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A dissipative anisotropic fluid model for non-colloidal particle dispersions

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This article examines a reduced form of the 'purely dissipative' model proposed several years ago as a general continuum model for the rheology of non-colloidal particle dispersions, ranging from Stokesian suspensions to non-cohesive granular media. Essential to the model is a positive-definite viscosity tensor $\boldsymbol{\eta}$, depending on the history of deformation and providing a crucial restriction on related models for anisotropic fluids and suspensions. In the present treatment, $\boldsymbol{\eta}$ is assumed to be as an isotropic function of a history-dependent second-rank 'texture' or 'fabric' tensor $\boldsymbol{A}$. A formula for $\boldsymbol{\eta}(\boldsymbol{A})$ borrowed from the analogous theory of linear elasticity, and its subsequent expansion for weak anisotropy provides an explicit expression for the stress tensor in terms of fabric, strain-rate and eight material constants.

Detailed consideration is given to the special case of Stokesian suspensions, which represent an intriguing subset of memory materials without characteristic time. For this idealized fluid one finds linear dependence of all stresses, including viscometric normal stress, on present deformation rate, with the provision for an arbitrary fabric evolution ('thixotropy') in unsteady deformations. As a concrete example, a co-rotational memory integral is adopted for $\boldsymbol{A}$ in terms of strain-rate history, and a memory kernel with two-mode exponential relaxation gives close agreement with the rather sparse experimental data on transient shear experiments. In the proposed model, an extremely rapid mode of relaxation is required to mimic the incomplete reversal of stress observed in experiments involving abrupt reversal of steady shearing, supporting the conclusion of others that non-hydrodynamic effects, with breaking of Stokesian symmetry, may be implicated in such experiments.

Qualitative comparisons are made to a closely related model, derived from a micro-mechanical analysis of Stokesian suspensions, but also involving non-Stokesian effects.

The present analysis may point the way to improved micro-mechanical analysis and to further experiments. Possible extensions of the model to the viscoplasticity of dry and liquid-saturated granular media also are discussed briefly.

1. Introduction

The main focus of this paper is the continuum mechanics of Stokesian suspensions, i.e. idealized suspensions of rigid neutrally buoyant particles in Newtonian liquids, in which inertia and all forces other than those arising from viscosity are negligible. With impetus from the seminal work of Batchelor (1970), which *inter alia* introduces the idea of a viscosity tensor, considerable progress has been made over the past three decades in the basic theoretical understanding and description of suspension micro-mechanics. Stickel & Powell (2005) cover several aspects of dense-suspension rheology, including a discussion of both Brownian and inertial effects. The latter represent prominent examples of Stokesian symmetry breaking, with significant qualitative changes in suspension rheology. More subtle effects may arise, however, from multi-body hydrodynamic interactions, which recent theoretical studies (Drazer *et al.* 2002, 2004) have definitively established as chaotic in nature. Stokesian chaos provides a compelling theoretical explanation of effects such as hydrodynamic diffusion (Leighton & Acrivos 1987; Acrivos *et al.* 1992; Drazer *et al.* 2002) and the 'loss of memory', which was dubbed 'effaceable memory' by Goddard (1982) without elucidation of its origins. Moreover, chaos may be implicated in several remarkable aspects of suspension rheology, including the development of anisotropic microstructure (Husband & Gadala-Maria 1987; Parsi & Gadala-Maria 1987), with linear dependence of viscometric normal stress on shear rate (Zarraga, Hill & Leighton 2000; Singh & Nott 2003), and the peculiar transient shear behaviour observed by Gadala-Maria & Acrivos (1980); Kolli, Pollauf & Gadala-Maria 2002).

With this as backdrop, the present study revisits a continuum theory of dissipative materials without characteristic time proposed by Goddard (1984) as models for Stokesian suspensions and non-cohesive granular media. We set aside here the general issues of material symmetry addressed in that paper, which, incidentally, overlooks the revision by Noll (1972) of his previous and widely known classification scheme. The narrower objective here is to set down a special continuum framework for particulate dispersions, as a guide to further micro-mechanical analysis and experiment.

2. Simplified model of dissipative materials

In the following, boldface symbols are employed for tensors, with lower- and upper-case Roman denoting vectors and second-rank tensors, respectively, lower-case Greek denoting tensors of higher rank, and superscript T denoting transposition of a second-rank tensor. Cartesian components, with the standard summation convention, are displayed where necessary for clarity. The standard mathematical symbol $\otimes$ is employed for tensor products, so that $\boldsymbol{\alpha} = \boldsymbol{a} \otimes \boldsymbol{B}$ stands for $\alpha_{ijk} = a_i B_{jk}$, etc. In the usual way, colons indicate ordered pairwise contraction of the trailing prefactor indices with the leading postfactor indices. A single dot denotes the scalar product of vectors, but is omitted from products indicating linear transformation of vectors by second-rank tensors. $\boldsymbol{v}(\boldsymbol{x}, t)$ denotes the velocity field at spatial position $\boldsymbol{x}$ and time t, with notation for material points suppressed and local kinematics assumed to represent materially homogeneous deformations.

2.1. *Reduced viscosity-fabric dependence*

In the literature on complex fluids such as suspensions, liquid crystals, polymers and granular media (Hand (1962); Barthès-Biesel & Acrivos 1973; Cowin 1985; Bird, Armstrong & Hassager 1987; Phan-Thien 1995; Goddard 1998; Larson 1999; Fang *et al.* 2002), the microstructure and its evolution is often described by means of internal variables or order parameters, given by certain 'texture' tensors. These generally represent various statistical moments

$$\langle \boldsymbol{a} \rangle, \langle \boldsymbol{a} \otimes \boldsymbol{a} \rangle, \ldots, \langle \boldsymbol{a} \otimes \boldsymbol{a} \otimes \boldsymbol{a} \otimes \boldsymbol{a} \rangle, \ldots, \tag{1}$$

with respect to a microstructural probability distribution $f(\boldsymbol{a}, t)$ of vector-valued 'director' $\boldsymbol{a}$. When $\boldsymbol{a}$ is a unit vector, these are sometimes referred to as 'fabric tensors' in the literature on image processing and granular media (Kanatani 1984; Goddard 1998).

Basic analyses of the statistical micro-mechanics are usually couched in terms of probability distributions $f(\boldsymbol{a}, t)$, as illustrated by various suspension theories (Hand 1962; Barthès-Biesel & Acrivos 1973; Hinch & Leal 1975; Szeri & Leal 1994; Brady & Morris 1997), where $\boldsymbol{a}$ represents particle orientation or pairwise separation. The usual goal of such analyses is to extract continuum-level constitutive equations for the evolution of quantities like those in (1) and derived quantities such as the stress tensor $\boldsymbol{T}$. To obtain tractable equations, one often seeks closure approximations giving higher-order moments in terms of a select set of lower-order ones (Hinch & Leal 1975; Szeri & Leal 1994; Phan-Thien 1995), and similar ideas motivate our simplification of the general continuum model considered here.

