Couplage rhéologie – microstructure d'une suspension de particules dans un fluide à seuil

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Résumé :

De nombreux matériaux naturels ou à enjeu industriel sont constitués de particules macroscopiques dans un fluide à seuil, comme le béton frais ou les laves torrentielles. Une suspension concentrée de particules présente des propriétés rhéologiques complexes, telles que élasto-plasticité, rhéofluidification, différence de contraintes normales,… Certaines de ces propriétés macroscopiques non triviales peuvent être expliquées par le comportement de la suspension à l'échelle locale, en particulier par sa microstructure (organisation spatiale des particules). Sous cisaillement stationnaire, une suspension Newtonienne développe une microstructure anisotrope, mise en évidence par la fonction de distribution de paires [1, 2], qui peut notamment expliquer l'apparition de différences de contraintes normales [3]. Que se passe-t-il dans le cas où la rhéologie du fluide suspendant est complexe ? Nous étudions le cas de sphères dures non Browniennes en suspension dans un fluide à seuil. Ce matériau, qui présente un comportement élasto-visco-plastique, est ici une émulsion concentrée. Nous étudions le couplage entre comportement macroscopique et microstructure de cette suspension en associant rhéométrie classique dans un dispositif de cisaillement plan entre deux plateaux rotatifs d'espacement variable et imagerie 3D par microtomographie X. L'étude de la fonction de distribution de paires à 3D en cisaillement simple stationnaire montre une anisotropie de la microstructure de la suspension dans le plan vitesse-cisaillement, pour des écoulements rotationnel et d'écrasement. Le régime instationnaire est également étudié suite à diverses préparations (i.e., diverses histoires de cisaillement). Les propriétés élasto-plastiques (contrainte seuil de plasticité, module élastique de cisaillement) du matériau montrent alors de remarquables propriétés d'écrouissage (durcissement, modification de la ductilité) et de variations du module élastique de cisaillement, qui peuvent être reliées à sa microstructure. Il est ainsi possible de contrôler les propriétés élasto-plastiques initiales d'une suspension de particules dans un fluide à seuil via l'histoire des sollicitations.

Abstract :

Numerous industrial and natural fluids, such as fresh concrete or debris flows, are made of coarse particles suspended in a yield stress fluid. Such suspensions exhibit complex rheological behaviours: elasto-plasticity, shear-thinning (non constant viscosity), normal stress differences, ... , that result from the local behaviour of the suspension, and eventually from its microstructure (spatial organization of particles) [3]. In the light of results about shear-induced microstructure in concentrated suspensions of spheres in Newtonian fluids [1, 2], we experimentally addressed the role of the particle microstructure in this complex rheology. We use a model system made of non-Brownian spherical hard particles suspended in a concentrated emulsion. We associate rheological measurements to X-ray microtomography imaging techniques allowing for the determination of high resolution pair distribution functions in three dimensions. We characterize the rheological behavior and the microstructure for various shear histories (squeeze flow and rotational flow, transient and steady state, various low and high shear rates) and we relate it to their elastic and plastic properties. We show that the elastic and plastic properties of the suspensions depend strongly on shear history; in particular, for certain initial states, the materials display a strain hardening behavior. We relate all these complex properties to the observed shear-history-dependent microstructures.

Mots clefs : Suspension, Fluide à seuil, Microstructure, Rhéologie, Elasto-plasticité, Ecrouissage, Tomographie X, Fonction de distribution de paire

1 Introduction

The presence and the spatial organization of spheres in a concentrated suspension control its mechanical behaviour, as microstructure is related to interactions between spheres and intrinsic rheology of the fluid. Whereas numerous experimental and theoretical studies focused on the dependence of the mechanical properties of a suspension with the concentration of particles [4-6], fewer studies addressed the coupling between rheology and microstructure of a suspension [2, 7-8]. In the case of particle suspension in a Newtonian fluid, a simple shear flow induces an anisotropic and asymmetric microstructure in the shear-velocity plane 1-2]. This anisotropy originates from direct interparticle interactions, and is at the origin of the development of normal stress differences. Moreover, any change in the spatial characteristics of flow is shown to result in a progressive change of the microstructure and, as a result, to a time-dependent macroscopic behaviour. The study of the microstructure and more generally the small-scale behaviour of a suspension allows explaining numerous complex rheological properties, such as flow threshold, non constant viscosity (shear-thinning or shear-thickening), normal stress differences. Here, we associate rheological measurements to 3D imaging in a dedicated experimental set-up (Figure 1a) to characterize the mechanical properties and the microstructure of a suspension. As concentrated suspensions in general exhibit structuration under shear –colloidal particles [7], macroscopic hard spheres [2], deformable spheres [8] in a Newtonian fluid–, our approach and our results about suspensions of spheres in a yield-stress fluid should be extended to other systems.

