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The electrical properties of heterogeneous mixtures containing an oriented spheroidal dispersed phase

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Abstract: Theoretical equations for the permittivity and conductivity of a heterogeneous mixture containing an oriented dispersed phase of spheroidal shape are limited to low volume fractions of dispersed phase. Two new sets of equations have been derived here for such systems using approaches which have been shown previously to be applicable to higher concentrations. One set has generalised forms of equations given by Bruggeman and Hanai and includes simplifications applicable to oil in water (O/W) and water in oil (W/O) systems. The other set has generalised forms of an equation given by Looyenga.

Tabulated mixture permittivity ε and dielectric increment $\Delta\varepsilon$ values are presented and, with the exception of $O\langle W$ systems containing prolate spheroids, the two types of equation show significant differences in both ε and $\Delta\varepsilon$. For both types $\Delta\varepsilon$ is strongly dependent on the ratio of the conductivities of the two media of a W/O system.

The relevance of the mixture equations to the electrical behaviour of micellar, microemulsion and liquid crystalline systems is considered.

Key words:

Introduction

There are many equations available for determining permittivity and conductivity values for heterogeneous mixtures containing a dispersed phase of spherical shape; these have been reviewed by van Beek [1] and Hanai [2]. Most formulae are only applicable to low volume fractions of dispersed phase. The equations of Bruggeman [3] and Looyenga [4], which are extensions of formulae applicable to low volume fractions up to higher concentrations, by a successive incrementation method, have been shown to be among the best for predicting permittivity values.

In conductivity studies, the equations of Hanai [5], which are an extension of the Bruggeman equation to the more general case, in which the conductivity component is significant, are favoured [5–7]. These Hanai-Bruggeman equations do not enable the complex permittivity to be found explicitly, but the high and low frequency limiting values for both permittivity and conductivity in a Maxwell-Wagner type dispersion [8, 9] can be determined.

As well as being applicable to mixtures in which the dispersed component is of macroscopic size, it has been shown that heterogenous mixture equations can be usefully applied to systems containing a much smaller sized dispersed component, such as in micellar and microemulsion systems [10–13]. Further, it has been shown that in some of these systems, the dispersed phase is of a spheroidal rather than spherical shape [10, 14, 15]. In these and many other fluid systems, the dispersed phase will have rotational mobility and will thus, as pointed out by Schwarz et al. [16], be oriented by an applied electric field.

Ther are relatively few equations for oriented spheroids, with those due to Sillars [17], Mandel [18] and van Beek [1] for permittivity and Fricke [19] for conductivity being the most commonly used. However, these equations are of limited use in that they are only applicable to low volume fractions of dispersed phase [1, 2, 17]. A more satisfactory situation would be to have mixture equations for spheroidal shape based on the more widely applicable ones of Bruggeman, Hanai and Looyenga.

While Reynolds and Hough [20] have extended the Bruggeman equation for the case of randomly oriented spheroids and this has been further extended by Boned and Peyrelasse [21] to include conductivity behaviour, the equations are unwieldy and do not directly yield permittivity and conductivity values except in a few special cases. As the concentration of dispersed phase is increased, orientation becomes progressively more probable on packing grounds, particularly at larger deviations from spherical shape [22]. Thus there are benefits in the generation of equations which are easy to apply to both directed and randomly oriented shapes in both mobile and immobile systems.

It has been claimed that the Looyenga equation is in principle applicable to all spheroidal shapes [4]. This is to be disputed however because its derivation involves the use of an equation only applicable to spheres, such as that due to Böttcher [23].

For comparison, derivations of extensions to both the Hanai-Bruggeman and the Looyenga equations for the case of an oriented dispersed phase of spheroidal shape are presented here. The latter equation is also generalised to include conductivity effects.

Derivation of new equations

Generalised Bruggeman equation

The Bruggeman integral method can be applied to oriented spheroidal shapes by using the equations of Sillars [17], Mandel [18] or van Beek [1].

