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# An investigation of bubble coalescence and post-rupture oscillation in non-ionic surfactant solutions using high-speed cinematography

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#### ABSTRACT

Most processes involving bubbling in a liquid require small bubbles to maximise mass/energy transfer. A common method to prevent bubbles from coalescing is by the addition of surfactants. In order to get an insight into the coalescence process, capillary bubbles were observed using a high speed cinematography. Experiments were performed in solutions of 1-pentanol, 4-methyl-2-pentanol, tri(propylene glycol) methyl ether, and poly(propylene glycol) for which information such as the coalescence time and the deformation of the resultant bubble upon coalescence was extracted. It is shown in this study that the coalescence time with surfactant concentration until the appearance of a plateau. The increase in coalescence time with surfactant concentration could not be attributed only to surface elasticity. The oscillation of the resultant bubble was characterised by the damping of the oscillation. The results suggested that a minimum elasticity is required to achieve an increased damping and considerable diffusion has a detrimental effect on the dynamic response of the bubble, thereby reducing the damping.

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#### 1. Introduction

Bubble swarms are often encountered in industrial processes for applications such as waste water treatment, bio-reactions, and flotation. It is, in such circumstances, difficult to have an insight into the sub-processes taking place. It has been found that a relatively good correlation can be found between the stability of a single film and static foams [1], which could also be relevant in the case of bubble pairs. Although suffering from a potential lack of accuracy in mimicking real, thick foams, simpler foam systems are better defined along with the quantities derived from them [2]. A common technique used to decouple the multiple interactions occurring in gas emulsions includes capillary bubbles. This practice was for example employed by Kazakis et al. [3] to simplify the phenomena of bubble formation at a porous sparger interface. Most studies, however, only focus on bubble-bubble interaction to characterise the coalescing properties of solutions. One pioneering work in the use of capillary bubbles is that of Lessard and Zieminski [4]. These authors produced bubble pairs on inverted capillaries using a constant gas flow rate to look into the effect of electrolyte solutions over a wide range of concentrations. It was found that water and diluted solutions of electrolytes did not prevent bubbles from coalescing whereas increasing the concentration of the solutions appeared to decrease the probability of bubble coalescence.

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In addition to electrolytes [4,5], the technique has also been applied to surfactant solutions [6–9]. The experimental setup can be slightly modified by changing the orientation of the capillaries [10–14]. Most of these works have used growing bubbles, which, if too stable, detach from the capillaries without coalescing. However, captive bubbles of constant volume allow extended contact times. A major development has been the use of optical techniques to measure coalescence times (e.g. [11,15,16]). When two bubbles merge, the resultant bubble undergoes a chaotic oscillation. The advantage of using an optical technique like high-speed cinematography is the ability to study the oscillation following the merging of two bubbles or post-rupture oscillation [12,14,17–19]. Interestingly, the oscillation of the resultant bubble was shown to be closely correlated to the detachment of micron-size particles [12,17], a process that is important in fields like mineral processing. Post-rupture oscillation was used to understand the adsorption and desorption behaviour of sub-micrometer-size latex particles at the air-water interface under various pH conditions [20] and at the interface between the oil and water droplets [21]. The coalescence technique was also successfully applied to study the effect of ageing (and the adsorption of impurities) and temperature on the coalescence between two separate kerosene drops [22]. These studies suggest that, although simplistic, the technique offers a powerful way to measure coalescence time, post-rupture oscillation, and to characterise particle-bubble aggregate stability.

This study investigates the properties of non-ionic surfactants using the binary bubble coalescence technique. Capillary bubbles



are brought into contact in a controlled environment and allowed to coalesce providing an elementary assessment of pure industrial surfactants (i.e. frothing agents used in froth flotation technology). Characteristics such as coalescence time and the oscillating behaviour of coalesced bubbles are analysed. Whenever possible, these fundamental properties are interpreted in terms of interfacial properties.

## 2. Methodology

# 2.1. Materials

Solutions of 1-pentanol (CH<sub>3</sub>(CH<sub>2</sub>)<sub>3</sub>CH<sub>2</sub>OH, MW = 88.15; Sigma–Aldrich, ≥99%), 4-methyl-2-pentanol (methyl isobutyl carbinol, MIBC) ((CH<sub>3</sub>)<sub>2</sub>CHCH<sub>2</sub>CH(OH)CH<sub>3</sub>, MW = 102.17; Sigma-Aldrich,  $\geq$  98%), tri(propylene glycol) methyl ether (TPM) (CH<sub>3</sub>(- $OC_3H_6)_3OH$ , MW = 206.28; Sigma–Aldrich,  $\geq$  97.5%), and poly(propylene glycol) (PPG 425) (H[OCH(CH<sub>3</sub>)CH<sub>2</sub>]<sub>7</sub>OH,  $M_n \approx$  425; (Sigma-Aldrich,  $\geq$  99%), were prepared at concentrations ranging from  $1 \times 10^{-5}$ -1  $\times 10^{-1}$  M,  $5 \times 10^{-5}$ -1  $\times 10^{-2}$  M,  $5 \times 10^{-6}$ -1  $\times 10^{-2}$  M, and  $1 \times 10^{-7}$ -1  $\times 10^{-4}$  M, respectively for bubble coalescence experiments. The former polyglycol is the main constituent of Dowfroth 200 while the latter is a component of Dowfroth 400. Dowfroth 200 and 400 are commercial frothers used in froth flotation technology to generate fine bubbles and, to some extent, to stabilise the froth phase. All glassware used in this experiment was soaked in a mixture of 30%v/v of HNO3 (analytical grade, Chem-Supply) in H<sub>2</sub>SO<sub>4</sub> (analytical grade, Ajax Finechem Pty Ltd.) for 5-10 min and then thoroughly rinsed with water. The water used for preparing solutions and rinsing glassware was dispensed from a Milli-O Pure water system and had a resistivity of 18.2 M $\Omega$  m<sup>-1</sup>. All experiments were carried out in a temperature controlled room at  $20 \pm 2$  °C. Before each experiment, the water and glassware were tested for contamination. The test for cleanliness consisted of coalescing bubble pairs in water after ageing the bubbles for 20 s for MIBC and 90 s for all other surfactants and insuring that coalescence took place within 3.33 ms, which is the time resolution of the experiment. It is noted that the ageing times to test for cleanliness were longer than the ageing at which the surfactants were tested.

