Acoustic cavitation in emerging applications : membrane cleaning and flow processing

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**NANYANG
TECHNOLOGICAL
UNIVERSITY**

**ACOUSTIC CAVITATION FOR EMERGING
APPLICATIONS: MEMBRANE CLEANING AND FLOW
PROCESSING**

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ACOUSTIC CAVITATION FOR EMERGING APPLICATIONS: MEMBRANE CLEANING AND FLOW PROCESSING

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Acknowledgments

A thesis is a culmination of a PhD work; it summarizes in some details the important results of a research work. However, in the body of the thesis there is not any chance to tell that research is actually not only a cumulative activity of some geeks exchanging scientific ideas, tinkering the experiments, deriving formulas, discussing results and writing reports, but also made possible by the supports of not-so-geek individuals, keeping the humanity side of the researchers intact. Therefore, here I would like to thank some people (geeks and non-geeks) for being there during my PhD pursuit.

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Contents

Acknowledgements	i
List of Figures	v
List of Tables	xi
Abstract	xiv
1 Introduction	1
1.1 History and motivation	3
1.2 Organization of the thesis	10
2 Bubble radial dynamics	12
2.1 RPNP equation	12
2.2 Compressibility correction	16
2.3 Damping mechanisms	18
2.4 Bubble near boundaries	21
3 Small amplitude oscillations of bubble near a rigid boundary and the generation of wall shear stress	24
3.1 Introduction	24
3.2 Theory	26
3.2.1 Bubble model	26
3.2.2 Effect of the wall	29
3.2.3 Wall shear stress formulation	29
3.3 Steady wall shear stress	31
3.4 Parameter study of the wall shear stress	33
3.5 Discussions	39
3.6 Conclusions	42
4 Surface oscillations and jetting from surface attached acoustic driven bubbles	44
4.1 Introduction	44
4.2 Experimental setup	46
4.3 Experimental results	49

4.4	Discussion and Conclusions	55
5	Improved ultrasonic cleaning of membranes with tandem frequency excitation	59
5.1	Introduction	59
5.2	Experimental setup	63
5.3	Results	67
5.3.1	Bare membrane	67
5.3.2	Membrane with filter cake	69
5.3.3	Tandem frequency validation experiments	74
5.4	Discussion	77
5.5	Conclusions	79
6	Multi-bubble sonoluminescence in flowing liquid	81
6.1	Introduction	81
6.2	Experimental setup	83
6.3	Experimental results	86
6.4	Discussion	98
6.5	Conclusions	101
7	Conclusions and outlook	103
7.1	Conclusions	103
7.2	Outlook	105
	Bibliography	107
A	Derivation of wall shear stress formulation	124
B	Linear wall shear stress	126
C	Numerical method	131

List of Figures

1.1	Jet formation at different dimensionless distances γ between the site of bubble generation and boundary. R_{max} is the maximum bubble radius and ϵ is the bubble elongation before collapse. The series were taken at 20,000 frames/s. The boundary is given by the darker part below the bubbles in the individual frames. The scale can be read from the maximum bubble radius noted. Figure is taken from [23].	5
1.2	Images of an oscillating bubble near a free surface, $\gamma \approx 2.6$. The surface was above the bubble. The bright spot in the center of each frame is plasma, while the black area on the brighter background is a cavitation bubble. Figure is taken from [34].	6
1.3	Interaction of a laser-produced bubble with an elastic boundary, $\gamma \approx 0.76$. The elastic boundary is compressed during bubble expansion and elevated during bubble collapse. The collapse results in bubble splitting with the formation of two liquid jets in opposite directions. (a) Side view, diffuse illumination, $\gamma = 0.77$; (b) side view, parallel illumination, $\gamma = 0.75$. Frame interval $20\mu s$. Frame width 3.5mm. Figure is taken from [37].	7
3.1	Detachment of particles on a flat silicone wafer after 3 hours of contact due to bubble oscillations. The particles are attracted toward the bubble. Figure is taken from [80].	25
3.2	Sketch of the geometry. The distance of the bubble center to the rigid boundary is $r' = r = \sqrt{d^2 + x^2}$, where d is the distance of the bubble center and the wall. The bubble's image center is located $D = 2d$ from its center.	26
3.3	Resonance plots for 5 different equilibrium bubble radii, R_0 , of 0.1, 1, 10, 100, and $1000\mu m$ from top to bottom row with and without a wall being at a distance $d = 1.5R_0$. The first column depicts the resonance parameter ω_0 and the true resonance frequency indicated with the circle. The second column plots the total damping coefficient β while the last column splits it into the contribution of acoustic, viscous, and thermal damping.	30

3.4	The wall shear stress as a function of time. After the initial transient dies out it is composed of a constant wall shear stress, τ_w^{avg} (red line), and a sinusoidal oscillating component τ_w^{osc} represented by its maximum and minimum values (blue lines).	32
3.5	Plot of average (left column) and oscillating (right column) wall shear stress as a function of distance from stagnation point (x) for some bubble center distances from wall (d). R_0 is set as $10\mu\text{m}$. The driving frequency is 298.12kHz. Dimensionless pressure amplitude perturbation, ϵ , is 0.1.	34
3.6	Plot of average (left column) and oscillating (right column) wall shear stress as a function of driving frequency. R_0 is set as $10\mu\text{m}$ and x is set as equal to R_0 . Dimensionless pressure amplitude perturbation, ϵ , is 0.1.	35
3.7	Plot of average (left column) and oscillating (right column) wall shear stress as a function of bubble equilibrium radius for some driving frequencies ($\omega/2\pi$). Bubble distance to wall, d , is set as $1.5R_0$. Distance from stagnation point, x , is set as $2.1R_0$. Dimensionless pressure amplitude perturbation, ϵ , is 0.1.	36
3.8	Plot of average (left column) and oscillating (right column) wall shear stress as a function of driving frequency ($\omega/2\pi$) for some pressure perturbations (ϵ). R_0 are set so that the bubbles are in resonance. Bubble distance to wall, d , is set as $1.5R_0$. Distance from stagnation point, x , is set as $2.1R_0$	37
3.9	Plots of steady (left) and oscillating (right) wall shear stress as a function of dynamic viscosity (μ) for some driving frequencies ($\omega/2\pi$). Dimensionless pressure amplitude perturbation, ϵ , is 0.1. R_0 is set as the resonant radius for each driving frequency. Bubble distance to wall, d , is set as $1.5R_0$. Distance from stagnation point, x , is set as $2.1R_0$	38
3.10	Plots of dynamic viscosities needed to get maximum steady wall shear stress, i.e. the optimum viscosity, (left) and their corresponding steady wall shear stresses (right) as a function of driving frequency. R_0 is set as the resonant bubble equilibrium radius for each driving frequency. Dimensionless pressure amplitude perturbation, ϵ , is 0.1. R_0 is set as the resonant radius for each driving frequency. Bubble distance to wall, d , is set as $1.5R_0$. Distance from stagnation point, x , is set as $2.1R_0$	38
3.11	The nonlinear bubble oscillation (left) and the wall shear stress generated (right). Note that the sinusoidal driving pressure amplitude is 900 kPa with frequency of 1 MHz and the bubble distance to the wall is $12\mu\text{m}$	41
4.1	Plot of the measured pressure amplitude as a function of time. The data are approximated by a straight line with a slope of 0.17 bar/s.	48

4.2	Picture of the attached bubble. Left: one frame taken from the high speed recording of an undisturbed bubble attached to the wall. Right: Sketch of the geometry for the analysis of the bubble shape. $r(\theta)$ is the bubble radius as a function of angle, R_0 is the radius of a circle fitted to undisturbed bubble image, and θ_c is the bubble contact angle with the surface.	49
4.3	Sketch of the experimental setup: The bubble located inside the acoustic chamber is attached to a glass plate. It is driven into radial and surface modes with a function generator and a power amplifier connected to a disc shaped piezoelectric transducer. The scene is illuminated with strong continuous illumination and observed with a long-distance microscope attached to a high-speed camera. . . .	50
4.4	The evolution of the bubble shape with increasing pressure driven at 16.27 kHz. White dashes represent minimum contours, black dashes represent maximum contour, and the bubble image is taken in the intermediate. The positions of the recording of A to D are indicated in Fig. 4.5	52
4.5	Evolution of the envelope of the radial amplitude (r^*) and envelopes of the Fourier mode amplitudes (c_n^*) of the 1st to 5th mode from bottom to top. The bubble is excited twice with a linearly increasing pressure amplitude from 0 to $p = 0.085$ bar. The letters A to D relate to the four frames in Fig. 4.4	54
4.6	Details of the jetting and droplet pinch-off. The number in each frame denotes the time in microseconds. The contrast of the bubble interior is adjusted to aid the reader. It shows the formation of a jet and the subsequent pinch-off of a droplet which impact onto a larger droplet (see arrow in frame j). The length of the bar in the last frame is $100\ \mu\text{m}$	55
4.7	Deformation parameter (α) plot as a function of time. The chaotic oscillations shown in Fig. 4.5 occur once the deformation parameter shows large variability, i.e. at $p = 0.0587$ bar.	57
5.1	Sketch of the experimental setup (A) and the geometry of the basin as seen from the camera position (B). The coordinate system is used in mapping of the pressure field, see Fig. 5.2. The plot in (C) sketches the timing of the two frequencies used in the tandem frequency driving.	64
5.2	(A) and (B): Typical waveforms at the origin showing at maximum driving for the high and low frequency driving, respectively. (C) and (D): Pressure map showing the amplitude distribution in the region of the membrane for the driving frequency of 220 kHz and 28 kHz, respectively. The "+" mark on (C) and (D) denotes the origin and center of the membrane, see Fig. 5.1(B). Note that (C) and (D) are recorded at a reduced driving voltage, therefore the pressure values are lower than in (A) and (B).	65

5.3	Upper row: frames taken from high speed recording of bubbles grown on the bare membrane. Time zero ($t = 0$) is set as the time when the acoustic driving is switched from high to low frequency. Hence, frames in the first column are in the high frequency regime while frames in the second and third columns are in the low frequency regime. Original frames from high speed recording. Lower row: frames subtracted with a background frame to enhance the contrast.	67
5.4	Comparison of the number of bubbles observed as a function of the non-dimensional time on a bare membrane with the TF method ($T = 36\text{ms}$ is the period of oscillation). The symbols represent different peak-to-peak voltage amplitudes of the high frequency driving. The amplitude of the low frequency remains unchanged. At time $t = 0$ the low frequency driving starts.	68
5.5	Frames of bubble activity on membrane covered with bentonite filter cake. (A)SF method. Time zero corresponds to the time when low frequency ($f_l = 28\text{kHz}$) driving starts. Upper: original frames from high speed recording. Lower: frames subtracted with background frame to enhance the bubbles visibility and parts of the cake disturbed by the bubble dynamics. (B) TF method. Time zero is the time when the low frequency driving ($f_l = 28\text{kHz}$) starts after 8 s of high frequency driving ($f_h = 220\text{kHz}$).	70
5.6	Plots of the number of bubbles observed on the membrane covered with bentonite filter cake as a function of time for the treatment with dual and single frequency method. Time zero starts when the low frequency driving starts. For the TF method, the low frequency regime ($f_l = 28\text{kHz}$) starts after 8 s of high frequency driving ($f_h = 220\text{kHz}$), see Fig. 5.1(C).	71
5.7	Comparison of the membrane surface covered initially with a bentonite filter cake before (#0) and after 1-9 cycles (#01-09) of single frequency (SF) and tandem frequency method (TF) on the left and right column, respectively. Both videos were captured at 250 fps.	72
5.8	Bubble growth below the bentonite filter cake. The dashed circles are put in some frames to aid the readers eyes. The arrow points on a cloud of particulates removed from the membrane by the bubble. Upper: original frames from high speed recording. Lower: frames subtracted with background frame to enhance the bubbles visibility.	73
5.9	Bubble oscillating while translating along the top part of the membrane and ejecting materials of bentonite filter cake upwards. Frames are subtracted with background frame to enhance the visibility of the ejected cake.	74
5.10	The transmembrane pressure TMP and the weight of liquid delivered for the three conditions tested. (A)The TMP as a function of time. Regions a-d correspond to the pump-on time during the tandem frequency tests. and (B) The weight delivered by the low pressure system for the three conditions tested, from the weight information the average flux obtained in regions a-d was obtained	76

6.1	Sketch of the experimental setup. Details of the cell is explained in Fig. 6.2. Note that for the first stage of experiment, the illuminations were not used and the camera used was a cooled ICCD camera. For the second stage of the experiment, PMT was not used and the camera used was high speed camera.	84
6.2	Sketch of the cell used. Left picture is the outline of the whole cell. The semi-transparent lines in the left picture represent the upper part of the cell. Note that liquid inlet and outlet are not drawn. The bold lines represent the bottom part of the cell, consist of triangular-shaped metal and piezoelectric transducer. The lines represent the tops of triangular metal parts. The lines also represent the elongation of the triangular metallic structure. Right picture is the cross section of bottom part of the cell which show the metal and transducer parts. Arrows are used to mark the part of left picture which is zoomed as cross section sketch in the right picture.	84
6.3	Impedance plot of the cell as a function of driving frequency. Data are by courtesy of Lam Research.	86
6.4	Sample of PMT output and the driving voltage applied on the piezoelectric transducer taken at zero flow rate (pump is off). Driving of piezoelectric transducer is switch on at $t = 0$ and off at $t = 3$ s. . . .	87
6.5	Samples PMT output signal for the first 4.5 ms after ultrasound irradiation with variation of 0, 10, 20 30, 40 and 45 ml/min flow rates (from top to bottom).	88
6.6	PMT output signal for the first 4.5 ms after ultrasound irradiation. Here, the before (left) and after (right) sonoluminescence events recognition result of the signals using Matlab are demonstrated; zoomed for $t = 2$ to 2.4 ms. Ultrasound of 438 kHz frequency is started at time zero. The event peaks are marked with red triangles in the right picture. Flow rate of water is set as zero (pump is off).	89
6.7	Number of sonoluminescence event captured during the first 4.5 ms of ultrasonic irradiation. The number of event drops drastically, to approximately zero, at flow rate of 45 ml/min.	90
6.8	Sonoluminescence structure captured by ICCD camera; camera and ultrasound exposures are 3 s. Top row: original images from ICCD camera. Bottom row: inverted (bright to dark and vice versa) and contrast-enhanced images. The white (top row) or black (bottom row) scale bars at the bottom of each frame has length of 1 mm. Frames A, B and C depict sonoluminescence at 0, 10 and 20 ml/min of flow rate, respectively.	91
6.9	Flow rate ramp up as a function of time. Letters A to G denote the corresponding flow rate at times 0.2, 0.4, 0.5, 0.55, 0.57, 0.59 and 0.61 s after start of high speed recording as depicted by frame snapshots in Fig. 6.10.	93

6.10	Frame of bubble motion affected by increasing the flow rate. The black scale bar at the bottom of each frame has length of $50\ \mu\text{m}$. Top row are the original frames from the high speed recording, taken at 54000 frame per second. Bottom row are the background subtracted frames of the top row. Letters A to G correspond to the flow rates as indicated in Fig. 6.9, i.e. 4.7, 10.4, 13.4, 14.7, 15.9, 16.2, and 17.2 ml/min, respectively.	94
6.11	Examples of ellipsoidal motion of the bubbles. The image has been processed for better viewing. The black scale bar in window A is $50\ \mu\text{m}$ in length. Window A is the frame showing the initial condition of bubbles a and b. Windows B and C consist of few frame overlays. In Window B, bubble a is moving in ellipsoidal/spiral-like upward motion while 1,2 and 3 denote its position at 20, 80, and $240\ \mu\text{s}$ respectively after window A, while bubble b is almost stationary. In window C, bubble a has moved to 1, 2 and 3 and bubble b has moved to 4 and 5. Positions 1, 2 and 4, 3 and 5 are at 780, 840, and $940\ \mu\text{s}$ respectively after window A. The arrows are added to aid the reader on the direction of bubble motion.	95
6.12	Bubble translation dynamics at no flow. The arrow in left of the figure represents the direction of the liquid flow and the elongation of triangular structure of the metal plate (see Fig. 6.2). The direction is the same for the rest of the figures. The black scale bar on the last frame is $50\ \mu\text{m}$ in length.	96
6.13	Bubble translation dynamics at 20 ml/min flow rate. Similar to Fig. 6.12, upward is the direction of the liquid flow and the elongation of triangular structure of the metal plate (see Fig. 6.2). The black scale bar on the last frame is $50\ \mu\text{m}$ in length.	97
6.14	Bubble translation dynamics at 40 ml/min flow rate. Similar to Fig. 6.12, upward is the direction of the liquid flow and the elongation of triangular structure of the metal plate (see Fig. 6.2). The white scale bar on the first frame is $50\ \mu\text{m}$ in length.	97
C.1	Plot of wall shear stress calculated using analytical and numerical solution.	132

List of Tables

5.1	Average flux obtained in regions a-d with the three different conditions tested, pure water, water-bentonite solution and tandem frequency.	77
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List of Symbols

Romans

A Hamaker constant

A_p Projection area

c Speed of sound in the liquid

d Distance of the bubble center to the rigid wall

D Distance between the centers of real and imaginary bubbles

D_{th} Thermal diffusivity

f Driving frequency

P_a Acoustic pressure amplitude

$p(R)$ Pressure at bubble wall

p_∞ Pressure at infinity; static pressure

$p(t)$ Varying pressure field

p_v Vapor pressure in the bubble interior

R Bubble radius

$\dot{R}$ Bubble wall velocity

$\ddot{R}$ Bubble wall acceleration

R_0 Bubble equilibrium radius

R_ϵ Small radial amplitude perturbation of the bubble

$\Re(\Phi)$ The real part of a complex variable Φ

$\Im(\Phi)$ The imaginary part of a complex variable Φ

Z_0 Distance between the particle and a flat surface

Greeks

β Total damping

β_{ac} Acoustic damping

β_{th} Thermal damping

β_{vis} Viscous damping

$\Delta(R)$ Pressure difference at the bubble wall

κ Polytropic exponent

ω Driving frequency

ω_0 Resonance frequency

ν Kinematic liquid viscosity

μ Dynamic liquid viscosity

σ Surface tension coefficient

ρ Liquid density

τ_w Wall shear stress

τ_w^{avg} The average wall shear stress

τ_w^{osc} The oscillating wall shear stress

θ Angle

ϕ Velocity potential

Φ Thermodynamics property of the gas inside the bubble

Σ_{rr} Normal stress in the liquid

Acronyms

MBSL Multi-bubble sonoluminescence

PSL Polystyrene latex

RPNNP Rayleigh-Plesset-Noltingk-Neppiras-Poritsky

SBSL Single-bubble sonoluminescence

TF Tandem frequency

TMP Trans-membrane pressure

US Ultrasound

Abstract

Using theoretical and experimental methods, we investigate potential applications of acoustic cavitation bubbles for cleaning and flow processing.

An important physical parameter for a successful cleaning is the wall shear stress generated on the contaminated surface. In this thesis, a theoretical investigation of wall shear stress generated from weakly oscillating bubbles near a rigid boundary was conducted. We approximate the effect of the rigid boundary using image theory. The important damping parameters, i.e. acoustic, viscous, and thermal damping, are included for radially oscillating bubbles. The model is a modified Keller-Miksis equation for two interacting bubbles. For small-amplitude oscillations, we obtain an analytical solution for the wall shear stress. Parameter study finds that the wall shear stress has maxima with varying frequency, equilibrium bubble radius, lateral distance, and viscosity. At resonance, we observe maximum wall shear stress of up to 1 kPa despite the limitation of the small-amplitude oscillations. The wall shear stress generated from this oscillating bubbles may already be sufficient to clean water filtration membranes used in low pressure system. Yet, to remove very small particulates which are found in the semiconductor industry, it is necessary to drive bubbles above their linear regime.

Here we also study experimentally oscillating gas bubble attached to a rigid surface. By varying the amplitude of the sinusoidal driving, we observed the onset of surface oscillations and the growth of surface oscillation modes. To quantify the shape of surface oscillations, we employ a Fourier transform on the contour. With increasing the driving amplitude three distinctive regimes are observed: radial oscillation with a monotonous increase of the first and second mode, sudden

jump and excitation of higher surface modes, and chaotic surface oscillations. We suggest that the chaotic oscillations are the result of circumferential travelling surface modes. Interestingly we find low velocity jetting. Repeated jetting build up a liquid cushion on the surface. Hence, we conclude the jets may not contribute to surface erosion or cleaning.

The cleaning of water filtration membrane is investigated experimentally. We propose a novel tandem frequency (combination of low and high frequency) cleaning method. Here, the high frequency is used to grow bubbles on the membrane surface and the low frequency to drive these bubbles into large amplitude oscillations which are the cause of the cleaning. We show experimentally using a high speed camera and compare the results with a single frequency method. We measure the trans-membrane pressure (TMP) of a fouled membrane before and after cleaning and find a TMP approximately equal to that of a clean membrane demonstrating the effectiveness of the tandem frequency method. This also suggests that the membrane is not damaged.