For general dissipative materials (Goddard 1984), the current (Cauchy) stress $\boldsymbol{T}(t)$ at a given material point is determined by the local strain rate $\boldsymbol{E}(t) = [\nabla \boldsymbol{v} + (\nabla \boldsymbol{v})^T]/2$ and a positive-definite fourth-rank viscosity tensor as

$$\boldsymbol{T}(t) = 2\boldsymbol{\eta}\{\mathfrak{h}\} : \boldsymbol{E}(t), \quad (T_{ij} = 2\eta_{ijkl}E_{kl} \quad \text{with } \eta_{ijkl} = \eta_{jikl} = \eta_{ijlk}), \tag{2}$$

where $\mathfrak{h}$ represents the history of deformation, to be specified by appropriate kinematic tensors in a frame-indifferent set of evolution equations. In the case of incompressible materials such as Stokesian suspensions, (2) defines stress only up to an arbitrary additive isotropic pressure or, with the restriction $\eta_{iijk} = 0 = \eta_{ijkk}$, it defines the deviatoric stress.

With the requirement of frame indifference and suitable restrictions on the evolution equations, (2) defines a class of 'simple materials', but one not necessarily endowed with fading memory (Coleman & Noll 1961; Truesdell & Noll 1965; Goddard 1984). Indeed, $\boldsymbol{\eta}\{\mathfrak{h}\}$ may even exhibit singular dependence on sets of zero measure, e.g. on $\boldsymbol{E}(t)$, as discussed further below.

As a major simplification, of the type employed by Cowin (1985) for the analogous problem in anisotropic elasticity, we assume that $\boldsymbol{\eta}$ is given uniquely as an isotropic function of a symmetric second-rank fabric tensor $\boldsymbol{A} = \boldsymbol{A}^T$, which in turn depends on $\mathfrak{h}$. It follows from (2) that $\boldsymbol{T}$ can then be written as an isotropic tensor polynomial in $\boldsymbol{A}, \boldsymbol{E}$, with scalar coefficients depending on their joint isotropic scalar invariants.

Before specializing to the linear forms in $\boldsymbol{E}$ appropriate to Stokesian suspensions, we note the relation to the Ericksen–Hand anisotropic fluid model, regarded by several past investigators as a possible model of particulate suspensions (Hand 1962; Barthès-Biesel & Acrivos 1973). Equation (2) of Hand (1962) gives the stress $\boldsymbol{T}$ as a linear combination of nine distinct symmetric tensors given, respectively, by symmetrization of the set:

$$\{\boldsymbol{P}^{(mn)} := \boldsymbol{A}^m \boldsymbol{E}^n \, : m, n = 0, 1, 2\}, \tag{3}$$

involving nine independent coefficients depending on the joint isotropic scalar invariants:

$$\{\operatorname{tr}(\boldsymbol{P}^{(mn)}) : 1 \leqslant n + m \leqslant 3\}, \tag{4}$$

where exponents zero represent the unit tensor $\mathbf{1}$.

We further recall that the evolution of $\boldsymbol{A}$ in Hand's model is represented by giving the co-rotational (Jaumann) rate

$$\mathscr{D}_t \boldsymbol{A} = d_t \boldsymbol{A} + \boldsymbol{A}\boldsymbol{W} - \boldsymbol{W}\boldsymbol{A}, \tag{5}$$

as an isotropic function of $\boldsymbol{A}, \boldsymbol{E}$ having the same general form as that for $\boldsymbol{T}$. Here, $\boldsymbol{W}(t) = [\nabla \mathbf{v} - (\nabla \mathbf{v})^T]/2$ denotes vorticity and $d_t = \partial_t + \boldsymbol{v} \cdot \nabla$ the material derivative.

The restriction to linearity in $\boldsymbol{E}$ (Hand 1962; Barthès-Biesel & Acrivos 1973), i.e. to $n = 1$ in (3), leads to a linear form in $\boldsymbol{E}$ with 11 independent coefficients depending only on the scalar invariants of $\boldsymbol{A}$, represented by $n = 0$ in (4). On the other hand, the corresponding expression for stress given by equation (5) of Cowin (1985) involves only nine independent coefficients, owing essentially to a required symmetry of the form:

$$\eta_{ijkl} = \eta_{klij} \tag{6}$$

and some further reductions arising from isotropy (see below). The symmetry of elastic moduli is guaranteed by the existence of a strain-energy function, whereas it may be viewed generally as an Onsager relation for linear dissipation. In the present context, symmetry is tantamount to the principle of minimum dissipation, a property of the underlying Stokesian dynamics, which we recall has been employed previously for suspension-viscosity estimates by Nunan & Keller (1984). Without a fully rigorous proof, we therefore adopt (6).

The required positivity of dissipation $\mathrm{tr}(\boldsymbol{TE})$ places additional restrictions on the nine coefficients in the above linear form, restrictions that we do not record here explicitly.

As a further simplification, we identify $\boldsymbol{A}$ with the deviator of a positive definite tensor, e.g. $\langle \boldsymbol{n} \otimes \boldsymbol{n} \rangle$, with $\boldsymbol{n}$ denoting a unit vector and $\mathrm{tr}(\langle \boldsymbol{n} \otimes \boldsymbol{n} \rangle) = 1$:

$$\boldsymbol{A} = \langle \boldsymbol{n} \otimes \boldsymbol{n} \rangle - \tfrac{1}{3}\mathbf{1}, \text{ and } \mathrm{tr}(\boldsymbol{A}) = 0. \tag{7}$$

In the usual interpretations of granular fabric, $\boldsymbol{n}$, is identified with the line of centres of neighbouring particles, as done by Phan-Thien (1995) for dense suspensions of nearly spherical particles.

Replacement of $\langle \boldsymbol{n} \otimes \boldsymbol{n} \rangle$ by any symmetric tensor with unit trace gives a somewhat more general result. Also, a tensor $\boldsymbol{A}$ with non-zero trace might serve to represent isotropic changes in suspension microstructure, such as those associated with granular dilatancy (Didwania, Ledniczky & Goddard 2001). Since the physical basis for such effects in Stokesian suspensions is less than evident, we invoke Occam's razor to reduce the number of parameters.

Then, the trace norm $|\boldsymbol{A}| = \{\mathrm{tr}(\boldsymbol{A}^2)\}^{1/2}$ provides a measure of anisotropy and, given a smooth dependence on $|\boldsymbol{A}|$, we may expand equation (5) of Cowin (1985) for small $|\boldsymbol{A}|$ to yield:

$$\begin{aligned}\boldsymbol{T} = -p\mathbf{1} + 2[\eta_0 + \eta_2 \mathrm{tr}(\boldsymbol{A}^2) + \eta_3 \mathrm{tr}(\boldsymbol{A}^3)]\boldsymbol{E} + [\nu_2 \mathrm{tr}(\boldsymbol{AE}) + \nu_3 \mathrm{tr}(\boldsymbol{EA}^2)]\boldsymbol{A} + \nu_3 \mathrm{tr}(\boldsymbol{AE})\boldsymbol{A}^2 \\ + [\mu_1 + \mu_3 \mathrm{tr}(\boldsymbol{A}^2)](\boldsymbol{AE} + \boldsymbol{EA}) + \mu_2(\boldsymbol{A}^2\boldsymbol{E} + \boldsymbol{EA}^2) + O(|\boldsymbol{A}|^4)\end{aligned} \tag{8}$$

involving an arbitrary isotropic pressure p and eight distinct material coefficients $\eta_0, \ldots, \nu_2, \ldots, \nu_3$, whose subscripts indicate the algebraic order in $|\boldsymbol{A}|$ of their postfactors. These coefficients depend generally on particle volume fraction ϕ and other non-dimensional parameters necessary to define particle geometry.

