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The viscosity of a concentrated suspension of rigid monosized particles

The rheological behaviour of concentrated suspension is of great importance in a wide variety of applications, in biology, food and engineering. There is, therefore, practical as well as fundamental interest in understanding the relationship between the concentration, particle shape and particle-size distribution on the one hand, and relative viscosity of the suspension (or slurry) on the other.

The concentration of concentrated slurries is highly sensitive to how this property is measured. Here the effective shear at zero-frequency is addressed. For dilute suspensions, the viscosity-concentration function can be linearized (e.g. the classical result of Einstein (1906, 1911). This linearized equation is based on no appreciable interaction between the particles, and the coefficient of which depends on particle shape only (and not on size distribution). As loading is increased, this universality is lost, and the viscosity diverges when the associated state of random close packing is approached, also depending on particle shape only. Numerous equations have been developed in efforts to extend the linear approximations to concentrated suspensions. These equations differ from each other, and none agrees well with data in the entire concentration range, from dilute to maximum concentration.

Farris developed and validated a theory to explain the viscosity reduction that follows from mixing discretely sized particles with sufficiently large size ratios. The suspension can then be represented as a coarse fraction suspended in a fluid containing the finer particles, all fractions behaving independently of each other. Here also a suspension of bimodal particles with small size ratio, i.e. interacting particles, is studied. The model of Farris (1968) of these suspensions is recapitulated. Next, the random close packing of these bimodal particle packings are addressed. The unimodal-bimodal limit as explained by Brouwers (2006) is studied to relate packing increase (when size ratio increases) and the associated apparent particle concentration reduction (fluid fraction increase). Combining the concepts of Farris and Brouwers for small size ratio, a general equation in closed form is derived that provides the viscosity of a suspension of monosized particles at all concentrations, from the dilute limit to the random close packing limit. This equation is governed by the single-sized packing of the particle shape considered (φ_1) and the dilute limit viscosity-concentration gradient (C_1), for spheres, $\varphi_1 \approx 0.64$ and $C_1 = 2.5$. This original expression for the viscosity is compared thoroughly with current models and with experiments in the full concentration range, and found to be in good accordance.

Suspensions of unimodal particles

The unimodal relative viscosity-concentration function is expressed as $H(\Phi)$, where H is the stiffening factor, the ratio of viscosity with particles divided by the viscosity of the pure fluid. H is a function of the particle volume concentration, Φ, and the particle shape. For dilute suspensions, the virial expansion of the relative viscosity to second order in Φ is:

$$\mu = \frac{\eta_{\mathrm{eff}}}{\eta_{\mathrm{f}}} = H(\Phi) = 1 + C_1\,\Phi + C_2\,\Phi^2 + O(\Phi^3) \quad . \tag{1}$$

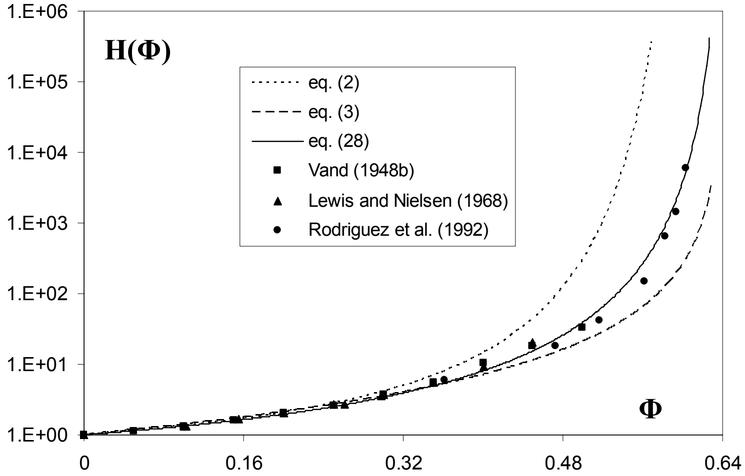

Figure 1: Stiffening function $H(\Phi)$ as function of particle volume fraction Φ for monosized spheres as predicted by eqs. (2), (3) and (28) and as measured.

