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Lifetime of microbubble-an interpretation

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Abstract : Microbubbles are miniature gas bubbles of less than 100 microns diameter in liquid. Microbubbles provides high gas dissolution, larger gas-liquid interfacial area and longer residence time. A theoretical interpretation of the lifetime of microbubble is presented in the present work. The classical Epstein and Plesset model is used to interpret the lifetime of microbubble. The influence of microbubble diameter and surface tension on microbubble lifetime are also analyzed. A comparative comparison of microbubble lifetime of differ-

ent gas microbubbles is also presented in this work. The minimum column length required to design an efficient microbubble column is also enunciated. It is observed that the lifetime as well as the minimum column length is significant based on the physiochemical property of liquid and microbubble, which is important to design the plant prototype for chemical process. © Global Scientific Inc.

Keywords : Dissolution; Diffusion; Life time; Microbubble; Minimum column length.

INTRODUCTION

In recent times, microbubble are gaining attention in scientific community. Microbubble offers large interfacial area than conventional bubbles. The physicochemical properties of microbubble like high inertial pressure, high gas-liquid interfacial area, negatively charged surface and high gas dissolution rate makes them extremely suitable for various engineering and medical applications^[1,2]. The charge on surface of microbubble is favourable to separate the opposite charge particles in mineral separation units. Microbubbles have also been used as a source for extreme temperatures in sonochemical reactions and to increase oxygen delivery in fermen-

tation process^[3,4]. Microbubble are used to drive mixing on a chip in microelectromechanical devices^[5]. Microbubbles are being developed to use as ultrasound contrast, drug delivery agents^[6]. When considering the industrial applications of microbubble, it is important to evaluate the benefits based on scientific principles, from an academic standpoint and to compare microbubble technology with existing technology both in terms of its functional quality and effectiveness. Thus it becomes very necessary to examine clearly the superior physical characteristics like dissolution, residence time and lifetime of microbubbles. The residence time of the microbubble is defined as total time that microbubble spent in the liquid, where lifetime of microbubble is the amount of time required for microbubble to com-

pletely dissolve in liquid. The magnitude of life time of microbubble depends on stability or diffusion of solute component of microbubble. The stability can be significantly enhanced by coating with surfactant^[7]. Presence of electrolyte in liquid can reduce the stability, whereas the effects of pH on microbubble stability have been inconsistent^[8]. Many authors Epstein and Plesset^[9], developed theoretical model to analyse the stability of gas bubble in liquid media based on diffusion mechanism with/without hydrostatic conditions. Readey and Cooper^[10] provided a numerical solution of dissolution of bubble in the absence of hydrodynamic instabilities. More accurate theoretical analysis, which takes into account the effect of motion of liquid was presented by Cable^[11] and Tao^[12]. The authors also established a relation for the life time of bubble. As per investigation by Weinberg^[13], the effect of surface tension on the rate of dissolution of an isolated single component stationary bubble is significant. Engelking^[14] derived an analytical expression for life time of metastable microbubbles. Ljunggren and Eriksson^[15] derived an expression for lifetime of colloidal-size bubble as a function of its radius. Several other studies are also available in literature^[16-21]. There is no work on the lifetime of microbubble based on diffusion and the residence time of microbubble to interpret the geometry of column for a chemical process. In the present work, therefore to understand the influence of microbubble gas diffusion on the stability and the residence time of microbubble, a theoretical interpretation has presented. The effect of physiochemical properties of encapsulated gas on the geometry of microbubble-induced column is also enunciated.

