Transient, near-wall shear-band dynamics in channel flow of wormlike micelle solutions

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Abstract – We report the spatiotemporal dynamics of shear bands in wormlike micelle solutions as generated in the near-wall region of rectangular channel flow. In this system, a very thin ($< 2 \mu\text{m}$) high shear rate band is generated, stacked along the gradient axis of the channel flow. By means of confocal microscopy and particle image velocimetry, we observe the effect of this thin band on both the steady-state flow profile and the transient variation in the flow-direction velocity, which is found to be non-uniform in the vorticity direction. The steady-state velocity profiles deviate from a parabolic shape above an applied wall shear rate of 6 s^{-1} ($Wi = 0.8$), in good correspondence with the measured rheology. Kinetically, the deviation from parabolic flow presents as the development of a thin, near-wall band flowing at a high shear rate. The band dynamics along the vorticity (neutral) axis of the flow are observed at distances from 2 to $10 \mu\text{m}$ from the wall. At the smallest near-wall position of $2 \mu\text{m}$, space-time plots reveal four distinct stages in the variation of the flow-direction velocity with time and with position along the vorticity axis. First, the flow accelerates at all points along the vorticity axis to about one-half of the final, steady-state velocity at that position in the gap. Second, a deceleration – again at all points in the flow-vorticity plane – abruptly slows down the fluid motion at this near-wall region. The total change in velocity resulting from the deceleration is about one-half that of the previous acceleration. Third, the flow partitions into multiple strips along the vorticity direction; these strips have non-uniform flow-direction velocity. The width of these strips is dependent on the wall shear rate; their width is $\sim 100 \mu\text{m}$ for flow with a wall shear rate of 8.4 s^{-1} . Finally, all the fast strips combine to form a single homogeneous flow with a fast velocity at $2 \mu\text{m}$ from the wall. This velocity is ~ 50 times that expected for unbanded flow with the same flow rate. We discuss these shear band

spatiotemporal dynamics in the context of transient vorticity structuring that occurs at applied wall shear rates within the stress plateau region.

Introduction – Wormlike micelles (WLM) are long, flexible, cylindrical aggregates of amphiphilic surfactant molecules¹. These flexible micelles form and increase in length above a critical surfactant concentration or upon addition of salt that screens the electrostatic interactions of the head groups². With further increase of surfactant or salt concentration, entangled and branched networks³ form. The linear viscoelastic rheological response of WLM solutions arises from reptation and reversible scission. The latter phenomenon is distinct from what is observed for polymers⁴. Reptation and scission results in linear viscoelasticity described by a single-mode Maxwell model⁵. Behavior deviates from the single-mode Maxwell model at high frequencies⁵.

The constitutive response of WLM solutions in steady shear flow yields a dependence of stress on shear rate that is non-monotonic⁶. In the region of this flow curve with zero or negative slope, homogeneous flow is unstable. The flow instead breaks into shear bands stacked along the gradient axis of the flow, with each band flowing at a different shear rate⁷. More generally, the nonlinear viscoelastic constitutive response of wormlike micelle solutions can generate flow instabilities⁶. In addition to the aforementioned (gradient) shear banding^{6,8}, vorticity banding^{9,10}, apparent wall slip^{11–13}, interfacial instability between shear bands at steady state^{14–16} and oscillating, chaotic bands^{17,18} are reported. Each of these phenomena is linked to the rheological response of the wormlike micellar microstructure to the imposed flow.

Gradient shear banding (i.e., with the stacking direction of the planar bands oriented along the velocity gradient axis) is widely observed in WLM solutions. The banding behavior of WLM solutions is attributed to the separation of the two regions into distinct microstructures¹⁹. These structural differences can arise from micellar alignment²⁰ or from concentration differences between the bands⁵. Several recent studies also show flow-direction velocity fluctuations along the vorticity direction in shear-banded flows at band interfaces^{14,15,21,22} or within the high shear rate band²³. This flow heterogeneity along the vorticity direction is referred to as vorticity structuring⁶. These structures may be generated by normal stress differences between the bands^{6,9}, fluctuating slip that causes instability in the high shear rate band²³, or an elastic instability of the high shear rate band²⁴.

Gradient banding and vorticity structuring are observed in both channel flow and Taylor-Couette flow geometries at steady state. In Taylor-Couette geometries, Salmon et al. observe a sharp interface between the two shear bands and an increase in the high shear rate band thickness with increasing applied shear rate for the cetylpyridium chloride and sodium salicylate system (CPyCl/NaSal)²⁵. Strong wall slip is also observed in these geometries in contact with the high shear rate band for the same system²⁴ or in the low viscosity phase for cetyltrimethylammomium bromide (CTAB) micelles in D₂O²¹. Channel flow is simple to generate, can be operated in the range of shear rates for which shear banding is observed, and is a good model of industrial conduit flows. In channel flow, Callaghan et al. observe a flow instability above a critical strain rate for the CPyCl/NaSal wormlike micelles and connect it to the spurt effect as identified by Vinogradov for polymer melts²⁶. Above a critical flow rate, the velocity profile changes

from parabolic with zero velocity at the wall to a parabolic profile with an apparent finite velocity close to the wall; at even higher flow rates, plug-like flow is observed^{11,26–29}. The plug-like flow is accompanied by a high shear rate band near the wall for CPyCl/NaSal wormlike micelles^{13,26,28,29} and/or a layer of apparent wall slip for both CPyCl/NaSal and CTAB/NaNO₃ systems^{11,13,14,27}. Nghe et al. also observe interfacial instability leading to velocity variation along the vorticity direction of shear-banded flows in microchannels at steady-state for the CTAB/NaNO₃ system, apparently induced by a normal stress jump across the bands¹⁴.

