Rectified Diffusion of Gas Bubbles in Molten Metal during Ultrasonic Degassing

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Abstract: In the present paper, an analytical solution of rectified diffusion of processes of gas bubbles in molten metal is derived for the purpose of predicting the diffusion behaviors of gas bubbles during ultrasonic degassing. In the present model, a theoretical threshold (in terms of the amplitude of the applied ultrasonic field) is determined for the evaluation of the ultrasonic degassing effects. The diffusion of hydrogen bubbles in molten aluminum is predicted, so as to provide examples to illustrate the important findings of the present work.

Keywords: cavitation; rectified diffusion; gas bubble; molten metal; ultrasonic degassing

1. Introduction

Ultrasonic degassing is an important topic for high-quality casting production [1–5]. For a comprehensive review of this technique, please see Eskin et al. [1]. During ultrasonic degassing, the existing bubbles could be forced to oscillate with significant amplitudes (termed as ‘acoustic cavitation’ [6–8]). During the oscillation process, gas shows prominent diffusion across the bubble interface with both the diffusion into and out of the bubble [9]. The overall diffusion direction is very important in regard to degassing efficiency. Based on our previous works on bubble dynamics in water [9], it has been proved that the amplitude of the ultrasonic field plays an important role on the determination of the diffusion direction. Hence, a rigorous theoretical determination of the phenomenon is necessary in order to promote further applications in the field.

First, the mechanisms of gas formations in the castings will be introduced. For this example, we will use aluminum alloys. At the aluminum melting interface with air, the existing moisture could react with the aluminum, forming atomic hydrogen and also molecular hydrogen. In conditions of oversaturation, hydrogen bubbles will be formed. During solidification, due to the prominent solubility of the hydrogen in the liquid aluminum, a great amount of porosity will be observed. As the porosity could significantly affect the performance of the alloys (e.g., yield strengths), some further treatments are essential to remove the pores.

When the ultrasonic fields are applied to the liquid aluminum, the existing bubbles will oscillate dramatically. During the oscillations, and in proper conditions, the dissolved hydrogen could be diffused into the bubble through the bubble–liquid interface [9]. With the growth of the bubble, the hydrogen in the liquid will be reduced, as the hydrogen will be inside the bubbles in increasing quantity. When the bubble is of a significant size, it will float to the liquid interface and then the hydrogen will
be removed. During the whole process, there is no introduction of additives into the system. Hence, compared with other mechanical methods (e.g., stirring facilities), the ultrasonic treatment is a clean method. Furthermore, ultrasonic treatment does not need expensive instruments, making it both a cheap and convenient method. It is necessary to mention that, except for the aforementioned degassing effects, ultrasonic treatment could achieve other benefits e.g., micro-structure modification [10,11] and grain refinement [12,13]. The applicable melts of ultrasonic treatment include aluminum–magnesium alloys [14]. Unfortunately, the theoretical basis for ultrasonic degassing is still absent in the literature (e.g., in terms of the theoretical predictions of the suitable conditions for the process).

In the present paper, an analytical solution to the rectified diffusion of gas bubbles in molten metal is derived and discussed, with the aid of typical examples. Specifically, the coupled equations of diffusion and bubble motion are solved together, with the corresponding initial and boundary conditions. Compared with the previous work, Sievert’s law is employed for the closure of the model. For the demonstrating cases, the diffusion process of the hydrogen bubbles in the liquid aluminum is analyzed in detail to illustrate the importance of the present paper.

2. Theoretical Analysis

In this section, the formulas for rectified diffusion of gas bubbles in molten metal under acoustic fields are derived. The following assumptions are employed:

1. The involved sizes of the bubble is far beyond the molecular level [15].
2. The interactions between the bubble and the boundaries are ignored. Here, we only consider a bubble in an infinite liquid melt. In fact, near the boundaries (e.g., wall and particle), the cavitation phenomenon is very complex [16–18].
3. The bubbles are assumed to be spherical. In fact, because of the high viscosity involved here, the bubble could be safely treated as spherical ones. For a more recent work on the bubble stability theory, readers could refer to Klapcsik and Hegedüs [19,20].
4. The viscosity and the compressibility of molten metal are both considered.
5. The relationship between the volume and the internal pressure of gas bubbles is described by the polytropic model [21].
6. Thermal damping is currently ignored. As shown in our previous work [21], this damping mechanism often does not serve as a dominant one.

