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Rheo-PIV of Aerosil® R816/polypropylene glycol suspensions

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A B S T R A C T

In this study we adapt a conventional rotational rheometer to perform PIV measurements on wide gap Couette flow. We measure the radial profile of the azimuthal velocity and the rheometric properties simultaneously. We use image deformation to convert the images from cylindrical coordinates to a rectangular frame, thus improving the PIV algorithm performance. This technique is validated with the Newtonian liquid phase of the rheological suspensions, showing an excellent agreement with the theoretical velocity profile. We also characterize the rheological properties of Aerosil® R816 Polypropylene Glycol suspensions, which show a shear-thickening behavior between two shear-thinning regions. The relation between the viscosity and the shear rate is computed in the whole radial domain between the inner and the outer cylinder. These Rheo-PIV measurements allow us to distinguish the onset of shear thickening and jamming, the former one being connected to a smooth change in the measured shear rate, whilst the latter corresponds to a sudden change in the measured shear stress value.

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1. Introduction

The experimental and theoretical knowledge of non-Newtonian fluids is relevant not only for applied research, but also for the fundamental understanding of flow mechanisms which take place inside these complex fluids. These are characterized by a viscous stress tensor that depends non-linearly on the shear-rate tensor. This dependence is generally studied using rheometers. In particular, one of the most used is the rotational rheometer. Due to the vast range of applications (plastics, concretes, asphalts, foods, etc) and the appearance of precise Computational Fluid Dynamics (CFD) to model these fluids, researchers need to improve the models used for the stress tensor. One of the main reason is that an accurate estimation of the stress-tensor can provide the dynamics of complex fluids by means of simulation [1]. Secondly, but not less important, numerics are able to "probe" inside the rheometers, giving some key physical points of the fluid studied. Nevertheless, tensor modeling involves some procedural errors which have lead the scientific community to directly measure the flow-field by non intrusive techniques. One of these errors in the experimental procedure stems from the possibility of slipping boundaries, that cannot be observed by rheometers. This has been studied with Particle Tracking Velocimetry (PTV) [2], and more recently using Particle

Image Velocimetry (PIV) [3–5]. Other sources of experimental error are related to the principle of operation of rotational rheometers, which only measure torque and rotation speed. To have an estimation of the shear-rate, rheometers employ some approximations such as the narrow gap, together with the assumption of a constant shear-rate (simple shear-rate). The viscosity can be obtained with the ratio of the stress and the estimated shear-rate. However, the real viscosity must be computed from the rheometer data solving the inverse problem [6–8]. The inverse Couette problem computes the shear rate for each rotation-rate that causes a given measured shear stress by the rheometer. An integral equation for the shear rate has to be solved as a function of the rotation-rate and the shear stress. This problem is ill-posed. The main drawback of this technique is the assumption of a relation between the local shear stress and the shear rate, hypothesis that has been invalidated recently [9,10]. Other procedures that can be used to obtain the shear rate is to measure it directly with any other experimental technique, for example PIV, as it has been reported recently in [11].

Shear-thickening fluids are those in which the shear stress increases with the shear rate more than what is predicted for a Newtonian fluid. This increase in the energy required to move the fluid may cause problems in its industrial processing, so understanding the physical cause of this behavior is of great scientific interest. One of the most intriguing features of these fluids is the appearance of this effect in very simple types of suspensions, for example hard spheres in a Newtonian fluid. From experiments, two types of

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shear-thickening can be characterized: continuous (CST) and discontinuous (DST). The first one corresponds to a case in which for a given critical value of the shear rate, $\dot{\gamma}_c$, the fluid needs a force bigger than the one predicted for a Newtonian fluid ($\tau \approx \dot{\gamma}^n$, with $n > 1$) to keep flowing. This is observed in the form of a smooth transition in the steady viscosity curve. For the second case (DST), the shear stress is necessary to keep the fluid flowing with a shear rate $\dot{\gamma} > \dot{\gamma}_c$ jumps suddenly to very high values (usually several orders of magnitude). This process is usually called *jamming* [9]. Three theories try to explain this flow behavior. The first one is hydroclustering, in which an increase of the shear-rate pushes the particles one into each other, and it becomes more difficult to move one apart from the others due to lubrication forces. This mechanism leads to the formation of clusters which increase the drag force. The second theory is that of order-disorder transition, which states the presence of a microstructural transition from ordered layers for lower shear-rates to disordered layers at high shear-rates. These two theories are able to predict accurately the onset shear rate $\dot{\gamma}_c$ but are unable to predict the magnitude of the jump in the stress for the DST. The third theory, called dilatancy, states that when the fluid is sheared, the particles try to go around each other and often they are not able to achieve this goal, so they collide and the packing fraction expands. This model is able to predict both the onset shear-rate and the upper stress bounds for the case of DST fluids. The interested reader is referred to an excellent review given in [12].

