RHEOLOGY OF NON-BROWNIAN FIBRES WITH ADHESIVE CONTACTS

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Abstract : An experimental investigation is undertaken into the shear thinning behaviour of non-Brownian rigid fibre suspensions in Newtonian fluids. In particular, we investigate the influence of the shear stress and the fibre concentration on the transient and steady shear viscosity of the suspension. The shear stress is adjusted by varying both the shear rate and the solvent viscosity. In the semi-dilute concentration regime (low collision frequency among the fibres), the suspension is found to be nearly Newtonian over the stress range we investigated. In the concentrated regime, the suspension becomes shear thinning below a certain shear rate. The shear thinning effects increase drastically with concentration and decrease with solvent viscosity. Although shear thinning behaviour of fibre suspensions has often been reported in the literature, its physical origins are not well understood. Here, our experiments are interpreted in the framework of formation and breaking of fibre flocs due to the competition between hydrodynamic and colloidal forces.

Keywords: Fibre suspension, shear flow, flocculation, shear thinning.

1. Introduction

During industrial processing of fibre composite materials or paper making, it is important to avoid flocculation of the fibres and the resultant fibre concentration heterogeneities which may have dramatic effects on the mechanical strength of the final product. In this work, we consider the shear flow of fibre suspensions in a Newtonian solvent, in which the structure evolution due to the flow is studied through the suspension rheological behaviour. In the case in which only hydrodynamic interactions are present, the effective stress generated by the flow of a fibre suspension has been a subject of many theoretical (1,2) and experimental (3,4) studies. In such situations (dilute and semi-dilute regimes), the experiments are generally in good agreement with the available theories. On the other hand, when the fibre concentration is higher or the flow is non-linear (5) so that non-hydrodynamic interactions such as physical contacts come into play, the discrepancy between experimental results and purely hydrodynamic theories becomes significant. The presence of mechanical contacts between the fibres increases considerably the effective stress of the suspension (6, 7). One might expect that lubrication forces would prevent contacts between particles in a viscous fluid. However it can be shown (6) that these forces are much less efficient when the particles are very thin.

The possibility of contact interactions between fibres in concentrated suspensions yields many non-linear rheological characteristics (8) such as finite normal stress differences, and shear thinning which we consider in detail in this study. Although this shear thinning property of fibre suspensions has already been reported in the literature (9), its origin remains unclear. For instance, it is generally explained (9) by refering to a mechanism in which the actual shear rate exerted on the solvent is higher than the apparent shear rate due to the presence of the particles, causing a drift of the solvent Newtonian plateau to low shear rates. For long and flexible fibres the shear thinning has been attributed to entanglement effects (10). In our experiments we found shear thinning behaviour even when the fibres are rigid, and our results cannot be interpreted in the framework of the reduction of the solvent Newtonian plateau due to the presence of the particles. Finally, it is to be noted that even when including Coulombic friction, Sundararajakumar and Koch (6) did not find shear thinning in their numerical simulations of rheological behaviour of concentrated fibre suspensions.

2. Experiments

The suspensions consist of nylon fibres in silicon oil (Rhodorsil). The fibres are obtained from a nylon thread whose diameter is fixed during spinning. The mean diameter d is 14.5 μm with a tolerance (given by the manufacturer) of about 2%. The process of cutting the thread into fibres, which is purely mechanical, induces a certain dispersion in their length distribution and a slightly irregular shape at each end. The length distribution is found to be approximately Gaussian with a standard deviation small enough (6%) for the fibres to be considered almost monodisperse. The mean value of the length L is 0.5 mm. The aspect ratio ($r=L/d$) is then 36. Using the criterion of Forgacs and Mason (11) for fibre bending in shear flow, it is found that fibre flexibility is unimportant in our experiments. This criterion has been derived by solving the shear-induced fibre buckling equation for an orientation for which the hydrodynamic compressive force is maximum (45° about the flow direction, in the flow-gradient plane), and taking the first mode of deformation. They found a 'critical' shear stress for fibre bending given by (11): $(\mu\gamma)_{crit}= E_b(\ln 2r-1.75)/2r^4$. The bending modulus E_b of nylon is about $3x10^9$ N/m^2, which gives a critical shear stress for bending of about 2.10^3 Pa in the case of our fibres, while the shear stresses involved in our experiments are smaller than 10^3 Pa. Experiments are conducted in three solvents (silicon oil) corresponding to different viscosities : 2.1 Pa.s, 12.2 Pa.s and 60.1 Pa.s. These values are determined at 25 _C in the Newtonian plateau. The three oils have the same density of 0.97 g/cm^3, while the fibre density is 1.13 g/cm^3; so there are some sedimentation effects. However, the fibre sedimentation time scale τ_S (the time required for a fibre parallel to the vertical direction to sediment over its length) is quite large: $\tau_S=8\mu l/\Delta\rho g d^2(\ln 2r-0.72)\sim 2$ h, for the smallest viscosity μ we used. Moreover, the measurements are conducted in a Couette device which is known to minimize the effects of sedimentation on rheological measurements. The Peclet number (hydrodynamic forces/ Brownian forces) can be estimated as : $Pe=\mu\gamma\pi L^3/3kT\ln(r)$. Pe is higher than 10^8, so the fibres can be considered non-Brownian. We used suspensions with four different concentrations corresponding to $nL^2d=0.5$, 1, 1.5 and 2. The variable nL^2d controls the importance of direct fiber-fiber contacts in a suspension. In the semi-dilute regime, $nL^2d<1$, fibers undergo purely hydrodynamic interactions. In more concentrated suspensions ($nL^2d>1$) the frequency of collisions between fibres in a shear flow is observed in numerical simulations (6) to increase drastically.