The equation of bubble motion is given as [9],

\[
(1 - \frac{\dot{R}}{c_l})R\ddot{R} + \frac{3}{2} \left(1 - \frac{\dot{R}}{3c_l}\right)R^2 = (1 + \frac{\dot{R}}{c_l}) \frac{p_{ext}(R, t) - p_s(t)}{\rho_l} + \frac{R}{\rho_l c_l} \frac{d[p_{ext}(R, t) - p_s(t)]}{dt},
\]

(1)

where

\[
p_{ext}(R, t) = P_g - \frac{2\sigma}{R} - \frac{4\mu_l}{R} \dot{R},
\]

(2)

\[
P_g = (P_\infty + \frac{2\sigma}{R_0})(R_0/R)^{3\eta},
\]

(3)

\[
p_s(t) = P_\infty + P_A \cos(2\pi ft).
\]

(4)

Here, \( R \) is the instantaneous radius of the gas bubble oscillating in the melting metals; overdot denotes the time derivative; \( \rho_l \) is the density of the molten metal; \( t \) is the time; \( c_l \) is the sound speed propagating in the molten metal; \( P_g \) is the instantaneous pressure in the bubble; \( \sigma \) is the surface tension coefficient; \( \mu_l \) is the viscosity of the molten metal; \( P_\infty \) is the ambient pressure; \( R_0 \) is the initial and equilibrium radius of the given bubble; \( \eta \) is the polytropic exponent; \( P_A \) is the amplitude of the applied acoustic field; \( f \) is the frequency of the applied acoustic field.
The mass transfer equation is \[ \frac{\partial c}{\partial t} + u \cdot \nabla c = D \nabla^2 c, \] (5)

where \( c \) is the concentration of certain gas in the molten metal; \( u \) is the velocity of the molten metal surrounding the gas bubble; \( D \) is the diffusion constant. The initial and boundary conditions are \[ c(r, 0) = C_i, \quad r > R; \] (6)

\[ \lim_{r \to \infty} c(r, t) = C_i; \] (7)

\[ c(R, t) = C_s, \quad t > 0. \] (8)

Here, \( C_i \) is the initial concentration or the concentration at infinity; \( C_s \) is the local gas concentration at the inner bubble wall. The solubility of gases in molten metal is controlled by Sievert’s law \[ C_0 = k_S \sqrt{P_\infty}, \quad C_s = k_S \sqrt{P_g}, \] as follows

Here, \( C_0 \) is the saturation concentration of gas in the molten metal; \( k_S \) is a constant. Equations (1) to (5) with initial and boundary conditions of Equations (6) to (8) can be solved following our previous framework \[ 9 \]. Because the solubility of gas in the molten metal is controlled by the Sievert’s law rather than Henry’s law, some modifications on the previous works will be performed as follows. The time derivative of the gas amount change (\( n \)) inside the bubble is given by \[ \frac{dn}{dt} = \frac{d}{dt} (\Delta_0 + \Delta_1), \]

where

\[ \Delta_0 = -8(\pi Dt)^{1/2} R_0^3 F_0 (\langle R/R_0 \rangle^4), \]

\[ \Delta_1 = -4\pi Dt R_0^3 F_0 (\langle R/R_0 \rangle^4), \]

\[ F_0 = \frac{1}{T_b} \int_0^{T_b} R^4 F dt, \]

with

\[ F(\tau) = C_s - C_i, \]

\[ \tau = \int_0^{t} R^4(t) dt', \]

\[ \tau_0 = \int_0^{T_b} R^4 dt. \]

Here, \( T_b \) is the bubble oscillation period; \( < > \) denotes time averages of the given parameter. By using the Sievert’s law, \( F_0 \) can be expressed as,

\[ F_0 = -C_0 \left( \frac{C_i}{C_0} - \frac{(R/R_0)^4}{(P_s/P_\infty)^{1/2}} \right). \]

The required time averages in \( \frac{dn}{dt} \) can be determined based on the solution of Equations (1) to (4) using the perturbation method with direct series expansions. Only second-order terms are considered,
and the harmonic term can be neglected ([22], p. 501). Following the framework of reference [9], the bubble growth rate in the given molten metal is as shown in [9]

\[
\frac{dR_g}{dt} = \frac{D_{R_g} T_0 c_0}{R_0^2 P_\infty} \left( \frac{R}{R_0} \right) + R_0 \left( \frac{\langle R/R_0 \rangle^4}{\eta} \right)^{1/2} \times \left( 1 + \frac{4\mu}{3\rho R_0^2} \right)^{-1} \left( \frac{C_i - \langle \langle R/R_0 \rangle^4 (P_\infty/P_\infty)^{1/2} \rangle}{\langle R/R_0 \rangle^4} \right)
\]

