Light-responsive viscoelastic timescales in bio-inspired metal-coordinate supramolecular hydrogel mechanics

SCOTT GRINDY, NIELS HOLTEN-ANDERSEN, Massachusetts Inst of Tech-MIT — Stimuli-responsive hydrogels are currently an active subject of research for biological structural applications such as adhesives. Conventional, covalently crosslinked hydrogels are typically too weak to function mechanically, so researchers have used dynamic and reversible crosslinks in attempts to improve mechanical performance. Recently, we showed that, in a PEG-based hydrogel crosslinked by bio-inspired Histidine:M2+ coordinate bonds, the characteristic mechanical relaxation timescale can be controlled by selecting the transition metal ion acting as the crosslink center and that by using mixed transition metals, we are able to design hydrogels with multiple hierarchical relaxation timescales while controlling the magnitude of the timescales by varying the relative metal concentrations. This platform of PEG-His hydrogels represents a straightforward method for creating hydrogel materials with precisely-engineered viscoelastic energy dissipation properties. Here, we expand on this platform by exploiting the chemistry of metal-coordinate complexes to create hydrogels with UV-responsive viscoelastic properties, where the oxidation state of the transition metal can be altered using UV light. Careful selection of the metal ion crosslinks and UV exposure allows a diverse set of (pre-UV properties) - (post-UV properties) pairs. Such a precise level of control over hierarchal energy dissipation modes enables optimization of hydrogel mechanics for a wide array of loading contexts.

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Date submitted: 11 Nov 2016
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