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PREDICTION OF ELECTROMAGNETIC PROPERTIES OF FERRITE COMPOSITES

Y. Ma, V. K. Varadan, and V. V. Varadan

9.1 Introduction

Ferrite composite materials as well as other ferromagnetic, carbonyl iron composites recently received wide attention due to their various applications in a quite broad microwave band. Electromagnetic shielding ability and wave absorbing efficiency are just two of their popular characteristics. In order to use these materials in different environments and to optimize their abilities, we must carefully tailor such composite materials. The tailoring work is a complex process. Though it can be done by examining a large number of laboratory-made samples, because of economic and time constraints, the goal can be easily achieved theoretically. For this reason, we provided a direct formalism which can be used to predict the effective electromagnetic properties of ferrite composite materials. The validity of the direct formalism, which later will determine the applicability of its inverse form, is discussed in this chapter.

When ferrite particles are dispersed in a resin matrix to form a composite, depending on the volume fraction of particles, single or multiple scattering dominates the scattered energy when waves are incident on them. Multiple scattering effect cannot be ignored when the concentration of particles is considerable as in the case of ferrite composites. In this chapter, we first review the multiple scattering dependent expressions for the effective permittivity $<\varepsilon>$ and permeability $<\mu>$ of microwave composite materials. These two expressions, which are required in the investigation of the reflection and transmission characteristics for a layer of microwave composite materials in response to incident millimeter-waves and microwaves, have been derived by the authors [Ma et al., 1988]. In the derivation, we employed the multiple scattering formalism, which has been previously applied to get the effective propagation constant for the ferrite composites [Varadan et al., 1986]. Although people have used Maxwell-Garnett equation, Clausius-Mosotti equation, and others to find $<\varepsilon>$ and $<\mu>$ of a two phase heterogeneous medium, none of them considered multiple scattering when the concentration of the inclusion phase became fairly large. In order to check the validity of the expressions, several cases involving lossy/lossless matrix and inclusion materials are discussed.

As one also knows, the wave absorbing power of ferrite-resin composites relies on the essential electric and magnetic properties of the inclusion phase in the microwave band in addition to its matrix properties, specific concentration, distribution, and geometry. Unfortunately, measured values of electric permittivity and magnetic permeability of most inclusion materials, for these applications, are not very common in the literature. Even so, depending on different manufacturing processes, the reported values vary from one place to another. Unless the properties are known, the design work will still remain in the mist. We can, however, predict these complex properties using the effective property measurements done for a high (or low) porosity microwave composites. The expressions for $<\varepsilon>$ and $<\mu>$ of ferrite composite materials can also be used to solve this inverse problem.

9.2 Dispersion Equation in the Long Wavelength Limit

The objective of the self-consistent multiple scattering formalism is to obtain an analytical expression for the dispersion equation of a wave propagating in the random medium. Details of this can be found in one of our papers published in this issue [Varadan et al., 1990] or in our previous publications [Varadan et al., 1986; Ma et al., 1988]. By solving the dispersion equation, we can obtain an effective propagation constant of the random medium. However, this is a forbidden task and can only be done numerically when the size of the scatterers is large compared with the incident wavelength.

Fortunately, for most microwave composite materials, the scatterers are much smaller than a millimeter in size. Therefore, at frequencies in the GHz range, the wavelength in the matrix material is much larger than the size a of particles and make the nondimensional frequency ka fall in the Rayleigh region. Thus, one can solve the dispersion equation in the long wavelength regime and obtain a closed form solution.

Retaining only the dipole terms, we have the following dispersion equation [Varadan et al., 1986]

$$\begin{vmatrix} T^1(JH_o + JH_2/2) - 1 & 3JH_1T^2/2 \\ 3JH_1T^1/2 & T^2(JH_o + JH_2/2) - 1 \end{vmatrix} = 0 \qquad (1)$$

