Short communication

An inverse method to fast-track the calculation of phase diagrams for sonoluminescing bubbles

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ABSTRACT
A sound driven air bubble can be transformed into an argon bubble emitting light pulses stably. The very foundation to investigate the sonoluminescing bubble is to accurately determine the ambient radius and gas composition in the interior. The conventional approach is to model the air-to-argon transformation process through a large number of bubble dynamics simulations to obtain the physical parameters of the ultimate argon bubble. In this paper, we propose a highly efficient method to pinpoint this information in a phase diagram. The method is based on the diffusive equilibrium for each species inside the bubble and derives the ambient radius and composition inversely. To calculate the former parameter, the bisection algorithm is employed to consecutively narrow down the searching range until the equilibria is approached. Afterward, several cycles of full dynamics simulations are conducted to refine the composition. The method is validated using published experimental data. The calculated ambient radii deviate from the test results by less than 1 μm, which falls within the margin of measurement error. The advantages of this method over the semi-analytical approach reported by Hilgenfeldt et al. [J. Fluid Mech. 365 (1998)] are also discussed. Our study provides a standard procedure to calculate the ambient radius and composition and is beneficial for the numerical simulation of sonoluminescing bubbles.

1. Introduction

Single bubble sonoluminescence (SBSL) refers to light emission from a levitated bubble trapped at the pressure antinode of a standing ultrasound. The catastrophic bubble collapse strongly compresses the gas inside, leading to intensive heating, gas ionization, and thermal bremsstrahlung in an extremely short time scale. A lasting interest centered on this subject has inspired sustained efforts trying to understand the detailed physics in SBSL as reviewed by Brenner et al. [1] and Suslick and Flannigan [2]. More recently, revived SBSL study has expanded to areas including exploring the emission in liquids other than pure water [3,4] and further refining the bubble dynamics model for numerical simulation [5,6]. One of the critical findings from the early SBSL studies is the parameter space within which the stably oscillating bubble can exist [7,8]. In this communication, we focus on the phase diagram of ambient radius versus acoustic pressure amplitude (R c –p c) in the allowable parameter space. This diagram is especially important for the numerical simulation of SBSL since the knowledge of R c as well as the gas composition of the bubble, must be first determined as a priori. As simple as it may seem, the ambient radius reflects profound physical characteristics of the sonoluminescing bubble. In a typical SBSL test, an air bubble is initiated in the fluid and pulsates under the driving pressure. In this process, the molecular constituents of the air would be dissociated and removed from the bubble in the course of numerous bubble collapses [8]. Through the chemical rectification, only the inert gas is left behind and stable light emission from the bubble is realized once every acoustical cycle [9,10]. Such an oscillating bubble is in diffusive equilibrium, meaning that each species inside is mass-conserving over each cycle. In this regard, the ambient radius and composition serve as the essential conditions for the diffusive equilibrium. Therefore, the ambient radius can’t be arbitrarily assigned but have to be calculated quantitatively. In practice, however, accurately determining the diffusive equilibrium following the air-to-argon transition route is a nontrivial effort. The difficulty is caused, firstly, by the large time scale of mass diffusion compared with that for bubble oscillation.
especially for the main species such as argon, oxygen, and nitrogen. Secondly, composition evolution due to chemical reactions is also slow. As a result, the air-to-argon transition requires thousands of acoustical cycles to be completed [8]. It is thus evident that modeling this process is rather computationally costly and time-consuming.

To accelerate the calculation, Toegel and Lohse [8] proposed using the Newton-Raphson algorithm to update the gas composition at the end of every acoustical cycle. With this method, they were able to reduce the required number of simulating cycles from thousands to hundreds. From the point of view of calculation efficiency, however, this forward method is still cumbersome. Especially at strong driving, the violent collapse and the extreme condition inside the bubble requires very small time steps to obtain calculation convergence, leading to exponential increase in the simulation time. In consequence, even the shortened simulation can be unacceptable in these cases.