It is argued by Ober et al. that observations of apparent wall slip is consistent with the hypothesis of gradient shear banding generated by a non-monotonic flow curve²⁹. Specifically, if we assume that the viscosity of each band differs by ~ 3 orders of magnitude, then the band thickness can be as small as ~ 100 nm in a flow with a gap ~ 1 mm. In this case, apparent wall slip and gradient shear banding are essentially the same phenomenon. (See also, Ref. 30 for an alternative view.)

The previous literature principally addresses banding at steady-state^{14,25–29}. Studies of band transient behavior^{15,21,23,24,30} upon start up of flow provide additional understanding of the flow-microstructure interactions that generate the instability. Moreover, certain industrially important flows, such as bottle filling, are unsteady³¹. Studies of transient banding can shed light on behavior in such flows.

Transient band phenomenology includes temporal fluctuations along the vorticity direction at band interfaces or near the wall in shear-banded fluids^{15,23,24}. Specifically, Becu et al. report a slow evolution and growth of the banded state (~ 6 hours) and

periodic oscillations at the band interface caused by oscillations in apparent wall slip velocities (CTAB in D₂O)²¹. Similar interfacial instabilities are observed by Lerouge et al. and Fardin et al. for CTAB/NaNO₃ systems^{15,22}. Other transient measurements focus on understanding the underlying mechanism of shear banding³⁰. Lopez-Barron et al. demonstrate that the microstructure of CPyCl/NaSal wormlike micelle solutions initially undergoes an elastic rheological response, with a large stress overshoot, followed by the formation of a transient, flow-aligned state.

The prior transient studies are all in Taylor-Couette flow. Curved streamlines contribute to elastic instabilities in this case; the flow geometry is therefore a factor in explanations of vorticity structuring. There is consequently value in studying transient banding in rectangular channel flow, as findings in this geometry (without curvature) can be compared to results in Taylor-Couette flow (with curvature). Such a comparison and identification of the near-wall dynamics of transient banding can assist the development of shear banding models^{6,32}.

Here, we present direct measurements of the spatiotemporal evolution of the flow-direction velocity in rectangular channel flow, with particular emphasis on the non-uniformity of the flow-direction velocity along the vorticity axis. Using particle image velocimetry, we obtain both the steady-state and transient velocity profiles along the gradient direction of the flow. The transient mean velocity profiles near the wall show fluctuating flow-direction velocities at intermediate times and an abrupt increase to the steady-state value after the onset of shear banding. We also observe that this thin, near-wall region (less than 2 μm) of locally high shear rate displays complex spatiotemporal dynamics along the vorticity direction. These near-wall dynamics accompany the

formation of the gradient shear band and are observed at distances up to 10 μm away from the wall in the gradient direction of flow.

Methods – Two anionic surfactant wormlike micelle solutions that show gradient shear banding at convenient shear rate ranges are studied. The first system is 42 wt.% sodium lauryl ether 3 sulfate (SLE3S), 24 wt.% sodium lauryl sulfate (SLS) and 3.5 wt.% sodium chloride (NaCl) in deionized water. In addition, at one condition, 1.9 wt.% NaCl is used instead to vary the internal microstructure of the WLM system. The second system is 10 wt.% sodium lauryl ether 1 sulfate (SLE1S) and 2.15 wt.% NaCl in deionized water. The amount of salt in these solutions is adjusted to obtain comparable entangled microstructures to the SLE3S/SLS system with 3.5 wt.% NaCl. Both solutions are found near the maximum point on the “salt curve” (cf. Supplementary Materials, SM E).

The linear viscoelasticity and steady shear rheology of the SLE3S/SLS solution is measured by controlled stress rheometry (AR-G2, TA Instruments) using a 40 mm diameter, 2° steel cone at 21.5°C. The storage (G') and loss (G'') moduli, as reported in Figure 1a-b, are measured at frequencies from 0.1 to 100 rad/s. A single-mode Maxwell model fit³³ yields the relaxation time, $\lambda_M = 0.14$ s, and the elastic modulus, $G_0 = 510$ Pa. Figure 1b indicates that deviations from single-mode behavior are small at low frequencies, with disagreement emerging for frequencies greater than about 30 s^{-1} . The steady-state viscosity is measured at shear rates from 0.1 to 100 s^{-1} , as shown in Figure 1c. The near independence of the shear stress for shear rates greater than 4.5 s^{-1} is consistent with the onset of shear banding⁵. Time dependent stress measurements are also made at

162 shear rates of 2.2, 6.1, and 11.7 s⁻¹ (Figure 1d). The stress overshoot at low times can be
163 explained as the initial elastic response of the WLM solution. For all measurements, a
164 solvent trap is placed over the sample to minimize sample evaporation.

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Figure 1. Linear viscoelasticity, steady shear rheology, and transient rheology measurements of SLE3S (42 wt.)/SLS (24 wt.%) WLM solutions with 3.5 wt.% NaCl. (a) Storage (G') and loss (G'') moduli are plotted as a function of angular frequency, ω , for oscillatory frequency sweep at $\gamma = 1$ and $T = 21.5^\circ\text{C}$. The solid black line corresponds to a single-mode Maxwell model fit with $G_0 = 510$ Pa and $\lambda_M = 0.14$ s. (b) Cole-Cole plot of the oscillatory frequency sweep results shows a Maxwellian behavior. The solid line is the single-mode Maxwell model fit. (c) Steady shear stress and steady shear viscosity of WLM solutions at 21.5°C . The plateau in the steady shear stress is observed at a shear rate of 4.5 s^{-1} . (d) Transient shear stress responses to start-up flow experiments at three shear rates (2.2, 6.1, and 11.7 s^{-1}).