The rheometer was a Stresstech (from Rheologica) which can detect a torque as small as 10^{-8} N.m, and the position resolution of the sensor is as accurate as 1 μrd. This enabled us to carry out measurements at quite low shear rates. All the measurements were made in a Couette geometry (concentric cylinders) in order to have shear flows uniform at the fibre length scale and to minimize sedimentation effects. The diameter of the inner cylinder was 50 mm, and the gap was 2 mm which is only 4 fibre lengths. Bibbo (3) showed that if the gap-to-fibre-length ratio is higher than 2, boundary effects are not significant for the viscosity results in the case of semi-dilute Newtonian fibre suspensions. One may however question whether boundary effects can also be disregarded in our experiments since the suspension is suspected to form fibre flocs and interact differently with the walls. This remains an open question.

3. Results and discussion

3-1 Transient behaviour of the viscosity

Viscosity measurements have been made as a function of time at different shear rates and for suspensions with different fibre concentrations and solvent viscosities. At relatively high shear stresses, the temporal evolution of measured viscosity shows the usual behaviour. It rises goes through a maximum and then decreases to a steady state value consistent with the well established orientation distribution for hydrodynamically interacting fibres (12). After establishing the suspension structure characteristic of a Newtonian fibre suspension sheared at a high shear rate, the shear rate is reduced to a much smaller value, $\gamma = 0.00334\ s^{-1}$. We observe a marked increase in the viscosity with time at this small shear rate (Figure 1). After the viscosity reaches a steady state corresponding to $\gamma = 0.00334\ s^{-1}$, we increase the shear rate to $\gamma = 0.0299\ s^{-1}$. The viscosity decreases due to the increased shear rate. This experiment can be understood only if we recognise that there has been a dramatic change in the microstructure of the suspension at the small shear rate. This structure must involve more than a change of orientation distribution, because the observed viscosity at the smallest shear rate is larger than the largest possible value that could be obtained for non-contacting semi-dilute fibres with any orientation distribution. The microstructure remained when we stopped the flow as was indicated by the fact that the viscosity after the shear was restarted was the same as that observed upon cessation of the flow. These results can be understood if we suppose that the fibres form flocs at low shear rates that are held together by attractive forces. Fibres in concentrated suspensions can be brought into contact by shear flows (6). If the shear stress is not strong enough to overcome the adhesive forces between the fibers, flocs of more and more fibres can be formed when increasing the shear strain. This may be the origin of the viscosity increase with shear strain obtained at small shear rates (Figure 1).

3-2 Shear thinning

The steady state viscosity has been considered as a function of shear rate for different solvent viscosities and suspension concentrations. We found that the shear thinning effects increase with concentration and decrease with solvent viscosity. In the semi-dilute regime the suspension is nearly Newtonian over the entire shear rate range (4 to 5 orders of magnitude) investigated. The solvents used in the study are Newtonian over the range of shear rates considered here.

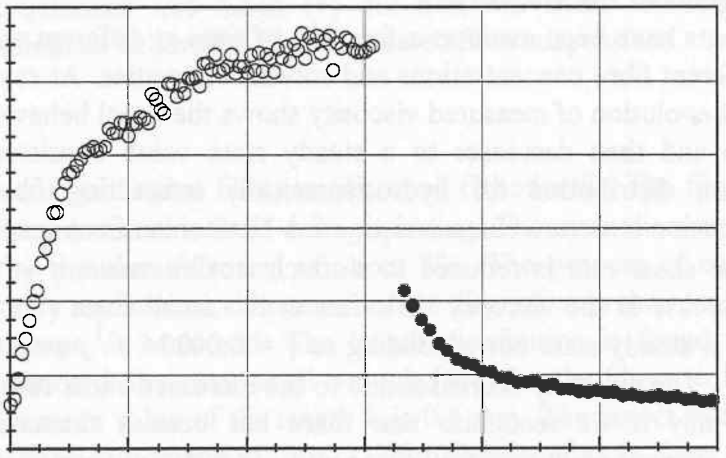

Figure 1 : Viscosity versus strain for 2 different shear rates : γ=0.00334 s^{-1}(○) followed by γ=0.0299s^{-1} (●). The suspension was presheared at a high shear rate (γ=9.68 s^{-1}) before initiation of the experiment.

