EXPERIMENTS IN DIFFUSIVE GAS-BUBBLE GROWTH

Oscar R. Enriquez[*a)], Devaraj van der Meer[*], Chao Sun[*], Andrea Prosperetti[*,**] & Detlef Lohse[*]
[*]*Physics of Fluids Group, Faculty of Technology, University of Twente, Enschede, OV, The Netherlands*
[**]*Department of Mechanical Engineering, Johns Hopkins University, Baltimore, MD, U.S.A.*

Summary This short paper describes a new experimental set-up for studying the growth of single and multiple gas bubbles in a supersaturated liquid. In these experiments, liquid supersaturation is carefully controlled. Results on the growth of a single bubble are compared with theory. We find that the theory accurately describes the growth of bubbles until dissolved gas concentration gradients trigger natural convection, which increases the growth rate.

INTRODUCTION

A gas dissolved in a liquid at a pressure P and temperature T will escape the solution whenever its concentration exceeds the equilibrium concentration given by Henry's law: ($c_{sat} = k_H(T)P$). The best known example of this phenomenon is perhaps the degassing of a carbonated beverage, which has been extensively studied (*e.g.* [2]). A liquid, initially saturated with CO_2 at pressure higher than atmospheric, becomes supersaturated when exposed to atmospheric pressure. Gas will then escape by diffusion through the free surface and also, when the conditions are appropriate, due to the formation and rising of small bubbles.

The main mechanism for bubble growth in this situation is the diffusion of gas through the liquid phase. The driving is provided by the supersaturation coefficient $S = (c_i - c_s)/\rho_g$, where c_i is the mass gas concentration in the liquid bulk, and c_s and ρ_g the concentration and density at the ambient pressure, respectively. The purely-diffusive growth of a gas bubble has been extensively studied theoretically [1] and numerically [3], but not so much experimentally [4]. A good understanding of the interaction of gas bubbles growing nearby each other while competing for a limited amount of dissolved gas would be very valuable for industrial applications. With a novel experimental set-up that allows for control of the supersaturation and bubble position, we study the purely-diffusive growth of gas bubbles; starting with the case of a single bubble to later expand to interacting growth.

EXPERIMENTS

The experimental setup (figure 1a) consists of two stainless-steel tanks connected between them and to water and CO_2 inlets and outlets through a series of electronically-controlled pneumatic valves. Pressure, temperature, and conductivity sensors constantly monitor the state of the system. Through the measurement of conductivity we keep track of the gas concentration. Water is saturated with CO_2 at a pressure of up to 10 bar in the larger (mixing) tank, with the aid of a magnetic stirring rod. A sample of the liquid is then transferred to a smaller (observation) tank with windows that allow optical access. The observation tank is equipped with a device that holds a square silicon chip, 1 cm by side, which has cylindrical pits, a few microns in diameter, etched into it and coated with hydrophobic material. Such pits work as nucleation sites of known size and position. The pressure in the observation tank is decreased at an accurately controlled rate (*e.g.* $0.5 - 1$ bar/h) and held at the target pressure. In order to isolate the diffusive process from effects like inertia, streaming, and convection the supersaturation coefficient S is kept small ($0 < S < 0.5$). When bubbles start growing, we image them and trace their contour to compute the radius as it evolves in time.

RESULTS

The theory [2], considering a spherical bubble of size R_0 at $t = 0$, predicts that a purely-diffusive growth rate will be described as

$$\frac{dR}{dt} = \frac{D(c_i - c_s)}{\rho_g}\left(\frac{1}{R} + \frac{1}{\sqrt{\pi D t}}\right), \qquad (1)$$

where D is the diffusivity of CO_2 in water. Laplace pressure effects can be neglected by taking a large enough R_0 ($> 10^{-5}$ m); hence, the gas concentration at the bubble surface is given by Henry's law at the system pressure and is equal to c_s. The $\sqrt{\pi D t}$ represents the width of the concentration boundary layer that grows around the bubble, and is 0 at $t = 0$.
Before studying the growth of interacting bubbles, we look at the diffusive growth of a single bubble. This case is well-understood theoretically, and works for testing whether we can indeed control the experimental parameters accurately. In

