PROBLEM OF DISSOLVING GAS BUBBLES IN A LIQUID

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UDC 532.73-1

The process of gas diffusion from bubbles, rising freely in a liquid, is investigated. The dependencies of the bubble radius on time and the coordinates are given.

The process of escape and dissolution of a gas rising in a liquid has been studied by a number of investigators both in the general formulation and in relation to individual problems of chemical technology, heat energetics, and other branches of engineering.*

The gas usually escapes from nozzles, perforated plates, or fine porous (for example, metallic–ceramic) membranes. Here different escape regimes can occur: jet ($Re_{ap} > 7 \cdot 10^3$), intermediate ($Re_{ap} = 2 \cdot 10^3$–$7 \cdot 10^3$), and bubble ($Re_{ap} < 2 \cdot 10^3$). However, in all cases (even at a small distance from the discharge apertures) the jets and bubbles of large dimensions lose stability and break up into smaller bubbles. According to experimental data air jets while escaping in outflowing into the water disintegrate into bubbles of diameter from 0.02 to 1.20 cm at distances on the order 100 mm above the aperture [2].

The critical radius of the bubble at which it starts disintegrating is [3]

$$r_{cr} \simeq \left(\frac{3}{c_f}\right)^{\frac{1}{3}} \frac{\sigma}{v^2 (\rho_l^2 \rho_g)^{\frac{1}{3}}}. \quad (1)$$

Therefore, in analyzing the process dissolving gases in a liquid in studies of an applied nature the elementary act of this process is most often investigated, i.e., the diffusion of the gas from individual bubbles rising under the action of bouyancy. The following assumptions are usually made [1]: the constancy of the bubble volume decreasing due to gas dissolution and increasing due to a hydrostatic pressure decrease; constancy of the gas density in the bubble and the rate of rise of the bubble; constancy of the concentration pressure.

Besides, if the bubbles contain different gases, then the approximation of their independent diffusion is used.

These simplifications are not always justified.

If an exact determination of the dimensions of the bubbles in different segments of high absorption columns is required, then in computing escape of the gas into a water tank of great depth or in computing bubbles of small dimensions the assumptions about the constancy of the volume and the rate of rise of the bubble (accordingly, also the dissolution rate) and also the constancy of the gas density in the bubble can lead to appreciable errors.

According to experimental data of a number of workers [4, 5] the rate of free rise of gas bubbles of radius larger than 1 mm is practically constant.†

The rate of rise of bubbles with $r > 0.2$ mm is linearly related to the radius. In the problem of rise of bubbles with $r < 0.2$ mm there have been disagreements between the theory and experiment, which are explained in [3] by the affect of bubbles of surface active materials, which are contained in impure liquids, on the hydrodynamic boundary layer. According to recent experimental data (for example, see [6]) the motion of bubbles for $Re_b = 2$–40 obeys the laws obtained for falling spherical liquid drops. The rate of rise of bubbles is proportional to the square of the radius.

*See bibliography in [1].

†The rate of rise of air bubbles $r > 1$ mm in water is about 30 cm/sec.

Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 34, No. 2, pp. 292-300, February, 1978. Original article submitted December 6, 1976.

TABLE 1. The Phase Equilibrium Coefficient (values of m for aqueous solutions in bar)

Gas	Temperature, °C	
	0°	20°
Hydrogen	58700	69300
Air	42400	62700
Oxygen	25700	40500
Nitrogen	53600	81500
Carbonic acid	737	1440

The validity of the assumption that $\Delta c = \text{const}$ depends on the specific conditions of the problem. Usually this condition is associated with the smallness of pressure change during the rise of the bubble or the proportionality of the gas density dissolved in the liquid to the pressure (the Henry's law). Generally speaking these conditions cannot occur (for example, in columns of large height).

The approximation of independent diffusion is applicable for binary diffusion in cases of dilute mixtures and for equal diffusion coefficients of the components [7].

We consider the problem of the rising and dissolving of a gas bubble in a thick layer of liquid taking into account the change in the rate of its rise, dimensions, and pressure.

The statics of the dissolution are estimated by the phase equilibrium coefficient m whose values for certain gases are given in Table 1 [8].