For spheres, dominating viscous effects, and ignoring particle interactions, Einstein (1906, 1911) computed the first-order virial coefficient C_1 as 2.5. For non-spherical particles, C_1 is larger (Boek et al. (1997), Wierenga and Philipse (1998), Van der Kooij et al. (2001)). The value has for instance been computed and measured for ellipsoids and slender rods (Jeffery (1922), Onsager (1932), Kuhn and Kuhn (1945), Berry and Russel (1987)). The second-order coefficient has among others been determined by Huggins (1942), Batchelor (1977), Wagner and Woutersen (1994), Boek et al. (1997), Wierenga and Philipse (1998) and Van der Kooij et al. (2001).

For more concentrated suspension, the most known phenomenological descriptions are the transcendental function by Vand (1948a) and Mooney (1951)

$$H(\Phi) = e^{\frac{C_1\Phi}{1-\Phi/\varphi_1}} \quad , \tag{2}$$

and the power-law function, attributed to Eilers (1941, 1943), Maron and Pierce (1956), Krieger-Dougherty (1959) and Quemada (1977), reading

$$H(\Phi) = \left(1 - \frac{\Phi}{\varphi_1}\right)^{-\varphi_1 C_1} \quad . \tag{3}$$

Both equations tend to eq. (1) for $\Phi \rightarrow 0$, and diverge for $\Phi \rightarrow \varphi_1$, i.e. the critical volume fraction. For low shear rates and without interparticle forces, this critical volume fraction is found experimentally to lie near the random close packing limit (Lee (1970), Marti et al.

(2005)). This represents the limiting packing fraction above which flow is no longer possible. For spheres, the random close packed fraction, φ_1, is about 0.64 (Scott (1960), Scott and Kilgour (1969)). Based on experiments with suspensions, Chong et al. (1971) and Van der Werff and De Kruif (1989) observed divergence for Φ tending to 0.61. In Figure 1, eqs. (2) and (3) are set out for $C_1 = 2.5$ and $\varphi_1 = 0.64$, which are the applicable values for hard spheres. For high shear rates some ordering is found, e.g. spherical particles tend to from crystalline clusters and the system seems capable to flow at volume fraction $\Phi > 0.64$ (Van der Werff and De Kruif (1989)), but this does not hold for zero and moderate shear rates, as addressed here.

Suspensions of bimodal particles with large size ratio

Eveson et al. (1951) conjectured that a bimodal suspension can be regarded as a system in which the large particles are suspended in a continuous phase formed by the suspension of the smaller particles in the fluid. Eveson (1959) further explored this geometric concept and by carefully executed experiments it could be confirmed. Farris (1968) used this concept to develop a model based on purely geometric arguments for the viscosity of multimodal suspensions. It was postulated that when large particles are suspended in a suspension of smaller particles, these fractions behave independently. The resulting viscosity can then be expressed in the unique viscosity-concentration behaviour of the unimodal suspension. Also the particle size distribution that results in the lowest viscosity, at a given solid concentration, was derived. Farris (1968) verified experimentally this geometrical concept for spheres. Also for non-spherical particle this concept was successful: for rods and spheres with large size ratio (length more than ten times the sphere diameter), see Mor et al. (1996) and Marti et al. (2005). So, to describe the viscosity of multimodal mixes, the unimodal concentration function, $H(\Phi)$, is of key importance.

Following the concept of Farris (1968), when coarse particles are added to the suspension of fines, the fine particles behave as a fluid towards the coarse. In this case of noninteracting particles, the relative viscosity reads

$$\mu = \frac{\eta_{\mathrm{eff}}}{\eta_f} = H(\Phi_L)\, H(\Phi_S) \quad , \tag{4}$$

in which Φ_L is the volume fraction of large particles in the total suspension volume, and Φ_S is the volume fraction of small particles in small particle plus fluid volume:

$$\Phi_L = \frac{V_L}{V_L + V_S + V_f} \quad , \tag{5}$$

$$\Phi_S = \frac{V_S}{V_S + V_f} \quad . \tag{6}$$

Note that this latter concentration is not equal to the volume fraction, defined as