The Silars equation will be used here and can be expressed in the form

$$\varepsilon = \varepsilon_1 + \phi \frac{(\varepsilon_2 - \varepsilon_1)\,\varepsilon_1}{\varepsilon_1 + A_a(\varepsilon_2 - \varepsilon_1)} \tag{1}$$

where ε_1, ε_2 and ε are the respective permittivities of the continuous phase, the dispersed phase and the mixture, ϕ is the volume fraction of dispersed phase and A_a is the depolarizing factor along the a-axis given by the equation

$$A_a = \frac{abc}{2} \int_0^\infty \frac{ds}{(s+a^2)^{3/2}(s+b^2)^{1/2}(s+c^2)^{1/2}} \tag{2}$$

where a, b and c are the lengths of the semi-axes of the spheroids, with $b = c$ [1].

For prolate spheroids [1, 17]:

$$A_a = \frac{-1}{\left(\frac{a}{b}\right)^2 - 1} + \frac{a/b}{\left[\left(\frac{a}{b}\right)^2 - 1\right]^{3/2}} \ln\left[\frac{a}{b} + \left\{\left(\frac{a}{b}\right)^2 - 1\right\}^{1/2}\right]. \tag{3}$$

For oblate spheroids

$$A_a = \frac{1}{1 - \left(\frac{a}{b}\right)^2} - \frac{\left(\frac{a}{b}\right) \arccos\left(\frac{a}{b}\right)}{\left[1 - \left(\frac{a}{b}\right)^2\right]^{3/2}}. \tag{4}$$

For spheres $A_a = \frac{1}{3}$ (In the original formulation Sillars used a factor n where $n = 1/A_a$.)

The Sillars equation is rendered into incremental form by adopting the following changes [1, 2]: ε_1 is replaced by ε, ε by $\varepsilon + \Delta\varepsilon$ and ϕ by $\frac{\Delta\phi'}{1-\phi'}$ where $\Delta\phi'$ is the incremental change in volume fraction and ϕ' is the volume fraction after a given addition. Inserting these permittivity and volume fraction changes into the Sillars equation (1) gives

$$\frac{[\varepsilon(1 - A_a) + A_a\varepsilon_2]\Delta\varepsilon}{\varepsilon(\varepsilon_2 - \varepsilon)} = \frac{\Delta\phi'}{1 - \phi'}. \tag{5}$$

Integration of (5) between the limits ε_1 and ε for the permittivity and 0 and ϕ for the volume fraction gives

$$\frac{\varepsilon - \varepsilon_2}{\varepsilon_1 - \varepsilon_2}\left(\frac{\varepsilon_1}{\varepsilon}\right)^{A_a} = 1 - \phi. \tag{6}$$

The same result is also obtained when the Sillars equation is replaced by either that of Mandel [18] or van Beek [1]. For spheres $A_a = \frac{1}{3}$, and the Bruggeman equation is obtained.

Generalised Hanai-Bruggeman equations

The Sillars equation can also be expressed in complex form for appropriate systems [17]

$$\varepsilon^* = \varepsilon_1^*\left[1 + \phi \frac{(\varepsilon_2^* - \varepsilon_1^*)\varepsilon_1^*}{\varepsilon_1^* + A_a(\varepsilon_2^* - \varepsilon_1^*)}\right]. \tag{7}$$

Applying the incremental changes, $\varepsilon_1^* \rightarrow \varepsilon^*$, $\varepsilon^* \rightarrow \varepsilon^* + \Delta\varepsilon$ and $\phi \rightarrow \Delta\phi'/1 - \phi'$ gives equation (5) in complex form.

Provided the complex permittivity is continuous during incrementation from ε_1^* to ε^* the complex integral can be solved [2, 24] giving

$$\frac{\varepsilon^* - \varepsilon_2^*}{\varepsilon_1^* - \varepsilon_2^*}\left(\frac{\varepsilon_1^*}{\varepsilon^*}\right)^{A_a} = 1 - \phi \tag{8}$$

which is the generalised Bruggeman equation in complex form.

This equation is then solved using the following simplified form of the Hanai method [2].

