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Theory of the nonlinear rheology of topologically entangled rod fluids KEN SCHWEIZER, DANIEL SUSSMAN, University of Illinois at Urbana-Champaign — Our first-principles microscopic theory of the tube confinement field and dynamics of topologically entangled rod fluids is extended to describe nonlinear rheology. Stress generically weakens tube constraints, resulting in a competition between reptation and transverse activated entropic barrier hopping. For a step-strain deformation, four distinct nonlinear relaxation regimes are predicted with increasing strain amplitude: quiescent-like reptation, strongly accelerated reptation due to stress-induced tube dilation, relaxation dominated by lateral barrier hopping, and, beyond a critical strain of order unity, an initially complete destruction of the tube constraint (microscopic yielding) followed by a re-entanglement process with complex kinetics and multi-step stress relaxation. A theory for continuous start up shear has also been formulated. In the nonlinear regime, deformation-rate-dependent stress overshoots are predicted. In the nonequilibrium steady state, strong shear thinning occurs determined largely by the rate-dependent dilated tube diameter. At very high Weissenberg numbers, a stress plateau in the flow curve is predicted and relaxation is controlled by a convective-constraint-release-like process that self-consistently emerges within the theory.

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