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Historical perspective

Measurement and modeling on hydrodynamic forces and deformation of an air bubble approaching a solid sphere in liquids



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ABSTRACT

The interaction between bubbles and solid surfaces is central to a broad range of industrial and biological processes. Various experimental techniques have been developed to measure the interactions of bubbles approaching solids in a liquid. A main challenge is to accurately and reliably control the relative motion over a wide range of hydrodynamic conditions and at the same time to determine the interaction forces, bubble-solid separation and bubble deformation. Existing experimental methods are able to focus only on one of the aspects of this problem, mostly for bubbles and particles with characteristic dimensions either below 100 µm or above 1 cm. As a result, either the interfacial deformations are measured directly with the forces being inferred from a model, or the forces are measured directly with the deformations to be deduced from the theory. The recently developed integrated thin film drainage apparatus (ITFDA) filled the gap of intermediate bubble/particle size ranges that are commonly encountered in mineral and oil recovery applications. Equipped with side-view digital cameras along with a bimorph cantilever as force sensor and speaker diaphragm as the driver for bubble to approach a solid sphere, the ITFDA has the capacity to measure simultaneously and independently the forces and interfacial deformations as a bubble approaches a solid sphere in a liquid. Coupled with the thin liquid film drainage modeling, the ITFDA measurement allows the critical role of surface tension, fluid viscosity and bubble approach speed in determining bubble deformation (profile) and hydrodynamic forces to be elucidated. Here we compare the available methods of studying bubble-solid interactions and demonstrate unique features and advantages of the ITFDA for measuring both forces and bubble deformations in systems of Reynolds numbers as high as 10. The consistency and accuracy of such measurement are tested against the well established Stokes-Reynolds-Young-Laplace model. The potential to use the design principles of the ITFDA for fundamental and developmental research is demonstrated.

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1. Introduction

1.1. Background and motivations

Interaction between bubbles and solid surfaces in aqueous solutions plays a crucial role in various industrial processes, most notably in froth flotation that is widely used in the separation of mineral particles, treatment of wastewater, recycling of fibers from waste paper, removal of toxic components from industrial effluent and separation of biological cells [1,2]. Since the selective attachment of air bubbles to target particles determines the separation between hydrophobic and hydrophilic particles in a flotation cell, understanding bubble-particle interactions in froth flotation is absolutely crucial [3,4]. An important feature of bubble-particle interaction is drainage of aqueous liquid films between air bubbles and solid surfaces under the influence of hydrodynamic and surface forces, compounded by bubble deformation. Accounting for such deformations under the hydrodynamic forces makes analysis of liquid film drainage dynamics much more challenging. Derjaguin and Kussakov [5] are among the first who analyzed non-equilibrium interactions between an air bubble and a flat mica plate. They showed for the first time dimple formation on the bubble surface.

A number of different experimental techniques have been used to study liquid film drainage dynamics and time dependent interactions between an air bubble and a solid surface immersed in a liquid [6]. One of the earliest methods to study the drainage dynamics of the liquid film involving deformable interfaces was based on the Scheludko cell although only the time evolution of the central liquid film thickness, h(t), was obtained.

The atomic force microscope (AFM), on the other hand, has been widely and effectively used to measure both static and dynamic interaction forces of deformable bubbles [7–10] or oil drops [11–20] approaching solid probe particles in aqueous solutions [21–23]. The AFM colloidal or bubble probe technique allowed direct measurement of interaction forces, but provided no direct information on bubble deformation. Different techniques such as free bubble rise method, bubble expansion method and surface force apparatus were used to study the thin film drainage between an air bubble and a solid surface.

However, none of these techniques is capable of determining simultaneously the deformation of air bubbles and colloidal forces. Moreover, the experiments conducted by the thin liquid film apparatus, surface force apparatus, bubble expansion method and AFM probe technique are mostly in the low Reynolds number regime. For example, the reported maximum bubble approach speed towards a particle in AFM measurement was ~100 μ m/s [24], corresponding to a bubble Reynolds number of particle–bubble encounters in a flotation cell.

To better understand interactions between air bubbles and solid particles in aqueous media as encountered in flotation practice, it is important to develop a device that measures both forces and bubble deformation in systems of higher Reynolds numbers. For this purpose, an integrated thin film drainage apparatus (ITFDA) was developed recently to measure the bubble-particle interactions over a wide range of dynamic conditions [25,26]. The ITFDA is capable of measuring simultaneously the dynamic forces and the geometric properties of the bubble interacting with solid particles. Using the diaphragm of a high frequency speaker as the drive of the bubble, the approach speed of the bubble to a solid particle can be as high as $5000 \,\mu\text{m/s}$, which gives a bubble Reynolds number of 10, making the ITFDA an ideal device to study the bubble-particle interactions under dynamic conditions. It should be noted that even though the Reynolds number that characterizes bubble motion can be as large as 10 with the ITFDA, the Reynolds number that characterizes drainage of intervening liquid film is small, typically Refilm < 1. Therefore the Stokes-Reynolds-Young-Laplace model based on the lubrication theory can still provide a quantitative description of film drainage dynamics and bubble deformation [27].

1.2. Coverage and scope

Historically, the systematic investigation of bubble-particle interactions in the context of colloid and interface science began in the late 1930s, with Derjaguin and Kussakov [5] as the pioneers who studied the behavior of a bubble in water rising under buoyancy towards a mica plate. The experiment was intended to measure surface forces that were the foundation of the Derjaguin-Landau-Verwey-Overbeek theory of colloidal stability [28,29]. The short-ranged nature of such forces required measurement using molecularly smooth surfaces such as a bubble-mica system. In a typical force measurement experiment, one either varies the separation between surfaces and measures the force, or imposes a known force and observes how the intervening liquid film thins. In the Derjaguin and Kussakov experiments, the buoyancy force was known. However, being a time-dependent dynamic experiment, it was necessary to track the position of the bubble and the separation between the bubble surface and the mica plate as a function of time. Furthermore for deformable bubbles, it is also necessary to measure variations of the interfacial deformation of the bubble as a function of position and time during the experiment. These technical and theoretical challenges were perhaps too overwhelming at the time for quantitative measurements. Nonetheless, Derjaguin and Kussakov were able to infer that the hydrodynamic repulsion that arose as the bubble approached the mica plate caused the bubble surface to form a dimple whose shape changed over the time. The work by Derjaguin and Kussakov demonstrated that any attempt to measure dynamic forces involving deformable bubbles has to be able to: i) control and/or measure the force as a function of time; ii) measure the spatial and temporal profile of the bubble or the film thickness between the bubble and the solid surface; and iii) control and/or measure

the position of the moving bubble. Omitting any of these elements in an experiment or a theoretical model to interpret the experimental results would limit the value of the work.