$$x_S = \frac{V_S}{V_L + V_S + V_f} \quad . \tag{7}$$

Only for the largest fraction the concentration and volume fraction coincide, so $\Phi_L = x_L$. The total solid volume fraction reads

$$x_T = x_L + x_S \quad , \tag{8}$$

which is not equal to $\Phi_L + \Phi_S$. The total solid volume fraction in the suspension, x_T, is related to the individual concentrations by

$$1 - x_T = (1 - \Phi_L)(1 - \Phi_S) \quad . \tag{9}$$

Farris (1968) demonstrated that for particles with large size ratio (typically 10 or so), a minimum viscosity is obtained when $\Phi_L = \Phi_S = 1 - (1 - x_T)^{1/2}$, and hence $\mu = H(\Phi_L)^2$. The volume fractions of large and small particles in the solid mixture follow as

$$c_L = \frac{x_L}{x_L + x_S}; \quad c_S = \frac{x_S}{x_L + x_S} \quad , \tag{10}$$

Suspensions of bimodal particles with small size ratio

Farris model

The geometric model of Farris is known for large size ratios, which validity has been extensively confirmed. What apparently has not been noticed over the years, or at least not has not been remarked upon, is that Farris (1968) also extended the model to finite and small size ratios. From theory and experiments it was concluded that for interfering particle sizes, eq. (4) is still applicable but a part f of the smaller fraction should be assigned to the larger fraction, and the remaining part, 1- f, to the small fraction, hence:

$$\Phi_L = \frac{V_L + fV_S}{V_L + V_S + V_f} \quad , \tag{11}$$

$$\Phi_S = \frac{(1-f)V_S}{(1-f)V_S + V_f} \quad , \tag{12}$$

whereby f, the so-called crowding factor, depends on the particle size ratio. For u = 1 (monosized particles), f = 1 and in such case μ becomes $H(\Phi)$ as Φ_S becomes 0 and Φ_L becomes Φ. That is to say, for $u \downarrow 1$, the total particle volume fraction x_T of the unimodal particle suspension reads

$$x_T = x_L + x_S = \Phi < \varphi_1 \quad . \tag{13}$$

On the other hand, f = 0 for $u^{-1} = 0$, i.e. non-interacting sizes as discussed in the previous Section. In the latter case, obviously eqs. (5) and (6) are obtained. For constant x_S and varying x_L, Furnas (1968) provided f as a function of u^{-1}. Obviously, for $u \downarrow 1$, $x_L < \varphi_1 - x_S$ as $\Phi < \varphi_1$. In the vicinity of $u^{-1} = 1$, f can be approximated

$$f = 1 + \omega(1 - u^{-1}) = 1 - \omega(u - 1) + O((u - 1)^2) \quad , \tag{14}$$

inserting eq. (14) into eqs. (11) and (12) yields the following expressions

$$\Phi_L = \frac{V_L + V_S}{V_L + V_S + V_f} - \frac{\omega(u - 1)V_S}{V_L + V_S + V_f} = \Phi - \omega\,(u - 1)x_S = \Phi - \omega\,(u - 1)x_S \quad , \tag{15}$$

$$\Phi_S = \frac{\omega(u - 1)V_S}{\omega(u - 1)V_S + V_f} = \frac{\omega(u - 1)x_S}{1 - \Phi} + O((u - 1)^2) \quad , \tag{16}$$

see eqs. (7) and (8). Inserting eqs. (15) and (16) into the stiffening functions appearing in eq. (4), their Taylor series expansion for $u - 1 \rightarrow 0$ yields the following expressions for them:

$$H(\Phi_L) = H(\Phi) - \omega(u - 1)x_S \left(\left.\frac{dH}{d\Phi}\right|_{\Phi} \right) + O((u - 1)^2) \quad , \tag{17}$$

$$H(\Phi_S) = H(0) + \frac{\omega(u - 1)x_S}{1 - \Phi} \left(\left.\frac{dH}{d\Phi}\right|_{0} \right) + O((u - 1)^2) = 1 + \frac{\omega(u - 1)x_S C_1}{1 - \Phi} + O((u - 1)^2) \tag{18}$$

