

Mixing Rules with Complex Dielectric Coefficients

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This article discusses the determination of effective dielectric properties of heterogeneous materials, in particular media with lossy constituents that have complex permittivity parameters. Several different accepted mixing rules are presented and the effects of the structure and internal geometry of the mixture on the effective permittivity are illustrated. Special attention is paid to phenomena that the mixing process causes in the character of the macroscopic dielectric response of the mixture when the losses of one or several of the components are high or when there is a strong dielectric contrast between the component permittivities.

1. Background

The objective of this article is to provide theoretical background for understanding and prediction of macroscopic dielectric properties of geophysical and especially subsurface materials. To provide an exact electromagnetic description for the structural complexity of materials occurring in nature is, of course, an impossible undertaking. Mixing rules are an attempt to perform this task approximately. The approach in analytical mixing formulas is to idealize the geometry and model the microstructure using simple forms, like spheres and ellipsoids. It is encouraging that, quite often, this strategy turns out to lead to mixing rules that approximate the real world to such an extent that the mixing predictions can be used in practical remote sensing work. Special emphasis in this article is given to moist substances and their dielectric behavior.

1.1. The Concept of Effective Medium

Dielectric mixing rules are algebraic formulas with which the effective permittivity of the mixture can be calculated as a function of the constituent permittivities, their fractional volumes, and possibly some other

parameters characterizing the microstructure of the mixture. The mixture can be discrete, which means that homogeneous inclusions are embedded in another homogeneous medium, or then the permittivity function can be continuous. Use of the concept of effective, or macroscopic permittivity, implies that the mixture responds to electromagnetic excitation as if it were homogeneous.*

This view to the homogenization of heterogeneous media is obviously not exact because by using electromagnetic waves of higher and higher frequency one can always “see” more accurately into the medium and probe its structural details. Therefore a quantity such as effective permittivity is only meaningful in the long-wavelength limit. This limit corresponds to low frequencies and indeed, the mixing rules are very often derived using static or quasi-static arguments. The size of the inclusions in the mixture and the spatial correlation length of the permittivity function need to be small with respect to the wavelength.

The effective permittivity can be complex, $\epsilon_{\text{eff}} = \epsilon'_{\text{eff}} - j\epsilon''_{\text{eff}}$, where the real and imaginary parts are certain averages of the real and imaginary parts of the components materials. To some extent, the validity of the effective permittivity can be generalized to include the first-order scattering effects of the inclusions. Because scattering entails losses, its effect gives a contribution to the imaginary part of the effective permittivity, which can be estimated by calculating the energy that the electric dipoles radiate which are induced within the inhomogeneities of the mixture. On the other hand, although the low-frequency character of the effective-medium description is a limitation of mixing theories, these mixing models can be extended to magnetic, anisotropic, and even magnetoelectric materials.

1.2. Quasi-Static Approach and Homogenization Principles

When a dielectric inclusion that is exposed to an electromagnetic field is small it can be safely assumed that its momentary internal field is the same as in the problem with a static excitation. The inclusion creates a perturbation to the field which to the lowest order is that of an electric dipole. The polarizability of the inclusion can be enumerated by solving the Laplace equation for the field inside the scatterer, in other words neglecting the dynamic wave processes altogether.

It is not easy to give an exact upper frequency limit for the validity of the concept of effective permittivity because the exact response of a randomly heterogeneous medium is beyond analysis. However, the following

*Often the term *dielectric constant* is used instead of *permittivity*. It may, however, be proper to remind that the dielectric constant of a material very seldom is constant with respect to temperature, frequency, or any material property.

rule of thumb is often used: the size of an inclusion in the mixture must not exceed a tenth of the wavelength in the effective medium. In fact, this criterion is an estimate towards the conservative side.

Once the polarizabilities of the inclusions composing the mixture are known, their interaction within the medium has to be taken into account. If the density of the inclusions is high it is important to analyze how the dipole moments “communicate” with each other. Another problem that one has to face in the theory of dielectric mixtures is the fact that it is not at all obvious how to enumerate the “real” excitation field for a given scatterer in a dense and random mixture. These questions are in fact being discussed in the present electromagnetics literature. Different solutions to these problems lead to different mixing rules that are applied today to various problems not only in remote sensing, microwave aquametry, and subsurface technology but also in materials science, for example in the design of new composite materials.

The early history of dielectric mixing rules can be traced back into the mid-1800's [1], and the turn of the century a hundred years ago saw dielectric mixing formulas in more or less in a present-day form [2]. Famous names like Clausius, Mossotti, Maxwell, L.V. Lorenz, H.A. Lorentz, Rayleigh, and Garnett are affiliated with the dielectric and optical properties of materials. Later on during the last century, as experimental results on dielectric properties of materials began to accumulate, shape effects in the microstructure of the mixture in the theoretical models were given particular attention. Another direction of research was pushed forward by the fact that different mixing rules predicted different results for the same mixture. This forced researchers into looking for upper and lower bounds for the effective permittivity of a given mixture, and there the pioneering work by Hashin and Shtrikman [3] is the basic reference. For a review of the mixing development throughout the century, see Landauer [4] and Sihvola [5]. The present-day research on mixing formulas focuses much on mixtures of complex materials, for example bi-anisotropic and non-linear media (see, e.g., [6,7]).

2. Basic Mixing and Maxwell Garnett Formula

This section presents the basic principle how the simplest mixing formula, so-called Maxwell Garnett rule, can be derived. The mixture to be analyzed consists of the background medium where spherical inclusions are embedded according to Figure 1. The two components composing the mixture are often called *phases*. The environment phase can also be termed *matrix* or *host*, and the inclusion phase as *guest*.

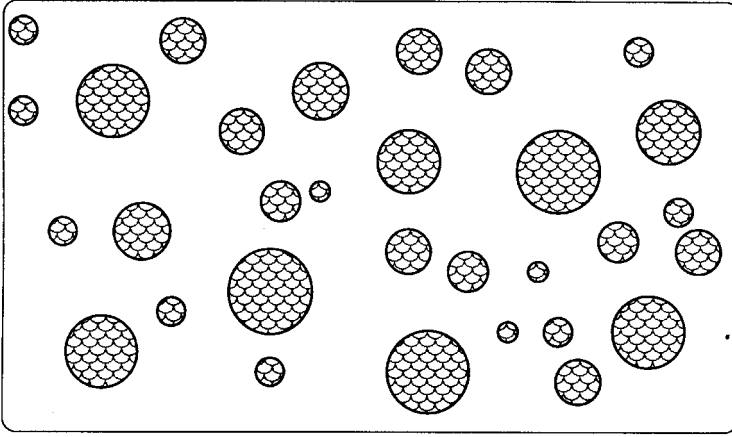


Figure 1. A simple mixture: spherical inclusions in a homogeneous background medium. The permittivity of the inclusions is ϵ_i and that of the environment ϵ_e .