The complex quantities are expressed as

$$\varepsilon^* = (\varepsilon^2 + \gamma^2)^{1/2} \exp\left[-j \arctan(\gamma/\varepsilon)\right] \tag{9}$$

where γ is the dielectric loss ($= j\sigma/\omega\varepsilon_0$ where σ is the conductivity).

$$\varepsilon^* - \varepsilon_2^* = [(\varepsilon - \varepsilon_2)^2 + (\gamma - \gamma_2)^2]^{1/2} \exp\left[-j \arctan\left(\frac{\gamma - \gamma_2}{\varepsilon - \varepsilon_2}\right)\right] \tag{10}$$

$$\varepsilon_1^* - \varepsilon_2^* = [(\varepsilon_1 - \varepsilon_2)^2 + (\gamma_1 - \gamma_2)^2]^{1/2} \exp\left[-j \arctan\left(\frac{\gamma_1 - \gamma_2}{\varepsilon_1 - \varepsilon_2}\right)\right]. \tag{11}$$

Substituting into equation (8) and separating real and imaginary parts yields:

$$\left[\frac{(\varepsilon - \varepsilon_2)^2 + (\gamma - \gamma_2)^2}{(\varepsilon_1 - \varepsilon_2)^2 + (\gamma_1 - \gamma_2)^2}\right]\left(\frac{\varepsilon_1^2 + \gamma_1^2}{\varepsilon^2 + \gamma^2}\right)^{A_a} = (1 - \phi)^2 \tag{12}$$

and

$$\arctan\left[\frac{\varepsilon\gamma_1 - \varepsilon_1\gamma}{\varepsilon\varepsilon_1 + \gamma\gamma_1}\right] = A_a \arctan\left[\frac{(\varepsilon - \varepsilon_2)(\gamma_1 - \gamma_2) - (\varepsilon_1 - \varepsilon_2)(\gamma - \gamma_2)}{(\varepsilon - \varepsilon_2)(\varepsilon_1 - \varepsilon_2) + (\gamma - \gamma_2)(\gamma_1 - \gamma_2)}\right]. \tag{13}$$

Equation (13) is solved to obtain ε and γ using the approximation $\arctan \psi = \psi$ and is thus only applicable for very low values of ψ. This approximation is valid for the following

a) The high frequency limiting case where $\varepsilon_1 \gg \gamma_1$, $\varepsilon_2 \gg \gamma_2$ and $\varepsilon \gg \gamma$ ($\varepsilon = \varepsilon_\infty$, $\sigma = \sigma_\infty$) for which

$$\frac{\varepsilon_\infty - \varepsilon_2}{\varepsilon_1 - \varepsilon_2}\left(\frac{\varepsilon_1}{\varepsilon_\infty}\right)^{A_a} = 1 - \phi \tag{14}$$

and

$$\sigma_\infty\left[\frac{1}{A_a(\varepsilon_\infty - \varepsilon_2)} - \frac{1}{\varepsilon_\infty}\right] = \frac{1}{A_a}\left[\frac{\sigma_1 - \sigma_2}{\varepsilon_1 - \varepsilon_2} + \frac{\sigma_2}{\varepsilon_\infty - \varepsilon_2}\right] - \frac{\sigma_1}{\varepsilon_1} \tag{15}$$

b) The low frequency limiting case where $\gamma_1 \gg \varepsilon_1$, $\gamma_2 \gg \varepsilon_2$ and $\gamma \gg \varepsilon$ ($\varepsilon = \varepsilon_S$, $\sigma = \sigma_S$) for which

$$\varepsilon_S\left[\frac{1}{A_a(\sigma_S - \sigma_2)} - \frac{1}{\sigma_S}\right] = \frac{1}{A_a}\left[\frac{\varepsilon_1 - \varepsilon_2}{\sigma_1 - \sigma_2} + \frac{\varepsilon_2}{\sigma_S - \sigma_2}\right] - \frac{\varepsilon_1}{\sigma_1} \tag{16}$$

and

$$\frac{\sigma_S - \sigma_2}{\sigma_1 - \sigma_2}\left(\frac{\sigma_1}{\sigma_S}\right)^{A_a} = 1 - \phi \tag{17}$$

These equations can be simplified when the two components of the mixture have significantly different conductivities as will be the case for dispersions of oil in water (O/W) and water in oil (W/O).