where,

$$JH_o = 3j\Omega/X_2^3 + w$$

$$JH_1 = 3j\Omega(K/k_2)/X_2^3$$

$$JH_2 = 3j\Omega(K/k_2)^2/X_2^3$$

$$\Omega = c/[1 - (K/k_2)^2]$$

$$T^1 = j(2X_2^3/3)[(\mu_1 - \mu_2)/(2\mu_2 + \mu_1)]$$

$$T^2 = j(2X_2^3/3)[(\varepsilon_1 - \varepsilon_2)/(2\varepsilon_2 + \varepsilon_1)]$$

$$w = (1 - c)^4/(1 + 2c)^2$$

$$X_2 = Re(k_2 a)$$

$$\mu_1 = \mu_1' + j\mu_1'' \quad \text{(scatterer)}$$

$$\varepsilon_1 = \varepsilon_1' + j\varepsilon_1'' \quad \text{(scatterer)}$$

$$\mu_2 = \mu_2' + j\mu_2'' \quad \text{(host)}$$

$$\varepsilon_2 = \varepsilon_2' + j\varepsilon_2'' \quad \text{(host)}$$

$$k_2 = \omega(\mu_2\varepsilon_2)^{1/2}/c_o$$

K = Effective propagation constant $= \omega(<\mu><\varepsilon>)^{1/2}/c_o$

$$c_o = \text{Light speed in vacuum}$$

$$c = \text{Volume fraction of scatterers}$$

The effective dielectric constant $< \varepsilon >$ and permeability $< \mu >$ can be easily obtained from (1) due to the fact that T^1 and T^2 are uncoupled and the effective propagation constant K is polarization independent in the Rayleigh region. This approach can also be applied to study electromagnetic scattering by a periodic array of particles.

For randomly distributed lossy/lossless scatterers with a high concentration in a lossy/lossless matrix, the effective permeability and permittivity are, respectively,

$$\begin{aligned} < \mu > /\mu_2 = &\{1 - [\beta w - (c\gamma - cj)/(1+\gamma^2)](M_2 + jM_1)\} \\ &/\{1 - [\beta w + (c\gamma - cj)/2(1+\gamma^2)](M_2 + jM_1)\} \end{aligned} \quad (2)$$

$$\begin{aligned} < \varepsilon > /\varepsilon_2 = &\{1 - [\beta w - (c\gamma - cj)/(1+\gamma^2)]\,(E_1 + jE_2)\} \\ &/\{1 - [\beta w + (c\gamma - cj)/2(1+\gamma^2)]\,(E_1 + jE_2)\} \end{aligned}$$

where,

$$M_1 = 2(B\gamma + A) \qquad M_2 = 2(A\gamma - B)$$

$$E_1 = 2(C\gamma - D) \qquad E_2 = 2(D\gamma + C) \quad (3)$$

$$A = [(\mu_1' - \mu_2')(2\mu_2' + \mu_1') + (\mu_1'' - \mu_2'')(2\mu_2'' + \mu_1'')]/\Delta$$

$$B = [(\mu_1'' - \mu_2'')(2\mu_2' + \mu_1') + (\mu_1' - \mu_2')(2\mu_2'' + \mu_1'')]/\Delta$$

$$C = [(\varepsilon_1' - \varepsilon_2')(2\varepsilon_2' + \varepsilon_1') + (\varepsilon_1'' - \varepsilon_2'')(2\varepsilon_2'' + \varepsilon_1'')]/\Delta' \quad (4)$$

$$D = [(\varepsilon_1'' - \varepsilon_2'')(2\varepsilon_2' + \varepsilon_1') + (\varepsilon_1' - \varepsilon_2')(2\varepsilon_2'' + \varepsilon_1'')]/\Delta' \quad (5)$$

$$\Delta = (2\mu_2' + \mu_1')^2 + (2\mu_2'' + \mu_1'')^2$$

$$\Delta' = (2\varepsilon_2' + \varepsilon_1')^2 + (2\varepsilon_2'' + \varepsilon_1'')^2 \quad (6)$$

$$\gamma = (X_2'^3 - 3X_2^2X_2')/(X_2^3 - 3X_2X_2'^2)$$

$$\beta = (X_2^3 - 3X_2X_2'^2)/3$$

$$X_2' = Im(k_2a)$$

9.3 Discussion of the Direct Formalism

Different from various equations, which are quite popularly used, such as Maxwell-Garnett equation, Clausius-Mosotti equation, and other equations like Lichtenecker logarithmic equation, our equations for independent permittivity and permeability can be used in either purely scattering domain or absorption domain. In other words, the matrix as well as the inclusion materials can both be lossy and lossless or combinations of the two. Furthermore, a rigorous multiple scattering theory has been employed in the derivations accounted for high concentrations of the inclusion phase while none of the mentioned equations has considered multiple scattering.

