Effect of liquid density on the Rayleigh-Taylor instability of sonoluminescing bubbles

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Rayleigh-Taylor instability of a radially oscillating gas bubble immersed in water is numerically studied. The model includes density changes in both gas within the bubble and liquid around its wall. The radial dynamics is described by Gilmore equation, which is coupled with the van der Waals (gas) and Tait (liquid) equations of state for simulating density variations. Shape instability is analyzed by the classical Hill’s equation, which describes the time evolution of the distortion amplitude. During the last moments of collapse, a sudden rise in gas and liquid densities is predicted; this exerts a stabilizing effect on the Rayleigh-Taylor instability. An instability borderline set by a new overwhelming criterion is in good agreement with an experimental boundary.

Keywords: Rayleigh-Taylor instability; single bubble sonoluminescence; surface distortion.

Se estudia numéricamente la inestabilidad de Rayleigh-Taylor de una burbuja de gas que oscila radialmente y está inmersa en agua. El modelo incluye cambios de densidad tanto del gas contenido en la burbuja como del líquido en su superficie. La dinámica radial se describe con la ecuación de Gilmore; la cual, esta acoplada a las ecuaciones de estado de van der Waals (gas) y Tait (líquido) para simular las variaciones de densidad. La inestabilidad superficial se estudia mediante la evolución de la amplitud de una distorsión descrita con la ecuación de Hill de la teoría clásica. Durante los últimos instantes del colapso, se predice un aumento súbito en las densidades del gas y del líquido ejerciendo un efecto estabilizador sobre la inestabilidad de Rayleigh-Taylor. Una línea fronteriza de inestabilidad calculada con un nuevo criterio de aplastamiento se aproxima bien a una frontera experimental.

Descripciones: Inestabilidad de Rayleigh-Taylor; sonoluminiscencia de una burbuja; distorsión superficial.

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1. Introduction

In typical experiments of single-bubble sonoluminescence (SBSL), a gas bubble in a liquid is acoustically levitated and made to oscillate so violently that pulses of light are emitted at the time of collapse [1]. Stable SBSL is characterized by light emission in each period of the acoustic driving at precisely the same brightness for millions of cycles [2]. However, stable SBSL occurs in a narrow region of the parameter space \((P_a, R_0)\); where \(P_a\) is the driving pressure amplitude and \(R_0\) is the equilibrium bubble radius [3]. This region is delimited by processes as: dissolved gas diffusion, chemical reactions, and spherical shape instabilities. This paper is concerned with the study of the later. In particular, the Rayleigh-Taylor instability (RTI) is considered. The RTI occurs near the minimum bubble radius when gas is accelerated towards the fluid and acts on the timescale of nanoseconds [3]. In Ref. 4, it is suggested that the RTI of the bubble surface is responsible for the extinction of the sonoluminescence when \(P_a\) exceeds a certain threshold. In fact, the onset of the RTI is connected with the destruction of the bubble into fragments.

Some numerical studies on shape stability of oscillating gas bubbles have shown that the shape and location of the RTI threshold lines in the \((P_a, R_0)\) space are strongly influenced by diverse assumptions as: the value of the boundary layer thickness \((\delta)\) [5] and the type of gas compression that depends on the polytropic index \((\gamma)\) [6]. Also, the trend and/or location of the RTI boundaries are very sensitive to peculiarities of the radial dynamics model; simplified versions of Rayleigh-Plesset equation (RPE) might result in bad predictions of the stability zone [7]. Thus, it is desirable to improve the models by introducing some corrections. In Ref. 6 and 7, this was done by taking into account the variation of gas density inside the bubble; and it was found a considerable suppression of the RTI and the threshold lines with this improvement approach better to some experimental data. In Ref. 5, the RTI lines were computed by assuming a small non-spherical perturbation in the driving sound field; the numerical results are in good agreement with many experimental data.