Due to technical constraints in instrumentation and allowable size of samples, it is not easy to incorporate all the above key elements in every experimental approach. As a consequence, many papers that appeared in the past that attempted to study bubble–particle interactions only provided partial or in some cases, even omitted such information. Nevertheless the experimental research coupled with a theoretical framework that includes all the aforementioned key information of dynamic force measurements has provided a good understanding of rather complex systems. The present overview on the use of the integrated thin film drainage apparatus (ITFDA) should be placed into the context with other complementary methods of studying the dynamic interactions between deformable bubbles and solid surfaces.

2. Dynamic experimental methods

In this section, we review complementary methods of studying dynamic bubble–solid interactions. Although no single approach was capable of including all key elements of dynamic experiments discussed in Section 1.2, each concentrated on certain key aspects of the system that taken together, will form a complete picture and foster an indepth understanding of a rather complex problem.

2.1. Free bubble rise method

There have been a number of experiments that studied the rise of mm-size bubbles in water impinging on a horizontal solid surface [30–33]. These studies focused on the trajectories of the bubbles that collided with and bounced from the surface. The bubbles in these studies were in the millimeter to hundreds of micron size range, with Reynolds number between 200 and 600, calculated based on the bubble terminal velocity. The initial bubble-solid surface encounter was dominated by inertia effect. The interaction forces in this regime were modeled successfully [31]. Recently, high-speed interferometry has been used to provide information on the film deformation and thin liquid film drainage mechanisms in this high Reynolds number regime [34].

Using smaller bubbles in the tens of micron size range allowed study of colloidal forces between rising bubbles and solid surfaces [35]. Under such conditions the role of surface forces was shown to play an important role in determining thin liquid film drainage dynamics between the bubble and the solid surface [36].

2.2. Bubble expansion method

With the bubble rise method, the buoyancy force can be modified by changing bubble size. But in order to measure the bubble deformation and film drainage, a larger interaction area between the bubble and the solid surface is needed. Detailed measurements of the spatial and temporal evolution of a draining aqueous film between a bubble and a smooth hydrophilic quartz plate were made in the early 1990s by Fisher et al. [37]. Their study concluded that quantitative comparison of the results from different laboratories was very difficult if not impossible, since the method of forming the draining film profoundly affected its shape and the dynamics of shape evolution. They pointed out that not all the authors publishing in the field were aware of these limitations. They noted a "scarcity of data where the initial conditions for film formation have been reliably and reproducibly controlled".

In the bubble expansion method [37-39], a bubble is forced to emerge rapidly (in less than 1 s) out of a (1 mm inner diameter) glass capillary tube held perpendicularly to a quartz plate placed 610 μ m away. The evolution of the trapped water film between the bubble and the plate was measured by interferometry from 5 s after the bubble expansion to over 200 s as the film drainage proceeded. This was a constant force experiment where the applied force on the bubble was estimated to be ~40 μ N. The detailed sets of experimental data were only analyzed quantitatively some 20 years later [40] using the model discussed in Section 4.

2.3. Scheludko cell

Fundamental studies of liquid film drainages have been conducted in specially designed capillary cells, referred to as Scheludko cells. In this technique, the captive air bubble was pressed against a flat silica surface through a capillary tube [41–44], or by pulling out the liquid between two approaching surfaces [45]. The film thickness between the two surfaces was measured using an interferometric method that was based on multiple reflection and interference of a monochromatic light. Using this method, the time evolution of the central liquid film thickness, h(t), was obtained while the quantitative details of the film profile and liquid withdrawal conditions were often not reported. Although the Scheludko thin liquid film apparatus allowed the thickness of the center film to be measured accurately, it was not capable of measuring the interaction force between an air bubble and a solid surface.

2.4. Surface force apparatus

The surface force apparatus, using fringes of equal chromatic order, has provided accurate measurement of film thickness at subnanometer resolution in elucidating the drainage dynamics of water films trapped between a molecular smooth mica plate and a mercury drop [46,47]. Although the apparatus could have been used to measure the force, the early experiments mainly focused on the measurement of film drainage process. Such studies provided valuable insights into the role of hydrodynamic forces along with repulsive and attractive surface forces in determining the film drainage dynamics that led to either a stable equilibrium film or rupture of the film, the latter leading to a three phase contact.

The same experimental technique has been used to study the interaction between a bubble and a mica plate in a range of monovalent electrolyte solutions of mM concentrations [48,49]. From these studies, the surface potential of the bubbles was found to be negative, with magnitude being less than 10 mV. However, the observed force displacement behavior remained unexplained [49]. This is an area worthy of further investigation, as it is likely to yield some extremely interesting results.

2.5. Atomic force microscope

Complementary to film drainage measurements using the bubble expansion method or the surface force apparatus, the atomic force microscope (AFM) has been adapted to measure bubble-particle interactions. The earliest attempts used the colloid-probe technique to measure the equilibrium force between a colloid particle and a sessile bubble on a substrate in electrolyte solutions [7,8]. With the development of the technique to attach small, ultrasonically generated bubbles (~100 µm diameter) in water onto the force-sensing cantilever [50], the time-dependent force between the bubble and a solid surface as the cantilever moved towards and away from the solid surface was measured and modeled. The deformation of the bubble and film drainage between a bubble and various substrate materials have been studied using this approach [51-53]. Unfortunately, the flexibility of the AFM in undertaking such measurements was offset by the fact that there was no method to measure directly the bubble-surface separation or to determine the extent of bubble deformation during the measurement. Such information had to be inferred from theoretical modeling of the drainage process.