In the following, we discuss some limiting cases when (2) is applied to compute the effective properties of microwave composite materials. Because the effective permittivity and permeability bear the same form, for simplicity, we use only the expressions for the effective permittivity. However, all the following discussion are exactly those for the effective permeability when all the $\varepsilon' s$ are replaced by the $\mu' s$ in the statements.

Case 1. Pure scattering

If neither the scatterers nor the matrix material are lossy, i.e., $\varepsilon_1'' = \varepsilon_2'' = 0$, only scattering contributes to the imaginary part of the effective dielectric constant $<\varepsilon>$.

$$<\varepsilon>/\varepsilon_2' = (1 + 2c\ \mathrm{T})/(1 - c\ \mathrm{T}) + j6\beta wc\ \mathrm{T}^2/(1 - c\mathrm{T})^2 \quad (7)$$

where $\mathrm{T} = (\varepsilon_1' - \varepsilon_2')/(\varepsilon_1' + 2\varepsilon_2')$. The effective dielectric constant $<\varepsilon>$ becomes ε_2' and ε_1', respectively, when c equals to 0 (no scatterer) and 1 (matrix material totally occupied by scatterers). Meanwhile, the imaginary part of $<\varepsilon>$ vanishes due to the disappearance of scatterers.

Case 2. Lossy scatterers in a lossless matrix

This is a common case happened in various problems, e.g., suspensions in fluids. For this case, $\varepsilon_2'' = 0$ but ε_1'' is not. Equation (2) can be reduced to the form as follows

$$<\varepsilon>/\varepsilon_2' = \{1+2(\beta w+cj)(D-jC)\}/\{1+2\,[\beta w-cj/2](D-jC)\} \quad (8)$$

Again, $<\varepsilon>$ becomes ε_2' when $c = 0$. However, when $c = 1$,

$$<\varepsilon>/\varepsilon_2' = (1 + 2C + 2jD)/(1 - C - jD) = (\varepsilon_1' + j\varepsilon_1'')/\varepsilon_2' \qquad (9)$$

It is easy to see that the effective dielectric constant becomes that of the lossy scatterers.

Case 3. Lossless scatterers in a lossy matrix

Although this problem has seldom been considered for electromagnetic case, e.g., particles in a viscous fluid, it is useful to check the validity of the formalism. For this case, $\varepsilon_1'' = 0$ and ε_2'' is finite. It is easy to show using (2), for $c = 0$, $<\varepsilon> = \varepsilon_2 = \varepsilon_2' + j\varepsilon_2''$. For $c = 1$, (2) reduces to

$$<\varepsilon>/\varepsilon_2 = (1 + 2C + 2jD)/(1 - C - jD) = \varepsilon_1'/(\varepsilon_2' + j\varepsilon_2'') \qquad (10)$$

which means the effective dielectric constant becomes that of the scatterers and is real.

Case 4. Lossy scatterers in a lossy matrix

For this case, the complete form of (2) must be considered. As for the relative contribution from scattering and absorption towards $<\varepsilon>$, one can easily show by using case 2 that

$$\begin{aligned}<\varepsilon>/\varepsilon_2' = &\left[(1 + 2c\,C)(1 - c\,C) - 2\,c^2\,D^2\right]/\left[(1 - c\,C)^2 + c^2\,D^2\right] \\ &+ j\left[3\,c\,D + 6\,\beta wc(C^2 + D^2)\right]/\left[(1 - c\,C)^2 + c^2\,D^2\right] \qquad (11)\end{aligned}$$

If $\varepsilon_2'' = \varepsilon_1'' = 0$ then D = 0, the above equation becomes that for case 1. Therefore, the scattering contribution to the imaginary part of $<\varepsilon>$ has a β dependence. Because β is of the order of $(k_2a)^3$, which is much less than one for the long wavelength limit, we know the scattering contribution is fairly small when compared with the absorption contribution which is frequency independent if ε_1 and ε_2 are also frequency independent. The real part of the effective dielectric constant is less affected by the absorption due to the fact that ε_1'' is, in general, fairly small.

