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Controlling toughness and dynamics of polymer networks via mussel-inspired dynamical bonds[1]

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For dry, thermoset, polymer systems increasing the degree of cross-linking increases the elastic modulus. However, it simultaneously compromises the elongation under tension, usually reducing the overall total energy dissipated before fracture (toughness). Dynamic reformable bonds and complex network topologies have been used to circumnavigate this issue with moderate success, mainly in hydrated network systems. Hydration, however, which swells these networks limits how far one could increase the modulus, while their chemistry prevents improvement of the mechanics upon drying. Employing the mussel byssus-inspired strategy of iron-catechol coordination bonds, we have synthesized and studied epoxy networks comprising covalently attached catechol moieties capable of forming additional iron-catechol complex cross-links that still function in dry conditions. In such a fashion, we create a high modulus, high elongation, high toughness material. The iron-catechol coordination bonds play multiple roles that enhance the mechanical performance of the system: at low strain and fast strain rates, they act like permanent cross-links with bonding strength similar to covalent bonds, but start disassociating at high elongation. They are also reformable, enabling material self-healing in a matter of minutes in the absence of load. Finally, the dissociative crosslink cleavage alters the local chain topology, creating length scales that unfold upon elongation. The elegance of this system lies on its general versatility. Both the polymer and metal ion can be used as control parameters to study the interplay of covalent and dynamical bonds as well as explore the limits of the design of elastomers with enhanced toughness.

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