a) Corresponding author. E-mail: oscarenriquez@gmail.com

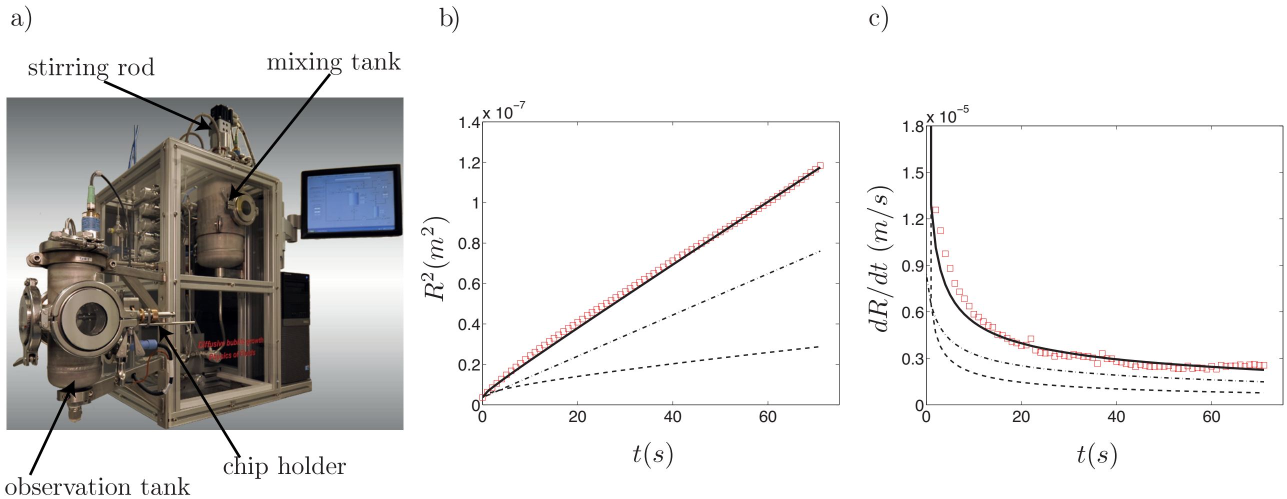

Figure 1. a) The experimental setup with its most important components indicated. b) R^2 as a function of time for one experimental realization. Symbols represent experimental data points. —: numerical solution of equation 1. -·- and — — show the influence of the first and second terms on the right-hand side of the equation, respectively. c) Equation 1 represented with the same experimental data. Symbols are the left-hand side of the equation. —: full right-hand side (RHS) of the equation. -·-: first term on RHS. — —: second term on RHS.

figures (1b) and (1c) we compare experimental observations of the growth of a single bubble with equation 1, showing the influence of each term in the right-hand side of the equation. We can see that the theory describes very well the evolution of the radius through the entire life of the bubble. Furthermore, we see that both terms in the equation play an important role, and an approximate solution neglecting one of them would not be accurate. However, for a bubble with longer lifetime we have observed the growth rate to increase considerably. We believe that this result is due to the combined influence of the deformation of the bubble due to buoyancy effects and a slight density-driven convective flow triggered by the local CO_2 depletion that lowers the liquid density. The quantitative determination of the influence of each effect will be determined in further experiments.

CONCLUSIONS

We have designed and constructed an experimental device that allows us to study the diffusive growth of gas bubbles. Our first results show that we can indeed control the experimental parameters accurately and isolate the influence of diffusion. However, we have also seen that the theory cannot describe the long-time growth rates. This is believed to be due to the onset of a slight density-driven convective flow combined with bubble deformation. We shall further deepen our understanding of these effects and extend the study to the interaction of bubbles growing near to each other.

References

[1] Liger-Belair G.: The Physics and Chemistry behind the Bubbling Properties of Champagne and Sparkling Wines: A State-of-the-Art Review. *J. Agric. Food Chem.* **53**:2788–2802, 2005.

[2] Epstein P.S. and Plesset M.S.: On the Stability of Gas Bubbles in Liquid-Gas Solution. *J. Chem. Phys.* **18**:1505–1509, 1950.

[3] Arefmanesh A., Advani S.G., and Michaelides E.E.: An Accurate Numerical Solution for Mass Diffusion-Induced Bubble Growth in Viscous Liquids Containing Limited Dissolved Gas. *Int. J. Heat Mass Transfer* **35**:1711–1722, 1992.

[4] Jones S.F., Evans G.M., and Galvin K.P.: The cycle of bubble production from a gas cavity in a supersaturated solution. *Adv. Colloid Interface Sci.* **80**:51–84, 1999.