9.4 Inverse Formulation

One can obtain the complex ε_1 and μ_1 of the scatterer (inclusion phase) by solving the equation for the effective properties of the ferrite composites. This approach is very useful in the sense that sometimes it is easier to obtain experimentally the properties of ferrite composites rather than the direct measurement of the properties of the inclusion phase such as ferrites. For a provided composite sample with limited information, we can retrieve some unknowns by this inverse approach.

Equation (2) can be used to get the solutions for ε_1 and $\mu_1(\varepsilon_2$ and $\mu_2)$ when the concentration $c, <\varepsilon>, <\mu>$, and ε_2 and $\mu_2(\varepsilon_1$ and $\mu_1)$ are known. The effective permittivity $<\varepsilon>$ and permeability $<\mu>$ are both complex if and only if the dielectric and magnetic properties of the matrix and the inclusion materials are different. Otherwise, e.g., the effective permeability $<\mu>$ is real for a nonmagnetic inclusion material. For each effective constant (presented is for effective $<\varepsilon>$, same for effective $<\mu>$), we can set up two equations for two unknowns E_1 and E_2. These two equations are essentially the equations for two circles and thus have the following form

$$(E_1 + p)^2 + (E_2 + q)^2 = f \tag{12}$$

and

$$(E_1 + p')^2 + (E_2 + q')^2 = f'$$

where p, q, f, p', q', and f' can all be determined by given input parameters (details in Appendix).

The solutions of E_1 and E_2 are geometrically the intersection points of two circles and thus can have at most two intersection points. The values of E_1 and E_2 are then substituted into (3) to solve for ε_1' and ε_1'' by use (4), (5), and (6). Again, we generate two equations for two circles as

$$(X + u)^2 + (Y + v)^2 = g \tag{13}$$

and

$$(X + u')^2 + (Y + v')^2 = g'$$

where u, v, g, u', v' and g' are all in terms of the given input and the previously solved E_1 and E_2. Finally, the real part of the permittivity ε_1' can be obtained by adding ε_2' to X while the imaginary part of the permittivity ε_1'' is by adding ε_2'' to Y. All the parameters appeared in (12) and (13) are given in the Appendix.

9.5 Discussion of the Inverse Formalism

The direct formalism has been successfully applied to the investigation of the phase velocity and attenuation, which can be expressed in terms of $<\varepsilon>$, of dielectric composites [Varadan et al., 1985]. However, a great care needs to be taken when using (12) and (13) to find the unknowns ε_1' and ε_1'' or μ_1' and μ_1''. Since the solutions are exact based on the input, any deviation (error allowance) from the measurements for the effective $<\varepsilon>$ and $<\mu>$ will cause unphysical results such as negative ε and μ although they still satisfy the equation for a circle. Nevertheless, by proper adjustment and judgment, we can pick up and modify the values from the intersection points for two circles to make them satisfy the direct equations. A graphical representation of the solutions of (12) and (13) is also given as Fig. 9.1. In Figs. 9.1c and 9.1d, two cases may also arise when examining the intersection of two circles. The case of Fig. 9.1d seldom happens because this implies a perfect match between the experimental results and the modeling while that of Fig. 9.1c can be avoided by modifying the model and by improving the experimental work.

Without proof, we picked up the experimental data for $<\varepsilon>$ and $<\mu>$ given by Ueno et al. [1980] to predict the permittivity and permeability of Fe_3O_4 they used to make the radio wave scattering suppressors. We assumed 70% volume fraction of Fe_3O_4 and the matrix material is PVC whose permittivity $\varepsilon_2 = 2.9 + 0.03j$ and permeability $\mu_2 = 1 + 0j$. The values of the permittivity ε_1 and permeability μ_1 of Fe_3O_4 for such a case are depicted in Fig. 9.2. One can tell that the dispersion characteristics of ferrite permittivity are very similar to those reported [Westphal and Sils, 1972] and the magnitudes of its real and imaginary parts depending on how the ferrite was made. The peak of $<\mu''>$ happens at about 2 GHz which is also the known low gigahertz magnetic resonant phenomenon for most ferromagnetic materials [Lax and Button, 1962].

