



Transient Marangoni transport of colloidal particles at the liquid/liquid interface caused by surfactant convective-diffusion under radial flow



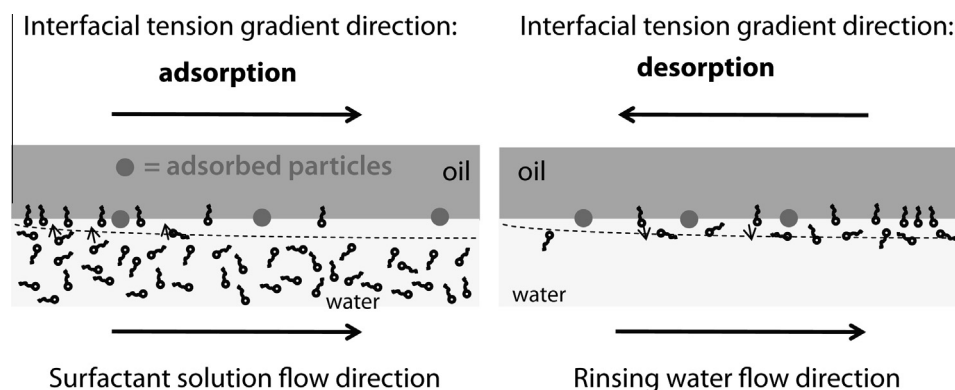
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GRAPHICAL ABSTRACT



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ABSTRACT

Hypothesis: Interfacial tension gradients at a liquid/liquid interface drive Marangoni flows. When colloidal particles are adsorbed to an interface in systems with spatial and temporal gradients of surfactant concentration, these interfacial flows can be potentially significant contributors to the direction and rate of particle transport.

Experiments: In this work, we use optical microscopy to measure the interfacial velocities of 5 μm diameter polystyrene latex particles adsorbed at an oil/water interface, using olive oil to represent polar oils often encountered in cleaning applications.

Findings: On surfactant adsorption the maximum interfacial velocity scales linearly with bulk surfactant concentration, even for concentrations exceeding the critical micelle concentration (CMC). The maximum interfacial velocity weakly decreases with increasing flow rate, but it varies non-monotonically with the radial distance from the inlet. Upon surfactant desorption into a rinse solution, the maximum velocity increases with increasing concentration of the original surfactant solution, but only up to a plateau near the CMC. These experimental trends are well-described by a convective-diffusion model for surfactant transport to or from the liquid/liquid interface coupled with Langmuir-type adsorption, using a constitutive relation between the interfacial tension gradient and interfacial velocity based on the interfacial tangential stress jump.

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1. Introduction

Marangoni flows occur at fluid interfaces in response to interfacial tension gradients. These gradients can be caused, for example, by differences in solvent composition or in surfactant surface excess concentration. Marangoni flows are important in the semiconductor and coating industries as well as in drug delivery and enhanced oil recovery applications. An example of the first is Marangoni drying, used in the semiconductor industry to obtain ultra-clean surfaces, wherein an organic vapor adsorbs near the three phase contact line on a partially submerged substrate, locally decreasing the liquid phase surface tension to drive the cleaning fluid to a bulk reservoir [1–4]. Marangoni flows associated with surfactant-laden aerosols are envisioned to enhance the uniformity of pulmonary drug delivery [5–7], and Marangoni stresses can help drive surfactant replacement therapy formulations to penetrate as liquid films deeply into the lung for treating respiratory distress syndrome [8–10]. Furthermore, surfactant-induced Marangoni stress can control the mobility of an interface, offering a means to engineer the lifetime of foams or emulsions relevant to enhanced oil recovery [11,12].

Marangoni spreading of localized surfactant deposits placed on fluid substrates has attracted much attention; see for example the review of Craster and Matar for a comprehensive summary [13]. Most of these studies consider the air–liquid interface. Considerably less effort has been devoted to liquid/liquid interfaces. A recent example of the latter is a study by Berg who used laser shadowgraphy to observe surfactant spreading along the water–decane interface [14]. When surfactants are deposited in a localized region on a fluid subphase, the expanding surfactant front takes the form of a capillary ridge that can travel in excess of 10 cm/s. Thus, surfactant driven Marangoni convection can be a potent transport mechanism at liquid/liquid interfaces. Another circumstance where Marangoni stresses have major effects is the control of drop or bubble shape when there is relative motion between two immiscible fluids when one of those fluids is a surfactant solution [15–18].

