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How polymer entanglement responds to fast large deformation: are we there yet?[1]

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Nearly all polymeric materials are of high molecular weight and therefore entangled in their liquid state. Significant melt elasticity arises from the transient networking due to chain entanglement. All rheological behavior stems from how the entanglement responds to external deformation of various forms. Unfortunately, the concept of entanglement still remains theoretically elusive to describe. On other hand, modeling the evolution of chain entanglement is the key to answering the core questions in polymer rheology: a) where chain deformation comes from? b) when affine-like elastic molecular deformation ceases? In other words, yielding at both macroscopic (which is obviously taking place, e.g., signified by the stress overshoot response to startup shear) and molecular levels (through chain disentanglement) is an essential ingredient of any theoretical description of nonlinear polymer rheology. Macroscopic observations are valuable to afford useful insights, but it is the molecular dynamics simulations that are expected to address the foundational issues. This presentation will attempt to make a coherent discussion of what is known and where we are going from here.

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