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Rheology of telechelic supramolecular polymers: Structure-properties relationship

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Supramolecular polymer networks offer unique properties due to their reversible and tunable bonds. However, understanding their viscoelastic response, particularly in the non-linear deformation regime, remains a challenge. In this work, we explore the interplay between entanglements and reversible junctions and their influence on the viscoelastic properties of model telechelic supramolecular networks.

We study well-defined systems composed of four-arm telechelic poly(n-butyl acrylate) stars (either entangled or not) physically crosslinked through terpyridine/metal ion bis-complexes (Zn^{2+}, Cu^{2+}, or equimolar blends forming dual networks), in melt.

To characterize the linear viscoelastic properties of entangled supramolecular networks, we extend a coarse-grained tube model approach [1]. Networks elasticity is governed by both the density of physical junctions and entanglements, while their relaxation time depends on the cooperative effects of sticker dynamics, dangling chains, and network topology. We compare systems with distinct compositions but similar linear viscoelastic responses, under high shear deformation, to rationalize the effect of strong flows on chain stretching, sticker dissociation and disentanglement mechanisms.

Understanding how these molecular-scale processes govern macroscopic response provides new opportunities for tailoring the mechanical properties of dynamic polymer networks.

References:

[1] de Wergifosse et al., Macromolecules 2025, 58, 222−239