ARTICLE IN PR

[Advances in Colloid and Interface Science xxx \(2014\) xxx](http://dx.doi.org/10.1016/j.cis.2014.04.007)–xxx

Contents lists available at ScienceDirect

Advances in Colloid and Interface Science

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

On the identification of liquid surface properties using liquid bridges

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article info abstract

Available online xxxx

Keywords: Liquid bridge Surface tension Thin film Surfactants Marangoni convection Gibbs elasticity

The term liquid bridge refers to the specific silhouette of a liquid volume when it is placed between two solid surfaces. Liquid bridges have been studied extensively both theoretically and experimentally during the last century due to their significance in many technological applications. It is worth noticing that even today new technological applications based on liquid bridges continue to appear. A liquid bridge has a well-defined surface configuration dictated by a rigid theoretical foundation so the potential of its utilization as a tool to study surface properties of liquids is apparent. However, it is very scarce in literature that the use of liquid bridges is suggested as an alternative to the well-established drop techniques (pendant/sessile drop). The present work (i) presents the theoretical background for setting up a liquid-bridge based surface property estimation problem, (ii) describes the required experimental equipment and procedures and (iii) performs a thorough literature review on the subject. A case with particular interest is that of liquid bridges made of electrically conducting liquids forming between two conducting solids; such a liquid bridge presents an integral electrical conductance value which is sensitive to the specific silhouette of the bridge. This enables the use of this integral conductance as shape descriptor instead of the conventional image processing techniques. Several attempts in literature for the estimation of liquid surface tension, liquid–solid contact angle and surfactant induced surface elasticity for conducting or non/conducting liquids are presented and the prospects of the technique are discussed. © 2014 Elsevier B.V. All rights reserved.

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1. Introduction

Small liquid amounts connecting two solid surfaces, apart by a short distance, are defined as liquid bridges. These solid surfaces can be surrounded by a gas phase or can be immersed in another liquid immiscible to that of the liquid bridge. The present work refers mainly to the first case which is the most common. The theoretical and experimental study of these liquid formations has been very extensive during the last

century. The shape of liquid bridges of pure liquids is determined by intermolecular forces (liquid to liquid molecular interaction determines surface tension whereas the three phase contact line is additionally influenced by liquid to solid molecular forces), by gravity and by external fields such as an electrostatic one. Although liquid bridges refer to small liquid volumes, they can be found in a range of size scales. Regarding the upper size scale there is a limit at the physical dimension of about 1 cm. At this size, gravity causes spontaneous collapse of liquid bridges. Even in the absence of gravity (space station experiments or by density matching with the surrounding fluid) there is the so-called Rayleigh stability limit at which some oscillation modes of the bridge

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<http://dx.doi.org/10.1016/j.cis.2014.04.007> 0001-8686/© 2014 Elsevier B.V. All rights reserved.

Please cite this article as: Kostoglou M, Karapantsios TD, On the identification of liquid surface properties using liquid bridges, Adv Colloid Interface Sci (2014), <http://dx.doi.org/10.1016/j.cis.2014.04.007>

become unstable leading to its disintegration to droplets. For bridges smaller than about 1 mm the effect of gravity is negligible and intermolecular forces completely dominate their shape. There is no lower limit to the bridge physical size beyond that imposed by the molecular nature of the liquid. The situation in the presence of surfactants is much more complex. These molecules not only affect the equilibrium surface tension of the liquid but also bring about a dynamic interfacial behavior of the bridge as they diffuse slowly from the bulk of the bridge towards the gas–liquid interface.

There are many technological applications where liquid bridges are encountered. A non-exhaustive list includes processes such as granulation, flotation and coating [1–[3\].](#page--1-0) In mechanical engineering, liquid bridges allow control of forces in micro-gripping processes [\[4\].](#page--1-0) In geology, liquid bridges determine the properties of moist soil. They are also related to the tertiary oil recovery from porous media. A particular recent area of interest refers to self assembly of particles for which surface tension is the main driving force [\[5\]](#page--1-0). New technologies for fabrication of micro-electromechanical systems based on liquid bridges have been proposed [\[6\].](#page--1-0) Liquid bridges have been employed as dispensing devices in medical applications [\[7\].](#page--1-0) Liquid bridges formed between the tip of the atomic force microscopy probe and the analyzed sample (in humid environment) affects significantly the measurements [\[8,9\]](#page--1-0). Despite the plethora of experimental and theoretical studies on liquid bridges it is surprising that very few refer to the presence of surfactant in the liquid bridge [\[10,11\]](#page--1-0). On the contrary, many works deal with the stability of the bridge with respect to stretching [\[12\]](#page--1-0), air shearing [\[13\]](#page--1-0) or thermocapillary effect [\[14\]](#page--1-0). The attempts of using liquid bridges as diagnostic tools to estimate liquid or surfactant properties are very limited. As a matter of fact, only one work apart from those examined in detail below was found to refer to a qualitative assessment of the rheological properties of biological fluids [\[15\]](#page--1-0).

