

**Introduction and Background** This research project studies electrocoalescence, the process of forcing the coalescence of water droplets in an oil or petroleum crude by the application of an electric field across the droplets. The field polarizes the droplets, creating a separation of charge and a dipolar attraction which results in the droplets merging. The focus of the study is to examine the role of colloidal particles, present in the oil phase, and adsorbed onto the water/oil interfaces of the droplet, on the coalescence. The particles provide a steric barrier to the coalescence, requiring a larger field to merge the droplets. The objectives are: (i) to use microfluidics to directly visualize the electrocoalescence of an emulsion in two dimensions, identify coalescence events, and measure the event frequency as a function of the applied electric field, the surface chemistry of the colloids (hydrophobic/hydrophilic) and the particle suspension density, (ii) design a microfluidic arrangement to measure the dielectric properties of colloidal particles by examining the motion of the colloids driven by dielectrophoretic forces, (iii) simulate the hydrodynamics of colloids trapped at, and moving along an interface, and (iv) simulate the electrostatics and hydrodynamics of the electrocoalescence process to predict merging events, in emulsions without and with particles. Progress on the last two objectives are reported below.

## Research Progress

### (i) Pairwise Hydrodynamic Interaction of Particles Along a Planar Fluid Interface

We studied the hydrodynamic interaction of two identical spherical particles (radius  $a$ ) floating on a flat gas-liquid interface (low capillary number). The particles translate along the surface with the particle immersion depth into the liquid ( $d$ , as determined from the contact angle or particle wettability) assumed fixed. The hydrodynamic resistance coefficients for Stokes flow corresponding to the complete set of pairwise relative motions were calculated as a function of the interparticle separation distance ( $\ell$ ) and the particle immersion depth ( $d$ ), including in-tandem motions along and perpendicular to the line of centers, and oppositely directed motion along and perpendicular to the line of centers. For each relative motion at a particular immersion depth, we have computed a scaled resistance or drag coefficient which is the resistance coefficient divided by the drag coefficient of an isolated particle translating along the surface at the same immersion depth. For in-tandem motions, we find that the scaled resistance coefficients for different immersion depths collapse into one curve as a function of separation distance. This curve corresponds to the well known curve for the hydrodynamic resistance of two particles completely immersed in the liquid moving in-tandem and along line of centers and perpendicular to line of centers. For oppositely direct motions, the scaled resistance coefficients do not collapse at different immersion depths to the corresponding curves for an infinite medium. These scaled resistances are a strong function of the immersion depth. In particular when the particle preferentially wets the liquid (contact angles less than  $\pi/2$ ) the scaled resistance increases without bound as the separation distance tends to zero because of the unbounded increase in the lubrication forces. For oppositely directed motions in which the particle resides predominantly in the air phase, the scaled resistance tends to a finite value at zero separation since at contact fluid remains entrained in the gap between the particles.

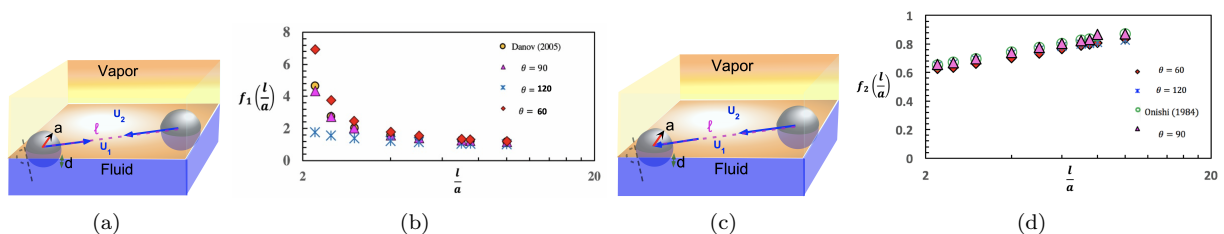
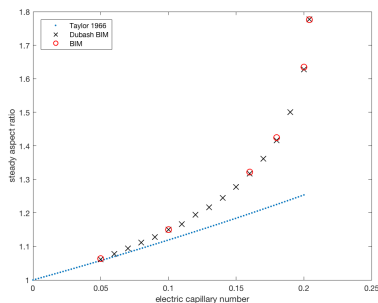


Figure 1: a) Oppositely directed motion along the line of centers for two particles translating towards each other on the surface of a semi-infinite liquid and (b) the normalized drag coefficient as a function of separation distance and (c,d) in tandem motion along the line of centers and the normalized drag coefficient.