whereby eq. (1) has been used in eq. (18), i.e. the first-order expansion of H(Φ) in the dilute limit. Substituting eqs. (17) and (18) in the bimodal stiffening function, eq. (4), yields a first-order expression

$$\mu = H(\Phi_L)\, H(\Phi_S) = H(\Phi) - \omega(u - 1)x_S \left(\left.\frac{dH}{d\Phi}\right|_{\Phi} - \frac{C_1}{1 - \Phi} H(\Phi) \right) \quad . \tag{19}$$

This equation, based on the concept of Farris (1968), expresses the relative viscosity of a monosized suspension a with total concentration Φ that becomes bimodal. The last terms on the right-hand side eq. (19) govern the stiffening reduction upon the transition of unimodal particles to bimodal particles ($u > 1$) in the suspension.

Excess fluid model
Robinson (1949) presented a modification of the Einstein equation by considering the “free fluid”, *i.e.* the fluid outside of the suspended particles. In this model the packing fraction of the suspended particles is relevant. Here this concept is applied to monosized particles and the case these particles become bimodal, that is to say, the packing increases and excess fluid is generated. By Brouwers (2006, 2007, 2008) it was demonstrated that for $u - 1$ approaching zero, the bimodal packing fraction can described by

$$\varphi_T(u \rightarrow 1, c_L) = \varphi_1 + 4\beta\varphi_1(1 - \varphi_1)c_S c_L(u - 1) \quad . \tag{20}$$

Both φ_1 and β depend on the particle shape and the mode of packing (e.g. dense, loose) only, for random close packing (RCP) of spheres, $\varphi_1 = 0.64$ and $\beta = 0.20$ (Brouwers (2007)). The parameter β follows from the gradient in packing fraction when a unimodal packing ($u = 1$) turns into a bimodal packing ($u > 1$). It follows that along ($u = 1$, $0 \leq c_L \leq 1$), the packing fraction retains it monosized value; physically this implies that particles are replaced by particles of identical size, i.e., maintaining a single-sized mixture, and $x_L + x_S = \varphi_1$. Also along ($u \geq 1$, $c_L = 0$) and ($u \geq 1$, $c_L = 1$), the packing fraction remains φ_1, as this corresponds to the packing of unimodal small and large particles, respectively.

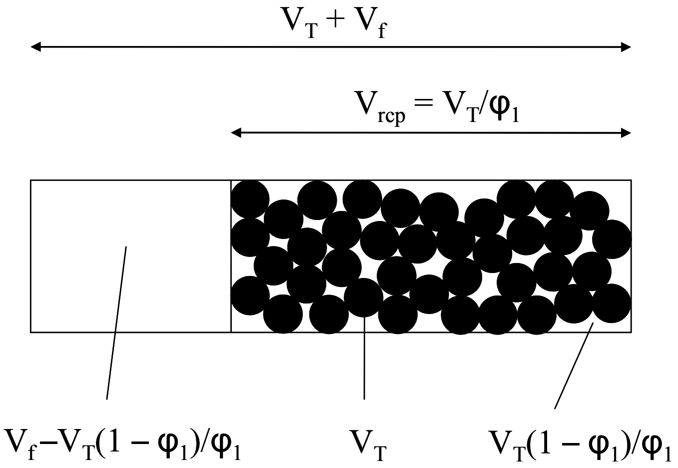

Figure 2: Schematic representation of a suspension of unimodal particles with total volume V_T and a fluid volume V_f, whereby the particles are arranged in a random close packing (packing fraction φ_1). The volumes of packed bed V_{rcp} and of the free (excess) fluid are indicated.

