**Elastic instabilities in parallel shear flows of a viscoelastic shear-thinning liquid**

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**Abstract** We report the results of an experimental study of the fully-developed flow of a viscoelastic, shear-thinning aqueous polymer solution through two large-scale parallel-shear flows: a pipe and channel-flow facility. We show that, at low flowrates, the velocity profile in both geometries is steady and is in good agreement with expected analytical solutions. However in both geometries at higher flowrates the flow becomes weakly time-dependent and the mean velocity profile is radically altered, exhibiting an overshoot near the duct centre, a reduced shear rate at the wall and an inflection point (i.e. a non-monotonic gradient). Although inertia is not completely negligible at instability onset, we speculate that the origin of these unstable flows lies in the combination of elasticity and strong shear thinning.

In this rapid communication we present experimental results from two canonical parallel shear flows – a fully-developed pipe and a channel flow – of a prototypical shear-thinning viscoelastic liquid (an aqueous polymer solution). One might imagine, perhaps naively, such simple flows to be rather benign and well understood. Once fully developed, the effects of fluid memory necessarily vanish as must, for straight constant-cross-section conduits, any effects of the elastic first normal-stress difference (*N*1) (which essentially can play no role in the macroscopic momentum balance and therefore cancels out). For such steady viscometric flows the velocity distribution should be solely governed by the steady-state shear viscosity of the solution [1], and in the limiting case of axisymmetric geometries – or planar geometries of infinite width – the precise form of the elastic second normal-stress difference (*N*2) is also unimportant and thought to play no role [2]. At low flowrates we confirm this viewpoint here. However, our results show that at higher flowrates instability arises and the flow becomes weakly time-dependent. Of course, much as for Newtonian and “inelastic” shear-thinning fluids, at sufficiently “high” Reynolds number (Re, defined below), a subcritical transition to classical inertial turbulence [3] is observed for such polymeric systems for Re in the range ~2,000-10,000 [4] (in pipe flow, being linearly stable for both Newtonian and simple inelastic shear-thinning models [5], this upper limit can in fact be much higher). What is observed here is at significantly lower levels of inertia and different in form to the mean-flow modification observed in classical turbulent flow [3-4, 6-7], thus what we describe here is distinctly different to classical inertial turbulence.

Although, in the zero inertia limit, constant-viscosity models of viscoelastic fluids predict parallel channel flows to be linearly stable [8], non-linear stability approaches [9] and microfluidic experiments show, with sufficiently large perturbations, these flows can be driven unstable in a sustained fashion [10]. The phenomenological picture here is that these perturbations are sufficiently large to locally curve the streamlines (usually considered a necessary condition for the onset of a purely-elastic instability [11-13]).

One study which has predicted a linear instability in such parallel-shear flows is that of Wilson and Rallison [14] who investigated shear-thinning elastic fluids in a channel with the White-Metzner model assuming a constant elastic modulus (G=η/λ where η is the shear viscosity and λ the fluid relaxation time). Instability, at Weissenberg number (Wi, defined below) ~ 2, was seen when the shear thinning was sufficiently strong e.g. shear-thinning index and and where subscript 1 implies the value at 1 s-1. (NB. For constant elastic modulus, the shear viscosity and relaxation time have to have the same functional dependence of the shear rate). Recently Bodiguel et al [15] demonstrated an elastic instability in a microfluidic channel flow for a concentrated polymer solution. In this case the combination of strong shear thinning of the viscosity coupled with elasticity was believed to be the root cause of flow instability which was found to occur at approximately a constant Weissenberg number Wi~5±3 (actual range 3-10). Although such results are in qualitative agreement with the earlier stability analysis of Wilson and Rallison [14], the base “laminar” profiles of Bodiguel et al [15] exhibit very large slip, being essentially plug-like across the channel. Subsequently, a supplementary linear stability analysis with a modified White-Metzner model [16] with non-constant G (i.e. as in the experiments of [15]) showed that such fluids are slightly more unstable (Wi ~ 1.8) in channel flow than the earlier constant modulus results of Wilson and Rallison [14].

