253i Dynamic Criterion for the Equilibrium Percolation Threshold of Weakly Attractive Colloids

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We investigate equilibrium structures and dynamic characteristics of percolating colloidal systems as a function of particle concentration and pair attraction. Previous measurements of a fluid-gel transition of attractive polymer coated colloids are compared with Monte Carlo (MC) simulations of colloids having either realistic van der Waals (vdW) or model adhesive sphere (AS) potentials. Analytical expressions are used to compute mutli-body self diffusion coefficients for the MC simulated configurations of vdW colloids to address two issues related to identifying the percolation threshold, including: (1) ambiguous definitions of connectedness, clustering, and percolation, (2) the possibility of an associated frequency dependent rheological transition. As expected, our results do not display any thermodynamic signatures of the percolation threshold based on relaxation rates proportional to $[1+2\Phi g(r=2a)]^{-1}$ that contribute to the long time self diffusion coefficient and zero frequency viscosity ($D_S^L \sim \eta_0^{-1}$). However, our results do indicate an abrupt change in the short time self diffusion coefficient and high frequency viscosity ($D_S^S \sim \eta_\infty^{-1}$) for vdW colloids at the same percolation threshold conditions as AS colloids with equivalent second virial coefficients. Because D_S^S is determined exclusively by multi-body dissipative hydrodynamic interactions, the percolation threshold corresponds to a purely dynamic transition of weakly attractive vdW colloids. This result indicates a unique dynamic criterion for the equilibrium percolation threshold and provides novel insights into frequency dependent relaxation mechanisms of weakly attractive colloids short of irreversible gelation or arrested glass formation.