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Single polymer dynamics in semi-dilute unentangled and entangled solutions: from molecular conformation to normal stress

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Semi-dilute polymer solutions are encountered in a wide array of applications such as advanced 3D printing technologies. Semi-dilute solutions are characterized by large fluctuations in concentration, such that hydrodynamic interactions, excluded volume interactions, and transient chain entanglements may be important, which greatly complicates analytical modeling and theoretical treatment. Despite recent progress, we still lack a complete molecular-level understanding of polymer dynamics in these systems. In this talk, I will discuss three recent projects in my group to study semi-dilute solutions that focus on single molecule studies of linear and ring polymers and a new method to measure normal stresses in microfluidic devices based on the Stokes trap. In the first effort, we use single polymer techniques to investigate the dynamics of semi-dilute unentangled and semi-dilute entangled DNA solutions in extensional flow, including polymer relaxation from high stretch, transient stretching dynamics in step-strain experiments, and steady-state stretching in flow. In the semi-dilute unentangled regime, our results show a power-law scaling of the longest polymer relaxation time that is consistent with scaling arguments based on the double cross-over regime. Upon increasing concentration, we observe a transition region in dynamics to the entangled regime. We also studied the transient and steady-state stretching dynamics in extensional flow using the Stokes trap, and our results show a decrease in transient polymer stretch and a milder coil-to-stretch transition for semi-dilute polymer solutions compared to dilute solutions, which is interpreted in the context of a critical Weissenberg number Wi at the coil-to-stretch transition. Interestingly, we observe a unique set of polymer conformations in semi-dilute unentangled solutions that are highly suggestive of transient topological entanglements in solutions that are nominally unentangled at equilibrium. Taken together, these results suggest that the transient stretching pathways in semi-dilute solution extensional flows are qualitatively different than for both dilute solutions and for semi-dilute solutions in shear flow. In a second effort, we studied the dynamics of ring polymers in background solutions of semi-dilute linear polymers. Interestingly, we observe strikingly large fluctuations in steady-state polymer extension for ring polymers in flow, which occurs due to the interplay between polymer topology and concentration leading to chain 'threading' in flow. In a third effort, we developed a new microfluidic method to measure normal stress and extensional viscosity that can be loosely described as passive yet nonlinear microrheology. In particular, we incorporated 3-D particle imaging velocimetry (PIV) with the Stokes trap to study extensional flow-induced particle migration in semi-dilute polymer solutions. Experimental results are analyzed using the framework of a second-order-fluid model, which allows for measurement of normal stress and extensional viscosity in semi-dilute polymer solutions, all of which is a first-of-its-kind demonstration. Microfluidic measurements of extensional viscosity are directly compared to the dripping-onto-substrate or DOS method, and good agreement is generally observed. Overall, our work aims to provide a molecular-level understanding of the role of polymer topology and concentration on bulk rheological properties by using single polymer techniques.