Here we provide new experimental evidence for an elastic instability of the type postulated theoretically by Wilson and co-workers [14,16] and observed experimentally by Bodiguel et al [15]. In contrast to the microfluidic study of Bodiguel et al [15], where wall slip was a significant factor in all of the base velocity profiles, we provide data in macro-sized geometries (~25-100mm) where the base laminar profiles are in excellent agreement with simple analytical solutions (based on the “power-law” model of viscosity discussed above). In addition, we are able to capture the mean velocity flow profiles using a high resolution Laser Doppler Velocimeter (LDV) technique and quantify the significant modification to this base profile which occurs beyond instability onset. The LDV technique also enables spectral information to be provided and is sufficiently spatially-refined such that the profile can be differentiated to determine directly the shear rate (and hence shear stress in the steady-flow cases) profile across the ducts. Furthermore, by providing data in both a channel and a pipe, we are able to unequivocally remove the possibility of *N*2 playing a significant role.

The experimental pipe and channel flow rigs have been described in detail elsewhere [6-7] and therefore only the briefest information is provided here. Although completely individual facilities, both rigs are built around a common architecture. The fluid is driven from a 500-l capacity stainless steel tank by a progressive cavity pump (Monotype, E101 with a maximum mass flowrate  of approximately 25 kg/s). Three pulsation dampers located immediately after the pump outlet act to remove any potential unsteadiness resulting from the rotation of the monopump. A Coriolis mass flowmeter (Promass 63, manufactured by Endress +Hauser) is also incorporated into each experimental rig. The pipe rig comprises of 21 precision-bore borosilicate glass tubes, each about 1029±2mm long, a single glass tube 656 mm in length and one PVC plastic pipe, 1060mm long, at the test section entrance. The internal diameter (D) of the tubes is 100.4±0.1mm and measurements are taken approximately 460 pipe radii (R=D/2) downstream of the entrance, ensuring fully-developed conditions. The channel flow geometry has an overall length of 7.45 m, a width of 298 mm and a channel half height H=12.5 mm. Measurements were taken approximately 600 H downstream of the inlet, again ensuring fully-developed conditions. Mean streamwise velocity profiles and fluctuation levels were measured using a Dantec Fiberflow LDV system comprising of a 60X10 probe and a Dantec 55X12 beam expander in conjunction with a Dantec Burst Spectrum Analyzer signal processor (model 57N10).

The polymer used is a polyacrylamide, Separan AP 273 E supplied by Floerger (hereafter “PAA”), which is regarded as having a highly flexible linear molecular structure and so is strongly viscoelastic, at a nominal 1% (w/w) concentration in water (total volume of fluid in each experimental rig ~700 l). Measurements of the flow curves (shear viscosity η vs shear rate ) and first normal-stress difference were carried out with a TA Instruments AR1000N controlled-stress rheometer using a cone and plate geometry (60mm diameter, 2° cone angle). The intrinsic viscosity [η] was determined to be 4400 ml/g using a Cannon-Fenske u-tube viscometer and the critical overlap concentration c\* is estimated to be c\* = 1/[η] ~225 ppm: therefore our solution can be considered to be highly concentrated/entangled as c/c\* ~30. As shown in Fig. 1(a), the solution strongly shear thins, decreasing from ~100 Pa.s at low shear rates to ~0.01 Pa.s at the highest shear rates where data was obtainable. Over a very large range of shear rates the viscosity data is well fit with a power-law model  where =9.88 Pa.s0.19 for the pipe and =6.00 Pa.s0.19 for the channel. The blue line in Fig. 1(a) represents a Cross model [1] fit to the data with where =219 Pa.s, =1.08 mPa.s,= 38.5 s-1 and n1=0.834 for the pipe and =209 Pa.s, =1.41\*10-2 Pa.s,= 67 s-1 and n1=0.800 for the channel. The approximate range of the wall shear rates obtained in the pipe and channel flow experiments are also highlighted in Fig. 1(a) confirming that the experiments lie nicely in the power-law range. The first-normal stress data, plotted as both the first normal-stress difference coefficient () and as a relaxation time () are shown in Fig. 1(b). A power-law fit to the relaxation time data (where =2.8 s0.32 and m =0.68 for the pipe and =2.0 s0.36 and m = 0.64 for the channel) confirm that the fluid does not exhibit a constant modulus but that it weakly shear thins as  where p = 0.13 for the pipe and p = 0.17 for the channel. To ensure a well-mixed solution required mixing in the rig at a very low pump speed for a period of about one month. Intermittent rheological sampling of the fluid was used to determine homogeneity. Given the nominal nature of the concentration (i.e. exact fluid volume of the rig has to be estimated) there were slight rheological differences between the mixed fluids in each rig, as a consequence fit information is provided for both fluids but rheology data only shown for the pipe data for clarity.

