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A phenomenological approach to suspensions with viscoelastic matrices

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Abstract

A simple constitutive model for viscoelastic suspensions is discussed in this paper. The model can be used to predict the rheological properties (relative viscosity and all stresses) for viscoelastic suspensions in shear and elongational flow, and the constitutive equations combine a "viscoelastic" behaviour component and a "Newtonian" behaviour component. As expected, the model gives a prediction of positive first normal stress difference and negative second normal stress difference; the dimensionless first normal stress difference strongly depends on the shear rate and decreases with the volume fraction of solid phase, but the dimensionless second normal stress difference (in magnitude) is nearly independent of the shear rate and increases with the volume fraction. The relative viscosities and all the stresses have been tested against available experimental measurements.

Keywords : constitutive model, viscoelastic suspension, relative viscosity, first normal stress difference, second normal stress difference, volume fraction

1. Suspension rheology

There are many experiments and many theories of suspension and filled material behaviour; see for example reviews by Metzner (1985) and Barnes (2003). However, most of the experimental work is concentrated on suspension viscosity behaviour and there is comparatively little on normal stresses. In addition, theoretical work is usually confined to low shear rates. However, recently there have been proposals to use suspension theories (Tanner, 2002; Tanner and Qi, 2005) for studies of flow of crystallizing polymers and a general constitutive model of suspensions useful for such purposes seems to be lacking. Here we present a phenomenological model that should be useful for these applications.

To study the constitutive relation of suspensions, several models have been proposed to describe the behaviour of concentrated suspensions in the recent years. Constitutive equations have been given by using these models, for example, a diffusive-flux model, which is developed in the model proposed by Leighton and Acrivos (1987), was given by Philips *et al.* (1992); a balance model, which is phenomenologically similar to the model proposed by Jenkins and McTigue (1992) (there is no diffusive equation), has been made by Nott and Brady (1994); and lubrication models based on the lubrication approximation are given by Phan-Thien (1995) and Phan-Thien *et al.* (1999). However, most of these models can be only used to describe the behaviour for high concentrated suspensions with a Newtonian matrix, but can not give the correct behaviour for viscoelastic suspensions. For instance, the second normal stress difference for viscoelastic suspensions is negative in experimental measurements, but the lubrication models give a positive second normal stress difference.

Therefore, the aim of this work is to find a model to describe the behaviour for viscoelastic suspensions and some comparisons have been made between the calculated results by using the model with experimental measurements. In future work, the model may be useful for describing polymer crystallization in shear and elongational flows.

2. Experimental data

We will concentrate to begin on suspensions of non-colloidal spherical particles, focusing on normal stress and elongational effects.

It is clear that the simple rule for increasing viscosity (η) with volume concentration of particles (φ) advocated by Krieger (1972) and Metzner (1985) is generally adequate. Krieger (1972) and Metzner (1985) proposed that the relative viscosity (μ_r) could be described by

$$\frac{\eta}{\eta_o} \equiv \mu_r = \left(1 - \frac{\varphi}{A}\right)^{-2} \tag{1}$$

here η_o is the (Newtonian) solvent viscosity, η is the suspension viscosity, φ is the volume fraction of the solid

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phase and A is a constant (maximum value of allowable volume fraction). For smooth spheres A~0.6, and for rough spheres and non-spherical particles A can be adjusted to lower values (Tanner, 2000). While more complex forms than (1) are available, for example Frankel and Acrivos (1967) and Zarraga *et al.* (2000; 2001), they present few advantages; and so we shall use Eq. (1) here.

Some non-Newtonian effects have been seen, even in suspensions with Newtonian solvents. Zarraga *et al.* (2000; 2001) suggested a mild power-law behaviour of the shear stress (exponent $n \approx 0.8-1.0$) showing shear-thinning, even with a Newtonian solvent, while various writers have seen not only shear-thinning but also shear-thickening at high shear rates (for example, Nam *et al.*, 2004). We shall mainly be concerned with shear-thinning here. Zarraga *et al.* (2000; 2001) reported normal stresses measurements (N_1 and N_2) in Newtonian and viscoelastic matrix suspensions. Mall-Gleissle *et al.* (2002) also made such normal stress measurements. The Mall-Gleissle *et al.* (2002) paper covers a range of volume fraction $\varphi = 0-0.25$, while Zarraga *et al.* (1999; 2001) used $\varphi = 0.3-0.55$. For a Newtonian solvent Zarraga *et al.* (2000) found