Rectangular channel flow at constant volumetric flow rates is generated in a flow cell (IBI Scientific) of dimension 0.8 mm x 4 mm x 40 mm. Three walls of the rectangular channel are made of polycarbonate. The fourth boundary is a 160 μm thick glass cover slip through which the flow is visualized. The WLM solution is delivered to the flow cell using a syringe pump (10 ml capacity, KDS 100, Fisher Scientific). The syringe is connected to the flow cell using an aluminum pipe (1/8"). The flow apparatus has a slow time constant (~ 500 s) for reaching steady-state flow when pumping high viscosity fluids at low rates (below 2.2 s^{-1} , $Wi = 0.3$); the WLM solutions studied here are in this category. At high rates, above 20.1 s^{-1} ($Wi = 2.8$), the (rise) time constant to reach steady flow is much faster (~ 20 s). The regimes of transient shear banding dynamics reported in this paper do not overlap with the rise times of the flow. To ensure that the results are independent of these mechanical effects, all steady state measurements are taken long after the flow reaches its steady flow rates (15 minutes for the slowest flow case, 2.2 s^{-1} , to 3.5 minutes for the fastest flow case, 20.1 s^{-1}). Moreover, comparison of the flow rate rise times to the transient velocity measurement shows that the transient behavior observed (~ 100 s from flow start-up) is significantly later than the time constant (~ 20 s) of the flow (c.f. SM A). The (applied) wall shear rates of the flows are calculated at the channel wall based on the volumetric flow rates, which are directly measured.

The wormlike micelle solutions are seeded with 500 nm, yellow-green fluorescent dye polystyrene particles (Invitrogen) at a volume fraction of ~ 0.00001 . Particles are visualized with Nikon A1R confocal laser scanning microscope with a 488 nm laser. A 20X objective ($NA = 0.75$) is used under the resonant scanning mode (0.067 s/frame) to

generate 512 x 512 pixel images with a pixel resolution of 1.23 $\mu\text{m}/\text{pixel}$. As shown in the left most panel in Figure 2a, frames are taken at varying focal planes (xy plane, flow-vorticity plane) within the channel. To illustrate: two consecutive frames (in green and red in the right most panel) with $\Delta t = 0.067\text{s}$ are analyzed to extract particle velocities. For measurements at steady-state conditions, the depth of the focal plane is varied between 2 μm and 798 μm along the gradient direction (z-axis). At each gradient position, the middle 15% of the flow along the vorticity direction is imaged. Images are taken at 2, 10, 50, 100, 250, 400, 550, 700, 750, 790, and 798 μm above the glass cover slip for steady state velocity profiles at wall conditions varying from 2.2 s^{-1} to 20.1 s^{-1} ($Wi = 0.3$ to 2.8). For transient measurements, images are acquired at 2 μm above the cover slip. The duration of individual experiments varies based on the wall shear rate studied, ranging from 15 minutes for the slowest flow case (2.2 s^{-1}) to 3.5 minutes for the fastest flow case (20.1 s^{-1}).

We use an open-source PIV software, PIVlab, to analyze the flow behavior within the channel³⁴. Prior to image processing, contrast limited adaptive histogram equalization (CLAHE) is applied³⁵. Once image contrast modification is completed, each pair of frames is divided into interrogation windows of dimension 64 x 64 pixels, as shown in Figure 2a, middle panel. For each of these interrogation windows, a 2D Fast Fourier Transform (FFT) is calculated. Then, the FFT at one time point is multiplied by the conjugate of the interrogation area FFT at the second time point (Figure 2b). The cross correlation matrix of the pixels within the interrogation areas of the two frames is obtained by inverse FFT of the product, as per the correlation theorem. From the cross correlation matrix, the particle velocity is determined, as illustrated in Figure 2b.

Figure 2. Particle image velocimetry (PIV) in the rectilinear channel flow. (a) The three images depict the flow at different scales: 1 mm, 100 μm and 15 μm scale bars from left to right. All confocal micrographs (a, middle and right panel, b, left and right panels) are of the flow, imaged at the center of the xy (flow-vorticity) plane. (a, middle panel) For PIV analysis, each confocal image frame is divided into interrogation areas. (a, right panel) A particular interrogation area is shown at two time frames (one green and one red), with $\Delta t = 0.067$ s. (b) Steps used for PIV cross-correlation are shown. (b, left panel) Fast Fourier Transforms (FFT) of the images taken at two time frames are calculated. (b, middle panel) The intensity of the first image FFT is multiplied by the conjugate of the second image FFT. Then, the inverse FFT of the result yields the cross-correlation matrix. The location of the maximum intensity reports the direction and magnitude of the vector describing the movement of the particle patterns on the images. The scale bar here is 15 pixels, with a pixel resolution of 1.23 $\mu\text{m}/\text{pixel}$. (b, right panel) From the correlation matrix, the local velocities within the interrogation area can be calculated from the frame rate (0.067 s). The process is repeated until the entire vector field is derived for the image series.

This process is repeated to generate a vector distribution for the entire image. To obtain a high-resolution vector field, multiple passes are taken with decreasing interrogation window size; the initial displacement data from the large interrogation areas are used to spatially offset the interrogation windows in the following passes. Specifically, for our analysis, three passes of 256, 128, and 64 square pixel interrogation areas are used. An example of a final vector map is shown in Figure 2b. Some spurious vectors, resulting from poor illumination of local regions or particles traveling out-of-field, are removed and interpolated. (The number of these interpolated vectors is below 5% in each experiment). We further validate the PIV method by comparing results for several instances to those generated by direct particle tracking. Particle velocity information is extracted along a line (0.63 mm) in the vorticity direction at 6 time points (1.1, 1.7, 2.2, 2.8, 3.4, 3.9 minutes) and compared to the velocities from particle tracking along the same line (SM B). The mean deviation between PIV and particle tracking ranges from 0.4% to 16% at the highest case, with a mean deviation across all cases of 3%. More detailed description of the validation process and results can be found in Supplementary Materials B.