2.6. An important but less explored domain

Due to the experimental design, each of the experimental methods described above to study bubble–particle interactions suffers some inherent limitations although each has particular strength. The free bubble rise method is capable of monitoring the motion, collision and bounce of the bubble below a solid plate surface in the high Reynolds number regime. However, to quantify experimentally the effects of bubble deformation and film drainage during the course of collision and bounces between the bubble and the solid surface, significant technical challenges remain because of the disconnect between the temporal and spatial scales involved.

Studies of bubble–solid interactions using the bubble expansion method or the surface force apparatus yielded accurate and valuable information on bubble deformation and film drainage. The inherent design of the apparatus limited the experiments to a low Reynolds number regime. Although it is possible in principle to measure the time dependent force using this approach, this has yet to be attempted.

In contrast, experiments using the AFM are capable of direct measurement of colloidal forces at the nN sensitivity. There are also considerable flexibility and control in bubble approach/retract speeds. However, since only bubbles or particles of sizes ~100 µm or less can be used in the AFM experiments, direct and precise measurements of bubble deformation are difficult. Furthermore in common with SFA studies, experiments with AFM are typically confined to the low Reynolds number regime.

Therefore a relatively unexplored domain, defined by small to intermediate Reynolds numbers, the ability to control the bubble–particle collision trajectory, to measure the extent of bubble deformation and the drainage of the trapped film between the deformed bubble and the particle, presents fertile opportunities for a different experimental approach. The integrated thin film drainage apparatus, in its current stage of the development with potential extensions, has the promise of adding valuable findings and insights to this important knowledge domain.

3. Integrated thin film drainage apparatus (ITFDA)

The current version of the integrated thin film drainage apparatus (ITFDA) is designed to measure the force between a solid glass sphere, as a model particle, and a bubble in different liquids [25,26]. The design principle of the ITFDA is similar to that of an atomic force microscope (AFM) and the surface force apparatus (SFA). It operates in the millimeter size range of bubbles and particles - intermediate between that of the AFM and SFA, and allows measurement of interaction forces over a wider range of Reynolds numbers. The ITFDA incorporates the strength of both SFA and AFM that features the flexibility of measuring the timedependent forces between the bubble and the particle under controlled variations in their relative displacement. The radii of the bubble and particle, the initial bubble-particle separation and bubble deformation that occur during the interaction are measured from the images of real time videos. Another important attribute of the ITFDA is the ability to modify the surface properties of the bubble and the glass sphere in situ during the measurement. This capability offers the opportunity to study the effect of surfactants on the liquid-air interface, the wetting properties of solid surface and possible additives in solution on stability and thin drainage dynamics of intervening thin liquid films.

3.1. Design features

A schematic diagram of the key components of the ITFDA and a photo of the bubble–glass sphere configuration are given in Fig. 1. The apparatus is housed in a stainless liquid chamber with two optical grade transparent windows placed perpendicular to each other to allow the viewing and recording of bubble approaching the sphere particle during the measurement for subsequent analysis.



Fig. 1. (a) Schematic diagram of the key components and characteristic parameters of the ITFDA: the time-dependent separation distance between the lower surface of the bubble and the top surface of the glass sphere at a distance r from the axis of symmetry, h(r,t); inner radius of the glass capillary tube, r_c ; angle of the bubble on the bottom of the capillary tube (remains constant in the model), θ ; bubble radius, R_b ; glass sphere radius, R_g ; the position of the glass capillary tube relative to the fixed end of the cantilever, X(t); and the deflection of the bimorph cantilever, S(t). (b) Photograph of the bubble at the end of the glass capillary tube above the glass sphere. The red line and green square are drawn to help demarcate the interfaces and to confirm the symmetry of the bubble.

A bubble is generated at the end of a vertical glass capillary tube that is immersed in the test liquid. It is sealed when the bubble attains the required size to be used for force measurements. The glass capillary tube, with the bubble attached is positioned above the glass sphere. They are aligned in an axisymmetric configuration with the help of two perpendicularly positioned cameras in the plane perpendicular to the axis of symmetry, to provide views in orthogonal directions. The axisymmetric alignment facilitates theoretical analysis.

The time-dependent interaction between the bubble and the glass sphere is studied as the capillary tube drives the bubble to approach or retract away from the solid sphere in a well-controlled manner in terms of the extent of bubble-particle overlap and the bubble-particle approach/retract speeds.

3.2. Force sensing bimorph

The glass sphere is attached to the free end of a bimorph cantilever that is used as a force sensor. A piezoceramic actuator with a dimension of 20 mm \times 3 mm \times 0.3 mm and a capacitance of 20 nF (Fuji Ceramics Corp.) is used to fabricate the force sensor. The actuator is enclosed in a fluorinated ethylene propylene sheath and mounted on a stainless

steel chamber. The piezoelectric property of the bimorph material generates an electrical potential in response to deformation under an external force, *F* exerted on the glass sphere by the approaching bubble. The deformation of the bimorph cantilever is determined by measuring the voltage. The bimorph force sensor is calibrated by placing small pieces of platinum wire of known mass on the cantilever and determining the voltage generated. The force sensor used in this study has a sensitivity of 0.1 μ N.

3.3. The glass spheres

The glass sphere attached to the bimorph used in the current study is obtained by melting a 1.5 \pm 0.1 mm diameter Pyrex rod under a butane–oxygen flame until the surface tension of the melting Pyrex produces nearly a spherical surface with a diameter of 4.5 \pm 0.2 mm. The spheres prepared this way are molecularly smooth with a peak-to-peak roughness of less than 1.2 nm determined by AFM imaging.

3.4. Bubble generation and displacement control

The air bubble is generated using a micro-syringe at the end of the glass capillary tube of 1.10 ± 0.01 mm inner diameter (Fisher Scientific). The end of the glass tube is placed under a butane flame to create a smooth end appropriate for bubble generation and force measurements. Extreme caution is taken to avoid overheating of the tube to ensure a uniform geometric symmetry at the end of the capillary tube. The bubble generation process and its size are monitored by real-time video observations. This arrangement allows reproducible generation of bubbles with desired diameters of 1.46 ± 0.01 mm. With this capillary tube, the bubbles of sizes ranging from 1 mm to 3 mm can be reproducibly generated. By changing the size of the capillary tube, a wider range of bubble sizes can be achieved. The other end of the capillary tube is attached to the diaphragm of a speaker that is used to control the vertical displacement of the capillary tube. A computer-generated waveform is used to control the patterns of the diaphragm displacement that in turn drives the attached air bubble towards or away from the lower glass sphere in the desired manner.

