Shear and Extensional Rheology of Polystyrene Melts and Solutions with the Same Number of Entanglements - DTU Orbit (09/11/2017)

Shear and Extensional Rheology of Polystyrene Melts and Solutions with the Same Number of Entanglements

We investigate the nonlinear shear and uniaxial extensional rheology of entangled polystyrene (PS) melts and solutions having the same number Z of entanglements, hence identical linear viscoelasticity. While experiments in extensional flows confirm that PS melts and solutions with the same Z behave differently, respective transient and steady data in simple shear over the largest possible range of rheometric shear rates (corresponding to Rouse-Weissenberg numbers from 0.01 to 40) demonstrate that melts and solutions exhibit identical behavior. Whereas the differences between melts and solutions in elongational flows are due to alignment induced friction reduction (more effective in melts than in solutions), in shear flows they disappear since the rotational component reduces monomeric alignment substantially. Recent molecular dynamics simulations of entangled polymers show that rotation induces molecular tumbling at high shear rates, and here a tube-based model involving tumbling effects is proposed in order to describe the response in shear. The main outcome is that tumbling can explain transient stress undershoot (following the overshoot) at high shear rates. Hence, the combination of tumbling in shear and friction reduction in extension successfully describes the whole range of experimental data and provides the basic ingredient for the development of molecular constitutive equations.

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