



# Influence of acoustic pressure and bubble sizes on the coalescence of two contacting bubbles in an acoustic field



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## ABSTRACT

In this study, the coalescence time between two contacting sub-resonance size bubbles was measured experimentally under an acoustic pressure ranging from 10 kPa to 120 kPa, driven at a frequency of 22.4 kHz. The coalescence time obtained under sonication was much longer compared to that calculated by the film drainage theory for a free bubble surface without surfactants. It was found that under the influence of an acoustic field, the coalescence time could be probabilistic in nature, exhibiting upper and lower limits of coalescence times which are prolonged when both the maximum surface approach velocity and secondary Bjerknes force increases. The size of the two contacting bubbles is also important. For a given acoustic pressure, bubbles having a larger average size and size difference were observed to exhibit longer coalescence times. This could be caused by the phase difference between the volume oscillations of the two bubbles, which in turn affects the minimum film thickness reached between the bubbles and the film drainage time. These results will have important implications for developing film drainage theory to account for the effect of bubble translational and volumetric oscillations, bubble surface fluctuations and microstreaming.

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## 1. Introduction

Bubble coalescence has been extensively studied in the absence of an acoustic field. The coalescence process is divided into three steps [1,2] (see Fig. 1(a)): the first step is the contact of the two bubbles to form an initial thin liquid film between them. The second step is thinning of the liquid film to a dimension of about  $10^{-6}$  cm. The third step involves rupture of the film at a dimple point, followed by a rapid coalescence. These three steps are complicated processes which have been extensively studied [3–9] and have been shown to be sensitive to effects such as the approach velocity [7,10–12], the force between the bubbles [13–15] and the fluid viscosity [10,16–19].

The effect of approach velocity is reflected in the Weber ( $W$ ) number (Eq. (1)), which can be used to predict whether two approaching bubbles may coalesce:

$$W = \rho U^2 R_{eq} / \sigma \quad (1)$$

$\rho$  is the liquid density,  $U$  is the velocity of approach,  $\sigma$  is the surface tension and  $R_{eq}$  is the equivalent radius of the bubbles, defined as in Eq. (2) [7]:

$$\frac{2}{R_{eq}} = \frac{1}{R_1} + \frac{1}{R_2} \quad (2)$$

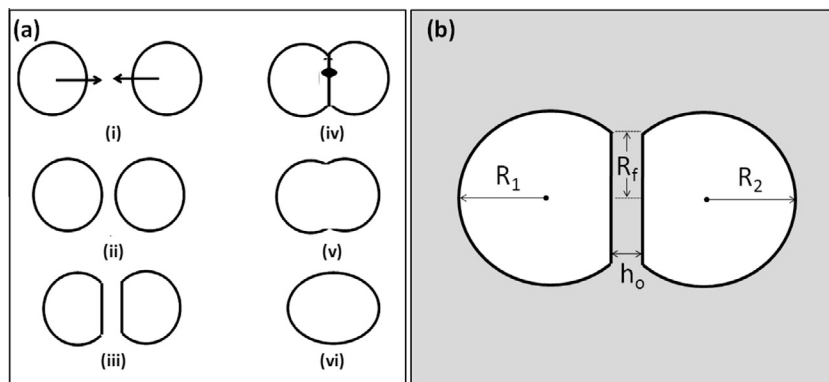
where  $R_1$  and  $R_2$  are the radii of the two approaching bubbles. If  $W < 0.18$ , the two bubbles will coalesce. Conversely, if the velocity of approach is too fast,  $W > 0.18$ , the bubbles will rebound and not coalesce [7]. Kirkpatrick [12] approximated the coalescence (or film drainage) time for free surfaces (in the absence of surface active solute) by using Eq. (3):

$$t_{Ds} \approx R_f \sqrt{\frac{\rho R_{eq}}{16\sigma}} \ln \left( \frac{h_o}{h_c} \right) \quad (3)$$

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**Fig. 1.** Schematic representation of (a) coalescing process of two colliding bubbles: approach [i–ii], flattening of the interposed film [iii], drainage to a critical thickness and there is a dimple on the film [iv], film rupture [v], and formation of a single bubble [vi], and (b) variables used for two bubble in contact.

where  $R_f$  is the radius of the contacting area,  $h_o$  is the initial film thickness and  $h_c$  is the critical film thickness at which the film ruptures (see Fig. 1(b)). The initial and critical thickness of this film has been reported [2] to be in the range of 1–10  $\mu\text{m}$  and 0.01  $\mu\text{m}$ , respectively.

