Ultrasonic velocimetry and the rheological investigation of complex fluids

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High-frequency ultrasonic velocimetry is used to explore the rheological behaviour of complex fluids sheared between concentric cylinders. The technique is based on time-domain cross-correlation of speckle signals backscattered either by the fluid microstructure or by contrast agents dispersed in the material. Velocity profiles with a 40 µm resolution can be measured every 0.1-10 s depending on the global shear rate. The technique is applied to various complex fluids that show "shear banding" (i.e. local heterogeneities of the shear rate due to local modifications of the microstructural organization) or shear-induced fractures.

Keywords: velocity profile,

1 INTRODUCTION

Contrary to Newtonian fluids, complex fluids may develop highly *inhomogeneous* flow patterns even under simple shear at low Reynolds numbers. This is due to the coupling between the fluid microstructure and the flow. Examples range from depletion-induced apparent wall slip in concentrated emulsions or fractures in gels to shear-banding in surfactant solutions, colloidal suspensions, foams, or granular materials [1]. In practical applications, such spatial inhomogeneity may be very misleading when interpreting rheological experiments based on the measurement of *engineering* quantities such as the shear stress or the viscosity averaged over the whole sample.

Moreover, spatial inhomogeneity often goes along with complex *temporal* behaviours and instabilities [2-3]. Due to a lack of temporally and spatially resolved instruments coupled to classical rheological tools, such *spatio-temporal* features remain difficult to assess. However, they may have a critical influence on the texture of the final product or on processing rates in industrial applications.

In this paper, we describe how high-frequency ultrasound can be used to measure and follow velocity profiles in sheared complex fluids with a 40 µm spatial resolution and a 0.1–10 s temporal resolution. Such ultrasonic velocimetry is coupled to a standard rheometer in Couette geometry (concentric cylinders). The resulting "ultrasonic rheo-velocimeter" is tested on various complex fluids, which illustrate the wide range of materials accessible to the technique and of possible subsequent characterizations.

2 COMPLEX FLUIDS AND THE NEED FOR LOCAL VELOCIMETRY TOOLS

2.1 Inhomogeneous flows in complex fluids

In complex fluids, the existence of a supramolecular organization (or "micro-structure") at some "mesoscopic" scale can lead to very complicated behaviours under flow: steady shear modifies the fluid microstructure and may even induce new structures or textures. For instance, shearing an initially isotropic wormlike micelle solution can lead to a nematic phase [4]. In concentrated granular materials and in soft glassy materials, solid-like states are observed that may coexist with shear-fluidized states [5]. Such transitions between shear-induced states are thus characterized by *inhomogeneous* flows featuring bands of different viscosities and supporting different shear rates. These effects may become crucial in a lot of applications where a good control of the product structure or texture is required.

Moreover, when a complex fluid is confined between two plates and depending on the roughness of the plates, the fluid velocity close to the walls may strongly differ from that of the walls: the fluid *slips*. Such apparent slippage is usually explained by the existence of thin lubricating layers close to the walls in which the fluid structure is very different from that in the bulk. For instance, in concentrated emulsions, apparent wall slip is attributed to the presence of very thin low-viscosity layers where the droplet concentration is much lower than in the bulk (depletion layers) [6].

2.2 Local velocimetry in complex fluids

Accurate time-resolved measurements of *local* velocities are thus crucial to the exploration of complex fluids during processing or quality control through rheological experiments. Since the microstructure of a complex fluid is very sensitive to local deformations, a *non-intrusive* technique is required.

The most popular non-intrusive optical techniques, namely particle imaging velocimetry (PIV), laser Doppler velocimetry (LDV), and dynamic light scattering (DLS), rely on the interaction between light and the microstructure (or seeding particles following the flow). However, many complex fluids, such as emulsions, slurries, or pastes, may not be

transparent enough to allow the use of PIV or LDV. Nuclear magnetic resonance (NMR) offers the possibility to image opaque media [7] but requires the use of powerful magnets and remains expensive and tricky to set up.

