

Tensorial rheological model for concentrated non-colloidal suspensions: normal-stress differences

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Most existing rheological models for non-colloidal suspensions fail to simultaneously capture the two main non-Newtonian trends of these systems, namely finite normal stress differences and transient effects. We address this issue by extending a previously-proposed minimal model accounting for microstructure anisotropy through a conformation tensor, and which was shown to correctly predict transient effects (Ozenda *et al.* 2018). The new model is compared to a large experimental dataset involving varying volume fractions, from dilute to concentrated cases. Both apparent viscosity and normal stress differences in steady state, are quantitatively reproduced in the whole range of volume fraction, and qualitative comparisons for transient evolution of apparent viscosity during shear reversal are provided. Furthermore, the model is validated against particle pressure measurements that were not used for parameter identification. Even if the proposed constitutive equation for the Cauchy stress tensor is more difficult to interpret than in the minimal model, this study opens way for the use of conformation tensor rheological models in applications where the effect of the second normal stress difference is prominent, like elongational flows or migration phenomenon.

1. Introduction

Concentrated suspensions of non-colloidal rigid particles present two main non-Newtonian rheological trends, namely finite *normal stress differences* under shear flow and *transient effects* (see, e.g., the recent review by Guazzelli & Pouliquen 2018). (i) Since the early work of Gadala-Maria (1979), *normal stress differences* have been investigated in different flow geometries (Zarraga *et al.* 2000; Singh & Nott 2003; Couturier *et al.* 2011; Boyer *et al.* 2011; Dai *et al.* 2013; Dbouk *et al.* 2013). These measurements consistently show that the second normal stress difference N_2 is negative, with an absolute value that grows with volume fraction ϕ . The first normal stress difference N_1 is generally found to be much smaller, in absolute value, than $|N_2|$. While most studies reported negative N_1 values, few also reported positive values especially at high volume fractions. (ii) Using shear reversal experiments, Gadala-Maria & Acrivos (1980) showed that non-colloidal suspensions present *transient rheological responses*, characterized by a *rate-independent*

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evolution of the shear viscosity over typical strains of a few units, before this quantity eventually reaches its steady-state value. These shear-reversal experiments were later revisited by [Kolli *et al.* \(2002\)](#) and [Blanc *et al.* \(2011\)](#). All these authors related the transient effects to the development of microstructure anisotropy under shear flow, and attempted to quantify this anisotropy using the pair distribution function.

Most non-Newtonian trends of concentrated suspensions can be captured by *particle-based numerical simulations*, which offer additional insights into the physical processes at play. Earliest approaches were based on Stokesian dynamics, in which hydrodynamic interactions are split into far-field and near-field (lubrication) contributions ([Brady & Bossis 1985](#)). Numerous studies showed that an essential ingredient to obtain realistic rheological behaviour in these simulations and, in particular, the existence of finite normal-stress differences, is to account for particle contacts through short-range repulsive forces ([Nott & Brady 1994](#); [Sierou & Brady 2002](#)). Numerical results also demonstrated that frictional interactions should be taken into account to recover correct orders of magnitude for these normal-stress differences, i.e. $|N_1| < |N_2|$ ([Sierou & Brady 2002](#); [Mari *et al.* 2014](#)). Similar conclusions concerning the critical role of particle friction were reached by [Gallier *et al.* \(2014\)](#), using a more accurate DNS method to model hydrodynamic interactions. More recently [Peters *et al.* \(2016\)](#) and [Chacko *et al.* \(2018\)](#) were also able to successfully simulate transient effects during shear reversal using DNS and simplified Stokesian dynamics approaches, respectively. These particle-based simulations make it possible to continuously vary physical parameters and perform numerical experiments, thereby providing datasets that are well complementary to “real” experiments. However, due to the high demand in computing power, they are still limited to small systems with a relatively low number of particles (a few thousands at most).

Unlike discrete particle-based models, *continuous rheological models* are suited to the simulation of large systems. However, the elaboration of constitutive laws able to reproduce both normal-stress differences and transient features has been proved a challenging task (e.g., [Denn & Morris 2014](#)). General expressions for the bulk Cauchy stress tensor of suspensions can be obtained in the frame of two-phase mixture models ([Jackson 2000](#); [Nott *et al.* 2011](#); [Baumgarten & Kamrin 2019](#)), and involve contributions from both the interstitial fluid and the particles. For practical applications, physical closures need to be postulated for these different contributions. [Nott & Brady \(1994\)](#) introduced the *suspension balance model* (later extended by [Morris & Boulay 1999](#); [Miller & Morris 2006](#)), which involves explicit empirical expressions for the particle-induced normal stress components. This model provides relatively accurate predictions for steady-state flows and, when coupled to an evolution equation for the volume fraction ϕ (that arises from mass and momentum conservation of the particle phase), can also account for particle migration processes. A similar type of approach based on explicit stress closures was recently followed by [Singh *et al.* \(2018\)](#), who proposed a model describing the transition to shear-thickening in highly-concentrated suspensions. By construction, however, these models are unable to capture transient rheological effects.

Transient effects can be described by introducing a *conformation tensor*, denoted \mathbf{b}_e in this paper, which represents microstructure anisotropy. Note that conformation tensors are also sometimes referred to as *texture tensors* (see e.g. [Lehoucq *et al.* 2015](#)). For non-colloidal suspensions, the conformation tensor \mathbf{b}_e needs to obey an evolution equation that ensures rate-independence, i.e. this equation should have no characteristic time (e.g., [Goddard 1982](#)). [Hand \(1962\)](#) formulated general properties for anisotropic fluids, and expressed the Cauchy stress tensor $\boldsymbol{\sigma}$ as a function of both \mathbf{b}_e and the strain

rate tensor $\dot{\gamma}$. Phan-Thien (1995) proposed a differential constitutive equation for the conformation tensor that led, for the first time, to predictions qualitatively in agreement with experimental observations during shear reversal. Goddard (2006) revisited this approach, and proposed a model involving twelve material parameters and two tensors for describing the anisotropy. By a systematic fitting procedure on a limited dataset, he obtained numerical results in quantitative agreement with experiments for transient effects and normal stress differences. Stickel *et al.* (2006) proposed a simplified expression for the Cauchy stress tensor, which is linear in terms of both the strain rate and the conformation tensors (see also Stickel *et al.* 2007; Yapici *et al.* 2009). While this latter model involves thirteen free parameters, it failed to provide quantitative comparisons with shear reversal experiments: an unexpected sharp spike in both apparent viscosity and normal stress differences was obtained at the time of the reversal. Using a much simpler model, involving only four free parameters and a linear evolution evolution for the conformation tensor, Ozenda *et al.* (2018) recently obtained a good quantitative agreement with the shear reversal measurements of Blanc *et al.* (2011) for a wide range of volume fraction ϕ . Furthermore, at the micro-structural scale, the model successfully reproduced both the pair distribution function and the depletion angles measured by Blanc *et al.* (2013). However, this simple model predicts unphysical normal stress differences.

