



Contents lists available at ScienceDirect

Colloids and Surfaces A: Physicochemical and Engineering Aspects

journal homepage: www.elsevier.com/locate/colsurfa



Effect of surfactant concentration and viscosity of outer phase during the coalescence of a surfactant-laden drop with a surfactant-free drop

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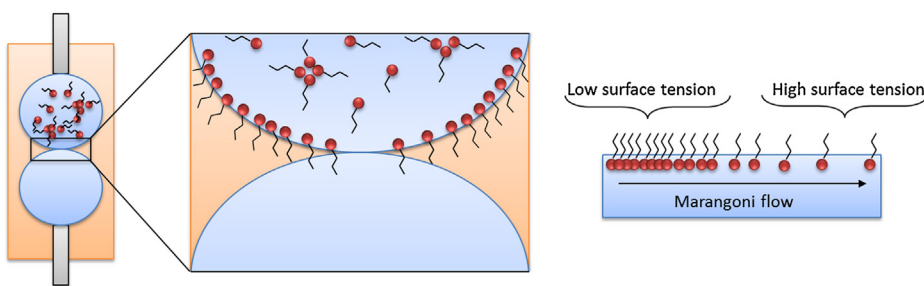
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HIGHLIGHTS

- Coalescence of drops in oils of various viscosities follows the same power scaling law.
- Marangoni flow induced by the interfacial concentration gradients is observed.
- Coalescence of dissimilar drops is accompanied by a considerable bulk convection.
- Convective patterns depend on the viscosity of the outer phase.
- Asymmetric bridge meniscus is due to local differences in interfacial tension.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 20 January 2016
Received in revised form 9 February 2016
Accepted 10 February 2016
Available online xxx

Keywords:

Kinetics of coalescence
Marangoni flow
Convective mixing
Surfactant transfer

ABSTRACT

This work focuses on the coalescence of two water drops, one of which contains surfactant. The coalescence was carried out in surrounding silicone oils of various viscosities. It has been found that, in all the studied cases, the outward motion of the liquid neck follows the power scaling law with exponent ~ 0.5 with the pre-factor dependent upon the surfactant concentration and viscosity of outer phase. Interfacial Marangoni flows arising at coalescence were visualised and quantified. Considerable convective bulk motion was observed by coalescence of surfactant-laden and surfactant-free drops with patterns depending on viscosity of outer phase. No noticeable convection was observed during the coalescence of two surfactant-free drops or similar surfactant-laden drops. It was confirmed that the rate of growth of the liquid bridge was reduced when the surfactant was present in the drops due to the lower interfacial tension in the bridge. Dependence of the reduction on surfactant concentration and viscosity of surrounding liquid is discussed. For the coalescence of a surfactant-free drop and a surfactant-laden drop, the curvature of the meniscus was different on either side of the growing bridge.

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1. Introduction

Rapid expansion of formulated products signifies a need to understand the development of bespoke drop designs. For example, recent developments in microfluidics convey new possibilities and precise control of drop coalescence that can be exploited for a plethora of functional products and processes. For instance, aqueous drops in oil can be considered as independent reactors, relevant for many applications [1], hydrogel bead generation [2] or nanopar-

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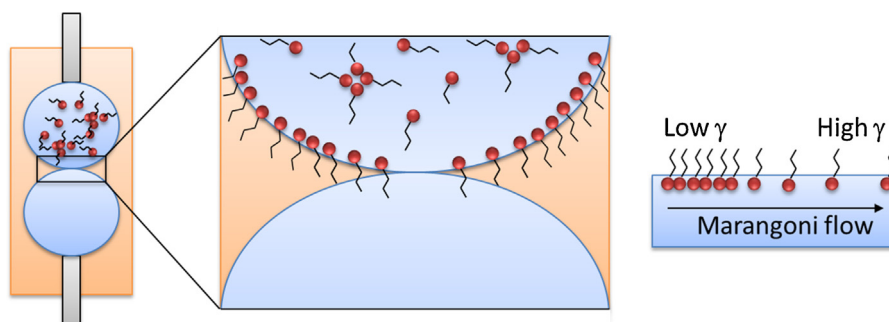


Fig. 1. Schematic representation of induced Marangoni stresses in the coalescence of drops with different composition.

ticle synthesis [3]. These applications and processes are not limited to the coalescence of drops that have similar composition, on the contrary, the possibility of merging dissimilar drops to initiate mixing, fusion, polymerisation or any kind of reaction in a highly controlled manner results in formulation of bespoke multiphase structures which deliver specific functions via novel manufacturing routes. Understanding of the detailed dynamics of such complex multiphase flows is of crucial importance for process development and optimization.

When the fluid film separating two drops ruptures, coalescence occurs spontaneously to form a bigger drop. The coalescence is driven by the capillary force and balanced by the viscous forces at early stages (or highly viscous drops) and inertial forces at longer timescales (or low viscosity drops) [4–6]. However, a few recent studies can be found that employ these theories developed for the dynamics of coalescence in inviscid surrounding liquid to the case where outer liquid is present [7,8].