To complete the constitutive model, we require a suitable evolution equation for $\boldsymbol{A}$. Ideally, this should be obtained from a detailed micro-mechanical analysis, of the type proposed for certain model suspensions (Barthès-Biesel & Acrivos 1973; Hinch & Leal 1975; Szeri & Leal 1994; Phan-Thien 1995; Morris & Brady 1996; Brady & Morris 1997; Morris & Katyal 2002), but for the present purposes, we shall adopt a phenomenological model, motivated by certain global aspects of Stokesian dynamics.

At this juncture, it is worth discussing the closely related work of Phan-Thien (1995), who proposes a model for Stokesian suspensions based on a special case of (8), with the only non-zero coefficients being η_0, μ_1, ν_2, plus a linear combination of terms $\boldsymbol{AEA}$ and $|\boldsymbol{E}|\boldsymbol{A}$. The first of these, which also appears in the model of Barthès-Biesel & Acrivos (1973), can be reduced to a linear combination of terms already appearing in (8) by means of a well-known result of Rivlin (1955).

The second term $|\boldsymbol{E}|\boldsymbol{A}$ is construed by Phan-Thien to represent statistical fluctuations, by analogy to Brownian effects in suspensions (Morris & Brady 1996; Morris & Katyal 2002). Terms of this type, homogeneous of degree one but nonlinear in $\boldsymbol{E}$, represent broken Stokesian symmetry, whose origin is not exactly clear, but whose effects are considered below. It is worth noting that the addition of a linear term in $|\boldsymbol{E}|$ to the Stokesian form (2) represents the viscous analogue of 'hypoplasticity' (Kolymbas 2000), where a similar term serves to break Hookean symmetry.

3. Stokesian suspensions

In addition to the property of strict dissipation, we can infer from the properties of the Stokes equations certain important aspects of the history dependence of various continuum properties. As an illustration, we first consider sphere suspensions, where angular orientation of particles can be ignored.

In an unbounded suspension of spheres subject to uniform global strain rate $\boldsymbol{E}(t)$ and vorticity $\boldsymbol{W}(t)$, the current value of all effective continuum properties, including the viscosity tensor $\boldsymbol{\eta}$, is presumably determined by $\boldsymbol{E}$, $\boldsymbol{W}$ and the instantaneous configuration $\mathscr{C}(t) = \{\boldsymbol{x}^\alpha(t),\ \alpha = 1, 2, \ldots\}$. where $\boldsymbol{x}^\alpha$ represents the centre of sphere α. The latter is governed by a frame-indifferent set of ODEs, linear in $\boldsymbol{E}$, $\boldsymbol{W}$, assumed in Goddard (1982) and given by Stokesian resistance formulae, e.g. in Bossis & Brady (1984):

$$\frac{\mathrm{d}\boldsymbol{x}^\alpha}{\mathrm{d}t} = \boldsymbol{W}\boldsymbol{x}^\alpha + \boldsymbol{\xi}^\alpha : \boldsymbol{E} \quad (\alpha = 1, 2, \ldots, N), \tag{9}$$

subject to an initial condition $\mathscr{C}(0)$ at an arbitrary time origin $t = 0$. Here $\boldsymbol{\xi}^\alpha$ is a third-rank tensor depending on $\mathscr{C}(t)$, with dilute (Einstein) limit

$$\boldsymbol{\xi}^\alpha : \boldsymbol{E} \rightarrow \boldsymbol{x}^\alpha \cdot \boldsymbol{E} \equiv \boldsymbol{E}\boldsymbol{x}^\alpha \quad \text{for } \phi \rightarrow 0,$$

where the spheres move as fluid particles. For larger ϕ, (9) apparently becomes chaotic owing to multi-body hydrodynamic interactions (Drazer *et al.* 2002).

Frame indifference permits the transformation of (9), by means of a particular time-dependent orthogonal transformation $\boldsymbol{Q}(t)$, to a co-rotational form:

$$\frac{\mathrm{d}z^\alpha}{\mathrm{d}t} = \boldsymbol{\zeta}^\alpha : \boldsymbol{H} \quad \text{with } z^\alpha = \boldsymbol{Q}^T\boldsymbol{x}^\alpha, \tag{10}$$

where

$$\boldsymbol{\zeta}^\alpha = \boldsymbol{\xi}^\alpha(\mathscr{C}_z(t)) \quad \text{with } \mathscr{C}_z(t) = \{z^\alpha,\ \alpha = 1, 2, \ldots\},$$

and $\boldsymbol{H}$ denotes the co-rotational strain rate:

$$\boldsymbol{H}(t) = \boldsymbol{Q}^T(t)\boldsymbol{E}(t)\boldsymbol{Q}(t) \quad \text{with } \frac{\mathrm{d}\boldsymbol{Q}}{\mathrm{d}t} = \boldsymbol{W}\boldsymbol{Q},\ \ \boldsymbol{Q}(0) = \mathbf{1}. \tag{11}$$

Thus, all flows with nominally constant velocity gradient are equivalent to oscillatory straining, as indicated explicitly for simple shear in (20)–(21) below.

It is clear from (10) that $\mathscr{C}_z(t)$ and all derived continuum properties should be determined by the history of $\boldsymbol{H}(t')$ on $(0, t)$:

$$\mathfrak{h}_z = \overset{t}{\underset{0}{\boldsymbol{H}(t')}}, \tag{12}$$

together with an initial condition $\mathscr{C}_z(0)$. Given the nature of the underlying Stokes equations, we expect the ODEs (10) to exhibit continuous and differentiable dependence on $\mathscr{C}_z(t)$, such that solutions $\mathscr{C}_z(t)$ exhibit no singular dependence on isolated values of $\boldsymbol{H}$ or on other sets of zero measure. The issue is crucial to the response of stress to discontinuities in $\boldsymbol{H}$, as in certain flow reversal experiments (Gadala-Maria & Acrivos 1980; Kolli *et al.* 2002) discussed below. In particular, and contrary to experiment, it follows from (2) that one should find strict linearity of stress $\boldsymbol{T}(t)$ in instantaneous strain rate $\boldsymbol{E}(t)$, with simultaneous reversal of the former upon abrupt reversal of the latter.

The possibility of chaos in (10) allows for loss of memory (Drazer *et al.* 2002) in the form of effaceable memory, with eventual independence from $\mathscr{C}_z(0)$.

Furthermore, the absence of characteristic time in the Stokes equations implies that the underlying dependence on history is time-scale invariant and hence rate-independent (Goddard 1982), a property already apparent from (9) and (10). Consequently, time can be replaced by a strain measure, with appropriate scaling of kinematic rates:

$$t \to \int_0^t |\boldsymbol{E}(t')|\,\mathrm{d}t', \quad \boldsymbol{E}(t) \to \frac{\boldsymbol{E}(t)}{|\boldsymbol{E}(t)|}, \quad \boldsymbol{W}(t) \to \frac{\boldsymbol{W}(t)}{|\boldsymbol{E}(t)|}, \quad \text{etc.}, \tag{13}$$

where again $|\ |$ denotes the trace norm. Thus, whenever shearing stops, 'time' stops, and the usual 'fading memory' due to thermally driven relaxation, becomes effaceable memory arising from shear-driven chaos.

Most of the preceding remarks for spheres carry over to suspensions of torque-free, non-spherical particles, with the ODEs for the additional configurational variables again indicating a direct dependence on (12).