$$\frac{N_1}{\alpha\tau} = -0.15 \pm 0.05 \tag{2}$$

$$\frac{N_2}{\alpha\tau} = -0.54 \pm 0.03 \tag{3}$$

where τ is the shear stress, N_1 and N_2 are the first and second normal stress differences respectively. Also they found experimentally that the parameter α is given by, over the narrow range $\varphi = 0.3-0.53$,

$$\alpha = 2.17\phi^3 e^{2.34\phi} \tag{4}$$

Thus the relative magnitude of (N_1/N_2) is completely different from the usual case for polymeric fluids, where $N_1 >> |N_2|$, and N_1 is positive. For a second paper Zarraga *et al.* (2001) used a Boger-type (PAA + corn syrup) fluid as matrix, so that the matrix viscosity was nearly constant and N_2 (for the matrix) was very small. The range of φ used was 0.3 – 0.53. Not much shear-thinning was observed, but N_1 was positive, and they wrote

$$\frac{N_1}{\tau} \sim \frac{\psi_1 \dot{\gamma}^2}{\tau} + \beta \tag{5}$$

where ψ_1 and β are constants, and

$$\frac{N_2}{\tau} \sim \frac{\psi_2 \dot{\gamma}^2}{\tau} + \delta \tag{6}$$

where δ has the same value as in the Newtonian case, approximately $-1.17\varphi^3 e^{2.34\varphi}$. In (5) and (6) $\psi_1\dot{\gamma}^2$ and $\psi_2\dot{\gamma}^2$ are the values of N_1 and N_2 for the matrix material alone.

Mall-Gleissle *et al.* (2002) used silicone oils as matrices and investigated the range $\varphi = 0.05-0.25$. Their results show the usual viscosity increase with φ and also a positive N_1 and a negative N_2. They found that N_1 and N_2 correlated with a power of the shear stress, with exponents in the range of around 1.63 – 1.66 (±0.66) for N_1 and 1.7 ± 0.08 for N_2, so that one may take a power of about 1.7 for both functions. There is, however, a considerable variation in the relative levels of N_1 and N_2; the investigators found that $|N_2|/N_1$, was only a function of φ.

In summary, Mall-Gleissle *et al.* (2002), following Zarraga *et al.* (2001) suggested that, to a reasonable approximation

$$N_1 \equiv N_{1,m} + N_{1,s} \tag{7}$$

$$N_2 \equiv N_{2,m} + N_{2,s} \tag{8}$$

where the suffixes m, s refer to the matrix and suspension respectively, and N_1 and N_2 are multiples of $\tau^{1.7}$; $N_{1,s}$ and $N_{2,s}$ resemble Equation (2) and (3).

3. A simple phenomenological model

Many suspension models, of various degrees of complexity, have been proposed, but many of them do not accurately reflect the behaviour discussed above. Here, we will simply take the suggestion of Zarraga *et al.* (2001) and Mall-Gleissle *et al.* (2002) and propose the simple addition of two kinds of stress term:

$$\tau = \tau_N + \tau_v \tag{9}$$

where the suffix N denotes "Newtonian" behaviour, and v denotes 'viscoelastic' behaviour; the τ is the extra-stress term; the complete stress $\sigma \equiv -p\boldsymbol{I} + \tau$. For τ_N we assume a Reiner-Rivlin (Tanner, 2000) model:

$$\tau_N = 2\eta_N \boldsymbol{d} - 4B\boldsymbol{d}^2 \tag{10}$$

where $\boldsymbol{d}$ is the rate of deformation tensor and the viscosity η_N is given by

$$\eta_N = \eta_0(\mu_r(\varphi) - 1)f(|\dot{\gamma}|) \tag{11}$$

where μ_r is given by Eq. (1) above, and $f(|\dot{\gamma}|)$ is a suitable (shear-thinning) function of $\dot{\gamma}$, where the shear rate is now defined as

$$\dot{\gamma} = \sqrt{2d_{ij}d_{ij}} \tag{12}$$

Although the Reiner-Rivlin model does not describe polymer rheology properly, it appears to be useful for the suspensions studied here. In the case when τ_v is negligible, τ_N should fall back to something like equations (2) to (3) in simple shearing. In this case it is known that this model shows, for the shear stress τ,

$$\tau = \eta_N \dot{\gamma}$$

and it gives the results

$$N_1 = 0, \quad N_2 = -B\dot{\gamma}^2$$

If we choose B appropriately, we can find

$$\tau_N = 2\eta_0(\mu_r - 1)\left(\boldsymbol{d} - \frac{\alpha^* \boldsymbol{d}^2}{\dot{\gamma}}\right) \tag{13}$$