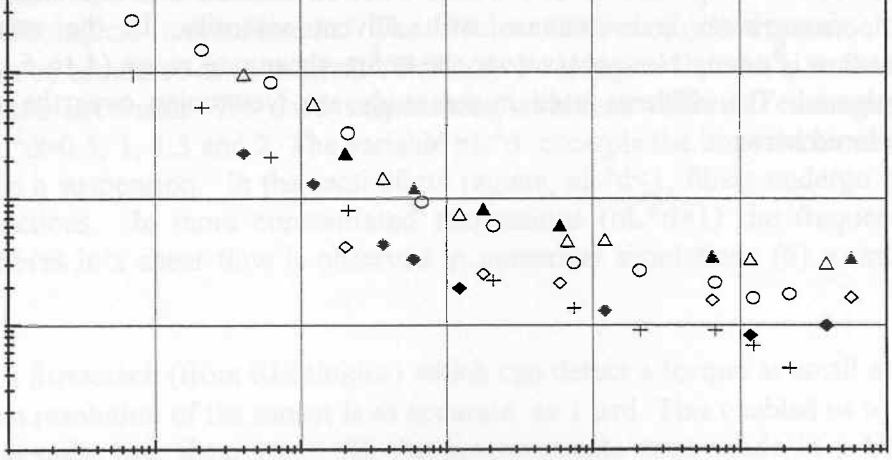

Figure 2 : Specific viscosity versus shear stress ($\mu\gamma$) for six suspensions corresponding to three different solvent viscosities and two concentrations. μ = 2.1Pas, nL^2d=2 (o) ; μ = 2.1Pas, nL^2d=1 (+) ; μ=12.2 Pas, nL^2d=2(Δ) ; μ=12.2 Pas, nL^2d=1 (◆) ; μ=60.1 Pas, nL^2d=2 (•) ; μ=60.1 Pas, nL^2d=1 (◊).

Figure 2 represents the specific viscosity (increase of the viscosity relative to the pure solvent value) versus shear stress for six suspensions corresponding to different solvent viscosities (2.1 Pas, 12.2 Pas, and 60.1 Pas) and for two concentrations (nL^2d=1 and nL^2d=2). This figure shows that for a given concentration the strength of the shear thinning is mainly determined by the shear stress. The shear thinning starts at approximately the same shear stress (about 10 Pa) for each of the six suspensions. One source of shear thinning is adhesive contacts between fibres. This adhesive force F_a may be related, for example, to the colloidal forces which have molecular origin and depend upon the physical and chemical properties of the solvent and the surface of the fibres. The hydrodynamic force that the shear flow exerts to pull two fibres apart is approximately F_h♠$\mu\gamma L^2$. The competition between the two forces should determine the shear thinning behaviour of the suspension. A dimensionless number can be defined as $\alpha=F_h/F_a=\mu\gamma L^2/F_a$ to quantify the shear thinning effects in the framework of breaking and formation of flocs. For $\alpha>1$, the suspension is Newtonian and for $\alpha<1$, the suspension is shear thinning. In our experiments, shear thinning occurs for shear stresses $\mu\gamma<10$ Pa. This would be consistent with the criterion $\alpha<1$, if we consider the adhesive force for contacting fibres as F_a♠2.510^{-6} N. Our experimental results shows that this adhesive force is roughly independent of the solvent viscosity and the shear rate. The concentration dependence of the rheological behaviour cannot be explained by this simple argument. Instead, one would need to conduct numerical simulations similar to those in (6) but incorporating adhesive forces to take into account the multi-body interactions associated with floc formation.

4. Conclusions

We have reported rheological measurements for *macroscopic* fibre suspensions which demonstrate that there can be a drastic change in the microstructure of the suspension at sufficiently small shear stresses. This structure formation has been attributed to fibre flocculation due to adhesive forces. Even though the colloidal forces are quite small between *millimeter* fibres, their effect on the suspension structure can be significant. For instance, this is the case in pulp fibre suspensions in water used for paper-making, in which hydrodynamic forces are comparable to colloidal forces due to the low viscosity. In practise one has to use turbulent flows to break the flocs. Note that in pulp fibres, flocculation is also made easier by the flexibility of the fibres. Despite the high viscosity of polymer solvents, there is a possibility of fibre flocculation due to colloidal forces in flow regions where the shear rate is sufficiently small. There may be other situations such as thickeners used in foods or pharmaceuticals where fibres with attractive forces give shear thinning

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