3.1. *Fabric evolution*

The representation of kinematic history by means of (Boltzmann) memory integrals is widespread in modern continuum mechanics, and has been justified by an appeal to fading memory (Coleman & Noll 1961). In the case of Stokesian suspensions, the preceding considerations suggest a co-rotational integral (Goddard 1967; Bird *et al.* 1987) for $\boldsymbol{A}(\mathfrak{h}_z)$ (although other integral models, exhibiting instantaneous elastic response, are favoured in the current literature on viscoelastic fluids).

To explore the potential use we adopt, as the leading term of a more general functional expansion (Goddard 1967), the deviatoric part of

$$\boldsymbol{A}(t) = -\int_0^t \dot{\psi}(t-t')[\boldsymbol{H}_t(t') + \chi \boldsymbol{H}_t^2(t')]\,\mathrm{d}t' + \boldsymbol{A}_0\psi(t), \tag{14}$$

where $\boldsymbol{A}_0$ denotes the initial fabric, $\psi(t)$ a memory function with $\psi(0) = 1$,

$$\boldsymbol{H}_t(t') = \boldsymbol{Q}_t^T(t')\boldsymbol{E}(t')\boldsymbol{Q}_t(t') \quad \text{with } \boldsymbol{Q}_t(t') = \boldsymbol{Q}(t')\boldsymbol{Q}^{-1}(t), \tag{15}$$

is the relative co-rotational strain rate, and $\boldsymbol{Q}(t)$ is given by (11).

The quadratic term in (14) is required to achieve a reasonably general response in simple shear and simple extension, and its scalar coefficient χ depends on the scalar invariants of $\boldsymbol{H}_t(t')$. However, since $\mathrm{tr}(\boldsymbol{H}_t(t'))=0$, and $\mathrm{tr}(\boldsymbol{H}_t^2(t'))=1$ by (13), the sole remaining invariant can be chosen as $\det(\boldsymbol{H}_t(t'))$. In the simple shear considered below, the latter vanishes, and χ reduces simply to a constant. Most of the algebraic detail and the consequences for viscometric functions can be found in Goddard (1967).

Based on the assumption of fading or effaceable memory, we take

$$\psi(t) \rightarrow 0 \quad \text{for } t \rightarrow \infty,$$

and replace (14)by

$$\boldsymbol{A}(t) = -\int_0^\infty \dot{\psi}(s)\big[\boldsymbol{H}_t(t-s) + \chi \boldsymbol{H}_t^2(t-s)\big]\, \mathrm{d}s, \tag{16}$$

where s is past time-lapse. Then, for $t > 0$, the portion of the integral over $0 < s < \infty$ serves to identify $\boldsymbol{A}_0$ in (14) with remnant anisotropy.

For purposes of illustration, we further employ a standard representation in terms of discrete exponentially relaxing modes (Bird *et al.* 1987; Larson 1999):

$$\psi(t) = \sum_{k=1}^{M} \hat{\psi}_k \mathrm{e}^{-t/t_k} \quad \text{with} \sum_{k=1}^{M} \hat{\psi}_k = 1, \tag{17}$$

with modal amplitudes $\hat{\psi}_k$ and relaxation strains $t_k > 0$.

It can be shown (Goddard 1967 and references therein) that (17) represents a linear Mth-order ODE for $\boldsymbol{A}$ and, hence, a generalized linear version of the equation of Hand (1962) in which the constants t_k represent ‘relaxation strains’. We recall that the description of structural evolution by means of ODEs is favoured in much of the literature on viscoelastic fluid mechanics, in part because they are adapted to contemporary numerical and analytical methods and also because they arise from simplified micro-mechanical analyses such as that of Phan-Thien (1995). In particular, his fabric-evolution equation can be written in the present notation as a first-order nonlinear ODE:

$$\lambda\left[\mathscr{D}_t\boldsymbol{A} - \boldsymbol{AE} - \boldsymbol{EA} - \tfrac{2}{3}\boldsymbol{E} + \tfrac{2}{3}\mathrm{tr}(\boldsymbol{AE})\mathbf{1} - \frac{2}{\eta_1}\boldsymbol{T}'(\boldsymbol{A},\boldsymbol{E})\right] + \boldsymbol{A} = 0, \tag{18}$$

where $\lambda \propto |\boldsymbol{E}|^{-1}$, and $\boldsymbol{T}'(\boldsymbol{A},\boldsymbol{E})$ is the deviatoric stress given by the modified form of (8) discussed above in the final remarks of § 2. Hence, up to an additive non-Stokesian term $|\boldsymbol{E}|\boldsymbol{A}$, (18) has the same form as the evolution equation of Barthès-Biesel & Acrivos (1973).

4. Unsteady simple shear

For the standard matrix representation of unsteady simple shear, with shear rate $\kappa(t)$:

$$\{\boldsymbol{E},\boldsymbol{W}\} = \kappa(t)\{\boldsymbol{E}_0,\boldsymbol{W}_0\} \quad \text{where} \left.\begin{matrix}\boldsymbol{E}_0\\ \boldsymbol{W}_0\end{matrix}\right\} = \frac{1}{2}\begin{bmatrix} 0 & 1 & 0\\ \pm 1 & 0 & 0\\ 0 & 0 & 0\end{bmatrix}, \tag{19}$$

the co-rotational strain rate (15) becomes

$$\begin{aligned}\boldsymbol{H}_t(t') &= \exp\{-\gamma_t(t')\boldsymbol{W}_0\}\,\boldsymbol{E}_0\exp\{\gamma_t(t')\boldsymbol{W}_0\}\\ &= \cos\gamma_t(t')\boldsymbol{E}_0 + \sin\gamma_t(t')[\boldsymbol{E}_0\boldsymbol{W}_0 - \boldsymbol{W}_0\boldsymbol{E}_0],\end{aligned} \tag{20}$$

where

$$\gamma = \int_0^t \kappa(t')\,\mathrm{d}t', \;\; \gamma_t(t') = \gamma(t') - \gamma(t), \quad \boldsymbol{H}_t^2(t') \equiv \boldsymbol{E}_0^2, \tag{21}$$

with γ representing shear strain.

By standard symmetry arguments for stress or optical-index tensors in viscometric flows (Truesdell & Noll 1965; Coleman & Dill 1971), it can be shown that $\boldsymbol{A}(t)$ must assume the general form

$$\boldsymbol{A} = \begin{bmatrix} \lambda(t)+\beta(t) & \alpha(t) & 0 \\ \alpha(t) & \lambda(t)-\beta(t) & 0 \\ 0 & 0 & -2\lambda(t) \end{bmatrix}, \tag{22}$$

provided the contribution from initial or remnant anisotropy $\boldsymbol{A}_0$ has the same form. Here, α, β, λ are functionals depending on the history of $\kappa(t)$. whose description can be simplified as follows for rate-independent materials.

For the purpose of the calculation to follow and to conform to the usual definition of shear strain, the strain measure t is defined in terms of actual time t and shear rate κ by replacing $|\boldsymbol{E}|$ everywhere by $\sqrt{2}|\boldsymbol{E}|$ in (13), in order to agree with the definition employed in experiments cited below. Thus, with the kinematics (19), the substitutions

$$t \to \int_0^t |\kappa(t')|\,\mathrm{d}t', \;\; \kappa(t) \to \mathrm{sgn}[\kappa(t)], \tag{23}$$

now represent the time–strain transformation (13). Thus, for rate-independent materials, the unsteady velocity gradient can be replaced by the binary (± 1) 'telegraph signal' of κ *vs.* strain t, with (14) representing a 'filter' (cf. Jakeman & Ridley 1999). This is illustrated by the following examples.

