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Bio-mimetic metal-ligand crosslinks yield self-healing polymer networks with near-covalent elastic moduli NIELS HOLTEN-ANDERSEN, University of Chicago, MATTHEW HARRINGTON, Max Planck Institute for Colloids and Interfaces, HENRIK BIRKEDAL, University of Aarhus, BRUCE LEE, PHILLIP MESSERSMITH, Northwestern University, HERBERT WAITE, University of California, Santa Barbara , KA YEE LEE, University of Chicago — Growing evidence supports a load-bearing role for metal-polymer interactions in biological protein networks. In particular, the strength of the coordinate bonds in metal-ligand coordination complexes combined with their capacity to reform after breaking has been proposed as a source of the high toughness and potential self-healing in certain natural materials. Some of the highest stabilities among metal-ligand coordination complexes are found between Fe3+ and catechol ligands at alkaline pH where the tris-catecholato-Fe3+ stoichiometry prevails, yet the effect of such crosslinks on material properties has not been fully characterized due to the low solubility of Fe3+ at high pH. Inspired by the pH jump experienced by marine biomaterials during secretion, we have developed a simple method to control catechol-Fe3+ inter-polymer crosslinking via pH. The resulting gels display elastic moduli (G') that approach covalently crosslinked gels as well as self-healing properties.

Niels Holten-Andersen
University of Chicago

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