Capillary Stress and Structural Relaxation in Disordered Granular Materials

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Abstract

Capillary condensation is a ubiquitous process of vapor-liquid phase transition in porous media, such as sand piles, plaster, silica gels, cementitious materials, which has an important yet poorly understood effect on mechanical behavior. The confined fluid can generate enormous local stresses, as observed in granular material aging, wet floor friction, nanotribology, cement drying shrinkage and in everyday life experiences such as hardening of a drying sponge or building a sand castle on the beach. Capillary condensation and evaporation potentially bring undesirable fracture processes,as in drying cracking of colloidal films and paints, but capillary stress can also can be exploited innano-materials fabrication by capillary force lithography, capillary rise infiltration, evaporation-driven assembly and self-organization and composite imbibition and even used to evaluate the atmosphere of planets. Despite the broad importance of capillary forces, they remain challenging to predict in complex porous materials over the full range of liquid saturation, either in equilibrium or during a dynamical process of drainage/imbibition. For granular or colloidal materials, existing models addressing partial saturations are oversimplified and only apply either to low humidity (so called pendular, funicular regimes) or to idealized geometries (slit/cylindrical independent pores or single sphere against a wall). At higher saturation, models based on geometrical analysis of Young-Laplace equation for smaller clusters are proposed but restricted to only equilibrium liquid distributions inside mono disperse packings. Molecular simulations methods are also difficult to use, since the porous structure considered at the mesoscale (micron-scale).
In this work, we propose a numerical and theoretical framework to quantitatively predict capillary condensation/evaporation, compute associated capillary forces and structural relaxation in a 3D realistic nano-granular cement paste model using lattice-gas simulations of

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adsorbed water parametrized from atomistic simulations. In particular, this allowed us to access the adsorption-desorption mechanism and assess the role of (metastable) cavitation. In addition, we present for the first time to our knowledge a phase field model of the liquid-vapor mixture spatial distribution, whose inhomogeneous stress tensor is integrated over Voronoi polyhedra in order to calculate forces on each grains. Minimizing system free energy upon the conjugate action of capillary forces applied to the cement hydrate nano-grains, together with the cohesive interactions between these nano-grains predict the overall stress relaxation. We show that capillary stress is an effective mechanism for eigenstress relaxation in granular heterogeneous porous media, which contributes to the durability of cement.

Keywords: capillary stress, drying shrinkage, mesoscale modeling, fluid solid interaction