Most studies on the coalescence of surfactant-laden drops in a surrounding liquid focus on the liquid film drainage just before the drops make contact [9,10]. In this case, a viscous surrounding liquid is present and the flow type and rate of the liquid film drainage from in-between the drops is considered [11,12]. However an open question remains on how the surfactant affects the coalescence after the drops make contact. Presence of the surfactant on the interface reduces the surface tension, γ , and non-uniform surfactant concentration results in surface tension gradients, i.e. the tangential stresses on the liquid interface. These tangential stresses cause surface flow which propagates into the liquid bulk (Marangoni effect, Fig. 1). Marangoni flow results in an additional bulk mixing, which is of importance for many industrial applications. Surface flow causes redistribution of surfactant and changes in the local capillary pressures affecting the kinetics of coalescence. The experimental study by Blanchette et al. [13] describes the effect of surface tension gradients on the coalescence in air of liquid drops with a reservoir of miscible but different liquid having a distinct surface tension. It was observed in [13] that the regime of coalescence depends crucially on the drop/reservoir surface tension ratio. The presence of capillary waves during the coalescence of two drops made from two different miscible liquids that had different surface tension was observed in [14]. Note, both above mentioned studies have been performed employing pure liquids. To the best of the authors' knowledge there are no studies reported on the coalescence of surfactant-laden drops in surrounding liquids bearing different surfactants and with different surface tensions, despite the obvious practical importance of such studies.

Therefore, the aim of this study is to fill this gap and to understand the dynamic processes i.e. the kinetics of bridge expansion as well as surface and bulk motion during the coalescence of two liquid drops, one of them having a much lower interfacial tension due to dissolved surfactant. It should be stressed that this study is devoted to the processes which occur after the fluid film separat-

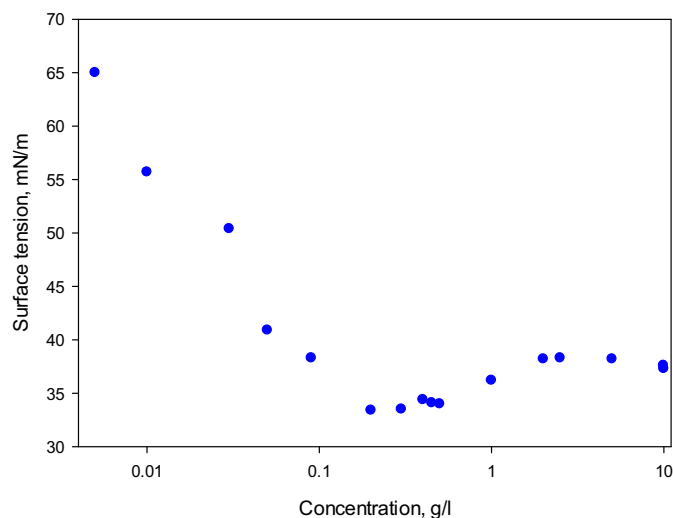


Fig. 2. Surface tension isotherm of sodium lauryl ether sulphate (SLES).

ing the two drops is ruptured. As a plethora of applications deals with the drops merging in an immiscible liquid, this study considers aqueous drops coalescing in silicone oils of various viscosities.

2. Materials and methods

The aim of this work is to investigate the coalescence of drops with an induced interfacial tension gradient along the joining liquid bridge. The gradient was created by contacting two drops, one of which was composed of pure or dyed water and the other was an aqueous solution of sodium lauryl ether sulphate (70%, ES-70, R&D Laboratories Ltd), an anionic surfactant commonly used in industry. The critical micelle concentration, CMC, of SLES was found from the surface tension isotherm presented in Fig. 2 as CMC ~ 0.2 g L⁻¹. The minimum on the curve is due to impurities always present in industrial grade surfactants. The CMC in this case correspond to the minimum in surface tension, after which impurities solubilise in micelles and the surface tension increases. The concentration of surfactant, c , used in experiments, $c = 0.84$ g L⁻¹ was essentially above cmc. Solutions were prepared in double distilled water produced by Aquatron A 4000 D, Stuart. Nigrosin (Alfa Aesar, Johnson Matthey Company) was used at concentration 0.1 g L⁻¹ as a dye to visualise the bulk flow. Hollow glass spheres with average particles diameter of 10 μ m, purchased from Dantec were used to visualise the surface flow. Surface and interfacial tensions were measured with Wilhelmy plate method, using a Krüss K100 tensiometer. The interfacial tensions between water and silicone oil was ~ 32 mN m⁻¹, that of Nigrosin solution, ~ 25 mN m⁻¹, both are significantly higher than the interfacial tension between aqueous SLES solution and silicon oil, ~ 9 mN m⁻¹ for $c = 0.84$ g L⁻¹. The coa-

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