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# Viscosity of concentrated noncolloidal bidisperse suspensions

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D. He (🖂) · N. N. Ekere School of Aeronautical and Mechanical Engineering, The University of Salford Salford M5 4WT, UK e-mail: d.he@salford.ac.uk **Abstract** At the same solid volume fraction ( $\Phi$ ) the relative viscosity ( $\eta_r$ ) of a concentrated noncolloidal bidisperse suspension of hard spherical particles is lower than that of a monodisperse suspension. In this paper a semi-analytical viscosity model of noncolloidal bidisperse suspensions is derived using an integration method. In this model the random loose packing density obtained by computer simulation is taken as the limit of solid volume fraction  $\Phi_m$  which depends upon both the diameter ratio ( $\lambda$ ) of large to small particles and the volume fraction of large particles ( $\xi = \Phi_l/\Phi$ ). This model shows that at high solid

volume fraction,  $\Phi > 0.40$ , both  $\lambda$  and  $\xi$  significantly influence  $\eta_r$ . For example, at  $\Phi = 0.5$ , it predicts that for monodisperse suspensions  $\eta_r = 70$ , while for bidisperse suspensions ( $\lambda = 2$  and  $\xi = 0.7$ )  $\eta_r = 40$ . Comparison shows that, at high solid volume fraction (0.4–0.5), the relative viscosity predicted by this model is in good agreement with that predicted by the work of Shapiro and Probstein (1992) and of Patlazhan (1993), but is higher than that predicted by the work of others.

**Key words** Concentrated suspensions · Bidisperse spherical particles · Rheology · Viscosity

### Introduction

In the study of the rheological properties of concentrated monodisperse suspensions of hard spherical particles, one of the principal objectives is to understand the influences of particle size d and solid volume fraction  $\Phi$ on the viscosity. The flow of a concentrated suspension may be dominated by Brownian motion, surface chemical forces or by the hydrodynamic forces between nearest neighbouring particles. A detailed rheological classification of concentrated suspensions can be found in a recent review by Coussot and Ancey (1999). The hydrodynamic interacting forces between nearest neighbouring particles dominate the flow of a concentrated non-colloidal suspension. At a given shear rate the hydrodynamic forces are dependent on the gaps between the nearest neighbouring particles (Cox 1973), hence on the solid volume fraction  $\Phi$ . Thus, it is convenient to use the relative viscosity  $\eta_r$ , the ratio of suspension

viscosity to liquid viscosity, which allows the independent investigation of the effect of  $\Phi$  on  $\eta_r$ . Many experimental and theoretical studies on the viscosities of monodisperse suspensions have been reported in the literature. It is well understood that in a suspension there exists a limit of the solid volume fraction  $\Phi_m$  that is termed *viscosity percolation threshold* by Bicerano et al. (1999). When the solid volume fraction  $\Phi$  is lower than  $\Phi_m$ ,  $\eta_r$  increases with  $\Phi$ ; when  $\Phi$  approaches  $\Phi_m$ ,  $\eta_r$  tends to infinity; and when  $\Phi$  exceeds  $\Phi_m$ , the suspension becomes immobile and behaves as a solid.

The particles of many physical systems and industrial materials have size distributions, such as soil, coal slurry, paint pigment (Rumpf 1989), metal and ceramic powders (Shiau et al. 1997). In practise, it has been found that high solid volume fractions can be attained without significant increase in viscosity by using suspensions of polydisperse particles rather than monodisperse particles. This finding is important in many applications