The kinetics of the process are determined by the mass-transfer coefficient K

$$\frac{1}{K} = \frac{1}{\beta_g} + \frac{1}{\beta_{g.l}} + \frac{m}{\beta_l}. \tag{2}$$

Only gases with $m < 1$ (for example, NH_3, HCl) are relatively easily dissolvable. For the majority of gases which are of practical interest, $m/\beta_l \gg 1/\beta_g$ and the resistance to mass transfer in the gaseous phase can be neglected. According to experimental data on dissolving O_2 and CO_2 in water the coefficient $\beta_{g.l}$, characterizing the resistance at the phase-separation boundary for liquids not subjected to special purifying, reaches relatively large values (0.3-4.0 cm/sec).

For the discussion presented below it is assumed that the kinetics of gas dissolution determined only by the mass transfer in the liquid and the problem is solved in the approximation of the diffusion boundary layer.

The following assumptions are made: Gas dissolution obeys Henry's law and its state is described by the equation of state of an ideal gas; the gas and liquid temperature on the entire path of the rise of the bubble and the kinetic diffusion coefficient are constant.

According to the experimental data presented above the entire process of rise of a gas bubble of subcritical size can be divided into four stages, each of which is characterized by a certain rate of rise as function of the bubble radius: for $1\text{ mm} \le r \le r_{cr}$, $v = \text{const} = k_1$; for $0.2\text{ mm} \le r \le 1\text{ mm}$, $v = k_2 r$; for $r_m \le r \le 0.2\text{ mm}$, $v = k_3 r^2$; for $r_m \ge r$, $v = 0$ (here r_m is the radius of the nonrising bubble).

Using this approximation we can obtain an equation of the type $r = r(t)$ or $r = r(H)$ for all stages of rise of the bubble, from which we can find such parameters as the lifetime of the bubble, the path of rise along the vertical, etc.

Applying the law of conservation of mass to a single gas bubble we have

$$dM = -I dt, \tag{3}$$

where the diffusion flux is obtained by integrating the equation of stationary convective diffusion. During the rise of the bubble in an unperturbed liquid [3]

$$I = 8\sqrt{\frac{\pi}{2}}(Dv)^{\frac{1}{2}} r^{\frac{3}{2}} (c_{eq} - c_\infty). \tag{4}$$

The mass of the gas bubble is equal to

$$M = \frac{4}{3}\pi\rho r^3. \tag{5}$$

In accordance with the assumptions made above we have

$$\rho = \frac{\rho_0}{H_0} H, \tag{6}$$

$$c_{eq} = \frac{c_{eq0}}{H_0} H, \tag{7}$$

and the vertical coordinate can be expressed in the form

$$H = H_0 - vt, \tag{8}$$

so that

$$dM = \frac{4}{3}\pi \frac{\rho_0}{H_0}[3(H_0 - vt) r^2 dr - vr^3 dt]. \tag{9}$$

The concentration of the gas dissolved in the liquid away from the bubble is generally an arbitrary function of the coordinate H. However, for most cases of practical interest it can be described by the formulas

$$c_\infty = \frac{c_{\infty 0}}{H_0} H, \tag{10}$$

where

$$c_\infty = \text{const} = c'. \tag{11}$$

In the first stage of rise ($v = k_1$) substituting the values of c_p and c_∞, determined from formulas (7) and (10), respectively, into Eq.(4) we get

$$I = 8\sqrt{\frac{\pi}{2}}(Dk_1)^{\frac{1}{2}} r^{\frac{3}{2}} \frac{H_1 - k_1 t}{H_1}(c_{eq1} - c_{\infty 1}). \tag{12}$$