The objective of this paper is to propose a continuous rheological model that provides quantitative predictions for both normal stress differences and transient effects, for dilute to concentrated suspensions. For that purpose, we revisit and extend the model proposed by Ozenda *et al.* (2018) based on the general expansion of Hand (1962). We limit consideration to *rate-independent* constitutive responses, i.e. to suspensions dominated by hydrodynamic (and possibly frictional) interactions. We also restrict attention to concentration ranges sufficiently below jamming, such that granular plasticity effects arising from sustained frictional contacts between particles (e.g. granular dilatancy, Pailha & Pouliquen 2009; Baumgarten & Kamrin 2019) do not come into play. In particular, our model does not include the shear-thinning and shear-thickening effects observed in more complex systems and/or close to jamming transition (see, e.g., Royer *et al.* 2016; Singh *et al.* 2018). When compared with previous tensorial approaches (Phan-Thien 1995; Goddard 2006; Stickel *et al.* 2006), the present model involves only seven adjustable parameters, and compares well with experiments. Section 2 presents the rheological model. Section 3 is devoted to simple shear flows. The system of time-dependent equations is expanded for this case, and the stationary solution is explicitly exhibited. The dependence of the material parameters upon volume fraction ϕ is investigated, based on asymptotic analyses in the dilute and highly concentrated limits. In section 4, model predictions are compared with a large number of experimental results regarding both transient effects and steady-state normal stresses. Finally, results are summarised and discussed in section 5.

2. Mathematical model

2.1. Problem statement

Following Ozenda *et al.* (2018), let us introduce the *conformation tensor* $\mathbf{b}_e = d_0^{-2} \langle \boldsymbol{\ell} \otimes \boldsymbol{\ell} \rangle$, where $\boldsymbol{\ell}$ is the branch vector joining the centres of two neighbouring particles and d_0 is the average distance between neighbouring particle centers in an isotropic configuration at rest. In the isotropic configuration at rest, we have $\mathbf{b}_e = \mathbf{I}$, where \mathbf{I} is the identity matrix.

For convenience, we introduce the tensor $\gamma_e = c_0(\mathbf{b}_e - \mathbf{I})$, where c_0 is a dimensionless constant that will be chosen later. The tensor γ_e interprets as the deformation of the micro-structure with respect to rest configuration. As in [Ozenda et al. \(2018\)](#), we assume a linear evolution for γ_e :

$$\frac{\mathcal{D}_a \gamma_e}{\mathcal{D}t} + \delta_1 |\dot{\gamma}| \gamma_e - \dot{\gamma} = 0, \quad (2.1a)$$

where $\dot{\gamma} = \nabla \mathbf{u} + \nabla \mathbf{u}^T$ is the strain rate tensor, \mathbf{u} is the velocity field of the suspension, and δ_1 is a positive dimensionless material parameter. The matrix norm $|\xi|$ is defined as $|\xi|^2 = (\xi : \xi)/2$ for any matrix ξ , with $(\cdot : \cdot)$ denoting the double contracted matrix product. We use in (2.1a) a general Gordon-Schowalter tensor derivative ([Gordon & Schowalter 1972](#); [Saramito 2016](#)):

$$\frac{\mathcal{D}_a \gamma_e}{\mathcal{D}t} = \frac{\partial \gamma_e}{\partial t} + (\mathbf{u} \cdot \nabla) \gamma_e - W(\mathbf{u}) \gamma_e + \gamma_e W(\mathbf{u}) - a(D(\mathbf{u}) \gamma_e + \gamma_e D(\mathbf{u}))$$

where $D(\mathbf{u}) = (\nabla \mathbf{u} + \nabla \mathbf{u}^T)/2 = \dot{\gamma}/2$, $W(\mathbf{u}) = (\nabla \mathbf{u} - \nabla \mathbf{u}^T)/2$, and $a \in [-1, 1]$. Note that [Ozenda et al. \(2018\)](#) used an upper-convected derivative, corresponding to $a = 1$. Equation (2.1a) has no characteristic time, such that its solutions are *rate-independent*: In simple shear, changing the amplitude of the strain rate $\dot{\gamma}$ leaves the evolution of $\dot{\gamma}_e$ unchanged when expressed in terms of the dimensionless time $|\dot{\gamma}|t$ (that interprets as a strain).

The Cauchy stress tensor $\boldsymbol{\sigma}$ of the suspension is assumed to express as an analytical function of the two tensorial variables $\dot{\gamma}$ and γ_e . Following [Hand \(1962\)](#), and by virtue of Cayley-Hamilton theorem, only the zeroth, first and second powers of $\dot{\gamma}$ and γ_e can contribute to the expression. Furthermore, to preserve rate-independence, terms in $\dot{\gamma}^2$ should also be discarded. After extensive tests to assess the effects of the different possible terms, we chose to consider the following expression for the Cauchy stress tensor:

$$\boldsymbol{\sigma} = -p_b \mathbf{I} + \eta \dot{\gamma} + \eta_e \left\{ \delta_1 |\dot{\gamma}| \gamma_e + \beta \left(\frac{\gamma_e^2 \dot{\gamma} + \dot{\gamma} \gamma_e^2}{2} - |\dot{\gamma}| \gamma_e^2 \right) + \delta_2 \left(\frac{\gamma_e \dot{\gamma} + \dot{\gamma} \gamma_e}{2} \right) \right\} \quad (2.1b)$$

where p_b is the bulk pressure of the suspension, η and η_e are characteristic viscosities, and β and δ_2 are additional dimensionless material parameters. Note that the terms in factor of δ_1 and δ_2 are linear in γ_e , while the term in factor of β is quadratic in γ_e .

The present model represents an extension of that presented by [Ozenda et al. \(2018\)](#), which involved only one linear and one non-linear terms. For the sake of simplicity and of formulating a minimal model, the scalar coefficients η , $\eta_e \beta$, $\eta_e \delta_1$ and $\eta_e \delta_2$ are assumed to be independent of the invariants of γ_e , which is a restriction compared to the general theory of [Hand \(1962\)](#).

Constitutive equations (2.1) are coupled with mass and momentum conservations to obtain a closed problem for three unknowns, namely the suspension pressure p_b , the suspension velocity \mathbf{u} , and the tensor describing the anisotropy of the micro-structure γ_e :

$$\frac{\mathcal{D}_a \gamma_e}{\mathcal{D}t} + \delta_1 |2D(\mathbf{u})| \gamma_e - 2D(\mathbf{u}) = 0 \quad (2.2a)$$

$$\rho \left(\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla) \mathbf{u} \right) - \operatorname{div} \boldsymbol{\sigma} = 0 \quad (2.2b)$$

$$\operatorname{div} \mathbf{u} = 0 \quad (2.2c)$$

where the Cauchy stress tensor $\boldsymbol{\sigma}$ is expressed by (2.1b). Note that p_b in (2.1b) should be

regarded as the Lagrange multiplier associated to the incompressibility constraint (2.2c). This set of equations is closed by appropriate boundary and initial conditions for \mathbf{u} and γ_e .

From (2.2a), remark that the conformation tensor $\mathbf{b}_e = c_0^{-1}(c_0\mathbf{I} + \gamma_e)$ satisfies:

$$\frac{\mathcal{D}_a \mathbf{b}_e}{\mathcal{D}t} - c_0 \delta_1 |2D(\mathbf{u})| \mathbf{I} + c_0 \delta_1 |2D(\mathbf{u})| \mathbf{b}_e = 2(1 - ac_0)D(\mathbf{u})$$

Hence, by choosing the dimensionless constant $c_0 = 1/a$, the right-hand-side in the previous relation is zero. In the following, we also assume $a \in]0, 1]$. According to Hulsen (1990), and since $\delta_1 > 0$, these choices guarantee that the conformation tensor \mathbf{b}_e is positive definite at any time, if this property is satisfied at initial time.