Results – Strong deviations from a parabolic profile are observed at wall shear rates above 6 s^{-1} ($Wi = 0.8$) for the SLE3S/SLS solution (Figure 3a). Under these conditions, a thin region of high velocity exists at the wall, because the measurement at the nearest wall position ($2 \text{ }\mu\text{m}$) is already at finite speed at these high wall shear rates. The high velocity region is observed at both the top and bottom boundaries, and for two different boundary materials –borosilicate glass and polycarbonate. The near-wall velocities,

taken at 2 μm from the cover slip, increases linearly as the applied wall shear rate is increased from 6.1 s^{-1} to 20.1 s^{-1} ($Wi = 0.8$ to 2.8) (Figure 3b). The flow conditions at which increased velocities are observed coincide with the shear rates of the stress plateau in the steady-shear rheology curve (Figure 1b). The appearance of a high velocity region near the wall, initiated at a shear rate corresponding to the plateau region of the shear rheology, is consistent with shear banding.

Figure 3. Steady-state velocity profiles and near-wall velocities for the WLM solution. (a) Velocity profiles are parabolic with nearly zero velocities at the wall for shear rates below 6.1 s^{-1} ($Wi = 0.8$). Above wall shear rates of 8.4 s^{-1} ($Wi = 1.2$), deviations from the parabolic behavior are observed at the wall; increasingly high velocities are measured 2 μm from the wall as applied wall shear rate is increased. Error bars plotted for the 11.7 s^{-1} data are standard deviations of the mean of 3 data sets. (b) The near-wall velocities at 2 μm from the wall (bottom cover slip) are plotted as a function of applied wall shear rate. An increase in these velocities is measured above a wall shear rate of 6.1 s^{-1} . All experiments are carried out for SLE3S (42 wt.%) / SLS (24 wt.%) WLM solutions with 3.5 wt.% NaCl solution at 21.5°C. (c) WLM near-wall measurements are reported at 11.7 s^{-1} ($Wi = 1.6$). Velocity measurements of WLM solutions within this small region show a near-flat velocity profile. The dotted line is an estimate for parabolic flow at 11.7 s^{-1} shear rate flow in an 800 μm channel, added to a constant near-wall velocity of 750 $\mu\text{m/s}$. Error bars indicate standard deviations of the mean.

Velocity profiles similar to that of Figure 3a are measured by Ober et al. In that case, slip-like layers are observed for flows of moderate Weissenberg numbers ($Wi < 5$)²⁹. In the present case, the Wi number for all flow conditions studied varies from 0.3 to 2.8, with the onset condition (6.1 s^{-1}) corresponding to $Wi = 0.83$. The Weissenberg number is defined as $\lambda_M \dot{\gamma}$, where λ_M is the Maxwell model relaxation time (0.14 s), $\dot{\gamma}$ is the applied wall shear rate calculated from the volumetric flow rate and the channel height (800 μm).

Figure 3c reports high-resolution microscopy measurements that resolve the region very close to the wall. A 100X objective ($NA = 1.45$) is used to generate 512 x 512 pixel images with a pixel resolution of 248 nm/pixel. In a region 4 μm and less from the wall, control measurements with a Newtonian fluid show linear behavior, consistent with the wall shear rate applied (11.7 s^{-1} , $Wi = 1.6$) and the no-slip boundary condition (cf. SM C). Based on the sizes of the tracer particles (500 nm), the nearest wall measurement is made at 250 nm from the wall. For the WLM solution, a large velocity is measured over the entire 4 μm region and a flat profile persists to the minimum dimension that the PIV method could resolve (Figure 3c). The near-flat profile indicates that the high shear rate band next to the wall must be less than 250 nm thick. (A tighter bounding of the depth of this thin shear banded region is beyond the capabilities of our optical microscopy methodology.) The dotted line is an estimate of parabolic flow in an 800 μm channel within the 4 μm region, given the existence of a near wall velocity equal to the magnitude measured at the point nearest the wall. The data agree well with parabolic estimate, suggesting that the near-wall region is consistent with the parabolic velocity measured in the remainder of the low-rate band.

Figure 4. Shear rates of the low rate band. (a) Velocity profiles from Figure 3a are adjusted by subtracting the near-wall velocities across the entire profile. Parabolic fits (solid lines) show good performance at all wall shear rate conditions. (b) Steady-state shear rates of the flow are extracted from the analysis of (a) and plotted against the nominal wall shear rate applied to the whole flow. The red line shows $\dot{\gamma}_{ss} = \dot{\gamma}_{nom}$. The blue line shows the critical shear banding condition, $\dot{\gamma}_{ss} = 4.5$ s⁻¹. At wall shear rates up to 6.1 s⁻¹ ($Wi = 0.8$), data approximately follow the red line. At wall shear rates above 6.1 s⁻¹, data fluctuate about the blue line. Error bars are calculated from the standard error of the fit in (a).

To analyze the shear rate dependence of the banded flow, the measured velocity profiles are fit to a parabolic velocity profile with an added slip velocity. The slip velocity values are as reported in Figure 3b – the measured velocity at a gradient height of 2 μm. The best-fit parabola, less the slip velocity, is compared to the measurements (also shifted by the slip velocity) in Figure 4a. The fit parameter is the apparent shear rate of the (low-rate) banded flow. These shear rates are plotted against the applied wall shear rates of the flow in Figure 4b. The two lines in Figure 4b represent bounding cases.