Substituting (9) and (12) into Eq. (3) we obtain the differential equation for the change of bubble radius with time

$$\frac{dr}{dt} + \frac{1}{3}\cdot\frac{k_1}{k_1 t - H_1} r = -\sqrt{\frac{2}{\pi}}(Dk_1)^{\frac{1}{2}} \frac{c_{eq1} - c_{\infty 1}}{\rho_1} r^{-\frac{1}{2}}, \tag{13}$$

which is an equation of Bernoulli type. Its general integral is

$$r = \left\{\left(\frac{k_1}{H_1 - k_1 t}\right)^{\frac{1}{2}}\left[C + \sqrt{\frac{2}{\pi}}(Dk_1)^{\frac{1}{2}} \frac{c_{eq1} - c_{\infty 1}}{\rho_1}\left(\frac{H_1 - k_1 t}{k_1}\right)^{\frac{3}{2}}\right]\right\}^{\frac{2}{3}}. \tag{14}$$

The constant of integration C in Eq.(14) is determined from initial conditions – for $t = 0$, $r = r_1$ – and thus we obtain solution of Eq. (13)

$$r = \left\{r_1^{\frac{3}{2}}\left(\frac{H_1}{H_1 - k_1 t}\right)^{\frac{1}{2}} - \sqrt{\frac{2}{\pi}}\left(\frac{D}{k_1}\right)^{\frac{1}{2}} \frac{c_{eq1} - c_{\infty 1}}{\rho_1}\left[H_1\left(\frac{H_1}{H_1 - k_1 t}\right)^{\frac{1}{2}} - (H_1 - k_1 t)\right]\right\}^{\frac{2}{3}}. \tag{15}$$

Making use of (8) we obtain the dependence of the bubble radius on the coordinate

$$r = \left\{r_1^{\frac{3}{2}}\left(\frac{H_1}{H}\right)^{\frac{1}{2}} - \sqrt{\frac{2}{\pi}}\left(\frac{D}{k_1}\right)^{\frac{1}{2}} \frac{c_{eq1} - c_{\infty 1}}{\rho_1}\left[H_1\left(\frac{H_1}{H}\right)^{\frac{1}{2}} - H\right]\right\}^{\frac{2}{3}}. \tag{16}$$

In the case when the gas density far from the bubble is described by function (11), it is more convenient to obtain a differential equation of the change in bubble radius as a function of the coordinate, having the following form:

$$\frac{dr}{dH} + \frac{1}{3}\frac{r}{H} = \sqrt{\frac{2}{\pi}}\left(\frac{D}{k_1}\right)^{\frac{1}{2}} \frac{c_{eq1}}{\rho_1}\left(1 - \frac{H_1}{H}\cdot\frac{c'}{c_{eq1}}\right) r^{-\frac{1}{2}}. \tag{17}$$

After integrating this equation with the initial conditions $r = r_1$, for $H = H_1$, we have

$$r=\left\{r_1^{\frac{3}{2}}\left(\frac{H_1}{H}\right)^{\frac{1}{2}}-\sqrt{\frac{2}{\pi}}\left(\frac{D}{k_1}\right)^{\frac{1}{2}}\frac{c_{eq1}}{\rho_1}\left[H_1\left(\frac{H_1}{H}\right)^{\frac{1}{2}}\left(1-3\frac{c'}{c_{eq1}}\right)H\left(1-3\frac{H_1}{H}\cdot\frac{c'}{c_{eq1}}\right)\right]\right\}^{\frac{2}{3}}, \tag{18}$$

or, passing over to the dependence of the radius on time we have

$$r=\left\{r_1^{\frac{3}{2}}\left(\frac{H_1}{H_1-k_1t}\right)^{\frac{1}{2}}-\sqrt{\frac{2}{\pi}}\left(\frac{D}{k_1}\right)^{\frac{1}{2}}\frac{c_{eq1}}{\rho_1}\left[H_1\left(\frac{H_1}{H_1-k_1t}\right)^{\frac{1}{2}}\left(1-3\frac{c'}{c_{eq1}}\right)+3H_1\frac{c'}{c_{eq1}}-(H_1-k_1t)\right]\right\}^{\frac{2}{3}}. \tag{19}$$