In this paper, we propose an inverse method to quickly obtain the diffusive equilibria. We directly apply the mass-conserving requirement for each species to derive the ambient radius and composition. Our approach circumvents the time-consuming air consumption process and can thus greatly fast-track the calculation. On average, only ten acoustical cycles of simulation is enough to obtain the equilibrium at given acoustic driving and air concentration. Using this method, we are able to map out the phase diagrams corresponding to several typical SBSL tests efficiently and the theoretical results agree well with the experimental data.

2. The principle of the method

We assume the air bubble has already been transformed into a stable sonoluminescing bubble. At acoustic pressure \( p(t) = 0 \), the bubble has a radius \( R_0 \) and the total number of gas particles is \( N_{\text{total}} \). The aim is to determine the value of \( R_0 \) and the makeup of \( N_{\text{total}} \) at this moment. For this purpose, we first introduce several assumptions derived from previous investigations [8,11]. In these two studies, full dynamics simulations were performed for the sonoluminescing bubble and the validated results shed light on the gas composition. The liquid water employed in the simulations is partly saturated with air. Therefore, the derived results apply to the argon bubble under the condition of SBSL. The water temperature in Ref [8] is 21.7°C while in Ref [11] it is 3°C. Based on these conditions, it is concluded that for sonoluminescing argon bubble levitated in the water at or below room temperatures, the bubble contents at ambient radius have the following characteristics:

1. The main components are Ar, \( \text{H}_2\text{O}, \text{O}_2, \text{N}_2 \), and \( \text{H}_2 \). Accordingly, we only consider these species in the calculation. Among all the species, the fraction of argon \( f_{\text{Ar,0}} > 90\% \) and its absolute quantity is orders of magnitude larger than other species.

2. The rectified diffusion of \( \text{Ar} \) is zero;
3. For \( \text{H}_2\text{O}, \text{O}_2, \) and \( \text{N}_2 \), the net increased amount by diffusion balances the decreased amount due to dissociation;
4. The chemically produced amount of \( \text{H}_2 \) from water vapor dissociation equals the lost amount from diffusion to the surrounding water.

The dominance of argon is consistent with the dissociation hypothesis. For the remaining species, water vapor is diffused from the surrounding water by evaporation during bubble expansion. At collapse, the majority of vapors are condensed and expelled to the outside while others are dissociated [12]. The chemical products of water dissociation include a variety of radical species as well as oxygen and hydrogen [13]. The radical species are assumed as highly unstable and soluble in water, and they are thus neglected. Conversely, the diffusion of oxygen and hydrogen is rather slow and considerable amounts remain inside when

The bubble rebounds to the ambient size. For nitrogen, it is supplied from the dissolved air in the surrounding water through diffusion and partly dissociated at bubble collapse.

In case that a different gas other than air is dissolved in the water, such as a mixture of argon and nitrogen as adopted in the test of Ketterling and Apfel [14], the dominating species is always the inert gas since the molecular species would be removed according to the dissociation hypothesis. The remaining minor components except water vapor may need to be adjusted depending on the possible chemical reactions and products [15]. However, this variation will not affect the determination of the ambient radius \( R_0 \) as will be illustrated below.

2.1. The iterative method to determine \( R_0 \)

To determine \( R_0 \), we evoke assumption (2) and applies the diffusive equilibrium condition [7,19]:

\[
\frac{c_{\text{Ar,0}}}{c_{\text{Ar,∞}}} = \frac{\langle p_{\text{Ar}}(t) \rangle}{p_0}
\]

where \( c_{\text{Ar,0}} \) is the argon concentration in water and \( c_{\text{Ar,∞}} \) is its saturation concentration at ambient pressure \( p_0 \). In cases where the air concentration \( c_{\text{air,∞}} \) is specified, the argon concentration is assumed as \( c_{\text{Ar,∞}} = 0.009000 \text{ air,∞} \) based on the argon content in the air. \( \langle p_{\text{Ar}}(t) \rangle \) denotes the weighted time-averaged partial pressure of argon over an acoustical cycle \( T \):

\[
\langle p_{\text{Ar}}(t) \rangle = \frac{\int_0^T p_{\text{Ar}}(t) R^4(t) \, dt}{\int_0^T R^4(t) \, dt}
\]

Our strategy is to perform iterative bubble dynamics simulations with an initial guess of \( R_0 \). Based on the calculated \( \langle p_{\text{Ar}}(t) \rangle \), \( R_0 \) is adjusted successively until Eq. 1 is satisfied. Then the corresponding \( R_0 \) is assumed as \( R_0 \).