3. Simple shear and shear reversal

3.1. The reduced problem

Let us consider a simple shear flow with uniform shear rate. The x axis is in the flow direction and the y axis is in the direction of velocity gradient, such that $\mathbf{u}(t, x, y, z) = (u_x(t, y), 0, 0)$. Let us denote $\dot{\gamma} = \partial_y u_x$ the uniform scalar shear rate, such that $|\dot{\gamma}| = |\dot{\gamma}|$. Evolution equation (2.2a) for $\gamma_e(t)$ then reduces to the following system of ordinary differential equations:

$$\partial_t \gamma_{e,xx} - (1+a)\dot{\gamma} \gamma_{e,xy} + \delta_1 |\dot{\gamma}| \gamma_{e,xx} = 0 \quad (3.1a)$$

$$\partial_t \gamma_{e,yy} + (1-a)\dot{\gamma} \gamma_{e,xy} + \delta_1 |\dot{\gamma}| \gamma_{e,yy} = 0 \quad (3.1b)$$

$$\partial_t \gamma_{e,xy} - \frac{1+a}{2} \dot{\gamma} \gamma_{e,yy} - \frac{1-a}{2} \dot{\gamma} \gamma_{e,xx} + \delta_1 |\dot{\gamma}| \gamma_{e,xy} = \dot{\gamma} \quad (3.1c)$$

$$\partial_t \gamma_{e,xz} - (1+a)\dot{\gamma} \gamma_{e,yz} + \delta_1 |\dot{\gamma}| \gamma_{e,xz} = 0 \quad (3.1d)$$

$$\partial_t \gamma_{e,yz} + (1-a)\dot{\gamma} \gamma_{e,xz} + \delta_1 |\dot{\gamma}| \gamma_{e,yz} = 0 \quad (3.1e)$$

$$\partial_t \gamma_{e,zz} + \delta_1 |\dot{\gamma}| \gamma_{e,zz} = 0 \quad (3.1f)$$

We assume $\gamma_{e,xz} = \gamma_{e,yz} = \gamma_{e,zz} = 0$ at $t = 0$, such that these components remain zero at any time. The constitutive equation (2.1b) for $\boldsymbol{\sigma}$ becomes:

$$\sigma_{xx} = -pb + \eta_e \left\{ \delta_1 |\dot{\gamma}| \gamma_{e,xx} + \beta \left(\dot{\gamma} (\gamma_{e,xx} + \gamma_{e,yy}) \gamma_{e,xy} - |\dot{\gamma}| (\gamma_{e,xx}^2 + \gamma_{e,xy}^2) \right) + \delta_2 \dot{\gamma} \gamma_{e,xy} \right\} \quad (3.2a)$$

$$\sigma_{yy} = -pb + \eta_e \left\{ \delta_1 |\dot{\gamma}| \gamma_{e,yy} + \beta \left(\dot{\gamma} (\gamma_{e,xx} + \gamma_{e,yy}) \gamma_{e,xy} - |\dot{\gamma}| (\gamma_{e,xy}^2 + \gamma_{e,yy}^2) \right) + \delta_2 \dot{\gamma} \gamma_{e,xy} \right\} \quad (3.2b)$$

$$\sigma_{zz} = -pb \quad (3.2c)$$

$$\begin{aligned} \sigma_{xy} = \eta \dot{\gamma} + \eta_e \left\{ \delta_1 |\dot{\gamma}| \gamma_{e,xy} + \beta \left(\frac{\dot{\gamma}}{2} (\gamma_{e,xx}^2 + \gamma_{e,yy}^2 + 2\gamma_{e,xy}^2) - |\dot{\gamma}| (\gamma_{e,xx} + \gamma_{e,yy}) \gamma_{e,xy} \right) \right. \\ \left. + \frac{\delta_2 \dot{\gamma}}{2} (\gamma_{e,xx} + \gamma_{e,yy}) \right\} \end{aligned} \quad (3.2d)$$

$$\sigma_{xz} = \sigma_{yz} = 0 \quad (3.2e)$$

The system (3.1) is linear and admits explicit solutions when the shear rate $\dot{\gamma}$ is given. Explicit expressions of the stress components are then obtained from (3.2). In the following, we will also consider situations in which the shear stress σ_{xy} is imposed. In this case, numerical solutions of the coupled equations (3.1)-(3.2) are computed using `lsode`

ordinary differential equation solver (Radhakrishnan & Hindmarsh 1993) implemented within the `numpy-scipy` environment (Jones *et al.* 2001–).

3.2. Explicit solution in steady state

The steady state solution of (3.1a)-(3.1c) writes:

$$\gamma_{e,xx} = \frac{1+a}{1-a^2+\delta_1^2} \quad (3.3a)$$

$$\gamma_{e,yy} = \frac{-(1-a)}{1-a^2+\delta_1^2} \quad (3.3b)$$

$$\gamma_{e,xy} = \frac{\delta_1 \operatorname{sgn}(\dot{\gamma})}{1-a^2+\delta_1^2} \quad (3.3c)$$

The steady-state apparent viscosity $\eta_{app} = \sigma_{xy}/\dot{\gamma}$ and normal stress differences $N_1 = \sigma_{xx} - \sigma_{yy}$ and $N_2 = \sigma_{yy} - \sigma_{zz}$ are then easily deduced from (3.2a)-(3.2d):

$$\eta_{app} = \eta + \frac{\eta_e}{1-a^2+\delta_1^2} \left(\delta_1^2 + a\delta_2 + \frac{((1+a^2+\delta_1^2) - 2a\delta_1)\beta}{1-a^2+\delta_1^2} \right) \quad (3.4a)$$

$$N_1 = \frac{2\eta_e|\dot{\gamma}|}{1-a^2+\delta_1^2} \left(\delta_1 - \frac{2a\beta}{1-a^2+\delta_1^2} \right) \quad (3.4b)$$

$$N_2 = \frac{\eta_e|\dot{\gamma}|}{1-a^2+\delta_1^2} \left(\delta_1(-1+a+\delta_2) + \frac{(2a\delta_1 - ((1-a)^2 + \delta_1^2))\beta}{1-a^2+\delta_1^2} \right) \quad (3.4c)$$

We also define the particle pressure $p_p = p - p_b$, where $p = -\operatorname{tr}(\boldsymbol{\sigma})/3$ is the total pressure of the suspension. In steady state:

$$p_p = -\frac{2\eta_e|\dot{\gamma}|}{3(1-a^2+\delta_1^2)} \left(\delta_1(a+\delta_2) + \frac{(2a\delta_1 - (1+a^2+\delta_1^2))\beta}{1-a^2+\delta_1^2} \right) \quad (3.4d)$$

3.3. Dependence of material parameters upon volume fraction

The rheological model presented in section 2 involves six material parameters: two viscosities η and η_e , and four dimensionless parameters a , δ_1 , δ_2 and β . Preliminary tests to fit experimental data for different volume fractions ϕ (see § 4), showed that all these parameters, except a , have to vary with ϕ . Following these preliminary investigations, we assumed a to be independent of ϕ , and the five remaining parameters to depend only upon the reduced volume fraction $\psi = \phi/\phi_m$, with ϕ_m the maximal volume fraction of the suspension. This latter assumption is required to obtain a model that can be applied to different experimental datasets characterised by different values of ϕ_m . In practice, ϕ_m can vary between 0.53 and 0.64, typically, depending on particle shape, particle roughness, etc (e.g., Guazzelli & Pouliquen 2018).