MATHEMATICAL FORMULATION

Lifetime of a microbubble

The stability of microbubble can be expressed in terms of its life time. Lifetime is a magnitude of time spent by bubble till it completely dissolves in liquid. In the present study, Epstein and Plesset model is used to calculate lifetime of microbubble^[9]. Let us suppose that at any time, t , a microbubble of initial radius r_0 is placed in large volume of liquid in an isothermal condition. The Young–Laplace equation for a microbubble can be written as:

$$p = p_0 + \frac{2\sigma}{r} \quad (1)$$

where p and σ denotes the excess pressure inside the bubble and surface tension respectively. p_0 represents atmospheric pressure. The parameter r is time dependant radius of the microbubble. The ideal gas equation for a microbubble yields the following equation:

$$p = \frac{\rho(r)RT}{M_g} \quad (2)$$

where $\rho(r)$ is density in the bubble, M_g is molecular weight of gas. R is universal gas constant and T is temperature. Substituting the value of p from equation (2) into equation (1), one gets

$$\left(p_0 + \frac{2\sigma}{r}\right) = \frac{\rho(r)RT}{M_g} \quad (3)$$

On rearranging the equation (3), it becomes

$$\rho(r) = \frac{M_g p_0}{RT} + \frac{2M_g \sigma}{RT r} \quad (4)$$

$$\rho(r) = \rho_g + \frac{2M_g \sigma}{RT r} \quad (5)$$

where ρ_g is density of gas under the same condition of pressure and temperature. The concentration of gas depends on time (t) and position (r) in the liquid. If we assume that the dissolution of gas is mainly governed by convective diffusion, then the transport of gas in the liquid can be given as

$$\frac{\partial c}{\partial t} = D \left(\frac{2}{r} \frac{\partial c}{\partial r} + \frac{\partial^2 c}{\partial r^2} \right) \quad (6)$$

where D is diffusion coefficient of gas in liquid. The solution of equation (6) with boundary conditions

$$c(r, 0) = c_0, r > r; \quad \lim_{r \rightarrow \infty} c(r, t) = c_0, t > 0$$

and $c(r, t) = c_r, t > 0$ can be written as

$$\left(\frac{\partial c}{\partial r} \right)_{r=r} = (c_0 - c_r) \left(\frac{1}{r} + \frac{1}{\sqrt{\pi D t}} \right) \quad (7)$$

The mass flux of bubble dissolving in the liquid can be expressed as

$$j = -\rho(r) \frac{\partial r}{\partial t} \quad (8)$$

From Fick's first law of diffusion

$$j = -D \frac{\partial c}{\partial t} \quad (9)$$

where D is diffusivity of gas in liquid. Thus from eqs. (6), (8) and (9), one can get the dissolution rate of microbubble as

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$$\frac{dr}{dt} = -\frac{D(c_r - c_0)}{\rho(r)} \left[\frac{1}{r} + \frac{1}{\sqrt{\pi Dt}} \right] \quad (10)$$

Now, substituting value of $\rho(r)$ from equation (5) in equation (10) one can get,

$$\frac{dr}{dt} = -D \left(\rho_g + \frac{2M_g \sigma}{RT r} \right)^{-1} (c_r - c_0) \left[\frac{1}{r} + \frac{1}{\sqrt{\pi Dt}} \right] \quad (11)$$

If microbubble tends to totally dissolve, then

$\frac{1}{\sqrt{\pi Dt}}$ is negligible as compared to $\frac{1}{r}$. Therefore equation (11) can further be reduced to

$$\frac{dr}{dt} = -D \left(\rho_g + \frac{2M_g \sigma}{RT r} \right)^{-1} (c_r - c_0) \left(\frac{1}{r} \right) \quad (12)$$

For convenience, equation (12) can be further modified by substituting $f = c_r/c_0$. If Henry's law is assumed to hold in this microscale system then, $K_h = c_r/p_0 M_g$ and $c_s/\rho_g = K_h RT^{[22]}$. For a finite surface tension (independent of radius) equation (12) becomes

$$\frac{dr}{dt} = -DK_h RT \left(1 - f + \frac{2M_g \sigma}{\rho RT r} \right) \left(1 + \frac{4M_g \sigma}{3\rho RT r} \right)^{-1} r^{-1} \quad (13)$$

Integrating equation (13) for air saturated solution (i.e. $f = 1$) with boundary condition $r = d_b/2$ at $t = 0$ and $r = 0$ at $t = \tau_{b-1}$ gives the life span of microbubble as

$$\tau_{b-1} = \frac{d_b^2}{12DK_h} \left[\frac{d_b \rho_g}{4M_g \sigma} + \frac{1}{RT} \right] \quad (14)$$

where τ_{b-1} represents lifetime of microbubble. In the present work six different gases were considered for analysis. The details of the physicochemical properties of gas and liquid are given in TABLE 1 and 2 respectively.

tively.

