Breakup of Gas Bubbles Rising in Molten Metals

K. Wichterle

VSB-Technical University of Ostrava, 70833 Ostrava Poruba, Czech Republic, kamil.wichterle@vsb.cz

A theory on the breakup of rising bubbles is developed on the basis of long-term experiments with bubbles rising in different liquids. The probability of bubble breakup is expressed in terms of half-life as a dimensionless function of the bubble volume and liquid properties. The results are applied to develop a mathematical model of bubbles rising in a column of high density liquid under a low external pressure. The model predicts that in typical steelmaking equipment the size of bubbles approaching the liquid level depends mostly on the external pressure and it is quite independent of the original size of the bubbles.

Keywords: gas bubbles in liquid, mathematical model, vacuum degassing

Submitted on 6 August 2008, accepted on 29 January 2010

Introduction

Larger bubbles rising in a liquid become unstable and of oscillating shape and sometimes they split. The probability of bubble breakup increases with the bubble size. Currently, the breakup is observed during the bubble formation and it is less significant during the bubble rise. However, when the bubble volume increases for different reasons, e.g. due to decreasing pressure, increasing temperature, boiling, desorption or chemical reactions, the breakup may occur all the time and it controls the bubble size. This paper presents a general mathematical model of bubble breakup for rising expanding bubbles. As an example, the model is applied to the vacuum degassing treatment of liquid metals.

State-of-the-art. One important process of steelmaking technology is argon-vacuum degassing [1–3]. Typically 200 t of molten steel is placed in a ladle. Argon bubbles are introduced at the ladle bottom to provide agitation by gas-lift effect, to strip other dissolved gases and to remove immiscible slag particles. The typical height of the steel level is 5 m. An external pressure of about 50 Pa (vacuum) is kept at the level. Bubble movement under these conditions is quite specific. The change from the ferrostatic pressure at the bottom, which is 0.4 MPa, to this vacuum means that the bubble volume expands by 4 decimal orders during its floating to the ladle surface level. According to these conditions, breakup of increasing bubbles might be extremely important here.

Direct observation of bubbles in liquid steel is quite complicated; measuring methods applicable either in industrial scale or in laboratory models are quite limited [4–7]. Therefore, modelling with other liquids is an essential way of the process investigation. Experimental modelling by using more user-friendly liquid metals like Wood metal [8–17] or mercury [18] was done in certain laboratories. Surprisingly, chemical engineering similarity analysis [19, 20] indicates that when modelling the bubble-liquid interactions, the simplest air-water system is more similar to liquid steel than these metals, which is shown in Table 1. Hundreds of experimental, theoretical and numerical studies on bubbles rising in water and other common liquids have been published as well as a number of monographs on the subject [21–28]. In relation to metallurgy ladles, degassing equipment, tundish, and continuous casting, the application of cold models is very popular. Few laboratories have presented hydrodynamic models with volumes of more than 1 m³ [29–31], most bench scale studies took place in volumes smaller than 100 l [32–45]. Vacuum is applied very rarely [46]. Under such circumstances rising of bubbles does not take more than few seconds and expansion of bubbles and consequential breakup could be neglected. Therefore, there is a lack of data on breakup which is particularly important in industrial metallurgy ladles; however it is also significant in large-scale airlift equipment [47–50].

Investigation of bubble breakup is mostly oriented either to the problems of bubble formation in orifices or to the breakup of bubbles subjected to external forces in ambient flow in pipes, sparged columns and in agitated vessels [51–65]. However, little attention is paid to the problem of breakup during the free rising of bubbles in liquid at rest.