From eq. (20) one can see that when a monosized packing becomes bimodal, the packing fraction increases, likewise when particles of large size ratio are combined (previous section). Mangelsdorf and Washington (1960) already expressed the increased packing fraction, by combining to particles sizes with small size, in terms of reduced void fraction of the packed bed and created excess volume. Hence, less fluid is needed to fill the voids and excess fluid is created when the size ratio is not unity. This means that a packed bed of monosized particles, i.e. $\Phi = \varphi_1$, becomes a suspension when $u > 1$. In Figure 2, this case corresponds to $V_f - V_{rcp}(1 - \varphi_1) = 0$ and $V_{rcp} = V_T/\varphi_1$: fluid volume V_f in the mixture is just sufficient to fill the voids of the close packed particles which have volume V_T.

When $u > 1$, the packed bed contracts, and the excess fluid becomes available to suspend the particles. This created excess fluid amounts

$$\Delta V_f = \frac{\varphi_T - \varphi_1}{\varphi_T} V_{rcp} = \frac{4\beta(1-\varphi_1)c_S c_L (u-1)V_T}{\varphi_1} + O((u-1)^2) \quad , \tag{21}$$

see eq. (20). The particle volume fraction then reads

$$\Phi = \frac{V_T}{V_T + V_f + \Delta V_f} = \varphi_1 - 4\beta\varphi_1(1-\varphi_1)c_S c_L(u-1) + O((u-1)^2) \quad , \tag{22}$$

as $V_T/(V_f + V_T) = \Phi = \varphi_1$. For a suspension, so when $V_f > V_1(1 - \varphi_1)/\varphi_1$ and hence $\Phi < \varphi_1$, see Figure 2, the reduction in particle volume fraction by letting $u > 1$, follows from

$$\Phi = \frac{V_T}{V_T + V_f + \Delta V_f} = \Phi\left(1 - 4\beta\frac{\Phi}{\varphi_1}(1-\varphi_1)c_S c_L(u-1)\right) + O((u-1)^2) \quad , \tag{23}$$

as $V_T/(V_f + V_T) = \Phi$. In other words, eq. (22) is the special case of eq. (23) when $\Phi = \varphi_1$, so a monosized random close packing as starting situation. Now, the bimodal stiffening function can expanded for $u \approx 1$:

$$\mu = H(\Phi) - \frac{4\beta(1-\varphi_1)c_S c_L(u-1)\Phi^2}{\varphi_1}\left.\frac{dH}{d\Phi}\right|_{\Phi} + O((u-1)^2) \quad . \tag{24}$$

In the previous subsection equivalent eq. (19) was derived, based on the model of Farris for u → 1. Both models and resulting equations will be combined in the next subsection.

Stiffening function H(Φ)

The bimodal relative viscosity is governed by both the Farris concept (eq. (19)) and by the excess fluid volume consideration (eq. (24)). Equating both equations, ignoring $(u - 1)^2$ and higher terms, and substituting x_S/Φ and x_L/Φ for c_S and c_L, respectively, yields

$$\omega\left(\left.\frac{dH}{d\Phi}\right|_{\Phi} - \frac{C_1}{1-\Phi}H(\Phi)\right) = \frac{4\beta(1-\varphi_1)x_L}{\varphi_1}\left.\frac{dH}{d\Phi}\right|_{\Phi} \quad , \tag{25}$$

and it can be seen that both $u - 1$ and x_S have cancelled out from the first-order terms. This implies that by combining both expansions (eqs. (19) and (24)), the actual bimodal character of the particle mix, governed by size ratio u and composition x_S (or x_L) is irrelevant.

In the limit of u → 1 and Φ tending to φ_1 (and hence $x_L \to \varphi_1 - x_S$), both dH/dΦ and H(Φ) tend to infinity, but dH/dΦ dominates H and H(Φ)/(dH/dΦ) tends to zero, i.e. the second term on the left-hand side of eq. (24) can be ignored. This feature of the stiffening function H(Φ) is confirmed by eqs. (2) and (3), and will here be verified *a posteriori* too. This insight implies that

$$\omega = \frac{4\beta(1-\varphi_1)(\varphi_1 - x_S)}{\varphi_1} \quad . \tag{26}$$

