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Effective permittivity of layered dielectric sphere composites

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A straightforward model is presented for analysing the effective permittivities of layered dielectric sphere composites. Using the present model, the effective permittivity, ε_{eff}, of layered dielectric sphere composites can be deduced using classical two-phase dielectric mixture formulae in two steps: first, the effective permittivity, $\varepsilon_{\text{incl}}$, of the inclusions is calculated by taking the layered dielectric sphere inclusions as sub-composites; and second, the effective permittivity, ε_{eff}, of the composites is found by substituting the layered dielectric sphere inclusions with homogeneous spheres whose permittivity is equal to $\varepsilon_{\text{incl}}$. The present model is applicable to multi-layer sphere composites. Experiments on resin-based hollow bead composites show that the present model accurately predicts the effective permittivity of layered dielectric sphere composites.

1. Introduction

For composite materials consisting of two homogeneous phases, host medium and inclusion, various dielectric mixture formulae have been presented in the research literature over the years [1–6]. Different formulae are derived using different approximations of the microstructure details of the composite, and the success of a mixture formula relies on its accuracy in the description of the real microstructure details. Several popular formulae for the effective permittivity, ε_{eff}, of two-phase non-polar dielectric mixtures with a host medium of permittivity ε_0, and spherical inclusion of permittivity ε_1 with volume fraction f_1, are given below.

Rayleigh's formula [4]

$$\frac{\varepsilon_{eff} - \varepsilon_0}{\varepsilon_{eff} + 2\varepsilon_0} = f_1 \frac{\varepsilon_1 - \varepsilon_0}{\varepsilon_1 + 2\varepsilon_0} \tag{1}$$

Looyenga's formula [5]

$$\varepsilon_{eff}^{1/3} = f_1 \varepsilon_1^{1/3} + (1 - f_1)\varepsilon_0^{1/3} \tag{2}$$

Beer's formula [6]

$$\varepsilon_{eff}^{1/2} = f_1 \varepsilon_1^{1/2} + (1 - f_1)\varepsilon_0^{1/2} \tag{3}$$

In the above formulae, the inter-particle actions between the inclusions are neglected. Although these formulae can be extended to multi-phase composites, they are not applicable to the composites with layered inclusions, because they ignore the correlation between different layers in an inclusion.

Recently, Sihvola *et al.* [7–10] proposed an approach to calculate the polarization, α, and dipole moment of a layered sphere inclusion by using Lorentz dielectric field concepts and finding a solution for Laplace's equation in spherical coordinates. The value of α obtained can then be substituted into the Clausius–Mossotti relation. Using this method, Sihvola obtained generalized formulae for layered sphere composites (Equation 16 in [8]). This method has also been successfully applied by Liu and Wilcox [11] to a porous composite composed of thin-walled hollow ceramic spheres. However, the mathematics involved in this approach is quite complicated.

In this paper, we propose a straightforward model based on classical two-phase mixture formulae to predict the effective permittivity of composites with layered inclusions. The present model has been compared with the precisely defined Sihvola's model, and verified by experiments on composite samples loaded with hollow microspheres.

2. Effective permittivity of layered dielectric sphere composites

In the two-layer dielectric sphere composite shown in Fig. 1, the permittivity of the host medium is ε_0; the permittivity and volume fraction of the shell are ε_1 and f_1, respectively; and the permittivity and volume fraction of the core are ε_2 and f_2 respectively. Here, we propose to calculate the effective permittivity, ε_{eff}, of

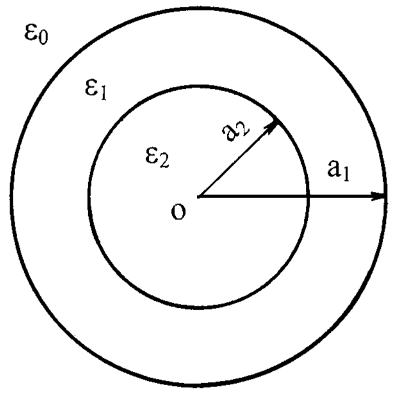

Figure 1 A two-layer dielectric sphere embedded in a host media. The two-layer sphere consists of a shell and a core. The shell is the space between the two sphere surfaces having the same center O, and with radii a_1 and a_2 respectively; and the core is the space enclosed by the sphere surface with radius a_2.