On the other hand, ultrasound appears as an efficient, cost-effective tool to measure time-resolved velocity profiles in a large range of fluids. Conventional ultrasound (frequency f= 1–10 MHz) has been used for in-line flow rate and velocity profile measurements [8-10]. However, "low" frequencies suffer from a poor spatial resolution (0.15–1.5 mm) incompatible with the study of complex fluid flows at the mesoscopic scale. This paper introduces high-frequency ultrasonic velocimetry coupled to a standard rheometer as a powerful characterization tool of the shear flow of various complex fluids.

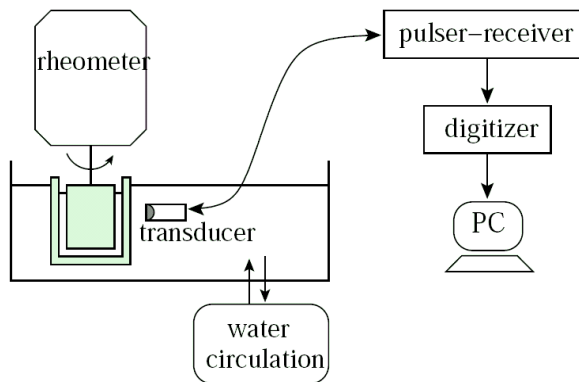## 3 ULTRASONIC RHEO-VELOCIMETRY SETUP

3.1 Apparatus

Figure 1: General sketch of the experimental setup.

Our ultrasonic "rheo-velocimeter" consists of two main components sketched in Fig. 1: (i) a commercial rheometer (TA Instruments AR1000N) that allows us to perform rheological measurements in homemade Couette cells and (ii) a high-frequency ultrasonic velocimetry system. The Couette cell is made of Plexiglas and has the following characteristics: inner radius R_1=24 mm, outer radius R_2, gap e=R_2-R_1=1mm, and height h=30 mm. The walls of both cylinders can be smooth or sand-blasted. Fig. 2 summarizes the notations used in this paper. The whole cell is surrounded by water whose temperature is kept constant to within 0.1 °C. The rheometer imposes a constant torque Γ on the axis of the inner cylinder, which induces a constant stress σ in the fluid, and measures the rotation speed Ω of the rotor. A computer-controlled feedback loop on Γ can also be used to apply a constant shear rate $\dot{\gamma}$ without any significant
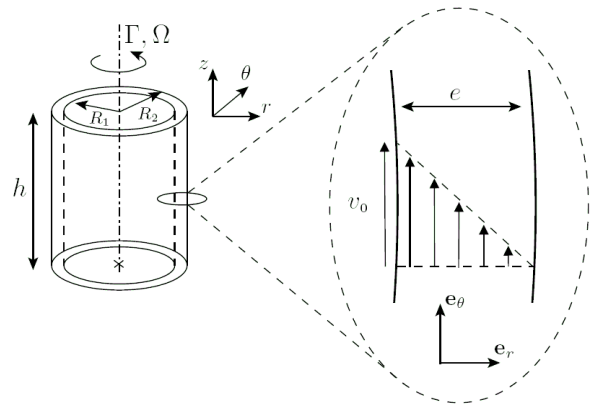
temporal fluctuations.

Figure 2: Sketch of the Couette cell used in our experiments and enlargement of the gap between the two cylinders in the case of a homogeneous flow.

High-frequency ultrasonic pulses are generated by a focused PVDF piezo-polymer transducer of central frequency f=36 MHz (Panametrics PI 50-2). The focal distance is 11.6 mm and the axial and lateral resolutions are 30 μm and 65 μm, respectively over a depth of field of about 1 mm.