2.1. Polarizability of a Dielectric Sphere

The polarizability of an inclusion is a measure for its response to an incident electric field. The polarizability of a particle α is the relation between the dipole moment \mathbf{p} that is induced in the inclusion by the polarization, and the external electric field \mathbf{E}_e :

$$\mathbf{p} = \alpha \mathbf{E}_e \quad (2.1)$$

For a sphere the polarizability is easy to calculate. It is proportional to the internal field within the inclusion, its volume, and the dielectric contrast between the inclusion and the environment. Since the electric field \mathbf{E}_i induced in a sphere in a uniform and static external field \mathbf{E}_e is also uniform, static, and parallel to the external field [8, Section 4.4],

$$\mathbf{E}_i = \frac{3\epsilon_e}{\epsilon_i + 2\epsilon_e} \mathbf{E}_e \quad (2.2)$$

the polarizability can be written immediately:

$$\alpha = V(\epsilon_i - \epsilon_e) \frac{3\epsilon_e}{\epsilon_i + 2\epsilon_e} \quad (2.3)$$

where the permittivities of the inclusion and its environment are denoted by ϵ_i and ϵ_e , respectively. The volume of the sphere is V . Note that the polarizability is a scalar quantity. This is because the inclusion material is isotropic and its shape is spherically symmetric.

2.2. Clausius–Mossotti Formula

Now that the polarizability of a single sphere is known, the effective permittivity of a mixture can be calculated as a function of the number density of the spheres in the background medium with permittivity ϵ_e . The effective permittivity is the relation between the external field and the average electric flux density $\langle \mathbf{D} \rangle$:

$$\langle \mathbf{D} \rangle = \epsilon_{\text{eff}} \mathbf{E}_e = \epsilon_e \mathbf{E}_e + \langle \mathbf{P} \rangle \quad (2.4)$$

where the average polarization $\langle \mathbf{P} \rangle$ is connected to the dipole moment density in the mixture:

$$\langle \mathbf{P} \rangle = n\mathbf{p} \quad (2.5)$$

and here n is the number density of dipole moments \mathbf{p} in the mixture.*

In a mixture, especially when it is dense, one cannot assume the field exciting one inclusion to be the external field \mathbf{E}_e . The surrounding polarization increases the field effect and has to be taken into account [9]. The field that excites one inclusion \mathbf{E}_L is often called as the *local field* or *Lorentzian field*. It is dependent on the shape of the inclusion [10], and for a sphere it is

$$\mathbf{E}_L = \mathbf{E}_e + \frac{1}{3\epsilon_e} \mathbf{P} \quad (2.6)$$

where $1/3$ is the depolarization factor of the sphere. Combining this equation with $\mathbf{p} = \alpha \mathbf{E}_L$ leaves us with the average polarization, and then the effective permittivity can be written (see Eq. (2.4)):

$$\epsilon_{\text{eff}} = \epsilon_e + \frac{n\alpha}{1 - n\alpha/(3\epsilon_e)} \quad (2.7)$$

The equation can often be seen in the form

$$\frac{\epsilon_{\text{eff}} - \epsilon_e}{\epsilon_{\text{eff}} + 2\epsilon_e} = \frac{n\alpha}{3\epsilon_e} \quad (2.8)$$

This relation carries the name *Clausius–Mossotti formula*, although it deserves the label *Lorenz–Lorentz formula* as well [11]. The dilute-mixture approximation can be written by taking the limit of small n :

$$\epsilon_{\text{eff}} \approx \epsilon_e + n\alpha \quad (2.9)$$

*Note the dimensions of the quantities: $[\mathbf{D}] = [\mathbf{P}] = \text{As/m}^2$, $[\mathbf{p}] = \text{Asm}$, $[\mathbf{E}_e] = \text{V/m}$, $[\alpha] = \text{Asm}^2/\text{V}$, and $[n] = \text{m}^{-3}$.

2.3. Maxwell Garnett Mixing Rule

In practical applications quantities like polarizabilities and scatterer densities are not always those most convenient to use. Rather, one prefers to play with the permittivities of the components of the mixture. When this is the case, it is advantageous to combine Clausius–Mossotti formula with the polarizability expression (2.3). Then we can write

$$\frac{\epsilon_{\text{eff}} - \epsilon_e}{\epsilon_{\text{eff}} + 2\epsilon_e} = f \frac{\epsilon_i - \epsilon_e}{\epsilon_i + 2\epsilon_e} \quad (2.10)$$

where $f = nV$ is a dimensionless quantity, the volume fraction of the inclusions in the mixture. This formula is called *Rayleigh mixing formula*. Note that because only the volume fraction and the permittivities appear in the mixture rule, the spheres need not be of the same size if only all of them are small compared to the wavelength.

Perhaps the most common mixing rule is the *Maxwell Garnett* formula which is the Rayleigh rule (2.10) written explicitly for the effective permittivity:

$$\epsilon_{\text{eff}} = \epsilon_e + 3f\epsilon_e \frac{\epsilon_i - \epsilon_e}{\epsilon_i + 2\epsilon_e - f(\epsilon_i - \epsilon_e)} \quad (2.11)$$

This formula is in wide use in very diverse fields of application. The beauty of the Maxwell Garnett formula is in its simple appearance combined with its broad applicability. It satisfies the limiting processes for vanishing inclusion phase

$$f \rightarrow 0 \quad \Rightarrow \quad \epsilon_{\text{eff}} \rightarrow \epsilon_e \quad (2.12)$$

and vanishing background

$$f \rightarrow 1 \quad \Rightarrow \quad \epsilon_{\text{eff}} \rightarrow \epsilon_i \quad (2.13)$$