Oscillatory shear

For a sustained periodic simple shear at circular frequency ω,

$$\kappa(t) = \kappa_0 \sin \omega t, \tag{24}$$

where t is actual time, it is easy to show that the transformation (23) gives, as a particularly simple example of the telegraph signal mentioned above, the unit $2t_0$-periodic square wave (Rademacher function):

$$\mathrm{sgn}(\kappa) = S\left(\frac{t}{t_0}\right) := \mathrm{sgn}\left\{\sin\left(\frac{\pi t}{t_0}\right)\right\} \text{ with } t_0 = \frac{2\kappa_0}{\omega}, \tag{25}$$

where t now denotes the strain measure (23). The actual shear strain, the time integral of (24), is therefore given in terms of the strain measure t as the $2t_0$-periodic triangle wave:

$$\gamma(t) = \int_0^t \mathrm{sgn}(\kappa)\,\mathrm{d}t = \frac{t_0}{2}\left[1 + (-1)^{\lfloor t/t_0 \rfloor}\left(2\left\{\frac{t}{t_0} - \left\lfloor\frac{t}{t_0}\right\rfloor\right\} - 1\right)\right], \tag{26}$$

where brackets $\lfloor\ \rfloor$ denotes the 'floor' function (the largest integer less than the argument). Substitution into (20)–(21) gives an explicit expression for the integrand of (16), which leads readily to the functions in (22), with $\alpha(t), \beta(t)$ given by $2t_0$-periodic linear combinations of $\cos\gamma(t)$, $\sin\gamma(t)$, and with $\lambda(t) \equiv \chi/12$, constant. The rather straightforward details are not recorded here since, apart from the extremely limited data of Gadala-Maria & Acrivos (1980) and Singh & Nott (2003), I know of no oscillatory-shear experiments that would allow for comparison with the above calculations. Instead, we turn to the case of transient shear.

4.1. Start-up of steady shear

By means of (20)–(21), with $\gamma \equiv t$, we readily obtain analytic expressions for the functions in (22) from (14) and (17), which reduce after considerable algebra to:

$$\left.\begin{aligned} \alpha(t) &= s\sum_{k=1}^{m}\hat{\psi}_k\cos\varphi_k\left[\cos\varphi_k - \mathrm{e}^{-t/t_k}\cos(t+\varphi_k)\right] + \alpha_0\sum_{k=1}^{m}\hat{\psi}_k\mathrm{e}^{-t/t_k},\\ \beta(t) &= \sum_{k=1}^{m}\hat{\psi}_k\cos\varphi_k\left[\sin\varphi_k - \mathrm{e}^{-t/t_k}\sin(t+\varphi_k)\right] + \beta_0\sum_{k=1}^{m}\hat{\psi}_k\mathrm{e}^{-t/t_k},\\ &\text{where } s = \mathrm{sgn}[\kappa(t)] \text{ and } \varphi_k = \tan^{-1}\left[t_k\big/\left(1+t_k^2\right)^{1/2}\right], \end{aligned}\right\} \quad (27)$$

with $\lambda(t) = \chi/12$, constant. These relations also define the steady-state values α_∞, β_∞. From standard symmetry arguments it follows that reversal at $t = 0$ of a previous steady-state shear corresponds to the initial condition:

$$\alpha_0 = -\alpha_\infty,\ \ \beta_0 = \beta_\infty,\ \ \lambda_0 = \lambda_\infty, \quad (28)$$

as suggested by (16) and (20). Substitution of (27) into (8), followed by elementary matrix algebra, yields explicit analytic forms for the viscosity $\eta(t)$ and the primary and secondary normal-stress differences $N_1(t)$, $N_2(t)$, including their steady-state values η_∞, $N_{1\infty}$, $N_{2\infty}$. These can be reduced to the more concise form:

$$\eta(t)/\eta_\infty = 1 + a[\alpha^2(t) - \alpha_\infty^2] + b[\beta^2(t) - \beta_\infty^2], \quad (29)$$

$$N_1(t)/N_{1\infty} = \frac{\alpha(t)\beta(t)}{\alpha_\infty\beta_\infty}, \quad (30)$$

$$\begin{aligned} N_2(t)/N_{2\infty} = 1 &+ c[\alpha(t) - \alpha_\infty] + f[\alpha(t)\beta(t) - \alpha_\infty\beta_\infty]\\ &+ g[\alpha(t)\beta^2(t) - \alpha_\infty\beta_\infty^2] + h[\alpha^3(t) - \alpha_\infty^3], \end{aligned} \quad (31)$$

involving three steady values η_∞, $N_{1\infty}$, $N_{2\infty}$, six coefficients $a, b, \ldots, h$ and functions $\alpha(t)$, $\beta(t)$ given by (8) and (14) in terms of $\psi(t)$ and ξ and the eight more fundamental coefficients in (8). The latter are not uniquely determined by $a, b, \ldots, h$, and we do not record the relations between them here. Positive dissipation is now guaranteed by $\eta(t) > 0$, which provides restrictions on the parameters in (29).

For simple shear, the relations (8), (14) and (17) introduce a total of $2M + 8$ adjustable parameters that are mapped onto the nine parameters in (29)–(31). This is illustrated by the following example, employing $M = 2$ exponential modes.

Comparison to experiments on shear reversal

Gadala-Maria & Acrivos (1980) and Kolli *et al.* (2002) report on the results of shear experiments involving abrupt reversal of steady shear. Both works allow for measurement of viscosity, whereas the second also involves the measurement of normal thrust in an annular ‘split-ring’ torsional-shear device, with ambient pressure maintained at the inner and outer free surfaces of the sample. Despite a possible reservation as to compatibility of these boundary conditions with the radial force balance, and setting aside a general concern as to the effect of inhomogeneous torsional shear in unsteady flow (V. Kolli & F. Gadala-Maria 2005, personal communication), we adopt the standard formula for steady torsional shear between parallel disks (Bird *et al.* 1987; Kolli & Gadala-Maria 2005, personal communication). This gives the ratio N of transient to steady normal thrust as

$$N(t) = \frac{N_1(t) - N_2(t)}{N_{1\infty} - N_{2\infty}} \equiv \frac{N_{1R} - R_\infty N_{2R}}{1 - R_\infty}, \quad (32)$$

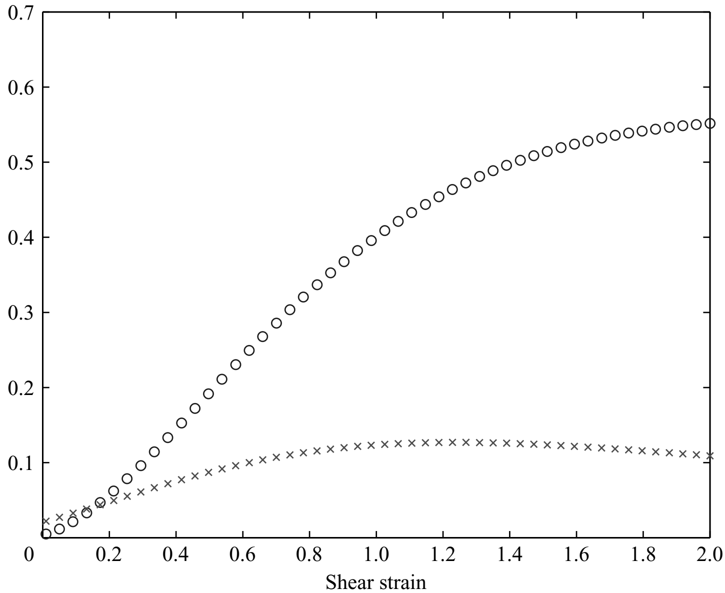

FIGURE 1. Structure functions ○, $\alpha(t)$; ×, $\beta(t)$ estimated from the data of Kolli *et al.* (2002) for $\phi = 0.4$, $\kappa_\infty = 0.5\,\mathrm{s}^{-1}$.

where

$$N_{kR} = N_k(t)/N_{k\infty}, \quad k = 1, 2, \quad R_\infty = N_{2\infty}/N_{1\infty}. \tag{33}$$

Also, since there was a slight departure from strict linearity of stress with shear rate, possibly due to instrument artefacts, we restrict our attention here to the experiments with steady shear rate $\kappa_\infty = 0.5\,\mathrm{s}^{-1}$.

