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Unsteady laminar flows of a Carbopol® gel in the presence of wall slip

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ABSTRACT

We present a comparative experimental study of unsteady laminar flows of a yield stress shear thinning fluid (Carbopol® 980) in two distinct configurations: a parallel plate rheometric flow and a pressure driven pipe flow. Consistently with the observations in the case of the rheometric flow, the in situ characterisation of the unsteady pipe flow reveals three distinct flow regimes: *solid (plug-like)*, *solid–fluid* and *fluid*. In both configurations and as the flow forcing is gradually increased, the yielding emerges via an irreversible transition. The irreversibility of the deformation states is coupled to the wall slip phenomenon. Particularly, the presence of wall slip nearly suppresses the scaling of the deformation power deficit associated to the rheological hysteresis with the rate at which the material is forced. An universal scaling of the slip velocity with the wall velocity gradients and a slip length which is independent on the degree of the flow steadiness is observed in the pipe flow.

1. Introduction

Yield stress fluids represent a broad class of materials made of high molar mass microscopic constituents which display a solid-like behaviour as long as the stress applied onto them does not exceed a critical value called the yield stress, τ_y, and a fluid behaviour beyond this threshold.

The constantly increasing level of interest of both theoreticians and experimentalists in yield stress fluids has a two-fold motivation. From a practical perspective, such materials have found an increasing number of applications for several major industries (which include foods, cosmetics, oil field, etc.) and they are encountered in the daily life in various forms such as food pastes, hair gels and emulsions, cement, and mud.

Understanding the unsteady flows of yield stress fluids is important in practical settings including industrially relevant flows of waxy crude oils [19,30], transport of ice slurries [35], coating flows employed in food industry and geophysical flows.

From a fundamental perspective and even in the case of minimal thixotropic effects, yield stress materials continue triggering intensive debates and posing difficult challenges to both theoreticians and experimentalists. From a historical perspective, the best known discussion is related to the very existence of the yield stress [1,2]. It was thus suggested that the yield stress is an *engineering* reality rather than a physical one or, in other words, the apparent yield stress behaviour observed during rheological tests is related to a very large Newtonian viscosity rather than to the onset of flow (yielding).

In context of the recent developments of the rheometric equipment (now able to resolve torques as small as 0.1 nNm and rates of deformation as small as $10^{-7}\ s^{-1}$), this debate seems to have reached an end. Indeed, recent and systematic rheological tests performed on various yield stress materials proved unequivocally the existence of a true yielding behaviour [21,32,6,12].

Whereas there exist numerous compelling investigations of the existence of yield stress, much less work has been done on the dynamics of the yielding transition. Thus, this topic interesting for both theoreticians and experimentalists, particularly in the case of realistic flows which are often unsteady and take place in the presence of wall slip. In practical flow situations involving pasty materials the yielding transition is often associated with the wall slip phenomenon which complicates even further the theoretical description. As in the case of pasty materials the wall slip manifests in a range of small shearing forces one can note that, for such materials, the wall slip and the yielding transition are coupled. This fact brings an additional experimental and theoretical difficulty in exploring the solid–fluid transition and modelling realistic flows of such materials in industrially relevant settings.

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Carbopol® gels have been frequently used in many rheological and hydrodynamic experimental studies of the yielding behaviour of viscoplastic fluids. They are synthetic polymers of acrylic acid initially introduced over six decades ago (B.F. Goodrich Co.). They are cross-linked with various chemical compounds such as divinyl -glycol, allyl-sucrose, and polyalkenyl polyether. Addition of a neutralising agent (e.g. sodium hydroxide, NaOH) leads to a decrease in the number of the positive hydrogen ions H^+ resulted from the dissociation of the polyacrylic acid in a polar solvent such as water. The un-compensated sodium ions Na^+ lead to an increase of the osmotic pressure and the individual polymer particles swell dramatically. Consequently, for polymer concentrations exceeding a threshold value c^* (overlap concentration), the swollen polymer particles jam forming a micro-gel system which can locally sustain finite deformations (behaving like an elastic solid) prior to damage. When the locally applied stresses exceed a threshold value the micro-gel system breaks apart and the material starts to flow. This is the commonly accepted microscopic scale origin of the macroscopic yielding of a Carbopol® gel.

Due to their low thixotropy, excellent micro-structural stability, optical transparency and highly reproducible and stable rheological properties, Carbopol® gels been considered for over a decade a *model yield stress fluids* [10,27].

It has only recently been shown that in a range of low applied stresses and depending on the manner they are forced these materials may actually depart from an ideal viscoplastic picture and that around the yield point thixotropic effects and irreversible deformation states may be observed [14,15,32].