Experimental

Rising bubbles levitating in diverging downstream liquid flow can be observed for quite a long period [66]. We have developed a similar equipment which enables to observe the behaviour of freely rising bubbles qualitatively and quantitatively under defined conditions. Breakup of rising medium size bubbles with diameters \( d_0 = 2.5 - 11 \text{ mm} \) was studied in our laboratory in water and glycerol solutions [67] and recently in butyl alcohol and in water with electrolyte or polymer additives [68]. Apparently the large bubbles are essentially unstable. Detailed analysis shows that there is a certain fuzzy region between the stable and broken bubbles. It can be characterized by the half-life of the bubbles which can be determined by observation of a statistically meaningful number of bubbles.
When starting with $N(0)$ bubbles, the number $N(t)$ of surviving bubbles decays exponentially according to

$$\frac{N(t)}{N(0)} = \exp(-rt).$$  \hspace{1cm} (1)

The breakup rate was characterized by the half-life of rising bubbles

$$t_{1/2} = \frac{\ln 2}{r}. \hspace{1cm} (2)$$

Generally, it can be assumed that $t_{1/2}$ depends on the bubble size, and on the liquid’s properties, such as density $\rho$ and viscosity $\mu$, and on the characteristics of the interface. As the experiments with glycerol solutions indicate, the effect of viscosity is minor, and the interface effects are essential. Assuming that they could be characterized by a single value, surface tension $\sigma$, a dimensionless quantity can be established as

$$\Theta_{1/2} = t_{1/2} \rho^{1/4} \sigma^{3/4}$$ \hspace{1cm} (3)

depending only on the Eötvös number

$$Eo = \frac{\rho g d^2}{\sigma}. \hspace{1cm} (4)$$

An empirical power-law function

$$\Theta_{1/2} = 5.9 \times 10^9 \, Eo^{-6} \quad (R^2 = 0.88). \hspace{1cm} (5)$$

fits approximately the experimental data [67]. According to this result, the kinetic constant in (1) can be expressed as a function of bubble volume

$$r = (KV)^a$$ \hspace{1cm} (6)

where

$$a = 4.$$ \hspace{1cm} (7)

The values of the instability parameter $K$ for particular materials are presented in Table 1. Here again, water is a fair model for studying bubbles in liquid steel.

### Analysis

Statistics evaluation of breakup events provided a simple model of the process. It is applied here for prediction of bubble size during motion under decreasing pressure.

Let us consider a given volume $Q_0$ of gas infused into a liquid as a large number of uniform size bubbles of volume $V_0$. The number of bubbles is $N_0 = Q_0/V_0$. The volume of gas is not constant when the hydrostatic pressure of the ambient liquid decreases from the viewpoint of the rising bubbles. Other reason for the volume change may be time change of external pressure during vacuum treatment, thermal expansion of gas, desorption of dissolved gas etc. Anyhow, the resulting volume of gas is a certain function $Q(t)$ and accordingly, the volume of a single bubble is

$$V_0(t) = V_0 \frac{Q(t)}{Q_0}. \hspace{1cm} (9)$$

The equivalent bubble diameter (diameter of a sphere of identical volume) is:

$$d_0(t) = \left(\frac{6 V_0(t)}{\pi}\right)^{1/3}. \hspace{1cm} (10)$$

Bubbles may split. Medium size ellipsoidal bubbles usually provide two daughter bubbles of nearly identical size. Large, unstable bubbles release chaotically groups of smaller daughter bubbles, nevertheless the size of grand-grand...-daughter bubbles becomes more and more uniform and the size distribution of medium size bubbles in resulting swarm is quite narrow as shown schematically in Figure 1.

This situation is considered as a starting point for the presented mathematical model. Accordingly, the volume of the first generation of daughter bubble is

$$V_1(t) = V_0(t)/2$$ \hspace{1cm} (11)

and for the $i$-th generation of daughter bubble it is

$$V_i(t) = V_0(t)/2^i$$ \hspace{1cm} (12)
The respective number of bubbles, $N_i$, in the $i$-th generation occupy the volume

$$Q_i(t) = N_i(t) V_i(t)$$

and the total gas volume is

$$\sum_i Q_i(t) = Q(t)$$

Then the volume fraction of the $i$-th generation bubbles is

$$m_i(t) = \frac{Q_i(t)}{Q(t)}$$

Mean (Sauter) equivalent diameter of all bubbles can be defined as

$$d_A = \frac{6 \sum_i N_i(t) V_i(t)}{\sum_i N_i(t) A_i(t)} = \sum_i \frac{d_0(t)}{m_i(t)^{2/3}},$$

where $A_i(t)$ is the surface area of $i$-th generation bubble.