As shown in appendix A, the parameter δ_1 is directly related to the *depletion angle* θ_e , i.e. the angle between the x axis and the direction of the eigenvector associated to the largest eigenvalue of $\boldsymbol{\gamma}_e$: $\delta_1 = \tan(2\theta_e)$. Based on the experimental measurements of θ_e provided by Blanc *et al.* (2013), the following dependence law is proposed for δ_1 (see figure 1):

$$\delta_1(\psi) = \bar{\delta}_1 \left((1-\psi)^{-1} - (1-b\psi) \right), \quad (3.5a)$$

where $\bar{\delta}_1$ and b are positive constants independent of ψ . Observe that $\delta_1(0) = 0$ and $\delta_1(1) = \infty$, such that the model predicts $\theta_e(0) = 0$ and $\theta_e(1) = \pi/4$.

To be consistent with numerous existing results (Maron & Pierce 1956; Morris & Boulay 1999; Guazzelli & Pouliquen 2018), the apparent viscosity η_{app} and the second normal stress difference N_2 should behave as $(1 - \psi)^{-2}$ in the $\psi \rightarrow 1$ limit. In addition, we expect $\eta_e = 0$ in the Newtonian limit $\psi = 0$. Accordingly, the following dependence laws are postulated for the viscosities η and η_e :

$$\eta(\psi) = \eta_0 \left(1 - \omega + \psi \left(\frac{5}{2} \phi_m - 2\omega \right) + \omega(1 - \psi)^{-2} \right) \quad (3.5b)$$

$$\eta_e(\psi) = \eta_0 \bar{\eta}_e \psi (1 - \psi)^{-2} \quad (3.5c)$$

where η_0 is the viscosity of the suspending fluid, and $\bar{\eta}_e > 0$ is a constant independent of ψ . The constant $\omega \in]0, 1[$ is introduced in (3.5b) in order to recover Einstein (1906)'s relation when $\psi \rightarrow 0$ (see §3.4).

Similar to η_e , the parameters δ_2 and β should vanish when $\psi = 0$, i.e. when the suspension reduces to a Newtonian fluid. For simplicity, we consider that δ_2 is proportional to δ_1 . We also assume that β behave as $(1 - \psi)^{-2}$ when $\psi \rightarrow 1$. Accordingly, the following dependence laws are postulated:

$$\delta_2(\psi) = \bar{\delta}_2 \left((1 - \psi)^{-1} - (1 - b\psi) \right) \quad (3.5d)$$

$$\beta(\psi) = \bar{\beta} \left((1 - \psi)^{-1} - 1 \right)^2, \quad i = 1, 2, 3 \quad (3.5e)$$

where $\bar{\delta}_2$ and $\bar{\beta}$ are constants independent of ψ .

At this stage, the suspension model involves seven constants independent of ψ , namely a , b , ω , $\bar{\eta}_e$, $\bar{\delta}_1$, $\bar{\delta}_2$, and $\bar{\beta}$ that need to be determined from experimental data. Note that with the dependence laws (3.5b)-(3.5c) for η and η_e , we obtain from (3.4) that the normal stress ratios

$$\alpha_1 = \frac{N_1}{\eta_{app} |\dot{\gamma}|} \quad \text{and} \quad \alpha_2 = \frac{N_2}{\eta_{app} |\dot{\gamma}|}$$

are independent of suspending fluid viscosity η_0 , in agreement with experiments (Dbouk *et al.* 2013; Guazzelli & Pouliquen 2018).

3.4. Dilute and concentrated limits

In the dilute limit $\psi \rightarrow 0$, a second-order Taylor expansion of (3.4a) leads to the following expression for apparent viscosity η_{app} :

$$\eta_{app} = \eta_0 \left(1 + \frac{5}{2} \phi_m \psi + \left(\frac{3\omega}{2} + \frac{a(1+b)\bar{\delta}_2 \bar{\eta}_e}{(1-a^2)} \right) \psi^2 \right) + O(\psi^3) \quad (3.6a)$$

At first order in ψ , the model agrees with Einstein (1906)'s relation $\eta_{app} \approx \eta_0(1 + 5\phi/2)$. The different constants associated to micro-structure evolution, a , b , $\bar{\eta}_e$ and $\bar{\delta}_2$, appear only at second order, consistently with the asymptotic expansion of Batchelor & Green (1972).

In the concentrated limit $\psi \rightarrow 1$, the expansion of η_{app} writes:

$$\eta_{app} = \eta_0 \bar{\eta}_1 (1 - \psi)^{-2} + O((1 - \psi)^{-1}) \quad (3.6b)$$

with $\bar{\eta}_1 = \left(\omega + \left(1 + \frac{\bar{\beta}}{\bar{\delta}_1^2} \right) \bar{\eta}_e \right)$

Hence, η_{app} grows as $(1 - \psi)^{-2}$, as required (Maron & Pierce 1956).

Let us now turn to the asymptotic behaviour of normal stress ratios α_1 and α_2 and particle pressure p_p . In the dilute limit $\psi \rightarrow 0$, from (3.4), we obtain:

$$\begin{aligned}\alpha_1 &= \frac{2\bar{\eta}_e\bar{\delta}_1(1+b)}{1-a^2}\psi^2 + O(\psi^3) \\ \alpha_2 &= -\frac{(1-a)\bar{\eta}_e\bar{\delta}_1(1+b)}{1-a^2}\psi^2 + O(\psi^3) \\ \frac{p_p}{\eta_{app}|\dot{\gamma}|} &= -\frac{2a\bar{\eta}_e\bar{\delta}_1(1+b)}{3(1-a^2)}\psi^2 + O(\psi^3)\end{aligned}$$

Observe that α_1 , α_2 and p_p all vanish for $\psi = 0$, as expected. Moreover, we obtain $\alpha_1 \geq 0$ and $\alpha_2 \leq 0$ when $\psi \rightarrow 0$. The sign of α_2 is consistent with experimental observations (Couturier *et al.* 2011; Dbouk *et al.* 2013; Dai *et al.* 2013).

In the concentrated limit $\psi \rightarrow 1$, the expansions of normal stress ratios and particle pressure write:

$$\begin{aligned}\alpha_1 &= \frac{2\bar{\eta}_e}{\bar{\eta}_1\bar{\delta}_1}(1-\psi) + O((1-\psi)^2) \\ \alpha_2 &= -\frac{\bar{\eta}_e}{\bar{\eta}_1}\left(\frac{-\bar{\delta}_1\bar{\delta}_2 + \bar{\beta}}{\bar{\delta}_1^2}\right) + O(1-\psi) \\ \frac{p_p}{\eta_{app}|\dot{\gamma}|} &= \frac{2\bar{\eta}_e}{3\bar{\eta}_1}\left(\frac{-\bar{\delta}_1\bar{\delta}_2 + \bar{\beta}}{\bar{\delta}_1^2}\right) + O(1-\psi)\end{aligned}$$

Hence the ratio α_1 vanishes when $\psi \rightarrow 1$. On the contrary, α_2 tends to a constant for $\psi \rightarrow 1$, in agreement with experiments (Leighton & Acrivos 1987; Guazzelli & Pouliquen 2018). Furthermore, it is sufficient to assume $\bar{\delta}_2 < 0$ and $\bar{\beta} \geq 0$ to obtain $\alpha_2 \leq 0$.