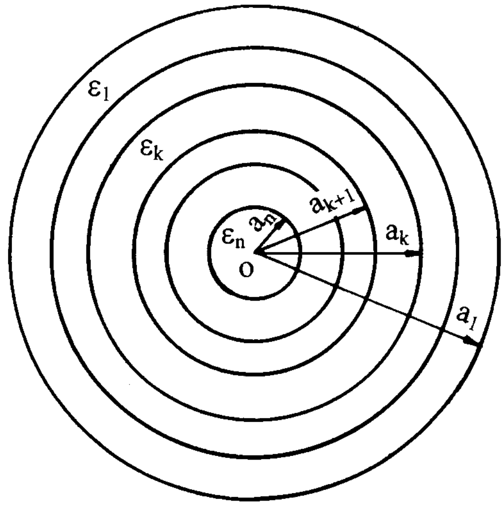

Figure 2 A dielectric sphere consisting of N layers.

the layered dielectric sphere composite in two steps: (i) by transforming the layered dielectric sphere into a homogeneous sphere of dielectric constant $\varepsilon_{\text{incl}}$ and radius a_1; and (ii) by approximating the layered dielectric sphere composite to a composite loaded with dielectric spheres of radius a_1 and dielectric constant $\varepsilon_{\text{incl}}$, and by calculating the effective permittivity, ε_{eff}, of the composite using a suitable two-phase dielectric mixture formula. The justifications of our approach are: (i) if the concept of effective permittivity stands for a composite, the wavelength, λ, of the operating electromagnetic field is much larger than the sizes of the embedded inclusions and the inner radius of the inclusions; therefore, the concept of effective permittivity still stands if we regard the inclusion as a sub-composite by taking its shell as the host medium, and its core as the inclusion; and (ii) with the same volume and shape, under the same electromagnetic field, a dielectric composite (whose effective permittivity is ε_{eff}) and a homogenous dielectric material (whose permittivity is equal to ε_{eff}) have the same polarization.

The effective permittivity of the inclusion can be derived from the model shown in Fig. 1. If the composite shown in Fig. 1 is placed in a homogeneous electric field, the potential can be calculated with the aid of the general solution of Laplace's equation. The electric field in the host media keeps the same, if we replace the layered sphere with a homogeneous sphere with radius a_1 and a permittivity $\varepsilon_{\text{incl}}$ [5]

$$\varepsilon_{incl} = \frac{1-2G}{1+G}\varepsilon_1 \tag{4a}$$

where

$$G = \frac{\varepsilon_1-\varepsilon_2}{2\varepsilon_1+\varepsilon_2}\left(\frac{a_2}{a_1}\right)^3 = \left(\frac{\varepsilon_1-\varepsilon_2}{2\varepsilon_1+\varepsilon_2}\right)\left(\frac{f_2}{f_1+f_2}\right) \tag{4b}$$

Equation 4 shows that $\varepsilon_{\text{incl}}$ is totally determined by a_1, a_2, ε_1 and ε_2, and is independent of ε_0.

The value of $\varepsilon_{\text{incl}}$ obtained can then be used to calculate the effective permittivity, ε_{eff}, of the composite using the Rayleigh mixture formula, Equation 1, by replacing the volume fraction as $(f_1 + f_2)$

$$\frac{\varepsilon_{eff}-\varepsilon_0}{\varepsilon_{eff}+2\varepsilon_0} = (f_1+f_2)\left(\frac{\varepsilon_{incl}-\varepsilon_0}{\varepsilon_{incl}+2\varepsilon_0}\right) \tag{5}$$

The present model can be easily extended to multi-layer dielectric sphere composites. In the calculation of the effective permittivity, $\varepsilon_{\text{incl}}$, of the multi-layer dielectric sphere, we should perform it layer by layer, starting with the innermost to the outermost. In Fig. 2, ε_k denotes the permittivity of the shell between the two sphere surfaces with radii a_k and a_{k+1}, and $\varepsilon_{\text{incl},k}$ denotes the effective permittivity of the layered sphere enclosed by the sphere surface of radius a_k. It is obvious that: $\varepsilon_{incl,n} = \varepsilon_n$ and $\varepsilon_{incl} = \varepsilon_{incl,1}$. By extending Equation 4 $\varepsilon_{\text{incl},k}$ can be deduced from $\varepsilon_{\text{incl},k+1}$ through