The perturbation expansion of the Maxwell Garnett rule gives the mixing equation for dilute mixtures ($f \ll 1$):

$$\epsilon_{\text{eff}} = \epsilon_e + 3f\epsilon_e \frac{\epsilon_i - \epsilon_e}{\epsilon_i + 2\epsilon_e} \quad (2.14)$$

Figure 2 shows the prediction of the Maxwell Garnett formula for different values of the dielectric contrast ϵ_i/ϵ_e . Shown is the susceptibility ratio

$$\frac{\epsilon_{\text{eff}} - \epsilon_e}{\epsilon_i - \epsilon_e}$$

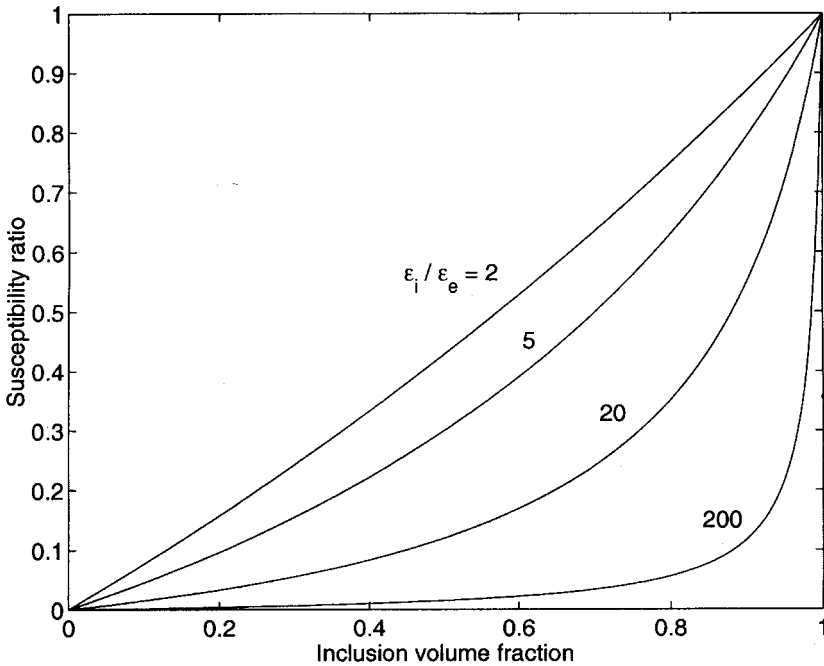


Figure 2. The susceptibility ratio $(\epsilon_{\text{eff}} - \epsilon_e)/(\epsilon_i - \epsilon_e)$ for the Maxwell Garnett prediction of the effective permittivity of a mixture with spherical inclusions of permittivity ϵ_i in a background medium of permittivity ϵ_e .

which vanishes for $f=0$ and is unity for $f=1$, independently of the inclusion-to-background contrast. The figure shows clearly the fact that the effective permittivity function becomes a very nonlinear function of the volume fraction for large dielectric contrasts.

3. Advanced Mixing Principles

The basic treatment of a mixture as an effective medium in the above section was the most idealized model that one can imagine: a two-phase mixture with isotropic dielectric components and the inclusion geometry was spherical. There are “materials” in the real world that follow this model quite well, for example clouds, fog, or even light rain to some extent. Fog is formed of water droplets in air that are certainly spherical, and the mixture is very dilute. As long as scattering effects can be neglected, the attenuation and phase delays in radio wave propagation problems in fogs and

clouds can be very accurately analyzed using the Maxwell Garnett principles.

But most classes of geophysical media that are encountered in remote sensing applications do not obey the assumptions of the basic Maxwell Garnett mixing. Therefore its generalization into more complicated heterogeneities is necessary. That is the aim of the present section.

3.1. Multiphase Mixtures

Wet snow, for example, is a mixture of three phases: air, ice, and liquid water. How to use the previous analysis for snow? To write down the mixing rule for mixtures where there are several components one has to choose one of the components as the host and then treat the polarization effect of all of the guest phases separately. The previous two-phase analysis can be followed very closely. Where above the total polarization was calculated from the individual dipole moments according to (2.5), now each guest phase contributes one such term to a sum of many. The final result is (compare (2.10))

$$\frac{\varepsilon_{\text{eff}} - \varepsilon_e}{\varepsilon_{\text{eff}} + 2\varepsilon_e} = \sum_{n=1}^N f_n \frac{\varepsilon_{i,n} - \varepsilon_e}{\varepsilon_{i,n} + 2\varepsilon_e} \quad (3.15)$$

where f_n is the volume fraction of the inclusions of the n th phase in the mixture, and $\varepsilon_{i,n}$ is its permittivity. And of course this can be solved for the effective permittivity:

$$\varepsilon_{\text{eff}} = \varepsilon_e + 3\varepsilon_e \frac{\sum_{n=1}^N f_n \frac{\varepsilon_{i,n} - \varepsilon_e}{\varepsilon_{i,n} + 2\varepsilon_e}}{1 - \sum_{n=1}^N f_n \frac{\varepsilon_{i,n} - \varepsilon_e}{\varepsilon_{i,n} + 2\varepsilon_e}} \quad (3.16)$$

Here again, all inclusions of all phases were assumed to be spherical.

3.2. Shape Effects of Inclusions

The assumption of spherical shape for the inclusions needs to be relaxed because many natural media possess inclusions of other forms. The polarizability of small particles can of course be calculated for any shape but in general this requires numerical effort. The only shapes for which simple analytical solutions can be found are ellipsoids. Fortunately, ellipsoids allow many practical special cases, like discs and needles, for example.

3.2.1. Depolarization Factors

The important parameters in the geometry of an ellipsoid are its depolarization factors. If the semi-axes of an ellipsoid in the three orthogonal directions are a_x , a_y , and a_z , the depolarization factor N_x (the factor in the a_x -direction) is

$$N_x = \frac{a_x a_y a_z}{2} \int_0^\infty \frac{ds}{(s + a_x^2) \sqrt{(s + a_x^2)(s + a_y^2)(s + a_z^2)}} \quad (3.17)$$

For the other depolarization factor N_y (N_z), interchange a_y and a_x (a_z and a_x) in the above integral.