Similarly after substituting the appropriate values of rate v in Eqs. (4) and (9) we obtain equations connecting the bubble radius with the coordinate for the second stage of rise ($v = k_2 r$):

$$r=\left\{r_2^2\left(\frac{H_2}{H}\right)^{\frac{2}{3}}-\frac{6}{5}\sqrt{\frac{2}{\pi}}\left(\frac{D}{k_2}\right)^{\frac{1}{2}}\frac{c_{eq2}-c_{\infty 2}}{\rho_2}\left[H_2\left(\frac{H_2}{H}\right)^{\frac{2}{3}}-H\right]\right\}^{\frac{1}{2}}; \tag{20}$$

$$r=\left\{r_2^2\left(\frac{H_2}{H}\right)^{\frac{2}{3}}-\frac{6}{5}\sqrt{\frac{2}{\pi}}\left(\frac{D}{k_2}\right)^{\frac{1}{2}}\frac{c_{eq2}}{\rho_2}\left[H_2\left(\frac{H_2}{H}\right)^{\frac{2}{3}}\left(1-\frac{5}{2}\frac{c'}{c_{eq2}}\right)-H\left(1-\frac{5}{2}\cdot\frac{c'}{c_{eq2}}\cdot\frac{H_2}{H}\right)\right]\right\}^{\frac{1}{2}}. \tag{21}$$

For the third stage of rise ($v = k_3 r^2$) we have

$$r=\left\{r_3^{\frac{5}{2}}\left(\frac{H_3}{H}\right)^{\frac{5}{6}}-\frac{15}{11}\sqrt{\frac{2}{\pi}}\left(\frac{D}{k_3}\right)^{\frac{1}{2}}\frac{c_{eq3}-c_{\infty 3}}{\rho_3}\left[H_3\left(\frac{H_3}{H}\right)^{\frac{5}{6}}-H\right]\right\}^{\frac{2}{5}}; \tag{22}$$

$$r=\left\{r_3^{\frac{5}{2}}\left(\frac{H_3}{H}\right)^{\frac{5}{6}}-\frac{15}{11}\sqrt{\frac{2}{\pi}}\left(\frac{D}{k_3}\right)^{\frac{1}{2}}\frac{c_{eq3}}{\rho_3}\left[H_3\left(\frac{H_3}{H}\right)^{\frac{5}{6}}\left(1-3\frac{c'}{c_{eq3}}\right)-H\left(1-3\frac{c'}{c_{eq3}}\cdot\frac{H_3}{H}\right)\right]\right\}^{\frac{2}{5}}, \tag{23}$$

here Eqs. (20) and (22) pertain to the case when the gas density in the liquid changes according to (10) and Eqs. (21) and (23) are valid for a constant concentration (11).

A comparison of the above solutions with the data obtained under the assumption of constancy of the gas density in the bubble, the concentration pressure, and the rate of rise of the bubble shows that for large bubbles ($r > 1$ mm) the results of computations are significantly different for small initial pressures and that they converge for increasing pressures. Thus, at an initial pressure of 15 atm for an air bubble rising in water the time for decreasing the radius from 2 to 1 mm, computed according to formula (15), exceeds by 100% the time computed from the formulas given in [9] assuming $\rho_g = \text{const}$, $\Delta c = \text{const}$, and $v = \text{const}$; for an initial pressure of 150 atm this difference is 15%.

For a small bubble ($r < 1$ mm) the computations using formulas (20)-(23) are in good agreement with the solutions for $p_g = \text{const}$, if only we assume as before that the rate of rise of the bubble is proportional to its radius and for $r < 0.2$ mm it is proportional to the square of the radius.

A number of experimental results indicate that the bubbles of sufficiently small radius do not rise in the liquid. Thus, under certain conditions sea water contains nonfloating air microbubbles of a radius on the order of 20 μ at different depths [10].

It can be assumed that the rise of the bubble stops when the rate of rise becomes comparable to the velocity of the thermal motion of the bubble, i.e., of a Brownian particle. The radius of a nonrising bubble computed on the basis of this assumption is in good agreement with experimental data.