The curve with slope one is for Newtonian (unbanded) flow; the horizontal line at 4.5 s^{-1} is the onset condition for banded flow, as deduced from Figure 1c.

The data, within error, appear to track the two bounds. First, at low applied wall shear rates (between 2.2 to 6.1 s^{-1} , $Wi = 0.3$ to 0.8), the measured shear rates agree with the imposed values, as indicated by the red curve. Second, at high applied wall shear rates (above shear rates of 6.1 s^{-1} , $Wi = 0.8$), the shear rates extracted from the parabolic fit scatter in the vicinity of the horizontal line of $\dot{\gamma}_{ss} = 4.5 \text{ s}^{-1}$. Recall that the horizontal bound is derived from the rheological measurements (Figure 1c). Therefore, the plateau observed above the critical shear rate (4.5 s^{-1}), suggests that the bulk contains a low shear rate band whose steady-state velocity profile is consistent with the measured shear rheology of this fluid.

The kinetics of the development and evolution of the high shear rate band are examined by measuring the time-dependent velocity at a gradient height of $2 \text{ }\mu\text{m}$ (Figure 5), averaged across the vorticity axis of the flow. Transient velocity profiles for the wall shear rate range below the critical shear rate for banding ($2.2 - 6.1 \text{ s}^{-1}$, $Wi = 0.3 - 0.8$) are shown in Figure 5a. A monotonic increase in $\langle v \rangle$ is observed for all conditions. Large time constants (e.g. $\sim 500 \text{ s}$ for 2.2 s^{-1}) result because of the very high viscosity of the WLM solutions at low wall shear rates (e.g. 41 Pa s at 2.2 s^{-1}), as discussed in the Methods section.

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341 **Figure 5. The effect of applied wall shear rate on the vorticity-averaged near-wall velocity in the flow**
 342 **direction.** All measurements are taken at $2 \mu\text{m}$ from the wall. (a) At wall shear rates 2.2 , 4.4 , and 6.1 s^{-1}
 343 ($Wi = 0.3$, 0.6 , and 0.8), transient velocity measurements show a monotonic increase in average velocities
 344 at this wall normal position. The large time constant ($\sim 500 \text{ s}$) for the steady-state flow is observed for the
 345 lowest shear rate value (2.2 s^{-1}) and is caused by the high viscosities of the fluid at low shear rates. (b) At
 346 wall shear rates 8.4 , 10 , and 11.7 s^{-1} ($Wi = 1.2$, 1.4 , and 1.6), large fluctuations in the average velocity are
 347 shown at intermediate times; at long times the near-wall $\langle v \rangle$ is large, consistent with the steady-state
 348 measurements of Figure 3. At higher wall shear rates, 14.0 , and 20.1 s^{-1} ($Wi = 1.9$ and 2.8), flow rapidly
 349 transitions to steady-state. (c) Time to reach steady-state (t_{ss}) extracted from (b) by taking the first time
 350 point at which $\langle v \rangle$ is 95% of its steady-state value.

For conditions above shear banding ($8.4 - 20.1 \text{ s}^{-1}$, $Wi = 1.2 - 2.8$), large fluctuations in $\langle v \rangle$ are observed at intermediate times. These fluctuations are followed by abrupt increases in the $\langle v \rangle$ to steady-state values (Figure 5b). This transition is most pronounced under the fastest flow condition, 20.1 s^{-1} ($Wi = 2.8$), where the increase in $\langle v \rangle$ occurred within $< 20 \text{ s}$ and only a brief intermediate period of fluctuations is measured. Overall, the time to steady banded flow decreases with the imposed wall shear rate (Figure 5c).

Figure 6. The effect of wall-normal distance (z-distance) on the transient velocity measurements. (a) Transient average velocities are measured at wall normal (gradient) distances of 2, 10, 50, 100, and 250 μm at an applied wall shear rate of 11.7 s^{-1} ($Wi = 1.6$). (b) Time to reach steady state (t_{ss}) is obtained from (a) by taking the first time point at which the $\langle v \rangle$ reaches 95% of its plateau value for all z-distance conditions.

Figure 6a plots the time-dependence of the average velocity at five distances from the wall (i.e., along the gradient direction) for an imposed wall shear rate of 11.7 s^{-1} ($Wi = 1.6$). Two main observations can be made. First, the velocity is non-monotonic at $2 \text{ }\mu\text{m}$ distance from the wall. The magnitude and the duration of the intermediate fluctuations vary from trial to trial, as shown by comparable wall shear rate conditions in Figure 5b of 11.7 s^{-1} flow. (Larger fluctuations are observed for the same wall shear rate condition in Figure 5b.) However, the overall qualitative characteristics of the flows are consistent. Second, the time it takes for the system to reach its steady-state (t_{ss}), is the longest at $2 \text{ }\mu\text{m}$ from the wall. Figure 6b plots t_{ss} as a function of distance from the wall. While the time measured at $2 \text{ }\mu\text{m}$ is indeed larger than the others, apart from this t_{ss} is largely independent of distance from the wall. Thus, only near the wall is the mean velocity non-monotonic, and this non-monotonicity may be correlated with an increased time to reach steady state.