It should be noted that the actual bubble approach velocity could be different from the capillary drive speed due to bubble deformation, in particular when the bubble approaches closer to the solid surface. This is taken into account in the theoretical model by analyzing the local velocity of the bubble surface, $\partial h(r,t)/\partial t$.

3.5. Force measurement protocol

For a typical force measurement, a fresh air bubble was generated at the end of the glass capillary tube. The bubble size and the gap between the bubble and the glass sphere were precisely controlled with the aid of two CCD cameras positioned perpendicular to each other. The glass capillary tube, with the attached air bubble, was driven towards and then away from the lower glass sphere by the diaphragm of the speaker. A typical approach-retract cycle that records variations of the capillary tube position with time, t, is shown in Fig. 2. The capillary tube with the bubble attached is first driven towards the lower glass sphere by decreasing the distance X(t) between the capillary tube and the fixed end of the cantilever (Fig. 1) in the approach part of the cycle. The voltage to the speaker is then reversed to move upward the air bubble away from the glass surface in the retraction phase. The time-dependent voltage applied to the speaker, the actual displacement recorded with a position sensitive displacement sensor and bimorph output are recorded for the approach-retract cycle and synchronized with the video recording the bubble shape using one of the two CCD cameras placed in orthogonal orientations in the plane normal to the direction of bubble displacement.

Change in the position of glass tube, $\Delta X(t)$, was measured by a displacement sensor with a detection sensitivity of 5 µm, mounted on the speaker diaphragm. The measured $\Delta X(t)$ (dotted line) in Fig. 2a was fitted with a polynomial function (separately for approach and retraction) to obtain a smooth curve (solid line), which facilitated the determination of capillary displacement speed. The instantaneous speed dX(t)/dt was obtained by differentiating this polynomial function as shown in Fig. 2b. The slight nonlinear-response of the speaker's diaphragm to the initial voltage applied or to changing directions of the displacement caused the displacement to deviate slightly from an ideal saw-tooth wave. In modeling the results, the measured capillary displacement speed, dX(t)/dt, from the measured displacement profile was used so that the observed nonlinearity had no adverse effect on modeling the experimental data.

The initial distance of the closest approach between the bubble and the glass spheres is set at $h_o = 120 \pm 10 \,\mu\text{m}$ and the maximum displacement of the capillary tube is set at $160 \pm 5 \,\mu\text{m}$ (see Fig. 2). Such a combination of settings translates to an overlap of the bubble and the solid sphere by 40 μ m. The ability to measure or specify the initial separation and the maximum displacement between the bubble and the sphere, and hence the bubble–particle overlap in the ITFDA is one of the advantages of the ITFDA over the AFM.



Fig. 2. (a) The measured (dashed line) displacement of the capillary tube $\Delta X(t)$ (the signal has been processed through a low pass filter with a cut-off frequency of 10 Hz) and the corresponding polynomial fit (line); and (b) the instantaneous speed dX(t)/dt (see Fig. 1) obtained by differentiating the polynomial, as a function of measurement time. The nominal speed of the approach-retract cycle is 33 µm/s. The maximum displacement is 160 µm. The resolution of the displacement sensor to determine X(t) is 5 µm.

Although ITFDA is capable of operating at a very high bubble approach speed, the bubble driving speed ranging from 30 μ m/s to 140 μ m/s was investigated in this study. In the experiment, at least 10 measurements were conducted for each set of conditions. For noise reduction, the force signal was processed through a low pass filter with a cut-off frequency of 10 Hz.

The initial separation between the bubble and the glass sphere, h_o , the radius of the bubble, R_b , the radius of the glass sphere, R_g , as well as the angle subtended by the bubble at the end of the capillary tube, θ (Fig. 1) can all be determined from the recorded images, using the image analysis program interfaced with LabVIEW 8.0.

3.6. Cantilever deflection

Quantitative modeling of the experimentally measured forces requires knowledge of the value of the instantaneous cantilever deflection, S(t). With the linear-dependence of F on the deflection S as verified independently, the measured deflection, S(t) of the force sensing bimorph allows the calculation of the applied force, F(t) by F(t) = K S(t), if the cantilever spring constant, K is available. The bimorph cantilever used in this study has a spring constant K in the range of 60–70 N/m. To determine S(t) from the measured voltage, the relation between the charge (or voltage) developed in the bimorph and the displacement of the bimorph, S(t) is first established by placing the glass capillary tube in the direct contact with the bimorph cantilever and measure the output voltage of the bimorph due to the change in the capillary tube displacement, $\Delta X(t)$ that is now equal to the change in the cantilever deflection $\Delta S(t)$ (see Fig. 1).

4. Theoretical model

The theoretical framework that we used to model the measurements of the hydrodynamic interaction between a deformable bubble and a glass sphere was established previously in the analysis of experimental results taken on the SFA and the AFM. These apparatus operate in a size range that encompasses the characteristic dimensions of the ITFDA. Readers are referred to [54] for a more detailed description of the theoretical model. In brief, drainage of the liquid film between the bubble and the sphere is described by the Reynolds lubrication theory under Stokes flow. This theory relates the evolution of the position- and time-dependent separation, h(r,t) to the hydrodynamic pressure, p(r,t) and the disjoining pressure, $\Pi(h(r,t))$ that characterizes surface forces in the film. The deformation of the liquid-air interface of the bubble is described by the nonequilibrium Young-Laplace equation. In this equation, it is assumed that the drop can adjust its shape immediately to accommodate changes in hydrodynamic and disjoining pressures. Since the bubble at the end of the capillary tube was sealed with liquid water to stabilize the size of bubble, the bubble is considered to be at constant volume. The approach is known as the Stokes-Reynolds-Young-Laplace (SRYL) model. In the experiments with the ITFDA, the displacement function, X(t) of the glass capillary tube drives the space-time evolution of these equations.

