MIT Chemical Engineering Department Fall 2019 Seminar Series

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"Phase Mechanics of Arrested Colloidal Gels: A New Paradigm for Non-Equilibrium Phase Transitions in Soft Matter"



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Abstract: In colloidal gels and glasses, kinetically-arrested phase transitions exert a remarkable influence on material behavior, yet their structure-property relationships remain challenging to model. The physical and chemical similarities between molecular systems and interparticle colloidal attractions led to the development of colloidal phase diagrams, where metastable and unstable phase separation closely parallels that seen in molecular systems. Thus, colloids have long been viewed as paradigmatic model systems for molecular phase transitions where the vast separation of timescales between colloidal and solvent particles provides a means by which to "slow down" relaxation processes and study phase behavior. However, colloidal gels represent "arrested" states of phase separation, where the same interparticle attractions that promote phase separation also inhibit it, freezing colloids into a non-equilibrium microstructure that forms a viscoelastic network. Despite attempts to place them on equilibrium phase diagrams, such gels do not belong there; rather, temporal evolution is required to describe their "state". We show that when interparticle bonds are O(kT), thermal fluctuations enable ongoing particle migration and a (logarithmically) slow march toward full phase separation. Our work reveals the surprising result that gel yield can occur with the loss of fewer than 0.1% of particle bonds and no network rupture; instead, localized re-entrant liquid regions permit yield and flow. Analysis of the evolving osmotic pressure and potential energy reveals the interplay between bond dynamics and external stress that underlies mechanical yield and provides a compelling connection to stress-activated phase separation. I will show that external forces open a pathway of escape from arrest toward equilibrium, and propose a non-equilibrium phase diagram as the foundation for "phase mechanics," a new view of states of arrested colloidal matter.