Rheology of Suspensions of Flat Elastic Particles

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We consider a suspension of noninteracting flat elastic particles in a Newtonian fluid. We model a flat shape as three beads, carried along by the flow according to Stokes law, and connected by nonlinear springs, chosen such that the energy is quadratic in the area. In analogy with common dumbbell models involving two beads connected by linear springs, we solve the stochastic equations of motion exactly to compute the constitutive law for the stress tensor of a flat elastic particle suspension. A lower convected time derivative naturally arises as part of the constitutive law, but surprisingly the rheological response in strong extensional and strong contracting flows is similar to that of the classical Oldroyd-B model associated with dumbbell suspensions.

The presence of elastic particles (e.g., high molecular weight polymers), is known to profoundly change the flow properties of complex liquids [1–3], such as blood, saliva, and many other biological and manmade substances. For example, the extensional flow accompanying the breakup of a liquid drop leads to the formation of long and thin threads, caused by the stretching of molecules in the flow [4–6]. To model such behavior, the equations of fluid motion are usually augmented with an extra, "polymeric" or "particle" contribution σ^p to the stress tensor, found by solving an additional constitutive equation, which couples the evolution of σ^p to the flow [1,7].

The derivation of such an equation using a realistic molecular description of a polymer has not been achieved. Instead, most constitutive equations have their basis in phenomenological models consistent with the invariances of the system as well as with thermodynamics, allowing for adjustable parameters to account for specific properties of the system [8,9]. In particular, Oldroyd [10] pointed out that for the description to be independent of the choice of coordinate system (known as frame invariance), the ordinary convected time derivative of the stress tensor has to be augmented with extra terms, which can only take two distinct forms: the upper-convected and lower-convected derivatives (and linear superpositions thereof). However, without reference to a specific microscopic system, the phenomenological approach does not reveal which frame-invariant derivative to use, although the rheological responses can be very different.

To address this, highly simplified model systems (known as dumbbell models, seen on the left of Fig. 1) have become very popular. They consist of two spherical beads, connected by a (harmonic) spring; their state is thus defined by the vector $\ell = \mathbf{r}_1 - \mathbf{r}_0$ alone. If both beads are convected passively by the flow, the material time derivative $\dot{\ell}$ (which we denote by a dot) is $\mathbf{v}(\mathbf{r}_1) - \mathbf{v}(\mathbf{r}_0)$; assuming that the flow varies on scales much larger than the particle size, we can expand $\mathbf{v}$ into a Taylor series to find to lowest order:
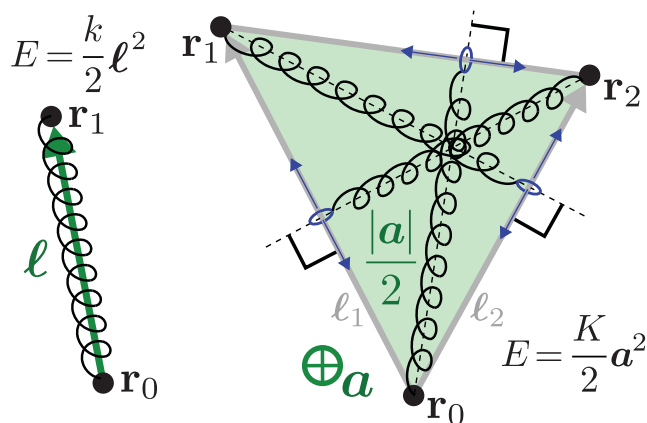
$$\dot{\ell} = \ell \cdot \nabla \mathbf{v}, \tag{1}$$

FIG. 1. Left (dumbbell model): Two beads, separated by a length vector ℓ, connected by a linear spring. Right (minimal flat elastic particle model): Three beads define a vector $\mathbf{a} = \ell_1 \times \ell_2$ normal to the plane of a triangle with surface area $|\mathbf{a}|/2$ that mimics a flat particle. An energy $E = K a^2/2$ is produced by three identical nonlinear springs that connect each bead along the triangle's heights to the bases as shown. Spring constants are proportional to the square of the corresponding base lengths. Thus, for triangle sides as labeled here, springs connected to beads at $\mathbf{r}_0$, $\mathbf{r}_1$, and $\mathbf{r}_2$ have spring constants $K|\ell_1 - \ell_2|^2$, $K|\ell_2|^2$, and $K|\ell_1|^2$, respectively.

with the strain rate tensor $(\nabla\mathbf{v})_{\alpha\beta} = \partial_\alpha v_\beta$ and $\alpha, \beta = x, y, z$. Using this together with Stokes drag law [11], and adding thermal noise, the thermal average of the polymeric stress yields the Oldroyd-B model [2], which has become an almost canonical description of a polymeric fluid [3], and which contains the upper-convected derivative only. However, depending on the geometry of the immersed particles, the lower-convected derivative should generically come in as well [12].

