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LETTER TO THE EDITOR

Possible shear instabilities for colloidal structures in a solvent

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Abstract. A colloidal aggregate in a moving fluid may suffer an instability of the plug-flow type for a large enough shear. The resulting non-uniform structure should be allowed for in shear-melting models.

Plug flow in suspensions is an experimentally well documented effect (see, e.g., [1]) and has recently been re-approached on the theoretical side [2, 3]. In Poiseuille flow, for example, above some threshold in the pressure head, that is in the shear, a flat segment will show up in the velocity profile around the axis of the tube and the suspended particles will tend to gather in that region. This is a typical two-parameter instability: local particle density, affecting the viscosity, and local shear strain, which results in a 'lift force' [2] related to the shear gradient.

Here, we address a related problem which we cast in formally similar terms. We consider shear flows—Couette between parallel plates to be specific—but, instead of a suspension, the solute is a colloidal structure now, say of the branched type.

Such a structure exhibits some elasticity, extending to finite lengths [3]. Imagine the colloid to be reticulated onto a cubic lattice (figure 1 provides a coarse-grained version of the orientational map). Without flow, the proportion p of links lying *perpendicular* to the plates, along $0x$, is $p_0 = \frac{1}{3}$. Under shear flow, this probability will decrease, whereas the probability of finding a link along $0z$, the direction of flow, increases above $\frac{1}{3}$ (the problem is in fact two dimensional since the links // $0y$ are not affected). This provides us with a natural definition of elastic strain at level x (of

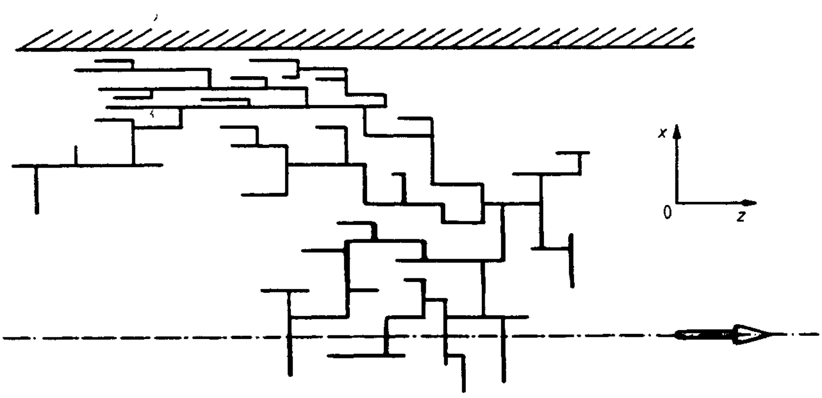

Figure 1. Schematic profile of the sheared structure above threshold. The arrow shows the direction of flow. The 'density' $p(x)$ decreases as one goes from the central symmetry plane (chain line) to the plate walls.

course, we take $x=0$ on the symmetry plane (statistical symmetry) between plates):

$$\varepsilon=\frac{p_0-p(x)}{p_0}=A\sigma \tag{1}$$

$$\sigma=\eta(p)\gamma=\eta(p)\,\partial v/\partial x \tag{2}$$

is the shear stress ($\eta(p)$ is the local viscosity, now depending on $p(x)$, $\gamma=\partial v/\partial x$, $v(x)$ is the velocity of flow along $0z$). We assume that the flow stress is entirely transferred to the colloid, where

$$A=\left(\frac{L}{b}\right)^{d}\frac{1}{\alpha\kappa n_a}\left(\frac{L}{a}\right)^{D_c} \tag{3}$$

is an elastic response factor, the expression of which (3) is due to Kantor and Witten [3]. The parameter notation is as in [3]: b, atomic size; a, colloidal particle size; d, space dimension (here three); D_c, scaling power (typically 1.5); $n_a\sim(a/b)^d$, number of atoms per particle; κ, shear modulus of a single particle; $\alpha(\approx 0.1)$, interparticle bonding efficiency. L is the length scale over which we probe the elasticity of the structure: here, it should be larger than the distance between plates.