where high solid loads are demanded but where reasonably low viscosities are desirable from the processing viewpoint, such as in the injection moulding of metal or ceramic pastes, the transportation of coal slurry and the use of dental pastes. The above facts underline the importance of the need to study the influence of particle size distribution on the viscosity. The viscosities of bidisperse suspensions of hard spheres have been studied experimentally by many researchers (Sweeny and Geckler 1954; Eveson 1959; Chong et al. 1971; Poslinski et al. 1988; Storms et al. 1990; Hoffman 1992; Shapiro and Probstein 1992; Logos and Nguyen 1996; Condred and Petit 1997). The results show that the main factors which influence the viscosities of bidisperse suspensions are the diameter ratio of large to small particles,  $\lambda = d_l/d_s$ , and the volume fraction of large particles,  $\xi = \Phi_I/\Phi$ . Sweeny and Geckler (1954) investigated the effects of  $\lambda$  and  $\xi$  on  $\eta_r$  by experiment. The diameter of large particles was fixed at 262  $\mu$ m and  $\lambda$  was varied from 1 to 20.76. They found that, at  $\Phi = 0.55$ , the viscosity of a bidisperse suspension with  $\lambda = 20.76$  and  $\xi = 0.75$  was less than 25% of that of a monodisperse suspension. Logos and Nguyen (1996) obtained similar results on coal slurries. For bidisperse coal slurry with 40% of coarse particles (208–279  $\mu$ m) and 60% of fine particles (smaller than 45  $\mu$ m) the viscosity was about 20% of that of slurry containing fine particles only. Eveson (1959) observed that, with  $\lambda < 8$  and  $\Phi < 0.2$ , the change in  $\eta_r$  was less than 5%. This means that at low solid volume fraction the influence of particle size distribution is insignificant. Gondret and Petit (1997) measured the dynamic viscosity of noncolloidal bidisperse suspensions. Their results show that, at finite frequency, the effects of  $\xi$  and  $\lambda$  on the dynamic viscosity are similar to the effects on the viscosity. Chang and Powell (1993) and Toivakka and Eklund (1996) confirmed the experimental results using Stokesian dynamics simulation. The experimental and simulation findings can be summarised in two parts. First, as  $\Phi$  and  $\xi$  are fixed,  $\eta_r$  decreases with the increase in  $\lambda$ . Second, when  $\Phi$ and  $\lambda$  are fixed, there exists a critical value of  $\xi$  at which  $\eta_r$  is minimised. Lower or higher than this value leads to the increase in  $\eta_r$ . Many investigators agree that  $\eta_r$  is minimised at  $\xi = 0.65 \sim 0.85$  (Sweeny and Geckler 1954; Chong et al. 1971; Hoffman 1992) and few agree that  $\eta_r$  is minimised at  $\xi = 0.5$  (Shapiro and Probstein 1992) or  $\xi = 0.4$  (Logos and Nguyen 1996).

For bidisperse suspensions, although the effects of  $\lambda$  and  $\xi$  on  $\eta_r$  have been well known from experimental observations, only a few empirical models are available in the literature (see next section). Therefore, more work is still needed to understand fully these effects. In this paper we propose a viscosity model of concentrated non-colloidal bidisperse suspensions. In the next section we review representative viscosity models of both monodisperse and bidisperse suspensions; by analysing

these models we derive the new model and discuss the physical meanings of the parameters involved. Then we apply this model to examine the effects of  $\lambda$  and  $\xi$  on the viscosity. The final section presents our conclusions.

# The new viscosity model

Review of viscosity models

Early this century, Einstein (1956) studied the energy dissipation due to particle motion in infinitely dilute suspensions and developed a theoretical relative viscosity model as

$$\eta_r = 1 + 2.5\Phi \quad \Phi \ll 1.0 \tag{1}$$

Following Einstein's work great efforts have been made to model the viscosity of concentrated suspensions theoretically. However, the complicated interactions between randomly suspended particles made it impossible to derive a closed theoretical model, since some empirical parameters had to be determined from measured data. For monodisperse suspensions, many viscosity models have been developed. A couple of representative semianalytical models are selected from the literature and listed in Table 1. The Frankel-Acrivos model and the Sengun-Probstein model were derived under the assumption that the hydrodynamic interactions (the lubrication forces) between nearest neighbouring particles dominate the suspension flow. So these two models are only applicable to suspensions of high concentration ( $\Phi$  > 0.2). Taking the limit  $\Phi \rightarrow 0.0$ , we find that the Arrhenius model, the Mooney model and the Krieger-Dougherty model all converge to the Einstein equation  $\eta_r = 1 +$  $2.5\Phi$ . This implies that these models could originate from a common analytical technique. Indeed Ball and Richmond (1980) and Sudduth (1993) found that these models have a common differential form as