In order to improve our intuition, and to expand the lexicon of distinct rheological models with an explicit microscopic underpinning, we develop in this letter an exactly solvable particle model that only contains the lower-convected derivative as part of its constitutive equation. According to long-established intuition [13], and supported by a recent penetrating analysis of ellipsoidal particles [14], our model involves ideally flat geometries, that are deformed by the flow. As a convenient abstraction, we consider three nearby Stokes beads (see Fig. 1, right) and two vectors $\boldsymbol{\ell}_1 = \mathbf{r}_1 - \mathbf{r}_0$ and $\boldsymbol{\ell}_2 = \mathbf{r}_2 - \mathbf{r}_0$ that uniquely define an "area vector"

$$\boldsymbol{a} = \boldsymbol{\ell}_1 \times \boldsymbol{\ell}_2, \qquad (2)$$

which is normal to the plane of the beads, and whose modulus is twice the area of the triangle defined by the beads.

In this Letter, we proceed by first deriving an equation of motion for the area vector $\boldsymbol{a}$ [Eq. (4)], which represents the kinematics of a flat particle in flow. When deriving the equation of motion for a suitable fabric tensor that encodes thermally averaged states of such particles, this kinematics naturally leads to a lower convected derivative [Eq. (15)]. Finally, by deriving the stress contributions that are due to the flat particles' elasticity and expressing them in terms of the fabric tensor [Eq. (19)], we arrive at a closed set of constitutive equations for a dilute suspension of flat elastic particles.

If each of the vectors $\boldsymbol{\ell}_1$ and $\boldsymbol{\ell}_2$ evolves according to (1), we find that $\dot{\boldsymbol{a}} = \dot{\boldsymbol{\ell}}_1 \times \boldsymbol{\ell}_2 + \boldsymbol{\ell}_1 \times \dot{\boldsymbol{\ell}}_2 = \boldsymbol{\ell}_1 \cdot \nabla(\mathbf{v} \times \boldsymbol{\ell}_2) + \boldsymbol{\ell}_2 \cdot \nabla(\boldsymbol{\ell}_1 \times \mathbf{v})$. Using the vector identity [15]

$$\boldsymbol{\ell}_1 \cdot \nabla(\mathbf{v} \times \boldsymbol{\ell}_2) + \boldsymbol{\ell}_2 \cdot \nabla(\boldsymbol{\ell}_1 \times \mathbf{v}) = -(\nabla\mathbf{v}) \cdot (\boldsymbol{\ell}_1 \times \boldsymbol{\ell}_2), \qquad (3)$$

which holds for $\nabla \cdot \mathbf{v} = 0$, it follows that in an incompressible flow the area vector obeys

$$\dot{\boldsymbol{a}} = -\boldsymbol{a} \cdot (\nabla\mathbf{v})^\top, \qquad (4)$$

instead of (1) for a length vector, as expected from the dynamics of a surface area element [16].

We now calculate the contribution $\boldsymbol{\sigma}^{\mathrm{p}}$ of a flat elastic particle suspension to the stress tensor, which is usually done in two steps [2,17]: first, one derives the equation of

motion for a "fabric tensor," which characterizes the state of the suspended particle. Second, the stress is calculated from the fabric tensor by a constitutive relation. As shown in [14], the equation of motion for a fabric tensor $\mathbf{L} = c_\ell \langle \boldsymbol{\ell} \otimes \boldsymbol{\ell} \rangle$, appropriate for a linear dumbbell (where $\langle \cdot \rangle$ denotes a thermal average), automatically involves upper-convected derivative. On the other hand, in deriving the equation of motion for a fabric tensor $\mathbf{A} = c_a \langle \boldsymbol{a} \otimes \boldsymbol{a} \rangle$ (constants c_ℓ and c_a serve to make fabric tensor dimensionless), based on our flat particle model (see Fig. 1, right), we now show that the upper-convected derivative is replaced by the lower-convected derivative.

