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Dynamics of drops — Formation, growth, oscillation, detachment, and coalescence

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Single drops or bubbles are frequently used for the characterization of liquid−fluid interfaces. Their advantage is the small volume and the various protocols of their formation. Thus, several important methods are based on single drops and bubbles, such as capillary pressure and profile analysis tensiometry. However, these methods are often applied under dynamic conditions, although their principles are defined under equilibrium conditions. Thus, specific attention has to be paid when these methods are used beyond certain limits. In many cases, computational fluid dynamics (CFD) simulations have allowed researchers, to extend these limits and to gain important information on the interfacial dynamics. Examples discussed here are the capillary pressure tensiometry used for short time and profile analysis tensiometry for long time dynamic interfacial tension measurements, the oscillating drop methods for measuring dilational visco-elasticity. For measuring the coalescence of two drops the liquid dynamics of the subsequently formed liquid bridges have to be considered. In this paper, a thorough review of important experimental and computational findings, related to the dynamics of drops, including its formation, growth, oscillation, detachment, and coalescence is presented. Emphasis is however on some selected important developments. In addition, the paper tries to predict the main directions of advancement in interfacial research for the near future.

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Contents

1. Introduction

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In the past decades, a wide range of experimental investigations have been conducted in order to study fluid–fluid interfaces. However,

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because of their simplicity, and at the same time their complexity, single drops/bubbles having different types of interfaces, have been the main concern of both numerical and experimental investigations for many years. In several of such studies, most dynamic effects have been neglected and thermodynamic equilibrium has been assumed. However, these studies have led to reliable, and at the same time relatively, simple and important theories.

More recently, it seems that emphasis has turned to interfaces in which dynamic effects play the main role. Especially, the complicated dynamic behaviors existing in both phases (e.g., inside and outside bubbles/drops) have attracted much interest in fundamental and applied research works. This for example has led to studying the mechanism of drops/bubbles formation and detachment from a capillary tip, which under certain conditions are of great interest.

To study the dynamics of interfacial layers, the interfaces are not necessarily in equilibrium conditions and considering hydrodynamic effects usually becomes very crucial. The complexities of fluid–fluid interfaces are mostly related to the mutual interactions between the interface and its adjacent bulk [\[1\].](#page--1-0) In addition, any change in the properties of the interface may directly affect the flow field in both phases. This includes interrelations between the physico-chemical properties and hydrodynamics, which usually make interfacial dynamics considerably complicated. Such mutual interactions are of course more complicated in systems containing surface active molecules (as the mechanism of transport of surfactant molecules to and from the interface). On the other hand, the quantitative understanding of the mutual dynamical bulk–interface interactions is the main challenge in studying the stability of many systems (like foams and emulsions). Thus, the properties of the adsorbed layers depend on rather complex mutual bulk–interface interrelations.

Various protocols and experimental techniques have been applied for such investigations. Among different available experimental techniques, the most recently developed capillary pressure technique (CPT) [2–[5\]](#page--1-0) and profile analysis technique (PAT) [\[6](#page--1-0)–9] represent the leading edge of the experimental investigations on dynamics of single drops and bubbles. Furthermore, these techniques have also the potential to be applied to study the bubble/drop coalescence and splitting processes [\[10,11\].](#page--1-0) Note, oscillating drop and bubble experiments are also very essential for studying rheological properties of interfacial layers [\[6,12,13\]](#page--1-0). A variety of other experimental techniques and protocols were developed to study the dynamics of interfacial layers, also based on single drop and bubble manipulations [\[11,14\].](#page--1-0) This is true also for rising drops and bubbles in surfactant solutions the rising velocity values of which depend strongly on the structure of the dynamic adsorption layer formed at the drop/bubble surface (see reference [\[15\]](#page--1-0) for more details).

Challenges in fluid–interface interactions are more complicated to be understood via experimental investigations alone and thus their comprehensive studies also need special attempts from the computational fluid dynamics (CFD) community. In about the past three decades, detailed CFD simulations have provided necessary insight required for understanding the actual flow properties and the mechanisms of transfer of mass and momentum in the bulk and at the interface. Of course, recent CFD simulations (validated with accurate experimental measurements) have also been effectively used to describe some of the complexities in the bulk–interface interactions [\[16\]](#page--1-0).