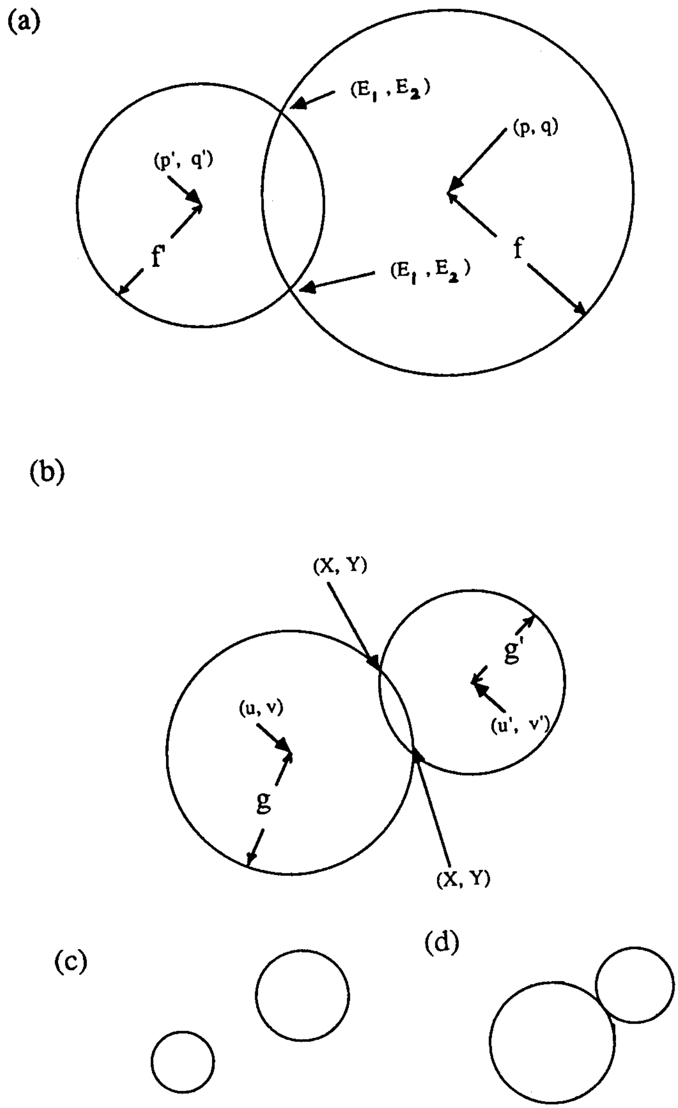

Figure 9.1 Graphical representation of (12) and (13).

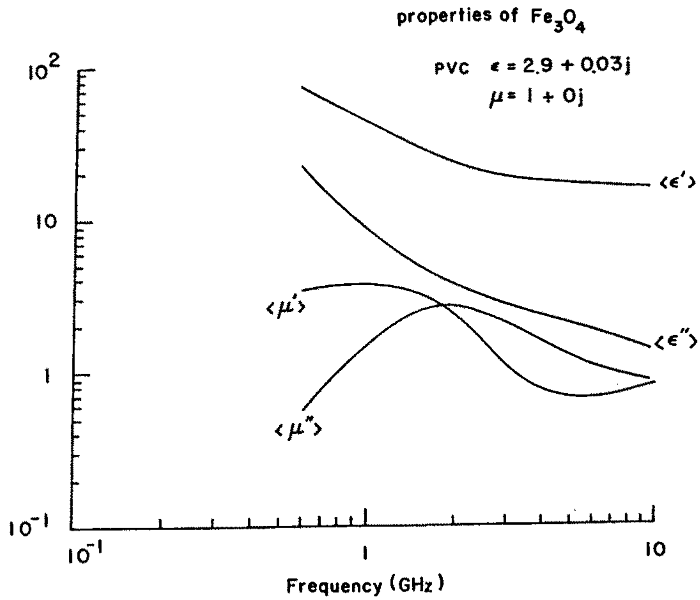

Figure 9.2 Electromagnetic properties of Fe_3O_4 (based on Ueno et al., 1980).