Finally, we investigate the effect of flow on acoustic cavitation bubble dynamics. We investigate the multi-bubble sonoluminescence (MBSL) in an acoustic cell with flowing liquid using photomultiplier and ICCD camera. We use the light emission as an indicator for the strength of bubble dynamics. It indicates how rapid the bubble dynamics during collapse. We find that low flow rates do not affect sonoluminescence, however at higher flow rates a strong decrease in sonoluminescence signal is found. Using a high speed camera, we investigate bubble dynamics and find that at low flow rate the typical bubble configuration is similar to Miller arrays, i.e. big bubbles surrounded by small satellite bubbles which are spiralling.

However, at high flow rate we find that the big bubbles are dragged away by the flow, hence disturbing the stable configuration of the Miller array. We conclude that the disappearance of the stable Miller array is the cause of the reduction in light emission.

Chapter 1

Introduction

A bubble, as stated by Prosperetti [1], is emptiness, non-liquid, tiny-cloud shielding a mathematical singularity. Bubbles are all around us and have many forms, including the familiar soap bubbles. However, in this thesis we will limit our discussion into specific kinds of bubbles. They are gas bubbles driven into small and large amplitude oscillations in liquid. They are commonly termed cavitation bubbles.

A gas bubble consists of a volume of permanent gas surrounded by liquid. This kind of bubble is relatively stable; its lifetime is given by the diffusion of the gas out of the bubble into the surrounding liquid. A cavitation bubble can be caused by a pressure drop below the liquid vapour pressure. The rupture of liquid which causing the formation of cavity due to sufficiently low pressure is commonly termed cavitation inception. Another way to generate a bubble is through deposition of energy in the liquid, e.g. using an intense focused laser pulse or an electrical discharge, to vaporize a volume of liquid. This leads to creation of small volume of vapour which expands explosively into a cavitation bubble.

In contrast with inertial cavitation which collapse due to inertia of the surrounding liquid, non-inertial cavitation bubbles are a type of bubbles which undergone forced oscillations due to an acoustic field [2]. In general, they grow from small gas bubble nuclei which already exist in the liquid. While oscillating, their gas content may increase due to a larger influx than outflux of gas during an oscillation cycle. This phenomenon is termed rectified diffusion [2, 3].

Cavitation bubbles which occur naturally may have unwanted effects. Aside from figuring how to avoid the effects, research has figured out some of their beneficial properties. Cavitation bubbles are the cleaning agents in ultrasonic cleaning bath which are used to clean glasses and other optical delicate surfaces, jewellery, watches, coins, tools and many more. They are also used for wafer cleaning in the semiconductor industry. Kidney stones may also be medically treated by shockwaves which lead to the formation of cavitation bubbles. This procedure is called extracorporeal shockwave lithotripsy [4]. In addition, gas bubbles with a shell (usually made from lipid, albumin, galactose or polymers) are used as ultrasonic contrast agents; their oscillations driven by diagnostic ultrasound head in the bloodstream provides a better contrast for medical diagnostic purposes [5]. Further, they may be used for stretching or breaking up cell; the latter is called cell sonoporation [6, 7]. Cavitation bubbles are also useful to enhance liquid mixing [8], an important feature in flow processing.

This chapter, as starting point of the thesis, begins with a brief literature review on cavitation bubble research. Early history and some important milestones, especially in acoustic cavitation bubbles, are described. Finally, the organization of the thesis is detailed. In this thesis, gas and cavitation bubbles will be in-

investigated theoretically and experimentally with applications to cleaning and flow processing in mind. To be specific, this thesis discusses theoretical investigation of a gas bubble weakly oscillating near a rigid wall, experimental investigations of oscillating bubbles attached on a rigid wall and on water filtration membrane. Their implications on cleaning are also discussed. Lastly, multi-bubble dynamics and sonoluminescence are also experimentally investigated and their interplay with a streaming flow is discussed.

1.1 History and motivation

Cavitation was first studied due to its damaging effect on mechanical tools, such as turbines and ship propellers. It was a serious problem which forced the British Admiralty in the early twentieth century to initiate a special commission to investigate and report that the problem was caused mainly by the cavitation collapse [9]. Theoretical analysis of a spherically collapsing bubble in an infinite incompressible liquid by Rayleigh [10] kick-started this research early last century. Rayleigh found that very high pressure is developed during the last stage of the bubble collapse, which may help to explain a possible cause of the damage. However, it took many years to understand it much better, see [11] and [12]).

In Rayleigh's first analysis he neglected surface tension and liquid viscosity, still he obtained rather accurate account for the collapse time. It is known as *the Rayleigh collapse time*:

$$\tau_c = 0.915 R_{max} \left(\frac{\rho}{p_\infty - p_b} \right)^{0.5}, \quad (1.1)$$

where ρ is density of the water, p_∞ is hydrostatic pressure and p_b is the water vapor

pressure.

By letting the ambient pressure vary in time, e.g. acoustic wave or in a liquid flow, many experiments results have already been understood, see e.g. [13]. Surface tension and liquid viscosity can be included in boundary conditions [14, 15, 16]. This formulation is commonly termed as the classical *Rayleigh-Plesset equation* [17, 2]. Lauterborn [18] proposed the equation to be termed Rayleigh-Plesset-Noltingk-Neppiras-Poritsky (RPNNP) equation, following the names of researchers contributing to its mathematical development. The equation will be explained in the next chapter.

A major simplification inherent in this equation is the assumption that the liquid is incompressible. Corrections of this equation which take into account the effect of liquid compressibility have been proposed by Gilmore [19] and Keller and Miksis [20]. It turned out later that these are mathematically equivalent, i.e. they are all first-order corrections. This was shown rigorously by Prosperetti and Lezzi [21]. Later Lezzi and Prosperetti [22] derived a corresponding theory which is correct to second-order in taking into account the liquid compressibility.

Near boundaries bubbles do not collapse spherically. To characterize this situation, the stand-off parameter, γ , defined as:

$$\gamma = \frac{d}{R_{max}} \quad , \quad (1.2)$$

where d is the distance of the bubble center to the boundary and R_{max} is the bubble radius at maximum expansion, is usually used. Near a rigid boundary, bubbles collapse while moving towards the boundary and may develop a liquid jet in the direction of motion [9]. An example of a bubble collapses in the vicinity of rigid boundary at different stand-off parameters γ is shown in Fig. 1.1 [23]. Pioneering

Figure 1.1: Jet formation at different dimensionless distances γ between the site of bubble generation and boundary. $R_{\max}$ is the maximum bubble radius and ϵ is the bubble elongation before collapse. The series were taken at 20,000 frames/s. The boundary is given by the darker part below the bubbles in the individual frames. The scale can be read from the maximum bubble radius noted. Figure is taken from [23].

Figure 1.2: Images of an oscillating bubble near a free surface, $\gamma \approx 2.6$. The surface was above the bubble. The bright spot in the center of each frame is plasma, while the black area on the brighter background is a cavitation bubble. Figure is taken from [34].

investigations of the phenomena are available in the works by Naude & Ellis [24] and Benjamin & Ellis [25]. Their work suggested a mechanism of cavitation damage: the bubble collapses aspherically near a rigid boundary and generates a fast liquid jet impacting the boundary. This observation stimulated other groups [26, 27, 28] to investigate in more details the bubble collapse near to rigid boundary. Among them, Lauterborn and Bolle [27] have validated experimentally numerical result from Plesset and Chapman [29].

Meanwhile, near a free surface, the aspherical bubble collapse was first investigated in detail by Voinov & Voinov [30], Chahine [31], and Blake *et al.* [32, 33]. In case of a free surface, the bubble moves away from the surface and again during collapse develops a jet in the direction of its motion [9]. This jet is now directed away from the free surface. An example of this dynamics is presented in Fig. 1.2 [34].

(a)

(b)

Figure 1.3: Interaction of a laser-produced bubble with an elastic boundary, $\gamma \approx 0.76$.

The elastic boundary is compressed during bubble expansion and elevated during bubble collapse. The collapse results in bubble splitting with the formation of two liquid jets in opposite directions. (a) Side view, diffuse illumination, $\gamma = 0.77$; (b) side view, parallel illumination, $\gamma = 0.75$. Frame interval $20\mu\text{s}$. Frame width 3.5mm . Figure is taken from [37].

Not too surprising, near an elastic boundary the bubble behaves somehow in between. Depending on the properties of the boundary, it may develop a jet toward or away from the boundary. The boundary may also store some of the kinetic energy of the liquid and deliver it back at later time [9]. Pioneering study in the fields were done by Gibson and Blake [26, 35, 36]. For an example of this phenomena, see Fig. 1.3 [37]. For further discussions regarding cavitation near boundaries, especially in the early years, readers are advised to consult the excellent review by Blake and Gibson [9].

Aside from the jetting collapse, an important aspect of bubble collapsing or oscillating near rigid and elastic boundary is the wall shear stress generated on the boundary due to liquid motion. It is the physical mechanism responsible for ultrasonic cleaning [38, 39]. It is also responsible for the rupture of cell membrane in sonoporation [40, 41]. Dijkink and Ohl [42] did the first detailed measurement of the wall shear stress generated from collapsing laser-induced bubbles near a rigid boundary. The study focused on the jet causing wall shear stress, such as in the work by Dijkink *et al.* [43] where the wall shear stress is estimated from the experiment using theoretical result from Glauert [44].

Image theory [45] has been used to model bubble oscillating near a rigid boundary. This is the first step before calculating the wall shear stress on the solid boundary due to bubble oscillations. This approach has been used in the microbubble and cell sonoporation applications [46, 47]. However the approach may not be accurate since cells are not truly rigid. A different approach, based on the assumption that the cell is a liquid with a different density, led to very different result [48].

The wall shear stress responsible for sonoporation is usually calculated following

acoustic streaming formulation by Nyborg [49]. However, this approach neglects the instantaneous oscillating wall shear stress. The steady wall shear stress, or the steady streaming, may be useful to transport the detached particulates away from the surface and keeping them to be reattached on the surface. For simplicity, one can assume small amplitude radial bubble oscillations and neglect wall shear stress generated by bubble translation. Small amplitude oscillations eliminate the possibility of jetting.

The wall shear stress has been shown as an important factor for cleaning application [42]. Aside from the movements of the bubble wall, this physical quantity may also come from microjets, generated during the asymmetrical collapse of the bubble, impacting on the surface. High jet impact velocities are leading to a high wall shear stress and successful cleaning [50, 42]. An oscillating attached gas bubble has been shown experimentally to generate rather low velocity jetting, even forming a liquid cushion on the surface. This renders it less useful for cleaning [51]. A particular case will be the subject of chapter 4.

Another phenomenon unique to bubble dynamics is sonoluminescence. It is the conversion of sound energy into light due to extreme conditions (i.e. temperature and pressure) reached inside bubble when it collapses sufficiently violent. Multi-bubble sonoluminescence (MBSL) was first discovered indirectly in 1933 by Marínescu and Trillat [52], then approximately a year later clearly demonstrated by Frenzel and Schultes [53]. Gaitan *et al.* [54] first discovered stable single-bubble sonoluminescence (SBSL) which is a huge advancement in the field. SBSL allows the pretty well-developed radial dynamics theory for single bubble to be applied directly to explain and predict experimental results.

Many aspects of SBSL has been investigated experimentally and the theory is quite matured, too. Most researchers now believe that the mechanism of the light emission rely on the very efficient process of energy focusing that cavitation bubble possess [55, 56, 57, 58]. The bubble dynamics from experiments fit quite well with RPNNP equation which is derived using classical hydrodynamics approach; some corrections (i.e. compressibility, gas thermodynamics of the bubble interior, etc.) usually increase the agreement even further.

In contradiction, a satisfying theory of MBSL has yet to be found despite the abundance of the experimental data available due to its earlier discovery than SBSL. One of the remaining problems is that the many bubbles system render the situation much more complex due to bubble–bubble interactions. Further discussions about SBSL and MBSL can be found in an excellent review by Brenner *et al.* [58].

1.2 Organization of the thesis

The thesis deals with the bubble dynamics and its application in cleaning and flow processing.

In chapter 2 we will describe and briefly derive the basic theory of bubble dynamics, especially RPNNP and Keller-Miksis equations as representations of spherical bubble dynamics theory in incompressible and compressible liquids, respectively. A brief description regarding gas thermodynamics theory of the bubble interior will also be given.

In chapter 3 we will cover the extension of the basic theory presented in the previous chapter. Here a bubble which is mildly oscillating near a rigid wall is con-

sidered, taking into account the liquid compressibility. Later the bubble dynamics will be used to calculate the wall shear stress on the rigid boundary where an analytical expression for the wall shear stress is derived. Parameter study affecting the wall shear stress and its relevance to cleaning application is done.

In chapter 4 we will discuss an experiment of bubble attached to a rigid wall. Amplitude modulation of the acoustic driving pressure and high speed recording enables the study of the onset of the surface oscillations. The Fourier transform is used to quantify the shape oscillations by asserting mode number. Predictions using potential theory are confirmed. Jetting is also observed and its contribution to surface cleaning and erosion is discussed.

Chapter 5 presents a novel method to improve the ultrasonic cleaning of water filtration membrane. Tandem frequency method, in which successive application of high and low frequency ultrasound is employed, is shown to be better in cleaning than with single frequency. We also discuss its connection to bubble dynamics, in fact it is the bubbles that do the cleaning as can be seen from the experiments.

In chapter 6 we will report on the sonoluminescence in a flowing liquid. Along with already existent small bubble nuclei, seed bubbles are also introduced with a bubble generator which become trapped and driven to volume oscillation in the acoustic field. High flow rates affect the sonoluminescence, as seen with a photomultiplier and an ICCD camera. The bubbles translation dynamics is also investigated by high speed recording.

In chapter 7 we give the summary of the thesis and list some important conclusions with suggestions for future direction of research.

Chapter 2

Bubble radial dynamics

2.1 RPNNP equation

Common model for the radial dynamics of a bubble in a liquid is the RPNNP equation. It can be derived from mass conservation and the Navier-Stokes equation in spherical coordinates using spherical symmetry. Here we follow a common approach, see for example Brennen [3] and Leighton [2]. First, due to conservation of mass the radial velocity, $u(r, t)$, is of the form:

$$u(r, t) = \frac{F(t)}{r^2} \quad , \quad (2.1)$$

where r is radial distance from the center of the bubble and $F(t)$ is an arbitrary function of time. If there is no mass transport across bubble wall, the velocity at the wall is

$$u(R, t) = \dot{R} = \frac{F(t)}{R^2} \quad , \quad (2.2)$$

where R is the bubble radius. Hence, one can see easily that $F(t) = R^2 \dot{R}$.

In a Newtonian liquid, the Navier-Stokes equation in spherical coordinates as-

suming spherical symmetry is

$$\rho \left(\frac{\partial u}{\partial t} + u \frac{\partial u}{\partial r} \right) = -\frac{\partial p}{\partial r} + \mu \left[\frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial u}{\partial r} \right) - \frac{2u}{r^2} \right] , \quad (2.3)$$

where ρ is the liquid density, p is the pressure, and μ is the dynamic viscosity of the liquid. Since the kinematic viscosity is $\nu = \mu/\rho$ and by substituting Eq. 2.1 one gets:

$$-\frac{1}{\rho} \frac{\partial p}{\partial r} = \frac{1}{r^2} \left(2R\dot{R}^2 + R^2\ddot{R} \right) - \frac{2R^4}{r^5} \dot{R}^2 , \quad (2.4)$$

where the viscous terms cancel during substitution. By separation of variables and integrating from the bubble wall, $r = R$, to infinity, $r \rightarrow \infty$, we obtain

$$\frac{p(R) - p_\infty}{\rho} = R\ddot{R} + \frac{3}{2}\dot{R}^2 , \quad (2.5)$$

where $p(R)$ and p_∞ are pressure at bubble wall and at infinity, respectively. Pressure at infinity for the simplest case may be static, p_∞ . A more general pressure term, p_s , may be expressed as

$$p_s = p_\infty + p(t) \quad (2.6)$$

which is a pressure as a function of time $p(t)$ superimposed onto static pressure p_∞ .

The normal stress in the liquid which points outwards radially from the bubble center is given by

$$\Sigma_{rr} = -p + 2\mu \frac{\partial u}{\partial r} . \quad (2.7)$$

At the bubble wall, the net force per unit area is zero if there is no mass transfer.

Hence the boundary condition at the bubble wall is

$$\Sigma_{rr}(R) + p_b - \frac{2\sigma}{R} = 0 , \quad (2.8)$$

where p_b and σ are pressure inside the bubble and surface tension, respectively.

Thus,

$$-p(R) + 2\mu \left[\frac{\partial u}{\partial r} \right]_{r=R} + p_b - \frac{2\sigma}{R} = 0 \quad . \quad (2.9)$$

Since no mass transfer across bubble wall is assumed, at the bubble wall one gets the stress as

$$-p(R) - \frac{4\mu}{R} \dot{R} + p_b - \frac{2\sigma}{R} = 0 \quad , \quad (2.10)$$

This leads to

$$p(R) = p_b - \frac{4\mu}{R} \dot{R} - \frac{2\sigma}{R} \quad . \quad (2.11)$$

The pressure inside bubble is often approximated with a uniform mixture of an ideal gas and vapour from the liquid. Assuming further that the vapour pressure p_v is constant and that the gas behaves polytropically with a constant polytropic exponent κ . The pressure inside the bubble may be expressed as [2]

$$p_b = \left(p_\infty + \frac{2\sigma}{R_0} - p_v \right) \left(\frac{R_0}{R} \right)^{3\kappa} + p_v \quad , \quad (2.12)$$

where R_0 is the equilibrium bubble radius.

Substituting Eq. 2.11 to Eq. 2.5 leads to

$$\frac{p_b - p_s}{\rho} - \frac{4\mu \dot{R}}{\rho R} - \frac{2\sigma}{\rho R} = R\ddot{R} + \frac{3}{2}\dot{R}^2 \quad . \quad (2.13)$$

By letting $\nu = \mu/\rho$, a familiar form of the RPNNP equation is retrieved as [3]

$$\frac{p_b - p_s}{\rho} - \frac{4\nu \dot{R}}{R} - \frac{2\sigma}{\rho R} = R\ddot{R} + \frac{3}{2}\dot{R}^2 \quad . \quad (2.14)$$

Analytical solutions of RPNNP equation, i.e. Eq. 2.14, are mostly unknown, however they can be obtained numerically. Lauterborn [18] was the first to report on extensive numerical solutions of RPNNP equation in mid-1970s. Nevertheless, if

one neglects surface tension and viscosity which transforms the RPNNP to Rayleigh equation [10], high-order analytical approximations are available [59].

In small-amplitude oscillation regime, the nonlinear RPNNP equation can be approximated as a linear resonator. Therefore, the amplitude of the radius change, R_ϵ must be small,

$$R = R_0 + R_\epsilon \quad . \quad (2.15)$$

For simplicity and without loss of generality, one may neglect the liquid viscosity and vapour pressure [2]. Then by first-order Taylor expansion of the equation in powers of R_0^{-1} , one will obtain

$$\ddot{R}_\epsilon + \omega_0^2 R_\epsilon = \frac{P_a}{\rho R_0} e^{i\omega t} \quad , \quad (2.16)$$

where P_a is the driving pressure amplitude. Eq. 2.16 is called the linearized equation of Eq. 2.14 excluding viscosity. Hence, this method is termed as a linearization about bubble equilibrium radius R_0 . One may recognize Eq. 2.16 as the familiar forced harmonic oscillator. The resonance frequency ω_0 is

$$\omega_0 = \sqrt{\frac{1}{\rho R_0^2} \left[3\kappa \left(p_\infty + \frac{2\sigma}{R_0} \right) - \frac{2\sigma}{R_0} \right]} \quad . \quad (2.17)$$

In the case when the surface tension terms is negligible, i.e. large bubbles, Eq. 2.17 reduces to

$$\omega_0 = \frac{1}{R_0} \sqrt{\frac{3\kappa p_\infty}{\rho}} \quad , \quad (2.18)$$

which is the same as the result derived by Minnaert [60]. At the other extreme, for very small bubbles so that surface tension dominates, Eq. 2.17 reduces to

$$\omega_0 = \sqrt{\frac{2\sigma}{\rho R_0^3} (3\kappa - 1)} \quad . \quad (2.19)$$

If one includes viscosity and vapour pressure, the full expression for resonance frequency is

$$\omega_0 = \frac{1}{R_0\sqrt{\rho}} \sqrt{\left[3\kappa \left(p_\infty + \frac{2\sigma}{R_0} - p_v \right) - \frac{2\sigma}{R_0} + p_v - \frac{4\mu^2}{\rho R_0^2} \right]} . \quad (2.20)$$

2.2 Compressibility correction

In the case of a sufficiently high bubble wall velocity, liquid compressibility become important. Different approaches have been taken by previous researchers. Most of them have been shown to be mathematically equivalent to a first-order compressibility correction. Here we will re-derive one famous version of these equations, the Keller-Miksis equation following Keller and Miksis [20].