The numerical model for bubble dynamics simulation is introduced in the Appendix. As in the study of Toegel and Lohse [8], the model takes into consideration several key processes, including heat conduction, gas diffusion, and chemical reactions. In this section, however, the simulation is simplified in two ways to improve computation efficiency: the chemical reactions are ignored and only two species, i.e., argon and water vapor, are included inside the bubble. The rationale for this treatment lies in assumption (1), i.e., the partial pressure of argon is hardly influenced by other minor components due to the large disparity in the absolute amount. A separate simulation using the complete model found no difference for the diffusion flux of argon in a cycle (not shown here). It should be noted that water vapor may comprise a non-negligible part of the bubble contents, e.g. in the high-temperature liquid where the saturation vapor pressure is high. More importantly, as the bubble expands during the rarefaction phase of the acoustic driving, a large number of water molecules would enter the bubble through evaporation and their proportion may overtake the argon fraction [12,16]. Therefore, we included it in the simulation and stressed that the pressure terms appearing in Eq. 1 and Eq. 2 correspond to the partial pressure of argon.

The inclusion of water vapor gives rise to the question of what the quantity ratio of these two components should be. Fortunately, our calculation shows that when the argon fraction \( f_{\text{Ar,0}} > 90\% \), argon diffusion is not influenced significantly by the exact value of their
quantity ratio. Table 1 displays the normalized partial pressure of argon under different $f_{\text{Ar},0}$. The calculation is based on the SBSL experiment of Holt and Gaitan [9], where the acoustic frequency $f = 20.6$ kHz, the ambient pressure $p_0 = 1$ bar, and the water temperature $T_0 = 21.7^\circ$C. At air saturation $c_{\text{air,0}}/c_{\text{air},0} = 14\%$, the ambient radius $R_0$ measured from the test is 2.61 $\mu$m for $p_a = 1.29$ bar (Case 1\textsuperscript{t}), 3.52 $\mu$m for $p_a = 1.3$ bar (Case 2\textsuperscript{t}), and 4.03 $\mu$m for $p_a = 1.32$ bar (Case 3\textsuperscript{t}). For the investigated three cases, $<p_{\text{Ar}}(t)>_4$ changes by less than 6\% when $f_{\text{Ar},0}$ varies from 90\% to 98\%. For simplicity, we assume $f_{\text{Ar},0}$ as 95\% in this simulation.

After the initial composition is determined, iterative bubble dynamics simulations are performed to identify the ambient radius. In the searching of $R_0$, the bisection algorithm is employed to update $R_0$ at the end of each iteration as illustrated in the flowchart of Fig. 1. For most sonoluminescing bubbles $R_0$ lies in the range of 2 $\mu$m to 7 $\mu$m [9,14,17]. We take them as the lower and upper limit of the initial searching range, respectively. Note that in some SBSL cases, the initial searching range should be expanded according to the test conditions. Then, the search is executed by consecutively halving the range until the difference between the $(p_{\text{Ar}}(t))_4 / p_0$ and $c_{\text{Ar,0}} / c_{\text{Ar},0}$ falls within the tolerance.

In practice, this method proves to be highly efficient. In most cases, only 3 ~ 4 iterations are required to obtain convergence for the tolerance $e = 0.005$. The high efficiency is due to the monotonous relation between $R_0$ and $(p_{\text{Ar}}(t))_4 / p_0$ as displayed in the left of Fig. 1. Note that the horizontal line in the figure refers to the diffusive equilibrium condition according to the argon concentration applied in the test. The crossing point marked by the pentacle is the equilibrium that we target. Using the bisection method, the equilibria can be located quickly.