Average bubble volume is defined as

$$V_A = \frac{\pi}{6} d_A^3 = \frac{V_0(0) Q(t)}{Q(0)} \left( \sum_i m_i(t)^{2/3} \right)^3$$

When the kinetics of bubble breakup is known, these values can be predicted by the following theoretical model.

**Cascade of bubble breakup.** Assume that the breakup of bubbles in a swarm follows the relation (6) determined for a single bubble. It is plausible at least for the systems with low gas holdup, where the bubble interactions are less significant. Then, the volume fraction of mother bubbles decreases

$$\frac{dm_0}{d\tau} = -r m_0$$

When the volume of gas in the system increases according to (9), application of (6) gives

$$\frac{dm_0}{d\tau} = -\left( KV_0 \frac{Q(t)}{Q(0)} \right)^a m_0.$$  \hspace{1cm} (19a)

The first generation of daughter bubbles can split as well and its balance is

$$\frac{dm_1}{d\tau} = -\frac{dm_0}{d\tau} \left( KV_0 \frac{Q(t)}{2 \frac{Q(0)}{Q(0)}} \right)^am_1;$$

Generally, for the $i$-th generation ($i = 1,2,3 \ldots$) it is

$$\frac{dm_i}{d\tau} = \left( KV_0 \frac{Q(t)}{Q(0)} \right)^a \frac{m_{i-1}}{2^{(i-1)a}} - \frac{m_i}{2^ia}.$$ \hspace{1cm} (19c)

We shall consider a simple starting point

$$m_0 = 1 \text{ and } m_i = 0 \text{ for } i = 1, 2, 3 \ldots \text{ at } \tau = 0$$ \hspace{1cm} (19d)

To formally simplify following formulas, we introduce the variables

$$\tau \equiv (KV_0)^a \int_0^\tau \left( \frac{Q(s)}{Q(0)} \right)^a ds,$$

and

$$\varepsilon = 2^{-a}.$$ \hspace{1cm} (21)

Then, the mathematical model of the process is the following set of ordinary differential equations

$$\frac{dm_0}{d\tau} = -m_0$$ \hspace{1cm} (22a)

and

$$\frac{dm_i}{d\tau} = \varepsilon^{i-1} m_{i-1} - \varepsilon^i m_i, \text{ for } i = 1,2,3 \ldots$$ \hspace{1cm} (22b)

with the condition

$$m_0 = 1 \text{ for } \tau = 0.$$ \hspace{1cm} (22c)

Its solution is simply

$$m_i = \sum_{j=0}^i b_{ij} \exp(-\varepsilon^{i-j})$$ \hspace{1cm} (23)

where

$$b_{0,0} = 1$$

$$b_{i,0} = \prod_{k=1}^{i} \left( -\varepsilon^{k+1} \frac{1}{1-\varepsilon^k} \right) \quad b_{i,i} = \prod_{k=1}^{i} \left( \frac{1}{1-\varepsilon^k} \right)^{i}$$ \hspace{1cm} (24a)
and generally

\[ b_{i,j} = \prod_{k=1}^{i-j} \left( \frac{a^k - 1}{1 - a^k} \right) \prod_{k=0}^{i-3} \left( \frac{1}{1 - a^k} \right) \]  

(24b)

The distribution of mother and daughter bubbles is plotted in Figure 2 for the values \( a = 4 \) and \( \varepsilon = 0.0625 \) from (7) and (21).