[†]Density of gases is taken from Perry and Green^[29]. [#]Diffusivity of gases in water is taken from Cussler^[30]. [§]Henry law constant for gases in water is taken from Battino and Clever^[31]. [§]The physicochemical properties of CO₂ in Sodium Carboxymethyl Cellulose (SCMC) is taken from Tan and Thorpe^[32].

RESULTS AND DISCUSSION

In this section the effect of different physicochemical properties of liquid on the lifetime of microbubble as well as the minimum column length is analyzed. It is clear from the equation (14) that the lifetime of microbubble is a function of gas diffusivity, molecular weight of gas, interfacial tension and properties of liquid. So it is very important to completely analyze the dissolution phenomena of microbubble to interpret life time of microbubbles.

Effect of microbubble diameter on the lifetime

The effect of diameter within the range 0 to 100 μm , on lifetime of microbubble is shown in Figure 1. It is seen that microbubble lifetime increases on increasing the diameter of microbubble. The size of a gas bubble in a liquid is control by flux of dissolved gas from the surface of bubble or toward the surface. It also depends on hydrostatic pressure and the partial pressure of gas in equilibrium with the concentration of gas dissolved in the liquid. In case, the

TABLE 1 : Physicochemical properties of gases

Gas	Density [†] (kg/m ³)	Diffusivity in water [#] (m ² /s)	Henry law constant in water [§]
O ₂	1.308	2.000 × 10 ⁻⁹	1.283 × 10 ⁻⁵
H ₂	0.082	4.500 × 10 ⁻⁹	7.698 × 10 ⁻⁶
CO ₂	1.808	1.920 × 10 ⁻⁹	3.356 × 10 ⁻⁴
N ₂	1.145	1.880 × 10 ⁻⁹	6.020 × 10 ⁻⁶
Ar	1.633	2.020 × 10 ⁻⁹	1.382 × 10 ⁻⁶
CO	1.145	2.030 × 10 ⁻⁹	9.376 × 10 ⁻⁶

TABLE 2 : Physicochemical properties of liquids[§]

Solution	Concentration (kg/m ³)	Density (kg/m ³)	Diffusivity (m ² /s)	Henry law constant
SCMC-1	0.25	1000.085	1.810 × 10 ⁻⁹	3.350 × 10 ⁻⁴
SCMC-2	0.50	1000.370	1.700 × 10 ⁻⁹	3.340 × 10 ⁻⁴
SCMC-3	0.75	1000.370	1.600 × 10 ⁻⁹	3.320 × 10 ⁻⁴
SCMC-4	1.00	1000.940	1.490 × 10 ⁻⁹	3.120 × 10 ⁻⁴

