## Elastic instabilities in a microfluidic cross-slot flow of wormlike micellar solutions

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## Supplemental Material

In addition to the linear rheological characterization provided in the paper, we have also included the large angle oscillatory shear (LAOS) measurements to investigate the non-linear viscoelasticity of the 0.100 M CTAB, R = 0.32 and 0.075 M CTAB, R = 0.32 solutions. Hyun et al.[41] have previously used LAOS to categorize the shear-induced formation of microstructures into four different types of behavior using the first order harmonic storage (G') and loss moduli (G''). Fig. 1 shows the reduced moduli  $G'/G'_o$  and  $G''/G''_o$  as a function of applied strain amplitude for the 0.100 M CTAB, R = 0.32 solution at two low oscillation frequencies (0.01 Hz and 0.1 Hz). At these low frequencies, the LAOS behavior corresponds to a Type I, strain thinning, where both  $G'/G'_o$  and  $G''/G'_{o}$  decrease as the strain amplitude increases. This type of behavior is similar to the origin of shear thinning in which the wormlike micelles disentangle and align with the flow field. When we probe the 0.100 M CTAB, R = 0.32 solution at higher frequencies (1 Hz and 10 Hz) we find that the LAOS behavior transitions to a Type III, weak strain overshoot, as shown in Fig. 2. This type of behavior is associated with an initial resistance against deformation of a complex structure until a critical strain (G'' increases), at which point the complex structure is destroyed by the large deformation and the wormlike micelles align with the flow field (G'' decreases). When we examine the LAOS behavior of the 0.075 M CTAB, R = 0.32 solution at the same oscillation frequencies (0.01 Hz, 0.1 Hz, 1 Hz, and 10 Hz), we see that this solution behaves as Type I for all frequencies as shown in Fig. 3. At present, we are not clear as to why there is a transition between Type I and Type III for only the 0.100 M CTAB, R = 0.32 solution. This dependence on oscillation frequency may suggest that the wormlike micelles in solution are forming different complex structures at varying times scales, which seems consistent from the linear rheological characterization. Although the plots of the reduced moduli provide a qualitative way to describe the different types of LAOS behavior, more extensive studies should be conducted as our future work (see Ewoldt et al. [42] and Hyun *et al.*[43]) with the higher harmonics analysis.



Figure 1: Reduced moduli  $G'/G'_o$  and  $G''/G''_o$  vs strain amplitude for the 0.100 M CTAB, R = 0.32 solution at low oscillation frequencies: 0.01 Hz, 0.1 Hz.



Figure 2: Reduced moduli  $G'/G'_o$  and  $G''/G''_o$  vs strain amplitude for the 0.100 M CTAB, R = 0.32 solution at high oscillation frequencies: 1 Hz, and 10 Hz.



Figure 3: Reduced moduli  $G'/G'_o$  and  $G''/G''_o$  vs strain amplitude 0.075 M CTAB, R = 0.32 solution at different oscillation frequencies: 0.01 Hz, 0.1 Hz, 1 Hz, and 10 Hz.