For O/W $\sigma_2 \ll \sigma_1$, $\sigma_2 \ll \sigma_S$ and the low frequency limiting values of ε and σ reduce to

$$\varepsilon_S = \frac{1}{1 - A_a}\left[\{\varepsilon_1(1 - A_a) - \varepsilon_2\}(1 - \phi)^{\frac{1}{1 - A_a}} + \varepsilon_3\right] \tag{18}$$

and

$$\sigma_S = \sigma_1(1 - \phi)^{\frac{1}{1 - A_a}} \tag{19}$$

For W/O $\sigma_1 \ll \sigma_2$, $\sigma_1 \ll \sigma_S$ and the low frequency limiting values reduce to

$$\varepsilon_S = \frac{\varepsilon_1}{(1 - \phi)^{\frac{1}{A_a}}} \tag{20}$$

and

$$\sigma_S = \frac{\sigma_1}{(1 - \phi)^{\frac{1}{A_a}}}. \tag{21}$$

For spheres $A_a = \frac{1}{3}$ and the Hanai equations are obtained.

Generalised Looyenga equation

Derivation of the Looyenga equation is also based on incremental changes, where in this case they are $\varepsilon_1 \rightarrow \varepsilon - \Delta\varepsilon$, $\varepsilon_2 \rightarrow \varepsilon + \Delta\varepsilon$ [4]. For the volume fraction term, the mixture is considered to contain a volume fraction ϕ' with a permittivity $(\varepsilon + \Delta\varepsilon)$ and $(1-\phi')$ with a permittivity $(\varepsilon - \Delta\varepsilon)$ such that

$$\phi(\varepsilon) = (1-\phi')\phi(\varepsilon - \Delta\varepsilon) + \phi' \cdot \phi(\varepsilon + \Delta\varepsilon) \qquad (22)$$

where $\phi(\varepsilon)$, $\phi(\varepsilon - \Delta\varepsilon)$ and $\phi(\varepsilon + \Delta\varepsilon)$ are all volume fraction functions. When equation (22) is expanded in a Taylor's series form to the second derivative, we obtain:

$$\phi' = \frac{1}{2} - \frac{1}{4}\Delta\varepsilon \frac{d^2\phi/d\varepsilon^2}{d\phi/d\varepsilon}. \qquad (23)$$

Inserting the incremental permittivity changes into the Sillars equation (1) gives

$$\phi' = \frac{1}{2}\left[1 + 2A_a \frac{\Delta\varepsilon}{\varepsilon}\right]. \qquad (24)$$

Combining equations (23) and (24):

$$\frac{1}{2A_a}\varepsilon \frac{d^2\phi}{d\varepsilon^2} + \frac{d\phi}{d\varepsilon} = 0 \qquad (25)$$

which on integration, together with the appropriate boundary conditions, gives

$$\varepsilon = [\varepsilon_1^{1-2A_a} + \phi(\varepsilon_2^{1-2A_a} - \varepsilon_1^{1-2A_a})]^{\frac{1}{1-2A_a}}. \qquad (26)$$

For spheres, $A_a = \frac{1}{3}$ and the Looyenga equation is obtained.