The current study concerns surfactant solution flow past a nominally flat interface with a more viscous, immiscible liquid. Rather than an instantaneous, highly localized placement of a surfactant deposit directly on the interface to create an abrupt interfacial tension gradient, it involves surfactant mass transfer between the interface and a solution in laminar flow. The surface excess concentration and interfacial tension gradients are created over significantly larger length- and timescales compared to the abrupt placement of a surfactant deposit.

This scenario is relevant to a variety of applications that involve unsteady mass transfer of surfactants to or from liquid/liquid interfaces under flowing solution conditions. Especially in cleaning applications, the interface may contain adsorbed particles, and these may be entrained in the resulting Marangoni flows. Surfactant adsorption and particle deposition are important factors in detergency, emulsion formulation, food processing, and a variety of personal care product applications. For example, in detergency applications, particles may adsorb at oil/water interfaces, and the transient Marangoni stresses that form during application of the surfactant solution as well as during the subsequent rinse may drive particle transport. In the realm of hair shampoos and dermatologic products, the sebum/water interface can be a transport locus for particulate active agents. The influence of Marangoni flows that arise during product application or rinsing on particle transport in such situations has not been considered previously.

The objective of the current study is to determine whether Marangoni stresses arising from non-uniform surfactant mass transfer rates significantly alter interfacial transport of particles relative to what occurs in the absence of surfactant effects and

the conditions that control the strength of these effects. The work is conducted with a radial stagnation point flow of an aqueous solution impinging from underneath a confined oil film to provide a well-defined flow field for analysis. Spatially varying surfactant adsorption and desorption rates arise as a result of the spatially varying concentration boundary layer thickness that forms as a surfactant solution flows over a clean interface or when a surfactant-free rinsing solution flows over an interface with pre-adsorbed surfactant [19,20]. On solid surfaces which do not allow a Marangoni flow, it is well-established that mass transfer rates of surface active molecules normal to the interface decrease in the direction of the solution flow under transport-limited conditions [19–21]. This leads to transient surface excess concentration gradients that eventually disappear as the surface saturates during adsorption or becomes bare during rinsing. If instead of an immobile solid surface, the interface is a liquid/liquid interface, the transient surface excess concentration gradients will produce interfacial tension gradients necessary for Marangoni flow.

We report on adsorbed particle displacements, velocities and timescales associated with adsorption and desorption of sodium dodecyl sulfate (SDS) in the aqueous phase impinging on a film of olive oil to represent polar oils commonly encountered in detergency, personal care and dermatologic applications. Results are reported for different bulk aqueous flow rates and different surfactant concentrations, both below and above the critical micelle concentration (CMC). We compare experimental results with a convective-diffusion model for surfactant adsorption and desorption with a Langmuir adsorption mechanism at the interface. The model imposes consistency between transient interfacial transport processes and equilibrium interfacial tension data. Comparing model and experimental results shows that spatially non-uniform surfactant mass transfer associated with laminar flow past an interface can produce strong Marangoni excursions in the interfacial particle velocity. The strength of the effect is highly sensitive to adsorption and desorption kinetic rate constants as well as surfactant concentration, solution flowrate and location in the flow field.

2. Experimental

2.1. Materials

All water was Milli-Q purified (18.2 M Ω cm resistivity, Millipore). Sodium dodecyl sulfate (electrophoresis grade, Fisher) was washed with hot acetone and dried to purify before use. All surfactant solutions were prepared just prior to use. Particles were surfactant-free, sulfate-stabilized polystyrene latex microparticles (5.2 μ m in diameter, Life Technologies). A single batch of extra virgin olive oil was used as the model polar oil.

2.2. Radial stagnation point flow cell

In order to quantify the Marangoni convection of particles adsorbed at the oil/water interface, a radial stagnation point flow cell was constructed, similar to that described by Goldstein and DiMilla [21], that would allow a radially dependent range of shear stresses to be sampled in each experiment. Optically clear poly(methylmethacrylate) (PMMA) served as both the flow cell body and the substrate supporting the oil film, allowing for direct microscopic observation of adsorbed polystyrene microspheres. Fluid entered through a 2 mm inlet and diverged radially through a 370 μ m gap past a 30 μ m deep oil film coating the PMMA substrate. Eight outlets for the aqueous flow were distributed uniformly around the outer circumference. See the [Supporting Information](#) for a schematic of the flow cell. The flow cell was

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