The response of this model gives the correct τ and N_2, and also $N_1 = 0$, which does not agree with Eq. (2). The factor $\dot{\gamma}$ in the denominator leaves an approximately linear law for stresses as a function of $\dot{\gamma}$, as one would expect with a Newtonian matrix. If we now add on a viscoelastic component τ_v we can introduce a positive N_1 while still retaining a large negative N_2. A wide set of choices is possible for τ_v; we will use a single-mode PTT model of the form (the exact form is not very important)

$$\lambda \frac{\Delta \tau_v}{\Delta t} + F(tr\,\tau_v)\,\tau_v = 2\eta_p \boldsymbol{d} \tag{14}$$

where λ is a time constant for a fixed value of φ, $\Delta/\Delta t$ is the upper convected time derivative (Tanner, 2000), η_p is a (possibly variable) viscosity, and F is a function of $tr\,\tau_v$. For simple shearing, (14) gives the result (Tanner, 2000)

$$N_{2v} = 0$$

$$N_{1v} = 2\eta_p \lambda \dot{\gamma}^2 F^{-2} \tag{15}$$

$$\tau_v = \eta_p \dot{\gamma} F^{-1} \tag{16}$$

where τ_v is the shear stress in the matrix. From (15) and (16) we find

$$N_{1v} = \frac{2\lambda}{\eta_p}\tau_v^2 \tag{17}$$

Hence the complete response, following (9), is

$$\tau = \tau_N + \tau_v = \eta_N \dot{\gamma} + \eta_p \dot{\gamma} F^{-1}$$

$$N_1 = \frac{2\lambda}{\eta_p}\tau_v^2; \quad N_2 = -0.54\,\alpha\tau.$$

4. Computed results

The model discussed above has been used to fit the rheological properties of some viscoelastic matrix suspensions. Here we use this model to compute the viscosity and stresses (N_1, N_2 and τ) for viscoelastic suspensions of non-colloidal spherical particles in shear and elongational flow. These computed results are shown against experimental measurements given by Mall-Gleissle *et al.* (2002) for shear flow at lower volume fraction ($\varphi = 0 - 0.25$), Zarraga *et al.* (2001) for shear flow at higher volume fraction ($\varphi = 0.3 - 0.55$) and Le Meins *et al.* (2003) for elongational flow. The matrix properties adopted in the calculations are taken to be the same as those used in the corresponding experiment.

4.1. Relative viscosity

Relative viscosities have been calculated for the suspensions used in experiments by Zarraga *et al.* (2001) in shear flow and Le Meins *et al.* (2003) in elongational flow. The relative viscosity of a suspension in shear flow is defined as

$$\eta_r = \frac{\tau}{\dot{\gamma}\eta_0} \tag{18}$$

where τ is the shear stress, $\dot{\gamma}$ is the shear rate and η_0 is the viscosity of the matrix (at zero shear rate); and the relative viscosity in elongational flow is defined as

$$\eta_r = \frac{\sigma_{11} - \sigma_{22}}{3\dot{\varepsilon}\eta_s} \tag{19}$$

in which σ_{11} and σ_{22} are the normal stresses, $\dot{\varepsilon}$ is the strain rate and η_s is the viscosity of the solvent. It is clear, for viscoelastic suspensions, that the relaxation time λ should no longer be a constant, but a function of the volume fraction φ same as the coefficient α^* (Eq. 13). It is a constant only when the volume fraction is a constant. For the suspension used by Zarraga *et al.* (2001), we found suitable values of λ and α^* have the form in terms of the volume fraction φ as

$$\lambda = 0.086 + 140\varphi^5(\varphi - 0.35)^2$$

$$\alpha^* = 0.19 + 200\varphi^3(\varphi - 0.3)^2. \tag{20}$$

In shear flow, the calculated relative viscosity dependence on the volume fraction φ is shown in Fig. 1 and compared to the experimental measurements given by Zarraga *et al.* (2001). Clearly, this fit is adequate. In the calculations for this set of data, the constant in Eq. (1) $A = 0.58$ and the parameter in Eq. (14) $F = 1$ (PTT model parameter $\varepsilon = 0$), are adopted.

