (EMT)

$$p\frac{\alpha-\beta}{\beta+1+n_0(\alpha-\beta)}+(1-p)\frac{-\beta}{\beta+1-n_0\beta}=0. \tag{2}$$

In Eqs. (1) and (2), the parameters $\alpha=\varepsilon_i/\varepsilon_h-1$ or μ_i/μ_h-1 and $\beta=\varepsilon_e/\varepsilon_h-1$ or μ_e/μ_h-1, are the normalized dielectric and magnetic susceptibilities of inclusions and composite, respectively. The parameter n_0 is the form factor (depolarization or demagnetization factor), the same for both the permittivity and permeability of a particular composite. Eqs. (1) and (2) are formulated rigorously for spherical inclusions with $n_0=1/3$, but they are frequently applied to non-spherical inclusions with an effective (average) form factor n_0 differing from 1/3 [9–11]. Mixing rules (1) and (2) are the quasi-static formulations, and the frequency dependence of effective material parameters appears mainly due to frequency dependence of the material parameters of inclusions, with some possible effect of the dispersive parameters of a matrix.

An applicability of any mixing rule to calculate effective material properties of a composite depends on its dielectric or magnetic contrast. The contrast is the difference between the corresponding material parameter of inclusions and of the host matrix [12]. A rigorous result for the case of small contrast ($\alpha\to0$) is the Landau–Lifshitz–Looyenga (LLL) mixing rule [13,14], which can be written as

$$\beta=p\alpha-p(1-p)\alpha^2/D \tag{3}$$

for a macroscopically isotropic composite in D dimensions. The practically important cases are $D=3$ (an isotropic 3D composite with non-aligned randomly distributed inclusions whose shape could be arbitrary in the general case) and $D=2$ (an assembly of infinitely long cylinders). As the permittivity and permeability of any material converge to unity at frequencies tending to infinity, the contrast $\alpha\to0$ and the LLL mixing rule governs the high-frequency asymptotic behavior of any composite. The result of the LLL mixing rule is independent of the form factor of inclusions. Eqs. (1) and (2) are consistent with (3) at $\alpha\to0$ only when $n_0=1/3$.

* Corresponding author.
E-mail address: krozanov@yandex.ru (K.N. Rozanov).

0304-8853/$ - see front matter © 2011 Elsevier B.V. All rights reserved.
doi:10.1016/j.jmmm.2011.10.028

For the microwave permeability of magnetic composites, it is frequently possible to obtain acceptable agreement with measured data with the use of the MGA and a fitted form factor. The reason is that over the microwave frequency range the intrinsic permeability of magnetic materials, especially of ferrites, does not exceed several units due to its fast decrease with frequency according to Snoek's and Acher's laws, and the contrast between the permeability of inclusions and that of host matrix is not large [12].

High contrast is typical for the permittivity of metal–dielectric composites, as the magnitude of the complex microwave permittivity of metal inclusions is so high that it can be considered as infinite. Therefore, the microwave permittivity of composites filled with metal powders is almost constant over a wide frequency range, and is determined by the morphology of the composite only. Most efforts made in the past have been aimed at deriving a mixing rule to predict the permittivity of a metal–dielectric mixture as a function of concentration, see, e.g., [15] for a review.

For the permittivity of a metal–dielectric mixture, the Bruggeman EMT is conventionally considered as the most suitable theory, because it allows for predicting the percolation threshold, p_c. The percolation threshold is the concentration, at which the mixture starts conducting the d.c. current, and the real part of effective permittivity of the mixture tends to infinity. From Eq. (2), it follows that $p_c = n_0$. However, in reality, p_c is known to vary within a wide range depending on the composite morphology. For example, the percolation threshold variation from 5 to 50% was observed in composites filled with the same carbon powder, but different host matrices [16]. The reason for this is that the structure and the rate of agglomeration of conducting particles in a composite depend on a host matrix type.

Also, the effective form factor found from measured data at low concentrations deviates from $n_0 = 1/3$ even in composites filled with almost spherical particles, e.g., carbonyl iron powder [17]. This could be due to agglomeration of the particles [17]. In turn, disagreement with the LLL theory results in distortions of the permeability frequency characteristics, because it is the LLL theory that governs the material parameters at very high frequencies.

The percolation threshold p_c can be used as a fitting parameter to ensure better agreement with measured concentration dependences of material parameters.