For determining the dissolution time of a nonrising bubble (fourth stage) we can make use of Langmuir formula

$$Sh=\frac{2r\beta_L}{D}=2. \tag{24}$$

Since at this stage the bubble does not rise, the change of gas density and concentration is determined only by the change in the surface tension force as the gas diffuses out of the bubble

$$\rho=\rho_4\left(1+\frac{2\sigma^*}{H_4 r}\right), \tag{25}$$

$$c_{eq}=c_{eq4}\left(1+\frac{2\sigma^*}{H_4 r}\right), \qquad (26)$$

where

$$\sigma^*=\sigma\frac{H}{M}\cdot 10^{-4}.$$

Considering that

$$\beta_l=-\frac{1}{4\pi r^2(c_{eq}-c_\infty)}\cdot\frac{dM}{dt}, \qquad (27)$$

substituting (25) into Eqs. (5) we obtain the following equation from (24):

$$\left(r+\frac{4}{3}\cdot\frac{\sigma^*}{H_4}\right)dr=-\frac{D(c_{eq4}-c')}{\rho_4}\left(1+\frac{2\sigma^*}{H_4 r}\right)dt. \qquad (28)$$

Integrating this equation under the initial conditions $r=r_4$ at $t=0$ we obtain the solution

$$t=\frac{\rho_4}{D(c_{eq4}-c')}\left\{4\frac{\sigma^*}{H_4}\left(\frac{\sigma}{H_4}-\frac{1}{3}\right)\ln\frac{r_4H_4+2\sigma^*}{rH_4+2\sigma^*}+\right.$$
$$\left.+\frac{1}{2}\left[\left(r_4+2\frac{\sigma^*}{H_4}\right)^2-\left(r+2\frac{\sigma^*}{H_4}\right)^2\right]-\frac{16}{3}\cdot\frac{\sigma^*}{H_4}(r_4-r)\right\}. \qquad (29)$$

For $H_4 > 10$ m and $r < 10^{-7}$ m in (29) we can neglect the terms containing σ^*/H_4 (σ^* is on the order of 10^{-7} m^2). Then we obtain the simple formula

$$t=\frac{1}{2}\frac{\rho_4}{D(c_{eq4}-c')}(r_4^2-r^2), \qquad (30)$$

which agrees with the formula given in [11].

The right-hand sides of Eqs. (16), (18), (20), (21), (22), and (23) represent the difference of two terms; the first of these contains the initial radius and the ratio of the initial coordinate to the vertical coordinate, while the second contains the kinematic diffusion coefficient.

Depending on the gas and liquid characteristics and also on the initial conditions during its rise the bubble can decrease in volume (if diffusion predominates over gravitational expansion), as well as it can increase.

In a number of practical cases the conditions under which the gas bubbles passing through a liquid do not reach the free surface are of interest. For determining these conditions we analyze the equations connecting the bubble radius with the vertical coordinate.

In the case when the gas concentration in the liquid changes according to (10), the condition of decrease in the bubble radius during its rise (in the first stage) is obtained from Eq. (16)

$$r_1^{\frac{3}{2}}\left(\frac{H_1}{H}\right)^{\frac{1}{2}}-\alpha\left[H_1\left(\frac{H_1}{H}\right)^{\frac{1}{2}}-H\right]<r_1^{\frac{3}{2}}, \qquad (31)$$

where

$$\alpha=\sqrt{\frac{2}{\pi}}\left(\frac{D}{k_1}\right)^{\frac{1}{2}}\frac{c_{eq1}-c_{\infty 1}}{\rho_1},$$

hence, it follows that

$$r_1<[\alpha(H_1+H_1^{\frac{1}{2}}H^{\frac{1}{2}}+H)]. \qquad (32)$$

Putting $H = 0$ we obtain the condition of monotonic decrease of the bubble from inequality (32)

$$r_1<(\alpha H_1)^{\frac{2}{3}}. \qquad (33)$$

If condition (33) is not fulfilled, function (16) can have an extremum. Equating the first derivative of the radius to zero and assuming $r \neq 0$ we can determine the value of the coordinate at which the radius goes through the extremum:

$$H = \left[\frac{1}{2\alpha} H_1^{\frac{1}{2}} \left(r_1^{\frac{3}{2}} - \alpha H_1\right)\right]^{\frac{2}{3}}. \tag{34}$$

It is obvious that the extremum value of the radius is meaningful for $H < H_1$; so it follows from formula (34) that

$$r_1 \leqslant (3\alpha H_1)^{\frac{2}{3}}. \tag{35}$$

If condition (35) is not satisfied, then during its rise the bubble increases monotonically in volume.