$$d\eta_r = \eta_r [\eta] (1 - k\Phi)^{-\alpha} d\Phi \tag{2}$$

where k is the crowding factor which is inversely proportional to the limit of solid volume fraction  $\Phi_m$ ,  $[\eta]$  is the relative intrinsic viscosity and  $\alpha$  is a constant. The physical meaning of Eq. (2) may be explained as follows. First, the effect of all particles on the viscosity is the sum of the effects of particles added to the suspension sequentially. Second, at high solid volume fraction, when a small portion of particles  $d\Phi$  is added to the suspension, the particles already in the suspension suffer a crowding effect that can cause an increase in viscosity  $d\eta_r$  which is proportional to  $(1 - k\Phi)^{-\alpha}$ . Thus the higher the solid volume fraction the stronger this crowding effect. Taking  $k = 1/\Phi_m$  and  $[\eta] = 2.5$ , we see that the integral of Eq. (2) becomes the Arrhenius model if  $\alpha = 0$ , the Krieger-Dougherty model if  $\alpha = 1$ , and the Mooney model if  $\alpha = 2$ . The model developed by Kitano

Table 1	Relative	viscosity	7
models			

Author(s)	Viscosity models
Arrhenius (1917)	$\eta_r = e^{2.5\Phi}$
Mooney (1951)	$\eta_r = e^{[2.5\Phi_m/(1-\Phi/\Phi_m)]}$
Krieger and Dougherty (1959)	$\eta_r = \left(1 - \Phi/\Phi_m\right)^{-2.5\Phi_m}$
Kitano et al. (1981)	$\eta_r = (1 - \Phi/\Phi_m)^{-2}$
Frankel and Acrivos (1967)	$\eta_r = \frac{9}{8} \frac{(\Phi/\Phi_m)^{1/3}}{1 - (\Phi/\Phi_m)^{1/3}}$
Sengun and Probstein (1989)	$eta = rac{(\Phi/\Phi_m)^{1/3}}{1 - (\Phi/\Phi_m)^{1/3}}$
	$\eta_r = 1 + \frac{3\pi}{8} \frac{\beta}{\beta+1} \left[ \frac{3+4.5\beta+\beta^2}{\beta+1} - \frac{3\beta+3}{\beta} \ln(\beta+1) \right]$

et al. can also be derived from Eq. (2) if  $k = 1/\Phi_m$ ,  $[\eta] = 2/\Phi_m$  and  $\alpha = 1$ .

For a bidisperse suspension with  $\lambda$  greater than ten, Farris (1968) used the following method to predict the viscosity. The suspension was initially assumed to contain the fine particles only and a monodisperse suspension model was employed to calculate its viscosity. Then the fine suspension was treated as a continuous phase and the coarse particles were added to it. Finally, the monodisperse suspension model was employed again to predict the viscosity of the bidisperse suspension.

Chong et al. (1971) correlated their experimental data, as well as those of Sweeny and Geckler (1954), by the following equation:

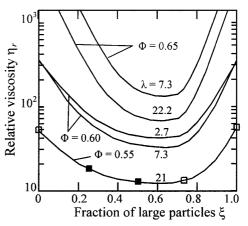
$$\eta_r = \left(1 + 0.75 \frac{\Phi/\Phi_m}{1 - \Phi/\Phi_m}\right)^2 \tag{3}$$

where  $\Phi_m$  depends upon both  $\lambda$  and  $\xi$  and was determined empirically. The effects of  $\lambda$  and  $\xi$  on  $\eta_r$  of Chong and Sweeny's work are shown in Fig. 1. A similar model was proposed by Storms et al. (1990) as

$$\eta_r = \left(1 + \frac{R\Phi}{1 - \Phi/\Phi_m}\right)^{3.3\Phi_m} \tag{4}$$

where R varies from 0.7 to 1.25 depending upon  $\xi$  and  $\lambda$ ,  $\Phi_m$  is also dependent upon  $\xi$  and  $\lambda$  and was determined from a series of algebraic expressions.