The three depolarization factors for any ellipsoid satisfy

$$N_x + N_y + N_z = 1 \quad (3.18)$$

A sphere has three equal depolarization factors of 1/3. The other two special cases are a disc (depolarization factors 1,0,0), and a needle (0, 1/2, 1/2). For ellipsoids of revolution, prolate and oblate ellipsoids, closed-form expressions for the integral (3.17) can be found [12].

Prolate spheroids ($a_x > a_y = a_z$) have

$$N_x = \frac{1 - e^2}{2e^3} \left(\ln \frac{1 + e}{1 - e} - 2e \right) \quad (3.19)$$

and

$$N_y = N_z = \frac{1}{2}(1 - N_x) \quad (3.20)$$

where the eccentricity is $e = \sqrt{1 - a_y^2/a_x^2}$. For nearly spherical prolate spheroids, which have small eccentricity, the following holds:

$$N_x \approx \frac{1}{3} - \frac{2}{15} e^2 \quad (3.21)$$

$$N_y = N_z \approx \frac{1}{3} + \frac{1}{15} e^2 \quad (3.22)$$

For oblate spheroids ($a_x = a_y > a_z$),

$$N_z = \frac{1 + e^2}{e^3} (e - \arctan e) \quad (3.23)$$

$$N_x = N_y = \frac{1}{2}(1 - N_z) \quad (3.24)$$

where $e = \sqrt{a_x^2/a_z^2 - 1}$. For nearly-spherical oblate spheroids,

$$N_z \approx \frac{1}{3} + \frac{2}{15} e^2 \quad (3.25)$$

$$N_x = N_y \approx \frac{1}{3} - \frac{1}{15} e^2 \quad (3.26)$$

For a general ellipsoid with three different axes, the depolarization factors have to be calculated from the integral (3.17). Osborn and Stoner have tabulated the depolarization factors of a general ellipsoid [13,14].

3.2.2. Aligned Mixture

Now consider a mixture where ellipsoids of permittivity ε_i are embedded in the environment ε_e . Let all the ellipsoids be aligned. Then the effective permittivity of the mixture is anisotropic, in other words it has different permittivity components in the different principal directions. The Maxwell Garnett formula for this mixture is

$$\varepsilon_{\text{eff},x} = \varepsilon_e + f\varepsilon_e \frac{\varepsilon_i - \varepsilon_e}{\varepsilon_e + (1-f)N_x(\varepsilon_i - \varepsilon_e)} \quad (3.27)$$

and for $\varepsilon_{\text{eff},y}$ and $\varepsilon_{\text{eff},z}$, replace N_x by N_y and N_z , respectively.

3.2.3. Random Mixture

If, on the other hand, all the ellipsoids in the mixture are randomly oriented, there is no longer any macroscopically preferred direction. The mixture is isotropic and the effective permittivity ε_{eff} is a scalar:

$$\varepsilon_{\text{eff}} = \varepsilon_e + \varepsilon_e \frac{\frac{f}{3} \sum_{j=x,y,z} \frac{\varepsilon_i - \varepsilon_e}{\varepsilon_e + N_j(\varepsilon_i - \varepsilon_e)}}{1 - \frac{f}{3} \sum_{j=x,y,z} \frac{N_j(\varepsilon_i - \varepsilon_e)}{\varepsilon_e + N_j(\varepsilon_i - \varepsilon_e)}} \quad (3.28)$$

Figures 3 and 4 illustrate the effective permittivity of mixtures where the inclusions are randomly oriented needles and discs. Shown is again the normalized susceptibility $(\varepsilon_{\text{eff}} - \varepsilon_e)/(\varepsilon_i - \varepsilon_e)$. A comparison with Figure 2 shows that the effective permittivity of mixtures is weakly dependent on the shape of inclusions if the dielectric contrast $\varepsilon_i/\varepsilon_e$ is small, close to one. But when the contrast increases, the macroscopic permittivity is strongly dependent on the inclusion form: spheres give the lowest permittivity, needles a larger permittivity, and discs will provide the largest effect.

It is an interesting detail to note that if the dielectric contrast between the inclusions and the environment are very large, the slope of the curve with spherical inclusions (Fig. 2) tends to zero whereas for randomly oriented needles (Fig. 3) and discs (Fig. 4) the slope is higher: 1/3 for needles and 2/3 for discs. This fact can be connected to the low depolarization factors: (no low depolarization components for a sphere because all three

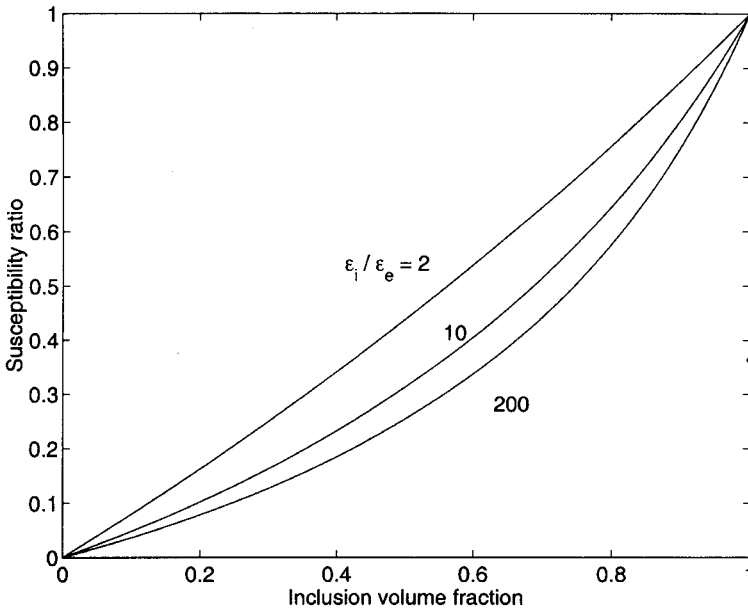


Figure 3. The susceptibility ratio $(\epsilon_{\text{eff}} - \epsilon_e)/(\epsilon_i - \epsilon_e)$ for the Maxwell Garnett prediction of the effective permittivity of a mixture with randomly oriented needle-shaped inclusions of permittivity ϵ_i in a background medium of permittivity ϵ_e .

components are of the value one-third; one of the three depolarization components is low for needles; and two of the three are low for the disc). The connection between the inclusion depolarization and effective permittivity is that a low depolarization means larger internal field in the inclusion which means larger polarization density and consequently a higher dipole moment density in the composite.

