

Dynamic Heterogeneity in Ring-Linear Polymer Blends Revealed by Single Molecule Studies

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Understanding the dynamics of ring polymers in fast flows is a particularly challenging yet interesting problem in polymer physics. Despite recent progress, a complete understanding of the nonequilibrium behavior of ring polymers has not yet been achieved. In this talk, I report the direct observation of DNA ring polymer dynamics in fast flows using single molecule techniques. In particular, we study ring polymer dynamics in semi-dilute blends of ring-linear polymers in planar extensional flow. Our results show that ring polymers fluctuate drastically in chain extension even at steady-state in extensional flow, yet ring polymers exhibit markedly less molecular individualism during transient stretching compared to linear chains. We hypothesize that ring polymer extension fluctuations arise due to threading of linear polymers through open ring polymer chains in flow. Ring fluctuation dynamics are quantified as a function of strain rate, concentration, and fraction of ring/linear chains in the blend. We further discuss the relaxation dynamics of single rings from high stretch in semi-dilute blends following cessation of extensional flow. Our results show the emergence of multiple molecular sub-populations underlying ring relaxation dynamics, and these results are compared to our recent work on the relaxation of purely linear polymers in entangled solutions. In particular, our results show dynamic heterogeneity in relaxation such that single polymer relaxation trajectories exhibit either a single-mode or double-mode exponential decay, which starkly contrasts relaxation behaviors from dilute solutions of ring or linear chains, and for semi-dilute unentangled solutions of pure linear chains. From a broad view, our results show that molecular behavior is markedly heterogeneous in non-dilute polymer solutions, which needs to be accounted for in model development and analytical theories.