Shapiro and Probstein (1992) measured the viscosity of bidisperse suspensions of glass beads ( $40 \sim 160~\mu m$ ). They found that if  $\lambda$  was not too far from one ( $\lambda \leq 4$  in their measurements), the viscosity of bidisperse suspensions could be predicted by the monodisperse model developed by Sengun and Probstein (1989) (see Table 1). The measurements showed that, for different  $\lambda$  and  $\xi$ , the ratio of the close random packing density to the limit of solid volume fraction was a constant that was equal to 1.19. Thus, they predicted the limits of the solid volume fractions from the measured random close packing densities of bidisperse particles. A least-squares error method was used to estimate the constant C which varied from 1.35 to 1.58 depending upon  $\lambda$  and  $\xi$ .



■ Sweeny & Geckler (1954) □ Chong et al. (1971) —— Prediction by Eq. (3)

**Fig. 1** Effects of  $\lambda$  and  $\xi$  on relative viscosity by Chong et al. ( $\blacksquare$  Sweeny and Geckler 1954;  $\square$  Chong et al. 1971;  $\longrightarrow$  Prediction by Eq. 3)

Since only one parameter  $\Phi_m$  is involved in Eq. (3) it can be seen that for a given  $\lambda$ ,  $\eta_r$  decreases to a minimum as  $\Phi_m$  increases to a maximum at the same value of  $\xi$ . While both Eq. (4) and the Sengun-Probstein model have two parameters,  $\Phi_m$  and R, or  $\Phi_m$  and C, the minimal  $\eta_r$  and the maximal  $\Phi_m$  may not correspond to the same  $\xi$ . For example, applying the Sengun-Probstein model with  $\lambda = 4$ ,  $\Phi_m$  is maximised at  $\xi = 0.65$  but  $\eta_r$  is minimised at  $\xi = 0.5$  (Shapiro and Probstein 1992).

Another interesting point is that, in the limit  $\Phi \rightarrow 0.0$ , if  $\Phi_m = 0.6$  in Eq. (3) and  $\Phi_m = 0.606$  and R = 1.25 in Eq. (4), then both models also converge to the Einstein equation. This implies that Eqs. (3) and (4) are also applicable to monodisperse suspensions if the above values are employed.

#### The new model

Although the models developed by Arrhenius, Krieger-Dougherty, Mooney, and by Kitano et al. were applied

to monodisperse suspensions, the physical meanings of Eq. (2) as explained above are tenable to bidisperse suspensions if  $\lambda$  is not too far from one. Therefore, it is expected that if the effects of  $\lambda$  and  $\xi$  are fully taken into account in the crowding factor  $k = 1/\Phi_m$ , the relationship between  $\eta_r$  and  $\Phi$  of bidisperse suspensions can also be expressed by Eq. (2). This is evidenced by the fact that the Sengun-Probstein model of monodisperse suspensions was successfully applied to bidisperse suspensions (Shapiro and Probstein 1992). Even if C is taken to be a single value in the Sengun-Probstein model, 1.45 for example, the maximal relative error is less than 10% because the effect of C on  $\eta_r$  is linear. It is also evidenced by the fact that, as reviewed above, the bidisperse suspension models, Eqs. (3) and (4), are both applicable to monodisperse suspensions. Thus, we may apply Eq. (2) to bidisperse suspensions.

In Eq. (2),  $\alpha$  is a very sensitive parameter that can dramatically influence the viscosity. Taking  $\Phi_m = 0.6$  and  $\Phi = 0.5$  for example, by the Krieger-Dougherty model ( $\alpha = 1$ )  $\eta_r$  is lower than 15.0, while by the Mooney model ( $\alpha = 2$ )  $\eta_r$  is higher than 1800; both are far from experimental results. If we take  $\alpha$  as a decimal parameter in the range  $1.0 < \alpha \le 2.0$ , then the integral of Eq. (2) becomes

$$\eta_r = e^{\left[(1 - \Phi/\Phi_m)^{-\alpha + 1} - 1\right] \frac{2.5\Phi_m}{\alpha - 1}} \tag{5}$$

Taking  $\Phi_m = 0.6$ , Fig. 2 shows the influence of  $\alpha$  on  $\eta_r$ . For comparison, values of  $\eta_r$  predicted by other models are also plotted. In Eq. (5), there are two parameters,  $\Phi_m$  and  $\alpha$ , whose physical meanings may be discussed as follows. The maximum solid volume fraction  $\Phi_m$  represents the effect of particle size distribution on  $\eta_r$ . For bidisperse suspensions  $\Phi_m$  is dependent upon  $\lambda$  and  $\xi$ . The constant  $\alpha$  represents the effects of the absolute particle size and the dominating force on  $\eta_r$ . As stated earlier, the hydrodynamic force dominates