4. Quantitative comparisons with experiments

4.1. Identification of model constants

As explained above, the rheological model proposed in this paper was built to recover qualitative agreement with experimental observations in the dilute and concentrated limits. More quantitatively, the seven model constants were identified through an adjustment to different experimental datasets reporting detailed rheological measurements on non-colloidal suspensions for different volume fractions ϕ (in the range $[0.2, 0.5]$). The four following datasets were considered for the adjustment:

- Experimental measurements of the depletion angle θ_e performed by Blanc *et al.* (2013). These authors used a suspending fluid of viscosity $\eta_0 = 0.85$ Pa.s and two sets of PMMA spheres of radius $r_p = 80$ and $90 \mu\text{m}$. They reported a maximal volume fraction $\phi_m = 0.57$.
- Measurements of transient apparent viscosity after a shear reversal performed by Blanc *et al.* (2011) for five different values of volume fraction ϕ . These authors used a suspending fluid of viscosity $\eta_0 = 1.02$ Pa.s, PMMA spheres of radius $r_p = 16 \mu\text{m}$, and reported a relatively low maximal volume fraction $\phi_m = 0.535$ presumably due to the presence of a residual cellulosic surfactant.
- Measurements of normal-stress differences in steady state performed by Couturier *et al.* (2011). These authors used a suspending fluid with viscosity $\eta_0 = 2.15$ Pa.s,

a	b	ω	$\bar{\eta}_e$	$\bar{\delta}_1$	$\bar{\delta}_2$	$\bar{\beta}$
0.51	1.5	0.52	0.089	0.47	-1.15	1.7

TABLE 1. Values of the seven dimensionless model constants identified from experimental data.

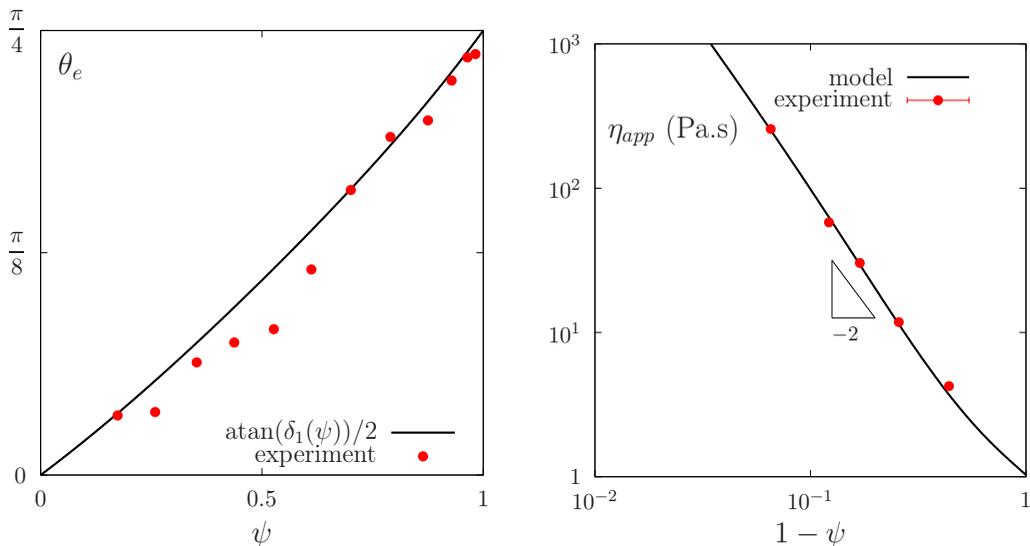


FIGURE 1. Evolution of depletion angle θ_e and steady-state apparent viscosity η_{app} versus reduced volume fraction ψ : comparison between model predictions and experimental data of [Blanc et al. \(2013\)](#) (θ_e) and [Blanc et al. \(2011\)](#) (η_{app}).

polystyrene spheres of radius $r_p = 35 \mu\text{m}$, and reported a maximal volume fraction $\phi_m = 0.62$.

- Measurements of normal-stress differences in steady state performed by [Dai et al. \(2013\)](#). These authors used a slightly shear-thinning suspending fluid with viscosity $1.17 \leq \eta_0 \leq 1.3 \text{ Pa}\cdot\text{s}$ for shear rates $0.01 \leq \dot{\gamma} \leq 100 \text{ s}^{-1}$, polystyrene spheres of radius $r_p = 20 \mu\text{m}$, and reported a maximal volume fraction $\phi_m = 0.62$.

For all these datasets, values of ϕ were rescaled in terms of reduced volume fraction $\psi = \phi/\phi_m$ by using the provided values of ϕ_m . As described in appendix B, the set of seven model constants $\{a, \omega, \bar{\eta}_e, \bar{\delta}_2, \bar{\beta}_1, \bar{\beta}_2, \bar{\beta}_3\}$ was *globally* identified from the datasets. The obtained values are indicated in table 1. Note that first attempts at trying to obtain a comparable quantitative agreement with both steady-state and transient measurements, failed. As a consequence, we chose to assign a stronger weight to the agreement with steady-state viscosity and normal-stress data. A lower weight was assigned to the agreement with transient experiments, although sufficient to ensure that model predictions present a correct qualitative behavior. More details on the identification procedure, including expression of the minimised cost function, are provided in appendix B.

Direct comparisons between model predictions with the constants given in table 1, and

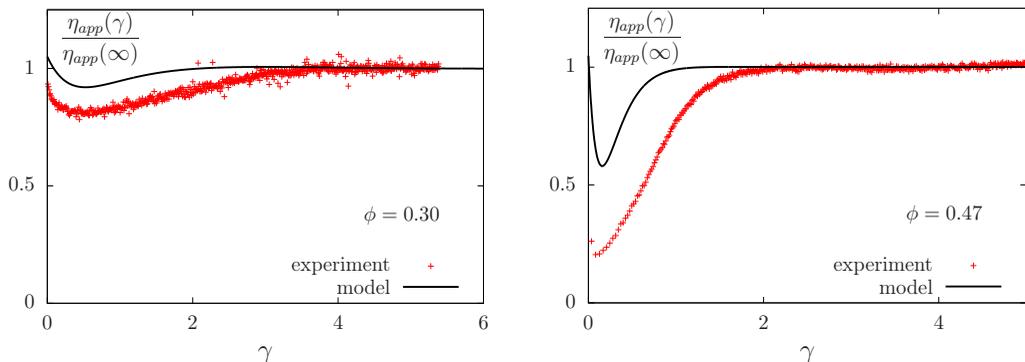


FIGURE 2. Evolution of apparent viscosity η_{app} versus shear strain γ after shear reversal: comparison between model prediction and experimental measurements of [Blanc *et al.* \(2011\)](#) for two volume fractions ϕ .

