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Physics and Mechanics of dual-crosslink gels[1]

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Dual crosslink hydrogels are swollen polymer networks containing a population of physical bonds dynamic and reversible), coexisting with a population of permanent covalent bonds. From the point of view of mechanical properties, the most interesting combination is a minority of sparse covalent bonds, providing extensibility, and a majority of dynamic bonds. At high strain rates the dynamic bonds provide additional stiffness while at low strain rates they are invisible. As the strain rate approaches the inverse of the relaxation time, the gel becomes markedly viscoelastic. We will present the physics and mechanics of several gels where the relative population of the two types of bonds and the lifetime of the dynamic bonds are systematically varied using complexation bonds. The dynamic properties will be characterized with linear rheology experiments while large strain and fracture experiments will be characterized with uniaxial tensile tests on notched and unnotched samples. Most notably we find that the presence of rapidly exchanging physical bonds can increase the extensibility at break, effectively delaying crack propagation, while having little effect on the large strain properties. We will discuss the mechanisms by which the cracks can be delayed and the connection between the relaxation time(s) of the gel in linear rheology and the fracture properties as a function of extension rate.

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