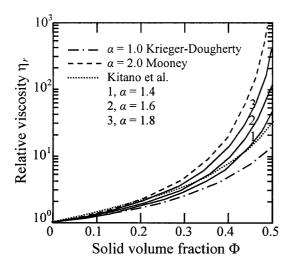


Fig. 2 Effect of  $\alpha$  on relative viscosity as calculated by Eq. (5)

the flow of concentrated noncolloidal suspensions and the relative viscosity is independent of the absolute particle sizes. Therefore, in Eq. (5)  $\alpha$  can be taken as a constant of the same value for any combination of  $\lambda$  and  $\xi$ . In contrast, for suspensions of very fine particles, such as colloidal particles ( $d < 10~\mu m$ ), the surface chemical forces and electrostatic forces, which depend upon absolute particle sizes, may significantly influence the viscosity. For example, at  $\Phi = 0.5$ , Greenwood et al. (1997) found that the viscosity of a monodisperse polymer latex with d = 84 nm was about seven times higher than that of latex with d = 434 nm.

For monodisperse suspensions, there is considerable variation in the reported limit of solid volume fraction  $\Phi_m$  depending upon the particle arrangements, from simple cubic packing  $\Phi_m = 0.524$  (Shapiro and Probstein 1992) to near face-centred hexagonal packing  $\Phi_m = 0.72$ (de Kruif et al. 1985). The commonly accepted value of the random packing density of monodisperse particles ranges from 0.59 to 0.63. Scott (1960) obtained the upper limit by shaking the packing beds and extrapolating the data measured in finite soft containers. Such a high value may not be attainable in a concentrated suspension because both the viscosity and the mass density of the liquid medium are much higher than that of the air. We agree that, for randomly suspended particles, the random loose packing density, which is about 0.59 to 0.60, should be the reasonable estimation of  $\Phi_m$  (Chong et al. 1971; Ball and Richmond 1980; Storms et al. 1990; Sudduth 1993). Taking  $\Phi_m = 0.595$ and applying the least-squares error method to fit the viscosities of monodisperse suspensions measured by other investigators to Eq. (5) we obtain  $\alpha = 1.45$ ; then Eq. (5) becomes

$$\eta_r = e^{5.56\Phi_m \left[ (1 - \Phi/\Phi_m)^{-0.45} - 1 \right]} \tag{6}$$

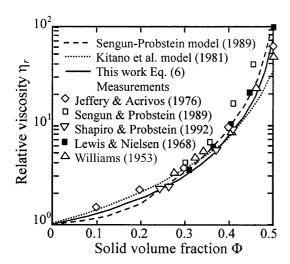
Figure 3 shows good agreement of this model with those developed by Sengun and Probstein and by Kitano et al., and with the results measured by other investigators. As discussed earlier, the effects of  $\lambda$  and  $\xi$  on  $\eta_r$  are fully taken account into  $\Phi_m$ ; hence Eq. (6) can be applied to bidisperse suspensions.

# Influences of $\lambda$ and $\xi$ on $\eta_r$

To apply Eq. (6) to bidisperse suspensions, the key is to obtain  $\Phi_m$  as a function of  $\lambda$  and  $\xi$ . It was suggested (Sudduth 1993; Greenwood et al. 1997) that the maximum packing density of bidisperse particles may be calculated by

$$\Phi_m = \Phi_{lm} + (1 - \Phi_{lm})\Phi_{lm} \tag{7}$$

where  $\Phi_{lm}$  is the random packing density of monodisperse particles. Thus  $\Phi_{lm} = 0.6$  leads to  $\Phi_m = 0.84$  and



**Fig. 3** Comparison of the new model with other models and measurement results ( $\diamondsuit$  Jeffery and Acrivos 1976;  $\square$  Sengun and Probstein 1989;  $\triangledown$  Shapiro and Probstein 1992; ■ Lewis and Nielsen 1968;  $\Delta$  Williams 1953)