The transducer is controlled by a pulser–receiver unit (Panametrics 5900PR) in the transmit-and-receive mode. The pulser generates 220 V pulses with a rise time of about 1 ns. The pulse repetition frequency is tunable from 0 to 20 kHz. The receiver is equipped with a 200 MHz broadband amplifier of maximum voltage gain 54 dB as well as a set of selectable high- and low-pass filters. Ultrasonic signals backscattered by the fluid (see below) are sampled at 500 MHz, stored on a high-speed PCI digitizer with 8 Mb on-board memory (Acqiris DP235), and transferred to the host computer for processing.

3.2 Measurement technique

Ultrasonic pulses are incident on the stator with a given angle $\theta_0 \approx 20°$ relative to the normal to the stator. Pulses travel through Plexiglas and enter the gap with an angle θ that is given by the law of refraction. Once inside the fluid, ultrasonic pulses get scattered by compressibility inhomogeneities that can be either naturally present due to the complex fluid's microstructure (oil droplets in an emulsion for instance) or artificially introduced to enhance the acoustic contrast. The total round trip for a pulse traveling from the transducer to the rotor and back to the transducer lasts about 15 μs. The position of the transducer is tuned so that the 1 mm gap lies inside the focal spot. Typical recorded backscattered signals are 1000 point long, which corresponds to a transit time of 2 μs inside the medium.
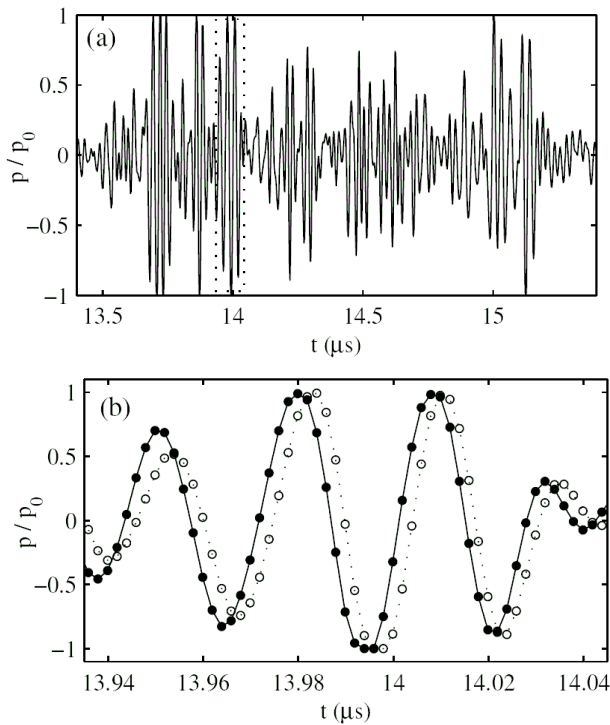
Figure 3: (a) Speckle signal recorded in a sheared Newtonian 1 wt.% suspension of polystyrene spheres of diameter 3–10 μm and normalized by the maximum amplitude p_0. (b) Zoom over four acoustic periods. ●: speckle signal p_T shown in (a) and received after a pulse is sent at time T. ○: signal $p_{T+\delta T}$ recorded over the same time window after a second pulse is sent 1 ms later.

Under the assumption of single scattering, the backscattered signal received at time t on the transducer can be interpreted as interferences of the various echoes from scattering particles located at position $y=c_0(t-t_0)/2$, where y is the distance from the stator along the ultrasonic beam and $t_0 \approx 13.5$ μs accounts for the fixed contribution of ultrasound propagation through water and Plexiglas. By analogy with optics, such a backscattered signal is called a "speckle" signal. Fig. 3(a) shows a typical speckle signal p_T backscattered by a dilute suspension of polystyrene spheres. When the fluid is submitted to a shear flow, the speckle changes as the scattering particles move along. As shown in Fig. 3(b), two successive pulses separated by ΔT lead to two speckle signals that are shifted in time. The time-shift δt between two echoes received at time t corresponds to the displacement $\delta y=c_0\delta t/2$ along the y-axis of scattering particles initially located at position $y=c_0(t-t_0)/2$. The velocity v_y projected along the y-axis, of the scattering particles located at y is then simply given by $v_y=\delta y/\Delta T$.