All of the above experiments indicate a pronounced departure from complete reversal of stress immediately following reversal of shear, which the present model can describe only by means of rapidly decaying exponential modes with extremely short relaxation strains. To illustrate this, we consider a simple two-mode exponential decay in (17), with $t_2 \ll t_1 = 0(1)$.

Figure 1 presents plots of the functions α, β in (22), and figure 2 shows the computed values of the normalized viscometric functions (29)–(31) for the following parameter values:

$$\left.\begin{array}{l} R_\infty = 4.9219, \, \hat{\psi}_1 = 0.5384, \, t_1 = 0.9407, \, t_2 = 0.0001, \, a = 1.0727, \\ b = -4.6557, \, c = 1.7024, \, f = -2.6318, \, g = -0.9751, \, h = 1.3578, \end{array}\right\} \tag{34}$$

obtained by a standard nonlinear least-squares technique (implemented by the Matlab® program 'LSQNONLIN'), to fit the experimental data of Kolli *et al.* (2002) at particle volume fraction $\phi = 0.4$ and $\kappa_\infty = 0.5\,\mathrm{s}^{-1}$. These data are shown in figures 5 and 6 of Kolli *et al.* (2002) and have been provided in a numerical format (Kolli & Gadla–Maria 2005, personal communication).

The value of R_∞ is within the range of those estimated by Zarraga *et al.* (2000) from their steady-shearexperiments, while any value of $t_2 \lesssim 0.001$ is sufficient to describe the initial steep relaxation. It can be seen from figure 2 that the values (34) result in a significant transient overshoot of N_1.

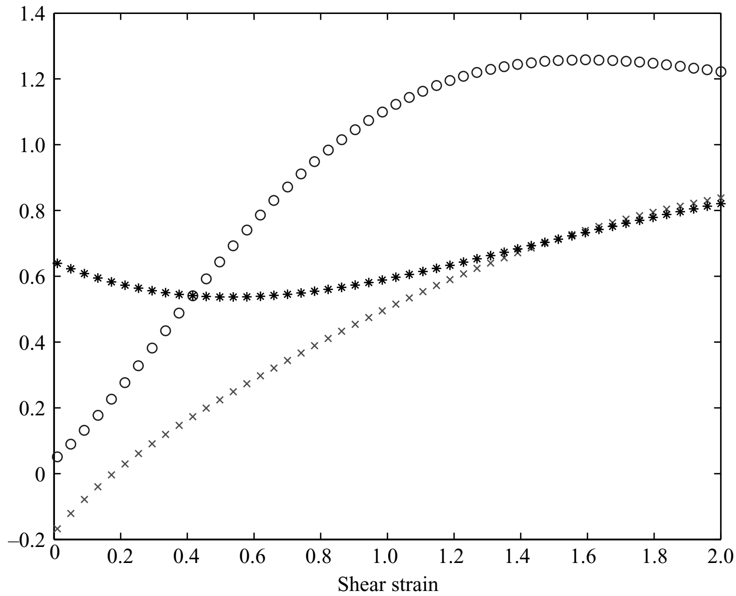

FIGURE 2. Normalized viscometric functions for reversal of steady shear estimated from the data of Kolli *et al.* (2002) for $\phi = 0.4$, $\kappa_\infty = 0.5\,\mathrm{s}^{-1}$. ○, N_1; ×, N_2; *, viscosity.

The same procedure was employed for the data of Kolli *et al.* (2002) at $\phi = 0.5$ and $\kappa_\infty = 0.5\,\mathrm{s}^{-1}$, with the resulting parameter values:

$$\left.\begin{array}{r} R_\infty = 10.96,\ \hat{\psi}_1 = 0.5733,\ t_1 = 0.7798,\ t_2 = 0.0001,\ a = 0.7798, \\ b = -2.8143,\ c = 0.9340,\ f = -1.9017,\ g = 2.4341,\ h = 1.1652, \end{array}\right\} \tag{35}$$

with figures 4–6 replacing figures 1–3, respectively.

Again, any value of $t_2 \lesssim 0.001$ is sufficient to describe the initial steep relaxation. However, the value of R_∞ now lies outside the range estimated by Zarraga *et al.* (2000), leading us to wonder about the strict validity of relation (32) for the apparatus of Kolli *et al.* (2002). In any event, it can be noted that the structure functions and normalized viscometric functions are not radically different for $\phi = 0.4$ and $\phi = 0.5$, as seen by a comparison of figures 1 and 2 with figures 4 and 5. This may suggest a potentially useful scaling of the transient structure and viscometric functions with their steady-state values.

While certain improvements to the above fit could be achieved by employing more exponential modes in (17), this would scarcely be justified by the existing experimental data. This suggests that other forms of unsteady shear may be called for, including more detailed versions of the oscillatory-shear studies of Gadala-Maria & Acrivos (1980) and Singh & Nott (2003).

As things stand, the values t_1, t_2 in (34) and (35) beg for a theoretical rationale. In particular, the strain t_2 is orders of magnitude smaller than the value $\kappa a^2/D \approx 10^2 - 10^3$ that one would infer from various experimental and theoretical estimates of hydrodynamic diffusivities D (see e.g. Drazer *et al.* 2002, figures 14–16, with $\phi = 0.4$). It is perhaps more plausible to attribute the small value of t_2 to non-hydrodynamic effects, for example, by identifying it with the small parameter $\epsilon \approx 0.0001 - 0.0004$ appearing in the interparticle force potential,

$$\varphi(r) = \ln(1 - \exp(-(r-2)/\epsilon)), \tag{36}$$

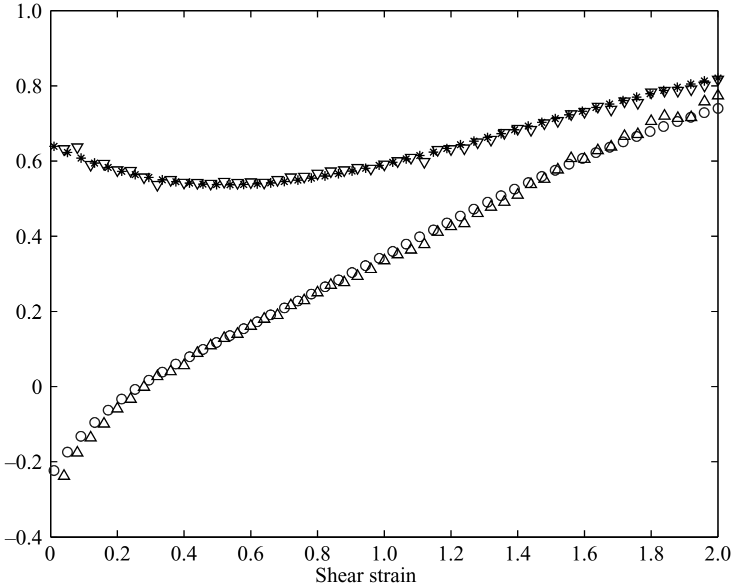

FIGURE 3. Relative torque (inverted triangles) and normal force (triangles) from of Kolli *et al.* (2002) for $\phi = 0.4$, $\kappa_\infty = 0.5\,\mathrm{s}^{-1}$ compared to the model with parameter values in (34). ○, N; *, viscosity.