Consider the velocity potential ϕ in the liquid, the governing equations in the liquid are the modified Bernoulli equation

$$\frac{\partial \phi}{\partial t} + \frac{1}{2} \left(\frac{\partial \phi}{\partial r} \right)^2 - \frac{4\mu}{3} \frac{\partial^2 \phi}{\partial r^2} + \int_{p(\infty)}^{p(r)} \frac{dp}{\rho} = 0 \quad (2.21)$$

and the wave equation

$$\frac{\partial^2 \phi}{\partial r^2} - \frac{1}{c^2} \frac{\partial^2 \phi}{\partial t^2} = 0 \quad (2.22)$$

where c is constant sound wave speed in liquid. The solution of the velocity potential and its derivatives are

$$\phi(r, t) = \frac{f(t - r/c) + g(t + r/c)}{r} , \quad (2.23)$$

$$\frac{\partial \phi}{\partial t} = \frac{f' + g'}{r} , \quad (2.24)$$

$$\frac{\partial \phi}{\partial r} = \frac{g' - f'}{rc} - \frac{f + g}{r^2} , \quad (2.25)$$

where f and g are arbitrary functions and primes means differentiation with respect to the corresponding argument.

The system enthalpy can be expressed as

$$\int_{p(\infty)}^{p(R)} \frac{dp}{\rho} = \frac{p - p_\infty}{\rho} \quad (2.26)$$

where p is the liquid pressure. At the bubble wall, the velocity and gas pressure inside the bubble are

$$\dot{R} = \frac{\partial \phi(R, t)}{\partial r} \quad (2.27)$$

and

$$p_b(R, t) = p(R, t) + \frac{2\sigma}{R} - \frac{4\mu}{3} \left(\frac{\partial^2 \phi(R, t)}{\partial r^2} - \frac{1}{R} \frac{\partial \phi(R, t)}{\partial r} \right) , \quad (2.28)$$

respectively. Substituting back to Eq. 2.21, one gets an expression for the pressure difference

$$\Delta(R) = \frac{p_b(R, t) - p_\infty}{\rho} = \frac{2\sigma}{\rho R} + \frac{4\mu}{\rho R} \frac{\partial \phi(R, t)}{\partial r} - \frac{\partial \phi(R, t)}{\partial t} - \frac{1}{2} \left(\frac{\partial \phi(R, t)}{\partial r} \right)^2 . \quad (2.29)$$

By substituting Eqs. 2.23, 2.24, 2.25, 2.27 and 2.28 into Eq. 2.29, one obtains

$$R\Delta(R) - cR\dot{R} = c \frac{f + g}{R} - \frac{R\dot{R}^2}{2} + \frac{4\mu R}{\rho} + \frac{2\sigma}{\rho} - 2g'(t - R/c) . \quad (2.30)$$

The time derivative of the velocity potential $d\phi/dt$ is found from the derivative of Eq. 2.30. Then, by substituting the result into Eq. 2.29 one gets

$$\begin{aligned} R\ddot{R} \left(1 - \frac{\dot{R}}{c} \right) + \frac{3}{2} \dot{R}^2 \left(1 - \frac{\dot{R}}{3c} \right) &= \Delta(R) \left(1 - \frac{\dot{R}}{c} \right) + \frac{R\Delta'(R)}{c} \\ &\quad - \frac{4\mu\ddot{R}}{\rho c} - \frac{2\sigma}{\rho R} + \frac{2}{c} \left(1 + \frac{\dot{R}}{c} \right) g''(t - R/c) . \end{aligned} \quad (2.31)$$

Assume the bubble is centred in a spherically symmetric incident sound field with velocity potential

$$Z(r, t) = \frac{g(t + r/c) + h(t - r/c)}{r} . \quad (2.32)$$

To avoid singularity in the origin, one must set $h = -g$. Thus, one may calculate the limit at the origin by L'Hopital rule. Thus,

$$Z(r, t) = \frac{g(t + r/c) - g(t - r/c)}{r} , \quad (2.33)$$

and by L'Hopital rule

$$Z(0, t) = \frac{2}{c} g'(t) . \quad (2.34)$$

After shifting the time by R/c then differentiating with respect to time, one obtains

$$2g''(t - \frac{R}{c}) = c \frac{\partial}{\partial t} Z(0, t - \frac{R}{c}) . \quad (2.35)$$

Suppose the incident pressure field is a plane wave with angular frequency ω and pressure amplitude P_a . Then

$$Z(r, t) = - \left(\frac{P_a}{\rho \omega} \right) \cos \omega \left(t - \frac{R}{c} \right) \quad (2.36)$$

and

$$2g''(t) = \frac{c P_a}{\rho} \sin \omega t . \quad (2.37)$$

Substituting Eq. 2.37 to Eq. 2.31 yields the so-called Keller-Miksis equation for the bubble radial dynamics

$$\begin{aligned} R\ddot{R} \left(1 - \frac{\dot{R}}{c} \right) + \frac{3}{2} \dot{R}^2 \left(1 - \frac{\dot{R}}{3c} \right) &= \Delta(R) \left(1 - \frac{\dot{R}}{c} \right) + \frac{R\Delta'(R)}{c} \\ &\quad - \frac{4\mu\ddot{R}}{\rho c} - \frac{2\sigma}{\rho R} \\ &\quad + \left(1 + \frac{\dot{R}}{c} \right) \frac{c P_a}{\rho} \sin \omega \left(t + \frac{R}{c} \right) . \end{aligned} \quad (2.38)$$

2.3 Damping mechanisms

When bubble oscillates, viscosity of the liquid dampens the oscillations. This is known as the viscous damping. Another damping mechanism is due to heat

transfer from the bubble interior to the surrounding liquid. These two dampings mechanisms, theoretically, are already apparent from the RPNNP equation.

Thermal damping is an important damping mechanism [61]; it is due to thermal behaviour of the bubble. The usual simplest approximation is to assume the gas inside the bubble behaves polytropically, with the polytropic exponent is set as constant, see previous discussions about RPNNP equation. Its value can be varied between the two extreme cases: adiabatic and isothermal. However, more complete treatments based on conservation equations are possible under the assumption of small amplitude bubble oscillation [61]. Further, assuming uniform pressure inside gas bubble [62, 63, 64] experimental results on the propagation of nonlinear pressure waves in bubbly liquids can be explained [65, 66].

Derivation of the thermal behaviour is quite involved, hence the reader is referred to the work by Prosperetti *et al.* [63, 64] for a complete picture. In brief, we will only show the result of expression for pressure perturbation amplitude P inside the bubble

$$P = - \left(\frac{3\gamma}{1 - 3(\gamma - 1)\chi \left[\sqrt{i/\chi} \coth(\sqrt{i/\chi}) - 1 \right]} \right) X . \quad (2.39)$$

Here γ is the ratio of specific heats, X is the amplitude of bubble pulsation and χ is the square of the ratio of thermal diffusion length to the bubble radius, i.e.

$$\chi = \frac{D_{th}}{\omega R_0^2}$$

where D_{th} is thermal diffusivity, ω is angular driving frequency and R_0 is the bubble equilibrium radius. Hence, small value of χ means a nearly adiabatic bubble, while large values of χ means a nearly isothermal behaviour of the gas inside the bubble.

To simplify the notation, following [63] Eq. 2.39 may be expressed as

$$P = -\Phi X$$

where Φ is the term in the bracket of Eq. 2.39.

Compressibility corrections (e.g. in Keller-Miksis equation) introduce another damping effect which is absent in incompressible liquid assumption, namely acoustic radiation [20]. High frequency driving renders acoustic radiation as a significant damping mechanism in bubble oscillations. This has been increasingly important because in more recent applications of bubbles, for example as ultrasound contrast agent in medical diagnostics and as cleaning agent in semiconductor industry, the typical driving frequency used is very high, i.e in the order of 10^6 Hz.

Based on the gas thermodynamics model mentioned previously and the Keller-Miksis equation, the linearized equations for small-amplitude oscillations give rise to the forced linear oscillator equation in the form of [65]:

$$\ddot{X} + 2\beta\dot{X} + \omega_0^2 X = \frac{\epsilon p_\infty}{\rho R_0^2} e^{i\omega t} \quad (2.40)$$

along with the expression for the resonance frequency

$$\omega_0^2 = \frac{p_0}{\rho R_0^2} \left(\Re(\Phi) - \frac{2\sigma}{R_0 p_0} \right) \quad , \quad (2.41)$$

where Φ is the term in bracket of Eq. 2.39 and $p_0 = p_\infty + 2\sigma/R_0$. Meanwhile the damping terms are expressed, if β denotes the total damping constant, as

$$2\beta = \frac{\omega^2 R_0}{c} + \frac{p_0}{\omega \rho R_0^2} \Im(\Phi) + \frac{4\mu}{\rho R_0^2} \quad , \quad (2.42)$$

in which the first to the third term are representing acoustic, thermal, and viscous dampings, respectively. Note that $\Re(\Phi)$ and $\Im(\Phi)$ mean real and imaginary part of the complex Φ , respectively.

2.4 Bubble near boundaries

Aspherical oscillations of bubbles due to the presence of a boundary have been investigated experimentally and theoretically. One of the most widely used and successful means for numerical investigations is the boundary element method, sometimes also known as boundary integral method. The method enables us to reduce the dimension of the problem by one due to the discretization of the boundary only [67]. Hence for three-dimensional problems such as a spherical bubble, only the two-dimensional bubble boundary (i.e. bubble wall) needs to be discretized. Details on this method as applied to bubble dynamics can be found on the literature by Blake *et al.* [32, 9, 33, 68], Pozrikidis [69, 67], and Klaseboer, Khoo *et al.* [70, 71, 72] among others.

Analytical solutions for the dynamics of the aspherical bubble collapse and oscillations due to the presence of boundaries are very difficult to obtain if not impossible. However, if the effect of the boundary is weakly coupled to bubble oscillations due to the distance is relatively far then analytical solution is possible. In this case, the bubble still oscillates spherically and in many case the oscillations are further assumed as small-amplitude oscillations. The solution will come from linearized RPNNP-type (or Keller-Miksis type) equation.

As an useful illustration, and because the results will be used in the following chapters, we present here the bubble oscillations near a rigid boundary. To first order, the rigid boundary can be modelled with image theory [45]. In potential flow, a rigid boundary is equivalent to an imaginary bubble located at twice the distance of the real bubble to the wall. Both bubbles are oscillating in phase.

Following Mettin [73], we assume two gas bubbles (real and imaginary) pul-

sating in a liquid driven by an acoustic field. The velocity field may be written as Eq. 2.1 and the liquid motion is generally expressed by Eq. 2.3. The viscosity terms and the nonlinear convective term $u\partial u/\partial r$ may be neglected. Thus, Eq. 2.3 reduces to

$$\rho \frac{\partial u_1}{\partial t} + \frac{\partial p_1}{\partial r} = 0 . \quad (2.43)$$

Here the index 1 denotes the first bubble, so that p_1 means the pressure emitted by the first bubble.

Substitution of Eq. 2.1 to Eq. 2.43 results in the expressions for the pressure gradient and pressure:

$$\frac{\partial p_1}{\partial r} = -\frac{\rho}{r^2} \frac{d}{dt} (R_1^2 \dot{R}_1) \quad (2.44)$$

and

$$p_1 = \frac{\rho}{r} \frac{d}{dt} (R_1^2 \dot{R}_1) , \quad (2.45)$$

respectively. The acoustic field can be represented by an external pressure

$$p(t) = P_a \sin(2\pi ft) \quad (2.46)$$

where f is the driving frequency.

Using the pressure (Eq. 2.45) emitted by bubble 1, one may modify Keller-Miksis equation (see Eq. 2.38) so that for bubble 2 [73]¹

$$\begin{aligned} \left(1 - \frac{\dot{R}_2}{c}\right) R_2 \ddot{R}_2 + \left(\frac{3}{2} - \frac{\dot{R}_2}{2c}\right) \dot{R}_2^2 &= \frac{1}{\rho} \left(1 + \frac{\dot{R}_2}{c}\right) [p(R, t) - p_\infty - p_{ex}] \\ &+ \frac{R_2}{\rho c} \frac{d}{dt} [p(R, t) - p(t)] \\ &- \frac{1}{D} \left(2\dot{R}_1^2 R_1 + R_1^2 \ddot{R}_1\right) \end{aligned} \quad (2.47)$$

where D is the distance between the two bubbles centers. The last term in Eq. 2.47 represents the effect of bubble 1 on bubble 2. Note that the Taylor expansion is

¹Here we have corrected typographical error in the article by Mettin [73].

used for $p(t + R/c)$ term in the original Keller-Miksis equation [20], so that it may be recasted as

$$p\left(t + \frac{R}{c}\right) = p(t) + \frac{R}{c} \frac{dp}{dt} + \frac{1}{2!} \left(\frac{R}{c}\right)^2 \frac{d^2p}{dt^2} + \dots \quad (2.48)$$

Due to the smallness of R/c , it is sufficient to truncate the terms after first order so that

$$p\left(t + \frac{R}{c}\right) \approx p(t) + \frac{R}{c} \frac{dp}{dt} \quad (2.49)$$

Here the total pressure also includes the pressure from bubble 2, so that the coupling terms of order

$$\frac{R}{D} \left(\frac{\dot{R}}{c}\right) \quad , \quad (2.50)$$

$$\frac{R}{D} \left[\frac{d^3}{dt^3} \left(\frac{R}{c}\right)\right] \quad , \quad (2.51)$$

and higher are neglected, hence the difference with the original Keller-Miksis equation [20] (Eq. 2.38). For the case of a bubble and its image, $R_1 = R_2$, and so do their derivatives. Thus the index can be dropped.

The next chapter will use the basic theory developed here to build a model of a bubble oscillating near a rigid boundary taking into account liquid compressibility and important damping terms.

Chapter 3

Small amplitude oscillations of bubble near a rigid boundary and the generation of wall shear stress

3.1 Introduction

Gas bubbles in liquids when driven by acoustic waves can create large forces on submerged structures. One important application of gas bubble pulsations is the cleaning of surfaces in ultrasonic cleaning bathes. At sufficiently small driving pressure, gas bubbles respond as damped harmonic oscillators with a characteristic resonance frequency. A boundary near to the bubble where the cleaning occurs affects the flow and reduces the resonance frequency.

Generally, the removal of particulate contaminants from surfaces is caused by a high magnitude of the wall shear stress generated from oscillating bubbles. The physical picture is that the inviscid flow of the bubble drives the boundary layer.

Figure 3.1: Detachment of particles on a flat silicone wafer after 3 hours of contact due to bubble oscillations. The particles are attracted toward the bubble. Figure is taken from [80].

In this boundary layer the velocity of the wall tangent flow reduces from the far field flow to zero on the boundary. This velocity gradient causes a twisting force to break adhesive bonds and a dragging of adherent particles which very effectively cleans surfaces [74]. Another interesting mechanism is the bubble while oscillating may also generate low pressure region which attract the particle towards it [80], see Fig.3.1 for the experimental evidence.

Bubble dynamics can be grossly separated into dynamics with small amplitude oscillations and more violent oscillations. The latter one leads to surface oscillation, jetting, and more complex phenomena classified under the term cavitation. Here, we'll focus only on mildly oscillating bubbles retaining their spherical shape. This simplified assumption allows to include dissipative mechanisms from the bulk viscosity, acoustic radiation, and thermal gas dynamics to determine the amplitude response of the bubble in presence of a wall. Therefore, we re-derive the Rayleigh-Plesset [17, 2, 61] equations now for a bubble oscillating near a boundary. The boundary layer flow is approximated using the Duhamel solution for the boundary layer. We present an analytical formula for the wall shear stress and analyze the steady and unsteady part of the wall shear stress as a function of the frequency,

Figure 3.2: Sketch of the geometry. The distance of the bubble center to the rigid boundary is $r' = r = \sqrt{d^2 + x^2}$, where d is the distance of the bubble center and the wall. The bubble's image center is located $D = 2d$ from its center.

distance to the boundary, and liquid parameters.

3.2 Theory

3.2.1 Bubble model

An irrotational flow field created by a bubble oscillating near wall may be constructed using the image theory [45]. In this model, the effect of wall is represented by interaction of the bubble with its own image at the same distance from the wall.

A sketch of the geometry is depicted in Fig. 3.2.

Assuming that the pressure inside the bubble is uniform and neglecting vapour the thermodynamics of the gas can be written as [63, 64]

$$\Phi = \frac{3\gamma}{1 - 3(\gamma - 1)i\chi \left[\sqrt{\frac{i}{\chi}} \coth \left(\sqrt{\frac{i}{\chi}} \right) - 1 \right]} \quad (3.1a)$$

and

$$\chi = \frac{D_{th}}{\omega R_0^2} \quad , \quad (3.1b)$$

where γ is the ratio of specific heats, D_{th} is gas' thermal diffusivity, ω is the angular driving frequency, and R_0 is the equilibrium bubble radius.

We use as a base equation for the bubble dynamics the Keller-Miksis equation [20, 75, 21] with a series expansion of the pressure driving term [73]. Additionally, we take the thermal dynamics into account, see Eq. (3.1). The equations read

$$\begin{aligned} & \left(1 - \frac{\dot{R}}{c}\right) R \ddot{R} + \left(\frac{3}{2} - \frac{\dot{R}}{2c}\right) \dot{R}^2 = \\ & \frac{1}{\rho} \left(1 + \frac{\dot{R}}{c}\right) [p(R, t) - p_s] + \frac{R}{\rho c} \frac{d}{dt} [p(R, t) - p_s] \quad , \quad (3.2a) \\ & -\frac{1}{D} (2\dot{R}^2 R + R^2 \ddot{R}) \end{aligned}$$

$$p(R, t) = p_0(1 - \Phi X) - \frac{2\sigma}{R} - \frac{4\mu}{R} \dot{R} \quad , \quad (3.2b)$$

$$p_0 = p_\infty + \frac{2\sigma}{R_0} \quad , \quad \text{and} \quad (3.2c)$$

$$p_s = p_\infty (1 - \epsilon e^{i\omega t}) \quad . \quad (3.2d)$$

Here, $R(t)$ is the instantaneous bubble radius, c is the speed of sound in the liquid, p is the pressure in the liquid at bubble wall, ρ is liquid density, p_s is the time varying total pressure far from the bubble, D is the distance between both bubbles, p_0 is pressure inside the bubble in absence of acoustic driving, Φ is a complex variable representing the thermal gas behaviour, X is the amplitude of the bubble pulsation, σ is surface tension coefficient, μ is dynamic viscosity of the

liquid, p_∞ is a constant pressure at infinity, and ϵ is the amplitude of the acoustic driving pressure.

A straightforward linearization of Eq. 3.2 around the equilibrium bubble radius, R_0 , yields

$$\ddot{X} + 2\beta\dot{X} + \omega_0^2 X = \frac{\epsilon P_\infty}{\rho R_0^2 \left(1 + \frac{R_0}{D}\right)} e^{i\omega t} \quad , \quad (3.3a)$$

$$2\beta = \frac{\omega^2 R_0}{c} + \frac{p_0 \Im(\Phi)}{\omega \rho R_0^2 \left(1 + \frac{R_0}{D}\right)} + \frac{4\mu}{\rho R_0^2 \left(1 + \frac{R_0}{D}\right)} \quad , \quad (3.3b)$$

$$\omega_0^2 = \frac{p_0}{\rho R_0^2 \left(1 + \frac{R_0}{D}\right)} \left[\Re(\Phi) - \frac{2\sigma}{p_0 R_0} \right] \quad . \quad (3.3c)$$

Note that even if Eq. 3.3a is identical to that of the familiar equation of harmonic oscillator, β and ω_0 are not constants but dependent on ω . The true resonance frequency is retrieved if $\omega_0 = \omega$ [75]. This distinguishes above equations from a formal ordinary damped harmonic oscillator, see [64].

The effective damping constant, β , is composed of an acoustic, a thermal, and a viscous contributions [75, 2]:

$$\beta = \beta_{ac} + \beta_{th} + \beta_{vis} \quad . \quad (3.4)$$

The individual damping coefficients are

$$\beta_{ac} = \frac{\omega^2 R_0}{2c} \quad , \quad (3.5)$$

$$\beta_{th} = \frac{p_0 \Im(\Phi)}{2\omega \rho R_0^2 \left(1 + \frac{R_0}{D}\right)} \quad , \text{ and} \quad (3.6)$$

$$\beta_{vis} = \frac{2\mu}{\rho R_0^2 \left(1 + \frac{R_0}{D}\right)} \quad . \quad (3.7)$$

In the limit of $D \rightarrow \infty$, Eq. 3.2 reduces to the famous equation of single oscillating bubble in a compressible liquid, see [20, 21]. Damping constants and ω_0 retrieved also reduce to their single bubble case [65].