Extension of Looyenga equation to include conductivity

As with the Bruggeman equation, the generalised Looyenga equation can also be expressed in complex form:

$$\varepsilon^* = [\varepsilon_1^{*\,1-2A_a} + \phi(\varepsilon_2^{*\,1-2A_a} - \varepsilon_1^{*\,1-2A_a})]^{\frac{1}{1-2A_a}}. \qquad (27)$$

Expressing the complex quantities in the same form as equations (9), (10) and (11) and substituting back into equation (27) gives on equating real and imaginary parts:

$$\left(\frac{\varepsilon^2 + \gamma^2}{\varepsilon_1^2 + \gamma_1^2}\right)^{\frac{1-2A_a}{2}} \cdot \cos\theta_1 - 1 = \phi\left(\frac{\varepsilon_2^2 + \gamma_2^2}{\varepsilon_1^2 + \gamma_1^2}\right)^{\frac{1-2A_a}{2}} \cdot \cos\theta_2 - \phi \qquad (28)$$

where $\theta_1 = (1-2A_a)\,[\arctan(\gamma/\varepsilon) - \arctan(\gamma_1/\varepsilon_1)]$ (29)

and $\theta_2 = (1-2A_a)[\arctan(\gamma_2/\varepsilon_2) - \arctan(\gamma_1/\varepsilon_1)]$ (30)

and

$$\left(\frac{\varepsilon^2 + \gamma^2}{\varepsilon_1^2 + \gamma_1^2}\right)^{\frac{1-2A_a}{2}} \cdot \sin\theta_1 = \phi\left(\frac{\varepsilon_2^2 + \gamma_2^2}{\varepsilon_1^2 + \gamma_1^2}\right)^{\frac{1-2A_a}{2}} \cdot \sin\theta_2. \qquad (31)$$

These equations are solved for the following two limiting cases:

(a) The high frequency limit $\varepsilon_1 \gg \gamma_1$, $\varepsilon_2 \gg \gamma_2$ and $\varepsilon \gg \gamma$ ($\varepsilon = \varepsilon_\infty$, $\sigma = \sigma_\infty$) for which conditions

$$\varepsilon_\infty^{1-2A_a} = \varepsilon_1^{1-2A_a} + \phi(\varepsilon_2^{1-2A_a} - \varepsilon_1^{1-2A_a}) \qquad (32)$$

and

$$\sigma_\infty = \frac{1}{\varepsilon_1}\left[\left(\frac{\varepsilon_2}{\varepsilon_\infty}\right)^{-2A_a} \phi(\sigma_2\varepsilon_1 - \sigma_1\varepsilon_2) + \sigma_1\varepsilon_\infty\right]. \qquad (33)$$

(b) The low frequency limit $\gamma_1 \gg \varepsilon_1$, $\gamma_2 \gg \varepsilon_2$ and $\gamma \gg \varepsilon$ ($\varepsilon = \varepsilon_S$, $\sigma = \sigma_S$) for which

$$\varepsilon_S = \frac{1}{\sigma_1}\left[\sigma_S\varepsilon_1 - \left(\frac{\sigma_2}{\sigma_S}\right)^{-2A_a} \phi(\sigma_2\varepsilon_1 - \sigma_1\varepsilon_2)\right] \qquad (34)$$

and

$$\sigma_S^{1-2A_a} = \sigma_1^{1-2A_a} + \phi(\sigma_2^{1-2A_a} - \sigma_1^{1-2A_a}). \qquad (35)$$

For these equations simplified forms for the cases of $\sigma_1 \gg \sigma_2$ and $\sigma_1 \ll \sigma_2$ cannot be given (see Table 5).

Discussion

The extended Hanai-Bruggeman values for the high and low frequency limiting mixture-permittivites, ε_∞

Table 1. Low and high frequency limiting values of permittivity given by extended Hanai-Bruggeman equation for a hypothetical oil in water system

ϕ	$a/b = 3$		$a/b = 1$		$a/b = 1/3$	
	ε_S	ε_∞	ε_S	ε_∞	ε_S	ε_∞
0.10	69.6	69.6	67.0	67.0	59.8	59.8
0.20	61.2	61.2	56.7	56.7	44.8	44.7
0.30	53.0	53.0	46.9	46.9	32.8	32.6
0.40	44.9	44.9	37.9	37.9	23.4	23.0
0.50	37.0	37.0	29.5	29.5	16.3	15.8
0.60	29.3	29.3	22.0	21.9	11.4	10.5
0.70	21.9	21.9	15.3	15.2	8.2	6.9
0.80	14.7	14.7	9.7	9.6	6.4	4.5
0.90	8.0	7.9	5.4	5.1	5.6	2.9

