ADMiER-ing Thin but Complex Fluids

Amarin G M^{C}Donnell, Pradipto K Bhattacharjee, Sharadwata Pan, David Hill, Michael K Danquah, James R Friend, Leslie Y Yeo, Ranganathan Prabhakar

Department of Mechanical & Aerospace Engineering,
Monash University, Clayton, Victoria – 3800

ABSTRACT

The Acoustics Driven Microfluidic Extensional Rheometer (ADMiER) utilises micro litre volumes of liquid, with viscosities as low as that of water, to create valid and observable extensional flows, liquid bridges that pinch off due to capillary forces in this case. ADMiER allows the study fluids that have been beyond conventional methods and also study more subtle fluid properties. We can observe polymeric fluids with solvent viscosities far below those previously testable, accentuating elastic effects. Also, it has enabled the testing of aqueous solutions of living motile particles, which significantly change fluid properties, opening up the potential for diagnostic applications.

INTRODUCTION

We have recently proposed a new device --- the Acoustics-Driven Microfluidic Extensional Rheometer (ADMiER) --- to create valid and observable extensional flows in low-viscosity fluids (Bhattacharjee *et al*, 2011). ADMiER implements a well-known technique for measuring extensional flow properties by observing the dynamics of capillary breakup of thin liquid bridges. Conventional rheometers based on this technique cannot work with samples of viscosities comparable to that of water, since inertiocapillary instabilities cause premature breakup of the filaments. ADMiER overcomes this problem by using surface acoustic waves to create bridges far more rapidly from microlitre-sized samples.

Here, we firstly demonstrate a novel application of this new device for biomedical diagnostics. Recent theoretical models predict that propulsive forces exerted by swimming microbes modify the viscosity of their suspensions. We show here how ADMiER can be used thus to detect and quantify the characteristics of swimming microbes, by comparing viscosity measurements of aqueous suspensions of live and dead cells, and relating them to motility analysis by microscopy.

Extensional flows are excellent for probing the elastic response of complex fluids. We show how ADMiER can serve as a research tool by using data obtained with this device to discriminate between several competing theoretical models for the viscoelasticity of dilute polymer solutions. With ADMiER, we can work with thin solvents such as water or toluene, in which many model parameters such as the size of the dissolved polymer molecules are accessible from other measurements. This enables a priori independent estimation of all model parameters and allows for unequivocal tests of models.

METHODS

The problem with low-viscosity fluids in extensional flow

Though shear flow experiments have been around for over one hundred years (Couette flow, 1890) rheometric extensional flow experiments have only really been around since the 1990's. Creating valid and controlled extensional flow has always been a challenge for rheologists and has only seen success with the development of

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techniques like the Filament-stretching extensional rheometry (FiSER; Tulahar & Mackley, 2008) and capillary break-up extensional rheometry (CaBER; Rodd *et al,* 2005) techniques; both of which involve creating thin filaments (or filament) from sample drops.

In FiSER, a liquid sample is placed in-between two end-plates and the plates are then moved apart at a controlled rate. In CaBER, on the other hand, the two end-plates plates are separated rapidly to a fixed distance, and then held stationary, while the resulting liquid bridge is allowed to neck and break-up under capillary forces. In either case, the tendency of the cylindrical column of liquid to neck due to capillary stresses is resisted by viscous, and in the case of viscoelastic fluids, elastic stresses. The evolution of the profile of the thinning filament with time is governed by these forces. Flow at the middle of a thin filament is uniaxial extension and the local strain rate at middle of filament can be related to change in filament radius. The mid-filament equation (Anna and McKinley, 2001) connects extensional strain-rate and radius through a balance of forces on the filament and thus infers the stresses at the middle of the filament. Both these techniques have found successful application with different types of fluids, but the study of polymeric fluids has received particular attention.