Finally, if the inclusions are neither aligned nor randomly oriented but rather follow an orientation distribution, the sums in (3.28) have to be replaced by terms where the dipole moment densities are weighed by the distribution function and integrated over all relevant spatial directions.

3.3. Anisotropic Materials

The aligned ellipsoid case above, Eq. (3.27), was an example where the geometry of the microstructure rendered the macroscopic permittivity anisotropic. How then about the case when the phases are anisotropic to begin with?

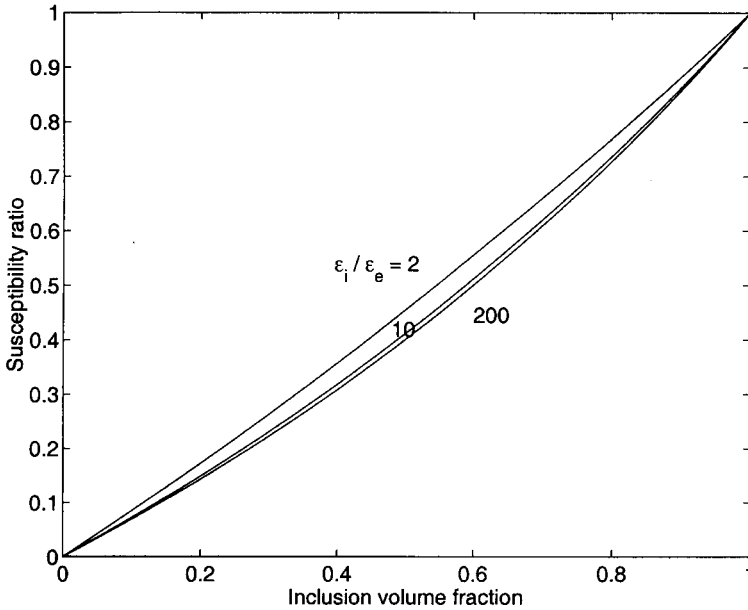


Figure 4. The same as in Figure 3, for a mixture where the inclusions are randomly oriented discs.

It turns out that a mixing rule for the case when the inclusions are anisotropic can be written formally in the same manner as the isotropic Maxwell Garnett formula. Now the permittivities are not scalars but 3×3 matrices, or dyadics, denoted by a double-bar over the symbol. Assume spherical inclusions, characterized by the permittivity dyadic $\bar{\bar{\epsilon}}_i$, embedded in isotropic environment of permittivity ϵ_e . Assume also that all inclusions are similarly oriented in the mixture. Then the Maxwell Garnett rule reads

$$\bar{\bar{\epsilon}}_{\text{eff}} = \epsilon_e \bar{\bar{1}} + 3f\epsilon_e [\bar{\bar{\epsilon}}_i + 2\epsilon_e \bar{\bar{1}} - f(\bar{\bar{\epsilon}}_i - \epsilon_e \bar{\bar{1}})]^{-1} \cdot (\bar{\bar{\epsilon}}_i - \epsilon_e \bar{\bar{1}}) \quad (3.29)$$

where $\bar{\bar{1}}$ is the unit dyadic.* Note also that the division with a scalar of the isotropic Maxwell Garnett rule (2.11) has been replaced by a dyadic inversion.

The case when the environment is anisotropic $\bar{\bar{\epsilon}}_e \neq \epsilon_e \bar{\bar{1}}$ is not as obvious. It turns out [15] that even if the inclusions are spherical, the depolarization characteristics are more complicated than the plain multiplication by

*A unit dyadic is a dyadic with the property that when it operates on any vector (from either side), the result is the same vector.

the factor 1/3 of the isotropic case. The generalization of Eq. (3.29) into the case that the external medium is also anisotropic is

$$\bar{\bar{\epsilon}}_{\text{eff}} = \bar{\bar{\epsilon}}_e + f(\bar{\bar{\epsilon}}_i - \bar{\bar{\epsilon}}_e) \cdot [\bar{\bar{\epsilon}}_e + (1-f)\bar{\bar{N}} \cdot (\bar{\bar{\epsilon}}_i - \bar{\bar{\epsilon}}_e)]^{-1} \cdot \bar{\bar{\epsilon}}_e \quad (3.30)$$

where the dyadic $\bar{\bar{N}}$ is dictated by the anisotropy of the environment:

$$\bar{\bar{N}} = \frac{1}{2} \int_0^\infty ds \frac{(s\bar{\bar{I}} + \bar{\bar{\epsilon}}_r^{-1})^{-1}}{\sqrt{\det(s\bar{\bar{\epsilon}}_r + \bar{\bar{I}})}} \quad (3.31)$$

and $\bar{\bar{\epsilon}}_r = \bar{\bar{\epsilon}}_e/\epsilon_0$ is the relative permittivity dyadic of the environment, ϵ_0 being the vacuum permittivity. The determinant (det) of a symmetric dyadic is the product of its three eigenvalues.

One could interpret (3.30) by saying that the anisotropy of the environment has “squeezed” the sphere into an ellipsoid whose depolarization factors are the components of the dyadic (3.31).

3.4. Inclusion Substructure

The analysis so far has been restricted to discrete mixtures. The inclusions are assumed to be dielectrically homogenous. The problem with non-homogeneous scatterers is again the difficulty of calculating the polarizability of such inclusions. There are, however, certain shapes for which analytical solution can be found in the electrostatic problem.

A layered sphere is one example of such a special case. There is no restriction for the number of layers, and even the case for a radially continuous permittivity profile of the sphere has been given solution [16].* As an example, the following is the generalization of the mixing rule (2.10) for the case when the inclusions are two-component spheres:

$$\frac{\epsilon_{\text{eff}} - \epsilon_e}{\epsilon_{\text{eff}} + 2\epsilon_e} = f \frac{(\epsilon_l - \epsilon_e)(\epsilon_c + 2\epsilon_l) + w(\epsilon_c - \epsilon_l)(\epsilon_e + 2\epsilon_l)}{(\epsilon_l + 2\epsilon_e)(\epsilon_c + 2\epsilon_l) + 2w(\epsilon_c - \epsilon_l)(\epsilon_l - \epsilon_e)} \quad (3.32)$$

The inclusion sphere consists of a spherical core with permittivity ϵ_c which is covered by a spherical shell with permittivity ϵ_l . The parameter $w = (b/a)^3$ is the fraction of the volume of the core from the total inclusion volume (a is the radius of the inclusion and b is the radius of the core), and f is the volume fraction of the inclusions in the mixture, as before.