Table 3. Low and high frequency limiting values of permittivity given by extended Looyenga equations for a hypothetical oil in water system with $\sigma_2/\sigma_1 = 0.001$. ($O/W$ and W/O systems generate the same permittivities for given proportions of water and oil)

ϕ	$a/b = 3$		$a/b = 1$		$a/b = 1/3$	
	ε_S	ε_∞	ε_S	ε_∞	ε_S	ε_∞
0.10	69.0	68.7	74.7	62.7	175.6	43.8
0.20	60.3	59.7	68.8	49.5	82.3	26.6
0.30	51.9	51.0	61.0	38.3	41.1	17.1
0.40	43.8	42.6	51.9	28.9	22.5	11.5
0.50	35.9	34.5	42.1	21.2	13.3	8.1
0.60	28.4	26.8	32.0	15.0	8.4	5.8
0.70	21.4	19.6	22.4	10.1	5.6	4.3
0.80	14.6	13.0	13.7	6.5	3.8	3.3
0.90	8.5	7.0	6.8	3.8	2.7	2.5

and ε_S, as a function of volume fraction of dispersed phase ϕ for axial ratios $a/b = 3$ (prolate spheroids), 1 (spheres) and 1/3 (oblate spheroids) are given for an O/W system in table 1 and for W/O table 2. A value of 78.0 for the permittivity of water and 2.0 for that of oil, corresponding to room temperature, has been used, with ε_S being calculated using one of the simplified equations, (18) or (20) as appropriate.

The extended Looyenga values for ε_∞ and ε_S at various values of ϕ for the above three axial ratios are given for O/W in table 3, where a conductivity ratio $\sigma_2/\sigma_1 = 0.001$ has been used in the evalutaion of ε_S. This type of equation generates the same permittivities for O/W and W/O for given proportions of water and oil, that is, a volume fraction ϕ of O/W has the same permittivity as a fraction $(1-\phi)$ of W/O.

Although orientation of oblate spheroids with their unique axis parallel to the applied field is physically unrealistic for many systems, because it is unlikely to be a direction of stable orientation [16, 17], it may well be appropriate in certain surfactant and liquid crystalline systems where the molecular dipole moment of the dispersed phase lies perpendicular to that of, for example, radially distributed molecules [25].

Table 2. Low and high frequency limiting of permittivity given by extended Hanai-Bruggeman equations for a hypothetical water in oil system

ϕ	$a/b = 3$		$a/b = 1$		$a/b = 1/3$	
	ε_S	ε_∞	ε_S	ε_∞	ε_S	ε_∞
0.10	5.3	4.1	2.7	2.7	2.4	2.4
0.20	15.5	7.6	3.9	3.6	2.8	2.8
0.30	52.7	12.8	5.8	5.1	3.5	3.4
0.40	216.9	19.6	9.3	7.4	4.5	4.3
0.50	1155.5	27.4	16.0	11.0	6.0	5.5
0.60	8950.7	36.4	31.3	16.5	8.5	7.5
0.70	$1.25.10^5$	45.9	74.3	25.1	13.3	10.9
0.80	$5.17.10^6$	56.1	251.2	37.6	25.2	17.6
0.90	$2.99.10^9$	66.8	$2.01.10^3$	55.0	75.1	33.0

The results given here show that the values of ε_∞ and ε_S are dependent both on dispersed phase shape and the mixture equation used, except for prolate spheroidal O/W systems where the two types of equation generate similar values. As the shape becomes progressively more prolate the permittivity values converge and in the limit of $a/b \gg 1$ both types of equation reduce to the simple ideal mixing equation

$$\varepsilon_S = \varepsilon_\infty = \phi\varepsilon_2 + (1-\phi)\varepsilon_1\,.$$

This is equivalent to two parallel layers aligned with the field and continuous between the two electrodes. The dielectric increment $\Delta\varepsilon(\Delta\varepsilon = \varepsilon_S - \varepsilon_\infty)$ is zero for this case.