By substituting an ansatz in the form of $X = X_m e^{i\phi} e^{i\omega t}$ as a solution for Eq. 3.3a, one gets the expression for the maximum radial amplitude perturbation

$$X_m = \frac{\epsilon p_\infty}{\omega_0^2 \rho R_0^2 \left(1 + \frac{R_0}{D}\right)} \left[\left(1 - \frac{\omega^2}{\omega_0^2}\right)^2 + \frac{4\omega^2 \beta^2}{\omega_0^4} \right]^{-\frac{1}{2}}. \quad (3.8)$$

3.2.2 Effect of the wall

We first present results on the effect of the wall on the resonance frequency and the damping coefficients in Fig. 3.3. Note, all calculations if not stated otherwise model an air bubble in water at room temperature with the following constants: $\gamma = 1.4$, $\rho = 998 \text{ kg/m}^3$, $\sigma = 0.0725 \text{ N/m}$, $p_\infty = 10^5 \text{ Pa}$, $D_{th} = 2.216 \times 10^{-5} \text{ m}^2/\text{s}$, $\mu = 10^{-3} \text{ kg/(m s)}$, and $c = 1500 \text{ m/s}$.

We find that the wall reduces the resonance frequency noticeably, for the here considered case of $d = 1.5R_0$ it is a reduction of 13% independent of the bubble radius. The wall significantly decrease the damping coefficient but only when the bubble is driven below or around the resonance frequency. The reduction for $d = 1.5R_0$ amounts to 25% independent of the bubble radius.

3.2.3 Wall shear stress formulation

The flow close to the boundary, i.e. the viscous boundary layer, is modeled following the work of Rau *et al.* [76] and Vos *et al.* [47]. The assumption is that the flow field can be separated into a potential flow around the bubble and a viscous boundary layer flow next to the surface. The linearity of the governing equation, i.e. the diffusion equation which is used in Stokes 1st problem, allows to derive an integral formulation commonly termed the Duhamel's integral. This approach is basically constructing a solution of a boundary value problem, with space- and

Figure 3.3: Resonance plots for 5 different equilibrium bubble radii, R_0 , of 0.1, 1, 10, 100, and 1000 μm from top to bottom row with and without a wall being at a distance $d = 1.5R_0$. The first column depicts the resonance parameter ω_0 and the true resonance frequency indicated with the circle. The second column plots the total damping coefficient β while the last column splits it into the contribution of acoustic, viscous, and thermal damping.

time-varying boundary condition, to a step function.

The acoustic boundary layer thickness may be estimated as [77]:

$$\delta = \sqrt{\frac{2\nu}{\omega}} \quad . \quad (3.9)$$

Hence for driving frequency range of 10kHz to 100MHz, $\delta \approx 5.64\mu\text{m}$ to 56.4nm.

The bubble oscillation start at time $t = 0$, thus the wall shear stress builds up from 0. In Appendix A, based on approach similar to reach the solution of Stokes 1st Problem but changing the boundary condition so that the fluid velocity is zero at the wall and equal to the bubble wall velocity at infinity, we derive the expression for the wall shear stress:

$$\tau_w = \rho \sqrt{\frac{\nu}{\pi}} \frac{x}{(d^2 + x^2)^{\frac{3}{2}}} \times \left[2 \int_0^t \frac{\ddot{R}R^2}{\sqrt{t-t'}} dt' + 4 \int_0^t \frac{\dot{R}^2 R}{\sqrt{t-t'}} dt' \right] \quad (3.10)$$

An analytical solution to Eq. (3.10) in the limit of a harmonic oscillating bubble can be derived and is stated in the Appendix B. The analytical solution has been validated with numerical solutions of the integral, see Appendix C.

3.3 Steady wall shear stress

The history term in the kernel of Eq. (3.10) leads to a transient behaviour of the wall shear stress when starting from time $t = 0$. This is insofar important as this transient solution depends on the phase, δ , i.e.

$$R = R_0 [1 + X_m \sin(\omega t + \delta)] \quad . \quad (3.11)$$

Figure 3.4: The wall shear stress as a function of time. After the initial transient dies out it is composed of a constant wall shear stress, τ_w^{avg} (red line), and a sinusoidal oscillating component τ_w^{osc} represented by its maximum and minimum values (blue lines).

For the following parameter studies we neglect the initial transients, see Fig. 3.4, and evaluate the shear stresses only after sufficient number of oscillations, here we use at least 10^{10} . However, 10^6 oscillations are indeed already sufficient. This does not add computational costs as we do have an analytical solution for the wall shear stress. We see that the wall shear stress is composed of a steady shear stress and an oscillating shear stress. In all cases presented the steady wall shear stress is about an order of magnitude smaller than the oscillating wall shear stress. We present the oscillating wall shear stress with two values, the minimum and maximum value at steady state.

The averaged wall shear, τ_w^{avg} is calculated using

$$\tau_w^{avg} = \frac{1}{2} \left[\max \left(\lim_{t \rightarrow \infty} \tau_w(t) \right) + \min \left(\lim_{t \rightarrow \infty} \tau_w(t) \right) \right] \quad . \quad (3.12)$$

The oscillating wall shear stress, τ_w^{osc} is determined from the envelope of the wall shear stress. Figure 3.4 depicts the transient behavior and the approximately constant averaged wall shear stress. Please note that in Fig. 3.4 only the first few oscillations are plotted. Here, the wall shear stress has not reached a steady state.

All later results presented are approximately the steady state. It is important to note that the steady wall shear stress, i.e. for $t \rightarrow \infty$ does not depend on the initial conditions, while the transient values may differ largely. This *physically correct* long time behavior has been checked for oscillations starting at different phases, see Appendix B for details.

3.4 Parameter study of the wall shear stress

We now conduct a parameter study and start with a strength of the wall shear stress plotted as a function of the distance from the stagnation point ($x = 0$) while the distance, d is varied from $2.5R_0$ to $1.5R_0$, Fig. 3.5. For this and the later plots, the left column shows the average wall shear stress and the plot on the right shows the oscillating wall shear stresses. The bubble in Fig. 3.5 has an equilibrium radius $R_0 = 10 \mu\text{m}$ and is driven at a frequency of 298.12 kHz which is the resonance frequency for $d \rightarrow \infty$.

As shown in Fig. 3.5, by symmetry, at $x = 0$ the wall shear stress is zero. It will also approach zero in the limit of $x \rightarrow \infty$, too. The wall shear stress has a maximum magnitude of 13.5 Pa of average wall shear stress at approximately $x = 2.1R_0$ for $d = 1.5R_0$, but for larger d/R_0 the maximum position is shifted to a larger value of x/R_0 . For $d = 2R_0$ the maximum magnitude of 11.8 Pa of average wall shear stress is achieved at approximately $x = 2.8R_0$, while for $d = 2.5R_0$ average wall shear stress magnitude of 10.2 Pa is achieved at approximately $x = 3.5R_0$. The maximum and minimum oscillating wall shear stress values at the same x as above respectively are 163.8 Pa and -190.8 Pa, 115.1 Pa and -138.8 Pa, 85.9 Pa and -106.3 Pa.

Figure 3.5: Plot of average (left column) and oscillating (right column) wall shear stress as a function of distance from stagnation point (x) for some bubble center distances from wall (d). R_0 is set as $10\mu\text{m}$. The driving frequency is 298.12kHz . Dimensionless pressure amplitude perturbation, ϵ , is 0.1 .

In Fig. 3.6, the wall shear stress is plotted as a function of driving frequency for different bubble distances from wall, $d = 1.5, 2, 2.5R_0$. The equilibrium bubble radius, R_0 , is also set to $10\mu\text{m}$, while x is set to the same value of R_0 . As expected, the wall shear stress has a maximum near the resonance frequency of the bubble, approximately at 257.9 kHz , 266.9 kHz and 272.5 kHz . At those respective frequencies, the average wall shear stress magnitudes are 48.8 Pa , 22.3 Pa and 11.9 Pa . Meanwhile, their respective maximum and minimum values of oscillating wall shear stress are 262.9 Pa and -360.6 Pa , 121.9 Pa and -166.6 Pa , 65.6 Pa and -89.4 Pa .

In Fig. 3.7, the wall shear stress is plotted as a function of equilibrium radius for a few driving frequencies, ranging between 100kHz and 100MHz . The bubble is located $1.5R_0$ away from the wall. To maximize the wall shear stress, using the result in Fig. 3.5, x is set as $2.1R_0$. For each driving frequency, the maximum wall shear stress is found when the bubble radius is near resonant condition. For 100

Figure 3.6: Plot of average (left column) and oscillating (right column) wall shear stress as a function of driving frequency. R_0 is set as $10\mu\text{m}$ and x is set as equal to R_0 . Dimensionless pressure amplitude perturbation, ϵ , is 0.1.

MHz the maximum wall shear stress is achieved at bubble equilibrium radius of approximately $0.09\mu\text{m}$, for 10 MHz is $0.43\mu\text{m}$, for 3 MHz is $1.09\mu\text{m}$, for 1 MHz is $2.77\mu\text{m}$, for 500 kHz is $5.23\mu\text{m}$, and for 100 kHz is $26.27\mu\text{m}$. The average wall shear stress magnitude at those equilibrium radii respectively are 1.6 Pa, 22.9 Pa, 77.3 Pa, 130.3 Pa, 107.9 Pa and 41.2 Pa. Meanwhile, the maximum and minimum values of oscillating wall shear stress for the respective equilibrium bubble radii are 433.2 Pa and -436.4 Pa, 631.4 Pa and -677.2 Pa, 734.2 Pa and -888 Pa, 720.7 Pa and -981 Pa, 554.7 Pa and -770.5 Pa, 223.1 Pa and -305.5 Pa.

In Fig. 3.8, the wall shear stress is plotted as a function of the driving frequency, but the bubble equilibrium radius is always set so that the bubble oscillates in resonance. In other words, the bubble equilibrium radius for each driving frequency is calculated as the R_0 which satisfy Eq. 3.3c when $\omega = \omega_0$. The pressure perturbation, ϵ , is also varied ($\epsilon = 0.1, 0.07, 0.05$). The maximum magnitude of the average wall shear stress is achieved at approximately 959.36 kHz, which results in a magnitude of 131.2 Pa for $\epsilon = 0.1$. For $\epsilon = 0.07$ is 70.6 Pa at 916.14 kHz. For

Figure 3.7: Plot of average (left column) and oscillating (right column) wall shear stress as a function of bubble equilibrium radius for some driving frequencies ($\omega/2\pi$). Bubble distance to wall, d , is set as $1.5R_0$. Distance from stagnation point, x , is set as $2.1R_0$. Dimensionless pressure amplitude perturbation, ϵ , is 0.1.

$\epsilon = 0.05$ is 38 Pa at 891.15 kHz.

Meanwhile, for $\epsilon = 0.1$, the maximum value of the oscillating wall shear stress is 755.1 Pa at 1.7149 MHz and the minimum value is -999.3 Pa at 1.2650 MHz. For $\epsilon = 0.07$ the values are 532.3 Pa at 1.9155 MHz and -654.6 Pa at 1.3872 MHz. For $\epsilon = 0.05$ the values are 385.7 Pa at 1.9511 MHz and -448.5 Pa at 1.4797 MHz.

Next, we study the change of the wall shear stress as a function of dynamic viscosity, ranging between 10^{-4} and 1 Pa.s. The wall shear stresses for three driving frequencies are plotted, while the corresponding R_0 are set so that the bubbles are in resonance. Results are depicted in Fig. 3.9. Calculated τ_w^{avg} have maximum values of 144.08 Pa at $\mu = 0.0439$ Pa.s, 87.38 Pa at $\mu = 0.0151$ Pa.s and 168.29 Pa at $\mu = 3.08 \times 10^{-4}$ Pa.s for driving frequencies of 10 kHz, 40 kHz and 1.3 MHz, respectively.

The oscillating wall shear stresses also have peak (maximum and minimum) values. For driving frequency of 10 kHz, they are 703.80 Pa at 0.0757 Pa.s and

Figure 3.8: Plot of average (left column) and oscillating (right column) wall shear stress as a function of driving frequency ($\omega/2\pi$) for some pressure perturbations (ϵ). R_0 are set so that the bubbles are in resonance. Bubble distance to wall, d , is set as $1.5R_0$. Distance from stagnation point, x , is set as $2.1R_0$.

−982.56 Pa at 0.0595 Pa.s. For 40 kHz, they are 547.94 Pa at 0.0375 Pa.s and −704.63 Pa at 0.0262 Pa.s. For 1.3 MHz, they are 786.95 Pa at 4.79×10^{-4} Pa.s and −1116.6 Pa at 3.95×10^{-4} Pa.s.

To further investigate the viscosity effect, calculations of the optimum viscosity (μ_{opt}) needed to generate maximum average wall shear stress (τ_w^{avg}) are done. For each frequency driving, a resonant R_0 is assigned. Dimensionless pressure amplitude perturbation, ϵ , is fixed to 0.1. The corresponding plots, including the magnitude of τ_w^{avg} are depicted in Fig. 3.10.

Interestingly, in Fig. 3.10 τ_w^{avg} shows a maximum and a minimum. It is minimum at driving frequency of approximately 142 kHz with corresponding μ_{opt} of approximately 4.98×10^{-3} Pa.s and $|\tau_w^{avg}|$ of approximately 65.12 Pa. Meanwhile, it is minimum at driving frequency of approximately 3.5 MHz with corresponding μ_{opt} of approximately 11.45×10^{-5} Pa.s and $|\tau_w^{avg}|$ approximately 200.5 Pa.

Figure 3.9: Plots of steady (left) and oscillating (right) wall shear stress as a function of dynamic viscosity (μ) for some driving frequencies ($\omega/2\pi$). Dimensionless pressure amplitude perturbation, ϵ , is 0.1. R_0 is set as the resonant radius for each driving frequency. Bubble distance to wall, d , is set as $1.5R_0$. Distance from stagnation point, x , is set as $2.1R_0$.

Figure 3.10: Plots of dynamic viscosities needed to get maximum steady wall shear stress, i.e. the optimum viscosity, (left) and their corresponding steady wall shear stresses (right) as a function of driving frequency. R_0 is set as the resonant bubble equilibrium radius for each driving frequency. Dimensionless pressure amplitude perturbation, ϵ , is 0.1. R_0 is set as the resonant radius for each driving frequency. Bubble distance to wall, d , is set as $1.5R_0$. Distance from stagnation point, x , is set as $2.1R_0$.

3.5 Discussions

First we discuss the bubble behaviour near a solid wall. The wall effect is much more pronounced on β than ω_0 . This is similar to the theoretical result reported by Keller [78] for bubbles which undergo damped radial oscillations. Keller's result is later generalized to any damped harmonic oscillator by Lippmann [79]. Since ω_0 is an intrinsic property of the oscillator (gas bubble), it is not much affected by perturbation in the liquid. Meanwhile, β is by contrast more affected by perturbation in the liquid, say by introducing an object which can exchange energy with the oscillator (gas bubble) in the liquid. Eventhough in our case the bubble is subjected to acoustic forcing and it is not an ordinary oscillator since β and ω_0 is dependent on ω , we propose that the same line of physical reasoning is also applicable in the linear regime.

Next, we discuss the wall shear stress simulation for potential cleaning application. Suppose an initially far away oscillating gas bubble, driven on its free bubble resonance frequency, is approaching the wall. If the wall shear stress due to bubble translation is neglected, the wall shear stresses can be calculated as depicted in Fig. 3.5.

One may notice from Fig. 3.5-3.9 that negative (oscillating) wall shear stress are higher than the positive one, which means that liquid near the wall is more being pulled toward the stagnation point than pushed by the bubble oscillations. By observing Figs. 3.8 and 3.9, one may also notice that the oscillating wall shear stress peaks are less symmetrical than in Figs. 3.5-3.7. In other words, the maximum and minimum wall shear stresses do not lie at the same position of x-axis. This difference is probably due to the setting of the resonant bubble in Fig. 9-10, hence

each ω will have its own corresponding R_0 .

Fig. 3.6 shows that wall shear stress is maximum at resonance, and in Fig. 3.7 the highest wall shear stress is achieved for driving frequency in the order of 10^6 Hz. This is the typical frequency used in megasonic cleaning application, which may explain the success of this cleaning equipment in the semiconductor industry. By setting the resonance condition for all driving frequencies, the maximum wall shear stress is achieved at 1.3 MHz, as depicted in Fig. 3.8.

In megasonic cleaning, since van der Waals force is the main contributor to the adhesion force of micron- and submicron-sized particles to a solid surface [77], order of magnitude estimation of adhesion force of a spherical polymer particle on a silicon surface can be done. Following the analysis of Kim *et al.* [80], the adhesion force is

$$F_a = \frac{1}{6} \frac{AR}{Z_0^2} \left(1 + \frac{a^2}{RZ_0} \right) \quad , \quad (3.13)$$

where A is the Hamaker constant ($A = 2.08 \times 10^{-20}$ J for PSL particle on Si surface immersed in water), R the particle radius, a the particle-substrate contact radius and Z_0 the distance between the particle and the flat surface normally set to 0.4 nm [77].

The cleaning mechanism is assumed mainly due to rolling [77, 80], hence the adhesion torque is the torque needed to overcome the adhesion force and may be estimated as

$$T_a \sim aF_a \quad (3.14)$$

which is at least in the order of $T_a \sim 10^{-4}$ pN.m for 0.2 μ m radius particle. Meanwhile, the torque generated by the wall shear stress may be estimated as

$$T_w \sim |\tau_w^{osc}| A_p R \quad (3.15)$$

Figure 3.11: The nonlinear bubble oscillation (left) and the wall shear stress generated (right). Note that the sinusoidal driving pressure amplitude is 900 kPa with frequency of 1 MHz and the bubble distance to the wall is 12 μm .

where the projection area $A_p \sim \pi R^2$. Since $|\tau_w^{osc}| \sim 10^3$ Pa, the estimation for the torque is $T_w \sim 10^{-6}$ pN.m, which is two order of magnitude lower than T_a . Hence, for megasonic cleaning application, it is necessary to drive the bubble above its linear regime for successful cleaning.

To show the feasibility of my point, a wall shear stress simulation based on the full Keller-Miksis equation for two interacting bubbles is done. The numerical scheme used, due to the absence of any analytical solution, is explained in Appendix C. The simulation result is shown in Fig. 3.11. Note that the parameters are not optimized for the whole parameter space due to the expensive computational cost. Furthermore, the wall shear stress do not have chance to build up due to very short period. Nevertheless, the maximum wall shear stress generated is approximately 60 kPa, which is almost two order of magnitude higher than the maximum wall shear stress generated by linear oscillation of the bubble previously shown. Vos *et al.* [47] used similar numerical scheme for their wall shear stress calculation, but using experimental data for the nonlinear bubble radius oscillation and found higher wall

shear stress, exceeding 300 kPa. Moreover, nonlinear and violent bubble oscillations are usually related to phenomena such as jettings, bubble shape oscillations and bubble translations along the surface which has been experimentally shown to help in cleaning [50, 80].

Next, we discuss the effect of viscosity. By varying dynamic viscosity, as depicted in Fig. 3.9, we found the maximum of the wall shear stresses. This is due to the two competing effects of viscosity: it adds the damping in bubble oscillation but it also increase the wall shear stress. Note that only viscosity is changed, while all other physical parameters are still kept as water at room temperature. One may notice from Fig. 3.9 that the optimum viscosity for $f = 1.3$ MHz is pretty close to the actual viscosity for water at room temperature.

Setting the optimum viscosity, as depicted in Fig. 3.10, leads to an interesting wall shear stress for resonant bubble case. As the viscosity is optimized, the highest wall shear stress is in the driving frequency of order 10^6 Hz. However, continuing to increase the frequency will make the wall shear stress start to decrease due to increasing acoustic damping. This suggest that changing the viscosity, by heating up or changing the liquid, may help in cleaning. Hence, heating up the liquid may not only help to clean by increasing reaction rate (e.g for wetting with water) and dissolving ability of the water, but also to achieve a viscosity better suited for generating higher wall shear stress on the surface.

3.6 Conclusions

Linear theory predicts that the wall shear stress due to bubble oscillations near a rigid wall in an incompressible liquid will reach steady-state after sufficiently

long period of insonation. This is relevant to ultra/megasonic cleaning applications because usually the insonation takes few minutes, very long compared to one period of bubble oscillation under frequency driving of the order 10^6Hz .

The magnitude of the oscillating and the steady wall shear stress may differ by two orders of magnitude. The much higher oscillating wall shear stress is contributing to particulates/contaminants detachment, due to inertia of the particulate, while the steady wall shear stress may contribute mainly to keep the detached particulates away from the surface [80].