### 2.2. The full dynamic simulation to determine the equilibrium composition

With the knowledge of the ambient radius $R_0$, the next procedure is to determine the composition of the bubble content at $R = R_0$. We still use the sonoluminescing bubble in the test of Holt and Gaitan [9] as an example to illustrate this procedure. By performing the iterative calculations introduced in Section 2.1, the ambient radius is determined as $R_0 = 3.87 \, \mu\text{m}$ at $p_0 = 1.3$ bar. Furthermore, by applying pressure balance at the bubble interface specified by Eq. A3, the total number of particles $N_{\text{total}} = 8.19 \times 10^9$ at the ambient condition.

As noted in assumption (1), we only consider the following main species: Ar, H$_2$, O$_2$, N$_2$, and H$_2$O, with the condition that $f_{\text{Ar},0} > 90\%$. As a first guess, we assume the initial composition following the studies of Toegel and Lohse [8] and Yasui [11] as: $f_{\text{Ar},0} = 95\%$, $f_{\text{H}_2\text{O},0} = 4\%$, $f_{\text{O}_2,0} = 0.5\%$, $f_{\text{N}_2,0} = 0.4\%$, $f_{\text{H}_2,0} = 0.1\%$. Then, we activate the full bubble dynamics simulation including the chemical reactions, and observe the composition evolution. The particle number of each species at the end of every acoustical cycle is displayed in Fig. 2. As is expected, the quantity of argon is unchanged due to the inverse method in deriving the ambient radius. For other species, it is observed that their amounts quickly converge as the simulation proceeds. After seven acoustical cycles, their quantities are stabilized and the equilibrium composition is achieved.

To test the validity of this variation trend, we vary the initial quantities of the two main species, i.e., argon and water vapor, while leaving the fractions of other minor components unchanged. The result is
3. Application and discussion

Using the inverse method, we mapped out the phase diagrams for several previously reported SBSL tests [9,14,17] as depicted in Fig. 3. To obtain the ambient radius at specific test conditions, only the iterative bubble dynamics simulation without chemical reactions is conducted. We stress again that, on average, only 3 - 4 iterations are needed to obtain R₀ for each case. More importantly, the neglect of chemical reactions can significantly relax the time step required in the calculation. As a result, the computation efficiency is very high. The average error between the calculated and experimentally measured ambient radius ΔR₀ is 0.70 μm for the argon bubble in Ref [9], 0.40 μm for the nitrogen-xenon bubble in Ref [17], 0.35 μm for the nitrogen-xenon bubble in Ref [14], and 0.44 μm for another set of argon bubble in Ref [17]. The test parameters for each case are presented in the legend of Fig. 3. Given that the error bar of R₀ in these tests is about ± 1 μm, the deviation between numerical prediction and experimental result lies within the margin of test error, showing the accuracy and robustness of the proposed method.

To further check the reliability of the method, we referenced another SBSL test where the ambient pressure was varied whilst the acoustic pressure was kept constant [18]. The test parameters are: f = 17.5 kHz, p₀ = 1.29 bar, T₀ = 21°C, ε_air,0/ε_air,0 = 23.3%. The ambient pressure p₀ is reduced from 1 bar to 0.9 bar in the test. At each p₀, the ambient radius measured from the test and predicted from our calculation is displayed in Fig. 4. The liquid in the test is a mixture of degassed water and glycerin, while in the simulation we used pure water. Despite this difference, the increase of R₀ with the reduction of p₀ is well captured. The average error in R₀ between the numerical calculation and test measurement is 0.61 μm, which again demonstrates the reliability of the method.