Figure 7 identifies the origin of these near-wall flow behaviors. The velocity field in the flow/vorticity plane at a gradient height of $2 \text{ }\mu\text{m}$ is reported at six different time points varying from $t/t_{ss} = 0.40$ to 0.93 . Measurements are for an imposed wall shear rate 8.4 s^{-1} ($Wi = 1.2$). At short times, the velocity increases monotonically at all points in the flow-vorticity plane (Figures 7a,b). At $t/t_{ss} = 0.77$, the flow-direction velocity begins to become non-uniform in the vorticity direction, breaking into thin, randomly positioned, high velocity strips (Figure 7c), coexisting with slowly-moving regions (Figure 7c). Over time, the high velocity strips grow thicker by merging together, until only 1-2 thin slow moving regions persist, as shown in Figures 7d,e. By $t/t_{ss} = 0.93$, a homogeneous flow with a fast velocity is achieved at all points along the vorticity direction (Figure 7f). This

steady-state flow-direction velocity near the wall is ~ 50 times greater than expected for an unbanded flow of a Newtonian fluid at the same flow rate. The observation of these transient dynamics indicates that the flow-direction velocity fluctuations and the long times to steady-state (Figure 6b) are strongly coupled with the spatially-dependent velocity field near the wall.

Figure 7. Transient fluctuations in the velocity along the vorticity axis at a gradient height of $2 \mu\text{m}$. Velocity fields are shown for a flow (8.4 s^{-1} , $Wi=1.2$) observed at the center of the flow-vorticity plane at $2 \mu\text{m}$ from the bottom wall at 6 distinct time points, t/t_{ss} (0.40, 0.61, 0.77, 0.81, 0.87, and 0.93). (a-b) Flow-direction velocity increases over time as observed by increasing vector lengths. (c) Thin strips flowing at fast velocities are observed along the vorticity direction. (d-e) Fast strips grow and merge until very thin strips of the slow velocity regions remain. (f) Homogeneous velocity vector field is observed at steady-state, where all the particles are traveling at the large velocity observed at $2 \mu\text{m}$ from the wall.

Figure 8. Space-time plots of flows below and above shear banding conditions. (a) Space-time plot of the flow at 4.4 s^{-1} ($Wi = 0.6$), prior to shear banding, is measured along the vorticity direction over time. (b) Flow profile along the vorticity direction is measured over time for 8.4 s^{-1} ($Wi = 1.2$) wall shear rate condition. (c-f) Similar 4-step sequence is observed for the flow under 10.0 , 11.7 , 14.0 , and 20.1 s^{-1} ($Wi = 1.4$, 1.6 , 1.9 , and 2.8) conditions. The fluctuating step, however, occurs for a shorter duration ($\sim 15 \text{ s}$) as compared to the slower wall shear rate conditions in (b and c) ($\sim 50 \text{ s}$).

Space-time plots of the flow are generated for six different flow conditions (4.4, 8.4, 10.0, 11.7, 14.0, and 20.1 s⁻¹, Wi = 0.6, 1.2, 1.4, 1.6, 1.9, and 2.8) as shown in Figure 8. These plots record spatiotemporal patterns of the flow along the vorticity direction, labeled as “y.” They are constructed from the flow-direction velocities (v_x) along the vorticity axis (y) taken at each time frame (0.067 s) over the duration of the flow. Below the critical shear rate for banding, a gradual increase in velocity is observed (Figure 8a) as similarly reported by transient average velocity measurements. There is no strong spatial variation in the flow along the vorticity axis.

Figures 8b-f report the vorticity variation in the flow-direction velocity for shear banding conditions (8.4, 10.0, 11.7, 14.0, and 20.1 s⁻¹, Wi = 1.2, 1.4, 1.6, 1.9, and 2.8). The space-time plots show the temporal development of high velocity regions at the walls. The process has four distinct steps. Each step is observed for all conditions; here we describe the development for 8.4 s⁻¹ (Wi = 1.2) (Figure 8b). First, the flow-direction velocity accelerates uniformly across the vorticity plane. The acceleration time ranges from ~ 55 to 150 s. The acceleration reaches about one-half of the steady-state, average velocity at this gradient position. Second, the observed flow-direction velocity decelerates to ~ one-half of the mean velocity observed in the first step. The deceleration is short lived (~ 5 – 10 s). Third, the flow-direction velocity becomes non-uniform in the vorticity direction; the flow breaks into multiple strips of differing velocity at points along the vorticity direction. The width of the fast moving strips depends on the imposed wall shear rate and is ~100 μm for 8.4 s⁻¹. A single, wide fast strip is observed for the 11.7 s⁻¹ flow. The strips grow in width over time during this step by the mechanism of strip combination and growth. The duration of this phase is dependent on the wall shear

rate conditions studied. For 8.4 s^{-1} ($Wi = 1.2$) condition, the characteristic time for the vorticity fluctuation phenomenon is $\sim 40 \text{ s}$. For faster flow conditions, 11.7, 14.0, and 20.1 s^{-1} ($Wi = 1.6, 1.9$, and 2.8), the characteristic time is shorter, $\sim 15 \text{ s}$. Finally, all strips combine to produce a homogeneous, fast flow-direction velocity. This steady-state velocity is now ~ 50 times that expected for an unbanded, Newtonian flow. While detailed observation of this transient vorticity dynamics is described here for the SLE3S/SLS WLM solutions (3.5% salt), the same 4-step process with transient vorticity structuring is observed for SLE3S/SLS (1.9% salt) and SLE1S WLM solutions (cf. SM D).

Figure 9. Characteristic transition times for deceleration, fluctuation and steady-state. Transition times for deceleration (t_{decel}), fluctuations (t_{fluc}), and steady-state (t_{ss}) are calculated and plotted for wall shear rates of 8.4, 10.0, 11.7, 14.0, and 20.1 s^{-1} ($Wi = 1.2, 1.4, 1.6, 1.9$, and 2.8). All curves lie close together for the range of wall shear rates reported, with the largest variation from the duration of the fluctuation ($t_{\text{fluc}} - t_{\text{ss}}$) for 8.4 and 10.0 s^{-1} conditions.