$$\varepsilon_{incl,k} = \frac{1-2G_k}{1+G_k}\varepsilon_k \tag{6a}$$

where

$$G_k = \frac{\varepsilon_k-\varepsilon_{incl,k+1}}{2\varepsilon_k+\varepsilon_{incl,k+1}}\left(\frac{a_{k+1}}{a_k}\right)^3 \tag{6b}$$

and $k = n-1, n-2, \ldots, 2, 1$. Step by step, we can finally get $\varepsilon_{\text{incl},1}$ from $\varepsilon_{\text{incl},2}$ through

$$\varepsilon_{incl} = \varepsilon_{incl,1} = \frac{1-2G_1}{1+G_1}\varepsilon_1 \tag{7a}$$

where

$$G_1 = \frac{\varepsilon_1-\varepsilon_{incl,2}}{2\varepsilon_1+\varepsilon_{incl,2}}\left(\frac{a_2}{a_1}\right)^3 \tag{7b}$$

After the value of $\varepsilon_{\text{incl}}$ is obtained, the effective permittivity, ε_{eff}, of the multi-layer dielectric sphere composite can then be deduced by using Rayleigh's formula.

3. Experimental procedure

Experiments have been made to verify the present model. We select resin-based hollow ceramics bead composites as our model samples.

3.1. Preparation of the samples

Our samples were prepared by using hollow beads, Microcells SL 300 (Microcell Australia Pty. Ltd.), as inclusions. The hollow beads were made from modified mullite with permittivity of 6.00. The sizes of the hollow beads were between 150 and 300 μm. The volume fraction of the shell and core were calculated from the density of the hollow beads and the density of modified mullite. The epoxy resin and the hardener used were LY5052 and HY5052 (Ciba Geigi), respectively. The viscosity of the mixture of epoxy resin and hollow beads could be further adjusted by either using a few percentage of MIBK (a thinner with boiling point at 115 °C), in case where the viscosity is too high, or adding less than 1% fumed silica powder in order to increase the viscosity to prevent the hollow beads from settling when the viscosity is too low. The mixture was then degassed in vacuum and cured into a composite in a silicone rubber mould with an inner diameter of 4 cm and height of 2.5 cm at room temperature for 24 h. The moulded composite was then ground with sandpaper to produce a flat and smooth surface for measurement.

In the sample preparation, the ratio of the epoxy resin LY5052 (100 parts) and the hardener HY5052 (38 parts) was kept the same for all samples. By measuring the permittivity of a sample without any inclusions, it was found that $\varepsilon_0 = 3.42$ at 1.0 GHz. The thinner MIBK had been evaporated after the mixture was cured into the composite, and thus the effect of MIBK on the effective permittivity of the samples was ignored. As the amount of silica powder added to the samples was very small, and the permittivity of the silica powder was about 3.8, which is close to ε_0, the effect of the silica powders on the effective permittivity of the samples was ignored in our calculation.

In our experiments, four groups of samples were made to study the effects of the sizes of the inclusions. The size ranges of the hollow beads for the four groups of samples were (1) 150–200 μm, (2) 200–250 μm, (3) 250–300 μm, and (4) 150–300 μm. In each group, there were five samples with different inclusion volume fractions.

3.2. Measurement of the effective permittivity

The measurement of the effective permittivity of composites was carried out using a microwave reflection method as shown in Fig. 3. The value of the permittivity of the sample was obtained from the reflection signal (the scattering parameter $S11$) at a defined plane. This is a non-destructive method, but requires good contact between the probe and the sample. In our experiment, a coaxial dielectric probe was used to measure the permittivity. A vacuum pump was used to pump out the air gap between the probe and the sample, so that the sample and the probe had very good contact [12]. By this technique, more reliable and accurate results could be obtained. Our experiments were carried out on a Vector Network Analyzer (HP 8719C) at 1.0 GHz. The values of permittivity were calculated from $S11$, which were collected and processed by a computer connected to the Vector Network Analyzer (HP 8719C) through an IEEE-488 data bus.

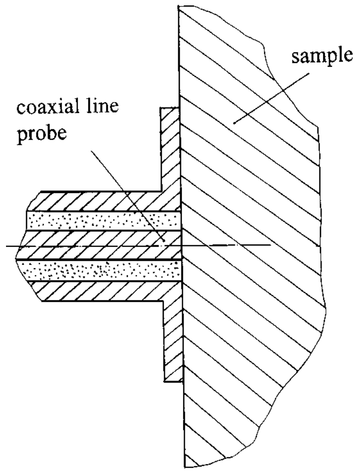

Figure 3 Measurement of dielectric permittivity using a coaxial line probe.