According to Stokes law [11], the motion of a bead $\mathbf{r}_i$ relative to the fluid equals the force $\mathbf{F}_i$, divided by the drag coefficient $\zeta = 6\pi\eta r_b$, where η is the fluid viscosity, and r_b the radius of a bead:

$$\zeta(\dot{\mathbf{r}}_i - \mathbf{v}(\mathbf{r}_i)) = \mathbf{F}_i \equiv -\partial_{\mathbf{r}_i} E(\boldsymbol{a}), \qquad i = 0, 1, 2. \quad (5)$$

For simplicity, we focus on the deterministic part of the equation, and add thermal noise later. From now on, we assume a quadratic energy $E(\boldsymbol{\ell}_1, \boldsymbol{\ell}_2) = K\boldsymbol{a}^2/2$. We present a possible mechanical realization of this energy below (see Fig. 1, right). Because of the translational invariance of $E(\boldsymbol{\ell}_1, \boldsymbol{\ell}_2)$, the particle dynamics separates into the motion of its center of mass $\mathbf{r}_c = (\mathbf{r}_0 + \mathbf{r}_1 + \mathbf{r}_2)/3$ (which is immaterial), and equations of motion for $\boldsymbol{\ell}_i$, where $\mathbf{r}_0 = \mathbf{r}_c - (\boldsymbol{\ell}_1 + \boldsymbol{\ell}_2)/3$, $\mathbf{r}_1 = \mathbf{r}_c + (2\boldsymbol{\ell}_1 - \boldsymbol{\ell}_2)/3$ and $\mathbf{r}_2 = \mathbf{r}_c + (2\boldsymbol{\ell}_2 - \boldsymbol{\ell}_1)/3$. Thus transforming to $\boldsymbol{\ell}_i$ derivatives and expanding $\mathbf{v}(\mathbf{r}_i) - \mathbf{v}(\mathbf{r}_0) \approx (\mathbf{r}_i - \mathbf{r}_0) \cdot \nabla\mathbf{v}$ as before, one finds

$$\dot{\boldsymbol{\ell}}_1 = \boldsymbol{\ell}_1 \cdot \nabla\mathbf{v} - \frac{1}{\zeta}\left(\frac{\partial E}{\partial \boldsymbol{\ell}_2} + 2\frac{\partial E}{\partial \boldsymbol{\ell}_1}\right), \qquad (6)$$

and similarly for $\boldsymbol{\ell}_2$ via a permutation $1 \leftrightarrow 2$. Equation (6) is a generalization of (1), describing the slip of beads relative to the flow, as a result of interbead forces.

Using that

$$E = K(\boldsymbol{\ell}_1^2\boldsymbol{\ell}_2^2 - (\boldsymbol{\ell}_1 \cdot \boldsymbol{\ell}_2)^2)/2, \qquad (7)$$

one finds

$$\frac{\partial E}{\partial \boldsymbol{\ell}_1} = K(\boldsymbol{\ell}_1\boldsymbol{\ell}_2^2 - \boldsymbol{\ell}_2\boldsymbol{\ell}_1 \cdot \boldsymbol{\ell}_2) \qquad (8)$$

(and correspondingly for $\boldsymbol{\ell}_2$), so that the last term in (6) reads

$$\frac{\partial E}{\partial \boldsymbol{\ell}_2} + 2\frac{\partial E}{\partial \boldsymbol{\ell}_1} = \boldsymbol{\ell}_1(2\boldsymbol{\ell}_2^2 - \boldsymbol{\ell}_1 \cdot \boldsymbol{\ell}_2) + \boldsymbol{\ell}_2(\boldsymbol{\ell}_1^2 - 2\boldsymbol{\ell}_1 \cdot \boldsymbol{\ell}_2). \qquad (9)$$

Then from the product rule $\dot{\boldsymbol{a}} = \dot{\boldsymbol{\ell}}_1 \times \boldsymbol{\ell}_2 + \boldsymbol{\ell}_1 \times \dot{\boldsymbol{\ell}}_2$, and repeating the calculation leading to (4), we arrive at

$$\dot{\boldsymbol{a}} = -\boldsymbol{a} \cdot (\nabla\mathbf{v})^\top - \frac{9KR^2}{\zeta}\boldsymbol{a}. \qquad (10)$$