 To quantify the importance of inertial and elastic forces we define characteristic Reynolds (ReCH) and Weissenberg (WiCH) numbers: they are “characteristic” values as we use the shear-rate dependent viscosity (ηCH) and relaxation time (λCH) determined at a shear rate =Ub/L where Ub is the average or bulk velocity in the pipe or channel and L is a length scale which for the channel is the half height H and, for the pipe, the radius R. The Reynolds number is then ReCH=ρUbL/ηCH and the Weissenberg number WiCH=λCHUb/L. In the channel flow experiments (0 <  < 11 kg/s), 0 < ReCH < 150 and 0 < WiCH < 11 and in the pipe (0 <  < 11 kg/s), 0 < ReCH < 105 and 0 < WiCH < 8. (NB. Defined using the zero-shear rate viscosity (~200 Pa.s), the Reynolds numbers would appear vanishingly small in all cases). Thus it needs to be recognised that although inertial effects are small, they are not completely negligible. Weak inertia was seen to be destabilising in recent related linear stability calculations [16].

In Fig. 2 we show results from the channel-flow facility and, in Fig. 3, from the pipe. Broadly, the same trends are observed in both geometries highlighting the robustness of our observations. At low flowrates the mean streamwise/axial velocity profiles, shown in Fig. 2(a) and Fig. 3(a) with open symbols, are in excellent agreement with analytical solutions based on the power-law model shown with the continuous red lines (expressions for which are not repeated here given they are available in many standard rheology/fluid mechanics textbooks e.g. [1]). Finite-volume numerical calculations of the Cross model at the same conditions show minimal differences to the simpler power-law model (not shown). As the rheological data is very well fit by a power-law model, plotted in non-dimensional form all velocity profiles should collapse regardless of flowrate (which we confirmed again numerically for all flowrates studied). However in both the channel and the pipe we observed that this flowrate-independent profile was only maintained up to some critical flowrate (for the channel ~6 kg/s corresponding to WiCH ~9 and for the pipe ~4 kg/s corresponding to WiCH ~6). Beyond this critical flowrate, a systematic change to the mean velocity profile was observed in combination with an increase in the measured velocity fluctuation levels.

The mean velocity profile which emerges at the highest flowrates exhibits the same features in both geometries: presenting an overshoot in the centre of the duct, a reduced shear rate at the wall in comparison to that expected from the power-law model and an inflection point at some distance from the wall (~0.1-0.2 L). This inflection point is illustrated in Figs 2(b) and 3(b) where the shear rate profile is determined by differentiating the mean velocity profiles using central differences. It is important to note that the form of the mean velocity modification is distinctly different to that observed in Bodiguel et al [15] where the plug-like flow (blamed on slip due to wall depletion) is replaced beyond onset instability by a more parabolic profile, seemingly monotone in gradient and where the largest velocity fluctuations occur in the centre of the channel. Whether such differences are driven by the differences in base profile (slip [15] versus no-slip here), for example by the instability arising from the slip layer itself, or by differences in the levels of inertia (essentially inertialess [15] versus modest inertia here, Re~50 at onset) presents an open question which potentially may be answerable by full non-linear simulations of viscoelastic constitutive models which include significant shear-thinning effects.