4.1. Governing equations

The Stokes-Reynolds equation for film evolution is given by

$$\frac{\partial h(r,t)}{\partial t} = \frac{1}{12\mu r} \frac{\partial}{\partial r} \left(rh^3(r,t) \frac{\partial p(r,t)}{\partial r} \right)$$
(4.1.1)

where μ is the shear viscosity of the Newtonian liquid. Implicit in Eq. (4.1.1) is that the hydrodynamic boundary condition at the bubble surface is the same as the tangentially immobile condition that holds at the glass surface. The usual assumption of a zero tangential stress

condition at the bubble surface was found to result in a drainage rate that is too fast as compared to the observations with the ITFDA. Furthermore, the acceptable agreement achieved in the current study between the experimental data and the SRYL model with immobile boundary condition at the bubble surface suggests that in spite of extreme care in surface and solution preparation, there appears the existence of a trace amount of impurities which is beyond the detection limit of surface tension measurement but is responsible for immobile boundary condition at the bubble surface [55]. The liquid–air interface of the film deforms as a result of the hydrodynamic pressure in the film. According to the Young–Laplace model, if the deformation is small as compared to the radius of the bubble, the film thickness is governed by the equation

$$\frac{\gamma}{r}\frac{\partial}{\partial r}\left(r\frac{\partial h(r,t)}{\partial r}\right) = \frac{2\gamma}{R} - p(r,t) - \Pi(h(r,t))$$
(4.1.2)

where γ is the surface tension of liquid [54]. The mean radius, *R* is defined as a characteristic value of the pressure: (γ/R) in this problem.

The Laplace pressure, that is the pressure difference between the interior and the exterior of a spherical bubble of radius R_b , is given by $(2\gamma/R_b)$. If the bubble at the end of the capillary tube is deformed as a result of interaction, its Laplace pressure will change to $(2\gamma/R_L)$ where R_L is the Laplace radius. If the deformation is small then the approximation: $R_L \approx R_b$ would hold. We can then approximate R in Eq. (4.1.2) by

$$\frac{1}{R} \equiv \frac{1}{R_L} + \frac{1}{R_g} \approx \frac{1}{R_b} + \frac{1}{R_g}$$
(4.1.3)

with the initial parabolic profile

$$h(r,0) \equiv h_0 + \frac{r^2}{2R}.$$
 (4.1.4)

Eqs. (4.1.1) and (4.1.2) are solved in the domain of $0 \le r \le r_{max}$ where the motion of the glass capillary tube enters in the boundary condition at $r = r_{max}$

$$\frac{\partial h(r_{max},t)}{\partial t} = \frac{dD(t)}{dt} - \frac{1}{2\pi\gamma} \frac{dF(t)}{dt} \left\{ log\left(\frac{r_{max}}{2R}\right) + B(\theta) \right\}$$
(4.1.5)

with $D(t) \equiv S(t) + X(t) = F(t) / K + X(t)$,

$$B(\theta) = 1 + \frac{1}{2} log \left(\frac{1 + \cos\theta}{1 - \cos\theta} \right)$$
(4.1.6)

and the force F(t) is given by

$$F(t) = 2\pi \int_{0}^{\infty} \left[p(r', t) + \Pi(h(r', t)) \right] r' dr'.$$
(4.1.7)

For a given system, the angle θ in Eqs. (4.1.5) and (4.1.6) is a fix system parameter and does not change with bubble displacement or deformation. In the current study of hydrodynamic interactions using the ITFDA, the effect of the disjoining pressure, $\Pi(h(r,t))$ due to surface forces is negligible because the liquid film between the bubble and the glass sphere remains much thicker than the operative range of Π , which was confirmed experimentally by observing a negligible effect of changing aqueous solution pH and electrolyte concentration on the measured force profiles [26].

4.2. Scaling and universality of the governing equations

Due to the widely different length scales involved in the problem and complexity of numerical solution, the Stokes–Reynolds–Young–Laplace equations become dimensionless by using the scaling parameters give in Eq. (4.2.1) [37]

film thickness : $h \equiv \left[Ca^{1/2}R\right]h^*$ radial coordinate : $r \equiv \left[Ca^{1/4}R\right]r^*$ time : $t \equiv \left[Ca^{1/2}R/V\right]t^*$ pressure : $p \equiv \left[\gamma/R\right]p^*$ disjoining pressure : $\Pi \equiv \left[\gamma/R\right]\Pi^*$ force : $F \equiv \left[(\gamma/R)\left(Ca^{1/4}R\right)^2\right]F^* \equiv \left[Ca^{1/2}\gamma R\right]F^*$ (4.2.1)

where $V \sim dX(t)/dt$ is a characteristic speed and $Ca \equiv \mu V/\gamma$ is the capillary number that measures the ratio of viscous forces to capillary forces. In the present ITFDA experiments, the capillary number is small: $Ca \sim 10^{-6}$.

In terms of the scaled variables, the Stokes–Reynolds equation that describes film drainage between a bubble and a solid sphere, Eq. (4.1.1), becomes

$$\frac{\partial h^*}{\partial t^*} = \frac{1}{12r^*} \frac{\partial}{\partial r^*} \left(r^* h^{*3} \frac{\partial p^*}{\partial r^*} \right)$$
(4.2.2)

and Eq. (4.1.2) for film thickness becomes

$$\frac{1}{r^*}\frac{\partial}{\partial r^*}\left(r^*\frac{\partial h^*}{\partial r^*}\right) = 2 - p^* - \Pi^*.$$
(4.2.3)

It is remarkable that the scaled Stokes–Reynolds–Young–Laplace equations in Eqs. (4.2.2) and (4.2.3) have a universal form independent of any physical parameters apart from the scaled disjoining pressure. In particular, the fluid viscosity does not appear explicitly.

The initial condition, Eq. (4.1.4) now has the form

$$h^{*}(r^{*},0) = h_{0}^{*} + \frac{(r^{*})^{2}}{2}$$
 (4.2.4)

The boundary condition at r^*_{max} , Eq (4.1.5), with $dX(t)/dt \sim -V$, has a weak logarithmic dependence on the capillary number *Ca*:

$$\frac{\partial h^*(r_{\max}^*, t^*)}{\partial t^*} = -1 + \frac{1}{2\pi} \frac{dF^*}{dt^*} \left\{ \frac{2\pi\gamma}{K} - \log\left(\frac{1}{2}Ca^{1/4}r_{\max}^* - B(\theta)\right) \right\}.$$
 (4.2.5)

Eq. (4.2.5) provides the dependence of film drainage on the interfacial tension and the fluid viscosity.