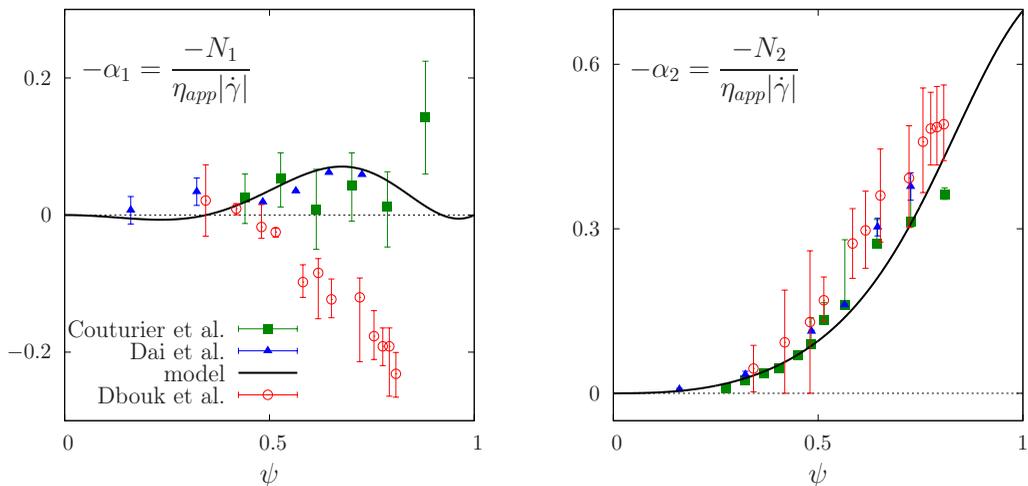


FIGURE 3. Evolution of steady-state normal stress ratios α_1 and α_2 versus reduced volume fraction ψ : comparison between model predictions and experimental measurements of [Couturier *et al.* \(2011\)](#), [Dai *et al.* \(2013\)](#) and [Dbouk *et al.* \(2013\)](#).

the experimental data used for the identification, are shown in figures 1, 2 and 3. Figure 1 shows that predicted steady-state depletion angle and apparent viscosity are indeed in excellent quantitative agreement with the experimental data. Figure 2 presents comparisons for the transient response, namely evolution of apparent viscosity $\eta_{app}(t)$ versus strain $\gamma(t) = \int_0^t |\dot{\gamma}(s)| ds$ after a shear reversal. Let us recall that the experiments of [Blanc *et al.* \(2011\)](#) were performed at imposed shear stress, while $\dot{\gamma}(t)$ varies. Note that both model predictions and experimental data are normalised by their respective steady state-values $\eta_{app}(\infty)$ in the figure. It is observed that, although the model generally tends to underestimate the amplitude of the transient response, a satisfactory qualitative behavior is nevertheless obtained for all values of ϕ . In particular, the successive phases present in the measurements, namely the initial brutal drop in apparent viscosity, the smooth evolution to a minimum, and the relaxation towards the steady state, are all reproduced. The typical duration of the transient phase is also correctly captured.

Figure 3 presents the evolution of steady-state normal stress ratios α_1 and α_2 as a function of reduced volume fraction ψ . The model appears to successfully capture the

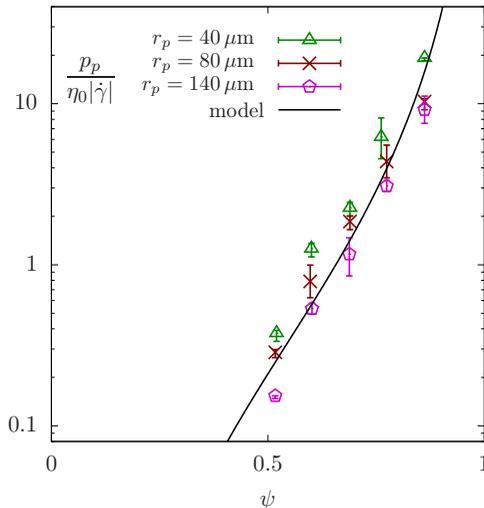


FIGURE 4. Evolution of steady-state normalized particle pressure $p_p/(\eta_0|\dot{\gamma}|)$ versus reduced volume fraction ψ : comparison between model predictions and experimental measurements of Deboeuf *et al.* (2009). The consistent data obtained for three different values of particle radius r_p are shown.

main features of the normal stress measurements reported in the studies of Couturier *et al.* (2011) and Dai *et al.* (2013), namely $N_2 < 0$ and $|N_1| < |N_2|$. Predictions for the second normal-stress ratio α_2 show a strong monotonic decrease with ψ , and are in excellent quantitative agreement with the data (figure 3-right). Regarding the first normal-stress difference, the model predicts non-monotonic variations of the ratio α_1 with ψ (figure 3-left). Values of α_1 are positive but very small for $\psi \lesssim 0.4$, negative and decreasing with ψ for $0.4 \lesssim \psi \lesssim 0.7$, and then increasing again towards the concentrated limit $\alpha_1(1) = 0$ (see § 3.4). Here also, these predictions appear to be in good quantitative agreement with the measurements in terms of overall magnitude, albeit the existence of non-monotonic variations cannot be confirmed from existing data due to the level of experimental noise. For comparison, experimental measurements obtained by Dbouk *et al.* (2013) (not used for the adjustment) are also shown in figure 3, using a value $\phi_m = 0.58$ for the volume fraction scaling. These measurements are fully consistent with the two other datasets and the model for α_2 , but show positive values of α_1 , thereby highlighting the need for additional characterisations of the first normal-stress difference in these systems.

4.2. Comparison with independent particle pressure measurements

Model predictions were also assessed against steady-state particle pressure measurements obtained by Deboeuf *et al.* (2009). These authors used a suspending fluid of viscosity $\eta_0 = 3$ Pa.s and different sets of particles with radii ranging between 40 and 140 μm . As they did not provide the value of maximum volume fraction ϕ_m in their systems, we used $\phi_m = 0.58$ to rescale their data. We insist that this new dataset was not used for the identification of model constants. As shown in figure 4, model predictions obtained with the values of constants given in table 1, show here also a good agreement with the experimental measurements. This remarkable result validates the introduction of reduced volume fraction $\psi = \phi/\phi_m$ in the dependence laws for the material parameters (see §3.3), and indicates that the model constants involved in these dependence laws

can be regarded, at least to a first approximation, as independent of the experimental conditions. In other words, the influence of experimental conditions is fully encoded in the maximum volume fraction ϕ_m used to rescale the volume fraction values.

5. Discussion and conclusions

An improved, rate-independent rheological model for non-colloidal suspensions of rigid spheres, involving the evolution of a conformation tensor, was presented. The model extends that of [Ozenda *et al.* \(2018\)](#) in several respects. While the structure of the linear evolution equation (2.1a) for the conformation tensor is unchanged, a more general Gordon-Schowalter tensor derivative is used, with a material parameter $a \in]0, 1[$. In addition, the expression of the Cauchy stress tensor (2.1b) is modified by the inclusion of additional linear and non-linear terms. While still keeping with the goal of a minimal model involving as few adjustable parameters as possible, these amendments were required to predict both the transient evolution of apparent viscosity after a shear reversal, and realistic values for the normal stress difference N_1 and N_2 in steady state.

The model was compared to different experimental datasets involving various suspending fluids and particle sizes. To that aim, the material parameters were all expressed in terms of reduced volume fraction $\psi = \phi/\phi_m$, where the maximum volume fraction ϕ_m varies among the experiments. The good agreement observed with steady-state data, in terms of apparent viscosity, normal stress differences and particle pressure, suggests that the rheological model, together with the constants given in table 1, can be applied to large range of non-colloidal suspensions. Regarding the transient responses, only qualitative agreement was obtained with experimental data. Further improvements of these transient predictions would certainly require complementing our minimal model with additional terms, for instance polynomial dependences of the material parameters upon the invariants of the texture tensor ([Hand 1962](#)), or introduction of additional microstructural tensorial variables ([Goddard 2006](#)). Along the same line, introduction of a plasticity term to the constitutive law would open interesting perspectives to tackle highly-concentrated regimes close to, or above, the jamming transition, in which the rheological behavior becomes dominated by sustained granular contacts ([Pailha & Pouliquen 2009](#); [Baumgarten & Kamrin 2019](#)).