The major drawback with both of these techniques, however, is that their mechanical operation can induce perturbations, which can seed inertiocapillary instabilities and hasten its break-up. Further, CaBER requires the initial end-plate opening time to be as short as possible, but current designs are limited to opening times greater than about 50ms. The opening time needs to be lower than the viscous breakup time $t_v \geq \delta t_o$ to avoid break-up before the liquid bridge is fully realised (Rodd *et al,* 2005). Therefore, given the viscous time is $t_v = 14.1\eta R_0 / \gamma$ (14.1 factor as seen in Rodd *et al,* 2005) where η is viscosity, R_0 is initial filament radius, and γ surface tension, we are limited to 70 mPa·s (using the nominal experimental values given by Rodd *et al* (2005)). The above reasons make fluids with a viscosity less 100 mPa·s difficult to analyse, thus aqueous solutions are beyond the range of these techniques; thus, no observations of extensional behaviour with CaBER or FiSER have been reported for liquids, without a polymer component, with shear viscosities less than 10mPa·s. This is of particular concern to this study as microorganism suspensions are typically in low-viscosity (~ 1 mPa·s) aqueous buffers. Attempting to increase the viscosity of these suspensions for testing with additives may affect biological activity and even halt a microorganism's motility altogether.

Furthermore, relatively large sample sizes are required for both techniques (D ≈ 6mm). The Bond number $Bo = \rho g R_0^2 / \gamma$, where ρ is density and g is gravity, characterises the gravitational effects versus surface tension. A larger sample size will thus result in gravitational sagging of the filament and asymmetric fluid flow in the filament around the mid-filament region. Larger sample sizes also subject the experiment to more dominant inertial effects, which are of particular concern when the mechanical vibrations produced in these systems is considered.

ADMiER

Creating valid extensional flow of aqueous (thus low viscosity) fluids is the crucial issue at hand in this study. As previously mentioned, conventional extensional flow techniques that measure mid-filament stress and strain have several drawbacks that make them unsuitable for low viscosity applications; their comparatively slower opening time lower-limits mean that filaments can break-up before a stable liquid bridge is created, larger sample sizes of low-viscosity fluids are prone to result in asymmetric liquid bridges and result in inertiocapillary instability, and the mechanical nature or their operation can produce perturbations that disrupt filament decay.

We have developed a technique, ADMiER, which addresses the above issues in order to make low viscosity fluids accessible to stress and strain measurement in extensional flow (Bhattacharjee *et al*, 2011). It combines the concept of CaBER with the novel idea of using a burst of surface acoustic wave (SAW) energy to create the liquid bridge. The technique's opening time to create a liquid bridge is an order of magnitude faster than its mechanical counterparts (>5ms as opposed to ~50ms), the diameters of the necessary sample sizes are less than half (>3mm as opposed to 6mm), and the use of SAW energy negates the need for any mechanical components. These qualities allow us to create valid extensional flows of viscosities down to the order of magnitude of water and collect data reliably.

