Contribution ID: **76** Type: **Poster Only**

Measuring and modeling non-Gaussian deformations of topologically-complex polymers using in situ capillary rheo-SANS

Macromolecular topology provides an opportunity to engineer polymers that provide orthogonal control over rheological performance and mechanical stability in high-shear rate applications. In this work, we combine topology-controlled polymer chemistry with in situ neutron scattering measurements in a high-shear capillary device (capillary rheo-SANS) that can probe the rheology and microstructure of polymers in shear flows exceeding rates of 106 s-1. We synthesize and study high molecular weight, low-dispersity, topology-defined poly(methyl methacrylate-co-stearyl methacrylate) polymers with linear and star-shaped architectures for model studies of flow-induced deformation in dilute solutions at extreme shear rates. The resulting scattering is interpreted using a new modeling framework, the Gram-Charlier analysis of polymer scattering (G-CAPS) that fingerprints polymer conformations through non-Gaussian moments of the segment density distribution. Brownian dynamics simulations are used to show that the moments extracted from G-CAPS can be used to distinguish effects of finite extensibility in large strain-rate flows. We show that these measures, when extracted from capillary rheo-SANS measurements, provide a molecular-level explanation for differences in rheology and mechanical stability between linear and star polymers, as well as molecular-weight dependent effects of finite extensibility. More generally, we anticipate that capillary rheo-SANS in combination with G-CAPS will provide powerful new tools to engineer polymers rheology through macromolecular architecture.

Topical Area

Soft matter: polymers, and complex fluids

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