Here some ideas are presented on using liquid bridges as vehicles to measure surface properties of liquids and the up to now progress is reported. It is well established that the shape of a liquid bridge depends on surface properties. So the main idea lies at the possibility to deduce surface properties by tracing this shape. The straightforward way would be to employ optical images of a liquid bridge and to process it appropriately through the solution of the Young–Laplace equation. This technique is similar to the respective technique developed for pendant drops [\[16,17\]](#page--1-0). However, optical imaging is hampered by inevitable ambient vibrations which blur the bridge shape lowering the accuracy of determinations. Proper illumination conditions yielding high contrast at the bridge borders, high resolution of optical and digital devices and slow image recording/processing steps are further drawbacks of this approach. A different (integral) technique is based on the measurement of forces produced by liquid bridges between the two supporting solid surfaces. This type of measurement suffers excessively from ambient vibrations which cause stability problems and add high background noise to force measurements lowering the accuracy and requiring extensive data filtering [\[18\]](#page--1-0). Another integral technique measuring a property indicative of the shape of a bridge is based on the liquid bridge's electrical conductance. This is a much simpler technique than optical imaging or force measurement and calls for passing low intensity alternating electrical current through the bridge and measuring current intensity and voltage. This technique must be not confused with the extensively studied application of electrical fields to create/stabilize liquid bridges of dielectric or slightly conducting liquids [\[19,20\]](#page--1-0) or soap films of conducting liquids [\[21\].](#page--1-0)

The proposed electrical technique cannot be applied to a pendant or sessile drop without interfering with the drop shape. In the case of a liquid bridge, the technique is applied naturally as long as the employed liquid is electrically conductive and the bridge solid supports are from conductive material. Apparently, the possibility of deriving surface properties based on a fast, integral, non-interfering technique needs to be exploited. Several ideas have emerged on this context. The first idea refers to the identification of surface tension. The shape of a liquid bridge is the result of the competition between gravity and surface tension. The conductance of a bridge depends on its shape and, accordingly, on surface tension. The second idea refers to the identification of contact angle. For small liquid bridges of insignificant Bond number the shape of the bridge is fully determined from the contact angle with its supports. In this case the conductance is directly related to the contact angle. The third idea refers to the creation of a thin liquid film between the surrounding gas and a bubble produced inside the bridge and to examine its conductance in order to identify surface elasticity, e.g., induced by a surfactant in the liquid.

The structure of the present work is the following: At first the theoretical foundation behind the examined experimental techniques is discussed. Assumptions, simplifications and solution techniques are presented in detail. Then the experimental setup and the corresponding strategies are described. Finally, up to now findings of applying the theory to experimental data are presented and the strategy for the future is discussed.

2. Theoretical framework

2.1. The general problem

Let us consider the most general case considered in the present work, that of a liquid bridge in the presence of a surfactant with the possibility of producing a bubble internally to the bridge. Three types of experiments have been realized differing by the way motion is imposed externally to the system. These types of experiments are: (i) increasing the distance between the solid rods supporting the bridge, (ii) reducing the volume of the bridge by liquid evaporation or by withdrawing of liquid through a hollow supporting rod and (iii) growing the internal bubble by feeding gas through a hollow supporting rod. The direct approach to model the motion of the liquid bridge system is complicated due to the involvement of phenomena occurring simultaneously at different size and time scales. To be more specific the Navier–Stokes equations must be solved for the liquid and the gas in the internal bubble combined to the surfactant mass conservation equation in the liquid phase. The boundary condition for the Navier–Stokes equations includes the continuity of the normal and tangential stresses at the gas–liquid interfaces [\[22\]](#page--1-0). The boundary conditions for the surfactant conservation equation includes surfactant adsorption–desorption and its surface diffusion on the gas–liquid interfaces. Interfacial surfactant non-uniformities create surface tension gradients which through the stresses boundary condition lead to the so called Marangoni effect.

In the particular case of the creation of a very thin liquid film in the system a disjoining pressure term must be included in the equations [\[23,](#page--1-0) [24\]](#page--1-0). The above described system of equations is a free boundary problem (the shape of the interface is part of the solution) and its numerical solution is extremely difficult so some assumptions are necessary to proceed with approximate solutions. Recently, numerical techniques capable of incorporating all the above phenomena have been reported but they are still applicable to simpler geometries than the present one [\[25\]](#page--1-0).

A first assumption is that the velocities of the fluid induced by the slow external motion are relatively small compared to the pressure terms in the normal stress boundary condition. A second assumption is that the mixing induced by convection ensures that the surface concentration of a surfactant is always uniform at its equilibrium value as the gas–liquid interfaces evolve. These two assumptions permit the decomposition of the initial problem to two subproblems: The first sub-problem stands for the evolution of the shape of the liquid bridge (and internal bubble) considering a quasi-steady equilibrium shape at every instant. This shape is dictated by a static equilibrium condition and the current value of parameters such as length of liquid bridge (distance between rods), liquid volume, internal bubble size or pressure in gas line. The second sub-problem stands for the flow field in the evolving fluid domain defined by the first subproblem. In the present

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