If surface active solutes are present at a sufficient surface concentration, the interface could be considered to be immobile (no-slip) [2,4,20] and Eq. (4) is then used to approximate the drainage time [21]:

$$t_{Dn} \approx \frac{3\eta R_{eq} R_f^2}{8\sigma h_c^2} \quad (4)$$

where  $\eta$  is liquid dynamic viscosity.

When bubbles are subjected to an acoustic field, the coalescence process is further complicated by the Bjerknes forces exerted on the bubbles. Depending on their size, bubbles can be drawn towards each other by both primary and secondary Bjerknes forces, with the latter force being dominant at close ranges [22]. The secondary Bjerknes force can also cause the bubbles to repel each other, if the oscillations of the two bubbles are strongly antiphase [23,24]. Details regarding the theoretical calculation of the secondary Bjerknes force were provided by Doinikov [25–27] and in our previous reports [22,28]. The time averaged secondary Bjerknes force ( $F_B$ ) is related to the average volume oscillation of two bubbles within one acoustic cycle ( $\langle \dot{V}_1 \dot{V}_2 \rangle$ ) and is described by Eq. (5) [25]:

$$F_B = \langle F_{12} \rangle = \frac{\rho}{4\pi r_{12}^2} \langle \dot{V}_1 \dot{V}_2 \rangle = \frac{4\pi\rho}{r_{12}^2} \langle R_1^2 \dot{R}_1 R_2^2 \dot{R}_2 \rangle \quad (5)$$

where  $r_{12}$  is the separation distance between the two bubbles (taken from the center of the bubbles), and the over-dot denotes the time derivative. Eq. (5) shows that the volume change in one acoustic cycle is related to the changes in both radii of the two bubbles ( $R_1$  and  $R_2$ ) and their radial velocities ( $\dot{R}_1$  and  $\dot{R}_2$ ).

Theoretical studies have assumed if two bubbles are drawn together by Bjerknes forces, they will coalesce at encounter [29,30]. There are a few studies on bubble coalescence in an acoustic field. Lee et al. [31] used a total bubble volume method, which stems from the same principle as that reported by Labouret et al. [32], to examine the effect of surface-active solutes on bubble coalescence in the presence of ultrasound (multibubble cavitation). Sunartio et al. [33] and Browne et al. [34] extended the study to investigate various frequency, power, and water-soluble additives on bubble coalescence. However, these studies were performed under multi-bubble systems and at frequencies where coalescence between bubbles would be difficult to image. In order to

understand the role of the secondary Bjerknes force on bubble coalescence, there is a need to study the coalescence of two isolated bubbles in an acoustic field. Crum [35] reported that coalescence of bubbles is normally observed under the influence of Bjerknes forces in a stationary wave field. However, Duineveld [36] demonstrated that this was not always the case and found when two equal size bubbles are driven near resonance, coalescence is inhibited. This was attributed to Bjerknes force induced bubble oscillations causing the contacting surfaces to fluctuate too fast for coalescence to take place. This was further substantiated by the Weber number, which was calculated to be near the critical value of 0.18. In a more recently study, Postema et al. [21,37] investigated the coalescence of lipid coated ultrasound contrast agent bubbles in an acoustic field. They showed that some bubbles bounce off each other, while others show very fast coalescence during bubble expansion caused by the rupturing of the lipid shell to expose the clean bubble surface. This was supported by a good agreement between the experimental coalescence time and that calculated using Eq. (3) for free bubble surface. These studies were performed at high frequencies (0.5 MHz and 1.7 MHz) and high acoustic pressures (0.66–0.85 MPa) and for small microbubbles (radii < 2.5  $\mu\text{m}$ ). It is unclear whether secondary Bjerknes force played a role, or perhaps was insignificant under the high frequency conditions studied.

It has been shown both experimentally and theoretically that at low acoustic frequencies the magnitude of the secondary Bjerknes force is strongly affected by the size of the two approaching bubbles [22,23,28]. This study aims to compare variations in the secondary Bjerknes forces calculated theoretically to the experimentally measured film drainage time for two bubbles driven at an acoustic frequency of 22.4 kHz and acoustic pressures from 10 kPa to 120 kPa.

## 2. Experimental and theoretical methods

### 2.1. Experimental method

The details of the experiment setup used to study the bubble coalescence process were described in our previous report [38]. A transducer (American Piezo Ceramics Inc. Z7) was driven at 22.4 kHz by a frequency generator (HM8131-2) coupled to a power amplifier (Krohn-Hite 7500). The bubble images were magnified using a long-distance microscope, and recorded using a high-speed video camera (Y4-PIV, IDT) at a frame rate of 4000 frames per second. The acoustic pressure was measured using a needle hydrophone (Precision Acoustics Ltd., 1.0 mm needle with preamp).

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