In case $x_S = 0$ and hence $x_L = \Phi$, $\omega = 4\beta(1 - \varphi_1)$, and combining eqs. (25) and (26) now yield as governing differential equation of the mono-sized system in the entire concentration range

$$\left(\left.\frac{dH}{d\Phi}\right|_{\Phi} - \frac{C_1}{1-\Phi} H(\Phi) \right) = \frac{\Phi}{\varphi_1} \left(\left.\frac{dH}{d\Phi}\right|_{\Phi} \right) \quad . \tag{27}$$

Separation of the variables H and Φ, integration and application of $H(\Phi = 0) = 1$ yields

$$H(\Phi) = \left(\frac{1-\Phi}{1-\dfrac{\Phi}{\varphi_1}} \right)^{\frac{C_1\varphi_1}{1-\varphi_1}} \quad . \tag{28}$$

This equation is an analytical expression for the unimodal stiffening function, and is derived employing theoretical considerations only. It contains two parameters, the first-order virial coefficient C_1 of the considered particle shape ($C_1 = 2.5$ for spheres, the Einstein result), and the random close packing fraction φ_1 of the considered particle ($\varphi_1 \approx 0.64$ for spheres). Hydrodynamic effects are accounted for by C_1 only, governing the single particle hydrodynamics, and the remaining part of the model is governed by geometric considerations. The stiffening function diverges when the particle concentration Φ approaches φ_1.

The derivation presumed that $H(\Phi)/(dH/d\Phi) \rightarrow 0$ for $\Phi \rightarrow \varphi_1$. From eq. (28) it readily follows that this condition is met. It also follows that in the entire concentration range $0 \leq \Phi < \varphi_1$, $dH/d\Phi > C_1H(\Phi)/(1 - \Phi)$, so that the last two terms on the right-hand side of eq. (19) imply a viscosity reduction indeed.

Comparison with experimental data

For one case, $x_S = 0.25$, Farris (1968) presented stiffening functions versus x_T for various u^{-1} ("Figure 4"), and values of the stiffening factor f against the inverse size ratio u^{-1} ranging from zero to unity follow. In Table I these values of f versus u^{-1} are summarized, and they are set out in Figure 3. From this data, $\omega = 0.18$ can be derived (eq. (14)). Substituting $\varphi_1 = 0.61\text{-}0.64$, $\beta = 0.20$ and $\varphi_S = 0.25$, the right-hand side of eq. (26) also yields 0.18 (actually 0.176-0.184). This comparison indicates that Farris' concept for interacting sizes is valid up to the situation of random close packing, and that ω is related to φ_1, β and x_L indeed, see eq. (26).

Next, the obtained stiffening function is compared with experimental data unimodal suspensions, from dilute to concentrated (close to divergence). In Figure 1 measured relative viscosity values are set out, taken from Vand (1948b), Lewis and Nielsen (1968) and Rodriguez et al. (1992).

Vand (1948b) and Lewis and Nielsen (1968) used glass spheres of a very narrow distribution for their viscosity measurements. Rodriguez et al. (1992) used monodisperse samples of crosslinked polystyrene microgels dispersed in bromoform, and determined the zero-shear viscosity. These suspensions were found to take the same φ_1 as in macroscopic random close packings (e.g. of glass spheres). From Figure 1 it follows that all three sets of measured relative viscosities closely agree with each other in the entire concentration range.

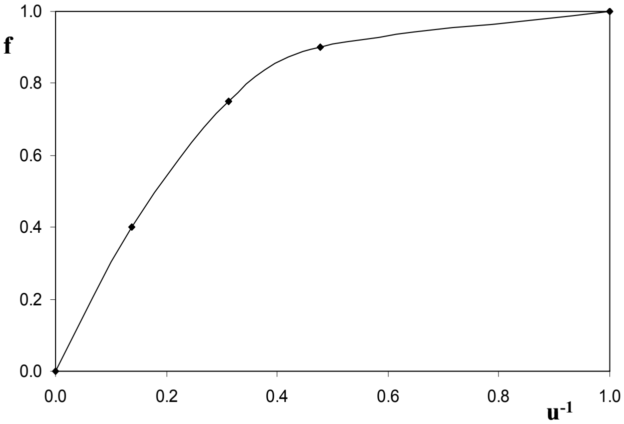

Figure 3 Values of crowding factor f versus inverse size ratio u^{-1}, taken from Table I. Lines are drawn to guide the eye.

u^{-1}	f
1	1
0.477	0.9
0.313	0.75
0.318	0.4
0	0

Table I Crowding factor f versus inverse size ratio u^{-1} as extracted from “Figure 4” from Farris (1968).