The extended Hanai-Bruggeman equations give a very small dielectric increment for all O/W systems considered except for the oblate shape at high concentrations. For W/O $\Delta\varepsilon$ is generally significant, and is largest for the prolate shape, with extremely large $\Delta\varepsilon$ values being obtained at higher concentrations. For oblate shaped W/O systems $\Delta\varepsilon$ is small over most of the concentration range and even at the highest concentration it is only moderate. This latter behaviour differs from that obtained by Boned and Peyrelasse [21] for random orientations where very high $\Delta\varepsilon$ values were obtained for both oblate and prolate shapes.

Table 4. Dielectric increment $\Delta\varepsilon$ for hypothetical water in oil systems for $\phi = 0.5$ given by extended Hanai-Bruggeman equations

σ_1/σ_2	$\Delta\varepsilon$ $a/b = 3$	$a/b = 1$	$a/b = 1/3$
0.3	85.1	89.4	77.3
0.1	83.3	70.6	43.4
0.03	84.3	48.1	17.3
0.01	89.6	26.5	7.3
0.003	111.8	13.4	2.6
0.001	171.2	5.4	1.2
0.0001	593.5	5.3	0.6
0.00001	1027.6	5.3	0.5

Table 5. Dielectric increment $\Delta\varepsilon$ for hypothetical water in oil systems for $\phi = 0.5$ given by extended Looyenga equations

σ_1/σ_2	$\Delta\varepsilon$ $a/b = 3$	$a/b = 1$	$a/b = 1/3$
0.3	1.9	7.6	11.0
0.1	0.5	2.4	2.6
0.03	0.0	0.1	0.0
0.01	0.2	1.2	0.7
0.003	0.8	7.3	2.9
0.001	1.7	20.9	5.2
0.0001	3.7	116.5	9.8
0.00001	7.7	547.0	13.2

The extended Looyenga equations give a small dielectric increment for the prolate shape at all concentrations, but $\Delta\varepsilon$ is significant for spheres and oblate spheroids, and largest for the oblate shape at low concentrations.

The equations given here show major differences in the dependence of the dielectric increment on the ratio of the conductivities of the two components. Tables 4 and 5 give the extended Hanai-Bruggeman and extended Looyenga equation values respectively for $\Delta\varepsilon$ as a function of σ_1/σ_2 for W/O systems with $\phi = 0.5$.

For the Hanai-Bruggeman type $\Delta\varepsilon$ progressively falls with decreasing σ_1/σ_2 for oblate spheroids and spheres to attain limiting values of 0.5 and 5.3 respectively, compared with values of 0.5 and 4.9 given by the simplified equation (20). For the prolate shape $\Delta\varepsilon$ progressively increases and has not attained the value of 1128.5 given by equation (20) even when $\sigma_1/\sigma_2 = 10^{-5}$.

For the Looyenga type, for all three shapes, $\Delta\varepsilon$ initially falls with decreasing σ_1/σ_2 becoming zero at $\sigma_1/\sigma_2 \simeq 0.03$ and then increases again. For spheres large increments are predicted when σ_1/σ_2 is small. These important features show the relevance of conductivity data to complement dielectric data in studies of heterogeneous systems.

Although the theoretical bases for the Hanai-Bruggeman and the Looyenga equations are probably equally valid [26], dielectric data for both O/W and W/O systems of assumed spherically-shaped dispersed phase gives a dielectric increment which is in reasonable agreement with the Hanai-Bruggeman approach [12, 27–29]. This suggests that in such systems the latter is more appropriate. Also, data on certain spheroidal micellar systems are better accounted for by the extended Bruggeman equation rather than the Looyenga type [11].

However, a lack of experimental data on systems with dispersed phases of various shapes at high concentrations prevents an assessment of the usefulness of the equations given here. Although the internal field of the dispersed globule, together with bound water effects and their magnitudes, present considerable computational difficulties [25, 30, 31], further studies on micellar and microemulsion systems, together with studies on certain liquid crystalline structures should provide data which will yield to analysis.

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