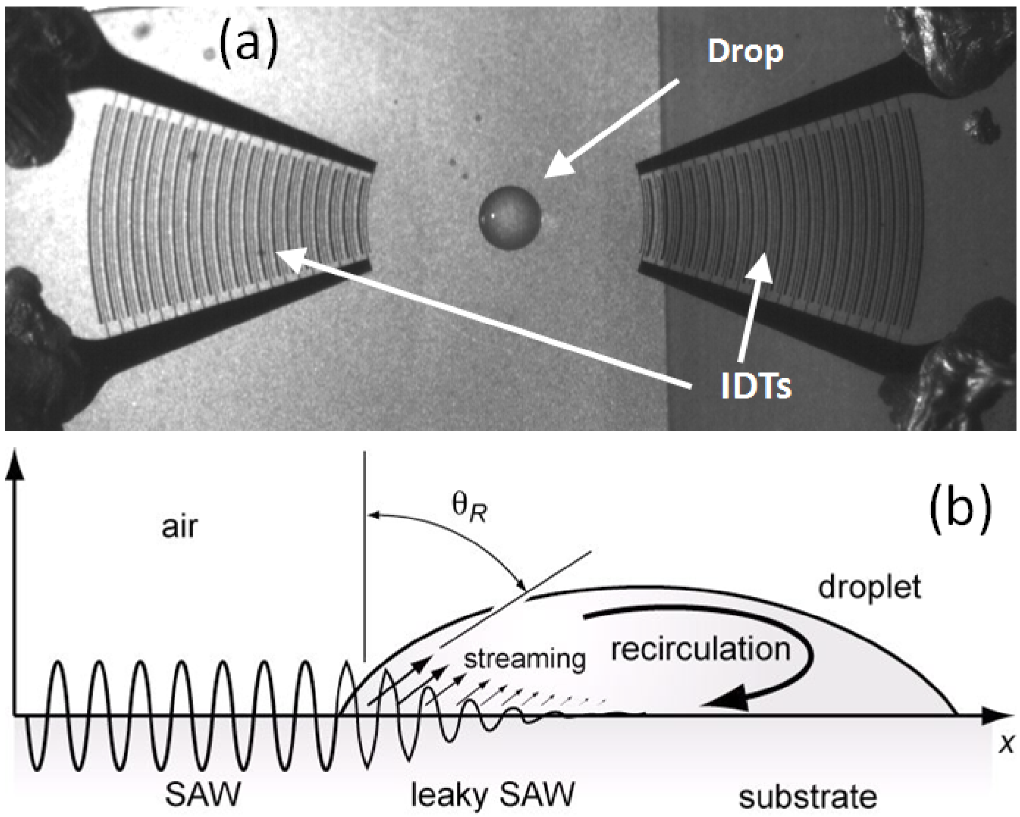

Fig. 1 (a): Photograph of a drop at the focal point of curved SAW IDTs (b) Diagram (Yeo and Friend, 2009) illustrating how SAW energy enters a droplet at the Rayleigh angle, θ_R, and causes recirculation.

An arrangement of inter-digitated transducers (IDTs) is bonded to a piezoelectric substrate (lithium niobate in this case). They act as capacitors and an applied voltage will cause the piezoelectric substrate to produce Rayleigh waves that travel across the surface; these are the surface acoustic waves. Typical SAW surface velocities and frequencies upon a piezoelectric surface, such as lithium niobate, are in the order of 0.5m/s and 37MHz, respectively, but can vary significantly due to IDT configuration and other factors. As is clearly seen in (Fig. 1 (a)) the IDTs curve to a focal point thus concentrating the SAW energy in the centre of the device.

When the SAW energy thus focused enters a sessile droplet resting upon the substrate surface, the difference in the speed of sound between the substrate and the liquid causes the SAW energy to leak into the fluid at a specific angle, the Rayleigh angle θ_R (see Fig. 1 (b)). This results in a net transfer of momentum into the drop, which has been modeled as an additional body force on the droplet itself in the direction of the SAW (Yeo & Friend, 2009). A bulk liquid recirculation (acoustic streaming) is produced within the droplet which can cause a range of behaviours of the drop, (Bhattacharjee *et al*, 2011); but it is this technique's ability to manipulate a droplet into an elongated jet (jetting) that we exploit; with this ability we create liquid bridges rapidly for use in the ADMiER device. The operation of the ADMiER involves delivering a sufficient burst of power to the sessile droplet, upon receiving the SAW energy bulk motion will occur and the droplet will form a jet that spans across (with gravity) to the opposing plate. The burst of SAW energy ends after the liquid bridge is formed and capillary forces are left to collapse the liquid bridge. SAW power and burst length can be adjusted to accommodate different fluids of varying properties.

Successful liquid bridge formation and therefore filament break-up are extremely sensitive events; excessive power can lead to disruptive oscillations, too little power can lead to inadequate wetting of the opposing plate, too close a distance and a permanent liquid bridge can form, too far and the jet will form droplets without even creating the liquid bridge, these are just some of the problems that can be encountered. Hence, the ability to reliably control the distance between the SAW device and the opposing plate, to ensure that both surfaces are parallel, and to very importantly ensure that experiments are repeatable required a purpose built rig (Fig. 2).

