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Impact of surface dilation rate on dynamic surface tension

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ABSTRACT

In past studies, estimating the dynamic surface tension in processes involving significant surface deformation relied heavily on empirical relations. Despite its importance, the detailed dynamics of surfactant adsorption onto a dilating surface have not been investigated extensively. In this study, we modified the pendant bubble tensiometer to provide direct observation on the dynamics of surfactant adsorption during rapid surface dilation. The relaxation of dynamic surface tension for bubbles rapidly expanding in the solution of four different bulk concentrations of Triton-X 100 was studied. It was found that surface tension rose with increasing surface area due to the decrease in surface concentration. Moreover, when surface dilational rate was increased, it was found that the relation between surface tension and surface area came to resemble the equation of state of two dimensional surfactant phase when dilational rates were larger than a certain threshold value, indicating that the adsorption of surfactant became negligible. This threshold value was found to increase with surfactant concentration. The results showed that the procedure established in this study could serve as a simple guideline to determine whether adsorption can be neglected in a process involving dilating surfaces. In addition, the detailed dynamical data reported in this work can serve as an experimental reference for multiphase flow simulations involving species that adsorb on interfaces.

1. Introduction

In surfactant solutions, surfactants tend to adsorb onto the liquid-vapor interface, lowering the surface tension of the liquid-vapor interface. However, the adsorption process requires finite time. When a new surface is created, it takes time for the surfactant to diffuse and repopulate the surface, and hence the surface concentration of surfactants does not always remain at the same value throughout the process. As a result, unlike the case for pure liquids, it is not always valid to consider the dynamic surface tension of surfactant solutions to be identical to the equilibrium surface tension. The relaxation of surface tension of surfactant solutions after a rapid change of surface area is an important subject in various physical phenomena where large surface deformation is involved.

One particular subject where dynamic surface tension is important is the impact of surfactant-laden drops on solid walls. During the impact of a liquid drop, very large deformation is experienced by the drop in a short time. For pure liquid drops impacting on several solid substrates, it was observed that the shape of drops was deformed drastically within several milliseconds [1,2]; large deformation was also observed for surfactant-laden drops [3]. Hence, the relaxation process of surface tension and the adsorption kinetics of surfactants are crucial in the understanding of the impact of surfactant-laden drops.

The impact of surfactant-laden drops on solid walls can be found in many applications. However, while there are extensive studies on the dynamics during the impingement of pure liquid drops onto solid walls, the number of studies on the dynamics during drop impact for surfactant-laden drops is relatively limited. Pasandideh-Fard et al. studied the dynamics of the impact of surfactant-laden drops. They found that the addition of surfactant enhanced the spreading. Zhang and Basaran investigated the effect of dynamics surface tension of Triton X and sodium dodecyl sulfate (SDS) solutions and found that Marangoni stresses play important roles during drop spreading [4]. Mourougou-Candoni et al. conducted systematic studies on the effect of various surfactants on the dynamics of drop impact [5,6]. With the aid of the empirical model proposed by Hua and Rosen [7], they were able to relate the dynamic surface tension in drop impact experiments to the values measured by the maximum bubble pressure method (MBPM) by comparing the surface dilational rate during the processes.

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Crooks et al. and Cooper-White et al. also studied the effect of dynamic surface tension on the dynamics of drop impact; in addition, they also investigated the effect of viscoelasticity of the fluid on the dynamics [8,9]. In two recent studies, Gatne et al. and Aytouna et al. compared the dynamic behavior of drops on hydrophobic substrates with surfactants with different adsorption/diffusion rate [10,11]. Other studies also investigated the dynamics of surfactant-laden drops on various substrates [12–17].

In the studies mentioned above, it was shown that the addition of surfactants greatly modified the dynamics of drop impact. However, to systematically study the effect of surfactants, the dynamic surface tension during the impact process must be determined. This is difficult due to the large deformation in surface and the highly far from equilibrium nature of the process. However, a clever resolution was proposed in the pioneering work of Mourougou-Candoni et al. They found that the dynamic surface tension in different processes with the same surface dilational rate could be compared [5]. By linking the dynamic drop impact process to the adsorption of surfactant to freshly formed surface in dynamic surface tension measurements, they were able to identify the dynamic surface tension during drop impact. With the help of the empirical relation proposed by Hua and Rosen [7], Mourougou-Candoni et al. were able to use the dynamic surface tension data obtained through the maximum bubble pressure method and the model proposed by Fainerman [18] to establish the relation between dynamic surface tension and surface dilational rate. Then, the dynamic surface tension during could be calculated as a function of surface dilational rate found in drop impingement experiments. Other studies followed the procedure of Mourougou-Candoni et al. [5,6] and also determine the dynamic surface tension through relating between the surface dilational rates in drop impact experiments and that in the dynamic surface tension measurement with the maximum bubble pressure method [8–11].