In both the results observed here and those in Ref. [15] the onset of instability leads to a drag reduction: here we observe a reduction in the wall shear rate from that expected from the power-law model of about 35-40% for both geometries at the highest flowrates. Given the extremely shear-thinning nature of the solutions this only results in a small reduction in the wall shear stress compared to that expected from the power-law model as ~ 0.60.19 ~ 0.92 i.e. a 40% reduction in wall shear rate only results in an 8% reduction in wall shear stress. This mean flow modification is associated with the onset of weak time dependency in the near-wall region as shown in Fig. 2(c) and (d) where a systematic increase in this quantity above the LDV noise level (~2%) can be observed. In Fig. 2(c) these fluctuations can be seen to be confined to about the first 50% of the channel half height and in Fig 2.(d) beyond onset they can be seen to grow continuously with flowrate at a fixed wall-normal location (y/H=0.25) before saturating at higher flowrates. Similar observations were observed in the pipe flow geometry. Much as with the mean flow, the fluctuations are different in form to those observed in Ref. [15] where they are concentrated in the channel centre (whereas in the current flows at the duct centres the flow appears essentially steady).

Measurements at different streamwise locations (e.g. Fig. 3(c)) – only possible in the pipe geometry because of a lack of optical access in the channel – confirmed that development length effects are not significant. Given the viscoelastic nature of the solutions, this invariance with streamwise distance suggests these changes to the mean flow profile are not a consequence of the fluid “remembering” an instability arising from the complex shear and extensional flow at the inlet.

Well beyond onset, the velocity fluctuations – shown in Fig. 2(e) – are non-periodic, approximately collapse and exhibit a broad-band frequency response over two orders of frequency (scaling like f-4/3) indicating that the flow is excited at many time-scales. By contrast, the low flowrate case exhibits a relatively flat power spectra consistent with noise. Closer to instability onset (~ 6 kg/s), no distinct peak can be observed suggesting that non-linear effects rapidly become important. For both flows, the increase in fluctuation level appeared quite gradually with flowrate and the “critical” flowrate did not apparently exhibit any hysteresis. However, given the relatively weak level of fluctuations observed here, increasing from a noise level of ~2% to a maximum ~4%, it is quite difficult to precisely define a “critical” flowrate and some small level of hysteresis may be occurring.

For a range of less elastic, but still significantly shear-thinning fluids, the mean axial velocity profiles in these two facilities [6-7,17] at “low” Re (on the order of 103 say) have been found to be in excellent agreement with the expected power-law analytical solutions e.g. for 0.2% PAA up to at least Re =3840 [17], for a 0.25% carboxymethylcellulose solution up to at least Re=1790 [18] and for a 0.14% Carbopol solution up to at least Re=1970 [18]. Thus, we do not believe that the effect observed here is due to shear-thinning alone. For the pipe, beyond these Reynolds numbers, an intriguing mean-flow asymmetry [17-20] develops, apparently for all shear-thinning fluids, but the effects observed here are far removed from that both in form and parameter space (we confirmed that the present results always exhibited symmetry both in the pipe and the channel although only half profiles are shown). When fully turbulent flow does occur, say Re > 10,000, the effect on the mean velocity profile is the exact opposite of what is observed here, i.e. the mean profile in turbulent flow is flatter. In addition, as is well known, classical turbulence leads to a drag increase whereas we observed a drag reduction here. Finally, the instability observed here also seems distinct from that observed for a Boger fluid in a microfluidic channel [10] where the fluctuations are largest close to the channel centreline, the mean velocity profile remains parabolic, and very strong inlet perturbations are required to locally curve the streamlines. Therefore the experimental evidence suggests, coupled with the existing results in the literature [10, 14-16], that the effects observed here are novel, apparently unrelated to classical inertial transition and are due to a combination of *both* elasticity and shear-thinning.