The left hand side of Eq. (4.2.3) represents the small deformation approximation of the mean curvature of h^* . If the disjoining pressure is negligible, that is $\Pi \ll (\gamma/R)$, then this curvature changes sign when the scaled pressure $p^* \equiv (R/\gamma) p$ has the numerical value of 2. Indeed, for $Ca \le 10^{-4}$ numerical solutions of these equations in the absence of the disjoining pressure, $\Pi = 0$, revealed that with the approach at a constant speed, the initial parabolic film profile will develop a dimple when the central separation reaches the value

$$h(r = 0, t) = cRCa^{1/2} \equiv h_{dimple}.$$
 (4.2.6)

For interactions in related geometries such as between a bubble and a plate or between two bubbles, the observed value of the constant c is in the range of 0.4–0.7 [31,54]. The universal nature of the governing equations, Eqs. (4.2.2) and (4.2.3) together with the weak dependence on the capillary number provides a consistency check on results obtained with the ITFDA using fluids of different interfacial tensions and fluid viscosities. We will see a demonstration of this application in Section 5.

Table 1

Material constants and ITFDA parameters.

| _ | | |
|---|--|----------------------|
| | Bubble radius, R _b | $730\pm5\mu m$ |
| | Particle radius, R _g | $2200 \pm 50 \mu m$ |
| | Geometric mean radius, R | 550 µm |
| | Bubble angle at capillary tube, θ | 132° |
| | Surface tension, γ | |
| | – KCl solution | 71 mN/m |
| | – Ethanol | 22 mN/m |
| | – Silicone oil | 22 mN/m |
| | Viscosity, μ | |
| | – KCl solution | 1.0 mPa∙s |
| | – Ethanol | 1.2 mPa∙s |
| | – Silicone oil | 55 mPa∙s |
| | Bimorph cantilever constant, K | 64 ± 4 N/m |
| | Initial separation, <i>h</i> (0,0) | $120\pm3\mu m$ |
| | Maximum displacement, ΔX_{max} | 160 µm |
| | | |

5. Experiments with hydrophilic glass

Now we present results of force measurements between a bubble and a glass sphere in aqueous electrolyte solutions, ethanol and silicone oil using the ITFDA. These fluids are chosen to demonstrate the flexibility of the ITFDA: being able to measure hydrodynamic interaction in a fluid over a range of surface tension, fluid viscosity and driving speed.



Fig. 3. Variation of the force F(t) – experiment (dashed line) and model (solid line), calculated central film thickness h(0,t) and central pressure p(0,t) as a function of measurement time for an air bubble approaching a solid sphere in aqueous electrolyte solution of 1 mM KCl and pH = 5.6 at nominal bubble drive speed of 33 µm/s, corresponding to a Reynolds number Re = 0.05. Insets of (a): photographs of the bubble and the glass sphere at times corresponding to points B, D and F on the force curve.

The results will be compared with the Stokes–Reynolds–Young–Laplace (SRYL) model outlined in Section 4.

To facilitate comparison across the three liquids, the glass sphere is rendered hydrophilic to avoid any possible hydrophobic attraction between the bubble and the glass that might rupture the intervening liquid film. A summary of material properties and characteristics of the ITFDA is given in Table 1. Aqueous KCl electrolyte solution and ethanol have nearly identical viscosities but with surface tensions that differ by a factor of 3, whereas ethanol and silicone oil have nearly identical surface tensions but have viscosities that differ by a factor of 55. Thus measurements using this triplet of fluids allow us to delineate the effect of interfacial tension and fluid viscosity on thin liquid film drainage dynamics and validate the applicability of the SRYL model over a wider range of fluids and Reynolds numbers.

5.1. Surface treatment

The capillary tube and the glass sphere were treated in freshly prepared piranha solutions (3:1 v/v H₂SO₄:H₂O₂) at 80 °C-90 °C for 30 min, rinsed thoroughly with Milli-Q water and blow-dried using an ultra-pure nitrogen stream. The surfaces prepared in this manner were free of contamination and completely water wettable (i.e., water contact angle, $\theta = 0^{\circ}$). The sample chamber was cleaned in the anhydrous ethyl alcohol (Commercial Alcohols Inc.) under ultrasonication for half an hour, rinsed with de-ionized water and then blow-dried with ultrapure nitrogen. The bimorph beam was mounted on to the chamber wall. A glass sphere was clamped at the free end of the bimorph in a dust-free laminar flow environment. The chamber was then filled with test solutions and placed on a three-dimensional translation stage. The capillary tube was filled with fresh air before being immersed into the solution. The system was then left for 2 h to equilibrate the interfaces and to stabilize the bimorph signal before any measurement.

5.2. Interaction in aqueous electrolyte solutions

An example of the time-dependent force profile between an air bubble and a hydrophilic glass sphere in 1 mM KCl aqueous electrolyte solution of pH 5.6, measured using the ITFDA, is shown in Fig. 3a. The bubble, initially at a separation of 120 μ m from the glass sphere, is first driven towards the sphere by moving the capillary tube at a nominal speed of 33 μ m/s for 160 μ m and is then retracted at the same speed (see Movie 1 in the Supplementary material). The repulsive force, rising towards the maximum during the approach phase is solely due to hydrodynamic repulsion, counterbalanced by the Laplace pressure force from the bubble. The force then decreases at the commencement of the retraction phase. The attractive (negative) force minimum is attributed to hydrodynamic suction as the bubble separates from the glass sphere. Using the system parameters in Table 1, the prediction of the time dependent force from the Stokes–Reynolds–Young–Laplace model is in good agreement with the experimental data.

In Fig. 3b, we show the predicted film thickness at the axis of symmetry, h(0,t) as a function of measurement time. When the force is repulsive, h(0,t) remains approximately constant at 310 nm, but briefly attains a minimum value of around 61 nm at the attractive force minimum. In Fig. 3c, we show the hydrodynamic pressure, p(0,t) at the same position. When the force is repulsive, p(0,t) reaches the constant value of 129 Pa which is the same as the Laplace pressure of the bubble. The force minimum is the consequence of the hydrodynamic suction at the negative pressure minimum of -860 Pa and $h(0,t) \sim 61$ nm.