In most literature including the above mentioned works, the liquid density at the bubble wall is considered a constant at room conditions. This approximation is well justified in most of the acoustic cycle, but when the bubble is close to its minimum radius, it is expected that liquid compressibility effects become important. The aim of this paper is to improve the shape stability analysis of a radially oscillating gas bubble by taking into account liquid density variations on its surface and gas density changes within it.
2. Model and methodology

2.1. Radial dynamics

The Gilmore model [8] describing the radial motion of a bubble is used.

\[
\left(1 - \frac{\dot{R}}{C}\right) R \ddot{R} + \frac{3}{2} \left(1 - \frac{\dot{R}}{3C}\right) \dot{R}^2 = \left(1 + \frac{\dot{R}}{C}\right) H + \left(1 - \frac{\dot{R}}{C}\right) \frac{R}{C} \frac{dH}{dt},
\]

(1)

where \(R\) is the bubble radius, \(C\) is the universal gas constant, \(\dot{R}\) is the effective polytropic exponent, \(H\) is the ambient liquid pressure, \(P\) is the speed of sound in the liquid, its density, and the pressure at the bubble

\[
H = \int p \rho^{-1} dP,
\]

(2)

\[
C = [c_0^2 + (m - 1)H]^{1/2},
\]

(3)

\[
P = P_g - \frac{2\sigma}{R} - \frac{4\mu}{R} \dot{R},
\]

(4)

Here \(R\) is the bubble radius; \(C\), \(\rho_i\), and \(P\) are the speed of sound in the liquid, its density, and the pressure at the bubble wall, respectively. \(H\) is the difference between the liquid enthalpy at the bubble wall and at infinity, and \(c_0\) is the speed of sound in the liquid at room conditions.

The pressure at infinity is \(P_{\infty} = P_0 + P_a \sin(2\pi ft)\), \(P_0\) is the ambient liquid pressure; \(P_a\) is the amplitude of the driving sound field.

It is assumed that gas pressure inside the bubble obeys a van der Waals type equation of state [6]:

\[
P_g = \left(\frac{P_0 + 2\sigma}{R_0^3} \right) \left(\frac{R_0^3 - h^3}{R^3 - h^3}\right)^\gamma,
\]

(5)

where \(R_0\) is the ambient bubble radius, \(h\) is the hard core van der Waals radius, and \(\gamma\) is the effective polytropic exponent.

Gas density \(\rho_g\) inside the bubble varies according to

\[
\rho_g = \frac{P_g M_g}{RT_g + P_g b},
\]

(6)

where \(R\) is the universal gas constant, \(M_g\) is the mass per mole of gas, and \(b\) is the excluded van der Waals hard core volume.

The following version of Tait’s equation [9] is used for modeling the liquid density \(\rho_i\):

\[
\rho_i = \rho_0 \left(\frac{P + B}{P_0 + B}\right)^{1/m},
\]

(7)

where \(B\) and \(m\) are constants depending upon the liquid, and \(\rho_0\) is the ambient liquid density.

In order to solve (1), it is necessary to know \(H\) as function of \(R\), which at the same time is function of \(R\). From (2) and (7) it is readily found that

\[
H = \frac{m (P_0 + B)^{m-1}}{(m - 1) \rho_0} \left[ (B + P)^{m-1} - (B + P_\infty)^{m-1} \right].
\]

(8)

2.2. Dynamics of the distortion amplitude

The growth or decay of a small initial shape distortion from the spherical bubble interface is expressed in a form of spherical harmonics expansion:

\[
r = R(t) + \sum_{n=2}^{\infty} \sum_{q=-n}^{n} a_n^q(t) Y_n^q(\theta, \phi),
\]

(9)

where \(R(t)\) is the undistorted bubble radius, \(Y_n^q\) is a spherical harmonic with \(q\) and \(n\) denoting its degree and mode respectively, and \(a_n^q\) is the corresponding amplitude. Thus, the differential equation that describes the time evolution of the distortion amplitude is [6]:

\[
\ddot{a}_n(t) + B_n(t) \dot{a}_n(t) + A_n(t) a_n(t) = 0,
\]

(10)

\[
B_n(t) = \frac{3 \dot{R}}{R^2} + \frac{\left[\left(n(n+2)\right) - \beta_n\right] 2\mu}{\rho_i R^3}
\]

\[
+ \frac{\beta_n - \left(n(n-1)\right) (n+2)}{1 + 2\delta/R^3} \frac{2\mu \dot{R}}{\rho_i R^3}
\]

\[
\times \left[1 + \frac{n+1}{\frac{n}{\rho_i}} \frac{P_g}{\rho_l}\right],
\]

(11)

where \(\beta_n = (n-1)(n+1)(n+2)\), dots denote time derivatives, \(\mu\) is the dynamic viscosity and \(\sigma\) the surface tension of the surrounding liquid. Equations (11) and (12) employ a bubble boundary layer type approximation (BLA) [3] with a thickness \(\delta\) defined by:

\[
\delta = \min \left(\sqrt{\frac{\mu}{\rho_i \omega}}, \frac{R(t)}{2n}\right),
\]

(13)

where \(f = \omega/2\pi\) is the frequency of the driving sound field.