In contrast to the large number of studies of the yielding of Carbopol® gels in rheometric flows, the body of literature concerning experimental studies of the solid–fluid transition of Carbopol® gels in industrially relevant flows is, to our best knowledge, more limited. An experimental study of the yielding and flow properties of a 0.2% Carbopol® in a capillary tube has been performed by González and his coworkers [29]. By combining classical rotational rheometry techniques with Digital Particle Image Velocimetry (**DPIV**) measurements of steady flows driven at various pressure drops the authors present a detailed experimental characterisation of the yielding process and the flow patterns. As the driving pressure drop is gradually increased, they observe a smooth solid–fluid transition rather than a sharp one and correctly identify this as one of the factors that lead to uncertainties in the determination of the yield stress. To overcome the difficulty of accurately measuring the yield stress triggered by the smoothness of the solid–fluid transition, the authors of the study propose an alternative method of measuring this parameter based on the analysis of the measured velocity profiles [29]. As the flows studied in Ref. [29] were stationary, this work did not capture the dynamic coupling (in unsteady flow conditions) between yielding and wall slip which, in part, motivates the present study.

Ma and his coworkers reported an experimental investigation of pipe flows of a petroleum-coke sludge slurry in the presence of wall slip [19]. They assess the wall slip behaviour using the Mooney method and correlate the slip behaviour with both the rheological properties of the slurries and the driving pressures.

The body of existing literature on unsteady pipe flows of Carbopol® gels is, to our best knowledge, quite limited which inherently limits the extent of this bibliography review.

A theoretical study of the dispersion of a solute in the pulsatile pipe flow of a yield stress fluid was recently presented by Nagarani and Sebastian [23]. Using the Casson model, they demonstrate that the periodic flow of the fluid contributes significantly to the total dispersion process compensating for the effect of the yield stress.

The development of the laminar unsteady pipe flow of a thixotropic fluid was studied by Corvisier and his coworkers [8]. By measuring the flow fields via the Particle Image Velocimetry technique, they have found a clear coupling between the kinetics of the micro-structural changes and the evolution of the flow.

Park and his coworkers have studied oscillatory flows of a 0.075% Carbopol® solution in a acrylic made U-shaped tube with a circular cross section [28]. A compelling experimental evidence of the significant role played by the elasticity on the measured unsteady velocity profiles (particularly near the centreline of the flow channel) is provided. Due to the elastic effects associated to the presence of un-yielded bands in the flow, strongly nonlinear time histories of the strain are recorded. To model their experimental findings, Park and his coworkers propose an elasto-viscoplastic model which fits their data better than the regularised Bingham model. Some discrepancies between theory and experiment still exist and may be due to the fact that their model does not account for the velocity slip near the wall.

The aim of the present study is to present a detailed comparative experimental characterisation of laminar unsteady flows of a Carbopol® gel in two distinct flow configurations: a rheometric flow between parallel disks and a pressure driven pipe flow.

The paper is organised as follows. The experimental setup, the measuring technique and the physical and rheological properties of the solutions are detailed in Section 2. The experimental results are presented in Section 3. A systematic description of the solid–fluid transition observed in a rheometric flow with a particular emphasis on the role of wall slip and its coupling to the irreversibility of the deformation states is presented in Section 3.1. A detailed characterisation of inertia-free unsteady pipe flows of a Carbopol® solution is presented in Section 3.2. The paper closes with a discussion of our main findings, of their impact on the current understanding of flows of yield stress fluids, Section 4. Several directions worth being pursued by future studies will also be discussed.

2. Experimental setup and methods

2.1. Preparation and characterisation of the Carbopol® solutions

A 0.08% (by weight) solution of Carbopol® 980 has been used as a working fluid. The procedure for the preparation of the solution is described as follows. First, the right amount of anhydrous Carbopol® 980 has been gently dissolved in water while continuing stirring the mixture with a commercial magnetic stirring device. The stirring process was carried on until the entire amount of polymer was homogeneously dissolved. A particular attention has been paid to the homogeneity of the final mixture, which has been assessed visually by monitoring the refractive index contrast of the mixed solution. Next, the pH of the mixture (initially around 3.2, due to the dissociation of the polyacrylic acid in water) has been brought to a neutral value by addition of about 140 parts per million (ppm) of sodium hydroxide (NaOH). The final value of the pH has been carefully monitored using a digital pH-meter (from Grosseron).

2.2. Rheological measurement protocol

The rheological properties of the solutions were investigated using a Mars III (from Thermofischer) rheometer equipped with a Peltier system able to control the temperature with an accuracy better than 0.1 °C and a nano-torque module which allows one to accurately explore very small rates of deformation, $\dot{\gamma} \approx 10^{-5}$ s^{-1}.

To quantitatively assess the role of the wall slip on the unsteady yielding process, two alternative geometries have been used: a smooth parallel plate system and a serrated one. The radius of each geometry was $R = 35$ mm and the gap between the parallel plates was $d = 1$ mm. Measurements of flow curves performed with the rough parallel plate system for various gaps