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Disentangling the Role of Entanglement Density and Molecular Alignment in the Mechanical Response of Glassy Polymers[1] THOMAS O'CONNOR, MARK ROBBINS, Johns Hopkins Univ — Glassy polymers are a ubiquitous part of modern life, but much about their mechanical properties remains poorly understood. Since chains in glassy states are hindered from exploring their conformational entropy, they can't be understood with common entropic network models. Additionally, glassy states are highly sensitive to material history and nonequilibrium distributions of chain alignment and entanglement can be produced during material processing. Understanding how these far-from equilibrium states impact mechanical properties is analytically challenging but essential to optimizing processing methods. We use molecular dynamics simulations to study the yield and strain hardening of glassy polymers as separate functions of the degree of molecular alignment and inter-chain entanglement. We vary chain alignment and entanglement with three different preparation protocols that mimic common processing conditions in and out of solution. We compare our results to common mechanical models of amorphous polymers and assess their applicability to different experimental processing conditions.

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