For suspensions used in elongational flow by Le Meins *et al.* (2003), in which the diameter of the particle $D =$

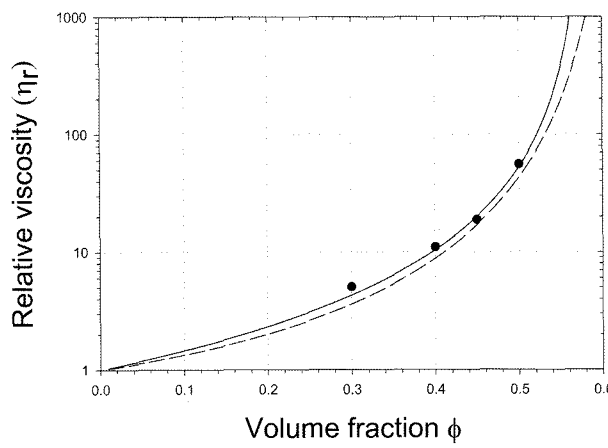

Fig. 1. Relative viscosities dependence on volume fraction at $\dot{\gamma} = 10(s^{-1})$. — calculated results; • and - - - Zarraga's data.

1.4 μm, we found that a step function could be used for the coefficient α^*. The relaxation time λ and α^* for these suspensions are given by

$$\lambda = 7.9 + 44800\varphi^2(\varphi - 0.05)^4$$

$$\alpha^* = \begin{cases} 0.25 & (\varphi \le 0.13) \\ 0.29 & (\varphi > 0.13) \end{cases} \tag{21}$$

In these calculations, $A = 0.65$ (constant) and PTT model parameter $\varepsilon = 1.0$ were used, and F is therefore given by

$$F = e^{\frac{\lambda(\sigma_{11}+\sigma_{22}+\sigma_{33})}{\eta_0}}. \tag{22}$$

Relative elongational viscosities as a function of volume fraction are plotted in Fig. 2 at various strain rates. Close agreement with the experimental measurements can be observed at a strain rate $\dot{\varepsilon} = 0.025$. These results show that the relative elongational viscosity not only increases as the volume fraction increases, but increases as the strain rate increases as well. Fig. 3 shows that relative elongational viscosities versus Weissenberg number at different volume fractions.

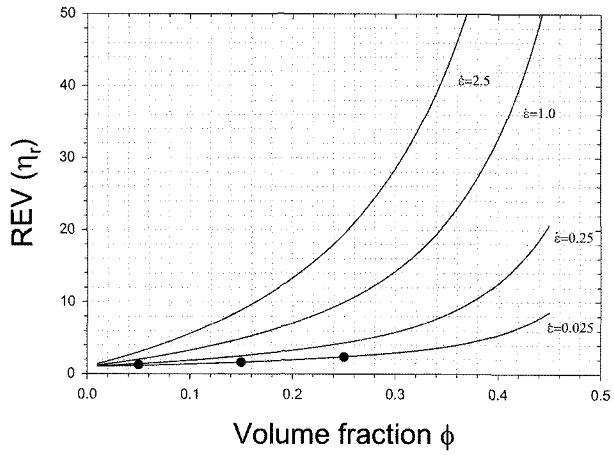

Fig. 2. Relative elongational viscosities (REV) as a function of the volume fraction φ. • experimental measurements ($\dot{\varepsilon} = 0.025$ $D = 1.4$ μm); curves represent the computed results.

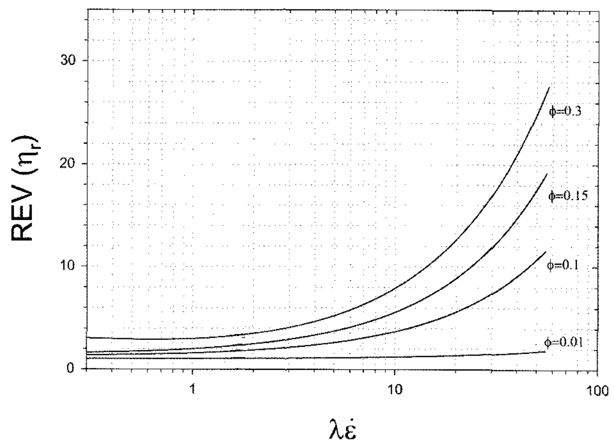

Fig. 3. Relative elongational viscosities (REV) as a function of Weissenberg number.