(9)

with

\[
\langle R/R_0 \rangle = 1 + K_\alpha^2 (P_A/P_\infty)^2,
\]

(10)

\[
\langle \langle R/R_0 \rangle^4 \rangle = 1 + (3 + 4K)\alpha^2 (P_A/P_\infty)^2,
\]

(11)

\[
\langle \langle R/R_0 \rangle^4 (P_\infty/P_\infty)^{1/2} \rangle = \left( 1 + \frac{2\mu}{R_0^2 P_\infty} \right) \left[ 1 + (4 - 3\eta/2)K_\alpha^2 (P_A/P_\infty)^2 + \frac{3(\eta/2-1)(3\eta/2-4)}{4} \alpha^2 (P_A/P_\infty)^2 \right]
\]

(12)

\[
a = -\frac{P_\infty}{\rho l R_0^2} \left( \frac{1 + (\omega R_0/ci)^2}{(\omega^2 - \omega_0^2)^2 + (4\mu/\rho l R_0^2 + \omega_0^2 R_0/ci)^2 \omega^2} \right)^{1/2},
\]

(13)

\[
\omega_0^2 = \frac{1}{\rho l R_0^2} \left[ (3\eta + 1 - \rho \omega^2 R_0^2 = 3\eta P_\infty) + (\alpha/2 R_0 P_\infty) (3\eta + 1 - 2/3\eta) \right],
\]

(14)

\[
K = \frac{(3\eta + 1 - \rho \omega^2 R_0^2 = 3\eta P_\infty) + (\alpha/2 R_0 P_\infty) (3\eta + 1 - 2/3\eta) \cdot \rho_1 R_0^2}{1 + (2\alpha/\rho_0 R_\infty P_\infty)(1 - 1/3\eta)}.
\]

(15)

Here, \( R_0 \) is the universal constant; \( T \) is the temperature (unit: K). Compared with previous work (e.g., [9]) based on the Henry’s law, Equations (9) and (12) are different owing to the use of Sievert’s law (e.g., the terms with \( P_S \) in the Equations (9) and (12)). Based on Equations (9) to (15), one can find that the growth or dissolution rate of gas bubbles in molten metal under acoustic excitation is dependent on many parameters, e.g., acoustic pressure, amplitude, and frequency, saturation conditions, ambient pressure, temperature, bubble radius, and time. By integration of Equation (9), the quasi-equilibrium bubble radius could be obtained. If one set \( dR_g/dt = 0 \) in Equation (9), the corresponding acoustic amplitude will be the threshold for the diffusion \( (P_T) \) as follows

\[
P_T^2 = \frac{P_\infty^2}{\alpha^2} \left[ (1 + 2\alpha/\rho_0 P_\infty - C_i/C_0) \right] \left( 3\eta/2 - 1 \right)^2 \left( 3\eta/2 - 4 \right)^2 + (4 - 3\eta/2) K (1 + 2\eta/\rho_0 P_\infty) \]

(16)

For a given bubble radius, the bubble will grow if \( P_A > P_T \) and dissolve if \( P_A < P_T \).

3. Results and Discussions

In this section, the predictions of the rectified diffusion of gas bubbles in molten metal are demonstrated and discussed. Here, hydrogen bubbles in molten aluminum is considered as an example. For the physical properties of molten aluminum, the following constants are used [24]: melting point \( T_m = 933.4 \) K; \( \sigma = 868 - 0.152(T - T_m) \) dyn/m; \( \rho_l = 2375 \) kg/m³; \( c_l = 6187 \) m/s; \( \mu_l = 0.1492 \exp(1984.5/T) \) Pa-s.

The solubility of hydrogen in molten aluminum can be expressed as [25],

\[
S = 10^6 \exp(-2550/T + 2.62)
\]

Here, \( S \) is the solubility of hydrogen in aluminum.

The diffusion coefficient of hydrogen in molten aluminum is [26]

\[
D = D_0 \exp(-Q/R_S T)
\]
Here, $D$ is the diffusion coefficient (m$^2$/s); $D_0$ is the maximum diffusion coefficient at the infinite temperature (m$^2$/s); $Q$ is the activation energy for diffusion. Here, a group of values suggested by [26] are used: $D_0 = 3.8 \times 10^{-6}$ m$^2$/s; $Q = 19320$ J/mol. The constants are highlighted in Table 1 with the following acoustic parameters given: $P_\infty = 1.01 \times 10^5$ Pa; $f = 18$ kHz; $P_A = 1.1 \times 10^5$ Pa. For some cases, the polytropic exponent could not be regarded as a pure constant. Instead, it will depend on the external frequency and the bubble radius. For more details for the predictions of the polytropic exponent, readers could consider our previous model [21].

