Comment on "A New Model for the Viscosity of Asphaltene Solutions"

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In a recent article[1] a "new" model for the viscosity of asphaltene solutions is proposed. The main result of that work can be summarized by the two equations:

$$\eta_r(\phi) = \left(1 - \phi_{eff}\right)^{-[\eta]}, \quad (1)$$

and

$$\phi_{eff} = \left[1 + \left(\frac{1-\phi_m}{\phi_m}\right)\left(\sqrt{1 - \left(\frac{\phi_m - \phi}{\phi_m}\right)^2}\right)\right]\phi, \quad (2)$$

where $\eta_r(\phi)$ is the relative viscosity of the solution, ϕ is the volume fraction of the dispersed phase, $[\eta]$ is the intrinsic viscosity, and ϕ_m is the maximum packing volume fraction of particles. The somewhat cumbersome expression for the effective volume fraction ϕ_{eff} given by Equation (2) is chosen ad hoc so that it satisfies four requirements:

(a) In the dilute limit $\phi \to 0$, $\phi_{eff} = \phi$
(b) In the limit $\phi \to \phi_m$, $\phi_{eff} = 1$
(c) The slope of ratio ϕ_{eff}/ϕ with respect to ϕ is positive
(d) The ratio ϕ_{eff}/ϕ becomes constant at high ϕ.

Then, this model is compared with the "key" literature models and used to accurately correlate a number of different experimental results.

However, the author, apparently unaware of the recent progress in the calculation of the viscosity of colloidal suspensions, does not mention that Equation (1) was first obtained a few years ago in Mendoza and Santamaria-Holek[2] for the case of spherical particles for which $[\eta] = 2.5$, and in Santamaria-Holek and Mendoza[3] for arbitrary-shaped particles (see Equation (22) of Mendoza and Santamaria-Holek[2] and Equation (17) of Santamaria-Holek and Mendoza[3]).

Furthermore, Equation (1) is obtained in Pal[1] through a differential effective medium procedure with the effective volume fraction ϕ_{eff} as an incremental variable (see Equation (23) of Santamaria-Holek and Mendoza[1]). This way of obtaining Equation (1) was also first proposed in Mendoza and Santamaria-Holek[2] and Santamaria-Holek and Mendoza [3] (see Equations (20) and (21) of Mendoza and Santamaria-Holek[2] and Equations (15) and (16) of Santamaria-Holek and Mendoza[3]).

In Mendoza and Santamaria-Holek[2] it was explained that the effective viscosity of suspensions depends on a particular scale variable ϕ_{eff} that should be used in the differential procedure in

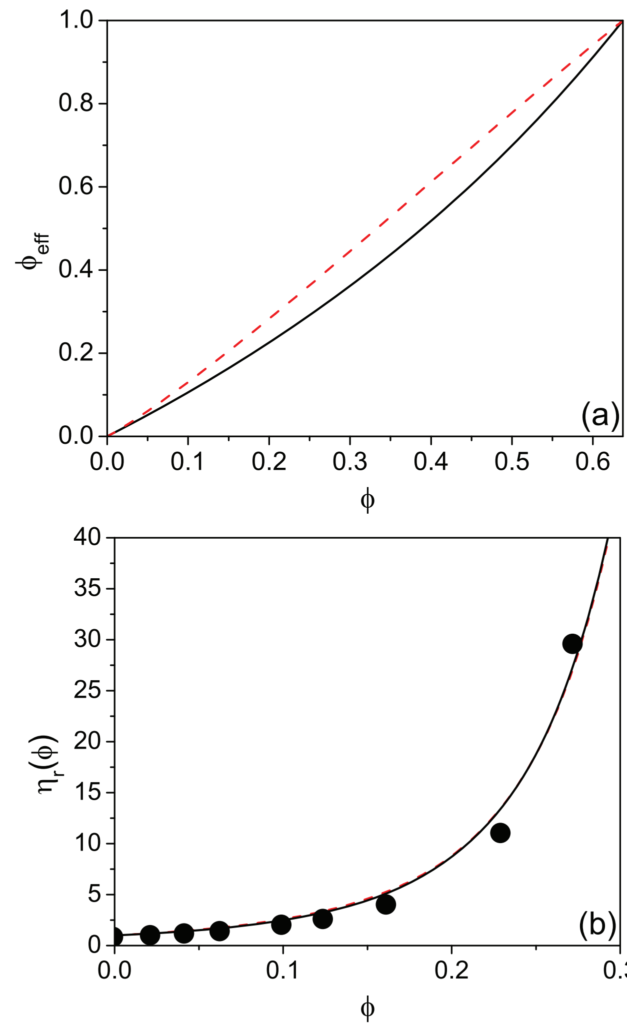

Figure 1. (a) Effective volume fraction versus volume fraction, as given by Equations (3) (solid line) and (2) (dashed line). (b) Relative viscosity vs volume fraction, as given by Equation (1) together with Equation (3) (solid line) and Equation (2) (dashed line). The fitted intrinsic viscosities were $[\eta] = 8.5$ and $[\eta] = 6.5$, respectively. In both cases $\phi_m = 0.637$.

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order to give a consistent description of the experimental data. This is reflected in the universal nature of the variable leading to a master curve [see Figure 5 of Mendoza and Santamaria-Holek[2]].

Thus, the only difference between the "new" model of Pal[1] and the previous one[2,3] is the expression for the effective volume fraction which is given by.[2,3]

$$\phi_{eff} = \frac{\phi}{1 - \left(\frac{1-\phi_m}{\phi_m}\right)\phi}. \tag{3}$$

This expression was introduced and physically justified in Mendoza and Santamaria-Holek[2] in terms of the volume that is not accessible to the dispersed phase of the suspension. Moreover, the fact that ϕ_{eff} satisfies requirements (a) and (b) was also explicitly shown previously.[2,3]

If we expand Equation (3) to second order in ϕ we get

$$\phi_{eff} \approx \left[1 + \left(\frac{1-\phi_m}{\phi_m}\right)\phi\right]\phi, \tag{4}$$

a form very similar to Equation (2) except for the factor ϕ within the square brackets which is replaced by the term with the square root.

In Figure 1a we plot both expressions for ϕ_{eff}, Equations (2) and (3). The difference is not significant when compared with experimental data as shown in Figure 1b. Here we plot the relative viscosity using both expressions for ϕ_{eff}. The values of $[\eta]$ used to fit the experimental data[4] were $[\eta] = 6.5$ and $[\eta] = 8.5$, respectively. As shown, both curves are essentially indistinguishable for the considered range of ϕ. The same conclusion is drawn when considering the rest of the experimental data sets used in Pal.[1]

In view of the above reasons, the novelty of the viscosity model "proposed" in Pal[1] is unjustified.

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