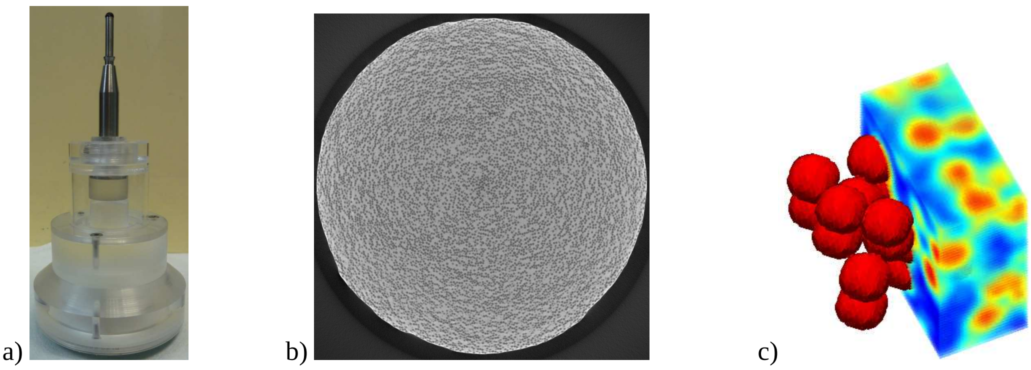


FIG. 1 – a) Parallel plates geometry allowing for a simple shear flow (either by rotation or by squeeze) and X-ray microtomography imaging. b) One horizontal slice of the suspension at 35% volumetric concentration. c) 3D reconstruction of a sub-volume of a suspension and segmentation of particles.

2 Experiments and methods

Our model suspension is made of non-Brownian spherical hard particles of crystalline polystyrene of diameter d=140µm suspended in a concentrated emulsion: an aqueous iodine-loaded phase is dispersed in an oil phase. The experimental set-up (Figure 1a) allows viscosimetric measurements in a parallel plates geometry (radius R=1cm and gap h=2mm) allowing both for squeeze and rotational flows and X-ray microtomography imaging conducted at Laboratoire Navier with a spatial resolution of 12µm (Figure 1b). In practice, a desired shear history is imposed to the suspension in the rheometric cell, then stopped by controlling zero rotation and squeeze velocities for the complete scanning. The suspension microstructure is ensured to be exactly the same just before and after the interruption of shear because of the existence of the yield stress. From the 3D image intensity, particles are first detected as local minima, then their position are systematically computed as the symmetry center of the intensity signal, defined as the position minimizing its standard deviation as a function of the distance to the center (Figure 1c). By interpolating linearly the image of the particle, we achieve to the desired precision (1µm) on the position of the particle. From the particle positions, we compute the pair distribution function g(**r**), representing the probability of a particle pair along the vector **r**, characterized by a distance and a spatial direction.

3 Rheological behaviour: Influence of history and strain hardening

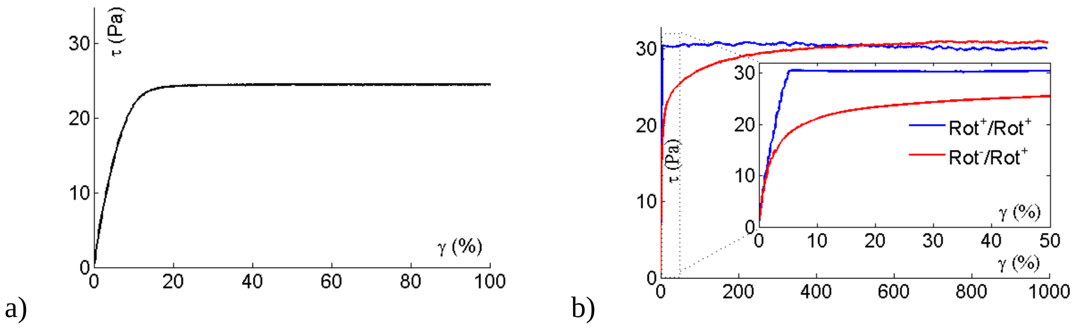


FIG. 2 – Stress- strain response to a quasi-static shear at constant rate ($10^{-2}s^{-1}$) of a yield-stress fluid (a) and of two suspensions of spheres in a yield-stress fluid, prepared by different shear histories (b) : the blue and red have been pre-sheared with the same and opposite shear than the imposed solicitation respectively.