In our problem, the local probability $p(x)$ plays the role of local concentration in the sheared-suspension problem. Following [2], we expect that, above some shear-flow threshold γ_c, the system will separate in a central region—the 'plug': low γ, large p—and two symmetric regions next to the plates, where γ is large and p low, i.e. where the rigidity of the structure is overcome by the flow and few bonds are left along $0x$ (see figure 1). To check this prediction, let us now turn to the equations of motion. The equation for flow is trivial:

$$\rho\dot{v}=\frac{\partial\sigma}{\partial x}=\eta\frac{\partial\gamma}{\partial x}+\gamma\eta'\frac{\partial p}{\partial x} \tag{4}$$

where $\eta'=\partial\eta/\partial p$ (>0). The equation for the dynamical variable $p(x)$ is more interesting. $\dot{p}$ is made up of three terms. Just as in Nozières and Quemada [2], there is a chemical potential term and a lift term, here corresponding to the 'orientation current' J along $0x$:

$$J=\alpha_0 p\left(-\mu'\frac{\partial p}{\partial x}+\Gamma_{\text{lift}}\right)\qquad \Gamma_{\text{lift}}=-\beta\gamma\frac{\partial\gamma}{\partial x}. \tag{5}$$

The chemical potential $\mu(x)=\int\mu'\,\mathrm{d}p$ expresses an elastic reaction† of the colloidal structure and, as such, is related to the rigidity A^{-1}; α_0 ($\neq\alpha$!) is a 'mobility' and Γ_{lift} is a force (the gradient of an energy varying as γ^2; physically, α_0 is related to an *orientational* mobility of each bond lying in the flow and Γ_{lift} correspondingly relates to a torque). The origin and sign of β are discussed in detail in [2]. In our problem, β should be positive in general due to entropy-controlled clustering contributions [4, 5]. As in [2], we assume α_0 and β to be independent of p but, in contrast to the suspension problem, the density p is of course *not* conserved, and $\dot{p}$ contains a third term, a structural *rigidity* term, obtained from (1). So we write

$$\dot{p}=-p_0A\dot{\sigma}-\frac{\partial J}{\partial x}=-p_0A(\eta\dot{\gamma}+\eta'\gamma\dot{p})$$
$$+\alpha_0\beta\left[\gamma\frac{\partial\gamma}{\partial x}\frac{\partial p}{\partial x}+p\left(\frac{\partial\gamma}{\partial x}\right)^2+p\gamma\frac{\partial^2\gamma}{\partial x^2}\right]+\alpha_0\mu'\left[\left(\frac{\partial p}{\partial x}\right)^2+p\frac{\partial^2 p}{\partial x^2}\right]. \tag{6}$$

† I am indebted to a referee who pointed out the need for such a term in an early report on this letter.

We now carry out a linear stability analysis (i.e. discuss the compatibility of (4) and (6)), whereby higher powers of fluctuations drop out. Taking small fluctuations to be of the form $[\mathrm{i}(qx-\omega t)]$ and putting $y=\mathrm{i}\omega$ (>0), the compatibility condition is

$$y^2[\rho(1+p_0A\eta'\gamma_0)]-y[q^2(\eta+\rho\alpha_0\mu'p_0)]+q^4\alpha_0p_0(\eta\mu'-\gamma_0^2\eta'\beta)=0. \quad (7)$$

Two limiting cases are readily distinguished.

(*a*) *Rigid aggregate.* The deformability A is small or moderate. An instability sets in when the average shear γ_0 exceeds a threshold given by

$$\gamma_c^2=\frac{\eta\mu'}{\eta'\beta} \quad (8)$$

independent of A. This critical shear is identical in form to that found in [2]. The distorted structure will look qualitatively as shown in figure 1. As usual in plug flow, the flow impedance will suddenly decrease above threshold.

(*b*) *Soft deformable aggregate.* A is large (of course, if A is too large, the elasticity theory of [3] breaks down). An instability of a different type appears above some—relatively low—threshold given, to first order, by

$$\gamma_c'=\frac{(\eta-\rho\alpha_0\mu'p_0)^2}{4\rho\alpha_0\mu'p_0^2\eta'\eta}\frac{1}{A}. \quad (9)$$

The eigenfrequencies are now *complex* and can be related to some kind of assisted wavy Taylor instability. In a very recent experiment on colloidal *crystals* [6], comparable features have been demonstrated.

In this letter, we have introduced a number of simplifying assumptions, some of which may turn out to have a rather limited range of validity. For instance, we have not considered the possible excluded-volume effects in the sheared 'phase' close to the wall. Also, we have not attempted to discuss the distorted regime above threshold and, in particular, the positioning of the 'Maxwell plateau' [2] associated with the instability, nor the extension of the model to multiconnected 'percolating' structures [5]. Nevertheless, our considerations might be related to a few problems of practical interest in large-scale hydrology, rheology, industrial flows and perhaps even in biology. In view of the broad range of variation open to the different parameters (see, e.g., (9)), it seems pointless to give numerical applications here.

Our main conclusion is that, when addressing problems such as shear-induced instabilities or melting, one should allow for non-uniform, domain-like, configurations such as found, for example, in plug flow.

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