However, the variation of both the film thickness profile, h(r,t) and the pressure profile, p(r,t) within the film are more complex and interesting. In Fig. 4a and b, we show the film profile, h(r,t) at various times along the force profile, F(t) indicated in Fig. 3a. One key feature is the initial development of a dimple at point B, when the film thickness is ~310 nm. The dimple grows to a maximum size at the force maximum (point D) with a dimple rim radius of ~110 µm. The minimum film thickness of ~170 nm occurs at the dimple rim.



Fig. 4. Thickness of the aqueous film, h(r,t) between the bubble and the glass sphere at different measurement time during (a) approaching (points A to D in Fig. 3a) and (b) retraction (points D to G in Fig. 3a); and the hydrodynamic pressure profile, p(r,t) scaled by (g/R) = 64.5 Pa in the water film during (c) approaching and (d) retraction. As in Fig. 3, the electrolyte concentration is 1 mM KCl and the nominal approaching/retraction speed, 33 mm/s.

As discussed in Section 4.2, the initial formation of the dimple at a film thickness of ~310 nm is due to hydrodynamic repulsion, as the disjoining pressure arising from surface forces at such large separations is negligible. Indeed, turning now to Fig. 4c for the hydrodynamic pressure at point B, the hydrodynamic pressure *p* reaches $(2\gamma/R)$, scaled Laplace pressure $p^* \equiv (R/\gamma) p = 2$ of the bubble, when the dimple begins to develop. During the retraction phase (points D to G in Fig. 4b), the thickness of most parts of the film increases with retraction from D to G, except around the axis of symmetry r = 0. In fact, we see in Figs. 3b and 4b that h(0,t) actually decreases to a minimum value of ~61 nm at point F, corresponding to the force minimum, before increasing with further retraction. This negative pressure is the hydrodynamic suction that gives rise to an attractive retraction minimum in the force profile.

The experiment with the results shown in Fig. 3 corresponds to a Reynolds number, $Re = 2R\rho V/\mu = 0.05$. In Fig. 5 we compare the



experimental and theoretical results taken at Re = 0.2, corresponding to a nominal bubble speed of $134 \,\mu\text{m/s}$ which is 4 times larger than that for the experiment in Fig. 3, with all other experimental parameters such as bubble size, initial separation and total displacement being the same as those used in the experiment described in Fig. 3. Qualitatively the results in Figs. 3 and 5 are similar. However, we note that the force maximum at a higher approach speed shown in Fig. 5a is smaller than that shown in Fig. 3a. The result can be understood by investigating bubble deformation shown in Fig. 5d and e. Since the bubbles have identical radius in both experiments, they have the same Laplace pressure: $2\gamma/$ R = 129 Pa. As the hydrodynamic pressure in the water film due to the approach of the bubble increases to $2\gamma/R$, the bubble surface will flatten, resulting in an increase in the intervening film (interaction) area. At a higher approach speed of 134 µm/s or higher Re of 0.2 (Fig. 5), the hydrodynamic pressure reaches the Laplace pressure at a larger bubble-particle separation of 635 nm, as compared to the



Fig. 5. (a): Variation of the force F(t)-experiment (dashed line) and model (solid line) with the insets being the photographs of the bubble and glass sphere at times indicated on the force curve; (b) and (c): Calculated central film thickness h(0,t) and central pressure p(0,t), respectively, as a function of measurement time for an air bubble approaching a solid sphere in an aqueous electrolyte solutions (1 mM KCl, pH = 5.6) at normal approaching/retraction speed of 134 µm/s, corresponding to a Reynolds number Re = 0.2. (d) and (e): Bubble surface profile at points B and D marked in (a) for bubble approaching speed of 33.4 µm/s (Fig. 3) and 134 µm/s, respectively.

Fig. 6. (a): Variation of the force F(t)-experiment (dashed line) and model (solid line) with the insets being the photographs of the bubble and glass sphere at times indicated on the force curve; (b) and (c): Calculated central film thickness h(0,t) and central pressure p(0,t), respectively, as a function of measurement time for an air bubble approaching a solid sphere in ethanol at normal approaching/retraction speed of 33 µm/s, corresponding to a Reynolds number Re = 0.05. (d) and (e): Bubble surface profile at points B and D marked in (a) for bubbles approaching solid sphere in 1 mM KCl aqueous solutions of pH 5.6 (Fig. 3) and ethanol, respectively.

separation of 310 nm for bubble approaching at 33 μ m/s or Re = 0.05 (Fig. 3), see curve B in Fig. 5d and e. The radial extent of the dimple at 134 μ m/s or Re = 0.2 is smaller, expanding to around 75 μ m (curve D in Fig. 5e) in comparison to around 110 μ m for the case of bubble approach speed of 33 μ m/s or Re = 0.05 (curve D in Fig. 5d).

It is interesting to note that the minimum film thickness reached before retraction is 50% thinner at 61 nm for the lower bubble approach speed, as compared to 126 nm for the higher bubble approach speed. In the context of interactions involving deformable surfaces, an important finding from this study is that with all other things being equal, slower bubble approach speed will result in the formation of thinner liquid films before retraction. This is in contrast to rigid bodies in which a faster drive speed will lead to smaller separations. As shown in Figs. 3a and 5a, the attractive hydrodynamic pull-off force $(-0.145 \,\mu\text{N})$ is lower for the lower bubble retracting speed (33 μ m/s) as compared with the



Fig. 7. (a): Variation of the force *F*(*t*-experiment (dashed line) and model (solid line) with the insets being the photographs of the bubble and glass sphere at times indicated on the force curve; (b) and (c): Calculated central film thickness $h(0,t \text{ and central pressure } p(0,t, respectively, as a function of measurement time for an air bubble approaching a solid sphere in silicon oil at normal approaching/retraction speed of 33 µm/s, corresponding to a Reynolds number Re = <math>10^{-4}$. (d) and (e): Bubble surface profile at points B and D marked in (a) for bubbles approaching solid sphere in Ethanol (Fig. 5) and silicon oil, respectively.

pull-off force (-0.4 $\mu N)$ for the higher retraction speed (134 $\mu m/s),$ as anticipated.