Despite the strict limitation of small amplitude oscillation, the bubble is capable of generating wall shear stress in the order of 10^3 Pa . This may be sufficient for less demanding cleaning application, e.g. water filtration membrane cleaning in low pressure system. By contrast, it is necessary to drive the bubble beyond its linear limit in megasonic cleaning to ensure successful cleaning.

Chapter 4

Surface oscillations and jetting from surface attached acoustic driven bubbles *

4.1 Introduction

A stable bubble attached to a wall and exposed to a time varying pressure field not only pulsates radially but also translates perpendicular to the wall. The oscillatory translational motion can be explained through the interaction of the bubble with its mirror image. The radial pulsation and the translational motion may lead to the excitation of non-spherical shapes of the bubble. Already in 1944 Kornfeld and Suvorov [81] reported on surface attached bubbles exposed to a sound field that are exhibiting polygonal shapes. They noted that the number of cusps increased with increasing size of the bubble for constant driving amplitude and

*This chapter has been published as [51]

frequency. Although surface modes radiate less sound as compared to the monopole radiation from the radial pulsation, they may have an important contribution for the enhancement of mass transfer [82]. This mass transfer may be even further enhanced when the surface oscillation develops into a fast focused flow which has been observed by Crum [83]. He investigated large, millimeter sized bubbles close to a periodically oscillating boundary and found a jet flow towards the boundary. These jets resembled the jetting of transient cavitation bubbles which have been considered responsible for the erosion of surfaces [24]. The jets reported in Crum's experiments, in contrast, develop over many oscillations. The bubble oscillates periodically with the jet accelerating during the shrinkage and decelerating during the expansion of the bubble. So far only a few simulations [68] studied the formation of this cyclic jet reported by Crum [83].

Recently, research on bubble oscillations close to surfaces has gained interest due to its importance for ultrasonic cleaning and in medical application. In particular Marmottant *et al.* [84] studied micrometer-sized bubbles attached to a surface. They observed a superposition of a stable radial pulsation and a translation. The phase shift between the radial and the translational motion leads to a large scale streaming pattern outside the acoustic boundary layer of the bubble (Marmottant & Hilgenfeldt [6]).

Far from boundaries, the coupling of the volume oscillation with surface modes of bubbles is a century old and intensively studied fluid mechanics problem. A first model was developed by Lord Rayleigh in 1879 [85] and later refined by Plesset [86] and many others. Longuet-Higgins [87] [88] and Mei & Zhou [89] predicted that chaotic dynamics of the surface modes may evolve once the coupling between

the surface modes is taken into account.

In many applications such as ultrasound cleaning and surface processing, acoustically driven bubbles are not only found in the bulk but also attached to rigid surfaces where they grow from gaseous stabilized nuclei. In the present work we study the onset and development of surface modes of gas bubbles attached to surfaces and exposed to an increasing driving pressure. We focus on bubbles relevant to ultrasonic cleaning equipment; and we decided to study bubbles resonant in the 20kHz frequency regime due to experimental constraints. We want to visualize the bubble interior thus we have to study relatively large bubbles, yet the driving frequency range is limited by the specification of the high-speed camera, too. We were particularly interested if the jet observed earlier by Crum [83] also develops at frequencies relevant for ultrasonic applications.

4.2 Experimental setup

Figure 4.2 presents a picture of an adherent bubble and the corresponding sketch of its geometry. The bubble is attached due to buoyancy to a glass surface and forms a nearly spherical shape with a contact angle, θ_c , of 60° . The bubble's center vertical coordinate is defined as the center of a circle fitted to the bubble's contour at rest; the circle radius is denoted as R_0 , see Fig. 4.2. Yet, the horizontal coordinate of the center may change with time as we identify its position through the center of mass of the bubble. This position can vary slightly once surface oscillation set in. The angle θ is measured clockwise and is zero along the horizontal x-axis passing through the bubble center. At rest, the radius of the bubble $r(\theta)$ in Fig. 4.2 is approximately constant for the gas-liquid interface, but varies at the gas-glass

interface. Thus, to define a unique quantity to measure the radial response we define the averaged radius of this bubble as $r_{\text{avg}} = (2\pi)^{-1} \int r(\theta) d\theta$, which is here $142 \mu\text{m}$. This bubble is inside a small container as shown in the sketch of the experimental setup, Fig. 4.3. The bubble is introduced with a microliter syringe into the water filled container of inner dimensions $12.1 \times 12.1 \times 44.5 \text{ mm}^3$ (Polystyrene cuvette, CXA-105-010Q, Einst Technology, Singapore). A disk-shaped piezoelectric transducer (SMD25T21F1000R, 2.1 mm thickness, 25 mm diameter, Steiner & Martins, USA) is glued onto the cuvette's outer wall. It is driven with a sinusoidal signal from an arbitrary function generator (33220A, Agilent) and amplified with an audio amplifier (XM-2002GTR, Sony) which is fed into a voltage transformer (XC462, Amplimo, The Netherlands). The voltage amplitude of the piezoelectric transducer reaches up to 200 V. These high voltages deemed necessary because the cuvette does not show acoustic resonances in the current frequency regime. To study the effect of the driving amplitude on the bubble oscillation we take recordings of the bubble while the amplitude is increased linearly from 0 to 100% of its set value, which corresponds to values of 0 to 200 V voltage amplitudes and 0 to 0.085 bar pressure amplitudes. This linear pressure increase is done on a rather long time interval of $\Delta t = 0.5 \text{ s}$.

The dynamics of the bubbles is recorded with a high-speed camera (Photron SA1.1, Japan) at frame rates of 180,000 frames per second. We recorded about 11 frames per period and around 8,100 periods per Δt . This allowed us to record twice the same slowly increasing pressure amplitude using the 4 GB frame memory of the camera.

To relate the position in time of the linearly increasing driving signal and thus

Figure 4.1: Plot of the measured pressure amplitude as a function of time. The data are approximated by a straight line with a slope of 0.17 bar/s.

the frame number of the high-speed recordings with the acoustic pressure amplitude we used a thin needle-type hydrophone (1 mm diameter, HP Series, Precision Acoustics Ltd, U.K.). It is equipped with a pre-amplifier and a booster amplifier and inserted into the cuvette after the recording of the bubble oscillation. Simultaneous recording were not possible due to the size constraints of the cuvette. Because one of the small faces of the cuvette is removed and the sensor is inserted during the acoustic measurements the values may differ. Still, we find that the acoustic pressure increases linearly with the driving voltage amplitude. The plot of pressure amplitude as a function of time is shown in Fig. 4.1 which may be well approximated by a straight line with a slope of 0.17 bar/s. This allows us to relate the time of the high-speed recording with a pressure value:

$$p(t) = \frac{\Delta p}{\Delta t} t \sin 2\pi f t \quad , \quad (4.1)$$

where $\Delta p/\Delta t$ is the pressure slope which is 0.17 bar/ Δt and $f = 16.27$ kHz is the driving frequency.

Figure 4.2: Picture of the attached bubble. Left: one frame taken from the high speed recording of an undisturbed bubble attached to the wall. Right: Sketch of the geometry for the analysis of the bubble shape. $r(\theta)$ is the bubble radius as a function of angle, R_0 is the radius of a circle fitted to undisturbed bubble image, and θ_c is the bubble contact angle with the surface.

A careful illumination is needed to visualize the interior of the bubble during the oscillation. Therefore, two positive lenses (with 40 and 45 mm focal distance) collimate and image the light from a high-power metal halide lamp (ILP-2, Olympus). A diffuser attached to the cuvette's wall diffuses the intense light sufficiently to visualize the interior of the bubble.

4.3 Experimental results

From the recorded movies, the contour of the bubble in each frame is extracted and analyzed (see below). We first present a few examples of the bubble shape evolution in Fig. 4.4 for increasing driving pressures. Therefore, in each frame, A to D in Fig. 4.4, the contour at minimum and maximum bubble radius is plotted onto the image of the bubble. This image of the bubble shown is recorded between the minimum and maximum size. Figure 4.4 demonstrates the morphological changes of the bubble shape oscillation: for low driving amplitudes the bubble oscillates

Figure 4.3: Sketch of the experimental setup: The bubble located inside the acoustic chamber is attached to a glass plate. It is driven into radial and surface modes with a function generator and a power amplifier connected to a disc shaped piezoelectric transducer. The scene is illuminated with strong continuous illumination and observed with a long-distance microscope attached to a high-speed camera.

radially (A), then slight non-radial modes set in (B) which develop into strong surface oscillations (C). At a later stage circumferential waves appear, and the bubble loses its axisymmetric shape (D). Possibly some asymmetries in the acoustic driving are responsible for the specific shape bifurcation observed. High-speed recordings of this last stage are discussed below.

The contours shown in Fig. 4.4 are obtained using standard image processing routines from MATLAB (The Mathworks, Natick, MA). From the contour, the radius as a function of the angle, $r(\theta)$, is obtained, see Fig. 4.2. For the later analysis the oscillation of the bubble is characterized with the envelope of the radial amplitude, $r^* = r_{avg}(t) - r_{avg}(t = 0)$, and angle dependent amplitude of the radius $\Delta r(\theta, t) = r(\theta, t) - r_{avg}(t)$. To study Δr , we follow a previous approach from Dollet *et al.* [90] using Fourier mode decomposition:

$$a(n) + ib(n) = \frac{1}{2\pi} \int_0^{2\pi} \Delta r(\theta) e^{in\theta} d\theta \quad , \quad (4.2)$$

where n is an integer mode number. We can now analyze the temporal development of the mode amplitude $c_n = \sqrt{a_n^2 + b_n^2}$. By Fourier transformation from 0 to 2π , the mode numbers are identifiable with simple shapes, e.g. $n = 1$ represents spherical cap shapes, $n = 2$ prolate and oblate shapes, $n = 3$ triangular shapes, $n = 4$ squarish shapes, and $n = 5$ pentagonal shapes.

In Fig. 4.5 we present the envelope of the radial amplitude, r^* , and the envelope of Fourier mode amplitudes, $c_n^* = c_n(t) - c_n(t = 0)$, as a function of the pressure for two consecutively increasing amplitudes. Initially, r^* and the first mode of c_n^* increases linearly up to $p = 0.0378$ bar. This first mode is caused by the translatory motion perpendicular to the boundary. Suddenly, from $p = 0.0378$ bar all modes

Figure 4.4: The evolution of the bubble shape with increasing pressure driven at 16.27 kHz. White dashes represent minimum contours, black dashes represent maximum contour, and the bubble image is taken in the intermediate. The positions of the recording of A to D are indicated in Fig. 4.5 .

show a rapid increase. Concurrently, we find in the recording the onset of surface oscillations, see Fig. 4.4 B. Interestingly, the onset of surface oscillations is occurring together with a jump in the radial amplitude of approx. $1.4 \mu\text{m}$. The fourth mode remains approximately constant while the third mode shows the largest gain and reaches at point C an amplitude of about $4 \mu\text{m}$, thus of the same magnitude as the radial mode. The prevalence of this "triangular"-mode is obvious from the shape shown in Fig. 4.4 C.

Similarly as abrupt as the onset of surface oscillation occurs, so does the onset of a noisy or chaotic regime at $p = 0.0587 \text{ bar}$. Although the radius for all modes, especially most obvious in mode 3, continue to increase linearly with the pressure amplitude strong fluctuations set in, which are even more pronounced in the amplitudes of the modes. In this chaotic regime, the bubble's position remains fixed. At even higher pressures which are not shown here, we observed that the bubble detaches and translates along the surface. The dynamics of the detachment is not studied here, yet may be of interest for cleaning applications.

The repeatability of the events is demonstrated in the second part of Fig. 4.5.

Note that the repeatability is considered quite good especially for the system investigated here, due to high nonlinearity inherent on it. Nonlinearity renders the bubble behaviour (i.e. radial amplitude and shape oscillations) sensitive to variations in initial conditions and bubble oscillations history, nevertheless we manage to consistently resolve the bubble's global behaviour. Immediately after the first slowly increasing driving pressure, an identical second ramp is applied to the same bubble. Here it takes about 13 ms for the acoustic field to decay inside the cuvette such that the bubble starts over with a small amplitude response. The second linearly increasing driving pressure reveals basically the same features as the first one with very similar magnitudes of the oscillation amplitudes. We find again 3 regimes, the linear increase of the radial amplitude, the sudden appearance of the surface mode, and the chaotic regime. Only modes 3 and 2 show some slight differences between the first and second pressure increase.

Next, we focused on the formation of large surface undulations which may evolve into a jet, e.g. we looked for similar shapes as found by Crum for his large bubble and low frequency driving regime. Indeed, we find a parametric instability which leads to the formation of a oscillating jet penetrating deep into the bubble, see Fig. 4.6. A thin liquid column oscillates upwards in frames a to e, and downwards in frames f to j of Fig. 4.6 during one period of the acoustic field. During the deceleration of the jet, a droplet is pinched-off from the tip of the jet (between frames f and g of Fig. 4.6) and propagates towards the rigid boundary, while the jet retreats back to the main bubble interface. Interestingly, a larger droplet is visible at the side where the pinched-off droplet would impact on the surface, see arrow in frame j of Fig. 4.6. This larger droplet is created due to the repetitive

Figure 4.5: Evolution of the envelope of the radial amplitude (r^*) and envelopes of the Fourier mode amplitudes (c_n^*) of the 1st to 5th mode from bottom to top. The bubble is excited twice with a linearly increasing pressure amplitude from 0 to $p = 0.085$ bar. The letters A to D relate to the four frames in Fig. 4.4

Figure 4.6: Details of the jetting and droplet pinch-off. The number in each frame denotes the time in microseconds. The contrast of the bubble interior is adjusted to aid the reader. It shows the formation of a jet and the subsequent pinch-off of a droplet which impact onto a larger droplet (see arrow in frame j). The length of the bar in the last frame is $100\ \mu\text{m}$.

impacts of pinched-off droplets. The smaller pinched-off droplet therefore does not hit the surface but its impact is cushioned by the larger droplet.

We measured the vertical speed of the jet and find a maximum at frame b of approx. $5.0\ \text{m/s}$. The pinched-off droplet impacts with a velocity of $2.5\ \text{m/s}$ onto the larger droplet.

4.4 Discussion and Conclusions

The attached bubble has the shape of a spherical cap for which the natural frequency of volume pulsations has been estimated by Maksimov [91]. Maksimov's potential flow model predicts for the contact angle of $\theta_c = 60^\circ$ a natural frequency of about 0.8 times the resonance frequency of a spherical bubble with the same volume. Neglecting surface tension and liquid viscosity we estimate the resonance frequency [2] of a spherical bubble to $f_r = R_0(3\kappa p_0/\rho)^{1/2}$, where κ is the effective poytropic index, $p_0 = 10^5\ \text{Pa}$ the atmospheric pressure, and $\rho = 1000\ \text{kg/m}^3$. We

estimate the effective polytropic index from Peclet number [2], $Pe \approx 90$, to be around $\kappa = 1.28$ for a diatomic gas, i.e. air. The estimated resonance frequency of the spherical cap bubble shown in Fig. 4.2 is 16.5 ± 0.4 kHz thus it is very close to the driving frequency in the experiment of 16.27 kHz.

With increasing the driving amplitude three distinctive regimes are observed:

- radial oscillation with a monotonous increase of the first and second mode (regime 1),
- sudden jump and excitation of all surface modes (regime 2), and
- chaotic surface oscillations (regime 3).

We can emphasis on the last point using the normalized standard deviation or deformation parameter as introduced by Dollet *et al.* [90]:

$$\alpha = \frac{1}{r_{avg}} \sqrt{\frac{1}{\pi} \int_0^\pi \delta r(\theta)^2 d\theta} \quad (4.3)$$

where $\delta r = r(\theta) - r_{avg}$. This deformation parameter, see Fig. 4.7, shows a strong variability when the bubble enters the chaotic regime.

We suggest that the chaotic oscillations are the result of circumferential traveling surface modes.

The jetting observed for large amplitude driving found in regimes 2 and 3, see Fig. 4.6, demonstrates the pinching-off of small droplets which accumulate into a big sessile droplet on the surface. This dynamics together with the rather low velocities of around 5 m/s in the chaotic regime and 3.4 m/s in regime 2 suggests that only a mild wall shear stress is created.

Figure 4.7: Deformation parameter (α) plot as a function of time. The chaotic oscillations shown in Fig. 4.5 occur once the deformation parameter shows large variability, i.e. at $p = 0.0587$ bar.

Philipp & Lauterborn [12] report for impact velocities of water jets below 25 m/s onto a aluminum surface with a corresponding water hammer pressure below 34 MPa no surface erosion. Thus, we expect that the weak jetting observed may not contribute to surface erosion and may not even remove strongly adherent contaminants. This jetting is in contrast to the strong jetting found for example by Ohl *et al.* [50] from cavitation bubbles. Crum [83] measured similar low velocities of the jet of around 5 m/s for bubbles ranging between 1 – 1.5 mm in radius. He also observed the pinch-off of small droplets from the jet. The similarity between the present sub-millimeter and Crum’s millimeter sized bubbles study provokes the question if this jetting at low velocities is found also for higher driving frequency, thus smaller bubbles relevant in medical and ultra clean processing applications. In a recent study of Zijlstra *et al.* [92] investigating at 1MHz – the so-called Mega-

sonic cleaning regime strong hints for cyclic jetting from an attached bubble has been gathered. The bubble driven at 1 MHz and recorded at framing rates at up to 13MHz shows non-spherical pulsations. Although their picture resolution does not allow for an unambiguous measurement of the jet velocity we may infer a velocity close to the radial velocity of the bubble which is of the order of $20 - 40$ m/s. This might indicate that larger bubbles driven in resonance develop jetting of moderate velocity whereas smaller bubbles may exert a higher jetting velocity on the surface. Although, the jetting [93] may be one of the contributors to mechanical effect, bubbles translating along the surface, thus bubble which are not attached, may create considerable wall shear stressed through the volume pulsations from the radial and surface mode [94] [95].

In summary we have studied the onset of surface modes of an adherent bubble driven in resonant pulsations at 16.27 kHz. The oscillation can be well characterized through Fourier modes of the bubble surface and show characteristic features. Interestingly, the bubble develops a parametrically excited jetting flow. The details of its dynamics suggests that this jetting flow is not contributing to mechanical effects on the surface, i.e. cleaning or erosion.

Chapter 5

Improved ultrasonic cleaning of membranes with tandem frequency excitation*

5.1 Introduction

Membrane separation process is a widely used technology in areas such as food and dairy products processing, and water purification. Membrane fouling, one of the major problems encountered in membrane filtration, occurs by irreversible deposition of retained particles, colloids, macromolecules, salts, etc. Consequently, fouling causes significant decrease in permeate flux [97]. Currently, the most common methods used to clean membranes include backflushing/backwashing and chemical cleaning of membranes, but these methods have some drawbacks and limitations [98]. Though backflushing is used in both hollow fiber [99] as well as

*This chapter has been published as [96]

flat sheet membranes [100, 101], it results in degradation of maximum flux after repeated backflushes. Chemical cleaning may damage the membrane and cause secondary pollution [102].

Ultrasound (US) is used for the cleaning of contaminated surfaces [103, 104]. Various studies report the enhancement of permeate flux using US [105, 106, 107]. This enhancement is attributed to phenomena related to bubble oscillations, acoustic streaming, and heating [108, 109, 110] which increase flux by affecting the concentration polarization at the membranes surface [111].

The frequency of the US driving is an important parameter. For example the effect of permeate flux with US irradiation at three different frequencies of 28 kHz, 45 kHz and 100 kHz had been investigated by Kobayashi *et al.* [112]. They found that lower frequencies (here 28 kHz) was more effective in cleaning fouled membranes. Similar results were obtained by Lamminen *et al.* [110]. Maskooki *et al.* [113] tested three ultrasound frequencies as well as a combination of these both with and without a chemical cleaning agent (ethylenediaminetetraaceticacid, EDTA). Interestingly, they found the best cleaning performance when the frequency was alternated between 28 kHz, to 45 kHz, and 100 kHz. Kim *et al.* [80] have demonstrated that high frequency can be applied to remove particles from silicon wafers, by means of high speed images they show that the bubble oscillating close to their resonant size induce fluid motion that causes the detachment of the solid particles attached to the silicon wafer. More recently, Thiemann *et al.* [114] studied the bubble structure present in a 230 kHz ultrasonic field; their work is relevant because their studied ultrasound frequency is similar to the high frequency of this work. They captured high speed video and performed sonoluminescence and surface

cleaning tests; two groups of bubbles were investigated, bubbles larger than the linear resonant size and bubble near or slightly below the linear resonant size. They concluded that both bubble populations showed cleaning potential, although they acknowledged that the mechanisms associated with particle removal may be different.

It is commonly accepted that the large amplitude bubble oscillations from so-called cavitation bubbles are responsible for the cleaning [110, 80, 114]. Even though the detailed mechanisms are not fully understood, experimental evidence points to the importance of the wall shear stress [80, 74]. The wall normal velocity gradient is caused by radial and surface oscillations as well as fast jet like flows.