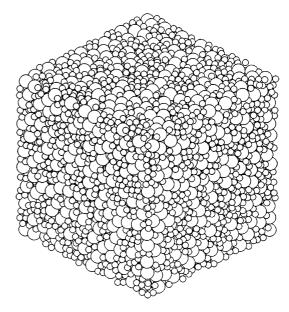
 $\xi = \Phi_{lm}/\Phi_m = 0.714$ . Equation (7) implies that only the large particles are used to obtain the packing density  $\Phi_{lm}$  first; then the interstices between the large particles are taken as an integral space to pack the small particles that gives an additional packing density  $(1 - \Phi_{lm})\Phi_{lm}$ . Such an assumption is not acceptable even if the particle size ratio  $\lambda$  is large enough to allow the small particles to fill the interstices created by the large particles. If each interstice amongst the large particles is filled by one small particle, then it is expected that in a packing the number of small particles equals the number of large particles. Then the packing density is given by

$$\Phi_m = \Phi_{lm} + \lambda^{-3} \Phi_{lm} \tag{8}$$

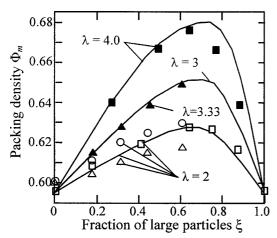
If  $\Phi_{lm} = 0.6$  and  $\lambda = 4$ , then by Eq. (8)  $\Phi_m < 0.61$  which is much lower than that given by Eq. (7). Therefore, Eq. (8) is also unacceptable.

We have developed a computer simulation model for the random loose packing of bidisperse particles (He et al. 1999). This model randomly places 10,000 particles within a cubic domain. For given values of  $\lambda$ and  $\xi$  the particle diameters are decided by sampling. In the initial positions many particles overlap with each other. A relaxation iteration is then applied to reduce the overlap values; meanwhile the packing domain is gradually expanded. The simulation is completed when the mean overlap value falls below a pre-set tolerance, which is  $10^{-4}$  times the radius of the small particles. Further reducing the tolerance has little influence on the simulation results, e.g. the relative change in the packing density is smaller than 1%. Figure 4 shows the threedimensional view of one simulation result. Figure 5 plots the random loose packing density as a function of  $\lambda$  and  $\xi$ . The random packing densities of bidisperse particles predicted by Ouchiyama and Tanaka (1981) and by Song et al. (1997) are also plotted. Shapiro and Probstein (1992) found that for both monodisperse and bidisperse suspensions the ratio of the random close packing density to the to the limit of the solid volume fraction is a constant of 1.19. For monodisperse particles the ratio of the random loose packing density to the random close packing density is about 0.95. For comparison, we scale the random close packing densities of bidisperse suspensions given by Shapiro and Probstein with this value. One can see that our simulation results are in good agreement with those of others.

Taking the random loose packing density from Fig. 5 as the limit of solid volume fraction  $\Phi_m$  in Eq. (6), Fig. 6 shows the influences of  $\lambda$  and  $\xi$  on  $\eta_r$  at  $\Phi = 0.5$ . For the range of solid volume fraction  $0.4 \le \Phi \le 0.5$ , Fig. 7 plots  $\eta_r$  against  $\Phi$ . It is seen from Figs. 5 and 6 that the value of  $\xi$ , at which  $\Phi_m$  is maximised and  $\eta_r$  is minimised, varies with  $\lambda$ . For example, with  $\lambda = 1.5$ ,  $\Phi_m$  is maximised and  $\eta_r$  is minimised at  $\xi$  around 0.6, while with  $\lambda = 3.0$  and 4.0,  $\xi$  is about 0.75. This agrees with the conclusion of other investigators that  $\xi$  is greater than 0.5 and in the range of our simulation  $(\lambda \le 4) \xi$  increases with  $\lambda$ . Figures 6 and 7 also show that our model is in good agreement with that of Shapiro and Probstein (1992) and of Patlazhan (1993). Comparing our results with the work of others we can observe qualitatively the similar effects of  $\lambda$  and  $\xi$  on the relative viscosity. However, the values of  $\eta_r$  predicted by our model, and by the work of Shapiro and Probstein, and of Patlazhan are higher than the results predicted by others as shown in Fig. 1. There may be two reasons for this. The first is the different value of  $\Phi_m$  used by