5.3. Interaction in ethanol and silicone oil

In Fig. 6 we show the effects of varying the surface tension of liquid by using ethanol with a surface tension about one third of the surface tension of water. The effect of liquid viscosity is shown in Fig. 7 by conducting the measurements in silicone oil which has a viscosity 55 times higher than the viscosity of ethanol but the same surface tension as ethanol (see Table 1). (See Movie 2 (ethanol) and Movie 3 (silicone oil) in the Supplementary material.)

The essential difference between the results obtained in water and in ethanol can be understood in terms of the difference in Laplace pressure, $2\gamma/R$, for the two liquids. For the same size of air bubble, the Laplace pressure of air bubble in ethanol is 40 Pa, in comparison to 129 Pa for the air bubble in water. Such a difference in the Laplace pressure of the air bubble means that with all other things being equal, bubble deformation in ethanol will occur at smaller hydrodynamic forces, rendering larger separation distances as shown in Fig. 6e. By the same reasoning developed to account for the decreased maximum repulsive forces with increasing bubble approach speed in water, the maximum repulsive force in ethanol at the same bubble approach speed is found to be only about one-thirds of the value obtained for water, as scaled by surface tension ratio.

For an air bubble approaching a solid sphere at 33 μ m/s in the silicone oil having a viscosity 55 times higher than the viscosity of ethanol, although the Reynolds number remains small ~10⁻⁴, the hydrodynamic pressure on the solid sphere from the motion of the bubble is much higher. As a result, a larger repulsive force is measured in silicon oil (2.4 μ N, Fig. 7a) than in ethanol (1.67 μ N, Fig. 6a), although both liquids have the same surface tension. The attractive hydrodynamic pull-off force is also stronger in silicon oil (-0.6μ N, Fig. 7a) than in ethanol (-0.125μ N, Fig. 6a) as anticipated. Such differences are again accounted for by the deformation and flattening of the bubble, now occurring at a much larger separation distance of about 4400 nm in silicon oil, as compared to 600 nm for the bubble approaching the solid sphere at the same bubble speed in the ethanol that has the same Laplace pressure of 40 Pa.



Fig. 8. A comparison between the measured and the calculated contact diameter between the bubble and the glass sphere in water, ethanol and silicone oil under different approach speeds in the repulsive part of the force profiles given in Figs. 3 and 5–7.



Fig. 9. Correlation between the measured repulsive forces normalized by capillary pressure $(2\gamma/R_b)$ and the film area between the bubble and the glass sphere in water, ethanol and silicone oil under different drive conditions.

5.4. Scaling of experimental results

The above experiments of measuring the hydrodynamic forces between bubbles and solid glass spheres in water, ethanol and silicone oil of different surface tensions and viscosities are conducted using the ITFDA at small Reynolds number, *Re* (<0.2) and small capillary number, *Ca* (~10⁻⁶), which allows us to compare the experimental results and theoretical predictions.

For accurate image analysis to determine the contact angle and/or diameter of bubble-solid contact, the inset image as shown for example in Fig. 8 was further zoomed in. The three phase contact points between the glass, the bubble and the test solution on the left and right hand sides were selected, and the distance between these two three phase contact points was measured as the contact diameter. The measurements were repeated three times using the same procedures. The average and the standard deviation of the three measurements were calculated. The absolute standard deviation of the contact diameter measurement using this method is about 15 µm. The contact diameters at different stages of bubble-glass interaction were determined as a function of experiment time using the method mentioned above. Once the film thickness was calculated by the model, the distance between the two barriers or diameter of the dimple could be determined and plotted as a function of experiment time. The measured contact diameter and the calculated dimple diameter corresponding to the same experiment time were then compared and plotted in Fig. 8.

As can be seen in Fig. 8, the measured contact diameters between the bubble and the glass sphere from the side view cameras for all the systems studied are in excellent agreement with the predicted contact diameter or twice the dimple radius, *R*_f, under this range of experimental conditions, demonstrating not only the excellent capability of the newly designed ITFDA, but also the applicability of the Stokes–Reynolds–Young–Laplace model to studying hydrodynamic interactions between deformable surfaces.

To further understand the measured variation of repulsive forces with bubble approach speed, liquid surface tension and viscosity, the measured repulsive forces scaled by capillary pressure $(2\gamma/R_b)$ are plotted as a function of film area in Fig. 9. The good linear correlation between the two parameters suggests that the Laplace pressure force during interaction counterbalances the hydrodynamic force which drives bubble deformation.

6. Conclusions and future perspectives

With the ability to measure forces and deformations simultaneously, the development of the ITFDA makes it possible to delineate details of the system physics. The ITFDA offers the flexibility to vary parameters such as displacement speed, interfacial tension and viscosity of the fluids and the chemistry to modify the solid surface to variable wettability. The excellent agreement between the experiment and theory of dynamic forces in the three fluids of very distinct physicochemical properties (viscosity, surface tension and polarity) demonstrates that the SRYL model can be applied to the systems of a wider range of bubble approach speed, and liquid interfacial tension and viscosity. Such good agreement also makes it possible to use the model to infer quantitative information about film profiles during the bubble approach-retraction. The simulation results indicate that for a given set of conditions the minimum film thickness in the approach phase is smaller for the system of low bubble approach speed, and/or low viscosity and high surface tension of the liquids. The agreement between the measured and the predicted contact diameter shows the capability of ITFDA to accurately measure bubble deformation. The linear relationship between the normalized repulsive force by Laplace pressure and bubble deformation suggests that for the studied range of experimental conditions (Reynolds number, Re < 0.2 and film thickness greater than 150 nm of hydrophilic solid surfaces), the contribution of disjoining pressure on total force is minimum and the force can be estimated from the product of the Laplace pressure of the undeformed bubble and the radius of the flattened film areas. It is our intention to further measure the bubble profile with similar experimental systems to fill the gap of confirming theoretical prediction with experimental results. In principle, the film thickness profile between the glass sphere and the deforming bubble can be measured by interferometry. This is an obvious direction for further development of this apparatus.

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