Department of Chemical Engineering Seminar Series

"Phase Mechanics" of arrested colloidal gels: A new paradigm for non-equilibrium phase transitions in soft matter

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Abstract

Understanding kinetically arrested phase transition in complex media, and its influence on structure-property relationships, has been identified as one of the Grand Challenges for the future of soft matter science. Colloidal gels and glasses are an important class of such materials and are the subject of an emergent field of study in which much focus is placed on predicting yield behavior. More fundamentally, the physico-chemical nature of inter-particle colloidal attractions has permitted the construction of colloidal phase diagrams via molecular theories, where metastable and unstable phase separation closely parallels that in molecular systems. As such, colloids have long been viewed as paradigmatic model systems for molecular phase transitions, but 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 in a non-equilibrium microstructure to form a visco-elastic network. In contrast to attempts to place them on equilibrium phase diagrams, we argue that such gels must exit the equilibrium phase diagram. 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 loss of fewer than 0.1% of particle bonds, with no network rupture; rather, 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 fields and forces open a pathway of escape from arrested phases toward equilibrium, and I will propose a 'non-equilibrium phase diagram as the foundation for "phase mechanics", a new view of states of arrested colloidal matter..

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