Appendix

Parameters in (12) and (13)

$$p = (\alpha_1 + \eta_1 - 2\eta_1 H)/2(H\eta_1^2 + H\eta_2^2 - \alpha_1\eta_1 - \alpha_2\eta_2)$$

$$q = (-\alpha_2 - \eta_2 + 2\eta_2 H)/2(H\eta_1^2 + H\eta_2^2 - \alpha_1\eta_1 - \alpha_2\eta_2)$$

$$f = (\alpha_1 + \eta_1 - 2\eta_1 H)^2/4(H\eta_1^2 + H\eta_2^2 - \alpha_1\eta_1 - \alpha_2\eta_2)^2$$

$$+ (-\alpha_2 - \eta_2 + 2\eta_2 H)^2/4(H\eta_1^2 + H\eta_2^2 - \alpha_1\eta_1 - \alpha_2\eta_2)^2$$

$$- (H - 1)/(H\eta_1^2 + H\eta_2^2 - \alpha_1\eta_1 - \alpha_2\eta_2)$$

$$p' = (\alpha_2 - \eta_2 - 2\eta_1 G)/2(G\eta_1^2 + G\eta_2^2 + \alpha_1\eta_2 - \alpha_2\eta_1)$$

$$q' = (\alpha_1 - \eta_1 + 2\eta_2 G)/2(G\eta_1^2 + G\eta_2^2 + \alpha_1\eta_2 - \alpha_2\eta_1)$$

$$f' = (-\alpha_2 + \eta_2 + 2\eta_1 G)^2/4(G\eta_1^2 + G\eta_2^2 + \alpha_1\eta_2 - \alpha_2\eta_1)^2$$
$$+ (\alpha_1 - \eta_1 + 2\eta_2 G)^2/4(G\eta_1^2 + G\eta_2^2 + \alpha_1\eta_2 - \alpha_2\eta_1)^2$$
$$- G/(G\eta_1^2 + G\eta_2^2 + \alpha_1\eta_2 - \alpha_2\eta_1)$$

$$\alpha_1 = \beta w - c\gamma/(1+\gamma^2)$$

$$\alpha_2 = c/(1+\gamma^2)$$

$$\eta_1 = \beta w + c\gamma/2(1+\gamma^2)$$

$$\eta_2 = -c/2(1+\gamma^2)$$

$$H = [< \varepsilon' > \varepsilon_2' + < \varepsilon'' > \varepsilon_2'']/[\varepsilon_2'^2 + \varepsilon_2''^2]$$

$$G = [< \varepsilon'' > \varepsilon_2' - < \varepsilon_2' > \varepsilon_2'']/[\varepsilon_2'^2 + \varepsilon_2''^2]$$

$$u = (6C\varepsilon_2' - 3\varepsilon_2'')/2(C-1)$$

$$v = (6C\varepsilon_2'' - 3\varepsilon_2'')/2(C-1)$$

$$g = (6C\varepsilon_2' - 3\varepsilon_2'')^2/4(C-1)^2 + (6C\varepsilon_2'' - 3\varepsilon_2'')^2$$
$$/4(C-1)^2 - 9C[\varepsilon_2'^2 + \varepsilon_2''^2]/(C-1)$$

$$u' = (6D\varepsilon_2' + 3\varepsilon_2'')/2D$$

$$v' = (6D\varepsilon_2'' - 3\varepsilon_2')/2D$$

$$g' = (6D\varepsilon_2' + 3\varepsilon_2'')^2/4D^2 + (6D\varepsilon_2'' - 3\varepsilon_2')^2/4D^2 - 9[\varepsilon_2'^2 + \varepsilon_2''^2]$$

$$C = (E_2\gamma - E_1)/2(\gamma^2 + 1)$$

$$D = (E_2\gamma + E_1)/2(\gamma^2 + 1)$$

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