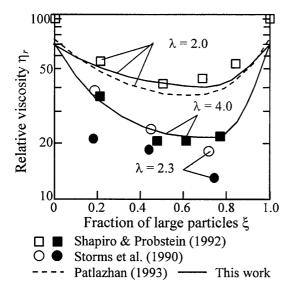


**Fig. 4** Random loose packing of bidisperse particles,  $\lambda = 2.0$  and  $\xi = 0.7$ 



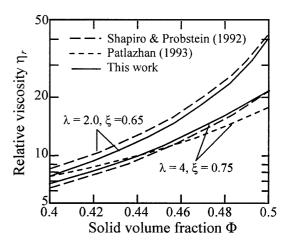
- ☐ Scaled results from Shapiro and Probstein (1992)
- O Prediction by Song at al (1998)
- ▲△ Prediction by Ouchiyama and Tanaka (1981)
- Random loose packing (He et al. 1999)

Fig. 5 Random packing density as function of  $\lambda$  and  $\xi$  ( $\blacksquare$ ,  $\square$  scaled results from Shapiro and Probstein 1992;  $\bigcirc$  prediction by Song et al. 1997;  $\blacktriangle$ , $\triangle$  prediction by Ouchiyama and Tanaka 1981; — random loose packing He et al. 1999)



**Fig. 6** Relative viscosity as function of  $\lambda$  and  $\xi$ ,  $\Phi = 0.5$  ( $\square$ ,  $\blacksquare$  Shapiro and Probstein 1992;  $\bigcirc$ ,  $\blacksquare$  Storms et al. 1990; — Patlazhan 1993; — this work)

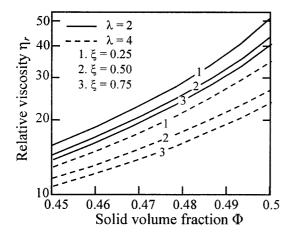
different investigators, which ranges from 0.524 to 0.72 for monodisperse particles. At the same solid volume fraction  $\Phi$ , the higher the solid limit  $\Phi_m$  the lower the relative viscosity  $\eta_r$ . As discussed in the previous section, the random close packing density may not be attainable in concentrated suspensions. Onoda and Liniger (1990) pointed out that for monodisperse suspensions, glass transition may happen as the solid volume fraction exceeds 0.555. We are concerned only with suspensions



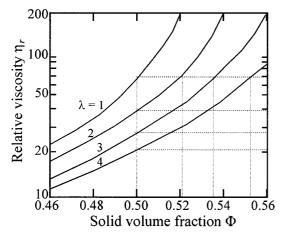
**Fig. 7**  $\eta_r$  as function of  $\Phi$  and comparison of different models (long dashes Shapiro and Probstein 1992; short dashes Patlazhan 1993; continuous line this work)

in which the particles are randomly suspended; therefore, we take the random loose packing density as the limit of solid volume fraction. The second reason may be that in viscosity measurement wall slip occurs at the solid boundaries which leads the measured viscosity to be lower than the truth (Cheng 1984; Jana et al. 1995). The effect of wall slip on the viscosity measurement is dependent on the geometry of the instrument; the viscosity of the same sample measured on different viscometers can be different. At high solid volume fractions, the effect of wall slip becomes more significant and difficult to control.