For example, substituting $\alpha \to \infty$ in Eq. (2), it can be easily obtained that

$$\beta = \frac{p}{n_0} \frac{1}{1 - p/n_0}. \tag{4a}$$

Since the percolation threshold is $p_c = n_0$,

$$\beta = \frac{p}{n_0} \frac{1}{1 - p/p_c}. \tag{4b}$$

Eq. (4b) coincides with the convenient fitting equation for the quasi-static effective permittivity of a metal–dielectric mixture, proposed by Odelevskiy [18], see [19] for details. It involves two fitting parameters to approximate the measured concentration dependence, n_0 and p_c. The parameter n_0 is determined by individual properties of inclusions and governs the permittivity at low concentrations. The fitting parameter p_c describes the permittivity behavior near the percolation threshold, where cooperative phenomena are dominant. Notice that the MGA also agrees with (4b) if $\alpha \to \infty$ and $p_c = 1$. The concentration dependence (4b) frequently provides an excellent fit to measured data on the effective permittivity of metal-dielectric composites [19], and hence will be employed in the further derivation.

Alternatively to the mixing rules, the effective material constants of composites can be considered in terms of the Bergman–Milton theory (BMT) [20] as

$$\beta = \int_0^1 B(n) \frac{\alpha}{1 + n\alpha} dn. \tag{5}$$

The spectral function $B(n)$ accounts for a distribution of the effective form factors n, arising from both a statistical spread in the shapes of inclusions and excitation of inhomogeneous fields within the inclusions due to interactions. For the spectral function, the following sum rules are valid, see, e.g. [21]:

$$\int_0^1 B(n) dn = p \quad \text{and} \quad \int_0^1 nB(n) dn = \frac{p(1-p)}{D}. \tag{6}$$

The sum rules are derived from the validity of the LLL mixing rule at the values of material constant close to unity.

Any feasible mixing rule corresponds to a particular form of $B(n)$. For example, the spectral function for the MGA is a delta-function, and therefore the MGA implies the same value of the effective form factor for all inclusions. The EMT accounts for a spread in effective form factors appearing due to the interactions, and involves the spectral function (see, e.g., [22]):

$$B(n) = \begin{cases} \frac{3}{4\pi} \frac{\sqrt{(n-n_1)(n_2-n)}}{n}, n_1 < n < n_2 \\ 0, \quad n < n_1 \quad \text{or} \quad n > n_2 \end{cases}, \tag{7}$$

where n_1 and n_2 are parameters of the theory, both dependent on p and D.

The current literature related to this topic lacks experimental data to explicitly formulate spectral functions of actual composites. Therefore, to apply the BMT to the analysis of the measured properties of composites, a parameterization of $B(n)$, which means the representation in the form with a number of fitting parameters, is needed [17]. For example, the Ghosh–Fuchs theory (GFT) [22] suggests the parameterization that produces a single broad peak of $B(n)$ as

$$B(n) = \begin{cases} C(n-n_1)^{1-A}(n-n_2)^B/n, & 0 \le n_1 \le n_2 \le 1 \\ 0 & \text{otherwise} \end{cases}, \tag{8}$$

where n_1, n_2, A, B, and C are the fitting parameters of the theory. It has been shown recently that the GFT provides an excellent agreement with the measured microwave material parameters of magnetic composites, both their permittivity and permeability [19]. Therefore a distribution of the form factors of inclusions is essential for an accurate prediction of the microwave permittivity and permeability. However, the GFT is inconvenient for use because of its complicated mathematical form: it produces an integral equation that relates α and β.

Herein, a simple analytical formulation of the Ghosh–Fuchs theory is suggested. The proposed mixing rule is based on the shape of the spectral function typical for the EMT with two fitting parameters: the averaged depolarization factor of inclusions n_0 and the percolation threshold p_c. The fitting parameters are found from the concentration dependence of permittivity of the composite, in accordance to the Odelevskiy equation (4b). The requirement of agreement with the LLL mixing rule at low contrast of inclusions provides a unique mixing rule.

2. Theory

The EMT stands out against all the existing mixing rules, because it incorporates the percolation threshold in a metal–dielectric mixture. The reason is that the EMT is a quadratic equation for the effective permittivity. At concentrations below and above the percolation threshold p_c, different solutions of the