Even though there has been a great deal of experimental and computational efforts in this field, the complete complex dynamics of most fluid–fluid interfaces is not yet fully understood. The main objective of this paper is to highlight some selected challenges involved in the two-way coupling of complex liquid–fluid interfaces. More specifically, this paper mainly deals with the latest progress in experimental and computational studies of drop formation and drop surface behavior (under different dynamical conditions). In addition, new findings on dynamical drop formation and detachment, drop oscillations and growth, direct drop/drop interactions, liquid bridges, and drop coalescence are discussed in details.

2. Drop profile analysis for growing drops

The shape analysis of pendent drops or buoyant bubbles (originally called axisymmetric drop shape analysis $-$ ADSA [\[17\]](#page--1-0)) is nowadays a routine tool for measuring the surface tension and other interfacial properties. The curvature of an interface corresponds to the physical properties of the interface as well as the dynamic interaction between the two adjacent phases. Therefore, a big advancement in the analysis of drop or bubble profiles has been performed so that modern commercial drop profile analysis tensiometers are available now for fundamental and industrial studies in interfacial science. There are many advantages with the profile analysis techniques, when compared to traditional methods like Wilhelmy plate, Du Nouy Ring, and drop volume tensiometry. The drop/bubble profile analysis technique is easy to handle and requires less amount of liquid for measurements. In addition, tracking the changes in the drop profile with time allows studying the dynamics of interfacial properties.

The process of drop formation and growth is one of the most important research fields because of the enormous number of scientific and industrial applications, like in food industry, extraction technology and petrochemical industry, pharmacy and biotechnology. Therefore, many investigations are performed with growing drops (e.g. [18–[20\]\)](#page--1-0). In addition, this is an interesting field of fundamental research for interfacial science [\[21,22\]](#page--1-0). During drop formation, the interface is getting out of equilibrium due to hydrodynamic effects. This allows deducing dynamic interfacial properties from the profile analysis technique (PAT), however, beyond interfacial properties and gravity as the body force, inertia force, and the drag force during drop growth at the tip of a capillary also contribute in the local drop curvature. According to literature, there are many experimental and numerical studies conducted on growing drops/bubbles, to the aim of studying the role of different contributions in drop and bubble surface deformation [\[3,9,16\].](#page--1-0)

The basis of PAT is an equilibrium force balance as given by the Gauss–Laplace equation. The parametric form of the equation can be presented as [\[8\]:](#page--1-0)

$$
\frac{dx}{ds} = \cos\theta \tag{1}
$$

$$
\frac{dz}{ds} = \sin\theta \tag{2}
$$

$$
\frac{d\theta}{ds} = \frac{2}{R_0} - \frac{\Delta \rho gz}{\gamma} - \frac{\sin \theta}{x}.
$$
\n(3)

Here $\Delta \rho$ is the difference of the densities of the two fluids, and g is the acceleration due to gravity. [Fig. 1](#page--1-0) shows the definition of the coordinates and other parameters used in the fitting of the Gauss–Laplace equation to an experimental axisymmetric drop profile.

To obtain the surface tension γ, the Gauss–Laplace equation is fitted to the experimental drop profile coordinates, as described in detail in [\[23,24\].](#page--1-0) As the technique is commonly in use, several attempts are performed to assess and to extend its capability to different experimental conditions or applications. For example, in [\[25\]](#page--1-0) overviews on the limitation of surface tension measurements with PAT are discussed. However, most of the work is concerned with the use of the technique for equilibrium or very slow dynamic conditions. Only recently, the profile analysis technique was used at rather dynamic conditions where the obtained dynamic profiles of growing drops are analyzed by fitting the Gauss–Laplace equation [\[9\]](#page--1-0). In this study, the applicability of this technique is examined for growing drops of water in air and in hexane, in a broad range of flow rates and drop sizes. For this aim, the values of the standard deviation of fitting the Gauss–Laplace equation to the dynamic drop profiles are obtained. The obtained surface tension values

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