The spatial resolution of the "ultrasonic rheo-velocimeter" is given by the acoustic wavelength $\lambda \approx 40$ μm. Its temporal resolution ranges between 0.1 and 10 s per profile depending on the shear rate. More details on the velocimetry technique including data storage, the cross-correlation algorithm, its accuracy and limitations, and the calibration procedure used to measure c_0, t_0, and θ can be found in Ref. [11].

4 APPLICATION TO WORMLIKE MICELLE SOLUTIONS

In this section we show a few results obtained using the above-described experimental setup on solutions of wormlike micelles.

4.1 Shear banding in wormlike micelles

Wormlike micelles solutions are self-assembled surfactant systems constituted of long, cylindrical, semiflexible aggregates (a few nanometers in diameter for lengths up to a few microns) [4]. These solutions undergo a shear-induced transition from a viscoelastic state of entangled, weakly oriented micelles to a state of highly aligned micelles above some critical shear rate. Such a transition is strongly shear-thinning, as the viscosity of the aligned state can be orders of magnitude smaller than the zero-shear viscosity of the system.

Above the critical shear rate, the system spatially separates into coexisting bands of high and low viscosities corresponding, respectively, to the entangled and aligned states. As the shear rate is increased, the shear-induced structure progressively expands along the velocity gradient direction at constant shear stress until the system is fully aligned. This phenomenon, known as "shear banding," has triggered lots of experimental and theoretical studies in last two decades [12,13].

4.2 Results from ultrasonic velocimetry

We have used ultrasonic velocimetry to confirm the above scenario that was independently established using NMR [14], DLS [15], and PIV [16]. Importantly, the fast temporal resolution of our technique has provided new insights into the dynamics of shear banded flows. As illustrated in Fig. 4, shear banding may involve strong wall slip, oscillations of the interface position between shear bands, and transient nucleation of three-banded flows [17]. Recent flow visualizations have evidenced an instability of the interface between shear bands along the vorticity direction [18]. These experimental results question the validity of the assumption of purely tangential flows. An interesting issue for future research is to explain the origin of the time scales involved in such temporal fluctuations and their order of magnitude (fractions of a second to minutes). A related question is whether such temporal fluctuations and instabilities are inherent to shear-banded flows and whether they may have some "universal" features regarding other shear-induced transitions.

5 APPLICATION TO FRACTURE IMAGING IN GELS

Ultrasound can not only be used to access time-

resolved velocity profiles but also to perform direct imaging of the material microstructure under shear.

An example is shown in Fig. 5 and 6 on an "organogel" that results from the crystallization of *N-5-hydroxypropyl dodecanamide* in toluene at a temperature of 5 °C. The gel is composed of anisotropic branched fibers of typical diameter 10 µm and length 100 µm. In this case, the microstructure is hard enough to scatter ultrasound efficiently and the fluid does not require seeding [19].
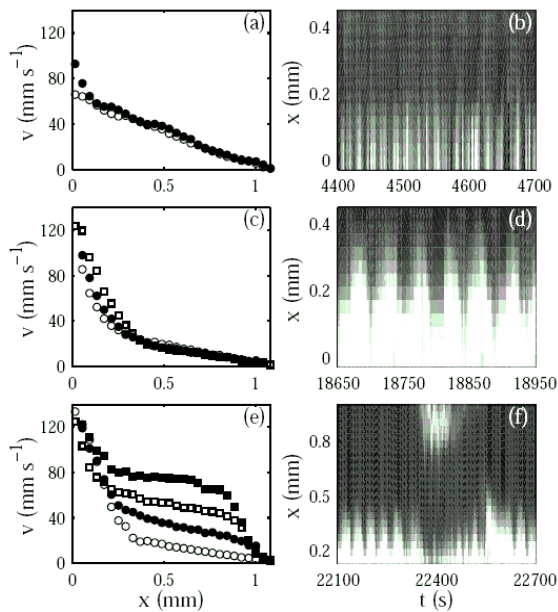