3.5. Lossy Materials

In the above analysis, it has been implicitly assumed that all permittivities are real numbers. Losses have not been addressed. The mixing rules

*See also the results for mixtures with dielectrically inhomogeneous ellipsoids [17].

that result from the real analysis can, however, be used in the complex domain with certain restrictions. What are these limitations?

The answer to this loss problem depends on the structure of the mixture, the size and form of the inclusions, and the loss mechanism itself. If the loss mechanism is the same in bulk material as in the case when this material is in the form of small particles, and the internal field calculation inherent in the derivation of the mixing formula is valid also for the lossy case, then also the imaginary part of the mixing formula correctly gives the mixture losses. This is the case in, for example, mixtures containing water, at microwave frequencies: the losses are of polar origin, and are due to the reaction of the polar water molecule to the incident field. As long as mixtures are concerned in which the inclusion dimensions are large compared with the molecular dimensions, the mixture loss calculation requires no special attention compared with the losses of homogeneous water.

It is worth noting that with the assumptions above, the mixing formula also takes into account the losses in the case that the inclusion phase is conducting. Consider a dielectric mixture where the background material is of permittivity ϵ_e and the inclusion phase has conductivity σ_i , and its complex permittivity is $\epsilon_i = \epsilon'_i - j\sigma_i/\omega$, ω being the angular frequency. If the volume fraction of the inclusion phase f is small, the effective conductivity $\sigma_{\text{eff}} = \omega \text{Im}\{\epsilon_{\text{eff}}\}$ of the mixture, calculated from the Maxwell Garnett formula, is

$$\sigma_{\text{eff}} = \frac{9\epsilon_e^2 f \sigma_i}{(\epsilon'_i + 2\epsilon_e)^2 + \sigma_i^2/\omega^2} \quad (3.33)$$

From this formula it is seen that the effective DC conductivity vanishes: $\sigma_{\text{eff}} \rightarrow 0$ as $\omega \rightarrow 0$. This is also intuitively clear: non-contacting conducting particles in a non-conducting matrix do not make the mixture conducting.

Equation (3.33) shows that the loss factor and the imaginary part of the permittivity, that varies as ω^{-1} for the conductive inclusion material, is converted to the so called Maxwell–Wagner losses of the mixture (see for example [18]): losslessness in the low-frequency region, and normal conducting losses at high frequencies. The imaginary part of the effective permittivity has its maximum value at the frequency

$$\frac{\omega}{2\pi} = \frac{1}{2\pi} \frac{\sigma_i}{\epsilon'_i + 2\epsilon_e} \quad (3.34)$$

This frequency is around 200 kHz for water with conductivity 1 mS/m.

Troubles in applying mixture formulas arise when the internal field of the inclusions, due to losses, is not calculable from the permittivity ratios as was done in the previous sections. This is the case for mixtures where the

inclusions are large compared to the penetration depth of the field into the inclusions. Inside lossy inclusions, the field decays exponentially, and quick decay is not compatible with the algebraic calculation of the inside field. Consequently the mixing formula predicts too large internal field, and gives too large losses for the mixture, for example, in the cases of metal spheres with radius considerably larger than the skin depth $\sqrt{2/\omega\mu\sigma}$, where μ is the magnetic permeability of the material.

Although the previous analysis has focused on the dielectric mixing problem, the solutions can be extended to other branches of physics. The same mathematical formulation as in the dielectric case will be encountered in the conduction problem, both in the transfer of electricity and heat, the magnetostatic permeability problem, and even in certain mechanical problems of elasticity. The mathematically equivalent character of these problems guarantees that the result from the effective permittivity can be directly used for calculating, for example, the effective magnetic permeability. One only needs [19, Section 4.2] to make the change $\epsilon \rightarrow \mu$ in all instances of a mixing formula. Concerning mixtures that consist of more complex materials, like chiral and bi-anisotropic media, the analysis is greatly rationalized by the use of six-vector algebra [20], which combines the electric and magnetic vector quantities into a supervector of six components.

4. Generalized Mixing Models

There is no exact result for the effective permittivity of a mixture with random geometry. In the analysis of random media, a major difficulty comes with the problem how to correctly take into account the interaction between the scatterers. For sparse mixtures, these effects of interaction are small and can be included by surrounding the inclusion with the average polarization $\langle \mathbf{P} \rangle$ as was done in the above derivation of the Maxwell Garnett rule. However, when dense mixtures are treated, this approach may not be correct. In the present section, mixing rules are presented for mixtures which predict different results compared to the Maxwell Garnett rule.

4.1. Polder–van Santen Formula

The important mixing rule which in the remote sensing community goes under the name *Polder–van Santen formula* [21] deserves special attention. In theoretical electromagnetics research, this formula is called as *Bruggeman formula* [22], and also the name *Böttcher formula* [23] can be found in the literature.

The basic form of this formula for spherical scatterers is

$$(1-f) \frac{\epsilon_e - \epsilon_{\text{eff}}}{\epsilon_e + 2\epsilon_{\text{eff}}} + f \frac{\epsilon_i - \epsilon_{\text{eff}}}{\epsilon_i + 2\epsilon_{\text{eff}}} = 0 \quad (4.35)$$

This Polder–van Santen rule formula has the special property that it treats the inclusions and the environment symmetrically. In fact, there is no difference between the two phases. The interpretation of Eq. (4.35) is that the formula balances both mixing components with respect to the unknown effective medium, using the volume fraction of each component as weight (f for the inclusions and $1-f$ for the environment). This symmetry property of (4.35) makes the radical distinction between the Maxwell Garnett rule and the Polder–van Santen rule. The Maxwell Garnett approach is inherently non-symmetric.

The Polder–van Santen formula for the case where the inclusions are randomly oriented ellipsoids is

$$\epsilon_{\text{eff}} = \epsilon_e + \frac{f}{3} (\epsilon_i - \epsilon_e) \sum_{j=x,y,z} \frac{\epsilon_{\text{eff}}}{\epsilon_{\text{eff}} + N_j (\epsilon_i - \epsilon_{\text{eff}})} \quad (4.36)$$

where now N_j are again the depolarization factors of the inclusion ellipsoids in the three orthogonal directions.