Generally, cavitation bubbles grow from stabilized micron and sub-micron sized bubbles which may be stabilized by some shell or are trapped in crevices. These crevices are found on surfaces of the container or on particles suspended in the liquids. Also microbubbles are shed from strongly oscillating bubbles which serve as nuclei for later cavitation bubble growth. The complex flow pattern and acoustic field in ultrasonic cleaning devices makes it to date impossible to predict the location of these "natural" cavitation bubble nuclei, nor to control them. Only nuclei close to the membrane contribute to the cleaning. Thus two strategies can be applied to improve the cleaning results, either increase the US power to create and drive more bubbles, or increase the number of nuclei close to the surface. It is important to note that Kim *et al.* [139] found a novel way to achieve high particle removal efficiency while minimizing surface damage using dual ultrasonic chambers. One chamber with high acoustic pressure transducer for bubble generation while the other chamber with low acoustic pressure transducer for the cleaning. In-

deed, increasing the power not only increases the energy cost, but also may cause side-effects such as membrane damage [107, 108] and a lesser control of the sound field due to nonlinearities. The second strategy of actively generating bubble nuclei near the membrane is more promising. In this work we report on a technique to populate the membrane with bubbles which allows to keep the US driving at low amplitudes. The cost evaluation of these two options could be compared in a similar way to that shown by Kang and Choo [115].

Membranes surfaces possess pores which may acts as a gas pocket and become the place of bubble nucleation. Bubbles in the liquid are attracted by a an attractive force to the membrane boundary. Once on the surface these seed bubbles may grow in size by coalescence or by rectified gas diffusion [116]. Bubbles of similar size and driven in phase experience an attractive interaction force, the so-called secondary Bjerknes force [3] . This is valid for sufficiently mild oscillations; some of the details are discussed in [73]. The secondary Bjerknes force is also responsible for the attraction of bubbles from the bulk towards the membrane. This field force can be simply explained using potential flow theory. In potential flow the oscillation of a bubble at a distances s from a boundary can be constructed from the oscillation of two bubbles oscillating in phase at a distance of $2s$ using image theory [3]. Hence, bubbles become attracted towards the boundary. Thus, our novel method accumulate bubbles directly on the membrane before they are activated into cavitation bubbles; the accumulation and coalescence is induced with a high frequency ($f_h = 220$ kHz) ultrasound. Then the driving sound field is quickly changed to a lower frequency ($f_l = 28$ kHz). At this lower frequency these bubbles undergo much larger radial oscillations which then lead to cleaning of the membrane

surface. We termed this technique *tandem frequency excitation*.

The objective of this study is to propose and test this method to clean and/or prevent fouling of microfiltration membranes.

5.2 Experimental setup

To evaluate the tandem frequency method we compare the bubble dynamics and resulting cleaning with that from a single frequency excitation. The water filtration membrane (hollow fiber membrane, 1.9 mm in diameter, 30 mm effective length, 0.035mm average pore size, GECorp.) was placed inside a basin filled with water. A focused piezoelectric transducer (80 mm diameter, 8.5 mm height) with a brass backing was placed below. The distance between the membrane and the focused transducer is 33 mm which is the distance where the largest acoustic pressure was measured with the PVDF hydrophones (RP Acoustics, Germany). The transducer was driven with a sine signal from a function generator (Model 33220A, Agilent, USA) connected to an amplifier (AG Series 1006, T&C Power Conversion Inc., USA). The transducer is operated at up to 136 V in the high frequency and at up to 370 V in the low frequency mode. For the tandem frequency driving a digital pulse/ delay generator (Model 575, Berkeley Nucleonics, USA) was used to set the desired timing of the two frequencies.

The bubble activity near the membrane was recorded using a high speed camera (Photron SA1.1, Japan) at frame rates of up to 112,500 frames per second. Care was taken for a good illumination of the scene. The membrane was illuminated from the front by a metal halide light source (LS-M250, Sumita, Japan) coupled to a dual light guide; a mercury lamp (Olympus ILP-2) was used to illuminate the

Figure 5.1: Sketch of the experimental setup (A) and the geometry of the basin as seen from the camera position (B). The coordinate system is used in mapping of the pressure field, see Fig. 5.2. The plot in (C) sketches the timing of the two frequencies used in the tandem frequency driving.

membrane from the back. Fig. 5.1A and B are a sketch of the experimental setup. The center of the membrane is the origin of the coordinate system, see Fig. 5.1B.

The filter cake of bentonite was deposited on the membrane in the following way: First, a clean membrane was immersed into a bentonite suspension; one end of the membrane was sealed while the other end was connected to a peristaltic pump operating at a flow rate of 6ml per minute. A thick bentonite cake deposits onto the membrane surface within approximately 2 h. Next, the wetted membrane still connected to the operating pump is now submerged into a basin shown in Fig. 5.1A and filled with fresh tap water. The pump continues to run at the set pump rate of 6 ml/min.

Membrane cleaning typically utilizes ultrasound excitation at a single frequency, most common at the acoustic resonance of the specific transducer. The focused transducer has two resonances, one at 28 kHz and a second at 220 kHz. For the

Figure 5.2: (A) and (B): Typical waveforms at the origin showing at maximum driving for the high and low frequency driving, respectively. (C) and (D): Pressure map showing the amplitude distribution in the region of the membrane for the driving frequency of 220 kHz and 28 kHz, respectively. The "+" mark on (C) and (D) denotes the origin and center of the membrane, see Fig. 5.1(B). Note that (C) and (D) are recorded at a reduced driving voltage, therefore the pressure values are lower than in (A) and (B).

tandem frequency (TF) method, the membrane was sonicated with a high frequency ($f_h = 220\text{kHz}$) for 8 s and then immediately the driving frequency was changed to the low frequency ($f_l = 28\text{kHz}$) for 2 s, see Fig. 5.1C. The TF method is compared to a single frequency driving by sonication of the membrane for 2s of the low frequency ($f_l = 28\text{ kHz}$) only.

To document the acoustic parameters of the sound field, we mapped the pressure in the area around the membrane with PVDF hydrophones. For the low frequency a hydrophone with 3.8 mm tip diameter and for the high frequency a hydrophone with a thinner tip of 1.4 mm in diameter is used (PVDF hydrophones, RP Acous-

tics, Germany, type p and l, respectively). The pressure measurement was done in the absence of the membrane. The region sampled has the same size as the region captured by the cameras field of view in the tests of low frame rate, the tests with the largest viewing area. The typical pressure values and waveforms for the actual maximum amplitude used are depicted in Fig. 5.2A and B. Here, only a few cycles of the strong ultrasound is applied to the hydrophone. For obtaining a map of the sound field (see Fig. 5.2C and D) the transducer was run at a lower voltage. Here, a purely sinusoidal pressure was measured. It is evident that the pressure map shows distinct differences for the two frequencies used. For the high frequency driving the pressure field has a strong maximum near the origin (Fig. 5.2C), while for low frequency a more homogeneous pressure distribution is found. This difference may be caused by the focusing property of the transducer operated at high frequencies while for low frequencies a standing field is generated. The structures in the sound field should be of the order of half the wavelength, $\lambda/2 = 27$ mm. The high frequency driving will create a standing wave, too, which is overlaid onto the focused field. Fig. 5.2A and B: Typical waveforms at the origin showing at maximum driving for the high and low frequency driving, respectively. C and D: Pressure map showing the amplitude distribution in the region of the membrane for the driving frequency of 220 kHz and 28 kHz, respectively. The "+" mark on (C) and (D) denotes the origin and center of the membrane, see Fig. 5.2B. Note that (C) and (D) are recorded at a reduced driving voltage, therefore the pressure values are lower than in (A) and (B).

Figure 5.3: Upper row: frames taken from high speed recording of bubbles grown on the bare membrane. Time zero ($t = 0$) is set as the time when the acoustic driving is switched from high to low frequency. Hence, frames in the first column are in the high frequency regime while frames in the second and third columns are in the low frequency regime. Original frames from high speed recording. Lower row: frames subtracted with a background frame to enhance the contrast.

5.3 Results

5.3.1 Bare membrane

At first we demonstrate that the high frequency driving is able to nucleate bubbles on the surface of a bare membrane. Unfortunately, the resulting size of most of the gaseous bubbles are below the spatial resolution of the camera, which is about $13 \mu\text{m}/\text{pixel}$. They only become visible once the low frequency driving is switched on, and they expand multiple times their rest radii.

Fig.5.3 depicts in the top row the bare membrane in the first frame after approx. 8 s at low frequency driving, just before the low frequency is switched on at $t = 0$.

Figure 5.4: Comparison of the number of bubbles observed as a function of the non-dimensional time on a bare membrane with the TF method ($T = 36\text{ms}$ is the period of oscillation). The symbols represent different peak-to-peak voltage amplitudes of the high frequency driving. The amplitude of the low frequency remains unchanged. At time $t = 0$ the low frequency driving starts.

Two round bubbles are visible on the upper part of the left frame (indicated with circles in Fig. 5.3). At time $t = 0$ the low frequency excitation is started (the high frequency is switched off). We see within less than 100 ms large bubbles in the recording. The number of visible bubbles is counted manually as a function of time. For the bubble detection the image contrast is enhanced by subtracting a background image from each image. The background subtracted frames are depicted in the lower row of Fig. 5.3. The relative high framing rate of 112,500 frames/s makes it possible to visualize the bubble oscillation within a period at $f_l = 28\text{ kHz}$.

Fig. 5.4 presents the number of bubbles as a function of time for different

driving voltage amplitudes of the high frequency. Note that the flow rate is kept at 6 ml/min, as stated earlier. For the highest voltage of $136V_{p-p}$ we find many bubbles already after 2 cycles of the ultrasound (period of oscillation TE 36 ms). The number of bubbles oscillates with time and increases. For voltages down to $48 V_{p-p}$ (which corresponds to 1.4 bars) of the high frequency we observe similar behavior, yet the bubble appearance is retarded and their number is less. For $44V_{p-p}$ and below no bubbles can be found. The oscillation of the number of bubbles is due to the limited image resolution. Thus when bubbles shrink below approx $13 \mu\text{m}$ in size they cannot be detected. Therefore, Fig. 5.4 relates indirectly to the bubble size as a function of time. Fig. 5.4 demonstrates that for lower acoustic pressures (lower voltage amplitudes) it takes more time for bubbles to appear on the membrane surface; also the number of bubbles detected is lower. If the acoustic pressure falls below 1.4 bars no more bubbles are created on the membrane that can be used as seeds for cleaning during the application of the low frequency. On the other hand, increasing the ultrasound power too much may lead to membrane damage [117]. Yet, we have not observed ultrasound induced membrane damage in all measurements, e.g. we find a recovery of the trans membrane pressure (see below) and by visual inspection no bentonite particles in the filtrate.

5.3.2 Membrane with filter cake

Now we show and compare the membrane cleaning using a single frequency (SF) ultrasound with the tandem frequency (TF) method, Fig. 5.5. Fig 5.5A (SF) and B (TF) show each a freshly prepared membrane with a bentonite filter cake. Fig. 5.5A depicts the cleaning for the first second of acoustic exposure showing very

Figure 5.5: Frames of bubble activity on membrane covered with bentonite filter cake.

(A) SF method. Time zero corresponds to the time when low frequency ($f_l = 28\text{kHz}$) driving starts. Upper: original frames from high speed recording. Lower: frames subtracted with background frame to enhance the bubbles visibility and parts of the cake disturbed by the bubble dynamics. (B) TF method. Time zero is the time when the low frequency driving ($f_l = 28\text{kHz}$) starts after 8 s of high frequency driving ($f_h = 220\text{kHz}$).

little bubble activity, hardly any activity is noticeable in the direct picture. Yet in the lower row of Fig. 5.5A the background subtracted frames reveal some bubbles, circled in Fig. 5.5A. Also fragments of the filter cake are ejected as visible in the upper left corner of the last frame. We contrast this finding with the tandem method where the membrane is first exposed to 8 s of high frequency (driving voltage $136V_{p-p}$). The first frame of Fig. 5.5A ($t = 240\text{ ms}$) depicts a very small bubble weakly visible in the background subtracted image. This bubble is probably only mildly oscillating as no filter cake ejection is observed till then. However, as the frequency is reduced at $t = 0$, a cluster of bubbles emanates. Also pronounced ejection of the filter cake from the membrane surface is found. This originates from the bubble indicated with the arrow ($t = 32\text{ ms}$). Again, this bubble develops into a cluster of bubble populating the upper side of the membrane and leading to the

Figure 5.6: Plots of the number of bubbles observed on the membrane covered with bentonite filter cake as a function of time for the treatment with dual and single frequency method. Time zero starts when the low frequency driving starts. For the TF method, the low frequency regime ($f_l = 28\text{kHz}$) starts after 8 s of high frequency driving ($f_h = 220\text{kHz}$), see Fig. 5.1(C).

ejection of the filter cake.

We quantify the number of bubbles found for the TF method vs. the single frequency excitation. Fig. 5.6 demonstrates that within 100 ms more than 40 bubbles appear whereas it takes more than 0.5 ms for the first bubbles to appear using the low frequency only. The total number of bubbles is an order of magnitude larger for the TF method. As already visible in Fig. 5.5, the bubble dynamics leads to ejection of filter cake material into the bulk liquid. We now study the progress of membrane cleaning using the tandem and the single frequency excitation.

We now apply the protocol for the US multiple times and follow the removal of the filter cake. Fig. 5.7 depicts in the left column the SF and in the right column

Figure 5.7: Comparison of the membrane surface covered initially with a bentonite filter cake before (#0) and after 1-9 cycles (#01-09) of single frequency (SF) and tandem frequency method (TF) on the left and right column, respectively. Both videos were captured at 250 fps.

Figure 5.8: Bubble growth below the bentonite filter cake. The dashed circles are put in some frames to aid the readers eyes. The arrow points on a cloud of particulates removed from the membrane by the bubble. Upper: original frames from high speed recording. Lower: frames subtracted with background frame to enhance the bubbles visibility.

the TF method. For the SF excitation 2 s is waited between runs while for the TF method the high/low frequency is applied consecutively. For the TF method after nine runs of the protocol large bright patches become visible indicating removal of the filter cake. In contrast, the SF treated membrane (left column in Fig. 5.7) is still fully covered with the filter cake. From frame 6 (seventh frame from the top, right column), pronounced ejection of debris into the bulk becomes visible.

We find much more bubble activity if we apply a high-frequency field prior to the cleaning with a low frequency US field. High-speed video reveal that the formation of bubbles close to the membrane surface is enhanced. Some more detail of this process is visible in Fig. 5.8. This recordings show the bubble dynamics on the membrane together with the filter cake removal. In the rightmost frame of Fig. 5.8 bentonite particles are ejected in a round plume originating from a single position. Likely a single bubble or dense cluster of bubbles grows between the membrane and the cake, once the low frequency is switched on particles become ejected from this position, visible after 4.5 ms (see arrow in the last frame of Fig. 5.8). We notice

Figure 5.9: Bubble oscillating while translating along the top part of the membrane and ejecting materials of bentonite filter cake upwards. Frames are subtracted with background frame to enhance the visibility of the ejected cake.

also that the bubble location is slightly moving to the right, it is translating as well as oscillating.

Besides the usual bubble random translation for the cleaning of membrane surface, we also noticed that bubble may translate along a straight path on top of the membrane, as can be seen in Fig. 5.9. While translating, the bubble ejects materials from the cake. The low frame rate used (250 fps) does not allow unambiguous measurement of the velocity in this measurement. However, we found that the lower bound of the velocity of the ejected materials is approximately 0.125 m/s. The frames of Fig. 5.9 have been subtracted with a background frame to enhance the bubbles motion and the particles removed by it.

5.3.3 Tandem frequency validation experiments

Of the two methods tested, only the tandem frequency is able to remove the accumulated bentonite particles on the membrane, the single frequency method does remove some of the cake but a large portion remains attached to the surface of the membrane. Besides this visual verification of the proposed membrane cleaning technique we tested the membrane cleaning performance by registering the trans

membrane pressure (TMP) and the flux through the membrane.

Our small scale experiment was setup and conducted as follows: First the membrane is immersed in a small beaker that contains a water and bentonite solution (12 g/l) to form a cake for 25 min, while the pressure at the exit of the membrane is recorded with a Wika pressure sensor (-100kPa to 100 kPa range) and the weight of the permeate is measured in an Ohaus scale (model PA413C, read out with a computer interface). When the cake is formed the peristaltic pump is turned off and the membrane is transferred to the container previously described that houses the focused transducer. Because of the rather small focal zone of the transducer we placed the membrane in clean water to make sure it is located at the right position. The time it takes to transfer the membrane from the small container with the water bentonite mixture to the container housing the focused ultrasound is less than 60 s; we do not expect a softening of the integrity of the cake from the time the pump is turned off to the time when the ultrasound is applied.

In the test shown in Fig 5.10a about 35 cycles of TF excitation were necessary to clean the membrane. Once the membrane is cleaned it is placed again in the small beaker with bentonite and the pump is turned on again. Fig. 5.10 shows the results of tests conducted in a hollow pressure membrane at constant flux. Fig. 5.10a depicts the change of TMP as a function of time. The "pure water" curve is a control for the TMP: no change of the TMP is observed. The "No US applied" curve represent the pressure obtained when the membrane is kept in the waterbentonite solution and no ultrasound is applied; this curve shows that the TMP increases with time. These two curves are used as a benchmark against which the tandem frequency is compared. The "Tandem Freq applied" curve depicts the

Figure 5.10: The transmembrane pressure TMP and the weight of liquid delivered for the three conditions tested. (A) The TMP as a function of time. Regions a-d correspond to the pump-on time during the tandem frequency tests. and (B) The weight delivered by the low pressure system for the three conditions tested, from the weight information the average flux obtained in regions a-d was obtained

TMP obtained with the application of ultrasound to remove the filter cake. When the test begins the TMP increases just as it does in the "No US applied" curve, then when the pump is turned off the TMP drops to zero. After the membrane is cleaned and the pump is turned on again the TMP reduces to approximately the same value it had at the beginning of the test, i.e., the value for the pure water case. Even after repeatedly fouling and cleaning of the membrane the minimum TMP returns to the control value, i.e., no residual fouling is observed [100].

We now analyze the mass flux. Fig. 5.10a depicts four regions labeled a-d and we compare the flux for the three cases, pure water, water bentonite, and TF excitation. Fig. 5.10b depicts the weight delivered during the time the tests are conducted. The flux delivered in the pure water case is practically constant. The "No US applied" curve follows the pure water curve closely at the beginning of the

Table 5.1: Average flux obtained in regions a-d with the three different conditions tested, pure water, water-bentonite solution and tandem frequency.

Region	Pure water (l/m^2h)	Water-bentonite (l/m^2h)	Tandem frequency (l/m^2h)
a	34.1	35.7	35.1
b	33.4	32.3	34.2
c	32.6	29.5	32.3
d	32.8	28.4	32.9
Average	33.2 ± 0.7	31.5 ± 3.3	33.6 ± 1.3

test but its slope decreases and after 90 min it is already noticeable that the flux is lower than that of the pure water test. Finally, the slopes of the TF curve in regions a-d are very similar to the slope of the "pure water" curve, the three flat sections shown in the "tandem frequency curve" correspond to the time when the pump was turned off thus no liquid was delivered.

Table 5.1 is a summary of the average flux in regions a-d. This table shows that the TF method is indeed able to maintain a steady liquid flux.

5.4 Discussion

The tandem frequency (TF) method presented here has some similarity to the cavitation control lithotripter work from Matsumoto's group [118]. There is a cloud of small and mildly oscillating bubbles are generated near a stone surface using a high frequency. Then the driving sound field is quickly switched to a low frequency which resonantly induces a concerted collapse of the cloud. In both settings a high frequency is used to nucleate bubbles close to the target. The difference here is that the bubbles in our experiments do not form a dense cloud. Instead of the clouds resonance frequency the lower frequency in our experiments

excites the non-linear "resonance" of the individual bubbles, the so-called dynamic Blake threshold [119, 120]. The bubbles thus oscillate at a much larger amplitude as compared to the high frequency excitation. A further difference between the cavitation control lithotripter [118] is that we use a low frequency resonance of the transducer while there the excitation frequency is matched to the resonance frequency of the cloud. The membrane cleaning method here presented shares some similarity to the study presented by Maskooki *et al.* [113]. They found best clean of microfiltration membranes by using an alternating sequence of frequencies (the direction and timing is not stated in their work). They report that the alternating sequence of 1 min each at 28 kHz, 45 kHz, and 100 kHz gave the best results. Unfortunately, no details on the order of application of the different frequencies are stated and the dead time between to frequencies is also not mentioned. With the reported data at hand we can only speculate that the order from low to high and back to low gave the best results. In that case the flux recovery may be similarly caused as reported here: the highest frequency may generate the bubbles on the surface, while the lower frequency similar to our method cause strong oscillations of these seed bubbles.