In the last, we briefly discuss another semi-analytical method to calculate R₀ proposed by Hilgenfeldt et al. [19] and the difference compared with our approach. Both methods are in the same spirit, i.e., derive the bubble ambient radius inversely that meets the requirement of diffusion equilibrium for the inert gas. Their strategy is to relate \( \langle p_{\text{gas}}(t) \rangle / p₀ \) to the expansion ratio \( R_{\text{max}} / R₀ \):

\[
\frac{\langle p_{\text{gas}}(t) \rangle}{p₀} = \xi [1 + 2\sigma / (p₀ R₀)] \left( \frac{R₀}{R_{\text{max}}} \right)^4
\]  

(3)

where \( \sigma \) is the surface tension and \( \xi \) the constant that is determined by

The success of identifying the equilibrium composition with a few iterations can significantly relax the time step required in the calculation. The average error in R₀ between the numerical calculation and test measurement is 0.61 μm, which again demonstrates the reliability of the method.

### Table 2

| Case                  | Ar     | H₂O    | O₂     | N₂     | H₂     |
|-----------------------|--------|--------|--------|--------|--------|
| n₉₀,H₂O, n₀,H₂O,0 = 90:9 | 7.37 × 10³(97.9%) | 1.39 × 10³(1.85%) | 1.00 × 10³(0.13%) | 9.11 × 10³(0.12%) | 1.63 × 10³(0.0002%) |
| n₉₀,H₂O, n₀,H₂O,0 = 95:4 | 7.78 × 10³(97.9%) | 1.48 × 10³(1.86%) | 1.15 × 10³(0.14%) | 9.08 × 10³(0.11%) | 2.02 × 10³(0.0002%) |
| n₉₀,H₂O, n₀,H₂O,0 = 98:1 | 7.94 × 10³(97.9%) | 1.53 × 10³(1.89%) | 1.19 × 10³(0.14%) | 8.94 × 10³(0.11%) | 2.32 × 10³(0.0002%) |

### Fig. 3.

The phase diagrams calculated with the inverse method and compared with the data from the SBSL test. The test parameters in Holt and Gaitan [9] are T₀ = 21.7°C, f = 20.6 kHz, ε_air,0/ε_air,0 = 0.14. The estimated measurement error in the test is ∆R₀ = ± 1 μm and ∆p₀ = ± 0.025 bar, respectively. In Ketterling and Apfel [14,17], T₀ = 20°C, f = 33.4 kHz, ∆R₀ = ± 0.9 μm and ∆p₀ = ± 0.07 bar. The first data set (marked with the superscript ‘1’ in the figure legend, and so forth) is for the “artificial air” consisting of 99% N₂ and 1% Xe saturated 20% to the water. The second is for the “artificial air” consisting of 99% N₂ and 1% Ar saturated 10% to the water. The third data set is for the air saturated 10% to the water.

### Fig. 4.

Variation of ambient radius as a result of the reduction in the ambient pressure. The lines are the theoretical prediction from our calculation and the asterisk denotes the test data from Dan et al. [18]. The calculation parameters are introduced in the text.
the $R(t)$ curve. Another relation between $R_0$ and the maximum radius $R_{\text{max}}$ is derived with the knowledge of acoustic driving (Eq.4.34 in the Ref [19]). With two equations for the two unknown parameters ($R_0$ and $R_{\text{max}}$), the ambient radius under a certain acoustic driving and gas concentration can be calculated. We note that this approximation method is heavily dependent on the characteristics of the radius curve, e.g., the fitting of the overall curve as a parabola to estimate $\zeta$, as well as the division of the expansion section into a linear part and a decelerating part to establish the relation between $R_0$ and $R_{\text{max}}$. Although the $R(t)$ curve for sonoluminescing bubbles follows such a pattern in general, the details vary with different acoustic driving (e.g., under dual-frequency excitation), which may lead to deviation in the calculated $R_0$. Also, this method doesn’t differentiate the various gas components in the bubble. The whole bubble contents are treated as an ensemble and the amount is assumed as invariant during bubble oscillation, thus providing no physical meaning to the calculated result regarding gas composition and evolution.