Appendix C presents a systematic sensitivity study of model predictions (steady-state normal-stress ratios and transient apparent viscosity) with respect to the different model constants. It appears in particular that none of the newly-introduced terms in the Cauchy stress equation can be omitted without significantly degrading the match with observations. We also note the existence of non-trivial couplings between these different terms, which explains the difficulties encountered in the identification procedure when trying to simultaneously reproduce steady-state and transient experimental data (see §4). The possibility to work out a more systematic identification procedure, including a physical interpretation of the different terms, shall be investigated in future work.

The ability of the presented model to predict accurate normal-stress differences potentially opens way to computations of elongational or more complex flows. The model could also be coupled to an evolution equation for the volume fraction in order to address migration phenomena ([Miller & Morris 2006](#)). Here also, detailed comparisons with experiments (e.g. [Dai & Tanner 2017](#)) will however be necessary to assess the validity of the model beyond the simple shear configuration investigated in this study. Beyond rheological experiments, addressing migration processes, e.g. around an obstacle ([Haddadi *et al.* 2014](#)), would test the effect of normal-stress differences on longer time

scales and allow for comparisons with other modelling approaches (e.g., Dbouk 2016). Prior to turning to such complex flows, a more in-depth analysis of the mathematical and thermodynamical properties of the model will also need to be undertaken. Finally, let us also note that, although the focus of the present paper was mainly on trying to reproduce physical experiments, systematic comparisons with discrete simulation results (e.g. Chacko *et al.* 2018; Singh *et al.* 2018) could constitute a fruitful avenue for further validating and improving our model. Such discrete simulations can indeed provide useful guidance for continuous rheological modelling, through the calculation of variables that remain currently unamenable to physical measurements (e.g., transient evolutions of normal stress differences).

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Appendix A. Eigen directions of γ_e

We prove here that, in stationary simple shear flow, the depletion angle, i.e. the angle between the x axis and the direction of the eigenvector associated to the largest eigenvalue of γ_e , expresses as:

$$\delta_1 = \tan(2\theta_e) \quad (\text{A } 1)$$

From (3.3a)-(3.3c), eigenvalues of γ_e , denoted as $\lambda_{min} \leq \lambda_{max}$, are both non-zero and write:

$$\lambda_{min} = \frac{\delta_1^{-1}}{1 + \delta_1^{-2}(1 - a^2)} \left(a\delta_1^{-1} - (\delta_1^{-2} + 1)^{1/2} \right)$$

$$\lambda_{max} = \frac{\delta_1^{-1}}{1 + \delta_1^{-2}(1 - a^2)} \left(a\delta_1^{-1} + (\delta_1^{-2} + 1)^{1/2} \right)$$

The corresponding eigenvectors are:

$$\mathbf{v}_{min} = \begin{bmatrix} \delta_1^{-1} - (\delta_1^{-2} + 1)^{1/2} \\ 1 \end{bmatrix} \quad \mathbf{v}_{max} = \begin{bmatrix} 1 \\ (\delta_1^{-2} + 1)^{1/2} - \delta_1^{-1} \end{bmatrix}$$

Accordingly, we obtain:

$$\theta_e = \text{atan} \left((\delta_1^{-2} + 1)^{1/2} - \delta_1^{-1} \right) = \frac{\text{atan}(\delta_1)}{2}$$

which is equivalent to (A 1). Observe that $\forall \delta_1 > 0$, $\theta_e \in]0, \pi/4[$.

Remark that, while the eigenvalues of γ_e depend on the parameter a of the tensor derivative, the eigenvectors are independent thereof. Hence, all the relations between microstructure orientation and model parameters derived in Ozenda *et al.* (2018), notably (A 1), remain unchanged in the present model.

Appendix B. Identification of model constants

The rheological model proposed in this paper involves seven constants to be identified: a , b , ω , $\bar{\eta}_e$, $\bar{\beta}$, $\bar{\delta}_1$, $\bar{\delta}_2$. Global identification of these parameters with respect to

removed

TABLE 2.

removed

FIGURE 5.

experimental data was achieved by minimising the following cost function:

$$\begin{aligned}
 J = & \frac{w_a}{\psi_2 - \psi_1} \int_{\psi_1}^{\psi_2} |\theta_e(\psi) - \theta_e^{obs}(\psi)|^2 d\psi + \frac{w_v}{\psi_4 - \psi_3} \int_{\psi_3}^{\psi_4} |\eta_{app}(\psi) - \eta_{app}^{obs}(\psi)|^2 d\psi \\
 & + \sum_{i=1}^2 \frac{w_{s,i}}{\psi_m} \int_0^{\psi_m} |\alpha_i(\psi) - \alpha_i^{obs}(\psi)|^2 d\psi \\
 & + \sum_{k=1}^5 \frac{w_{t,k}}{\gamma_k} \int_0^{\gamma_k} |\eta_{app}(\psi_k, \gamma) - \eta_{app,k}^{obs}(\gamma)|^2 d\gamma
 \end{aligned}$$

The function $\theta_e^{obs}(\psi)$ is a piecewise-constant interpolation of steady-state depletion angle measurements reported by [Blanc *et al.* \(2013\)](#) for $\psi \in [\psi_1, \psi_2]$, with $\psi_1 = 0.088$ and $\psi_2 = 0.98$ the range of reduced volume fraction explored in the experiments. The function $\eta_{app}^{obs}(\psi)$ is a piecewise-constant interpolation of steady-state values of apparent viscosity measured by [Blanc *et al.* \(2011\)](#) for $\psi \in [\psi_3, \psi_4]$, with $\psi_3 = 0.56$ and $\psi_4 = 0.93$ in this case. The functions $\alpha_1^{obs}(\psi)$ and $\alpha_2^{obs}(\psi)$ are piecewise-constant interpolations of steady-state values of normal stress ratios measured by ([Couturier *et al.* 2011](#)) and ([Dai *et al.* 2013](#)) for $\psi \in [0, \psi_m]$, with $\psi_m = 0.81$ the maximal reduced volume fraction reached in the experiments. The functions $\eta_{app,k}^{obs}(\gamma)$, $k = 1 \dots 5$, represent the transient evolutions of apparent viscosity during the five shear reversal experiments of [Blanc *et al.* \(2011\)](#) characterised by reduced volume fractions ψ_k . Here, γ_k , $k = 1 \dots 5$, represent the maximal strain reached in each experiment. Lastly, the function $\eta_{app}(\psi, \gamma)$ is associated to the transient solution of the model, while the functions $\theta_e(\psi)$, $\eta_{app}(\psi)$, $\alpha_1(\psi)$, and $\alpha_2(\psi)$ are associated to the steady-state solution. The factors w_a , w_e , $w_{s,i}$ and $w_{t,k}$ are the relative weights of the different terms of the cost function J .

The minimization problem of J was solved with a Levenberg-Marquardt method ([More *et al.* 1980](#)), using the optimisation library `lmfit` ([Newville *et al.* 2016](#)). As explained in §4, larger values had to be used for the weights w_a , w_e , $w_{s,i}$, with respect to $w_{t,k}$, in order to promote the agreement with steady-state measurements. In addition, after the global identification step, a secondary, one-by-one, adjustment of each model constants was performed in order to further improve the solution. Final values of identified model constants are indicated in table 1.

Appendix C. Sensitivity study

Figure 6 shows the sensitivity of steady-state normal stress ratios α_1 and α_2 with respect to the seven model constants a , b , ω , $\bar{\eta}_e$, $\bar{\beta}$, $\bar{\delta}_1$, $\bar{\delta}_2$. Similarly, figure 7 presents a sensitivity study for the transient apparent viscosity after a shear reversal.