Here, $R^2 = 2[\ell_1^2 + \ell_2^2 - (\ell_1 \cdot \ell_2)]/9$ denotes the mean-squared distance of the three beads to the center of mass $\mathbf{r}_c$ and is thus a measure of the flat particle's size [15]. In order to make (10) amenable to a statistical analysis in analogy to the classical dumbbell model [2], we assume a size-dependent bead drag $\zeta = 9R^2\zeta_a$ (ζ_a a constant), which makes (10) linear in $\boldsymbol{a}$. This assumption is consistent with the expectation of how elastic membranes interact with surrounding shear flow [18], which is in itself a challenging problem that has been studied in various contexts [19–23]. While mechanistically, the assumed drag could, in principle, be realized using beads that adjust their sizes appropriately, it also illustrates crucial subtleties that are involved in the modeling of flat particles and their interaction with flow.

Adding thermal noise to the area vector dynamics [24], we arrive at

$$\zeta_a(\dot{\boldsymbol{a}} + \boldsymbol{a} \cdot (\nabla\mathbf{v})^\top) = -K\boldsymbol{a} + \boldsymbol{\xi}, \quad (11)$$

where $\boldsymbol{\xi}$ denotes fluctuations around equilibrium with $\langle\boldsymbol{\xi}\rangle = 0$ and $\langle\xi_\alpha(t)\xi_\beta(t')\rangle = 2k_BT\zeta_a\delta_{\alpha\beta}\delta(t-t')$. The corresponding Fokker-Planck equation for the probability distribution $P(\boldsymbol{a}, t)$ to find a particle with area vector $\boldsymbol{a}$ at time t then reads [24]

$$\frac{\partial P}{\partial t} + \frac{\partial}{\partial\boldsymbol{a}} \cdot \mathbf{J} = 0, \quad (12)$$

where the probability current is given by

$$\mathbf{J} = -\boldsymbol{a} \cdot (\nabla\mathbf{v})^\top P - \frac{K}{\zeta_a}\boldsymbol{a}P - \frac{k_BT}{\zeta_a}\partial_{\boldsymbol{a}}P. \quad (13)$$

The thermal average to compute the fabric tensor is given by $\mathbf{A} = c_a \int \boldsymbol{a} \otimes \boldsymbol{a}P(\boldsymbol{a}, t)d\boldsymbol{a}$, such that the dynamics of the fabric tensor can be determined from the material derivative

$$\frac{1}{c_a}\frac{dA_{\alpha\beta}}{dt} = \int a_\alpha a_\beta \partial_t P\, d\boldsymbol{a} = \int (\delta_{\alpha\gamma}a_\beta + \delta_{\beta\gamma}a_\alpha)J_\gamma\, d\boldsymbol{a}, \quad (14)$$

having used (12) and then integrated by parts. Inserting (13) into (14), the first two terms on the right of (13) are expressible directly in terms of $A_{\alpha\beta}$, the last term, integrating once more by parts, and using that P is normalized, yields a constant. Thus the equation of motion (14) for the fabric tensor, our first main result, becomes

$$\overset{\triangle}{\mathbf{A}} = -\frac{2K}{\zeta_a}\mathbf{A} + \frac{2c_ak_BT}{\zeta_a}\mathbb{I} \equiv -\frac{1}{\lambda}(\mathbf{A} - \mathbb{I}), \quad (15)$$

where

$$\overset{\triangle}{\mathbf{A}} \equiv \frac{\partial\mathbf{A}}{\partial t} + \mathbf{v} \cdot \nabla\mathbf{A} + (\nabla\mathbf{v}) \cdot \mathbf{A} + \mathbf{A} \cdot (\nabla\mathbf{v})^\top \quad (16)$$

is the lower-convected derivative, and $\mathbb{I}$ the unit tensor. Here $\lambda = \zeta_a/(2K)$ is a relaxation time of a suspended particle, and $c_a = K/k_BT$ has been chosen to make $\mathbf{A}$ relax toward $\mathbb{I}$ in equilibrium [17]. As anticipated, (15) is the exact analog of the equation for the fabric tensor $\mathbf{L}$ of a dumbbell, but written with the lower-convected derivative.