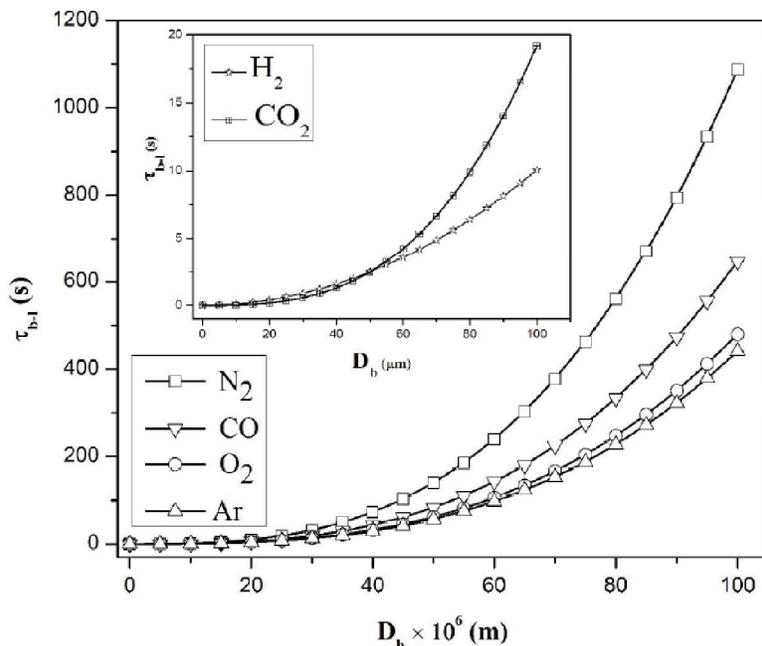


Figure 1 : Variation of lifetime of microbubbles with diameter for different gases in water

internal pressure of the bubble is lower than surrounding hydrostatic pressure, the size of bubble increases as the gas enters the bubble from the liquid. However due to their small size, the pressure inside the core region of microbubble is very high.

As the internal pressure is higher than the surrounding hydrostatic pressure, the microbubble dissolves in the liquid and shrinks. Microbubble lifetime is recognized by the length of time it takes either to dissolve or to be transported out of the solution. The pressure difference between the inside and the outside of a bubble (Laplace pressure) depends on the diameter of bubble. As the diameter of microbubble increases the Laplace pressure decrease. As the size of microbubble decreases, the chemical potential of gaseous component inside the microbubble becomes higher than the surrounding liquid, due to high internal pressure, which results in high dissolving of gas from inside bubble to the surrounding liquid. Hence the lifetime of microbubble decreases. It is also observed from the figure that life time of microbubble is significantly affected by the properties of gas. For same liquid and same diameter range, the theoretical lifetime of microbubble is different for different gases. Among all the six gases (Hydrogen, carbon dioxide, nitrogen, carbon monoxide, oxygen, argon) analysed theoretically, it is found that the nitrogen microbubble have the

maximum lifetime while H_2 microbubble has the least lifetime. The lifetime of nitrogen microbubble is approximately hundredfold higher than the H_2 microbubble. The lifetime of CO_2 microbubble is also very less. The diffusivity of the CO_2 in water is very high as compared to the other gases.

It is reported by previous studies that the dissolution of CO_2 in water form carbonic acid with water. However such chemical reaction is not observed in other gases^[23]. This causes the reduction of viscosity of the liquid at the interface, which reduces the resistance for gas to dissolve in liquid. Therefore the lifetime of CO_2 microbubble is too short. The dissolution of microbubble also depends on the liquid properties. Figure 2 shows, the effect of sodium carboxymethyl cellulose (SCMC) on life time of CO_2 -microbubble. The transfer rate of gas from bubble to liquid decreases with increasing viscosity and this can be attributed both to the reduction in mass diffusivity of gas and mixing occurring in the liquid phase. It is also observed that on increasing SCMC concentration the life time increases. Increase in viscosity decreases the diffusional between the bubble and liquid which causes the increases in life time of microbubble.