Table 1. Detailed values of the employed parameters.

| Parameters | Values   |
|------------|----------|
| $\rho_l$   | 2375 kg/m$^3$ |
| $T_m$      | 933.4 K   |
| $R_g$      | 8.314 J/mol/K |
| $T$        | 1073.15 K |
| $\eta$     | 1.2       |

Figure 1 shows the predictions of the threshold of rectified diffusion ($P_T$) based on Equation (16) for hydrogen bubbles in molten aluminum. The threshold is minimal near resonance and increases gradually for non-resonance conditions. For oversaturation conditions (e.g., $C_i/C_0 = 1.05$), the threshold is lower while for subsaturation conditions (e.g., $C_i/C_0 = 0.95$), the threshold is higher. In Figure 1, Eskin’s predictions [6] were not compared with ours because Eskin’s model cannot be used to predict the threshold phenomenon of the rectified diffusion owing to the assumption of gas always flowing into bubbles embedded in his model ([6], Equation (3)). It should be emphasized that for effective ultrasonic degassing, the amplitude of applied ultrasonic fields should be strong enough (i.e., above the threshold of rectified diffusion) to facilitate the growth of a gas bubble.

![Figure 1](image1.png)

**Figure 1.** Predicted threshold of acoustic pressure amplitude of rectified diffusion of hydrogen bubbles in molten aluminum. $C_i/C_0 = 0.95$, 1.00 and 1.05. $f = 18$ kHz.

Figure 2 shows the dynamic change of the equilibrium bubble radius of hydrogen bubbles in molten aluminum. The bubble growth rate (the gradient of the curves shown in Figure 2) firstly increases to a maximum value near the bubble resonance regime and then decreases quickly. It should be noticed that the growth of hydrogen bubbles is rather rapid on average, i.e., about fifteen-fold of the initial bubble radius after 0.1 s. When the bubble radius reaches a critical value (determined by the liquid densities), the dissolved hydrogen could be successfully removed through the bubble floatation. Figure 2 reflects the detailed process for hydrogen degasification in molten aluminum, which can be used for the further optimization of efficient ultrasonic degassing technique.
Finally, the validation of the present model will be briefly discussed. In our previous work, the present framework has been well proved by the experimental studies as shown in Figure 8 of Zhang and Li [9]. As shown in the above figure, the agreement between the theoretical predictions and the experimental data is fairly good. As a future work, an experimental system based on the high speed photography [27] will be employed to further reveal the interesting bubble phenomenon (e.g., micro-jet formation) and the complex fluid flow (e.g., vortex [28–30]). For related reviews on the cavitation phenomenon have been demonstrated through predictions of the behaviors of hydrogen diffusion processes of hydrogen bubbles in molten aluminum. Initial bubble radius ($R_0$) employed for the predictions are 14, 16 and 18 μm respectively. $C_i/C_0 = 1. f = 18$ kHz. $P_A = 1.1 \times 10^6$ Pa.

The limitations of the present model will be also further briefly discussed. When the acoustic wave passing through the liquids containing bubbles, many complex phenomenon will be induced by the mutual interactions between the acoustic waves and the bubbles. For example, the wave will be greatly attenuated by the oscillating bubbles within a short distance [33–35], leading to the frequency locking of the wave [34]. On the contrary, the acoustic wave could also manipulate the distributions of the bubbles through inducing the Bjerknes forces between them [36–39], leading to the coalescence of the bubbles. In the present model, for the sake of the simplicity, those effects are not accounted with neglecting the strong interactions between the waves and bubbles.

4. Conclusions

Rectified diffusion process of gas bubbles in molten metal during ultrasonic degassing has been theoretically investigated based on analytical analysis. Theoretical expressions of the threshold and the bubble growth (or dissolution) rate have been derived and interpreted. The features of the diffusion phenomenon have been demonstrated through predictions of the behaviors of hydrogen bubbles in molten aluminum. In the present work, the nonlinearity of the bubble oscillator (e.g., sub-harmonics [40,41]) is not fully considered, and its effects on bubble dynamics will be further explored in future work.

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