#### 2.2. Experimental procedure

#### 2.2.1. Surface tension

Surface tension of surfactant solutions were measured by the profile analysis tensiometry of an emerging bubble. The measurements were performed with a contact angle system OCA 20 (Data Physics Instruments) at a temperature of  $22 \pm 1$  °C.

#### 2.2.2. Bubble coalescence

The experimental apparatus was designed to allow the observation of coalescing capillary bubble pairs [11]. A detailed schematic representation of the setup can be found elsewhere [14]. Two stainless steel capillaries with 1.07 mm O.D. were coupled to linear stage translators such that the bubble pairs could be aligned in all three dimensions. A pair of capillary bubbles of approximately 2 mm in diameter was brought together using an electronic linear actuator (T-LA28A, Zaber Technologies Inc.) to move one capillary at a constant speed of 2.754 mm s<sup>-1</sup>. The bubbles formed in the solutions of MIBC and 1-pentanol were aged 10 s and 20 s, respectively, before they were moved together. This ageing time was sufficient to reach an adsorption equilibrium [23]. For larger molecules, the equilibrium time can be longer, especially at lower concentrations. To investigate the effect of ageing of the interfaces in solutions of polyglycols, bubble pairs were brought together after ageing times of 20 s, 40 s, and 80 s. A high speed video camera (Phantom 5, Vision Research Inc., USA) set at a capture rate of 300 frames per second (fps) and 6024 fps was employed to record video footage used in the evaluation of the coalescence time and the oscillation of the resultant bubble, respectively.

The coalescence time for each experimental condition was averaged over three replicated experiments, each composed of at least 15 measurements. The coalescence time was defined as the interval from the moment the bubbles are in contact to the rupture of the thin liquid film. Video footage recorded for the evaluation of the oscillation of the resultant bubbles was analysed using the imaging software package ImageJ (National Institutes of Health). The projected surface area was measured as a function of time where time zero was arbitrarily chosen as the first frame after the rupture of the thin liquid film. A sufficient number of frames were analysed to cover a length of time of 40 ms. Following the description of the oscillation [12], it was quantified using the equation suggested by Schulze [24], originally developed for a bubble impacted by a particle, given as

$$A' = A'_0 \times e^{-\delta t} \times \sin(\omega_0 t + \psi) + B \tag{1}$$

where *A*′ is a normalised relative projected area (i.e. the difference between the projected area and the average projected area divided by the average projected area),  $A'_0$  is the initial amplitude,  $\omega_0$  is the angular frequency (ms<sup>-1</sup>),  $\delta$  is a damping constant (ms<sup>-1</sup>),  $\psi$  is the phase shift, *t* is the time (ms) and *B* is an integration constant. The experimental data points were fitted by minimising the residual sum of squares.

#### 3. Results and discussion

# 3.1. Surface tension of surfactant solutions

The profile analysis tensiometry technique can be used to obtain dynamic surface tension data. However, the resolution in the early stages of formation of a bubble is in the order of one to a few seconds [25]. For most surfactants used in this study, this lack of resolution means that an equilibrium surface tension is already reached at the start of the measurement. The only surfactant showing a change in surface tension with time with the current technique was PPG 425. This is illustrated in Fig. 1 for some of the concentrations tested. It can be seen that the initial rate of change in surface tension is dependent upon the bulk concentration of surfactant. This is followed by sudden decrease in surface tension, before reaching a constant value. Some soluble polymers, like PPGs, are known to adsorb very quickly through a diffusion-controlled



**Fig. 1.** Dynamic surface tension of PPG solutions at concentrations (♦)  $5.5 \times 10^{-7}$  M, (□)  $2.2 \times 10^{-6}$  M, (▲)  $5.3 \times 10^{-6}$  M, (○)  $1.1 \times 10^{-5}$  M, (+)  $2.1 \times 10^{-5}$  M, (◊)  $5.2 \times 10^{-5}$  M, (■)  $1.0 \times 10^{-4}$  M, (△)  $2.1 \times 10^{-4}$  M, (●)  $5.0 \times 10^{-4}$  M, (×)  $2.1 \times 10^{-3}$  M, and (-)  $1.1 \times 10^{-2}$  M.

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