Effect of surface tension the lifetime of microbubble

The stability of microbubble depends upon its

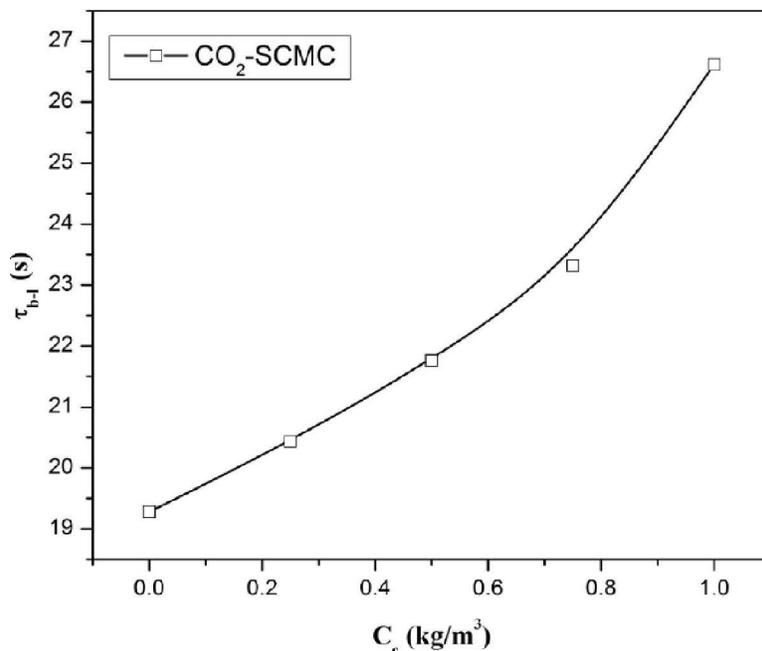


Figure 2 : Variation of life time of microbubble for CO₂ with SCMC concentration

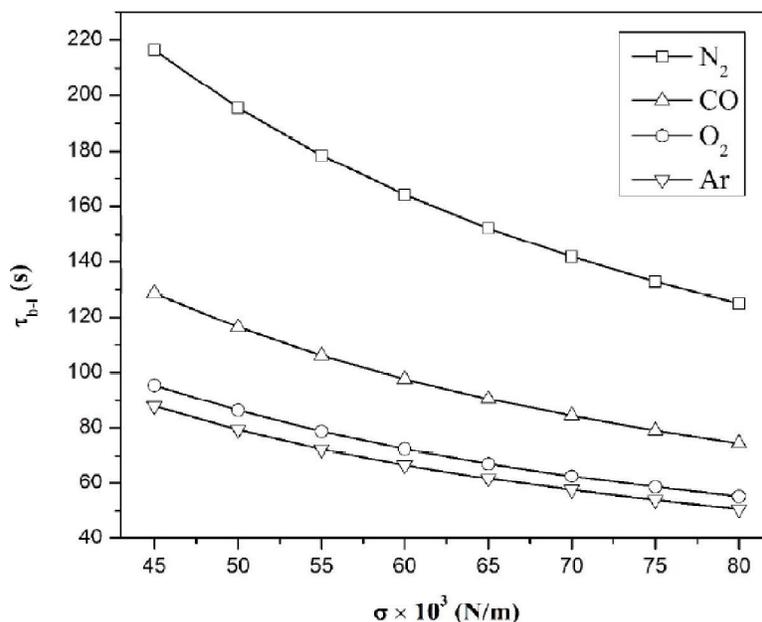


Figure 3 : Variation of lifetime of microbubbles with surface tension for different gases in water

dissolution characteristics. The dissolution time of microbubble is a function of bubble diameter, diffusivity of gas, density of gas etc. However, other parameters such as surfactant concentration, electrolyte concentration and pH also affect the dissolution.

Figure 3, shows the effect of surface tension on life time of microbubble for different gases. It is observed that the life time of microbubble decreases with increase in surface tension of liquid. Generally surfactant are

added in liquid to reduce the surface tension of liquid. Surface tension of water normally decreases with increasing surfactant concentration upto critical micelle concentration (CMC) and then remains constant with adding more surfactant^[24]. Addition of surfactant in water significantly effects the dissolution time of bubble. In general surfactants easily adsorbed on the surface of bubble, increases the diffusion barrier and decreases the dissolution rate, which ultimately increases the life-time. It is very difficult to produce high concentrated