For high sphere loads, $\Phi > 0.4$, eqs. (2) and (3) overestimate and underestimate, respectively, the measured values. Eq. (2) could be better fit to the data by augmenting φ_1, but this also implies that divergence will take place at a packing fraction higher than pertaining to random close packing. In Figure 1, also eq. (28) is set out from $\Phi = 0$ to Φ approaching φ_1, with $\varphi_1 = 0.64$. One can see that in the full concentration range, eq. (28) and experiments are lying close together. In Table II the computed values are included, as well those computed with eq. (28) using $\varphi_1 = 0.61$. For the glass sphere experiments and moderate Φ sphere loads, one can see that eq. (28) with $\varphi_1 = 0.61$ yields better agreement. This limiting value of Φ was observed by Chong et al. (1971) and Van der Werff and De Kruif (1989).

Summary

In the present paper the relative viscosity of concentrated suspensions of monosized and multimodal rigid particles, consisting of equally shaped particles, at zero shear rate is addressed. In the dilute limit, the hydrodynamics of the individual particle prevails, governed by the first-order coefficient C_1 (eq. (1)), which takes the well-known Einstein value of 2.5 for spheres. When particle interactions cannot be ignored anymore, it is known that for particles with large size ratios, the viscosity increase can be described by considering geometric considerations only.

To obtain an exact equation for the monosized particle viscosity-concentration relation, i.e. the stiffening function; $H(\Phi)$, two approaches are followed. Basically, both are related to packing considerations of bimodal suspensions and packings of discretely sized particles with small size ratio u.

Using the random close packing fraction of such bimodal packings, which contract upon combining two sizes, a differential equation for the apparent fluid increase (eq. (23)) and associated viscosity reduction is derived (eq. (24)). It turns out that the viscosity of these discrete bimodal particle suspensions is governed by the size ratio u, the gradient of the stiffening function for the concentration considered (C_1 for a dilute system), φ_1 and β. The latter two parameters follow from the random close packing of the considered particle shape, φ_1 is the monosized packing fraction and β the packing fraction gradient when a unimodal packing turns into a bimodal packing. By Brouwers (2006) the parameter β has been derived and values listed, and here it turns out that the bimodal random close packing and related parameter β can be employed to quantify viscosity reduction.

The second line of reasoning follows the observation by Farris (1968) concerning the viscosity reduction by combining particles of different size ratios, so not large size ratios only. Farris also considered the case of interacting sizes and found that all bimodal suspensions can be described using the same geometric concept, whereby a crowding factor f (the part of the finer fraction that behaves as large fraction) depends on size ratio u only. Here, this concept is employed to derive a second differential equation (eq. (19)) that describes the viscosity of a monosized suspension at the onset of turning into a bimodal suspension. This expression contains the gradient of f versus u at $u = 1$, viz. ω, governing the gradient when a unimodal suspension becomes a bimodal suspension.

Both approaches yield two differential equations for the bimodal suspension viscosity for small $u - 1$, by combining both governing equations it follows that ω can be expressed in φ_1 and β (eq. (26)). Using data provided by Farris (1968), close agreement with this theoretical expression for ω is obtained.

Finally, using both equations that govern the monosized relative viscosity (stiffening function) at the onset of bimodal suspensions, using the relation between ω on the one hand and φ_1 and β one the other, a governing differential equation (eq. (27)) for the stiffening function $H(\Phi)$ is derived, and solved in closed-form (eq. (28)). The resulting analytical expression is solely governed by C_1 and φ_1. The resulting stiffening function predictions are found to be in good quantitative agreement with the classical experiments by Vand (1948b), Lewis and Nielsen (1968) and Rodriguez et al. (1992).

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