2.3. Rayleigh-Taylor instability

The RTI is studied by following the Bogoyavlenskiy approach [10]; which consists of calculating the amplification factor \(a_n(t)/a_n(0)\) of an initial distortion of nano-meter sizes only during the primary collapse, which is defined as the period from the moment of maximum bubble radius \(t_{R_{\max}}\), when an initial perturbation \((a_n(0), \dot{a}_n(0))\) starts to grow, to the second bounce moment (as indicated by our previous numerical experiments) when the distortion \(a_n(t)\) stops growing.

The following criterion to determine the RT borderlines is adopted [3]:

\[
\left\{ \begin{array}{l}
t_{R_{\max}} < t < t_{R_{\min}} + t_{RT} \\
\max \left\{ \frac{|a_n(t)|}{R(t)} \right\} \geq 1,
\end{array} \right.
\]

(14)
The heat transfer rate. Nevertheless, the implementation of a more suitable since the bubble wall moves much faster than the final stages of its first and subsequent collapses-rebounds, the global dynamics of the bubble. But, when the bubble reaches the acoustic forcing is 32.8 kHz. Heat exchange between the bubble and the surrounding water is not taken into account the isothermal approximation temperature of the gas in the bubble is the same as that of the liquid. It is worthy of note that the isothermal approximation is assumed isothermal and for simplicity the gas compression during whole bubble oscillation is modeled by (6) and (7); of course $D_R$ is also a function of time.

Case III: The gas and liquid densities are varying as functions of time, so they are modeled by (6) and (7) respectively; in this case $D_R$ is also a function of time.

Figure (2) shows the computed RTI borderlines in the $(P_a, R_0)$ space for the three study cases. To the left of a RTI line the bubble is stable. The RTI line for case I has a negative slope, as well; it is located on the left and far from the experimental boundary. Clearly, this line significantly underpredicts the experimental threshold.

When including gas density variations, case II, the prediction shows a borderline located to the right of the line for

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**Figure 1.** Time dependence of the bubble radius in a typical single bubble sonoluminescence cycle, calculated by (1). The bubble has an equilibrium radius of $R_0=4 \mu$m, pressure and frequency of the driving are: $P_a=0.142$ MPa and $f=32.8$ kHz respectively.

which states that the distortion can overwhelm the bubble radius $R(t)$ within the collapse stage.

Here $t_{R_{\text{min}}}$ is the moment of minimum bubble radius, and $t_{RT}$ is set to 2 $\mu$s to cover the moment when the distortion stops growing. In Fig. (1) are indicated $t_{R_{\text{max}}}$, $t_{R_{\text{min}}}$ and $t_{RT}$ with the corresponding radial positions. It is worth noticing that both the displacement and velocity of an initial perturbation will affect the location of the RT borderline determined by (14).

As in Ref. 6, an initial perturbation of $(a_n(0)=10 \text{ nm}, \dot{a}_n(0)=0)$ represents well the actual imperfections that may occur, e.g., thermal fluctuations of molecules at the liquid-gas bubble wall [11].

Before calculating the RT borderlines, Eq. (1) was solved for the first 10 cycles to make sure that the bubble oscillations reached a steady state [12]. Then the RT boundaries are the result of an incremental search on the $(P_a, R_0)$ parameter space covering the ranges: $0.05 < P_a < 0.253$ MPa, $0.0 < R_0 < 7 \mu$m. The computational resolution to calculate the $(P_a, R_0)$ space was: $\Delta P_a=0.001$ MPa, $\Delta R_0=0.05 \mu$m.