4.2. Coherent Potential Formula

Another well-known formula which is relevant in the theoretical studies of wave propagation in random media is the so-called *Coherent potential formula* [24, p. 475]:

$$\epsilon_{\text{eff}} = \epsilon_e + \frac{f}{3} (\epsilon_i - \epsilon_e) \sum_{j=x,y,z} \frac{(1+N_j)\epsilon_{\text{eff}} - N_j\epsilon_e}{\epsilon_{\text{eff}} + N_j(\epsilon_i - \epsilon_e)} \quad (4.37)$$

This formula for spherical inclusions is

$$\epsilon_{\text{eff}} = \epsilon_e + f(\epsilon_i - \epsilon_e) \frac{3\epsilon_{\text{eff}}}{3\epsilon_{\text{eff}} + (1-f)(\epsilon_i - \epsilon_e)} \quad (4.38)$$

It is worth noting that for dilute mixtures ($f \ll 1$), all three mixing rules, Maxwell Garnett, Polder–van Santen, and Coherent potential, predict the same results. Up to the first order in f , the formulas are the same:

$$\epsilon_{\text{eff}} \approx \epsilon_e + 3f\epsilon_e \frac{\epsilon_i - \epsilon_e}{\epsilon_i + 2\epsilon_e} \quad (4.39)$$

4.3. Unified Mixing Formula

A unified mixing approach [25] collects all the previous aspects of dielectric mixing rules into one family. For the case of isotropic spherical inclusions ε_i in the isotropic environment ε_e , the formula looks like

$$\frac{\varepsilon_{\text{eff}} - \varepsilon_e}{\varepsilon_{\text{eff}} + 2\varepsilon_e + \nu(\varepsilon_{\text{eff}} - \varepsilon_e)} = f \frac{\varepsilon_i - \varepsilon_e}{\varepsilon_i + 2\varepsilon_e + \nu(\varepsilon_{\text{eff}} - \varepsilon_e)} \quad (4.40)$$

This formula contains a dimensionless parameter ν . For different choices of ν , the previous mixing rules are recovered: $\nu = 0$ gives the Maxwell Garnett rule, $\nu = 2$ gives the Polder–van Santen formula, and $\nu = 3$ gives the Coherent potential approximation.

4.4. Other Mixing Models

In practical remote sensing applications and random medium theories, a very large set of other mixing rules are being used. Of these, let us mention the following ones.

A widely used class of mixing models is formed by the “power-law” approximations:

$$\varepsilon_{\text{eff}}^a = f\varepsilon_i^a + (1-f)\varepsilon_e^a \quad (4.41)$$

For example, the Birchak formula [26] the parameter is $a = 1/2$, which means that the square roots of the component permittivities add up to the square root of the mixture permittivity.

Another famous formula is the Looyenga formula [27] for which $a = 1/3$. One can also find in the literature (see for example [28, p. 1080]) the linear law

$$\varepsilon_{\text{eff}} = f\varepsilon_i + (1-f)\varepsilon_e \quad (4.42)$$

which corresponds to $a = 1$ in (4.41). This mixing rule can be given theoretical configuration if the mixture is formed of plates or other inclusions for which no depolarization is induced. If the depolarization factor is $N_x = 0$, one can recover formula (4.42) from (3.27).

Other models resulting from a differential analysis are

$$\frac{\varepsilon_i - \varepsilon_{\text{eff}}}{\varepsilon_i - \varepsilon_e} = (1-f) \left(\frac{\varepsilon_{\text{eff}}}{\varepsilon_e} \right)^{1/3} \quad (4.43)$$

and its “complement” [29]

$$\frac{\varepsilon_{\text{eff}} - \varepsilon_e}{\varepsilon_i - \varepsilon_e} = f \left(\frac{\varepsilon_{\text{eff}}}{\varepsilon_i} \right)^{1/3} \quad (4.44)$$

which have the common feature of one-third powers.

There are also formulas for mixtures with spherical inclusions in a cubic array in a background matrix. These formulas can be seen as successive improvements to the classical Rayleigh result, Eq. (2.10). These have been presented by Runge [30], Meredith and Tobias [31], McPhedran, McKenzie and Derrick [32,33], Doyle [34], and by Lam [35]. However, these formulas are derived for ordered mixtures, though not all necessarily for cubic-centered lattices, and from the point of view of application to random media, they suffer from the disadvantages of predicting infinite effective permittivities as the inclusions come to contact with each other.

5. Consequences of Mixing Formulas for Moist and High-Loss Materials

The mixing rules that have been discussed so far are actually rather simple algebraic formulas that combine the component permittivities and some structural parameters of the mixture. It is actually amazing how many real-life phenomena are predicted by these rules. The mixing process sometimes gives rise to qualitatively new effects such that the mixture displays properties that are absent in the component materials. These phenomena are especially prominent when materials are moist, in other words, one of the components in the mixture is water. This is because of the strong dispersion in the permittivity of water on one hand, and the fact that water permittivity has very high values in the microwave frequencies on the other. The present section introduces some interesting effects in this vein.

5.1. Special Effects for High-Loss Mixtures

Although Figures 2–4 might leave the reader with the impression that the effective permittivity of a composite is a rather soft average of the permittivities of the components, the situation is in fact complicated to a great extent in cases when one (or several) of the components composing the mixture is highly lossy. This fact is seen clearly in the following example.