The respective time development of the Sauter mean diameter, according to (8), is shown in Figure 3.

Apparently, there is an asymptote

\[ \frac{V_A Q(0)}{V_0(0) Q(\tau)} = 1 \]  

for the start-up period \( \tau << 0.1 \)

when breakup can be neglected.

The average bubble size slightly oscillates about a line

\[ \frac{V_A Q(0)}{V_0(0) Q(\tau)} = 0.64 \left( \frac{\tau}{\tau^{1/a}} \right) \]  

(26a)

for developed breakup at \( \tau >> 0.1 \).

The last relation can be rewritten as

\[ V_A = \frac{0.64}{K^{1/a}} \Psi \]  

(26b)

with the correction factor

\[ \Psi = \frac{Q(t)}{Q(0)} \left[ \frac{1}{t} \int_0^t \left( \frac{Q(s)}{Q(0)} \right)^a ds \right]^{-\frac{1}{2}} \]  

(27)

which becomes \( \Psi = 1 \) when the total gas volume does not change. Evidently, the mean bubble size for larger \( \tau \) does not depend on the initial conditions (mother bubble size).

**Breakup cascade for bubble rising in hydrostatic (ferrostatic) column.** Bubbles rising in a larger column expand due to decreasing hydrostatic pressure. In particular, if gas is released in a depth \( H \) of a liquid column of density \( \rho \) (significantly higher than the gas density), then the gas volume \( Q \) depends on the bubble position and on the external pressure at the liquid level. It is known [21–28] that rising velocity, \( v \), of medium size bubbles in low viscosity liquids is nearly independent on Reynolds and Eötvös number and in water it is usually 0.2–0.3 m/s. Considering \( v \) to be nearly constant, the change of volume of an ideal gas with time is given by

\[ \frac{Q(t)}{Q(0)} = \left[ \frac{p_0 + \rho g H}{p_0 + \rho g H - \rho g v t} \right]^b \]  

(28)

where

\[ b = 1 \quad \text{for isothermal process} \]  

(29a)

\[ b = \frac{1}{\gamma} \quad \text{for adiabatic process} \]  

(29b)

Let us transform this relation using the dimensionless parameter

\[ B = \frac{p_0}{\rho g H} \]  

(30)

which characterizes the relation of external pressure to the hydrostatic pressure. The dimensionless coordinate

\[ X = \frac{t v}{H} \]  

(31)

is the ratio of the bubble path length to the column height.

Then the correction factor \( \Psi \) in Equation (27) is expressed by

\[ \Psi = \left[ \frac{(1 + B - X)}{X (ab - 1)} \left( 1 - \left( 1 - \frac{X}{B + 1} \right)^{ab-1} \right) \right]^{-\frac{1}{2}} \]  

(32)

If either \( t \to 0 \) or \( B \to 0 \), the volume change is negligible and \( \Psi \to 1 \).

Otherwise,

\[ V_A = \frac{0.64}{K^{1/a}} \Psi(X, B, a, b) \]  

(33)

where the function \( \Psi(X, B, a, b) \) rises with \( X \) as plotted in Figure 4.

The important parameter \( B \) is very high for common laboratory experiments in small vessels and the assumption of \( \Psi = 1 \) is acceptable in most cases here.
Other situation is in large airlift equipment [47–50], e.g. in pachuca tanks for leaching of ores where $H \approx 20$ m and $B \approx 0.5$.