Overall, the ratios α_1 and α_2 are only weakly sensitive to the constant ω (figure 6). Conversely, variations of all the other constants lead to significant qualitative changes,

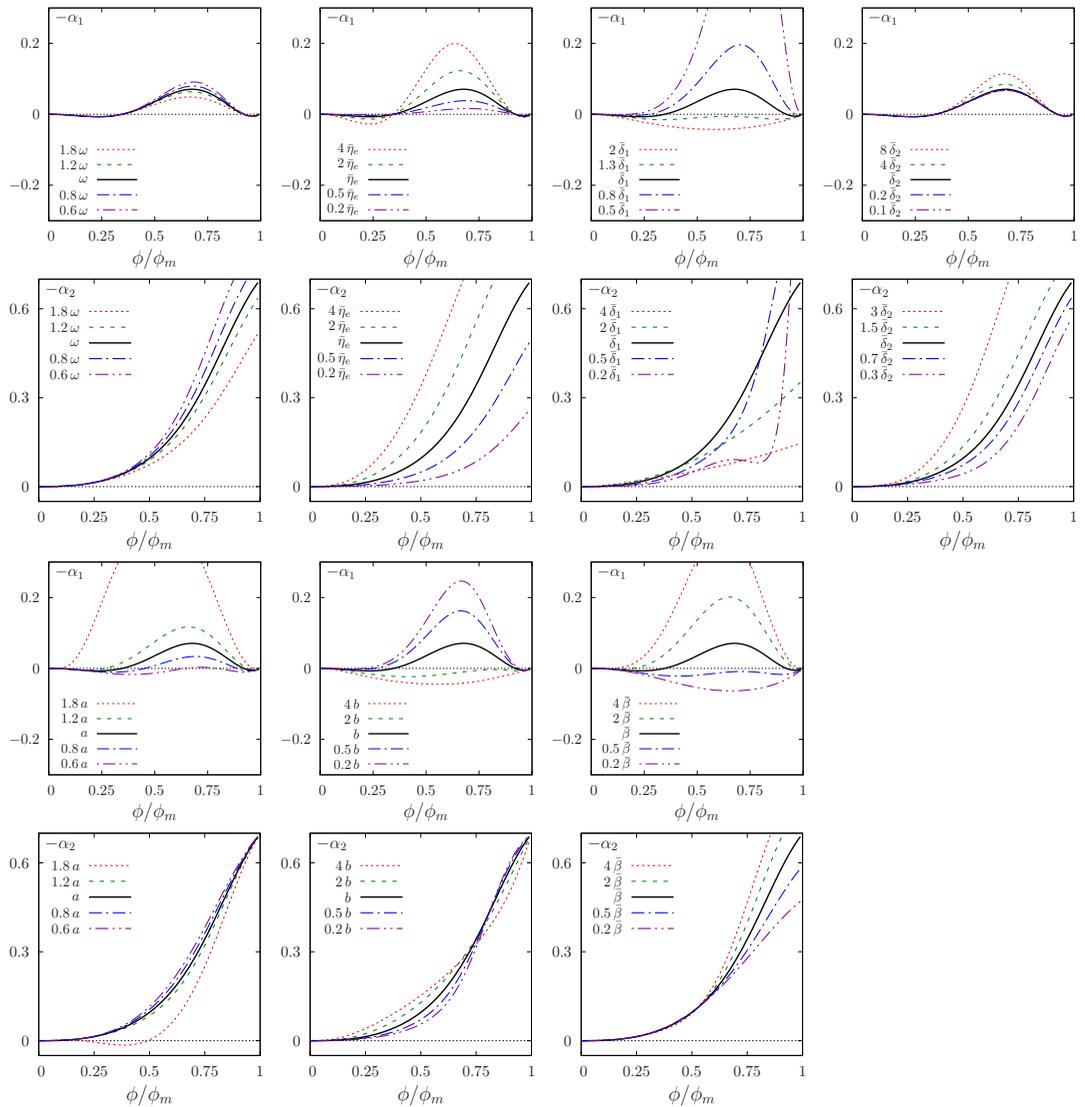


FIGURE 6. Sensitivity of normal stress ratios α_1 and α_2 with respect to the seven model constants: ω , $\bar{\eta}_e$, $\bar{\delta}_1$, $\bar{\delta}_2$, a , b , $\bar{\beta}$. Each of the constants was varied around the reference values indicated in table 1.

such as non-monotonous variations of α_2 with ψ . Note in particular that unphysical predictions $|\alpha_1| \geq |\alpha_2|$ are obtained when values of $\bar{\delta}_1$ or $\bar{\beta}$ are close to zero, or when $a \approx 1$. Similarly, an unphysical change of sign of α_2 is observed when a is large. These results show that all the terms introduced in the constitutive equations (2.1) contribute to obtaining correct normal stress predictions.

Figure 7 shows a weak sensitivity of the transient apparent viscosity with respect to model constants a and $\bar{\delta}_2$. Conversely, the constant ω has a direct influence on the value of the apparent viscosity minimum, while $\bar{\delta}_1$ affects the minimum position. Lastly, $\bar{\eta}_e$, $\bar{\delta}_1$ and $\bar{\beta}$ affect the steady-state regime.

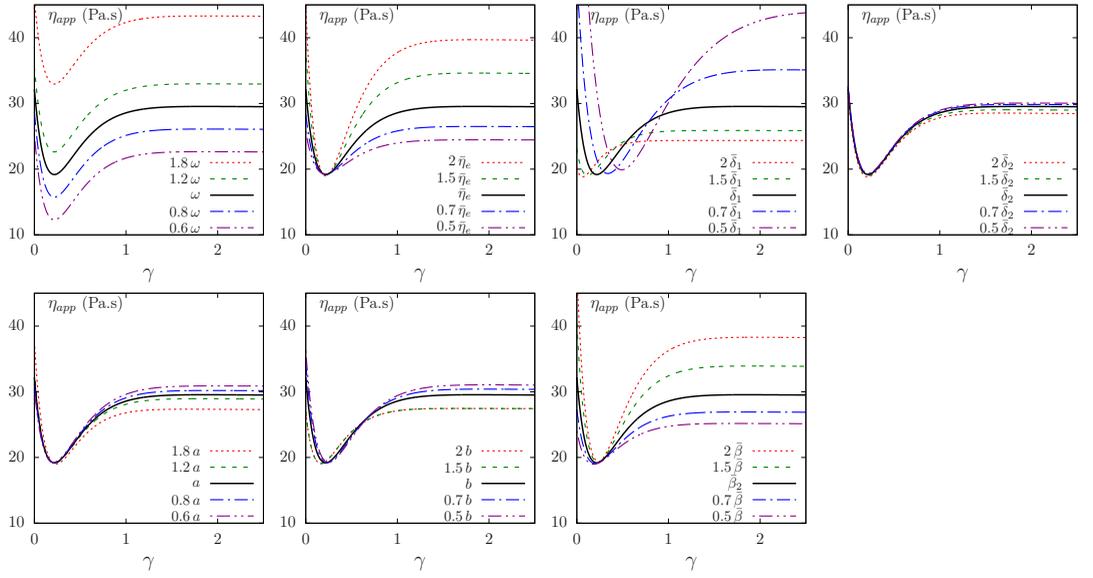


FIGURE 7. Sensitivity of transient apparent viscosity η_{app} with respect to the eight model constants: ω , $\bar{\eta}_e$, $\bar{\delta}_1$, $\bar{\delta}_2$, a , b , $\bar{\beta}$. Each of the constants was varied around the reference values indicated in table 1.