

Hysteresis in Contact Angle and Interfacial Tension: Implications on Multiphase Flow

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Abstract:

Multiphase fluids are common in the subsurface and affect a wide range of natural and engineering process from infrastructure performance above unsaturated soils, to contaminant transport, environmental remediation, oil and gas recovery, and CO₂ geological sequestration. The capillary pressure $P_c = \kappa \cdot \gamma \cos \theta$ between two immiscible fluids is proportional to the interfacial tension γ , the contact angle θ and the interface curvature κ . Both contact angle and interfacial tension may show significant hysteresis.

Instead of a single thermodynamic contact angle predicted by Young's equation, fluid-liquid-mineral interfaces exhibit contact angles that can vary between the asymptotic advancing θ_A and receding θ_R contact angles. Typical explanations involve surface roughness and chemical inhomogeneity; yet, contact angle hysteresis is observed even on smooth, homogeneous surfaces. The normal component $\gamma \cdot \sin \theta$ of the fluid-liquid surface tension underscores the attractive interaction between the solid surface and the contact line where the three phases meet. Experimental results show that contact line adhesion is highest under static conditions and decreases to the kinetic adhesion during sliding, causing the stick-slip motion of the fluid-liquid interface. Consequently, DI water, brine, surfactant solutions and nanofluid droplets resting on smooth and homogeneous silicon wafers and quartz glass substrates exhibit distinct advancing and receding behaviors. In particular, surfactant molecules and nanoparticles adsorbed at interfaces enhance contact pinning, alter the contact line behavior during advancing and receding and result in unique contact angle hysteresis.

The presence of surface-active species such as ions, surfactants and colloidal particles changes the interfacial tension. The Gibbs adsorption isotherm relates the interfacial tension to surface excess concentration relative to the bulk concentration under thermodynamic equilibrium. However, a migrating fluid-fluid interface is continuously evolving as it traverses interconnected pores through converging and diverging cross-sections. Interfacial tension hysteresis induced by surfactants, nanoparticles and asphaltene in crude oil exhibit distinct interfacial tension hysteresis patterns that reflect the properties of adsorbed species, and both the strain and strain rate the fluid-liquid interface experiences.

Experiments and pore network analyses show that contact angle hysteresis and interfacial tension hysteresis work collectively to affect the migration of fluid-fluid interfaces across a pore constriction, invasion patterns and the resultant distribution of phases.