<Supplementary Material>

Dissolution without disappearing: multicomponent gas exchange for CO_2 bubbles in a microfluidic channel

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1. Dissolution of a spherical bubble of single gas component in infinite liquid phase

Assume that there is a stationary spherical gas bubble in infinite liquid phase. Since the bubble is spherical, diffusion of gas into the liquid phase can be considered as one dimensional (radial only). Without considering surface tension and the liquid pressure change in the channel flow, the bubble radius change in time is[1],

$$\frac{da}{dt} = -\frac{D(c_s - c_\infty)}{\rho}\left\{\frac{1}{a} + \frac{1}{\sqrt{\pi D t}}\right\}. \quad \text{(S1)}$$

c_s is the mass concentration of CO_2 in the bubble, c_∞ is the initial mass concentration of CO_2 in the liquid, and ρ and D are the mass density and the diffusivity of CO_2, respectively. Together with the ideal gas law and considering surface tension and liquid pressure changes,

$$\frac{da}{dt} = -\frac{\frac{a}{3}\frac{dp_L}{dt} + DR_gT(c_s - c_\infty)\left\{\frac{1}{a} + \frac{1}{\sqrt{\pi D t}}\right\}}{\left(p_L + \frac{4\gamma}{3a}\right)}, \quad \text{(S2)}$$

and the radius change in time can be obtained.

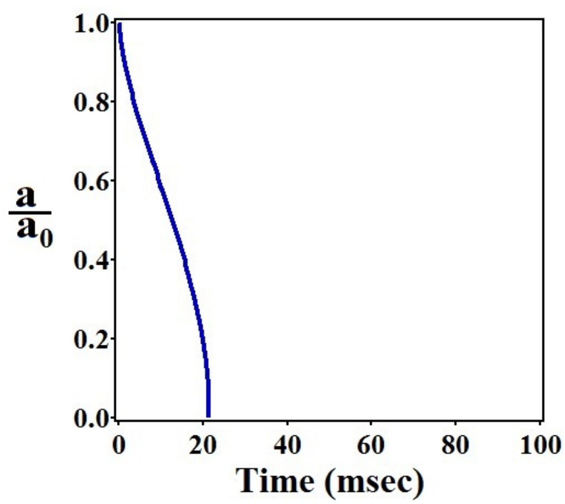

Figure S1 Radius change of a CO_2 bubble in an infinite liquid phase. When there is no other gas in the liq uid phase, a CO_2 bubble of initial radius $a_0 =$ 15 µm disappears within 20 ms.

2. Soluble amount of CO_2 in the channel

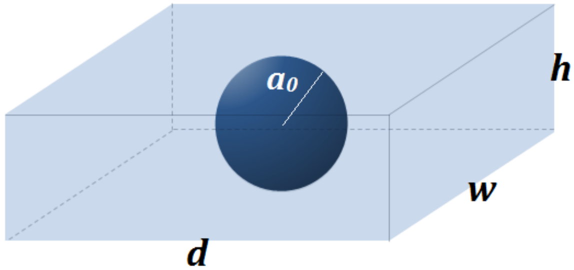

Figure S2 CO_2 bubble in a liquid box. The initial bubble radius is $a_0 = 15\ \mu m$, and the dimension of the l iquid box are $d = 150\ \mu m,\ w = 150\ \mu m,$ and $h = 38\ \mu m$ respectively.

The CO_2 solubility in pure water is 1.7 g per 1 kg of water[2]. Initially, the volume of liquid in the box (Figure S2) is

$$V_{liq} = V_{Channel} - V_{Bubble} = dhw - \frac{4}{3}\pi {a_0}^3 = 8.5 \times 10^5\ \mu m^3\,. \qquad \text{(S1)}$$

Assuming that the density of liquid phase is the pure water value, $\rho =$ 997 kg/m^3, then the initial liquid mass is

$$M_{liq} = \rho V_{liq} = 8.5\ \mu g\,, \qquad \text{(S2)}$$

and the mass of CO_2 soluble in this amount of liquid is 1.44 ng.

