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Bio-inspired Self-healing Composite Hydrogel with Iron Oxide Nanoparticle as Coordination Crosslinker QIAOCHU LI, Massachusetts Institute of Technology, DEVIN G. BARRET, PHILLIP B. MESSERSMITH, Northwestern University, NIELS HOLTEN-ANDERSEN, Massachusetts Institute of Technology — Polymer-nanoparticle (NP) composites have attracted renewed attention due to enhanced mechanical strength combined with various functionalities, but controlling the interfacial chemistry between NPs and polymer matrix, which is crucial for the composite's mechanical behavior, remains a major challenge. Inspired by the adhesion chemistry of mussel fibers, we investigated a novel approach to incorporate Fe<sub>3</sub>O<sub>4</sub> NPs into hydrogel matrix. A polyethylene glycol polymer is designed with both ends conjugated by catechol groups, which have strong coordination affinity to Fe. The polymer network is crosslinked via coordination bonding at the surface of  $Fe_3O_4$  NPs, yielding a stiff nanocomposite hydrogel. Due to the reversible nature of coordination bonding, the hydrogel presents self-healing behavior. Oscillatory rheology allows comparative kinetic studies of self-healing driven by catechol bonding at  $Fe_3O_4$  NP interfaces and by catechol- $Fe^{3+}$  coordination complexes. Furthermore, the superparamagnetic property of Fe<sub>3</sub>O<sub>4</sub> NP is preserved after gelation, allowing for response to external stimuli. This gelation motif can serve as a versatile platform for tuning functional and mechanical properties for future polymer nanocomposite materials.

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