Abstract Submitted
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Designing Stress-adaptive Dense Suspensions via Dynamic Covalent Chemistry[1] GRAYSON JACKSON, University of Chicago, JOSEPH DENNIS, Army Research Laboratory (ARL), STUART ROWAN, HEINRICH JAEGER, University of Chicago — Dense suspensions exhibiting reversible non-Newtonian flow behavior typically possess relatively weak and short-lived particle scale interactions, yet it is unclear how manipulating the strength and lifetime of microscopic contacts perturbs these macroscopic rheological properties. We utilize the catalyst-free, room temperature dynamic bonds that form between thiol-coated particles and ditopic benzalcyanoacetate-based Michael acceptors to engineer dense suspensions with well-defined and tunable contact lifetimes. Steady shear rate or stress ramps reveal pronounced negative thixotropy which relaxes upon shear cessation, which we attribute to shear-induced dynamic covalent crosslinks between particles. Constant stress measurements reveal a transition from suspensions whose viscosities diverge as a function of strain at low stresses to those which undergo continuous deformation at high stresses. The exact stress required for this transition systematically depends on the electron-donating/withdrawing nature of the Michael acceptor, which is known to control the dynamic bond strength and lifetime. We anticipate these connections between contact lifetimes and macroscopic rheology will aid in the design of smart materials which autonomously sense and adapt to applied stresses.

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