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Self-Healing Nanocomposite Hydrogel with Well-Controlled Dynamic Mechanics QIAOCHU LI, Massachusetts Inst of Tech-MIT, SUMEET MISHRA, North Carolina State University, PANGKUAN CHEN, Massachusetts Inst of Tech-MIT, JOSEPH TRACY, North Carolina State University, NIELS HOLTEN-ANDERSEN, Massachusetts Inst of Tech-MIT — Network dynamics is a crucial factor that determines the macroscopic self-healing rate and efficiency in polymeric hydrogel materials, yet its controllability is seldom studied in most reported self-healing hydrogel systems. Inspired by mussel's adhesion chemistry, we developed a novel approach to assemble inorganic nanoparticles and catechol-decorated PEG polymer into a hydrogel network. When utilized as reversible polymer-particle crosslinks, catechol-metal coordination bonds yield a unique gel network with dynamic mechanics controlled directly by interfacial crosslink structure. Taking advantage of this structure-property relationship at polymer-particle interfaces, we next designed a hierarchically structured hybrid gel with two distinct relaxation timescales. By tuning the relative contribution of the two hierarchical relaxation modes, we are able to finely control the gel's dynamic mechanical behavior from a viscoelastic fluid to a stiff solid, yet preserving its fast self-healing property without the need for external stimuli.

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