4. Conclusion

In this paper, we introduced an inverse method to efficiently calculate the phase diagrams for stable sonoluminescing bubbles. This method uses an iterative algorithm to calculate the ambient radius based on diffusive equilibrium, and full bubble dynamics simulation to refine the composition of bubble contents. The former procedure can be accomplished within $3 \sim 4$ iterations while the latter needs only about 7 acoustical cycles of simulation. With this method, we mapped out the phase diagrams for several typical SBSL tests and validated the result using existing test data. Our approach can significantly reduce the computational costs in calculating the equilibria for sonoluminescing bubbles and provides a convenient tool for performing numerical simulations of SBSL.

CRediT authorship contribution statement

Kewen Peng: Conceptualization, Methodology, Investigation, Formal analysis, Funding acquisition, Writing - original draft. Frank G. F. Qin: Formal analysis, Investigation, Funding acquisition. Shouceng Tian: Project administration, Investigation, Funding acquisition. Yiqun Zhang: Investigation, Data curation.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix: THE NUMERICAL MODEL

The model is based on Toegel & Lohse [8], Toegel et al. [20], Yasui [11], and our previous study [16]. However, several modifications were made to improve the computation efficiency while preserving the robustness of the original models. We consider a sonoluminescing bubble oscillating in an acoustic field. The pressure and temperature inside the bubble are treated as homogenous. The heat transfer between the bubble and surrounding fluid is assumed to occur through a thin thermal layer inside the bubble near the interface. The diffusions of the various species are differentiated and modeled by different formulas according to the solubility. The chemical reactions involving 48 reactions and 19 species are accounted for.

The Gilmore model is employed to simulate the radial oscillation of the bubble. This model is second-order accurate when considering liquid compressibility and can well capture the detailed bubble dynamics during violent collapse:

$$
\left(1 - \frac{\dot{R}}{C}\right) \dot{R} \ddot{R} + \frac{3}{2} \left(1 - \frac{\dot{R}}{3C}\right) \ddot{R}^2 = \left(1 + \frac{\dot{R}}{C}\right) H + \left(1 - \frac{\dot{R}}{C}\right) \frac{R}{C} H
$$

(A1)

where $R$ is bubble radius, $C$ and $H$ is the sound speed and enthalpy of water at the bubble wall, and the overdot denotes the time derivative. The thermodynamic state inside the bubble is described by the van der Waals equation of state with the inclusion of a hard-core radius [20]:

$$
P_s = \frac{N_{\text{mub}} k T_B}{4 \pi \left[R^3 - (R_0 / 8.86)^3\right]}
$$

(A2)

in which $p_s$ and $T_B$ is the pressure and temperature inside the bubble, respectively. The ambient radius $R_0$ is defined as the bubble radius when the acoustical pressure is zero. It is a function of the total number of particles inside the bubble $N_{\text{total}}$ and can be calculated based on the pressure balance at the bubble wall:

$$
\frac{4 \pi}{3} \left[ p_0 + \frac{2 \sigma}{R_0} \right] \left[ R_0^3 - \left( \frac{R_0}{8.86} \right)^3 \right] = N_{\text{mub}} k T_B
$$

(A3)

where $\sigma$ is the surface tension and $p_0$ is the ambient pressure.

The scheme of chemical reactions is taken from Yasui [21] and described by Arrhenius correlations. For the diffusion of chemical products between the bubble interior and the surrounding liquid, we adopt a hybrid approach based on Toegel & Lohse [8] and Yasui [11]: for the moderately soluble species, i.e., Ar, O$_2$, H$_2$, and N$_2$, their diffusion flux per acoustical cycle is approximated by [8]:

$$
\Delta n_{ij} = 4 \pi D_{ij} T_{\text{mub}} C_{i,j} \left( \frac{c_{i,m}}{c_{i,0}} - \frac{p(t)}{p_0} \right)
$$