Let us treat the basic Maxwell Garnett mixing model for a medium where lossy spherical inclusions occupy the volume fraction f in vacuum: the relative permittivity of the inclusions is $3-j10$ and the relative permittivity of the environment is unity. Then the real and imaginary parts of the effective permittivity, as a function of f , can be seen to follow the curves in Figure 5. Compared with the earlier results (Figs. 2–4) where the effective permittivity increased monotonously from the environment value to the inclusion permittivity value, the lossy case is clearly different. Although here the imaginary part of the effective permittivity behaves in an expected manner, the real part attains a maximum at about 80% volume fraction. In other

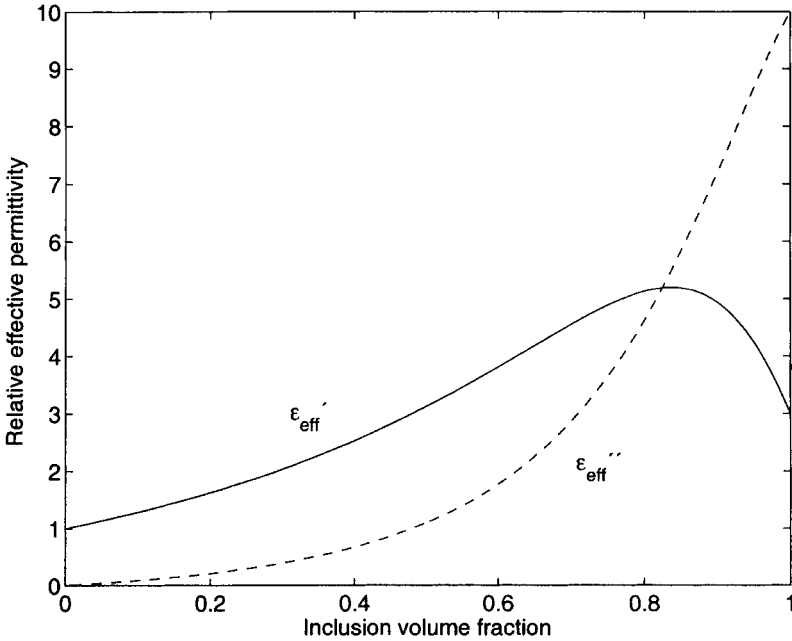


Figure 5. The real and imaginary parts of a mixture where highly lossy inclusions are embedded in lossless background medium. The permittivity of the inclusions is $\epsilon_i/\epsilon_e = 3 - j10$. The solid line is for the real part of the effective permittivity, and the dashed line for the imaginary part.

words, the real part of the effective permittivity may be higher than that of the components.

The enhancement phenomenon that Figure 5 illustrates is relevant in microwave aquametry and modeling of the effective properties of geophysical and moist substances. Depending on the shape of the water phase components in the mixture, the macroscopic dielectric response of matter may be very strong.

5.2. Frequency Dependence and Dispersion

Hidden inside the material permittivity ϵ there are many physical polarization mechanisms. A more correct description of the constitutive relation—than a plain dielectric “constant”—between the displacement and the electric field presents the dependence as a convolution operator:

$$\mathbf{D}(t) = \epsilon_0 \mathbf{E}(t) + \int_{-\infty}^t \chi(t - t') \mathbf{E}(t') dt' \tag{5.45}$$

where the first term is the free-space response which contains the vacuum permittivity ϵ_0 , and the material response is described by the susceptibility kernel $\chi(t)$, which, due to causality requirements, vanishes for negative arguments. In engineering applications, it is more convenient to work in the frequency-domain which means that the explicit time-dependence has been Fourier-transformed, and the permittivity is a function of frequency ω . The variation of the permittivity with respect to frequency is called dispersion.*

Different physical mechanisms in the dielectric response of materials can be described by certain dispersion models. Some important ones for remote sensing applications are

- the *Debye model*, which is suited for fluids and other materials that contain permanent electric dipole moments. A characteristic parameter for Debye materials is the relaxation time of the moments. This time determines the relaxation frequency, and radio waves of that frequency will be attenuated in the medium.
- for solid materials, the *Lorentz model* describes the resonant frequencies and damping for characteristic frequencies of the molecules of the medium. Around the resonance, a very strong dispersion takes place.
- for metals, the *Drude model* can be used to model their free-electron type behavior.

The mixing process can affect strongly the dispersive characteristics of materials. For example, if metal spheres are mixed in non-dispersive environment, the mixture becomes a Lorentz-medium with resonant behavior. Another interesting example is rain as a mixture of water drops in air. Water obeys Debye law, and the air permittivity is constant. It turns out that rain follows Debye-model, too, but the relaxation frequency will be shifted to a frequency far higher than that of water [36]. The features of the mixture can be quite accurately calculated with a simple Maxwell Garnett mixing rule [37]. Also more general mixing models have been applied to study the dispersion of heterogeneous substances [38].

5.3. Polarization Enhancement and Percolation

It is often that electromagnetic scattering analyses are interpreted with a tendency to underappreciate the richness of the dielectric response of small particles. In quasistatic modeling of mixtures, the dynamic nature of

*More accurately, the frequency dependence of ϵ is a manifestation of *temporal* dispersion. One has to bear in mind that media can also be spatially dispersive, for which case the permittivity is a function of the wavevector \mathbf{k} .

Maxwell equations is suppressed and one may think that the result is an uninteresting average of the component materials. But this is not the case as has already been pointed out above, and which is very convincingly argued in [39, Chap. 12].

The frequency dependence of the effective permittivity can be affected in a very nonlinear manner if the inclusion permittivity is dispersive, as can be observed from the mixing rule (2.11). Especially in frequency ranges (like in the optical regime) where negative real parts for the permittivity can be measured, it may happen that the denominator reaches very small absolute values.* The result is a strong absorption which explains the known fact that certain materials emit different colors in particulate form compared to the same material in bulk form.

A further effect which is predicted by dielectric mixing rules is the property called *percolation*. Percolation is a very nonlinear phenomenon. It is an abrupt change in the behavior of a certain parameter in the random medium as the volume fraction of the phases changes. Percolation theories have been developed in diverse fields, like ferromagnetism, forest fires, and even the spread of epidemics in animal and human populations [40].

It turns out that percolation behavior becomes pronounced if the mixture consists of components which have a strong contrast in their dielectric properties. Therefore one can neglect percolation discussion in dry snow modeling, but if the snow is wet, percolation effects may become important. In general, moist substances are more difficult to model as mixtures, and one of the reasons is actually the strong dielectric contrast between the permittivity of water and that of ordinary dry materials.

Mixing formulas predict percolation but the threshold volume fraction is different depending on the mixing rule. It turns out [41] that for the case of (three-dimensional) spherical high-permittivity inclusions, the percolation threshold (the volume fraction of the inclusion phase around which the sudden increase in the mixture permittivity ϵ_{eff} takes place) is 0.25 for the Coherent potential formula, 0.333 for the Polder–van Santen formula, and 1.0 for the Maxwell Garnett rule.

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*For dilute mixtures ($f \ll 1$), the condition $\epsilon_i = -2\epsilon_e$ gives this type of “catastrophe.”

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