Supporting Information

Calculation of the complex permittivity for concentrated suspensions of ellipsoidal particles

The effective complex permittivity for a dilute suspension of randomly oriented ellipsoids (of complex permittivity ε_p^*, and semiaxes R_x, R_y and R_z) in a continuous medium (of ε_a^*) at volume fraction P is given by[27]

$$\varepsilon^* = \varepsilon_a^* \left[1 + \frac{1}{3} P \sum_{k=x,y,z} \frac{\left(\varepsilon_p^* - \varepsilon_a^*\right)}{\varepsilon_a^* + \left(\varepsilon_p^* - \varepsilon_a^*\right) L_k} \right], \tag{S1}$$

where the depolarization factor L_k is given by

$$L_k = \frac{R_x R_y R_z}{2} \int_0^\infty \frac{ds}{\left(R_k^2 + s\right) R_s} \tag{S2}$$

with

$$R_s = \sqrt{\left(R_x^2 + s\right)\left(R_y^2 + s\right)\left(R_z^2 + s\right)}, \tag{S3}$$

$$\sum_{k=x,y,z} L_k = 1. \tag{S4}$$

For extending the mixture equation to high volume fractions, the effective medium theory proposed by Bruggeman provides a reasonable approximation. In the theory, each particle is supposed to be dispersed in an effective medium, though it includes particles, and the initially low volume fraction is gradually increased by infinitesimal additions of particles. When a small amount of particles of ε_p^* are added to an effective medium of ε^*, eq S1 may be applicable to every addition process. Hence, the increment in complex permittivity $\Delta\varepsilon^*$ due to an infinitesimal addition of particles is related to the increment in volume fraction $\Delta P'$ by substituting $\varepsilon^* + \Delta\varepsilon^*$, ε^* and $\Delta P'/(1-P')$ for ε^*, ε_a^*, and P in eq S1, respectively.

$$-\frac{\Delta P'}{1-P'}=\frac{3}{\varepsilon^*\left(\varepsilon^*-\varepsilon_i^*\right)}\left[\sum_{k=x,y,z}\frac{1}{\varepsilon^*+\left(\varepsilon_i^*-\varepsilon^*\right)L_k}\right]^{-1}\Delta\varepsilon^* \tag{S5}$$

By successive infinitesimal additions of particles, the system reaches the final volume fraction P and complex permittivity ε^*, and thus we obtain an integral equation as:

$$\int_0^{\Phi}-\frac{d\Phi'}{1-\Phi'}=\int_{\varepsilon_a^*}^{\varepsilon^*}\frac{3}{\varepsilon^*\left(\varepsilon^*-\varepsilon_p^*\right)}\left[\sum_{k=x,y,z}\frac{1}{\varepsilon^*+\left(\varepsilon_p^*-\varepsilon^*\right)L_k}\right]^{-1}d\varepsilon^* . \tag{S6}$$

Although eq S6 can be solved analytically, the obtained equation is complicated and its numerical solution requires iterative searching that often does not converge. Thus, an alternative algorithm has been developed, in which numerical integration is made with eq S7 derived from eq S5.

$$\Delta\varepsilon^*=\frac{\Delta P'}{3(1-P')}\sum_{k=x,y,z}\frac{\varepsilon^*\left(\varepsilon_i^*-\varepsilon^*\right)}{\varepsilon^*+\left(\varepsilon_i^*-\varepsilon^*\right)L_k} . \tag{S7}$$

The volume fraction P' was successively increased by adding $\Delta P'$ until the final volume fraction P was attained after n addition steps, i.e., $P=n\Delta P'$. The increment of complex permittivity $\Delta\varepsilon^*$ is calculated for every addition steps and then the final complex permittivity $\varepsilon_{final}{}^*$ is obtained after n^{th} step as follows:

1st step: to put $\varepsilon^*\left(\equiv\varepsilon_1^*\right)=\varepsilon_a^*$ and $P'\left(\equiv P_1'\right)=0$ in eq S7, and then to calculate $\Delta\varepsilon^*\left(\equiv\Delta\varepsilon_1^*\right)$.

2nd step: to put $\varepsilon^*\left(\equiv\varepsilon_2^*\right)=\varepsilon_1^*+\Delta\varepsilon_1^*$ and $P'\left(\equiv P_2'\right)=P_1'+\Delta P'$, and then to calculate $\Delta\varepsilon^*\left(\equiv\Delta\varepsilon_2^*\right)$.

jth step: to put $\varepsilon^*\left(\equiv\varepsilon_j^*\right)=\varepsilon_{j-1}^*+\Delta\varepsilon_{j-1}^*$ and $P'\left(\equiv P_j'\right)=P_{j-1}'+\Delta P'$, and then to calculate $\Delta\varepsilon^*\left(\equiv\Delta\varepsilon_j^*\right)$.

nth step: to put $\varepsilon^*\left(\equiv\varepsilon_n^*\right)=\varepsilon_{n-1}^*+\Delta\varepsilon_{n-1}^*$ and $P'\left(\equiv P_n'\right)=P_{n-1}'+\Delta P'$, and then to calculate $\Delta\varepsilon^*\left(\equiv\Delta\varepsilon_n^*\right)$ and $\varepsilon_{final}^*=\varepsilon_n^*+\Delta\varepsilon_n^*$.