(A4)

where $D_{ij}$ is the diffusion coefficient of the species $i$ into the species $j$. $C_{i,j}$ is the bulk concentration of species $i$ in the liquid, $c_{i,m}$ and $c_{i,0}$ are the maximum concentrations of species $i$ inside the bubble and outside the bubble, respectively. $p(t)$ is the pressure in the bubble, and $p_0$ is the ambient pressure.
Table A1
Comparison of several key parameters between the full PDE calculation [12] and the present simulation. \(R_{\text{max}}\) and \(R_{\text{min}}\) is the maximum and minimum radius, respectively. \(T_{\text{max}}\) is the maximum temperature in the collapsing bubble, \(f_{i,0}\) and \(n_{i,0}\) is the fraction and molecule number of the water vapors at \(R = R_{\text{min}}\), respectively.

| Case | Model | \(R_{\text{max}}/R_{\text{min}}\) (\(\mu\m)) | \(T_{\text{max}}\) (K) | \(f_{i,0}\) or \(n_{i,0}\) |
|------|-------|---------------------------------|----------------|------------------|
| II   | Storey and Szeri [12] | 31.3/0.7 | 9700 | \(f_{i,0} = 14.0\%\) |
|      | Present model          | 31.1/0.78 | 10,013 | \(f_{i,0} = 14.1\%\) |
| IV(a)| Storey and Szeri [12] | 31.7/0.65 | 7000 | — |
|      | Present model           | 31.4/0.73 | 7079 | \(n_{i,0} = 7.29 \times 10^8\) |

where \(D_i\) is the diffusion coefficient of species \(i\), \(c_{i,0}\) is the saturation concentration, and \(c_{i,\text{eq}}\) is the actual concentration in water. For the radical species, their instant dissolution into the ambient liquid is estimated from kinetic theory by assuming an uptake coefficient \(\Theta\) [21]:

\[
\dot{n}_i = 4\pi R^2 c_i \Theta \frac{kT}{2m_i}
\]

where \(k\) is the Boltzmann constant, \(m_i\) is the molecular mass and \(c_i\) the concentration of the species \(i\) inside the bubble. Following Storey and Szeri [12], the coefficient \(\Theta\) is taken as 0.001. We note that the treatment for the diffusion of radical species is simplified with the uptake method. The extreme condition at the violent bubble collapse renders it rather difficult to accurately evaluate the diffusion rate for these species. In addition, the Arrhenius parameters in describing the chemical reactions have error bars in order of magnitude. Therefore, there is significant uncertainty in the exact quantity of radical species inside the bubble. The various methods proposed in other literature can be interpreted best as crude estimations. Here, we employ the uptake coefficient method for its simplicity and lesser computation requirement.

For water vapor transport across the bubble, the diffusion-limited model is used:

\[
\Delta n_{i,\text{vapor}} = 4\pi R^2 D_{i,0} \frac{c_i - c_i}{\text{lag}}
\]

where \(\text{lag}\) is the thickness of the diffuse layer, \(\text{lag} = \min(RD_{i,0}/[R, R/\pi])\), \(c_i\) is the vapor concentration inside the bubble and \(c_{i,0}\) the equilibrium concentration. The latter equals the vapor density at the bubble wall, \(c_{i,0} = p_i(T_0)/kT_0\) with \(p_i\) the vapor pressure at ambient temperature \(T_0\). The diffusion is assumed to occur between vapor and argon, the two principal components inside the bubble. Correspondingly, \(D_{i,0}\) is their binary diffusion constant and is determined from the kinetic theory of gases [22]. For other details of the model, the readers are referred to the aforementioned publications [8,11,16,20].

The model is validated against the widely-accepted calculation by Storey and Szeri [12]. In their study, they solved the full partial differential equations (PDE) governing the gas dynamics and obtained several key parameters for a sonoluminescing bubble (\(T_0 = 25^\circ\text{C}, f = 26.5\text{ kHz}, p_0 = 1.2\text{ bar}, R_0 = 4.5\mu\m\)). For the two investigated cases: evaporation and condensation take place but no chemical reactions (Case II), and evaporation and condensation, chemical reactions, and diffusion of radicals all take place (Case IV(a)), the results from both studies are presented in Table A1. The model agreement is impressive and shows the high accuracy of the present model.