4.2. First normal stress difference, N_1

As we discussed in the previous section, this model always gives a positive first normal stress difference. At higher volume fractions of the solid phase ($\varphi = 0.3 - 0.5$), the dimensionless first normal stress differences (made dimensionless by dividing the shear stress τ) have been calculated for the suspensions used in the experiment by Zarraga *et al.* (2001). The relaxation time λ and the parameter α^* of these suspensions are given by Eq. (20). The calculated result is plotted as a function of shear rate and shown in Fig. 4. At lower volume fractions ($\varphi = 0 - 0.25$), the first normal stress differences have been calculated for suspensions used in the experiment by Mall-Gleissle *et al.* (2002). For these suspensions (Silicone-oil DOW310[5] used as the matrix), we found that the relaxation time λ and the parameter α^* as a function of the volume fraction have the form as

$$\lambda = 0.09 + 36\varphi(\varphi - 0.15)^2$$

$$\alpha^* = \begin{cases} 0.25 & (\varphi \le 0.13) \\ 0.29 & (\varphi > 0.13) \end{cases} \tag{23}$$

Calculated results of the first normal stress differences dependence on the shear stress are shown in Fig. 5, Fig. 6 shows the dimensionless first normal stress differences as a function of shear rate for these suspensions.

As shown in Fig. 4 and Fig. 5, calculated results of the first normal stress difference are a reasonably good fit to the experimental measurements given by Zarraga *et al.*

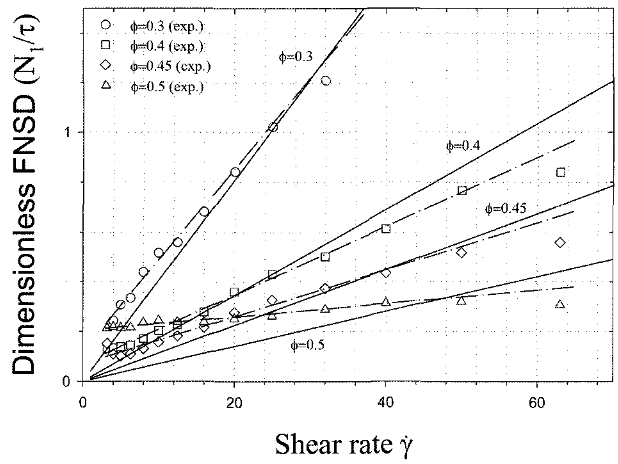

Fig. 4. Dimensionless first normal stress difference (FNSD) dependence on shear rate for suspensions at various volume fractions. Scatter plots and dash-dot lines are Zarraga's data; solid lines are calculated results.

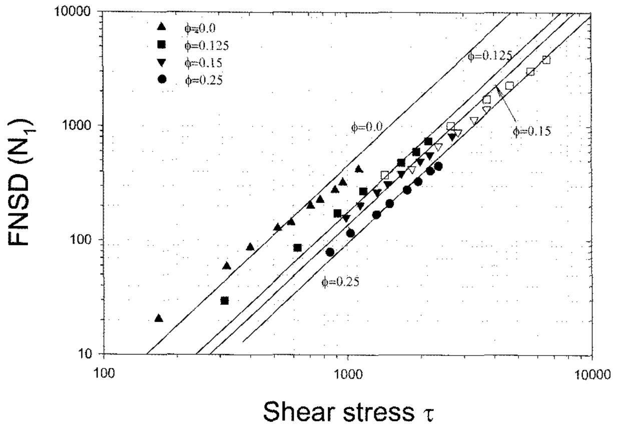

Fig. 5. The first normal stress difference (FNSD) as a function of shear stress for suspensions used in Mall-Gleissle's experiments at various volume fractions. Scatter plots are Mall-Gleissle's data; solid lines are calculated results.

