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Predictive relationships between crosslinker unbinding kinetics, gel stiffness, and plasticity in adhesive biopolymers

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We determine the viscoelastic responses of rigid rod polymer networks that have been strongly bonded by labile crosslinkers. Experimentally, we use microtubules, extremely stiff biopolymers that play important roles in maintaining the strength and organization of cells. We generate controllable adhesive bonds using well-characterized protein chemistries, such as biotin-streptavidin bonds, or using recombinant microtubule-associated proteins. Networks are visualized using confocal scanning fluorescence microscopy or transmission electron microscopy, and custom-built, high-force magnetic tweezers devices are used to apply localized forces to the gels. For rigid crosslinkers, we find that at short time scales, the networks respond nonlinearly to applied force, with stiffening at small forces, followed by a softening regime, which we attribute to the force-induced unbinding of crosslinkers. At long time scales, force-induced bond breakage leads to local network rearrangement and significant bead creep. Interestingly, the material retains its elastic modulus even under conditions of significant plastic flow, suggesting that crosslinker breakage is balanced by the formation of new bonds. These results provide important insight into the determinants of gel toughness, elasticity, and plastic deformation in rigid networks, but also suggest new avenues for materials optimization based on modulation of crosslinker kinetics. In particular, the incorporation of crosslinkers that break under force, but are competent to reform when the force is removed, significantly enhance gel toughness while minimizing material fatigue under cyclic loading.