To compute the particle stress tensor $\boldsymbol{\sigma}^\mathrm{p} \equiv \boldsymbol{\sigma}^a$ coming from our arealike elastic particles in the dilute limit, we use the Kirkwood formula [25]

$$\boldsymbol{\sigma}^a = -\rho_0 \sum_{i=0}^{2} \langle \mathbf{r}_i \otimes \mathbf{F}_i \rangle = \rho_0 \sum_{i=1}^{2} \left\langle \ell_i \otimes \frac{\partial E}{\partial\ell_i} \right\rangle, \quad (17)$$

where ρ_0 is the number density of three-bead particles. In reducing to a sum over internal forces in the second step, we have used that the model flat particle is force-free, such that $\mathbf{F}_0 = \partial_{\ell_1}E + \partial_{\ell_2}E$, $\mathbf{F}_1 = -\partial_{\ell_1}E$, and $\mathbf{F}_2 = -\partial_{\ell_2}E$. With $E(\ell_1, \ell_2)$ as given in Eq. (7), the stress tensor becomes

$$\boldsymbol{\sigma}^a = \rho_0K\langle\ell_1 \otimes \ell_1\ell_2^2 + \ell_2 \otimes \ell_2\ell_1^2 - 2\ell_1 \otimes \ell_2\ell_1 \cdot \ell_2\rangle, \quad (18)$$

and using the Levi-Civita tensor product identity for $\epsilon_{\alpha\beta\gamma}\epsilon_{\delta\epsilon\kappa}$ on $\mathbf{A} = c_a\langle(\ell_1 \times \ell_2) \otimes (\ell_1 \times \ell_2)\rangle$ yields

$$\boldsymbol{\sigma}^a = \mu[\mathbb{I}\mathrm{Tr}(\mathbf{A}) - \mathbf{A}], \quad (19)$$

our second main result; here $\mu = \rho_0k_BT$ is an elastic constant. Crucially, $\boldsymbol{\sigma}^a$ is not simply proportional to the fabric tensor, as would be the case for Oldroyd models A or B, but contains an extra term involving the trace of $\mathbf{A}$. It is straightforward to show [15] that the total stress in Eq. (19) corresponds to a particular case of Oldroyd's 8-constant model [1].

Finally, to complement model equations (15) and (19), the velocity field satisfies the momentum balance

$$\rho(\partial_t\mathbf{v} + \mathbf{v} \cdot \nabla\mathbf{v}) = -\nabla p + \nabla \cdot \boldsymbol{\sigma}^a + \eta\triangle\mathbf{v}, \quad (20)$$

where ρ is the density, and η the viscosity of the solvent.

To illustrate the consequences of (19), let us calculate $\boldsymbol{\sigma}^a$ for the two-dimensional extensional flow

$$u = \dot{\epsilon}x, \qquad v = -\dot{\epsilon}y, \quad (21)$$

which is stretching at a rate $\dot{\epsilon}$ in the x direction, and compressing in the y direction. Then the dynamics of the corresponding diagonal components of $\mathbf{A}$ follow from Eq. (15) as

$$\dot{A}_{xx} = (-2\dot{\epsilon} - \lambda^{-1})A_{xx} + \lambda^{-1}, \quad (22)$$

$$\dot{A}_{yy} = (2\dot{\epsilon} - \lambda^{-1})A_{yy} + \lambda^{-1}, \quad (23)$$

while for the fabric tensor **L** of dumbbell particles, whose dynamics are written with an upper-convected derivative, the signs of strain rate contributions are reversed:

$$\dot{L}_{xx} = (2\dot{\epsilon} - \lambda^{-1})L_{xx} + \lambda^{-1}, \tag{24}$$

$$\dot{L}_{yy} = (-2\dot{\epsilon} - \lambda^{-1})L_{yy} + \lambda^{-1}. \tag{25}$$

Thus for $\dot{\epsilon} > \lambda^{-1}/2$, L_{xx} grows exponentially following the stretching of the flow, while the lower-convected derivative implies that A_{yy} grows, in the direction in which the flow is contracting [5].