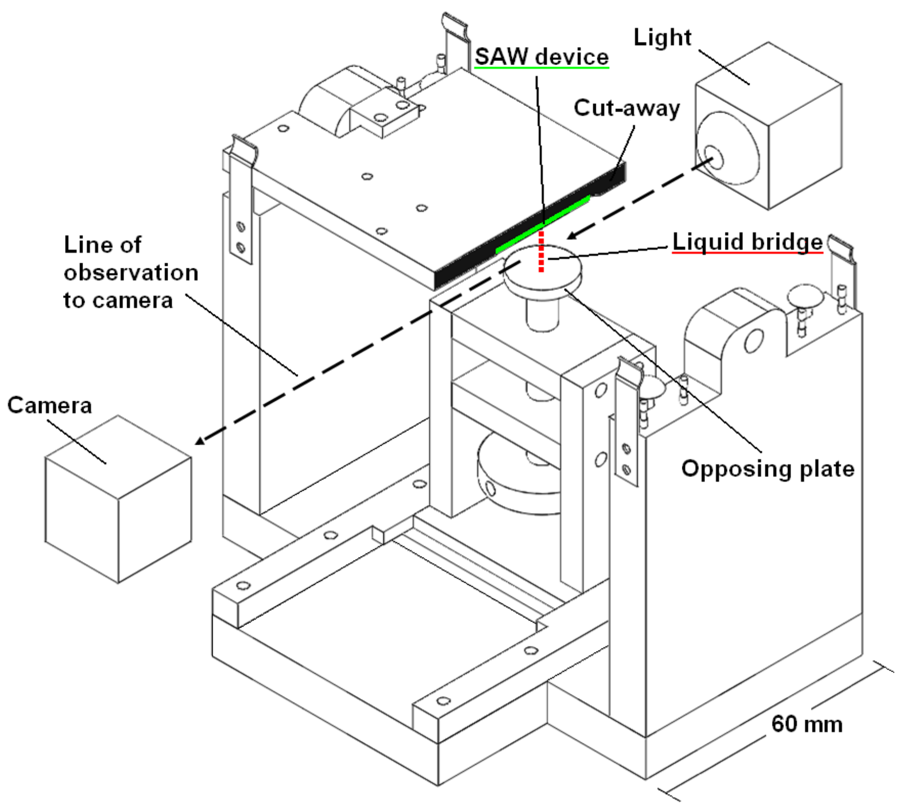

Fig. 2: CAD image of the experimental rig and set up that allows for mounting of SAW device, parallel alignment of device surface and opposing plate and high-contrast high-speed video to be obtained.

Polymer solutions

Solutions with polymer additives experience an increase in tensile stresses as polymer chains are elongated near filament break-up; these elastic stresses resist the pinching capillary stresses and result in a slowed filament break-up rate near pinch-off. In this study we tested the technique's abilities to observe the change in exponential decay as is expected to be observed in polymer solutions (Anna and McKinley, 2001) using a 1.95 Mw polystyrene(PS)/dioctyl phthalate solution (DOP).

Active particle suspensions

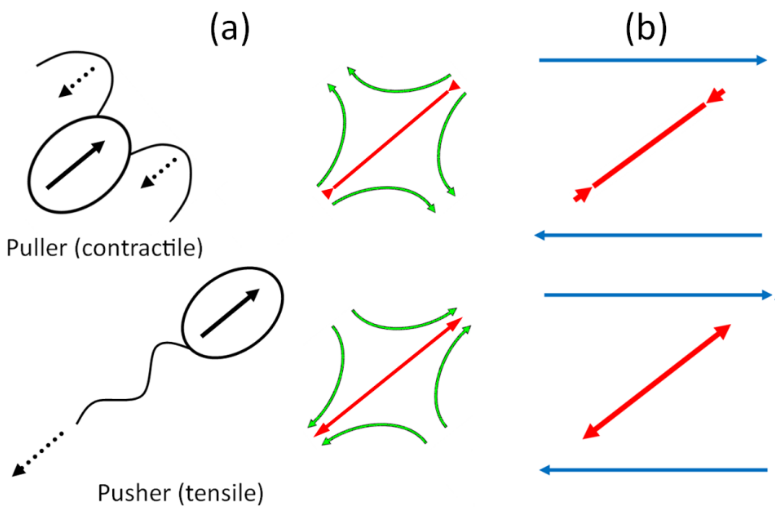

Fig. 3: (a) A puller microorganism performing a "breast-stroke" and a pusher microorganism effecting its swimming movement using its polar flagella; immediately to the right are shown the corresponding flow fields (green arrows) generated by the local hydrodynamic force dipoles (red arrows). (b) Diagram depicting how shear flow, blue arrows, is resisted by contractile activity vectors (red arrows, top) and how it is strengthened by tensile activity vectors (red arrows, bottom).