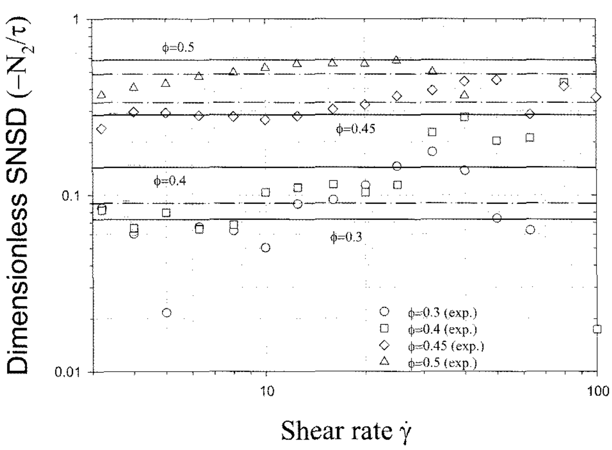

Fig. 7. Dimensionless second normal stress difference (SNSD) dependence on shear rate for suspensions at different volume fractions. Scatter plots and dash-dot lines are Zarraga's data; solid lines are calculated results.

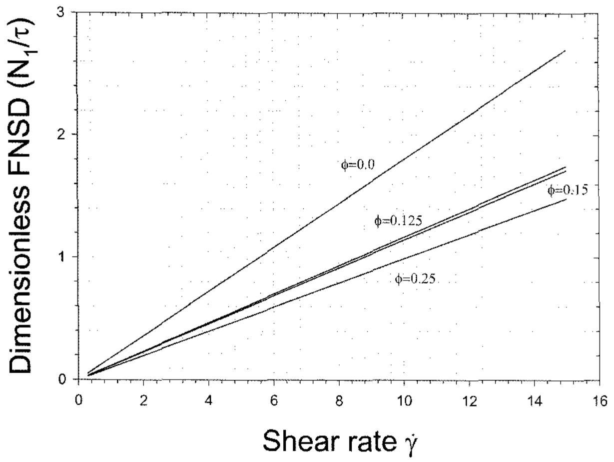

Fig. 6. Calculated results of dimensionless first normal stress difference (FNSD) as a function of shear rate for suspensions used in Mall-Gleissle's experiments at various volume fractions.

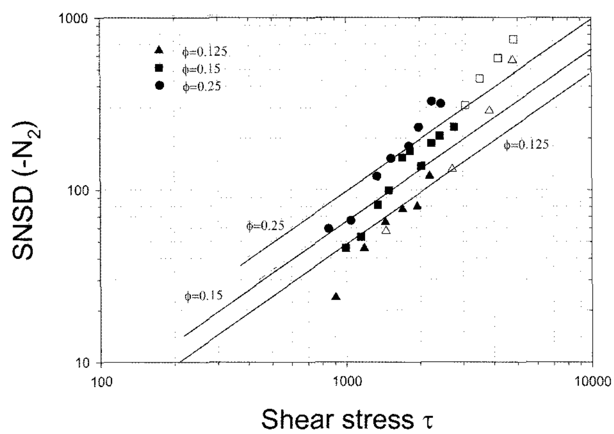

Fig. 8. Second normal stress difference (SNSD) dependence on the shear stress for suspensions at different volume fractions. Scatter plots are Mall-Gleissle's data and the solid lines are the calculated results.

(2001) and Mall-Gleissle *et al.* (2002). Clearly, the model shows that the dimensionless first normal stress difference decreases as the volume fraction of the solid phase increases. This behaviour can be seen in Fig. 4 and Fig. 6. This is consistent with experimental measurements (Zarraga *et al.*, 2001; Mall-Gleissle *et al.*, 2002), and also the observations of Ohl and Gleissle (1993) for suspensions of crushed limestone in a viscoelastic shear-thinning fluid.

4.3. Second normal stress difference, N_2

Similar to the result obtained in experiments, the model always gives a negative second normal stress difference. The calculated result for the dimensionless second normal stress difference at volume fraction ($\varphi = 0.3 - 0.55$) is plotted as a function of shear rate and shown in Fig. 7, and the second normal stress difference dependence on the shear stress at the volume fractions ($\varphi = 0 - 0.25$) is shown in Fig. 8. The calculated results are tested against experimental measurements given by Zarraga *et al.* (2001) and Mall-Gleissle *et al.* (2002) and show reasonably good fits. Fig. 9 shows that the dimensionless second normal stress difference versus the shear rate for suspensions used by Mall-Gleissle *et al.* (2002) and Fig. 10 shows the average of dimensionless second normal stress differences over the shear rate range in the calculations as a function of the volume fraction for the suspensions used by Zarraga *et al.* (2001).