In the Oldroyd-A model, the particle stress is given by $\boldsymbol{\sigma}^p = -\mu(\mathbf{A} - \mathbb{I})$, while for the Oldroyd-B model, $\boldsymbol{\sigma}^p = \mu(\mathbf{L} - \mathbb{I})$ (and stress from solvent viscosity is to be added). This means the Oldroyd-B model faithfully describes the buildup of stress in the spring of a dumbbell, as the two beads are convected by the flow (cf. Fig. 1, left). The Oldroyd-A model, on the other hand, describes the rather less intuitive situation of stress building up in a direction orthogonal to the stretching. This prediction of the Oldroyd-A model is, however, in stark contrast to our present finding for the behavior of idealized flat particles: Because the stress $\boldsymbol{\sigma}^a$ in Eq. (19) is not simply proportional to **A**, but contains an additional trace contribution, the stress in the *extensional* direction follows

$$\sigma_{xx}^a = \mu(\mathrm{Tr}(\mathbf{A}) - A_{xx}) \approx A_{yy} \propto A_{yy}^0 e^{(2\dot{\epsilon} - \lambda^{-1})t}, \tag{26}$$

and once more grows exponentially. Thus, surprisingly, the behavior of our flat elastic particle model is similar to that of a classical dumbbell model, even though the fabric tensor dynamics is given in terms of a lower-convected derivative. The two models, however, remain distinct. In an axisymmetric extensional flow, as it arises during the pinch-off of a polymeric thread [4,5], the buildup of axial stress is weaker in our flat particle model then in the dumbbell model. As a result, no uniform thread is formed, as is observed during the pinching of solutions of elastic polymers [4,5,26]. On the other hand, the response of our model to a simple shear flow of rate $\dot{\gamma}$ is identical to that of the Oldroyd-A model: the first and second normal stress differences are $N_1 = -N_2 = 2\mu\lambda^2\dot{\gamma}^2$ [1].

In conclusion, we have constructed an exactly solvable microscopic model for a dilute suspension of flat elastic particles, whose coarse-grained dynamics is described by the lower-convected derivative. Nevertheless, the rheological properties of this suspension are remarkably similar to those of conventional linear "dumbbell" particles.

In fact, even though this is assumed widely [12], it is very difficult to realize a flat particle model that would give rise to the Oldroyd A model macroscopically, and in which stress builds up orthogonally to the direction of an extensional flow. The reason is that elastic forces in an overall force- and torque-free flat particle must be acting within the plane of that particle. This precludes the possibility of stresses being generated in the direction normal to the particle's surface, which would be required to achieve the Oldroyd A-like rheological properties described above.

A mechanical realization of the flat elastic particles introduced in this model is depicted in Fig. 1 (right): three identical nonlinear springs with vanishing rest length connect beads along the triangle's heights to each base and have spring constants proportional to the square of the corresponding base lengths. For example, the force $\mathbf{F}_1 = -\partial_{\boldsymbol{\ell}_1} E$ acting on the bead at $\mathbf{r}_1$ can, according to (8), be written as

$$\mathbf{F}_1 = -K\boldsymbol{\ell}_2^2 \mathbf{P}_2^\perp \cdot \boldsymbol{\ell}_1, \tag{27}$$

where $\mathbf{P}_2^\perp = \mathbb{I} - (\boldsymbol{\ell}_2 \otimes \boldsymbol{\ell}_2)/\boldsymbol{\ell}_2^2$ is a projection that extracts the component of $\boldsymbol{\ell}_1$ that is orthogonal to $\boldsymbol{\ell}_2$. Therefore, $\mathbf{F}_1$ is parallel to the height that originates at $\mathbf{r}_1$ and passes through the base $\boldsymbol{\ell}_2$. From (27), we can identify the spring constant $K\boldsymbol{\ell}_2^2$. Similar arguments apply for the elastic forces $\mathbf{F}_2$ and $\mathbf{F}_0 = -\mathbf{F}_1 - \mathbf{F}_2$ acting on the beads at $\mathbf{r}_2$ and $\mathbf{r}_0$, respectively. More general microscopic particle models can be considered, for example, by using both length and area vectors as geometric building blocks. Similarly, it would be interesting to study the nonlinear dynamics and many-body effects in such a family of minimal particle models in numerical simulations.

Finally, it would be interesting to explore the implications of our results for rheological measurements of red blood cell suspensions [27–29], flat objects whose mechanical properties have previously been interpreted using insights from Oldroyd-B models.

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