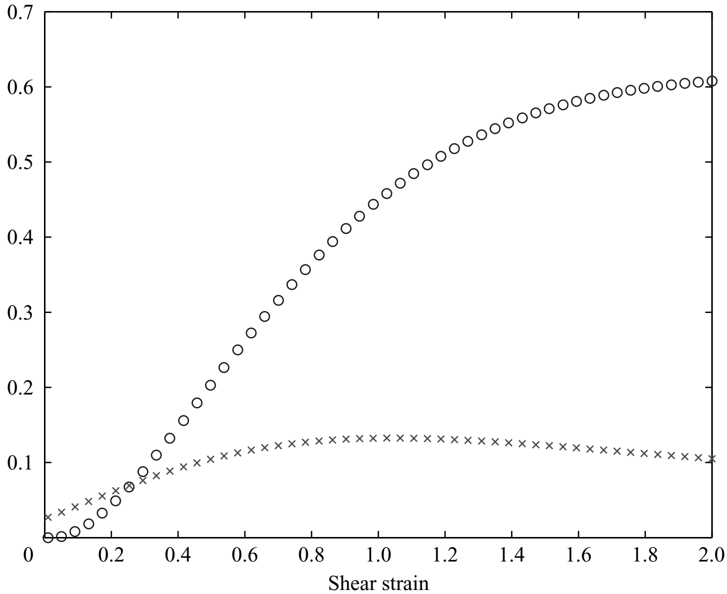

FIGURE 4. Structure functions ○, $\alpha(t)$; $\beta(t)$, for the parameter values estimated from the data of Kolli *et al.* (2002) for $\phi = 0.5$, $\kappa_\infty = 0.5\,\mathrm{s}^{-1}$.

employed in numerous simulations (Bossis & Brady 1984; Sierou & Brady 2002; Drazer *et al.* 2002) to break Stokesian symmetry for pairs of spheres in near contact. Thus, with $(r - 2)$ representing the non-dimensional sphere separation along direction

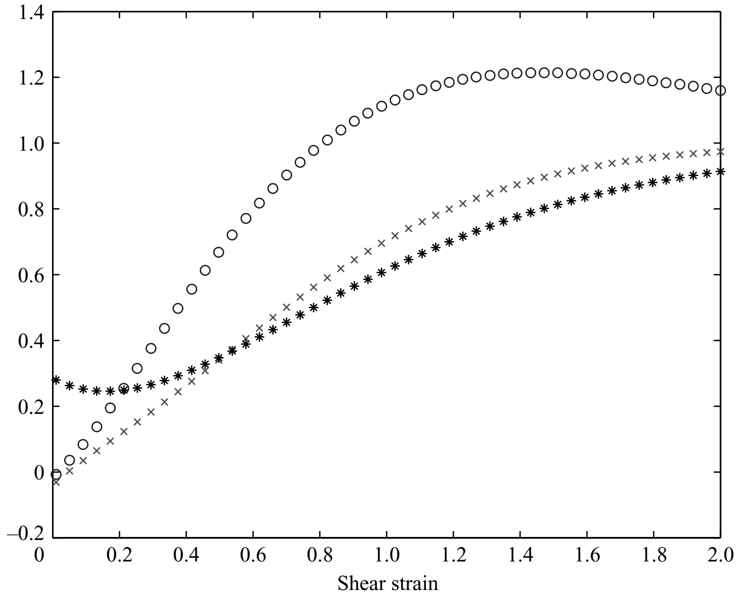

FIGURE 5. Normalized viscometric functions for reversal of steady shear, estimated from the data of Kolli *et al.* (2002) for $\phi = 0.5, \kappa_\infty = 0.5\,\text{s}^{-1}$. ○, N_1; ×, N_2; *, viscocity.

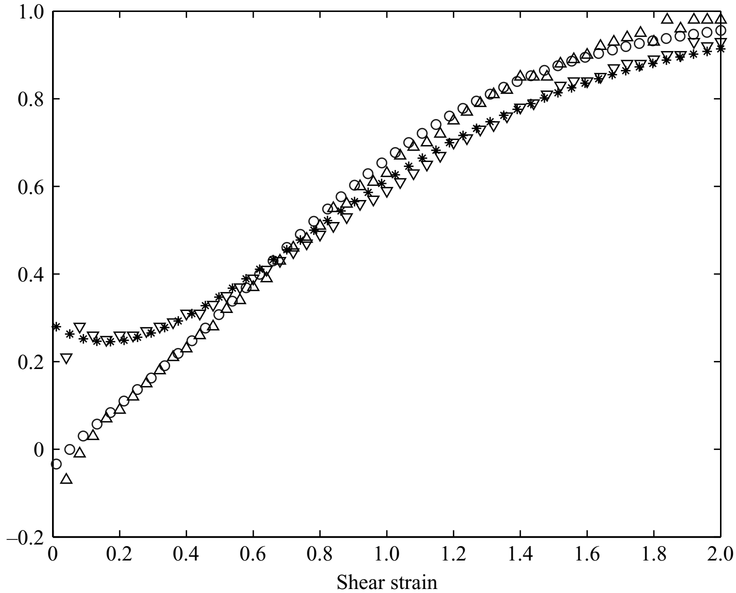

FIGURE 6. Relative torque (inverted triangles) and normal force (triangles) from Kolli *et al.* (2002) for $\phi = 0.5, \kappa_\infty = 0.5\,\text{s}^{-1}$, compared to the present model with parameter values in (35). ○; N; *, viscocity.

$\boldsymbol{n}$, the parameter ϵ represents the characteristic strain $\boldsymbol{n} \cdot \boldsymbol{E} \boldsymbol{n}$ associated with making or breaking contact.

Other possibilities for symmetry breaking may arise from small elastic effects in near contact, such as elastohydrodynamic deformation of particles, or viscoelastic response of an otherwise Newtonian suspending fluid. In any event, the associated small strain or time parameter can be regarded as a singular perturbation to Stokesian hydrodynamics, particularly in the description of evolution by ODEs.

By contrast, the value $t_1 = O(1)$ in (34) is suggestive of a large-scale hydrodynamic re-orientation of suspension microstructure. However, without investigating alternative representations to (14), we cannot rule out artefacts due to this assumed form of history dependence.

Regarding the last point, we recall that the evolution equation (18) of Phan-Thien (1995), which employs a much smaller number of adjustable parameters, predicts viscometric functions that are qualitatively similar to those shown in figures 2 and 4. The incomplete stress recovery in shear reversal is due to broken Stokesian symmetry arising from the term $|\boldsymbol{E}|\boldsymbol{A}$ discussed above. Given its relative simplicity, a slightly modified version of Phan-Thien's model could prove a useful adjunct or alternative to the general phenomenological approach employed here.