The technique has also been used to differentiate between fluids containing self-propelling particles. The theoretical approach to this developed by Saintillan, 2010, idealises particles as small hydrodynamic dipoles. The instantaneous orientation of each particle defines both the directions of the hydrodynamic dipole, and its motion. The hydrodynamic dipoles set off flows in the ambient fluid which then perturbs the orientation and motion of surrounding particle dipoles. These hydrodynamic interactions are believed to be responsible for the variety of bulk behaviours that seem to be common to different forms of active matter, such as vortices, jets, and flow instabilities (Koch & Subramanian, 2011). Dipoles are further classified as being "contractile" and "extensile". In the context of rod-like particles, as depicted in Fig. 3, contractile particles draw ambient fluid inwardly along their principle axis and expel it perpendicular to their principle axis; conversely, extensile particles will draw fluid inwardly perpendicular to their principle axis and expel it along their principle axis. Such particles are also referred to as "pullers" and "pushers", respectively.

In the case of active suspensions, one way the effect of pullers and pushers on a fluid is predicted to manifest itself is in a suspension's viscosity (Saintillan, 2010). Subjecting an active fluid suspension to a simple shear flow results in a mean alignment of the principle axis of the particles with the stretching direction of the flow. Given this alignment, a suspension of pullers will contract flow against the principal stretching direction of the shear (as described in (Fig. 3 (b), top), increasing the stress/strain rate ratio, and thus increasing the fluid's viscosity. On the other hand when pushers are aligned (Fig. 3 (b), bottom) with this flow they will push in the direction of the imposed flow, enhancing it, and thus reducing the fluid's viscosity. It is this effect on fluid viscosity that we wish to investigate. In order to do this we require model systems to test. We chose live and dead algal (*Dunaleilla tertiolecta*) and bacterial (*E. coli*) suspensions to be our puller and pusher examples, respectively. Importantly, it must be said that these suspensions are aqueous and thus beyond the range of the mechanical techniques aforementioned.

