STRESS RELAXATION IN COMPLEX MODEL POLYMERS: COMBS AND RINGS

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Abstract

We present an overview of rheological investigations with model branched polymers. They key element is synergy of chemistry, experimental rheology and modeling. We show that all three elements are necessary to obtain a fundamental understanding of the molecular origin of the viscoelastic response of these systems.
In this particular presentation, we focus on two types of 'nonlinear' polymers, rings and combs, synthesized anionically.

Comb polymers of different chemistries (polyisoprene, polybutadiene, polystyrene) serve as model branched polymers for exploring aspects of the complex rheology of commercial polymers. They consist of a linear backbone, onto which several branches are grafted. Selecting the molecular weights of backbone and branches as well as the number of branches provides the means to tune the rheology of these macromolecules. We demonstrate that the concept of hierarchical motion (branches relax stress first, followed by backbone relaxation) governs the stress response to an imposed strain. Moreover, the tube model, accounting for the dynamic dilution effects, works well for highly entangled branches.

Recently, it was shown that entangled ring polystyrenes relax their stress in a self-similar manner. Moreover, the purity of the rings was shown to be an issue of utmost significance, and critical fractionation emerged as the key process necessary for understanding ring dynamics. Pure rings were shown to be extremely sensitive to linear chains 'contaminants'. These results were explained by invoking entropy maximization arguments and ideas from percolation theory. Here, we extend this work and attempt at addressing a number of open issues. We present experimental results with moderately entangled 1,4-polyisoprene rings, prepared under different conditions (mainly good solvent), confirm and extend the earlier findings, show universalities in behavior and outline open challenges.

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