The characteristic time scales of the flow at all wall shear rate conditions are displayed in Figure 9. Three distinct times, t_{decel} , t_{fluc} , and t_{ss} , report the times at which

deceleration, fluctuation, and steady-state transition take place, respectively. The deceleration transition time, t_{decel} , is taken as the time at which the local maximum $\langle v \rangle$ is reached from flow start up. The fluctuation transition, t_{fluc} , is defined as the time at which no consecutive decrease in velocity is measured for Δt of 0.3 s. The steady-state transition times, t_{ss} , is calculated by taking the time at which 95% of the average steady state velocity is reached. Durations of the fluctuations are the largest for 8.4 and 10.0 s^{-1} ($Wi = 1.2$ and 1.4) wall shear rate cases. Overall, characteristic times for all three regimes decrease with applied wall shear rates for shear rate ranges of 8.4 to 14.0 s^{-1} ($Wi = 1.2$ to 1.9). However, similar time scales are observed for 14.0 and 20.1 s^{-1} ($Wi = 1.9$ and 2.8); these conditions also exhibit the shortest time scales for all three regimes. Relative to the overall time required to achieve steady-state flow, the duration of the acceleration, deceleration, and fluctuating regimes is small.

Discussion – Steady-state velocity profiles of wormlike micelle solutions in rectangular channel flow display gradient shear banding at flow conditions consistent with the region of stress plateau as measured by cone-and-plate rheometry. Under these conditions, steady-state profiles exhibit thin, near-wall, regions ($< 2 \mu\text{m}$) of high shear rate, high velocity flow. In these regions, the maximum velocity is ~ 50 times higher than the value predicted by extrapolating the parabolic flow observed in the bulk to the wall, corresponding to local, near-wall shear rates that are an equivalent factor larger than the nominally applied wall shear rate. Such a thin, near-wall region of high velocity flow coexisting with a parabolic inner flow is consistent with a number of literature studies^{13,26,28,29}.

At applied wall shear rates above the shear banding condition, steady-state velocities within the near-wall region increase linearly with the applied wall shear rate (Figure 3b). The linear increase in the near-wall velocity – an apparent slip velocity – is consistent with a shear band whose interface is closer to the wall than the $2\text{ }\mu\text{m}$ point of measurement. This bound on the band thickness is further refined to be less than $\sim 250\text{ nm}$ based on the near-flat velocity profiles within $4\text{ }\mu\text{m}$ of wall, as measured by high resolution microscopy (c.f. Figure 3c). The thin width ($< 250\text{ nm}$) of the high shear rate band implies that the high shear rate region of the flow curve appears only at shear rates greater than $\sim 10^4\text{ s}^{-1}$. The bounds on band dimension also imply that the viscosity within the band is low – no greater than 0.07 Pa s – and a factor of ~ 700 less than the zero-shear viscosity of the WLM as measured by cone and plate rheometry.

Prior to steady state, transient flows with complex spatiotemporal behavior are observed in the near-wall region ($< 10\text{ }\mu\text{m}$) for shear banding conditions. At a distance only $\sim 1\text{-}2\text{ }\mu\text{m}$ from the shear band interface, strong variations in the time dependent flow-direction velocity are observed along the vorticity direction. We describe the transient dynamics as a four-step process involving: (1) global acceleration of the flow to about half of the steady-state flow-direction velocity at all points along the vorticity axis, (2) global deceleration to about half of the magnitude of the previously accelerated velocity, (3) a regime in which the flow-direction velocity is non-uniform in the vorticity direction, with strips of high velocity observed, (4) attainment of the final, fast velocity characteristic of steady-state. In the third stage, the strips of varying flow-direction velocity are spatially chaotic; the number of strips is highly variable. The duration of this third, fluctuation regime ranges from 10 to 40 s, with shorter time scales for faster flows.

The spatiotemporal dynamics of gradient shear banding reported in this paper are distinct from vorticity banding, as generated in Taylor-Couette geometries by viscoelastic instability triggered by the curved streamlines of that flow^{10,22}. Indeed, the inhomogeneous flows observed here are not vorticity bands, because they do not extend more than 10 μm in the wall normal direction of the flow. Above this dimension, the flow (in the low rate band) is steady and homogeneous.

However, we argue a connection between the vorticity structuring of gradient bands observed here and recent high rate/stress experimental¹⁴ and theoretical¹⁶ studies of wormlike micelles in plane Poiseuille flow. Nghe et al. and Fielding et al. find – by both theory and experiment – that the interface between high and low shear rate bands is unstable; this interface supports flow velocity fluctuations along the vorticity axis at steady-state^{14,16}. These interfacial instabilities are induced by a jump in the second normal stress difference across the interface. This imbalance generates secondary flows. The theoretical analysis is for the case of a diffusive Johnson-Segalman fluid, a material with a viscoelastic constitutive response that generates a non-monotonic stress vs. shear rate curve and a high shear stress region that is Newtonian¹⁴. This constitutive model also predicts non-zero first and second normal stress differences³⁶.

There are similarities and differences between the observations of Nghe et al. and this work. First, Nghe et al. report that the wavelength of the interfacial undulations in the flow-direction velocity (along the vorticity direction) ranges from 1 to 2 times the height of the channel (in the gradient direction). In our case, the full height of the channel is 800 μm ; the corresponding wavelength of the vorticity fluctuation would be 800 – 1600 μm in length if there were full correspondence with Nghe et al. The widths

of the transient strips reported in this work range from 100 to 500 μm , with a maximum observable size of $\sim 600 \mu\text{m}$ as set by our observation window along the vorticity axis.