Particulate removal can be accomplished with the use of high frequency ultrasound, as shown in the work of Kim *et al.* [80] and Thiemann *et al.* [114]. However, in the current study the high frequency is only used to accumulate nuclei on the surface, while strong bubble/cavitation activity is to created with the low frequency driving. This application of short low frequency cycles may prolong the membranes lifetime. Yet, real cleaning arrangements uses not only a single hollow fiber but many bundled stacks. Although we still think that the generic

approach of a tandem-frequency driving is suitable, one would need to take care in the actual implementation of the acoustic field. In particular if the bundles are too dense they may prevent the penetration of the ultrasound field into the inner hollow fiber bundles. Thus a less densely packed bundled could be advantageous. Also, standing wave effects in the filter container housing may lead to preferential sites of cleaning of the bundles. Then a moderate modulation of the high and low frequencies may help.

The TF method was tested on a hollow membrane yet we expect that it is equally applicable to flat sheet membranes.

5.5 Conclusions

We find that the use of two ultrasound frequencies is more efficient in removing particulate matter deposited on a circular membrane. We attribute the better cleaning performance of the TF method to a larger number of bubbles created on the surface of the membrane prior to the application of a lower frequency which induces cavitation. These low frequency driven cavitation bubbles acts as scrubbers that perform the cleaning of the membrane. The technique may be generally applied to seed the surface to be cleaned before the high amplitude/low frequency US is applied. This splitting of the seeding of the surface from cleaning could lead to less damaging protocols in US cleaning. Still, the details of the enhanced bubble formation at high frequency need to be elucidated. The proposed technique was applied in a small scale experiment that showed that the flux obtained can be constant thus it can be implemented as a mechanism to de-foul membranes in low pressure ultra filtration applications.

Other issues such as the quality/type of the filter cake that can be removed with the present technique, long term effects of ultrasound on the membranes, and the energy balance need to be addressed. Nevertheless, we hope this work stimulates others to have a fresh look at US cleaning through the deeper knowledge of bubble dynamics and its correlation to the cleaning process.

Chapter 6

Multi-bubble sonoluminescence in flowing liquid

6.1 Introduction

Single-bubble sonoluminescence (SBSL) has been theoretically and experimentally investigated since its discovery in 1992 by Gaitan *et al.* [54]. Its investigation enjoys the robust experimental method developed and the relatively simple theory of single bubble dynamics derived from hydrodynamics point of view [121], which lead to a rapid progress of scientific understanding. The light emission from a cloud of bubbles (MBSL for multi-bubble sonoluminescence) in contrast has already been studied since 1933 [52]. Yet, level of maturity on the theory lags behind as compared to SBSL, primarily due to inherent complication of many bubbles system [58].

In SBSL experiments a bubble is trapped within a flask at the pressure antinode of an acoustic standing wave. While the typical experiment for MBSL is usually

involving an ultrasonic horn which vibrates strongly and is immersed in the liquid. Due to the rapid oscillations, the cavitation bubbles are excited which by some physical mechanisms are able to emit light during their violent collapse. From the nature of the experiments, one can easily see that MBSL is less predictable than SBSL, e.g. of bubbles location is not fixed, number of bubbles created are many and apparent as a cloud of bubbles, bubble dynamics are more complex due to bubble – bubble interactions, and so on.

The light emission occurs due to the violent collapse of bubble. It may serve as an indicator of how rapid the bubble dynamics is during the collapse. Further, bubble dynamics is important in flow processing, more rapid dynamics usually means more effective the bubble for flow processing. It may be an indicator of how well the bubbles act as catalyst for chemical reactions (i.e. sonochemistry), help with liquids mixing, disperse particulates and so on. For practical applications, cavitation clouds rather than single well-controlled bubble are needed. Hence, despite SBSL has been a very important to understand the physics of sonoluminescence, MBSL may be more practically important for real world applications.

Recent experiment show that sonoluminescence (i.e. MBSL) can be detected in a very confined geometry, i.e. microchannel [122]. This suggests that asphericity may be not too severely affect the final stage of bubble collapse. MBSL has been shown to be intensified by a forced flow in the form of circulation and stirring [123]. This result was further investigated for sonochemistry purposes by Kojima *et al.* [124]. He found that a combination of acoustic and mechanical flow enhances sonochemistry by reducing the possibility of bubbles to group or coalesce and maintain the supply of active bubbles.

In this chapter, we report on MBSL affected by streaming flow. Quite different from previous investigators [123, 124], in our experiment the bubbles are generated outside the acoustic cell and are transported with the flow into the acoustic field. Due to high frequency driving used, it is difficult to resolve the individual oscillation of the bubbles. However, sonoluminescence is used as an indicator of how violent the bubble dynamics is. We use a photomultiplier, an ICCD camera and a high speed camera to study the effect of the liquid flow on the sonoluminescence and the bubble dynamics.

6.2 Experimental setup

The experiments were done in two stages. First, the sonoluminescence was characterized using photomultiplier signals and recording of the intensified and cooled CCD camera. Second, the bubble translational dynamics was investigated using high-speed camera (Photron SA1.1, monochrome, Japan). Note that during the first stage, the experiments were done in the dark, meanwhile during the second stage extra illumination was needed. The complete setup for both experiments is depicted in Fig. 6.1.

An acoustic cell was the subject of the investigation. The flow direction, as seen in Fig. 6.1, was set in such a way that liquid is sucked from the top side with a gear pump. The initial pump (Cole-Palmer, Model 75211-15, USA) was later upgraded to a digital controlled pump (Cole-Palmer, Model 75211-35, USA). The flow rate in the system was measured with a flow meter (McMillan, Model 101, USA). Bubbles were generated with a home built generator (see below) and transported into the cell by the liquid flow. Singapore tap water without any special treatment was

Figure 6.1: Sketch of the experimental setup. Details of the cell is explained in Fig. 6.2. Note that for the first stage of experiment, the illuminations were not used and the camera used was a cooled ICCD camera. For the second stage of the experiment, PMT was not used and the camera used was high speed camera.

Figure 6.2: Sketch of the cell used. Left picture is the outline of the whole cell. The semi-transparent lines in the left picture represent the upper part of the cell. Note that liquid inlet and outlet are not drawn. The bold lines represent the bottom part of the cell, consist of triangular-shaped metal and piezoelectric transducer. The lines represent the tops of triangular metal parts. The lines also represent the elongation of the triangular metallic structure. Right picture is the cross section of bottom part of the cell which show the metal and transducer parts. Arrows are used to mark the part of left picture which is zoomed as cross section sketch in the right picture.

used. Bubble generation rate and flow rate can not be independently controlled.

The flow cell is a rectangular container ($22.5 \times 28 \times 80 \text{ mm}^3$) with one of its side is a piezoelectric transducer (depicted in Fig. 6.1 as a thick line at one side of the cell). The transducer is attached to a metal plate which connect directly to the water inside the cell. The metal plate has triangular structures, see Fig. 6.2 for an illustration. The piezoelectric transducer, connected to amplifier and a function generator, vibrates the cell at the resonance frequency of 438 kHz. An acoustic standing field is then formed and leading to the entrapment of bubbles at certain locations. It seems that due to the special shape of the metal plate a non trivial sound field is set up leading to a trapping of the bubbles and parallel to the structure. This is evident in the experimental results.

The bubble generator used is a lab-made device. It consists of a channel with a gradation of its cross-sectional area. The area is $1 \text{ cm} \times 1 \text{ mm}$ at the water inlet. The area is at its minimum, $0.5 \text{ mm} \times 0.5 \text{ mm}$ about 1 cm after the water inlet. Millimeter sized bubbles which enter the nozzle are split into many smaller bubbles. These bubbles are polydisperse with a typical radius range from 30 – 100 μm .

A photomultiplier (H5783, Hamamatsu) connected to a sampling oscilloscope (WaveRunner 64Xi-A, LeCroy) is used to record the time-resolved light emission generated by sonoluminescence from the bubbles. The PMT was set to almost highest amplification. The oscilloscope can store more than 10^7 samples which aids data analysis of many brief light emission events.

The cell has a characteristic impedance. As seen in Fig. 6.3, the low impedance of around 50Ω is at driving frequency of around 440 and 450 kHz. The amplifier

Figure 6.3: Impedance plot of the cell as a function of driving frequency. Data are by courtesy of Lam Research.

also has low impedance of $50\ \Omega$. Manual tuning revealed that the power amplifier gave approximately zero reflective power to the cell and the cell exhibited strongest bubble activities at 438 kHz, hence the driving frequency was set at that specific frequency.

6.3 Experimental results

We start with the results from the PMT measurements. Fig. 6.4 shows a typical PMT output where a sinusoidal driving voltage is applied on the transducer. The flow rate is zero. During sonication light emission is observed which stops abruptly after the transducer is switched off ($t = 3\text{ s}$). For a close look on the sonoluminescence, detailed measurement of the first 4.5 ms were done. This time limit is a constraint from the oscilloscope.

While step wise increasing the flow rate from 0 to approximately 45 ml/min, PMT signals retrieved during the first 4.5 ms are compared in Fig. 6.5. From typical

Figure 6.4: Sample of PMT output and the driving voltage applied on the piezoelectric transducer taken at zero flow rate (pump is off). Driving of piezoelectric transducer is switch on at $t = 0$ and off at $t = 3$ s.

results depicted in Fig. 6.5, it is clear that a drastic drop of sonoluminescence signals happen at approximately 45 ml/min. Increasing the flow rate even further did not change the result; the sonoluminescence signal is close to zero.

To quantify the number of sonoluminescence events, a simple Matlab code is developed. The code calculates how many peaks related to sonoluminescence exist in a given PMT recording, and marks each event. Yet event with a too small signal is not counted. The threshold value is calculated by taking into account the noise level without ultrasound excitation; hence for different measurements the thresholds may alter slightly.

Shown in Fig. 6.6 is the typical signal in detailed measurement of the first 4.5 ms since the start of the ultrasonic irradiation and the detected events of sonoluminescence. The left picture shows the original PMT signal. The result is shown in the right picture of Fig. 6.6. For better viewing, the result is zoomed from 2 to 2.4

Figure 6.5: Samples PMT output signal for the first 4.5 ms after ultrasound irradiation with variation of 0, 10, 20 30, 40 and 45 ml/min flow rates (from top to bottom).

Figure 6.6: PMT output signal for the first 4.5 ms after ultrasound irradiation. Here, the before (left) and after (right) sonoluminescence events recognition result of the signals using Matlab are demonstrated; zoomed for $t = 2$ to 2.4 ms. Ultrasound of 438 kHz frequency is started at time zero. The event peaks are marked with red triangles in the right picture. Flow rate of water is set as zero (pump is off).

ms. Each sonoluminescence event is marked with a red triangle.

The sonoluminescence events counting is performed for signals at different flow rates, i.e. 10, 20, 30, 40, and 45 ml/min. We got typically 4 – 7 measurements for each flow rates. Figure 6.7 depicts the number of sonoluminescence events captured during the first 4.5 ms of ultrasonic radiation. In Fig. 6.7, it is shown that the number of event drops drastically to approximately zero at flow rate of 45 ml/min and above. For smaller flow rates we find the events only occur after approximately $t = 100 \mu s$. The number of events seem to increase slightly when the flow rate is increased to around 10 ml/min.

To investigate the sonoluminescence structure, the ICCD camera is used. In general an ICCD camera has a much lower sensitivity than a PMT. The result is depicted in Fig. 6.8. To enhance the images quality, they are processed by inverting the intensity values (dark to bright and vice versa) and enhancing the contrast. Here, the flow rate is varied from zero to a sufficiently high flow rate with

Figure 6.7: Number of sonoluminescence event captured during the first 4.5 ms of ultrasonic irradiation. The number of event drops drastically, to approximately zero, at flow rate of 45 ml/min.

the acoustic field switched on. Without flow, the distinction between the non-sonoluminescing and sonoluminescing regions is clear. By slightly increasing the flow to 10 ml/min, the structure seems to begin spreading, in other words it seems that a large region is sonoluminescing. It is possible that due to lower sensitivity of the ICCD only streaks of the light emissions are captured. In other words, ICCD camera can only capture stably sonoluminescing areas which give stable emissions. At 20 ml/min, it seems that the sonoluminescing region is spreading. There is only small region distinguishable from others. The results for higher flow rates are not shown due to there is no sonoluminescence structure recognized. This may be caused by two reasons: Firstly, the sonoluminescing region is distributed more randomly, i.e. not much of repeating sonoluminescence event at a given spot, and secondly at high flow rates the number of sonoluminescence events is close to zero.

Figure 6.8: Sonoluminescence structure captured by ICCD camera; camera and ultrasound exposures are 3 s. Top row: original images from ICCD camera. Bottom row: inverted (bright to dark and vice versa) and contrast-enhanced images. The white (top row) or black (bottom row) scale bars at the bottom of each frame has length of 1 mm. Frames A, B and C depict sonoluminescence at 0, 10 and 20 ml/min of flow rate, respectively.

Since sonoluminescence is due to bubble dynamics, it is important to show what type of bubble dynamics is happening. Radial bubble dynamics can not be captured due to very high driving frequency use (i.e. 438 kHz), which exceeds the highest framing rate the high speed camera offers. Hence, we resort to investigating the bubbles translational dynamics. The most common bubbles configuration is where there is a big bubble with many small satellite bubbles around it. This bubble configuration is similar to what is first reported by Miller [125] in acoustic standing wave field at a driving frequency of 1 MHz, hence we name this phenomenon as Miller bubble configuration. Similar configuration is also observed by Thiemann *et al.* [114] recently at driving frequency of 230 kHz.

In our experiment with flow, different from Miller, the big bubbles are not due to rectified diffusion, but due to bubbles from the bubble generator. The big bubble is likely not at all oscillating by the acoustic field, due to its size being much larger than the resonance size. Meanwhile the small bubbles, due to their size closer to resonance (around 10 μm radius), are more likely to oscillate radially.

Figure 6.9 depicts the ramp up of the flow rate as a function of time. Here, we relate the time at certain flow rates to the high speed recording with the frames shown in Fig. 6.10. It is evident that at first the bubbles are at some distance from one another. Increasing the flow rate tend to align the bubbles and make them move closer to each other. The small bubbles are spiralling around a common axis to the bigger bubble. This bubble configuration is reported earlier by Miller [125].

In Fig. 6.10, the original frames from the high speed recording (taken at 54000 frame per second) are presented along with their corresponding image processed frames. The originals are in the top row while the processed frames are in the

Figure 6.9: Flow rate ramp up as a function of time. Letters A to G denote the corresponding flow rate at times 0.2, 0.4, 0.5, 0.55, 0.57, 0.59 and 0.61 s after start of high speed recording as depicted by frame snapshots in Fig. 6.10.

bottom row. Image processing is done by subtracting a background image (without bubble activity) from the original frame.

As mentioned before, the typical motion of the small bubbles is in ellipsoidal or spiral-like trajectory. This is true for zero and low flow rates up to approximately 20 ml/min. An example of the upward motion while spiralling is depicted in Fig. 6.11. Note that in the figure only processed images (i.e. after background subtraction) are shown for better viewing. The figure is an overlay of several frames to elucidate the different positions of the bubble and show that the bubble is indeed undergoing a spiralling motion, while moving slowly upward. Upward is the direction of the water flow.

For elucidating more on the bubble dynamics, Figs. 6.12 to 6.14 are presented. Figure 6.12 depicts the bubble dynamics at zero flow rate. Here, the bubble dy-

Figure 6.10: Frame of bubble motion affected by increasing the flow rate. The black scale bar at the bottom of each frame has length of $50\ \mu\text{m}$. Top row are the original frames from the high speed recording, taken at 54000 frame per second. Bottom row are the background subtracted frames of the top row. Letters A to G correspond to the flow rates as indicated in Fig. 6.9, i.e. 4.7, 10.4, 13.4, 14.7, 15.9, 16.2, and 17.2 ml/min, respectively.

Figure 6.11: Examples of ellipsoidal motion of the bubbles. The image has been processed for better viewing. The black scale bar in window A is $50\ \mu\text{m}$ in length. Window A is the frame showing the initial condition of bubbles a and b. Windows B and C consist of few frame overlays. In Window B, bubble a is moving in ellipsoidal/spiral-like upward motion while 1,2 and 3 denote its position at 20, 80, and $240\ \mu\text{s}$ respectively after window A, while bubble b is almost stationary. In window C, bubble a has moved to 1, 2 and 3 and bubble b has moved to 4 and 5. Positions 1, 2 and 4, 3 and 5 are at 780, 840, and $940\ \mu\text{s}$ respectively after window A. The arrows are added to aid the reader on the direction of bubble motion.

Figure 6.12: Bubble translation dynamics at no flow. The arrow in left of the figure represents the direction of the liquid flow and the elongation of triangular structure of the metal plate (see Fig. 6.2). The direction is the same for the rest of the figures. The black scale bar on the last frame is $50 \mu\text{m}$ in length.

namics is relatively stable, in particular that of the bigger bubble. There is not much change of the location of the bigger bubble even after 300 ms. Here the bigger bubble is still relatively small, approximately $20 \mu\text{m}$ in radius. They are grown at that size due to coalescence and possibly rectified diffusion. Slight change of the location may be due to the growth of the bubble, i.e. in the last frame it has grown approximately $4 \mu\text{m}$ in radius as compared to the first frame.

Figure 6.13 depicts the bubble dynamics at a flow rate of 20 ml/min . The bubble dynamics is less stable, where the bigger bubble has moved away after approximately 130 ms. The bigger bubble here is generated bubble generator. Hence, its typical size is bigger than in the case of zero flow rate. Here the bigger bubble radius is approximately $30 \mu\text{m}$. After the bigger bubble moves away, another bigger bubble with similar size comes into the field of view.

A case of high flow rate, 40 ml/min , is presented in Fig. 6.14. Here, it is difficult to capture small bubbles due to the abundance of the big bubbles. The small bubbles are hard to find, however still do exist. It is probably due to more bubbles are generated from bubble generator that they have bigger chance to coalesce and

Figure 6.13: Bubble translation dynamics at 20 ml/min flow rate. Similar to Fig. 6.12, upward is the direction of the liquid flow and the elongation of triangular structure of the metal plate (see Fig. 6.2). The black scale bar on the last frame is 50 μm in length.

Figure 6.14: Bubble translation dynamics at 40 ml/min flow rate. Similar to Fig. 6.12, upward is the direction of the liquid flow and the elongation of triangular structure of the metal plate (see Fig. 6.2). The white scale bar on the first frame is 50 μm in length.

increasing in size quickly. In Fig. 6.14 we show the typical big bubbles seen are not only in the size of around 30 μm in radii (frames 1 to 3 from left to right), but also of radii approximately 120 μm (frame 4 to 5). The even greater flow rate may cause the annihilation of the small bubbles due to coalescence.

For slow flow rates bigger bubbles and small satellite bubbles system resembling Miller configuration are found. While for higher flow rates (greater than 20 ml/min) many much bigger bubbles are seen with less stable dynamics of the small bubbles. At high flow rate (approximately 40 ml/min and higher), small bubbles can not be seen easily any more.

6.4 Discussion

The magnitude of the liquid velocity inside the cell can be estimated using the geometry of the cell and the flow rate. The cross-section area of the cell is approximately $A = 630 \text{ mm}^2$. The corresponding flow rate for sonoluminescence disappearance is approximately 45 ml/min. From $Q = A \times u$ we obtain a liquid velocity of $u = 1.19 \text{ mm/s}$.

Our experimental observation show that at sufficiently high liquid flow velocities Miller array can not be formed. We now do an order of magnitude estimate to see if the drag from the flow can overcome acoustic forces, i.e. the primary Bjerknes force. The ratio of the two forces is

$$\frac{F_D}{F_B} = \frac{6\pi\mu Ru}{\langle \nabla P \rangle V} \quad , \quad (6.1)$$

with F_D is the drag force due to liquid velocity in the limit of low Reynolds number, F_B is the primary Bjerknes force, μ is liquid dynamic viscosity, R is bubble radius, P is the acoustic pressure and V is bubble volume. Symbol $\langle \rangle$ denotes time average. The ratio may be approximated as

$$\frac{F_D}{F_B} \approx \frac{6\pi\mu Ru}{P_a \frac{2\pi}{\lambda} \frac{4}{3}\pi R^3} \quad , \quad (6.2)$$

where λ is the wavelength of the acoustic wave and P_a is acoustic pressure amplitude, and can be rewritten as

$$\frac{F_D}{F_B} \approx 0.7 \frac{\mu c u}{f P_a R^2} \quad (6.3)$$

where c is acoustic wave velocity and f is the driving frequency. To affect the bubbles, the drag force has to be larger than the Bjerknes force. Hence,

$$0.7\mu c u > f P_a R^2 \quad . \quad (6.4)$$

Estimating acoustic pressure amplitude as 10^4 Pa, the liquid velocity should be

$$u > (417 \times 10^7 R^2) \frac{m}{s} . \quad (6.5)$$

The resonance radius can be estimated as approximately $R_{res} \approx 3/f$, which yield $R_{res} \approx 6.8 \mu\text{m}$ for the driving frequency $f = 438$ kHz. From Eq. 6.5, the minimum velocity needed to overcome Bjerknes force for $R = R_{res}$ would be $u_{min} \approx 0.19$ m/s. For the bigger bubbles which can be seen in experiments, due their size of the order of 10^{-4} m in radius, the minimum velocity would be approximately $u_{min} \approx 1$ mm/s. This means that the small bubbles that sonoluminescing are not much affected by the drag force, yet big bubbles will be.