To examine the influences of  $\lambda$  and  $\xi$  on  $\eta_r$  at different solid volume fractions further, with  $\lambda = 2.0$  and 4.0 Fig. 8 plots  $\eta_r$  against  $\Phi$  at different values of  $\xi$ . With  $\xi = 0.75$ , Fig. 9 plots  $\eta_r$  against  $\Phi$  at different values of  $\lambda$ . Figure 8 shows that at high solid volume fraction  $\xi$ significantly influences the viscosity. Taking  $\Phi = 0.5$  for example, with  $\lambda = 2$  the relative viscosity at  $\xi = 0.25$  is about 1.3 times that at  $\xi = 0.65$ , and with  $\lambda = 4$  the relative viscosity at  $\xi = 0.25$  is about 1.7 times that at  $\xi = 0.75$ . Figure 9 shows that the use of bidisperse suspensions can provide two advantages. One is that, at a given solid volume fraction, the viscosity of a bidisperse suspension is lower than that of a monodisperse suspension and this effect becomes more significant as  $\lambda$  increases. Taking  $\Phi = 0.5$ , for example, the relative viscosity of a bidisperse suspension with  $\lambda = 2.0$ and  $\xi = 0.7$  is about 60% of that of a monodisperse suspension, while when  $\lambda = 4.0$  and  $\xi = 0.75$ ,  $\eta_r$  is only about 30% of that of a monodisperse suspension. The other advantage is that, for a desirable viscosity value, the use of a bidisperse suspension can attain a higher solid volume fraction than the use of a monodisperse suspension. For example, the viscosity of a bidisperse suspension with  $\lambda = 2.0$  and  $\Phi = 0.52$ , or  $\lambda = 4.0$  and  $\Phi = 0.55$ , equals the viscosity of a monodisperse suspen-



**Fig. 8** Effect of  $\xi$  on relative viscosity



**Fig. 9** Effect of  $\lambda$  on relative viscosity,  $\xi = 0.75$ 

sion with  $\Phi = 0.50$ . These advantages can benefit many industrial applications where suspensions of high solid volume fractions are demanded but reasonably low viscosities are desirable from the processing viewpoint.

Our model shows that as the solid volume fraction  $\Phi$  is lower than 0.3 the influences of  $\lambda$  and  $\xi$  on  $\eta_r$  become insignificant, which agrees with experimental results and other predictions. Although our model is derived using an integration technique, it coincides with the model derived using lubrication concept (Shapiro and Probstein 1992) in predicting the significant influences of  $\lambda$  and  $\xi$  on  $\eta_r$  at high solid volume fraction. In a suspension the mean rel-

ative gap between nearest neighbouring particles is estimated by  $\delta/d_{\rm a} = [(\Phi_m/\Phi)^{1/3}-1]$  (Hoffmann and Kevelam 1999), where  $\delta$  is the average gap and  $d_{\rm a}$  is the average diameter of particles. Then at  $\Phi=0.5$ , for monodisperse particles ( $\Phi_m=0.595$ )  $\delta/d_{\rm a}=0.060$ , and for bidisperse particles with  $\lambda=4$  and  $\xi=0.75$  ( $\Phi_m=0.68$ )  $\delta/d_{\rm a}=0.108$ . With very narrow gaps, the lubrication forces between nearest neighbouring particles significantly decrease as  $\delta/d_{\rm a}$  increases (Cox 1973). Therefore, the relative viscosity of the bidisperse suspension is significantly lower than that of the monodisperse suspension.

For high value of  $\lambda$ , higher than 5 for example, its influence on  $\eta_r$  is still an open question. In bidisperse suspensions if  $\lambda$  is very high the small particles are likely to fill the voids among large particles or settle down. Such a structure may have significant influence on the viscosity of the suspensions.

## Summary

In this paper a relative viscosity model of concentrated bidisperse suspensions of noncolloidal particles was derived. The random loose packing density obtained by a collective rearrangement simulation model was accepted as the limit of solid volume fraction. The influences of particle size ratio  $\lambda$  and the volume ratio of large particles to total particles  $\xi$  on the relative viscosity were investigated. At high solid volume fraction, results showed that both  $\lambda$  and  $\xi$  significantly influence  $\eta_r$ , which can be summarised in two parts. First, as  $\Phi$  and  $\xi$ are fixed,  $\eta_r$  decreases with the increase in  $\lambda$ . Second, when  $\Phi$  and  $\lambda$  are fixed,  $\eta_r$  decreases first as  $\xi$  increases from zero, reaches a minimum as  $\xi$  is around 0.60–0.75, then increases as  $\xi$  approaches unity. The value of  $\xi$  at which  $\eta_r$  is minimised varies with  $\lambda$ . At low solid volume fraction, the influences of  $\lambda$  and  $\xi$  on  $\eta_r$  are both insignificant. Results of this work can be used to obtain concentrated suspensions of higher solid volume fraction without significant increase in viscosity by controlling the particle size ratio and the volume ratio of large particles to total particles.

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