It is not difficult to verify that the second derivative of the radius is positive and hence the obtained extremum corresponds to the minimum of the radius. We find this value by substituting (34) into Eqs.(16):

$$r_{\min} = 1.53\alpha^{\frac{2}{9}} \left[H_1^{\frac{1}{2}} \left(r_1^{\frac{3}{2}} - \alpha H_1\right)\right]^{\frac{4}{9}}. \tag{36}$$

If it turns out that $R_{\min} < 10^{-3}$ m, then the further evolution of the bubble is determined by Eq. (20).

It follows from Eq. (36) that $r_{\min} < 10^{-3}$ m if

$$r_1 < \left(\alpha H_1 + \frac{0.705 \cdot 10^{-7}}{\alpha^{\frac{1}{2}} H_1^{\frac{1}{2}}}\right)^{\frac{2}{3}}. \tag{37}$$

It follows from Eq. (20) that at the second stage of rise the bubble radius will decrease monotonically if

$$r_{\min} < \left(\frac{6}{5} \alpha H_2\right)^{\frac{1}{2}}. \tag{38}$$

In order to determine what happens to the bubble after it reaches the minimum value $r_{\min} < 10^{-3}$ m in the second stage of rise in inequality (38) instead of H_2 we substitute the coordinate corresponding to the minimum radius given by formula (34). The bubble will decrease monotonically if

$$r < \left(\alpha H_1 + \frac{6.3 \cdot 10^{-3}}{\alpha^{\frac{1}{2}} H_1^{\frac{1}{2}}}\right)^{\frac{2}{3}}. \tag{39}$$

It is seen from a comparison of formulas (37) and (39) that when condition (37) is satisfied and the minimum value of the bubble radius falls within the radii characterizing the second stage of rise the bubble will always decrease in size thereafter.

A similar analysis can be done also for the case of constant gas concentration in the liquid far from the bubble. Omitting the intermediate computations we give the condition of monotonic decrease of the bubble radius

$$r_1 < \left[\xi H_1 (1 - 3\gamma)\right]^{\frac{2}{3}}, \tag{40}$$

where

$$\xi = \sqrt{\frac{2}{\pi}} \left(\frac{D}{k_1}\right)^{\frac{1}{2}} \frac{c_{\text{eq1}}}{\rho_1}, \quad \gamma = \frac{c'}{c_{\text{eq1}}},$$

and also the condition of decrease of the bubble radius in the second stage of rise (after reaching the minimum value of the radius $r_{\min} < 10^{-3}$ m in the first stage):

$$r_1 < \left[\xi H_1 (1 - 3\gamma) + \frac{2}{\xi^{\frac{1}{2}} H_1^{\frac{1}{2}}} \left(\xi\gamma H_1 + 1.055 \cdot 10^{-5}\right)^{\frac{3}{2}} \right]^{\frac{2}{3}}. \quad (41)$$

The above relations between the radius of the rising gas bubble and the coordinate enable us to compute the characteristics of the absorption process for all sizes of columns and their elements.

These relations are valid also for processes occurring at high pressures. Computational estimates show that the errors related to the assumptions made here do not exceed 5% up to pressures on the order of 200 atm for all gases that are not easily dissolved.

NOTATION

r	is the bubble radius;
v	is the bubble rise velocity;
t	is the time;
H	is the absolute height of liquid column (vertical coordinate);
M	is the mass;
I	is the diffusion flux;
ρ	is the density;
c	is the volumetric concentration;
D	is the diffusion coefficient;
σ	is the surface tension coefficient;
	is the mass-transfer coefficient;
m	is the phase equilibrium coefficient;
K	is the mass-exchange coefficient;
c_f	is the resistance coefficient;
Re	is the Reynolds number;
Sh	is the Sherwood number.

Indices

g	is the gas;
l	is the liquid;
g.l	is the gas–liquid interface;
0	is the initial value;
1, 2, 3, 4	are the initial values on corresponding bubble rise stages;
eq	is the value at equilibrium;
ap	is the aperture;
b	is the bubble.

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