In summary we have reported experimental results in two parallel-shear flows of a highly shear-thinning viscoelastic polymer solution. At low flowrates, the mean velocity profiles are steady and are in excellent agreement with those determined analytically from steady-shear rheology data. Beyond a critical flowrate, the mean velocity close to the wall becomes weakly time dependent and the magnitude of this fluctuation grows with increasing flowrate until saturating at higher flowrates. The effect of these fluctuations on the mean flow is actually quite significant: resulting in an overshoot of velocity close to the duct centre, a reduced wall-shear-rate and a non-monotonic velocity gradient distribution. The reduced wall-shear-rate in effect means that the instability results in a genuine drag reduction – i.e. a reduced wall shear stress or pressure-drop is required to drive a given flowrate of fluid than would otherwise be expected – although the actual level of drag reduction is actually rather modest due to the strongly shear-thinning nature of the solution. Frequency analysis of the velocity fluctuations indicate a broad-band response, even close to instability onset, suggesting a weakly turbulent state.

Finally, although the LDV technique used here has revealed the appearance of an instability, more sophisticated instrumentation may allow more insight into the physical mechanisms involved to be gained. For example, time-resolved stereoscopic PIV [20] could reveal the effect on the radial and tangential velocity components and potentially allow velocity gradient terms to be estimated. Ideally this would be coupled with time-resolved measurements of the stress field to determine if the mean-flow modification is responsible for the drag reduction i.e. by advecting/mixing regions of low/high viscosity fluid such that the overall stress experienced by the fluid is decreased.

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| (b)(a) |  |

**Figure 1:** Steady-shear rheology for 1% PAA in the pipe. (a) Viscosity versus shear rate including power-law model fit (n=0.19 red line i.e. ), Cross model fit (blue line) and indicative wall shear rates of shear-flow experiments; (b) first normal-stress difference coefficient (open symbols) and relaxation time (closed symbols) versus shear rate including power-law fit (). The power-law fits imply a non-constant elastic modulus . {100 words for single column figure + ~65 words caption}

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**Figure 2:** Channel-flow experimental results. (a) Mean velocity distribution; (b) shear rate variation; (c) velocity fluctuation distribution normalised by local mean velocity (u); low flowrate (open symbols, 3 kg/s, *Re*CH = 14, *Wi*CH= 7) and high flowrate (8.3 kg/s, *Re*CH = 88, *Wi*CH= 10) including channel-flow analytical solution for the power-law model (n=0.19, red line). (d) variation of velocity fluctuation level with flowrate at fixed wall normal location (y/H=0.25) normalized by both the bulk velocity (blue symbols) and by the local mean velocity (red symbols) (in both (c) and (d) horizontal dashed line (- - -) indicates approximate noise level in the LDV system); (e) velocity power spectra density variation with flowrate at y/H=0.25 (Δ 3 kg/s, ∇ 5.96 kg/s, ∇ 7.8 kg/s, ▲ 8.3 kg/s, Δ 9.5 kg/s) solid line shows power-law fit with exponent -4/3. {770 words for double column figure + ~140 words caption}

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**Figure 3:** Pipe-flow experimental results. (a) Mean velocity distribution; (b) Shear rate variation; low flowrate (open symbols, 1 kg/s, *Re*CH = 1.4, *Wi*CH= 3.8) and high flowrate (10 kg/s, *Re*CH = 89, *Wi*CH= 8.0) including pipe-flow analytical solution for the power-law model (n=0.19, red line). (c) mean velocity profile at different axial locations from inlet highlighting fully-developed nature of the flow at 10 kg/s (blue 230 D from inlet, red 100 D from inlet). {480 words for double column figure + ~75 words caption}