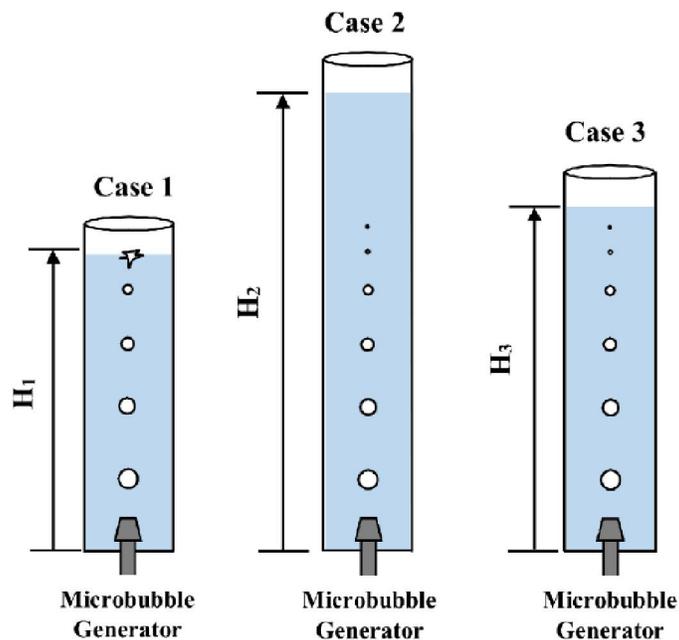


Figure 4 : Different cases of microbubble dissolution and column height (H)

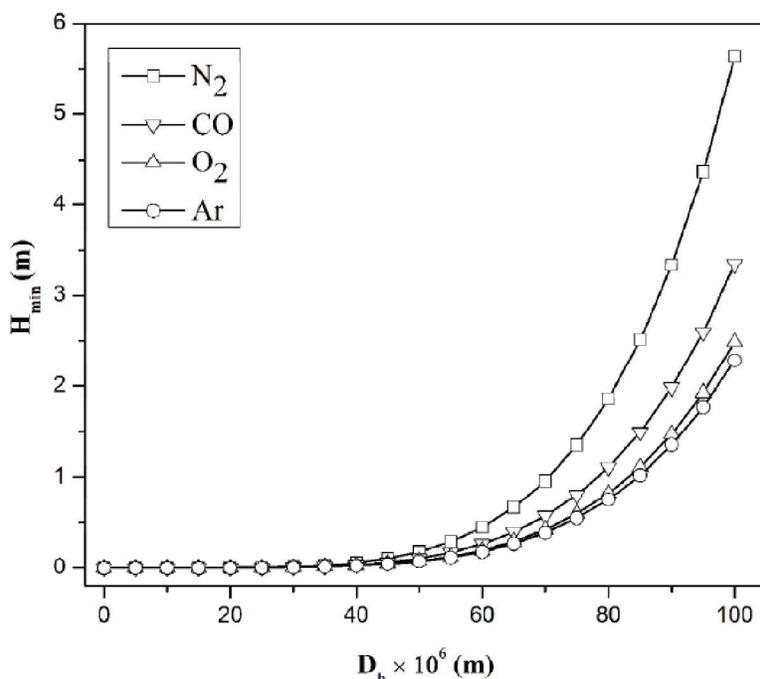


Figure 5 : Variation of minimum column length with microbubble diameter for different gases in water

microbubble in the pure liquid, so a minimum concentration of surfactant is essential for microbubble formation with good amount of microbubbles.

However, beyond CMC size and stability of microbubbles does not depend on concentration. The most stable microbubbles are produced when the concentration of surfactant is equal to critical micelle concentration^[25]. Soetanto and Chan^[26] observed that addition of surfactant can enhance life time of

bubble. Fenget al.^[8] also reported that increasing surfactant concentration increases life time. Weinberg^[13] reported that surface tension plays a prominent role in dissolution process of small bubbles. He observed that surface tension effect reduces the bubble dissolution time. Kwan and Borden^[17] examined the dissolution of SDS coated microbubble and observed that surfactant coated microbubble has strong effect on reaction of microbubble. They observed that the SDS coated

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microbubble dissolved as per Epstein and Plesset model, but lipid coated microbubble deviates significantly. The stability of microbubble can be increased by surface deposition of particle. The surface deposition can make microbubble stable up to three days.

