Hybrid metal-coordinate transient networks: using bio-inspired building blocks to engineer the mechanical properties of physical hydrogels

SCOTT GRINDY, Massachusetts Inst of Tech-MIT, DEVIN BARRETT, PHILLIP MESSERSMITH, Northwestern University, NIELS HOLTEN-ANDERSEN, Massachusetts Inst of Tech-MIT — Recently, metal-coordinate complex crosslinks have been suggested to contribute to the self-healing properties of mussel byssi. Two specific amino acid derivatives - 3,4 dihydroxy-L-phenylalanine (dopa) and histidine (his) - are known to form coordinate complexes with trivalent and divalent ions (respectively) in aqueous solutions. We show here that, by functionalizing poly(ethylene glycol) polymers with dopa and his we are (1) able to characterize the fundamental kinetics and energetics of each specific metal-ligand pair using small amplitude oscillatory shear rheology and (2) create hybrid networks using various mixtures of metals and ligands. From this information, we can design gels with specific target mechanical properties by tailoring the amounts and types of metal-ligand crosslinks present in the gel network, resulting in the ability to engineer the mechanical relaxation spectrum. This work provides basic understanding necessary to intelligently design materials which incorporate metal-ligand crosslinks in more complex architectures.

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