Only for neutral wetting do the scaled resistances collapse to the curves for particles immersed in an infinite medium, due to the symmetry of the motion (Fig. 1).



### (ii) Numerical Simulations of Electrocoalescence

The hydrodynamics of the electrocoalescence of a pair of droplets merging along their line of centers was simulated for the case of inertialess flow (low Reynolds number) by a boundary integral method. The simulation assumes the droplets are perfectly conducting liquids immersed in an immiscible continuous phase which is a non-conducting (dielectric) liquid with a viscosity ( $\mu$ ) equal to the droplet phase and a dielectric constant  $\epsilon_o\epsilon_{oil}$ . The interfacial tension of the drop interface is  $\gamma$ . The droplets are placed a distance  $\ell$  (edge-to-edge) apart in the dielectric liquid, where they are assumed spherical with radius  $a$ . A uniform electric field with magnitude  $E$  is imposed

along the line of centers, and the droplets proceed to deform into prolate-like shapes as they approach one another. The deformation, due to the action of the field on the accumulated surface charge on the drop surface, is characterized by the electrocapillary number,  $Ca = \epsilon_o\epsilon_{oil}E^2a/\gamma$  and simulations are undertaken at different initial separation distances and capillary numbers. As a first step, the code is verified by simulating the interfacial deformation of a single conducting droplet in the dielectric. In this case, the droplet deforms in a prolate shape along the direction of the field. The figure to the left reports the aspect ratio of the steady deformation as a function of the electrocapillary number; the turning point in the curve represents the critical capillary number beyond which steady shapes are not possible. The simulations are shown to be in good agreement with the simulations Dubash et al (JFM, 581, 469 (2007)) which verifies the code. Fig. 2a reports the deformation of a droplet pair with an initial separation equal to  $2a$  and  $Ca=0.2$  for a nondimensional time of approximately 17 (time scaled by  $a\mu/\gamma$ ). The interfacial shape, on the scale of the droplets (Fig. 2a), approaches a conical form for the portion of the droplets facing one another due to the intensification of the field between the droplets. The deformation on a finer scale is shown in Fig. 2a which demonstrates that the cone is flattened due to the viscous resistance to remove the fluid between the droplets in the region of closest approach. This figure shows the emergence of a small dimple due to the flattening of the cone tip. When the initial separation distance is reduced to  $0.1a$ , the viscous resistance of

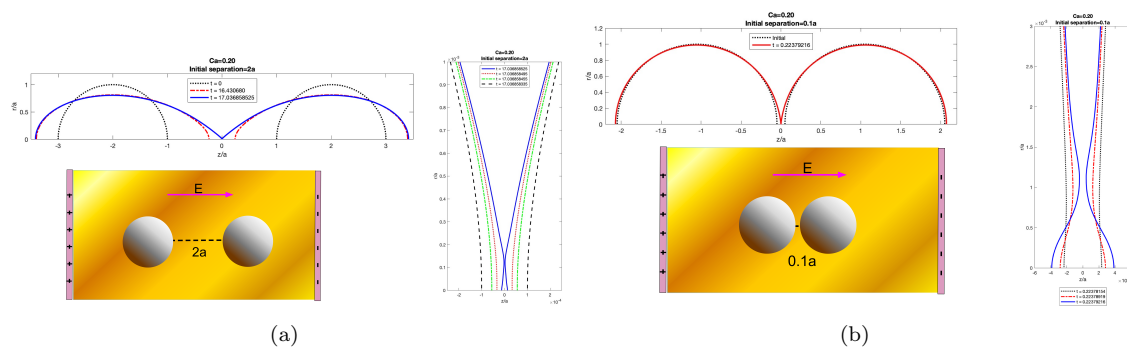


Figure 2: (a) the deformation and approach, due to electrocoalescence, on the droplet scale, of two droplets initially separated by  $2a$  for  $Ca=0.2$ , and the deformation on a finer scale in the region between the droplets; (b) deformation and approach, due to electrocoalescence, on the droplet scale, of two droplets initially separated by  $0.1a$  for  $Ca=0.2$  and the deformation on a finer scale in the region between the droplets.

the intervening fluid prevents the formation of a cone shape and a larger dimple forms (Figs.2b). This large dimple indicates that upon coalescence the conducting fluid may become trapped in the coalesced dielectric drop. Future work will reconstruct the drop interfaces upon coalescence and begin to treat the presence of colloid particles on the droplet surfaces.

Two doctoral student are undertaking the simulations and they are in their third year of study.