Second, the amplitude of the modulations at the shear band interface found by Nghe et al. ranges from 0.5 - 1% of the channel height. This is ~ 4 to $8 \mu\text{m}$ for the geometry studied in this work, which is consistent with the gradient height at which we observe vorticity-direction non-uniformity of the flow. This correspondence suggests that the fluctuation dynamics measured here could be an interfacial instability similar to that described by Nghe et al. and Fielding et al.

Notwithstanding these commonalities, there are a number of interesting differences in the channel flow results of Nghe et al. and this work. First, the fluid studied by Nghe et al. displays a stress plateau $\sim 100 \text{ Pa}$ that persists from a low-rate transition of 5 s^{-1} to a high-rate transition of $\sim 100 \text{ s}^{-1}$. The fluid they studied is a 0.3M CTAB solution in 0.405M sodium nitrate solution. In our fluid (1.2M SLE3S and 1.0M SLS solution in 0.7M NaCl solution), the stress plateau is at $\sim 180 \text{ Pa}$. However, the shear rate range of the stress plateau is very different in our fluid, with a low rate critical condition of 4.5 s^{-1} and a high rate transition estimated to be greater than 10^4 s^{-1} . The vorticity structuring observed in Nghe et al. is for flow at a wall shear stress greater than the stress plateau (30% greater than that plateau), and in regions for which the measured slip length is small – a mean value of $0.5 \pm 0.9 \mu\text{m}$ is reported. On the other hand, the vorticity structuring here is observed within the region for which shear bands coexist, and for which the extracted, average slip length is much larger: $144 \pm 14 \mu\text{m}$. (The slip length is extracted from the slope of the plot in Figure 3b.)

Another key difference between the present report and the work of Ref. 14 is that the vorticity-axis velocity fluctuations here are only observed transiently; the flow-direction velocity is homogeneous along the vorticity direction at steady-state. The velocity modulations reported in Ref. 14 occur at the shear band interface, again for wall shear stresses that are 30% above that of the shear-banding onset. The applied shear rate in their case is estimated to be ~ 25 times the critical shear rate (5 s^{-1}) based on the flow curve reported in Ref. 12. In comparison, the equivalent wall shear rate ranges probed in this work are at most 4 times the onset rate for shear banding. This comparison suggests two conclusions: (i) that vorticity structuring of the flow-direction velocity in the vicinity of the band interface is not solely a high-shear rate phenomena; it can occur at applied wall shear rates that are only just above the onset condition; (ii) that such low shear rate cases of vorticity structuring generate flow-direction velocity fluctuations that are only transient. That is, there is a mechanism at these lower shear rates by which vorticity disturbances initiated at the band interface are damped, and do not persist at steady state.

Therefore, the absence of steady-state vorticity structures in the present case could stem from a subcritical instability in the shear rate region studied. That is, fluctuations at the near-critical shear rate could dampen over time, leading to a secondary flow that is unsustainable at steady-state. Transient vorticity dynamics observed in this work could consequently be understood as the early transient effect of the interfacial instability of gradient shear bands.

The above comparison is a purely continuum description of the phenomenon, with a mechanism for transient instability driven by the second normal stress differences at the interface, as described by Fielding¹⁶. The question arises as to what behavior of the

micellar microstructure is generating these changes in continuum rheological properties. Ideas discussed in the literature include the development of a concentration gradient between the bands (demixing)⁵ or the generation of an aligned state of the WLM solution (a flow-induced isotropic-nematic transition)²⁰. The thin bands observed here suggest that the solution in the high shear band has a very low viscosity – no greater than 0.07 Pa s, which corresponds to a high to low shear phase viscosity ratio of 0.001. In comparison, Nghe et al. report a viscosity ratio of ~ 0.05 between the high and low shear phases¹⁴. One mechanism by which these low viscosities could be produced could be through the rejection of surfactants from the near-wall region. The channel flow geometry establishes a shear stress relation that varies linearly across the cell, with maximum shear stress at the walls. We speculate that large wall stresses and an inhomogeneous stress field could cause surfactants to migrate towards the center of the channel, forming a low surfactant concentration region with significantly reduced viscosities. The measured concentration-viscosity dependence for our surfactant system (data not shown) is consistent with that of literature¹. If concentration depletion by surfactant migration were the origin of the low viscosity (no higher than 0.07 Pa s) in the thin, high shear rate band, then the measured concentration viscosity dependence would require that the surfactant concentration in the high rate band be about 75% less than in the unbanded solution. Such surfactant migration would take time and be a complex process; therefore, it could contribute to the chaotic spatiotemporal dynamics in the velocity observed in the near-wall region of the flow.

Conclusion – We have carried out velocity profile measurements of wormlike micelle solutions in rectangular channel geometry. Using confocal microscopy and particle image velocimetry, we have characterized transient variations in the flow-direction velocity that are non-uniform along the vorticity direction for shear-banded flows. The transient process has four steps: flow acceleration, deceleration, fluctuations in the flow-direction velocity along the vorticity direction, and attainment of a steady-state flow that is uniform in the vorticity direction. The intermediate structuring of the flow is found at points up to 10 μm in the gradient direction; the characteristic time scale for its onset increases for both near-wall and low wall shear rate conditions. The observed phenomenon is distinct from vorticity banding generated by viscoelastic instabilities in curved streamlines, but could be explained by the presence of interfacial instabilities at the shear band interface. The vorticity structuring of gradient shear bands observed here occurs at applied wall shear rates that are as low as four times the onset condition for shear banding.

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