As shown in Fig. 7 and Fig. 9, the behaviour of dimensionless second normal stress difference (in magnitude) is, unlike the dimensionless first normal stress difference, which is strongly dependent on the shear rate and decreases as the volume fraction increases, independent of

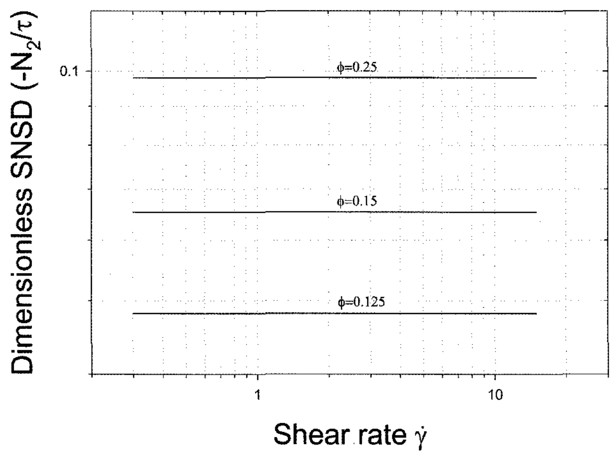

Fig. 9. Dimensionless second normal stress difference (SNSD) dependence on the shear rate for suspensions used in experiments by Mall-Gleissle *et al.* (2002) at different volume fractions.

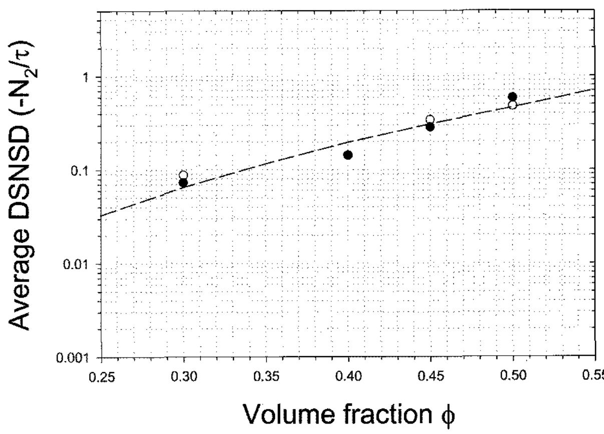

Fig. 10. The average of dimensionless second normal stress difference (DSNSD) over the shear rate range in calculations dependence on the volume fraction for suspensions used in experiments by Zarraga *et al.* (2001). ○ and - - - Zarraga's data; • calculated results.

the shear rate and increases as the volume fraction increases. This behaviour of viscoelatic suspensions has been observed in experimental measurements given by Zarraga *et al.* (2001).

For suspensions used in experiments by Zarraga *et al.* (2001), Mall-Gleissle *et al.* (2002) and Le Meins *et al.* (2003), the relaxation time λ as a function of the volume fraction φ is plotted in Fig. 11(a), and Fig. 11(b) shows the coefficient α^* dependence on the volume fraction for the suspension used by Zarraga *et al.* (2001).

4.4. Stopping and restarting shear flow

For Newtonian matrix suspensions, the behaviour of stopping and restarting shear flow has been discussed by Phan-Thien (1995) and in the experimental observations made by Gadala-Maria and Acrivos (1980). Our model can be used to describe this behaviour for viscoelastic suspensions. When the flow is stopped, the behaviour of the first normal stress difference and shear stress are, according to Phan-Thien (1995) and experimental measurements given by Gadala-Maria and Acrivos (1980), such that all the stresses or torques instantaneously decay to zero. In our model, there is a rapid, but not instantaneous, reduction to zero. A short relaxation period appears here. This obvious difference of the behaviour is due to the matrix being viscoelastic fluid (non-Newtonian) in the calculations, but it is a Newtonian fluid in the work of Phan-Thien (1995) and in the experiments of Gadala-Maria and Acrivos (1980). But the second normal stress difference shows no relaxation period here; and gives a behaviour which instantaneously goes down to zero when the shear rate vanishes, as given by Phan-Thien (1995). This is because the second normal

