

Viscoelasticity of transient networks from associating polymers: a mesoscopic modeling perspective

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Understanding and tailoring the viscoelastic response of polymer melts or concentrated solutions from the knowledge of their molecular structure (architecture) represents a formidable challenge and remains a prime field of soft matter research, with several important and fundamental questions still open. To this end, we have developed a general coarse-grained approach based on the tube model, that we are now using as a predictive tool in order to investigate the viscoelastic properties of complex, entangled polymer architectures. Our main objective is to elucidate the molecular origin of their relaxation processes and in particular, their constraint release mechanisms.

In this presentation, we first outline the construction of the time marching algorithm which represents the key ingredient of our approach, and then discuss some selected examples. Next, we extend this approach to describe the complex rheology of entangled macromolecular self-assemblies built from sticky polymers, and use our model as a new tool for an in depth analysis of their viscoelastic properties. In particular, we investigate the dynamics of a series of model telechelic linear and star polymers which create a reversible network through metal-ligand association, and study the effect of blending different architectures on the network relaxation. We also investigate the behavior of entangled polymers bearing stickers along their backbone. In all cases, we show that the dynamics of these associating polymers strongly depends on the ratio between entanglements and stickers density, as well as on the ratio between their association and disentanglement times, leading to a large variety of viscoelastic responses.