Minimum column length (H_{\min})

When the bubble size reaches to micro-scale, rate of dissolution increases because the surface area and internal pressure of bubble increases. Ohnari^[27] described the shrinking process of microbubbles in this regard. The internal pressure of microbubble is increased as the radius decreases. Due to this high pressure inside the bubble, gas tends to diffuse outside from a region of high pressure to a low pressure of surrounding. As a consequence of this, the microbubbles shrink and finally collapse. Microbubbles are often sparged in column to enhance the gas to liquid mass transfer^[28]. Due to high dissolution it is very essential to design the microbubble-aided column properly, otherwise the efficiency as well as the yield of the process may be affected. Residence time (τ_{b-r}) and microbubble lifetime (τ_{b-l}) are two important variables that has to keep in mind for proper design of column. Suppose a microbubble rising in liquid column and at the same time the microbubble dissolves in the liquid due to its high internal pressure. Three cases as shown Figure 4, can be explained as follows:

Case 1: $\tau_{b-r} \ll \tau_{b-l}$

Under this condition the microbubble will rise slowly due to buoyancy and before being fully dissolved in the liquid, it will reach to top of the column of length H_1 and will burst in the open atmosphere. As the gas inside the microbubble is not fully utilised in the liquid so this will reduce the efficiency in term of gas utilization. Therefore this case is not favourable in designing an efficient column.

Case 2: $\tau_{b-l} < \tau_{b-r}$

Under this condition the microbubble will fully dissolved in the liquid, before reaching the top of the column. If the column is very high and the microbubble is dissolved within the half or three fourth of column length (H_2). Then this case is also not favourable in term of designing, because in the lower

section the liquid will be saturated with the gas but in the top section the concentration of gas will be almost zero. Another drawback is that the extra height will increase the pressure drop and manufacturing cost of the column.

Case 3: $\tau_{b-l} \approx \tau_{b-r}$

In this case the microbubble will fully dissolved in the liquid, just before reaching the top of the column. The gas inside the microbubble will be utilised fully. The concentration of dissolving gas will be uniform through the column. This is most favourable condition in term of operating and manufacturing cost. So the microbubble column should be designed in such a way that the lifetime and residence time are approximately equal. If microbubble are generated at the bottom of column, it will rise due buoyancy force as shown in Figure 4. If it is assumed that the motion of rising microbubble is described by the Stokes' law, ignoring the effect of reduction of diameter, then the rise velocity (U_b) of microbubble can be expressed a

$$U_b = \frac{(\rho_l - \rho_g)gd_b^2}{18\mu_1} \quad (15)$$

where g and μ are the acceleration due to gravity and viscosity of liquid respectively. In order to completely utilise the dissolving microbubble, the length of the column should be taken in such a way that the microbubble completely dissolves in the liquid. The minimum column length will be equal to the maximum distance travelled by the microbubble till it completely dissolves. So the minimum column length can be expressed as

$$H_{\min} \approx U_b \cdot \tau_{b-l} \quad (16)$$

$$H_{\min} \approx \frac{gd_b^4(\rho_l - \rho_g)}{216DK_l\mu_1} \left[\frac{d_b\rho_g}{4M_g\sigma} + \frac{1}{RT} \right] \quad (17)$$

The effect of microbubble diameter on the minimum column length required is shown in Figure 5.