5. Granular plasticity

The dissipative model (2) is also applicable to the quasi-static mechanics of assemblies of non-cohesive rigid grains (Goddard 1984), for which the 'effective' granular stress, over and above the pressure of any interstitial ('pore') fluid, is given by (2), with

$$\boldsymbol{\eta} = p_s|\boldsymbol{E}|^{-1}\boldsymbol{\mu}\{\mathfrak{h}\} \tag{37}$$

where $\boldsymbol{\mu}$ is a non-dimensional fourth-rank (Mohr–Coulomb) friction tensor and $p_s > 0$ is the confining pressure on the solid granular phase. (Replacing p_s by a constant with units of stress gives a standard pressure-independent model of rigid plasticity.) With the interpretation of $\boldsymbol{T}$ as deviatoric stress, inversion of (37) formally yields a history-dependent conical yield surface (Goddard (1984, 1998); Didwania *et al.* (2001)), defined by:

$$\boldsymbol{T} : \boldsymbol{\mu}^{-2} : \boldsymbol{T} = p_s^2 \tag{38}$$

where $\boldsymbol{\mu}^{-2}$ denotes the square of the linear transformation $\boldsymbol{\mu}^{-1}$.

Granular media generally are subject to a dilatancy constraint (Goddard 1998; Didwania *et al.* 2001), which can be represented by specifying the dependence of $\mathrm{tr}(\boldsymbol{E})/|\boldsymbol{E}|$ on $\mathfrak{h}$. We recall that dilatancy vanishes at the so-called 'critical state' and also in the fully saturated state, which are contiguous to the maximally dense shearable state of particle suspensions.

In the absence of colloidal forces, micro-inertia, or pore-fluid viscosity, we can rule out any material time scale, which once again allows for the rate-independent description of kinematics implied by (13), a hallmark of standard plasticity theory. However, in contrast to Stokesian suspensions, the mechanics of granular media are dominated by direct, pairwise mechanical contact between particles, usually modelled by a Mohr–Coulomb type of sliding friction. The latter corresponds, incidentally, to a well-known micro-mechanical analogue of (37), with $\boldsymbol{T}$, $\boldsymbol{E}$ and p_s replaced, respectively, by tangential force, sliding velocity and normal force at a contact. This type of contact force lacks Stokesian linearity, so that the analogues of (9) or (10) should generally involve additional dependence of $\boldsymbol{\xi}$ or $\boldsymbol{\zeta}$, respectively, on $\boldsymbol{E}$ or $\boldsymbol{H}$ (as indicated in Goddard 1984).

From the preceding considerations, it follows that the analogous simplification to that made above for suspensions must now allow for the dependence of $\boldsymbol{\mu}$ on $\boldsymbol{E}(t)$ as well as on the fabric $\boldsymbol{A}$. However, since fabric depends only on current particle configuration, its history dependence should involve no singular dependence on $\boldsymbol{E}(t)$ or other sets of zero measure. In any event, the current stress $\boldsymbol{T}(t)$ must be given by an isotropic function of both $\boldsymbol{E}(t)$ and $\boldsymbol{A}$, whose general form, nonlinear in $\boldsymbol{E}$ and given in equation (2) of Hand (1962) (cf. Barthès-Biesel & Acrivos 1973), involves nine scalar coefficients depending the joint isotropic scalar invariants of $\boldsymbol{E}$ and $\boldsymbol{A}$. Even after reductions based on isotropy and rate-independence, there remain significantly more coefficients than in (8).

Without the history dependence represented by $\boldsymbol{A}$, the preceding model reduces to a well-known isotropic Reiner–Rivlin form, a polynomial in $\boldsymbol{E}(t)$ with coefficients depending on its scalar invariants (Goddard 1996). A special linear version of this model has been proposed to describe certain dense granular flows (Jop, Forterre & Pouliquen 2005, equation (4.2)), with inclusion of rate effects based on a micro-inertial time scale previously identified in Goddard (1996). However, it may be optimistic to expect the Reiner–Rivlin form, let alone simpler versions, to describe complex three-dimensional deformations of granular yield, given the anisotropic model required for suspensions. Of course, the latter requires evolution equations for $\boldsymbol{A}$, and the co-rotational memory integral (14) or equivalent ODEs appear worthy of further investigation.

Since particle diffusion and memory loss may arise from stochastic, possibly chaotic particle mechanics, even in quasi-static flow (Phan-Thien 1995; Didwania *et al.* 2001; Radjai & Roux 2005), there is a question as to the validity of the exponential relaxation assumed in (17). If valid, then much of the present development for suspensions would carry over to granular media. Leaving these issues to future investigation, we close with a brief mention of rate effects.

5.1. *Viscoplasticity*

Beyond the quasi-static regime, we must contend with strain-rate effects and departures from the scaling implied by (13). In addition to the micro-inertia mentioned above, dense rapid granular flows may also involve rate effects arising from pore-fluid viscosity. Conversely, dense suspensions with ϕ near the limit of shearability may involve non-hydrodynamic particle-contact effects as well as micro-inertia. Consequently, it will probably be necessary to replace (37) by a more general viscoplastic 'Oldroyd–Bingham' model (Goddard 1984):

$$\boldsymbol{\eta} = p_s|\boldsymbol{E}|^{-1}\boldsymbol{\mu}^{(0)} + \boldsymbol{\mu}^{(1)}, \tag{39}$$

where $\boldsymbol{\mu}^{(0)}$ and $\boldsymbol{\mu}^{(1)}$ refer, respectively, to plastic and viscous contributions of the type discussed above.

The immediately obvious time scale p_s/μ_0 in (39), where μ_0 is a characteristic viscosity, represents one of the micromechanical effects envisaged above and may be implicated in experiments on wet granular media by Tsai & Gollub (2004) and Huang *et al.* (2005). Although p_s arises from enduring particle contact in those experiments, it might loosely be interpreted as a limiting form of the 'particle pressure' appearing in certain suspension theories (Brady & Morris 1997). At any rate, the relative importance of terms on the right hand-side of (39) is governed by the relative magnitudes of forces due to particle contact and to fluid viscosity and is no doubt involved in the transition from granular medium to dense suspension. There are numerous questions as to the importance of rate effects on the evolution of anisotropy and as to the possibility of representing the latter by a single fabric tensor.

6. Conclusions

We have investigated a simplified version of the purely dissipative continuum model for the viscoplasticity of particulate systems. In this version, the history-dependence of the fourth-rank viscosity tensor is represented by a second-rank fabric tensor, whose kinematically driven evolution is described as a co-rotational memory integral. An expansion up to terms of third order in anisotropy, together with a single-integral co-rotational model and a strain-based exponential relaxation, gives a close fit to experimental shear- and normal-stress recovery following reversal of steady simple shear (Kolli *et al.* 2002). At least one rapidly relaxing mode is necessary to represent the partial reversal of stress observed in such experiments. The rapidity of this relaxation is highly incommensurate with estimates based on hydrodynamic diffusivity, suggesting non-hydrodynamic effects of the type invoked in numerous past studies.

A cursory survey of the mechanics of non-cohesive granular media suggests the potential use of the plastic and viscoplastic versions of the above model for application to dry and liquid-saturated granular media as well as dense suspensions. The present work may serve as a guide to further micromechanical studies, e.g. such as that of Phan-Thien (1995), and to crucial experimental studies of transient and oscillatory shear, in order to sharpen the underlying constitutive theory.

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