The ellipsoidal trajectory of the bubble has also some resemblance with the case of moving single-bubble sonoluminescence (m-SBSL) as found by Didenko *et al.* [126]. The m-SBSL path instability was theoretically explained by Toegel *et al.* [127] with an increased viscosity. Their analysis using Keller-Miksis equation¹ showed that the history force is the culprit leading to the instability. Different from m-SBSL case, in our experiments the ellipsoidal motion is not due to increased viscosity, since here we use water. We suspect that the cause is the interaction between bubbles and the flow. It is interesting to note that for MBSL the translational motion of the small bubbles are not severely affecting their light emissions.

Classical theory predicts that bubbles gather in pressure nodes or antinodes [128]. In sufficiently high intensity acoustic standing waves, bubbles have been shown to undergo erratic motions [129], zigzag motion in pressure variation direction [130], and forming halo around pressure antinode [131]. The shape oscillations need not be the case for the phenomena. Watanabe and Kukita [132] show theo-

¹See Chapter 2 for some discussion regarding Keller-Miksis equation.

retically that spherical bubble may have irregular translational motion if the radial pulsation is strong enough. This later extended by Doinikov [133] to take into account the effect of liquid compressibility. Hence, we suggest that the bigger bubbles are important for the formation of a stable bubble array as also suggested by Miller *et al.* [125] and Thiemann *et al.* [114]. Less stability may cause the bubble sonoluminescence to be reduced.

In highest liquid velocity, small bubbles can not be observed. Note that the flow rate is also affecting the typical bubble sizes produced by the bubble generator. The bubble generator working principle is mainly due to Venturi effect, where very low pressure are generated inside the device and thus generating cavitation bubbles. Typically, the higher the flow rate will yield bigger bubbles. Bubble generators using Venturi effect have been used by previous researchers, e.g. in [137] and [136]. Another mechanism of the bubble generator is by bubble fission. If a big bubble enters the bubble generator it will be broken into many small bubbles. This is similar to what is observed in [138]. By this mechanism, the typical bubble sizes produced are also increasing after bubble fission for higher flow rate. Furthermore, coalescence occurrence at downstream is also the cause of too many big bubbles and lack of small bubbles observed in this experiment at high flow rate. The water inside the cell may also be saturated with much bigger bubbles than the resonance size, which hinder the sufficiently long appearance of smaller resonant bubbles. This is due to the smaller bubbles are swept away by the bigger bubbles, or coalesce with other bubbles.

The application of the cell in flow processing also looks promising. The small bubbles activity especially can be used to disperse particulate, mixing and sono-

chemistry applications. If the flow rate is not too high (20 ml/min or less), the translational activity of the bubbles system may be beneficial. Hence, larger area may be swept by the active bubbles. Though the coalescence may happen sooner or later, the continuous flux of new bubbles due to bubble generator may help to renew the bubble activity.

This application is important for example for structural or material enforcement using carbon nanotubes (CNTs). CNTs often comes as a bulk where they stick to each other. To mix them in more homogeneous distribution with other materials is a challenge. For this purpose, bubbles may be used to induce local flow which disperse the aggregate. However, it would be more advantageous in an experimental point of view if the driving frequency can be adjusted in demand. In current setup this is difficult due to impedance matching between the cell and amplifier. This is possible to be addressed for example by using impedance matching circuit. If the resonance frequency can be reduced, it would be easier to resolve directly the individual bubble oscillations. Thus, results interpretations are more direct and less prone to error.

6.5 Conclusions

In conclusions, we have done experiments on multi-bubble dynamics and sonoluminescence and their interplay with streaming flow. Bubbles are introduced using bubble generator. We observe stable bubble arrays for low flow rates. At higher flow rates, big bubbles can be dragged by the flow. At certain flow rate the sonoluminescence cease to exist, the main reason is most probably due to destabilization of the bubble arrays. Another factor is the saturation of the cell with bubbles

bigger than resonance radii due to coalescence.

The cell has potential application for particulate dispersion using bubbles. The small bubbles will localize flow in this purpose. In particular, it may be useful for CNTs aggregate which needed to be distributed more equally to strengthen other materials. Better experiments are possible by usage of impedance matching circuit for varying the driving frequency.

Chapter 7

Conclusions and outlook

7.1 Conclusions

In this thesis, we have presented theoretical and experimental results related to bubble dynamics near a rigid boundary, in particular for membrane cleaning and flow processing applications. From our observations we reach to the following conclusions:

1. In the linear (i.e. small amplitude) oscillation regime, it is possible to derive analytical solution for the wall shear stress generated by gas bubbles oscillating spherically near a rigid boundary. The thermodynamics of the gas within the bubble is approximated by the assumption of a uniform pressure. Thermal, acoustic and viscous damping effects are included.
2. Even for small amplitude oscillations, the bubble may generate wall shear stress of up to 1 kPa. This may be sufficient for less demanding cleaning application, e.g. in water filtration membrane cleaning. The optimum frequency in resonance condition is of the order of a few MHz, which is in the range

used in megasonic cleaning. This may offer an explanation for the success of megasonic cleaning technique in semiconductor processing industry.

3. Besides bubble oscillations, jetting has also potential for cleaning since it may generate high wall shear stress too, as shown by previous research. But not all jetting flows contribute to cleaning and surface erosion. For micron-sized bubble attached on a surface, if the jet velocity is not high enough, the cleaning effect is maybe weak. Repeated low velocity jettings may also building up liquid cushion on the surface which weakens the cleaning effect. However, other factors such as surface oscillations and bubble travelling while oscillating on surface may contribute to cleaning.
4. The cleaning from bubbles travelling while oscillating along a surface is observed on fouled hollow fiber membrane. The conventional technique of ultrasound cleaning of membranes usually employ single-frequency driving. We have shown that a tandem-frequency driving, consisting of a low frequency for growing bubble on the membrane surface and a high frequency for driving bubble to do the cleaning is superior.
5. The bubbles activity may also used for flow processing. Here we found that for sufficiently low flow rate the sonoluminescence is only weakly affected. However at higher flow rates large bubbles become dragged and destabilize the condition necessary for driving the small bubbles in the light emission regime.

7.2 Outlook

The analysis method presented in Chapter 3 may be extended to incorporate many bubbles. Exploiting symmetry, i.e. in the case of monodisperse bubbles, analytical solution may still be possible to achieve. The nonlinear case however even for single bubble case can not be solved analytically. For nonlinearly oscillating polydisperse multi-bubble system, a computationally cheap algorithm is needed for steady state wall shear stress investigation.

In the bubble near wall modeling and wall shear stress simulations presented in the thesis, the bubble is limited as spherical hence the bubble oscillation amplitude and the wall distance are kept in check to maintain the validity of this assumption. However, as can be seen in experiments, for large-amplitude bubble oscillations and a sufficiently near wall this assumption will not hold. To investigate this effect, several approaches may be done in future study:

1. By numerically solving Laplace's equation using Boundary Element Method. However, due to the nature of Laplace's equation, viscosity is neglected. Hence a way to couple viscosity, especially at the surface of the wall and the bubble wall, to the numerical solution has to be found.
2. By numerically solving Stokes flow using Boundary Element Method. However, here the inertia is neglected. Hence, corrections on the numerical solution is necessary to take into account the inertia.
3. By numerically solving the full Navier-Stokes equation. However, the computational cost for this approach is very expensive. Massive computing power is needed to get steady state solution of the bubble and the wall shear stress.

Indeed this problem is still an open problem.

Theoretical analysis may be developed to support experimental results in Chapter 5, tandem frequency for membrane cleaning. The method can be extended to not only tubular hollow fiber membranes, but also to a flat sheet membrane. From an experimental point of view, the geometry of a flat sheet membrane is much better for image capturing and high speed video recording.

Lastly, the sonoluminescence in a flowing liquid experiments in Chapter 6 may be extended to bubbles of more controlled size and different driving frequencies. The cell may also be used as a flow processing device, in particular for mixing and dispersing particulates in water (e.g. for dispersing carbon nanotubes).

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Appendix A

Derivation of wall shear stress formulation

For completeness of Chapter 3, the wall shear stress is briefly derived as the following [47]. Using a potential flow, the velocity potential and the velocity at the wall can be expressed as

$$\phi(x, (y, z = 0), t) = -2 \frac{\dot{R}R^2}{\sqrt{d^2 + x^2}}$$

and

$$\frac{\partial \phi}{\partial x} = U_x = 2x \frac{\dot{R}R^2}{(d^2 + x^2)^{3/2}} \quad .$$

Hence, the acceleration of the liquid at the wall is

$$\frac{\partial U_x}{\partial t} = (2\ddot{R}R^2 + 4\dot{R}^2R) \frac{x}{(d^2 + x^2)^{3/2}} \quad . \quad (\text{A.1})$$

The problem has some similarities with Stokes 1st Problem, however the boundary condition needs to be changed. Here the velocity is zero at the wall. Hence, the velocity component parallel to the wall for this problem is

$$v_x(x, z, t) = \int_{-\infty}^t \frac{\partial U_x}{\partial t'}(x, t') \operatorname{erf} \left(\frac{z}{2\sqrt{\nu(t-t')}} \right) dt' \quad . \quad (\text{A.2})$$

Since

$$\tau_w(x, t) = \nu \rho_L \frac{\partial v_x}{\partial z}(x, z = 0, t) \quad ,$$

and assuming boundary layer is thin at the wall, the wall shear stress can be calculated as a function of distance and time:

$$\tau_w(x, t) = \rho_L \sqrt{\frac{\nu}{\pi}} \int_{-\infty}^t \frac{\partial U_x}{\partial t'}(x, t') \frac{dt'}{\sqrt{t - t'}} \quad , \quad (\text{A.3})$$

where ρ_L is the liquid density, and ν is the liquid kinematic viscosity.

Appendix B

Linear wall shear stress

For a sufficiently small forcing amplitude, the bubble will oscillate sinusoidally. Hence, the bubble radial oscillations may be expressed as

$$R = R_0 [1 + X_m \sin(\omega t)] \quad , \quad (\text{B.1})$$

which implicitly means that the bubble will start to oscillate from its equilibrium radius then followed by expansion to its maximum radius. Analytical solution of Eq. 3.10 can be retrieved if there exist analytical solutions for the integrals

$$I_1 = \int_0^t \frac{\ddot{R}R^2}{\sqrt{t-t'}} dt' \quad (\text{B.2})$$

and

$$I_2 = \int_0^t \frac{\dot{R}^2 R}{\sqrt{t-t'}} dt' \quad . \quad (\text{B.3})$$

From Eq. B.1 one may get the numerators inside integrals I_1 and I_2

$$\ddot{R}R^2 = -R_0 X_m \omega^2 \sin(\omega t) R_0^2 [1 + X_m \sin(\omega t)]^2 \quad ,$$

$$\dot{R}^2 R = R_0^2 X_m^2 \omega^2 \cos^2(\omega t) R_0 [1 + X_m \sin(\omega t)] \quad .$$

By substituting into Eqs. B.2 and B.3 then solving analytically the integrals and putting the results in Eq. 3.10, one finally derives

$$\tau_w = \rho \sqrt{\frac{\nu}{\pi}} \frac{x}{(d^2 + x^2)^{3/2}} \{2\zeta_1 + 4\zeta_2\} \quad , \quad (\text{B.4})$$

where

$$\zeta_1 = - \left(\frac{1}{48} + \frac{i}{48} \right) \omega^{3/2} X_m R_0^3 \times \quad (\text{B.5a})$$

$$[\zeta_{11} + \zeta_{12} + \zeta_{13} + \zeta_{14} - \zeta_{15}] \quad ,$$

$$\begin{aligned} \zeta_{11} = & -3i\sqrt{2\pi} (3X_m^2 + 4) \times \\ & (\cos(\omega t) - i \sin(\omega t)) \operatorname{erf} \left((1-i) \sqrt{\frac{\omega t}{2}} \right) \quad , \end{aligned} \quad (\text{B.5b})$$

$$\begin{aligned} \zeta_{12} = & 3\sqrt{2\pi} (3X_m^2 + 4) \times \\ & (\cos(\omega t) + i \sin(\omega t)) \operatorname{erf} \left((1+i) \sqrt{\frac{\omega t}{2}} \right) \quad , \end{aligned} \quad (\text{B.5c})$$

$$\begin{aligned} \zeta_{13} = & (-2 + 2i) \sqrt{6\pi} X_m^2 \times \\ & \left\{ \cos(3\omega t) \operatorname{S} \left(\sqrt{\frac{6\omega t}{\pi}} \right) - \sin(3\omega t) \operatorname{C} \left(\sqrt{\frac{6\omega t}{\pi}} \right) \right\} \quad , \end{aligned} \quad (\text{B.5d})$$

$$\begin{aligned} \zeta_{14} = & (24 - 24i) \sqrt{\pi} X_m \times \\ & \left\{ \cos(2\omega t) \operatorname{C} \left(2\sqrt{\frac{\omega t}{\pi}} \right) + \sin(2\omega t) \operatorname{S} \left(2\sqrt{\frac{\omega t}{\pi}} \right) \right\} \quad , \end{aligned} \quad (\text{B.5e})$$

$$\zeta_{15} = (48 - 48i) X_m \sqrt{\omega t} \quad , \quad (\text{B.5f})$$

and

$$\zeta_2 = \frac{\omega^{3/2} X_m^2 R_0^3}{12} \times \quad (B.6a)$$

$$[\zeta_{21} + \zeta_{22} - \zeta_{23} - \zeta_{24} + \zeta_{25} + \zeta_{26} + \zeta_{27}] \quad ,$$

$$\zeta_{21} = 3\sqrt{2\pi} X_m \sin(\omega t) \text{ C} \left(\sqrt{\frac{2\omega t}{\pi}} \right) \quad , \quad (B.6b)$$

$$\zeta_{22} = \sqrt{6\pi} X_m \sin(3\omega t) \text{ C} \left(\sqrt{\frac{6\omega t}{\pi}} \right) \quad , \quad (B.6c)$$

$$\zeta_{23} = 3\sqrt{2\pi} X_m \cos(\omega t) \text{ S} \left(\sqrt{\frac{2\omega t}{\pi}} \right) \quad , \quad (B.6d)$$

$$\zeta_{24} = \sqrt{6\pi} X_m \cos(3\omega t) \text{ S} \left(\sqrt{\frac{6\omega t}{\pi}} \right) \quad , \quad (B.6e)$$

$$\zeta_{25} = 6\sqrt{\pi} \cos(2\omega t) \text{ C} \left(2\sqrt{\frac{\omega t}{\pi}} \right) \quad , \quad (B.6f)$$

$$\zeta_{26} = 6\sqrt{\pi} \sin(2\omega t) \text{ S} \left(2\sqrt{\frac{\omega t}{\pi}} \right) \quad , \quad (B.6g)$$

and

$$\zeta_{27} = 12\sqrt{\omega t} \quad . \quad (B.6h)$$

Note that S and C are Fresnel sine and cosine integrals, and erf is the error function [134].

Another initial condition for the bubble oscillation can be represented with:

$$R = R_0 [1 + X_m \cos(\omega t)] \quad , \quad (B.7)$$

which means that the bubble start to oscillate from its maximum radius then followed by compression to its minimum radius. Following the same derivation approach as before, one arrives at the expression for the wall shear stress:

$$\tau_w = \rho \sqrt{\frac{\nu}{\pi}} \frac{x}{(d^2 + x^2)^{3/2}} \{2\xi_1 + 4\xi_2\} \quad , \quad (\text{B.8})$$

where

$$\begin{aligned} \xi_1 &= \left(\frac{1}{48} + \frac{i}{48} \right) \omega^{3/2} X_m R_0^3 \times \\ &[\xi_{11} + \xi_{12} + \xi_{13} - \xi_{14} - \xi_{15}] \quad , \end{aligned} \quad (\text{B.9a})$$

$$\begin{aligned} \xi_{11} &= 3i\sqrt{2\pi} (3X_m^2 + 4) (\cos(\omega t) + i \sin(\omega t)) \times \\ &\quad \text{erf} \left((1+i) \sqrt{\frac{\omega t}{2}} \right) \quad , \end{aligned} \quad (\text{B.9b})$$

$$\begin{aligned} \xi_{12} &= 3\sqrt{2\pi} (3X_m^2 + 4) (\sin(\omega t) + i \cos(\omega t)) \times \\ &\quad \text{erfi} \left((1+i) \sqrt{\frac{\omega t}{2}} \right) \quad , \end{aligned} \quad (\text{B.9c})$$

$$\begin{aligned} \xi_{13} &= (-2 + 2i) \sqrt{6\pi} X_m^2 \times \\ &\quad \left\{ \cos(3\omega t) \text{C} \left(\sqrt{\frac{6\omega t}{\pi}} \right) + \sin(3\omega t) \text{S} \left(\sqrt{\frac{6\omega t}{\pi}} \right) \right\} \quad , \end{aligned} \quad (\text{B.9d})$$

$$\begin{aligned} \xi_{14} &= (24 - 24i) \sqrt{\pi} X_m \times \\ &\quad \left\{ \cos(2\omega t) \text{C} \left(2\sqrt{\frac{\omega t}{\pi}} \right) + \sin(2\omega t) \text{S} \left(2\sqrt{\frac{\omega t}{\pi}} \right) \right\} \quad , \end{aligned} \quad (\text{B.9e})$$

$$\xi_{15} = (48 - 48i) X_m \sqrt{\omega t} \quad , \quad (\text{B.9f})$$

and

$$\xi_2 = -\frac{\omega^{3/2}X_m^2R_0^3}{12} \times \quad (B.10a)$$

$$[\xi_{21} + \xi_{22} - \xi_{23} + \xi_{24} + \xi_{25} + \xi_{26} - \xi_{27}] \quad ,$$

$$\xi_{21} = -3\sqrt{2\pi}X_m \cos(\omega t) \text{ C} \left(\sqrt{\frac{2\omega t}{\pi}} \right) \quad , \quad (B.10b)$$

$$\xi_{22} = \sqrt{6\pi}X_m \cos(3\omega t) \text{ C} \left(\sqrt{\frac{6\omega t}{\pi}} \right) \quad , \quad (B.10c)$$

$$\xi_{23} = 3\sqrt{2\pi}X_m \sin(\omega t) \text{ S} \left(\sqrt{\frac{2\omega t}{\pi}} \right) \quad , \quad (B.10d)$$

$$\xi_{24} = \sqrt{6\pi}X_m \sin(3\omega t) \text{ S} \left(\sqrt{\frac{6\omega t}{\pi}} \right) \quad , \quad (B.10e)$$

$$\xi_{25} = 6\sqrt{\pi} \cos(2\omega t) \text{ C} \left(2\sqrt{\frac{\omega t}{\pi}} \right) \quad , \quad (B.10f)$$

$$\xi_{26} = 6\sqrt{\pi} \sin(2\omega t) \text{ S} \left(2\sqrt{\frac{\omega t}{\pi}} \right) \quad , \quad (B.10g)$$

and

$$\xi_{27} = 12\sqrt{\omega t} \quad . \quad (B.10h)$$

Appendix C

Numerical method

Under non-linear oscillations, the bubble radial dynamics (Eq. 3.2) first needs to be solved, e.g. using `ode45` in MATLAB. The function `ode45` is in principle a 4th order Runge-Kutta ODE solver scheme with adaptive time stepping. However, it is accepted that the bubble will oscillate linearly under sufficiently small forcing amplitude. In this forcing regime, the bubble oscillates sinusoidally [2].

After $R(t)$, $\dot{R}$ and $\ddot{R}$ are solved then they are used to calculate $\partial U_x / dt$. The liquid acceleration at wall, $\partial U_x / dt$, is then used in the wall shear stress calculation. From $t = -\infty$ to $t = 0$, shear stress is assumed to be zero as $t = 0$ is taken as the start of the insonation [135].

Calculation of wall shear stress is not straightforward since there are history terms which may lead to singularity. To overcome the problem, $\partial U_x / dt$ is assumed constant at the limit t' approaching t [135, 47]. Hence at that limit, at $t' = [t - \Delta t, t]$, the integral in Eq. 3.10 reduces into $2\dot{V}\sqrt{\Delta t}$.

Wall shear stresses calculated with analytical and numerical solutions are then compared by means of plotting, see Fig. C.1, and results in a good fit.

Figure C.1: Plot of wall shear stress calculated using analytical and numerical solution.