It is observed that minimum column length is significant based on by the diameter of microbubble as well as physiochemical properties of gas. Within a short range of diameter (0 to 100 μm), minimum column length increases extremely. So for proper operation of microbubble-aided bubble column, the range of microbubble should be as minimum as possible. The range of bubble diameter can be reduced by addition a

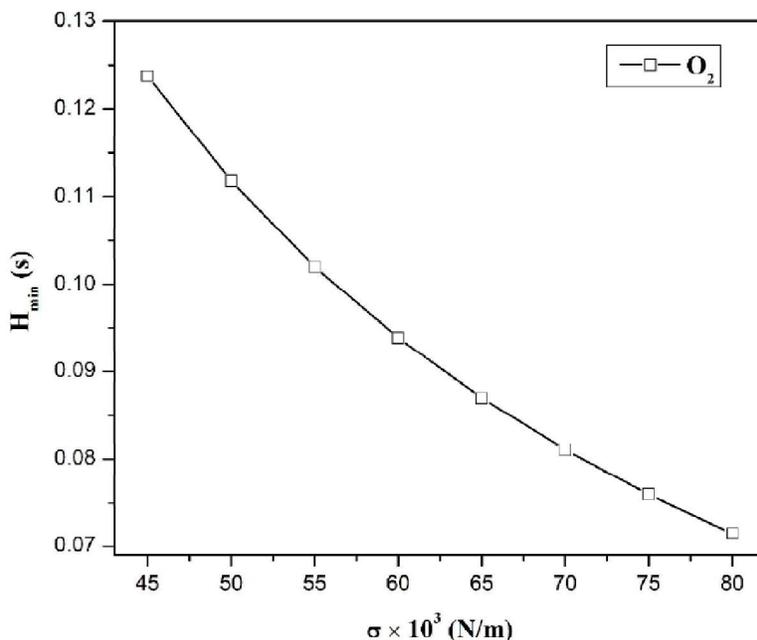


Figure 6 : Variation of minimum column length with surface tension for oxygen

proper additives or surface tension modifier. However addition of surface tension modifier or surfactant can significantly increase the stability of microbubble, which in turn increases the column length. The effect of surface tension on the minimum column length on a 50 μm O_2 microbubble is shown in Figure 6. It is clear from the figure that the minimum column length for microbubble increases with decreasing surface tension. So for designing an efficient column, one has to use a proper concentration of surfactant so that both problem can be overcome.

CONCLUSION

The present work theoretically investigates the lifetime and dissolution characteristics of microbubble. The Epstein and Plesset model was used to derive the lifetime of microbubble. It is found that the model can significantly determine the lifetime of microbubbles. The life time of microbubble is significantly affected by liquid as well as gas properties. Among all the gases (N_2 , CO , CO_2 , O_2 , H_2 and Ar) investigated theoretically, nitrogen has the highest lifetime or most stable in water, whereas H_2 has the least stable or minimum lifetime. The life time of microbubble was found to be decreased with increase in surface tension of liquid. The minimum column length, to be used for proper design of microbubble

column, which is affected by microbubble stability. The minimum column length required to completely dissolve a nitrogen microbubble of 100 μm is approximately equals to 5.6 m where for the same diameter CO_2 microbubble it requires only 0.1 m. The present study may be useful for further understanding and designing the microbubble aided process intensifications in various chemical process in this regard.

NOTATIONS

- c Concentration (mol/l)
- d_b Microbubble diameter (m)
- D Diffusion coefficient of gas in liquid (m^2/s)
- g Acceleration due to gravity (m^2/s)
- H_{min} Minimum height of column (m)
- Mg Molecular weight of gas (kg/kg mol)
- p Excess pressure inside the bubble (N/m^2)
- p_0 Atmospheric pressure (N/m^2)
- r Time dependent radius (m)
- R Universal gas constant (J/mol.K)
- T Temperature (K)
- U_b Rise velocity of microbubble (m/s)

Greek letters

- σ Surface tension (N/m)
- ρ_g Density of gas (kg/m^3)
- ρ_l Density of liquid (kg/m^3)

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K_h	Henry's law constant (mol/J)
τ_{b-l}	Lifetime of microbubble (s)
τ_{b-r}	Residence time (s)
μ_l	Viscosity of liquid (kg/s·m)

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