RESULTS AND DISCUSSION

Polymer

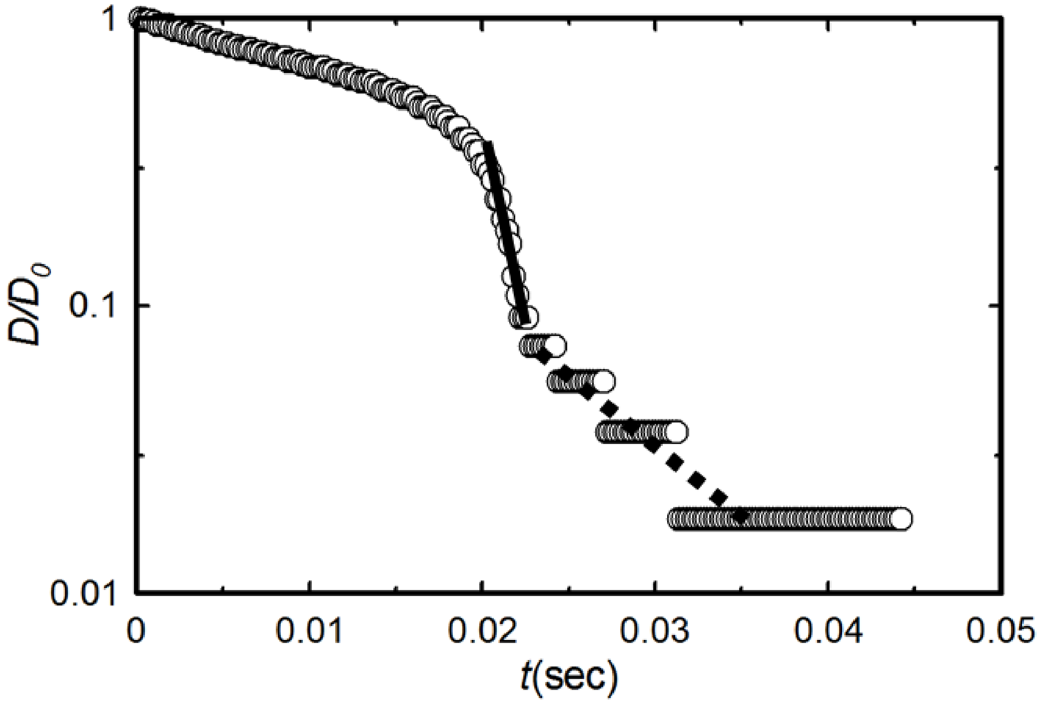

Fig. 4: PS/DOP solution, here we can see two distinct trends in the filament's evolution beginning with the linear trend of the Newtonian response (solid line) and then transitioning to the exponential trend (dotted line) as stresses induced by the elongation of the polymer chains begin to take effect

The transition from the linear Newtonian regime to the exponential regime caused by the elasticity of the polymers within the solution is clearly visible in Fig. 4 as is expected. From results like this, one can extract a fluid's extensional properties, such as extensional relaxation time, as is described in Bhattacharjee *et al*, 2011.

Active particle

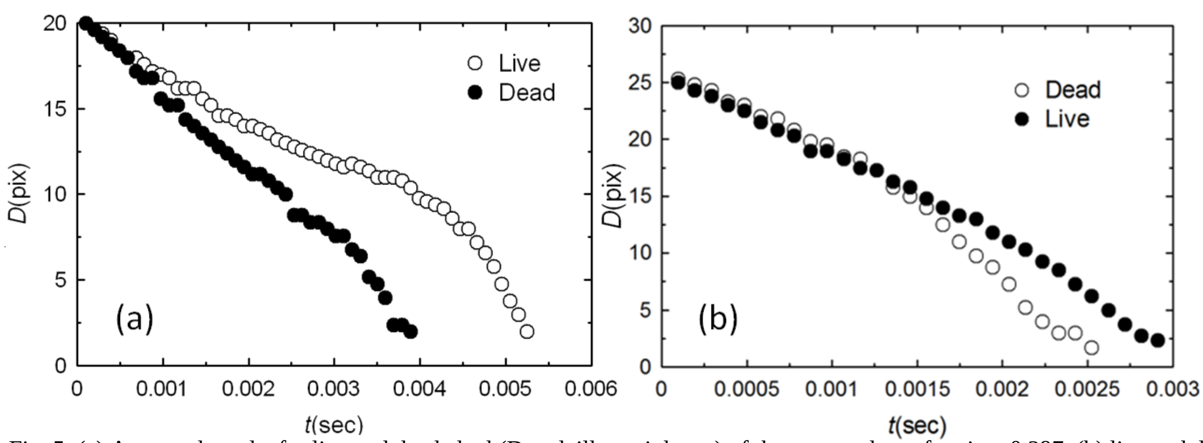

Fig. 5: (a) Averaged results for live and dead algal (Dunaleilla tertiol.ecta) of the same volume fraction, 0.297. (b) live and dead bacterial (E. coli) averaged results same volume fraction 0.351

For a given volume fraction, our puller system clearly exhibits a lengthened filament decay for the live sample, as can be seen in Fig. 5 (a), in other words an increase in viscosity is demonstrated. From the results of our pusher system seen in Fig 5 (b), however, we can see that viscosity is apparently decreased by the presence of live pusher particles. These results agree with theoretical predictions.

CONCLUSION

Rheological extensional flow investigation of fluids that were out of the reach of previous methods has been opened up by ADMiER. The technique has been qualitatively validated against established extensional rheometry techniques with a range of fluids (Bhattacharjee *et al*, 2011). Observing the results seen in Bhattacharjee *et al*, 2011, and in this paper we can see that this technique allows for rheology using extensional flow to be applied to fluids and indeed fields it was unable to explore before. It's the system's lack of mechanical parts, its small required sample volumes, and speed of operation that makes available a range of low-viscosity fluids that could not previously be explored. This technique is envisaged to have applications with biological fluids, printer inks, and industrial sprays like pesticides and paints.

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