4. Results and discussion

The measurement results of the resin-based samples are listed in Table I, and are compared with different models, including uncorrelated model, Sihvola's model and the present model. In the uncorrelated model, we take the two-layer sphere composites as three-phase composites: host media, and two inclusions (shell and core of the two-layer spheres). Neglecting the correlation between the shell and core, we use the three-phase Rayleigh's formula, which depicts

$$\frac{\varepsilon_{eff} - \varepsilon_0}{\varepsilon_{eff} + 2\varepsilon_0} = f_1 \frac{\varepsilon_1 - \varepsilon_0}{\varepsilon_1 + 2\varepsilon_0} + f_2 \frac{\varepsilon_2 - \varepsilon_0}{\varepsilon_2 + 2\varepsilon_0} \quad (8)$$

where ε_0 is the permittivity of the host medium; ε_1 and ε_2 are the permittivity values of the two inclusions, and f_1 and f_2 are the volume fractions of the two inclusions, respectively. Because Equation 8 does not concern the correlation between the two inclusions (the shell and the core), it cannot give accurate prediction of the permittivity of the layered dielectric composites, as reported in Table I.

In Sihvola's model, we use Equation 25 from Sihvola and Lindell [8]. To compare the calculated results from the different models, five decimal digits are listed for each theoretical calculation result. We find that the present model and the precisely defined Sihvola's model give the same results. Table I shows that the predictions according to the present model and Sihvola's model fit the experimental results very well.

It should be noted that in the present model, f_1 and f_2 are average values, and they do not convey any information about the sizes of the inclusions. In Table I, there are four groups of samples with different size ranges

TABLE I Comparison of the measurement results and different mixture models. In our calculations, $\varepsilon_0 = 3.42$, $\varepsilon_1 = 6.00$, $\varepsilon_2 = 1.00$. Our measurements were carried out at 1.0 GHz

Size range of the inclusions (μm)	f_1	f_2	ε_{incl}	Uncorrelated model (Eq. 8)	Sihvola's model (Eq. 25 in [8])	Present model (Eq. 5)	Measurement results
150–200	0.146	0.523	1.8389	2.2228	2.3054	2.3054	2.32
	0.133	0.474	1.8425	2.3241	2.4009	2.4009	2.37
	0.124	0.445	1.8377	2.3829	2.4555	2.4555	2.48
	0.117	0.417	1.8425	2.4433	2.5126	2.5126	2.53
	0.108	0.386	1.8405	2.5090	2.5738	2.5738	2.55
200–250	0.139	0.503	1.8319	2.2611	2.3406	2.3406	2.34
	0.130	0.471	1.8310	2.3267	2.4021	2.4021	2.38
	0.117	0.420	1.8375	2.4356	2.5048	2.5048	2.53
	0.104	0.375	1.8344	2.5311	2.5940	2.5940	2.57
	0.093	0.334	1.8371	2.6214	2.6786	2.6786	2.71
250–300	0.121	0.438	1.8317	2.3959	2.4671	2.4671	2.44
	0.108	0.392	1.8298	2.4932	2.5581	2.5581	2.57
	0.092	0.331	1.8359	2.6277	2.6844	2.6844	2.69
	0.085	0.307	1.8332	2.6804	2.7334	2.7334	2.76
	0.073	0.266	1.8271	2.7718	2.8182	2.8182	2.84
150–300	0.137	0.494	1.8343	2.2803	2.3589	2.3589	2.40
	0.118	0.428	1.8303	2.4166	2.4863	2.4863	2.53
	0.110	0.395	1.8372	2.4888	2.5546	2.5546	2.54
	0.101	0.364	1.8347	2.5551	2.6164	2.6164	2.59
	0.085	0.308	1.8310	2.6777	2.7307	2.7307	2.77

of the inclusions. Experiments show that the present model is applicable to composites whose inclusions are of varying sizes.

5. Conclusion

The effective permittivity of layered dielectric sphere composites can be calculated by replacing the actual inclusions with equivalent homogeneous sphere inclusions whose permittivity is equal to the effective permittivity of the actual inclusions. The inclusions in the composite may be of varying sizes. Although, in this paper, attention is focused on two-layer sphere composites, the present model is applicable to multi-layer sphere composites.

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