Under a quasi-static shear at constant rate ($10^{-2}s^{-1}$), the yield-stress fluid has an elastic response followed by a plastic stationary flow, as shown by the evolution of stress as a function of strain (Figure 2a). When particles are suspended in the yield-stress fluid, the suspension is still elasto-plastic, but its mechanical properties are modified and do depend strongly on shear history. This effect is visible on the different responses to a quasi-static constant shear of two suspensions characterized by different shear histories (Figure 2b). In blue, the suspension has been pre-sheared with the same shear than the solicitation: stress evolves linearly with strain until being constant equal to the yield-stress. In red, the suspension has been pre-sheared with the opposite shear than the solicitation: the suspension seems to be smoother and the transient regime is long before reaching the plastic stationary flow. Elastic and plastic contributions to the mechanical response were distinguished by instantaneous measurements of the elastic modulus G' with oscillation measurements (Figure 3a) and of the yield-stress τ_Y with load-unload cycles (Figure 3b). Here the instantaneous yield-stress τ_Y corresponds to the threshold stress between reversible and irreversible response of the suspension, but does not correspond to the threshold stress to get a stationary plastic flow. The different mechanical responses to the imposed shear are related to different transient values of elastic shear modulus and plastic yield stress, as shown by Figure 4a et 4b: when presheared in the opposite sense, the suspension has low initial values of G' and τ_Y, that increase transiently until their stationary larger values; contrary to the case of a suspension presheared in the same sense, that has constant elasto-plastic properties. What are the elementary mechanisms of these variations of mechanical properties? How a suspension of spheres in a yield-stress spatially organize under steady or transient flows?

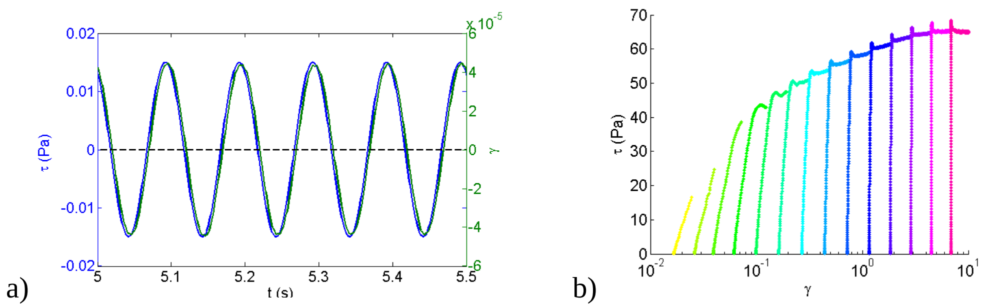
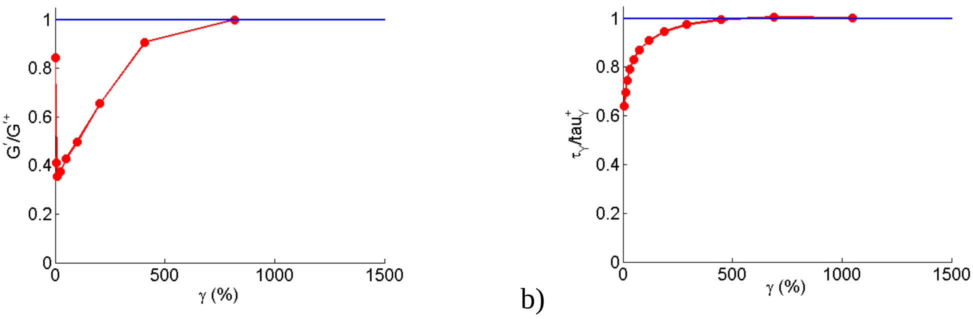


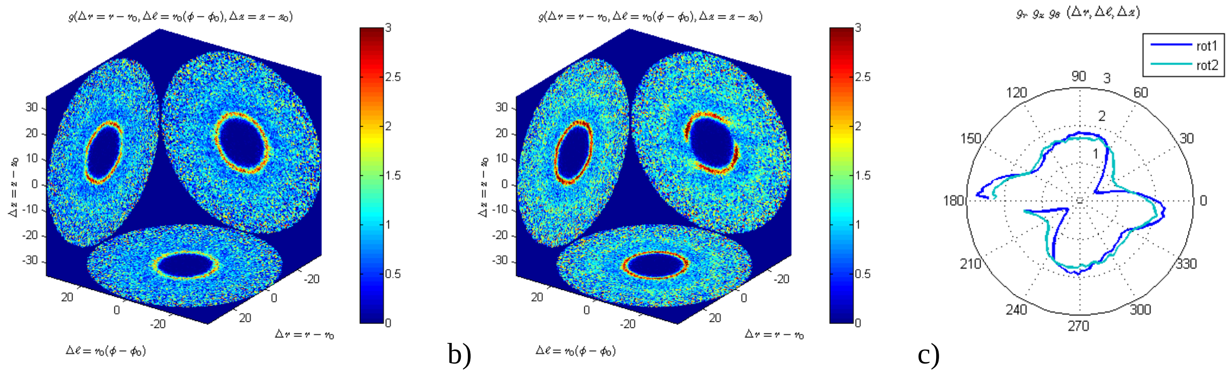
FIG. 3 – Illustration of the principles of oscillations at high frequency and low amplitude to measure the elastic modulus G' (a) and of load-unload cycles to measure the yield-stress τ_Y (b).



a) b)

FIG. 4 – Elastic shear modulus (a) and (plastic) yield stress (b) properties measured during the shear.

