ABSTRACT

Electrical modulation of interfaces has emerged as a thriving research topic owing to its promise towards defeating the fundamental challenge of overwhelming viscous forces compared to inertia in microscale fluid flow. Diverse practical applications, with droplet-based multiphase microfluidic devices on the one hand to targeted drug delivery using artificial microswimmers on the other, have inspired the researchers to exploit its advantages in several new prospects in this field. Despite outstanding advancements, however, numerous issues on interfacial electro-hydrodynamics remain to be poorly understood, including electrically modulated dynamics of chemically tuned droplets and their possible synergistic connectivity with the electrically modulated dynamics of complex active matter. Motivated by these, the present thesis explores several unresolved facets of the dynamics of surfactant-coated drops in an electric field and the influence of complex biofluids on the locomotion of auto-electrophoretic micromotors as a model active matter system.

Surfactants are essential ingredients of the droplet-based microfluidic technology, facilitating the stabilisation of droplet interfaces, augmenting the biocompatibility of the system and acting as media of molecular exchange between droplets. However, diffusive transport of surfactants leads to spontaneous gradients in interfacial tension, which in turn, may influence the electrically mediated dynamics of droplets in a rather profound manner. Motivated by this proposition, first, a generic theoretical formalism is developed that accounts for the interfacial tension gradients due to bulk-insoluble non-ionic surfactants. Interface deformability and the surface charge convection are implemented within the scope of the small deformation theory and small electric Reynolds number (defined as the ratio of the timescales of charge convection by flow and that for charge relaxation by ohmic conduction), respectively, while the electrical effects are modelled with the leaky dielectric model. It is shown that a non-trivial interplay between the hydrodynamic and Maxwell (electrical) stresses at the drop surface, being implicitly connected with the surfactant dynamics, as mediated by the incipient flow field, leads to unique facets of drop migration and emulsion rheology.

In a subsequent analysis, Marangoni effect, originated out of spatial variations in the interfacial tension at the drop interface, is shown to arrest an otherwise increment of the drop-settling velocity as attributable to surface charge convection, under gravitational settlement. For specific combinations of the electrical parameters, the Marangoni flow completely suppresses the additional secondary vortices observed in a clean drop. Further, under the influence of a background Poiseuille flow profile, contrasts in electrical conductivity and permittivity of the drop-surrounding fluid pair, as well as the magnitude and the orientation of the electric field play a significant role in selectively altering the implications of the Marangoni stress, thereby providing a means for selectively controlling the drop velocity in different directions. This leads to the design paradigm of switching the cross-stream migration of the drop and controlling the time for reaching the final state in the trajectory, by modifying the nature of the surfactant and the sensitivity of surface tension on the concentration of the same.

The deformation of a droplet in a simple shear flow is of fundamental importance in several industrial and biological processes, including the formation and rheology of emulsions, emulsifying devices, polymer blending, oil recovery, break-up of fuel droplets in a double-swirl combustion chamber of a jet engine, and the study of red blood cells in a physiological conduit. Accordingly, the electro-hydrodynamically mediated transport of a surfactant-laden droplet in a background shear flow is subsequently investigated. A three-dimensional asymptotic theory in this regard leads to an apparently surprising observation that at a critical value of the shear stress relative to the electrical stress, the Marangoni effects surprisingly vanish, despite the presence of surfactant gradients. Consequently, the bulk emulsion viscosity and the normal stress differences can either enhance or decline, as determined by unique discriminating functions.

The above investigations develop the necessary background of extending the electro-hydrodynamic transport of chemically functionalised inert droplets to biologically active micromotors. Such micromotors have emerging applications in medicine, with their potential capabilities of delivering materials to living cells within an organism. They also may turn out to be effective in degrading certain chemical and biological warfare agents. Motivated by such important implications, the swimming performance of an artificial micromotor driven by enzyme catalysis in a biophysical environment is subsequently investigated. The interplay of electrocatalysis phenomenon and non-Newtonian fluid rheology is captured by employing a unified model for shear-thinning and shear-thickening fluids that are otherwise attributed to distinctive time-relaxation characteristics. It is shown that shear-thinning fluids hold the exclusive capability to bring in massive augmentations in the mechanical power developed by the micromotor relative to the input chemical energy. Further, it is revealed that a critical competition between the increased swimming speed and the reduced drag force results in optimal efficiency points, as dictated by the fuel concentration, catalytic reaction rate constants, and the micromotor size.

While the studies on droplets and active micromotors, as reported in this work, are all centered around electrically-mediated manipulation over interfacial scales, the respective strategies for the same are subtly contrasting. Whereas the electric field effect considered for manipulating the droplet is externally applied, the electric field modulating the micromotor transport is self-generated by internal bio-chemical activation, very much akin to the electro-chemical activities in living systems. Despite such obvious disparities, the mathematical framework for delving into the underlying physics is grossly universal, connecting several paradigms of inert and active matters in an electrically modulated fluidic environment. These findings pave the way for optimising the performance of various droplet-based lab-on-a-chip devices, as well as apprehending the locomotion strategies of microswimmers in physiologically relevant complex environments in a unified paradigm, and venturing into further experimental and numerical studies in this field to build up specific focused applications in this emerging field, including a unified paradigm of electrically modulated functionality of biologically active droplets, bearing connectivities with features such as decay, collective behaviour, and self-division that are essential aspects of functioning of living organisms.

Keywords: leaky dielecric fluid; drop deformation; charge convection; electrohydrodynamics; surfactants; Marangoni stress; Janus particle; electrocatalysis; self-electrophoresis; biofluids; rheology.