The extreme cases occur just in metallurgy during vacuum degassing of liquid steel ($\rho = 7200$ kg/m$^3$) stirred by argon bubbles, where $p_0$ may be as low as 100 Pa and $H$ as high as 5 m which gives $B \approx 0.0005$. For small $B$, the formula (32) can be simplified as

$$\psi = \left(\frac{a - 1}{B}\right)^{0.5}$$

for $X \rightarrow 1$ and $B << 1$. (34)

A higher value of $\psi$ means that the bubble breakup is slower than gas expansion. When bubbles approach the liquid level, the expansion is fast and size of bubbles depends mainly on the external pressure, $p_0$. If the liquid column is high enough, according to (26b) the breakup process becomes independent of initial conditions and the mean bubble volume should be simply related to the local pressure, $p$,

$$V_A = 0.64 \frac{1}{K} \left(\frac{a - 1}{B}\right)^{0.5}$$

for $X = 1$ and $B << 1$. (35)

Bubbles approaching the liquid surface level – example. The values of mean bubble diameters, estimated by Eq. (34) with the constants (16–17) calculated for water and for liquid steel [18] are presented in Table 2.

While for water at external pressure $p = 100,000$ Pa the mean bubble size (bubbles at the sea level) is by our model $d_A \approx 9$ mm, the diameter of bubbles leaving water at a low pressure (100 Pa) space is $d_A \approx 16$ mm.

The diameters of gas bubbles in liquid steel are predicted to be approximately two times larger than in water.

Table 2. Size of bubbles approaching the liquid surface level from deep column as a function of external pressure.

<table>
<thead>
<tr>
<th>External pressure, $p_0$ [Pa]</th>
<th>100,000</th>
<th>10,000</th>
<th>1,000</th>
<th>100</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sauter diameter, $d_A$ [mm]</td>
<td>water</td>
<td>9.1</td>
<td>11.0</td>
<td>13.3</td>
</tr>
<tr>
<td></td>
<td>liquid steel</td>
<td>17.8</td>
<td>21.6</td>
<td>26.1</td>
</tr>
</tbody>
</table>

According to our estimate, the typical equivalent diameter of bubbles leaving the level of liquid steel in a ladle under vacuum degassing is about 30 mm. It is in accordance with the experimental results, e.g. by Iguchi et al. [70].

This size is nearly independent on the ladle depth when at least two breakup events occur during the bubble rise. In that case, the conditions at the ladle surface essentially control the bubble size irrespectively of the technique of gas introduction, temperature variation and the chemical reaction process.

Conclusion

The size of bubbles rising as a swarm in a large column where breakup occurs can be estimated using the developed model based on experimental data for single bubble rise and breakup probability under constant pressure conditions. Dimensional analysis offers predictions both for common low-viscosity liquids and for liquid metals. It can also be applied for estimating the heat and mass transfer processes in various metallurgical applications, including treatment by oxygen, argon, hydrogen, carbohydrates, etc. Concerning physical modelling of the systems with significant expansion, namely under vacuum, water models assuming normal ambient pressure can be still applied; in that case, other portions of gas should be subjoined at different liquid levels to the stream of bubbles introduced at the bottom to simulate the bubble expansion.

Nomenclature

$\alpha$ empirical exponent of breakup rate correlation (6)
$\beta$ expansion exponent (31)
$B$ dimensionless parameter (30)
$E_0$ Eötvös number (4)
$H$ liquid column depth
$K$ instability parameter (6)
$m$ volume fraction of given bubbles
$N$ number of bubbles
$p$ pressure
$Q$ gas volume
$r$ kinetics constant of breakup
$t$ time
$\tau_{1/2}$ half-life of breakup
$v$ bubble rise velocity
$V$ single bubble volume
$X$ dimensionless coordinate (31)
$\gamma$ Poisson constant
$\varepsilon$ parameter (21)
$\theta_{1/2}$ dimensionless half-life of breakup (3)
$\mu$ dynamic viscosity of liquid
$\rho$ liquid density
$\sigma$ surface tension
$\tau$ dimensionless time (20)
$\psi$ factor of breakup retardation (32)
Indexes

A average (Sauter)
0 initial
i daughter bubbles of i-th generation
w water

Acknowledgements

We gratefully acknowledge financial support by the grant No.104/07/1110 from the Grant Agency of the Czech Republic.

References