4 Coupling between microstructure and rheology



a) b) c)

FIG. 5 – a-b) The 3D pair distribution function in three orthogonal (horizontal, radial and azimuthal) planes for stationary squeeze (a) and rotational (b) flows. c) Polar plots of g(r) in the shear-velocity plane, averaged for pair distances between d and d+d/3, with d the particle diameter, for two shear rates of $10s^{-1}$ (rot1) and $10^{-2}s^{-1}$ (rot2).

For various steady and transient states, the microstructure is characterized via the probability density function g(**r**). Here g(**r**) is shown for particles located inside a ring centered at 3R/4 from the center of the geometry, with R its radius, to take into account the heterogeneity of shear stress, rate and strain in a parallel plates geometry.For stationary flows, the suspension structure is anisotropic in the shear-velocity plane -in the radial plane by squeezing (Figure 5a), in the azimuthal plane for a rotation shear (Figure 5b)-, while isotropic in the two other orthogonal planes. When the value of the shear rate for rotational flows is systematically varied , we observe a variation of the microstructure, as visible from the polar plot of g(r) in the shear-velocity plane, averaged for pair distances between d and d+d/3, with d the diameter of particles (Figure 5c). This may be attributed to different relative contributions of elastic and hydrodynamic interactions. During transient regimes, the evolution of the microstructure is shown to depend strongly on the initial preparation of the suspension and on its shear history (Figure 6). Two suspensions pre-sheared in one direction and the opposite direction have stationary mirror microstructures with respect to the shear axis, in particular with depletion of pairs at short distances in the region of extension (Figure 7). This follows that when they are sheared, one has a constant microstructure, while the other one continuously reorganizes to reach the stationary microstructure, with one transient microstructure being symmetric with respect to the shear axis (Figure 6b).

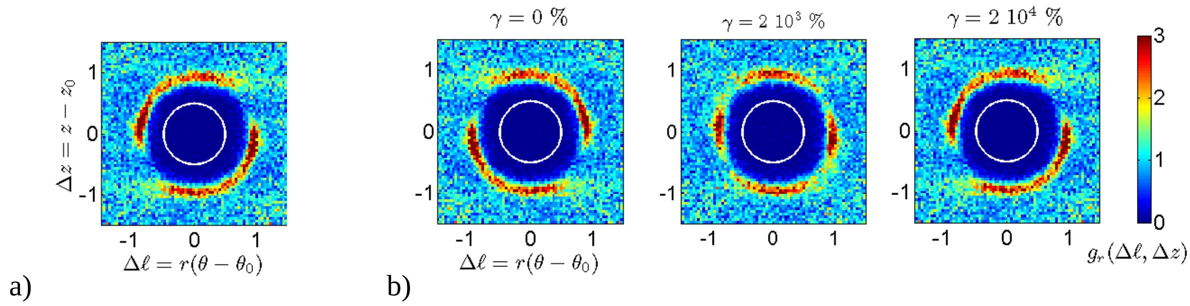


FIG. 6 – a) Probability density function g(r) in the shear-velocity plane for the stationary rotational shear at rate $10^{-2}s^{-1}$. b) Evolution of the microstructure in the shear-velocity plane during the shear of a suspension initially pre-sheared in the opposite direction.

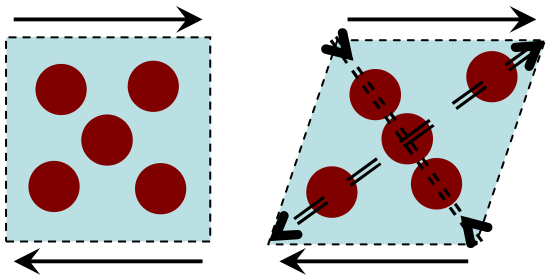


FIG. 7 – Scheme of an elementary volume of suspension in a plane shear, showing the main stress directions of compression and extension.

5 Conclusion

By associating rheological measurements to X-ray microtomography imaging techniques allowing for the determination of high resolution pair distribution functions in three dimensions, we characterize the rheological behavior and the microstructure for various shear histories (squeeze flow and rotational flow, transient and steady state, various low and high shear rates) and we relate it to their elastic and plastic properties. We show that the elastic and plastic properties of the suspensions depend strongly on shear history; in particular, for certain initial states, the materials display a strain hardening behavior. We relate all these complex properties to the observed shear-history-dependent microstructures.

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