Recently, new non-intrusive techniques have provided novel methods to study rheological properties of non-Newtonian fluids. For example, ultrasound velocimetry [13,14], MRI [9,15,16] or Particle Image Velocimetry PIV [3,17–22]. These techniques compute velocity profiles of the fluid, giving some insights into the dynamic behavior of the flow. On the other hand, MRI is able to measure 3D velocity fields and simultaneously the packing fraction inside the fluid, which determines whether the change in properties can be explained by particle migration, as it has been done in [9]. Other non-intrusive methods such as Small Angle Electron Scattering (SANS), are not able to provide the velocity field inside the fluid, but they are able to detect the size of the clusters formed, explaining the change in viscosity detected by rheometers [23]. PIV measurements also allow the measurement of velocity fields in two dimensions, thus being able to detect instabilities in the flow, at a lower cost than MRI techniques.

PIV technique can be applied in any geometry, for example microchannels [20], Hele-Shaw cells [22] or more complicated micro devices [21]. A PIV technique similar to the one presented for concentric cylinders is used in [18] to analyze a shear-thinning fluid, showing that it can provide accurate results in comparison to those given by a rheometer. The same setup is used in [17] to study particle migration in non-colloidal suspensions. There is a scarce number of references on fluid velocimetry in shear-thickening fluids [10]. Herle et al. [13] carried out an ultrasound velocimetry study of a worm-like micellar solution in a concentric cylinder setup, showing the appearance of shear bands in the radial profile. These bands started to oscillate when the shear-rate reached a minimum value, determining the threshold of an instability. When the flow was stable, the velocity profile was almost linear whereas it became banded when a critical shear rate was surpassed. On the other hand, Dehmoune et al. [14] did ultrasonic experimental measurements on a fluid that presented an apparent viscosity curve quite similar to the one studied in this manuscript. They did not report any unstable behavior although their velocity profiles presented the same shear bands when the fluid entered into the shear-thickening region.

In this paper, we study a complex fluid with an enhanced shear-thickening behavior preceded by a shear-thinning region at low shear-rates. A more diluted version has been the topic of previous studies from the traditional rheometry perspective [24]. Our main purpose is to measure velocity profiles aiming to obtain an experimental shear-rate measurement by means of the PIV technique to be compared with the shear-rate estimated by the traditional rheometer. To that end, different velocity profiles are measured at several rheological regions.

The paper is organized as follows. In Section 2 the rheology of the fluid of study and the PIV experimental procedure are described. In Section 3 we show the results for the non-Newtonian fluid. We discuss the results in Section 4 and finally, we draw some conclusions in section 5.

2. Materials and methods

2.1. Non-newtonian fluid

Aerosil® R816 (Degussa AG), a commercial hydrophobic fumed silica, which is obtained from Aerosil® 200 (Degussa AG) by means of replacing silanol (Si-OH) groups by alkyl groups ($C_{16}H_{33}$), has been used as solid phase. Its specific surface area BET is 190 ± 20 m^2/g and its primary spherical particles size is 12 nm [25]. Although the flame hydrolysis process initially produces spherical particles they fuse into fractal aggregates when they collide. These fractal structures can be considered as fumed silica primary structure, as the fusion process is irreversible [26]. We used polypropylene glycol (HO-[CH_2-CH(CH_3)-O]n-H) with a molecular weight of 400 g/mol (PPG400) on average (Sigma-Aldrich Chemical GmbH, Düsseldorf, Germany) as the continuous phase. This is a low-viscosity Newtonian liquid with a viscosity of 100 mPa·s (20°C, 1 atm).

The suspensions were prepared following a previously designed protocol [24,27], which is reproduced here. The liquid has been added to the silica powder and afterwards both were mixed in a stirrer for 10 min at 796 rpm. These values were determined as the minimum mixing time and velocity required to ensure reproducibility between batches. Samples were made in batches of 35 cm^3 and placed under vacuum and sonication at room temperature for about 1 h in order to remove air bubbles. Afterwards, the sample was allowed to rest for 12 hours in an airtight plastic bottle to avoid contamination by silicates from glass recipients. The volume fraction of Aerosil® R816 particles in the suspension studied here was the maximum we could make ($\phi_v = 0.096$), so the shear-thickening effect was enhanced. This concentration was calculated from bulk densities assuming silica particles as Euclidean solids. Higher solid concentrations are really difficult to obtain due to the fractal nature of silica aggregates in PPG400. Certainly, due to their fractal conformation, silica particles generate open loose primary aggregates [28] with an effective disperse phase volume fraction being much higher than what is expected from an Euclidean solid [29].

2.2. Rheological measurements

Steady shear rheological experiments were carried out on a RS600 rheometer (Haake-Thermo, Germany) using a cone-plate sensor system with a cone angle of 2° and a diameter of 35 mm. The temperature was kept constant with a controlled temperature bath (± 0.1 °C). Experiments were conducted under steady shear. The absence of wall slip was checked, comparing the results obtained with a cone-plate sensor system with a cone angle of 1° and a diameter of 20 mm.

The time-dependent behavior of fumed silica suspensions has been reported in [30,31]. To erase this pre-shear history-dependent behavior is a common practice to apply a pre-shear stage [32–34]. Therefore, to ensure that the results were reproducible, before each experiment, a steady pre-shear was applied up to an equilibrium