Figure 4: Spatiotemporal behaviors observed in a 20 wt% CTAB solution in D_2O under an imposed engineering shear rate of 186 s^{-1}. (a–b) Intermittent apparition of a highly sheared band at the rotor. (c–d) Oscillations of the position of the interface between shear bands. (e–f) Nucleation of a second highly sheared band at the stator. (a), (c), and (e) present individual velocity profiles, whereas (b), (d), and (f) show spatiotemporal diagrams of the local shear rate inferred from the local velocity measurements. Reprinted from Ref. [17].

The velocity profiles measured in a sand-blasted cell of gap 0.55 mm are strongly inhomogeneous (see Fig. 5(a)): a solid-like region close to the stator extends over about 100 µm. The rest of the fluid is sheared but very large error bars on the time-averaged velocities are recorded in the middle of the gap indicative of huge temporal fluctuations. Such large fluctuations most probably result from *fractures* that occur unevenly in space and time in the central region.

This picture is confirmed by directly looking at the spatiotemporal representation (A-scan) of the ultrasonic echoes shown in Fig. 5(b) and

corresponding to a start-up experiment at a very low shear rate. The solid-like region close to the stator is clearly identified by the fixed vertical echoes. Fractures show up as discontinuities (or even reverse motion) in the ultrasonic echoes, for instance at $t \approx 15$, 18, 27, or 50 s: some parts of the gel locally translate over typically 20 µm within less than $\Delta T = 0.2$ s.
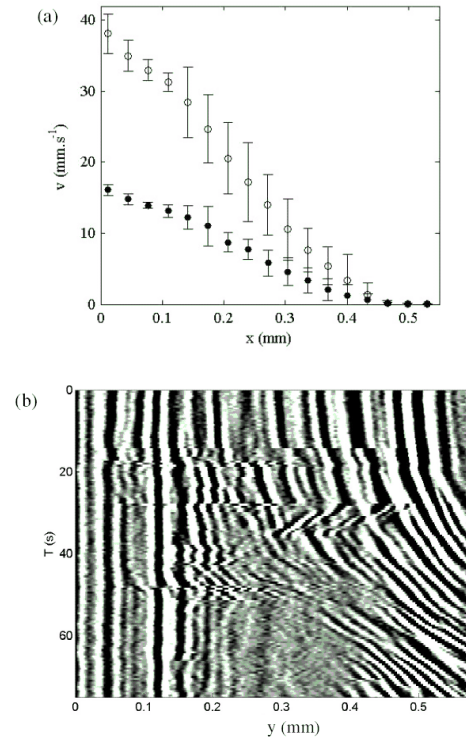

Figure 5: (a) Time-averaged velocity profiles in the organogel at $\dot{\gamma}$=0.05 s^{-1} (⊘) and 70 s^{-1} (o). (b) 50 speckle signals with ΔT=0.2 s recorded during a start-up experiment at $\dot{\gamma}$=0.05 s^{-1} and coded in grey levels. Shear is applied at t=0 s. The stator (rotor resp.) is located at y=0 (y=0.55 mm resp.).

6 CONCLUSIONS

Through the above examples, the ultrasonic rheo-velocimeter appears as a promising tool to perform time-resolved measurements of slip velocities and to study shear localization or fractures in complex fluids during rheological experiments. Such measurements are very important in industrial applications where wall slip, slow transients, temporal instabilities, or strong shear-induced effects are involved.

The main perspectives of the present work concern:

(i) the extension of the technique to *two-dimensional flow imaging* using a transducer array (instead of a single focused transducer) in order to measure the tangential velocity as a function of both radial and vertical positions;

(ii) its application to *small oscillatory displacements*

during rheological experiments in the linear regime in order to recover a spatiotemporal description of viscoelastic properties of complex materials and investigate the crossover from linear to nonlinear behaviours.

ACKNOWLEDGEMENT

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