The initial mass of CO_2 in the bubble is $M_{CO_2} = \rho_{CO_2} V_{Bubble}$, and the density ρ_{CO_2} can be obtain ed from the ideal gas law. When the liquid flow rate in the channel is Q = 25 μL/min, the initial p ressure in the bubble is

$$p_{Bubble}(0) = p_L(0) + \frac{2\gamma}{a_0} = -\frac{\mu Q \beta L}{wh^3} + p_{atm} + \frac{2\gamma}{a_0} = 1.29 \times 10^5\ Pa\,, \qquad \text{(S3)}$$

where μ is the viscosity of the liquid and β is a constant that can be determined by the geometry of the cross-section of the channel. $p_L(0)$ in equation (S3) will be explained in the next section. The corresponding density of CO_2 in the bubble is

$$\rho_{CO_2} = p_{Bubble}(0)\frac{M_w}{R_g T} = 0.1 kg/m^3 = 10^{-10}\ \mu g/\mu m^3\,. \qquad \text{(S4)}$$

Therefore, the initial mass of CO_2 in the system is

$$\therefore M_{CO_2} = \rho_{CO_2} V_{Bubble} = 1.43 \times 10^{-12}\ g\,, \tag{S5}$$

which is ~10^{-3} of the soluble CO_2 mass in the liquid phase. Since the soluble mass is much larger than actual mass of CO_2 in the bubble, no saturation is expected in the microfluidic channel.

3. Liquid pressure in the channel flow

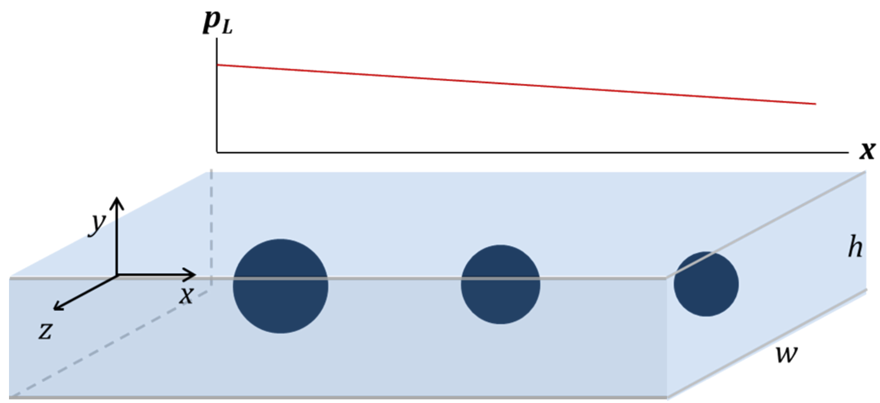

Figure S3 Flow in the rectangular channel. Liquid pressure gradient is constant along the channel, and the width and height of the channel are $w = 150\ \mu m$, and $h = 38\ \mu m$ respectively. Also, the origin of the axes is at the center of the cross-section of the channel.

In our microfluidic channel, liquid pressure gradient per unit length is constant. Therefore, liquid flow in such rectangular channel follows

$$\mu\left(\frac{\partial^2 u}{\partial y^2} + \frac{\partial^2 u}{\partial z^2}\right) = \frac{dp_L}{dx}\,, \tag{S6}$$

where μ, u, and p_L are, respectively, liquid viscosity, flow velocity, and liquid pressure.

Solving the partial differential equation with no-slip boundary conditions gives the velocity profi le of the liquid flow, and thus the corresponding flow rate Q for $0 \leq h/w \leq 1$ can be written as[3]

$$Q = -\frac{wh^3}{12\mu} \cdot \frac{dp_L}{dx}\left[1 - \frac{6(2^5)}{\pi^5}\left(\frac{h}{w}\right)\right]. \tag{S7}$$

Let $\langle v \rangle$ the average velocity of the flow, then

$$\langle v \rangle = \frac{Q}{wh}, \tag{S8}$$

and

$$\frac{dp_L}{dx} = \frac{1}{\langle v \rangle}\frac{dp_L}{dt} = \frac{wh}{Q}\frac{dp_L}{dt}. \tag{S9}$$

Substituting Q in equation (S9) with (S7) gives,

$$\frac{dp_L}{dt} = \frac{12\mu Q^2}{w^2h^4} \cdot \frac{1}{\left[\frac{6(2^5)}{\pi^5}\left(\frac{h}{w}\right) - 1\right]} = \frac{\beta\mu Q^2}{w^2h^4}. \tag{S10}$$

Given that the channel length is L, the liquid pressure can be written as

$$p_L = \frac{dp_L}{dx}(x - L) + p_{atm} = \frac{dp_L}{dt}\left(t - \frac{L}{\langle v \rangle}\right) + p_{atm}, \tag{S11}$$

where p_{atm} is the atmospheric pressure at the outlet of the channel. Therefore, substituting equati on (S10) into (S11) gives the liquid pressure equation:

$$p_L = \frac{\beta\,\mu Q^2}{w^2h^4}\left(t - \frac{Lwh}{Q}\right) + p_{atm}. \tag{S12}$$

References

1. P. B. Duncan and D. needham, *Langmuir*, 2004, **20**, 2567-2578.
2. N. A. Lange, *Handbook of Chemistry*, McGraw-Hill, New York, 10th edn., 1967.
3. F. M. White, *Viscous Fluid Flow*, McGraw-Hill, New York, 2nd edn., 1991.