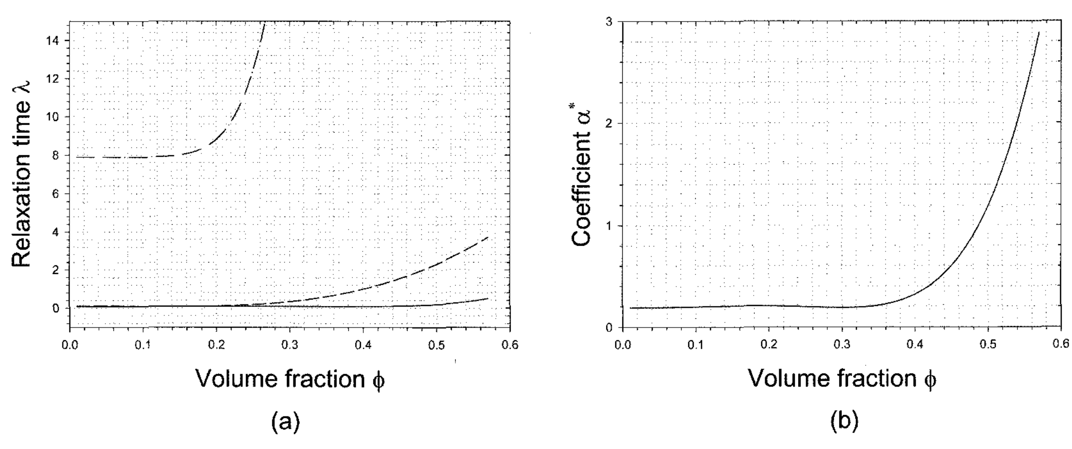

Fig. 11. (a), The relaxation time λ as a function of the volume fraction φ. Solid line is for the suspension used by Zarraga *et al.* (2001), dash line is for the suspension used by Mall-Gleissle *et al.* (2002) and dash-dot line is for the one used by Le Meins *et al.* (2003). (b), The coefficient α^* dependence on the volume fraction φ for the suspension used by Zarraga *et al.* (2001).

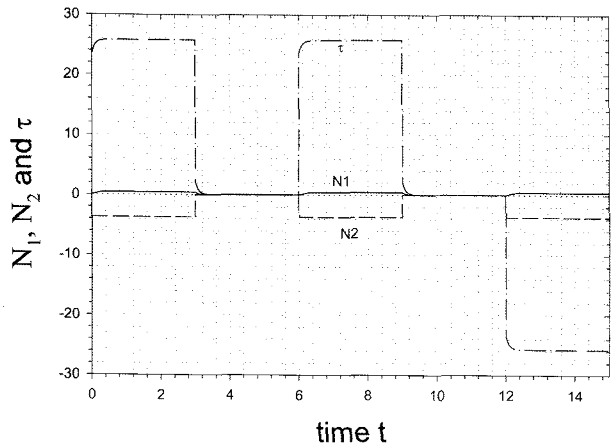

Fig. 12. The shear stress and normal stress differences as a function of time in stopping and restarting shear flow.

stress difference is given by the component of "Newtonian" behaviour in our model.

When the flow is restarted in the same direction, the first normal stress difference and the shear stress undergo a transient period similar to the starting flow and then go to the steady state. But the second normal stress difference will recover from where it was left. The corresponding steady state value of the shear stress will be almost instantaneously attained.

When the flow is restarted in the opposite direction, the model gives the same behaviour, but the shear stress is also in the opposite direction, for all the normal stresses are the same as these obtained when the flow is restarted in the same direction. A summary of the predictions is shown in Fig. 12. This calculated result is for the suspension used by Zarraga *et al.* (2001) at the volume fraction $\varphi = 0.4$ and shear rate $\dot{\gamma} = 1.0(-1.0)$.

5. Summary

The focus of this work is to discuss a new constitutive model for the description of viscoelastic suspensions. In summary, the constitutive model discussed above is quite simple and gives reasonably good predictions of the rheological properties for viscoelastic suspensions in shear and elongational flow. Features of predictions given by the model include:

- The positive first normal stress difference
- The negative second normal stress difference
- The dimensionless first normal stress difference strongly dependent on the shear rate and decreasing with volume fraction
- The dimensionless second normal stress difference is nearly independent of the shear rate and increases with the volume fraction
- Gives an approximate prediction for the viscoelastic suspension in stopping and restarting flow

For calculated results of relative viscosities and stresses, reasonably good agreements can be observed by comparing with the corresponding experimental measurements.

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