In the maximum bubble pressure method, gas is fed through a capillary tube, where liquid is displaced and bubbles form. The surface tension is calculating by measuring the maximum pressure difference between the tip of the capillary tube and the reference node, which happens when the radius of curvature of the liquid-gas interface is minimized. The lifetime of the bubble can be controlled through the flow rate of the gas, and hence the dynamic surface tension of different timescales can be measured. The effective age of the surface of the solution has been calculated by Kloubek, who studied the relationship between the interval between subsequent bubbles and the actual age of the surface [19]. Fainerman et al. showed that the maximum bubble pressure method can be used to measure the dynamic surface tension over a large range of timescales, from 1 ms to several seconds [20,21].

The maximum bubble pressure method is convenient and capable of observing fast adsorption dynamics. However, the procedure of relating the dynamic surface tension measured by MBPM to the dynamic surface tension during surface dilation is an indirect process that relies heavily on the empirical relation proposed by Hua and Rosen [7]. It would be more convincing and informative if the complete temporal relaxation of dynamic surface tension during a surface dilation process is directly measured. However, to the best of our knowledge, there is no systematic study reporting the direct observation of such process. In sight of this, in this article, we design a simple bubble expansion experiment to provide such experimental data. We focused on expanding surfaces because it was reported by Mourougou-Candoni et al. that during the initial spreading stage of drop impact, the surface dilational rate could be as large as 250–600 s^{-1} in drop impact experiments [5]. In this study, a modified pendant bubble tensiometer was employed to study the dynamic surface tension of expanding bubbles in aqueous Triton X-100 solution of different concentrations. We hope that our results could potentially serve as the bridge between numeric simulations and experiments on complex dynamics, and enhance our understanding of the effect of surfactant adsorption and dynamic surface tension in processes where substantial deformation is involved.

2. Materials and methods

2.1. Materials

The non-ionic surfactant octylphenoxypolyethoxyethanol, commercially referred as Triton X-100 (CAS 9002-93-1, $C_{14}H_{22}O(C_2H_4O)_n$, n = 9–10, Average Molecular Weight, M_w = 625 g/mol), was purchased from Sigma-Aldrich (T9284) and used in this study without modification. The water with which the aqueous solutions were made was purified via a Barnstead NANOpure water purification system, with the output water having a specific conductance of less than 0.057 μS/cm. The value of the air-water surface tension was 72.0 mN/m at 25.0 ± 0.1 °C [22].

2.2. Apparatus

A pendant bubble tensiometer enhanced by video digitization was employed in this study [23]. The video-image system (Optronis CR3000X2) digitized the pictures in 400 lines × 400 pixels. The rate of image acquisition was 6770 images per second. The video-image system was calibrated by digitizing a stainless-steel ball with a known diameter of 2.498 ± 0.002 mm. All experiments were undertaken at 25 ± 0.5 °C and the temperature variation of the aqueous solution was less than 0.1 °C. The captured images were processed to determine the bubble edge coordinates, according to the edge detection routine described by Lin et al. [22]. The uncertainty for the edge location was approximately 0.2 pixels [22]. The edge coordinates of the bubble were then best-fitted with a theoretical drop shape generated from the Young-Laplace equation, and the surface tension (γ) was calculated [22,24,25]. The accuracy of the γ measurements was 0.1 mN/m for equilibrium surface tension [22].

2.3. Methods

At the start of the experiment, an emerging air bubble with a diameter of ~2 mm was formed in the Triton X-100 solution. The bubble is maintained at constant volume until the system reached the equilibrium state. Then the liquid-gas surface was expanded and sequential digital images of the bubble were taken. The measurements were performed for four surfactant bulk concentrations, $C_0 = 3.18 \times 10^{-8}$, 1.43×10^{-8}, 8.05×10^{-9} and 3.07×10^{-9} mol/cm^3, at which the equilibrium surface tensions were γ_{eq} = 45.7, 51.1, 54.9 and 59.9 mN/m respectively [22]. The experiments were performed at eight different bubble expansion rates (dV/dt from ~3 to ~60 mm^3/s).

3. Results

The dynamic surface tension, $\gamma(t)$, and the surface area of the liquid-gas interface of the expanding bubble as a function of time, A(t), were derived for all experimental conditions. As illustrated in Fig. 1, after the original bubble was rested and had attained equilibrium, the bubble was inflated. As observed in the figure, during the rapid dilation of the surface, the surface tension began to increase, indicating that the surface concentration of surfactant decreased. The surface dilational rate was determined by linear fitting the time dependence of surface area, as shown with the solid line in Fig. 1.