The numerical results are compared to experimental data of Ketterling and Apfel [13], for argon bubbles in water. The liquid temperature and ambient pressure are at room conditions ($P_0=101.325$ kPa, $T_0=293.16$ K) and the frequency of the acoustic forcing is 32.8 kHz. Heat exchange between the bubble and the surrounding water is not taken into account and for simplicity the gas compression during whole bubble oscillation is assumed isothermal $\gamma =1$ [3]; i.e., the heat transfer is faster than the time scale of the bubble motion, thus the temperature of the gas in the bubble is the same as that of the liquid. It is worthy of note that the isothermal approximation holds in most of the oscillation cycle without affecting the global dynamics of the bubble. But, when the bubble reaches the final stages of its first and subsequent collapses-rebounds, the assumption of an adiabatic gas compression ($\gamma \approx 5/3$) is more suitable since the bubble wall moves much faster than the heat transfer rate. Nevertheless, the implementation of a numerical scheme by switching $\gamma$ would imply more difficulties in programming and longer computing time.

For modeling the distortion amplitude, the quadruplet mode (n=2) is considered. The remaining parameters for numerical simulations are set to: $c_0=1483$ m/s, $\sigma =0.073$ N/m, $\mu=0.001$ Pa s, $B=304.7$ MPa, $m=7$, $b=0.03219$ l/mole, $h=R_\text{g}/3.86$ m, $M_g=0.0399$ kg/mole, $R=8.3145$ J/mole K.

### 3. Results and discussion

Three study cases are considered:

**Case I:** The density ratio $D_R=\rho_g/\rho_l$ in (11) and (12) is equal to zero, employing the same formulas used in Ref. 3.

**Case II:** Gas density varies as a function of time and is modeled by (6), liquid density is taken as a constant $\rho_l=1000$ kg/m$^3$; of course $D_R$ is also a function of time.

**Case III:** The gas and liquid densities are varying as functions of time, so they are modeled by (6) and (7) respectively; in this case $D_R$ is also a function of time.

Figure (2) shows the calculated Rayleigh-Taylor instability borderlines for the three study cases. Symbols represent stability thresholds from experiments by Ketterling and Apfel [13], for argon bubbles periodically driven at $f=32.8$ kHz in liquid water at 293 K and at an ambient pressure of 0.101 MPa.

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**Figure 2.** Calculated Rayleigh-Taylor instability borderlines for the three study cases. Symbols represent stability thresholds from experiments by Ketterling and Apfel [13], for argon bubbles periodically driven at $f=32.8$ kHz in liquid water at 293 K and at an ambient pressure of 0.101 MPa.
Figure 3. (a) and (b) show the time development of the radius, and acceleration of a bubble during the brief stage of collapse and rebounce, for $R_0=4\mu m$ and $P_a=0.147$ MPa. $t=0$ indicates the moment of minimum bubble radius. (c) and (d) show the time evolution of the gas and liquid densities respectively. (e) shows the $D_R$ profile, and (f) shows the behavior of $A_2(t)$. Dotted lines correspond to case I, dash-dotted lines are for case II and solid lines are for case III.
case I, indicating a bigger stability region in the \((P_a, R_0)\) space and better agreement with the experiments, however this result remains conservative. The position change of line for case II, and the enlargement of the stability region are consequences of the stabilizing effect of gas density variations on the RTI according to [7]. Additionally, if liquid density varies, an extra widening of the stability zone in the \((P_a, R_0)\) space is found, see thick solid line in Fig. (2).

Although the line for case III indicates a stabilizing effect due to changes in \(\rho_g\) and \(\rho_l\), it does not fit well with the experimental boundaries. In fact, it over predicts the stability zones. Now, the effect of gas and liquid densities on the RTI from the \(A_n(t)\) coefficient in the third term of (10), is analyzed with major detail. As stated in Ref. 6, the density ratio \(D_R\) strongly affects \(A_n(t)\) changing the contribution of \(\dot{R}\), a dominant factor in the RTI.

In most time of a cycle or at \(P_a < 0.1\) MPa, \(D_R\) is small. But for larger \(P_a\) and when the bubble radius is near to its minimum, it becomes significant. Without considering gas density variations, case I, \(A_2(t)\) resembles a negative pulse implying instability, see dotted line in Fig. (3f). Figure (3c) shows the gas density profile for case II, which is identical to the \(D_R\) curve (but dimensionless); if \(R(t)\) equals the \(R_{\text{min}}\), the gas density reaches a maximum \((\sim 1200 \text{ kg/m}^3\) limited by the hard core van der Waals radius for argon bubbles) and \(A_2(t)\) becomes a positive pulse decreasing the amplitude of shape distortions, see dash-dotted line in Fig. (3f). In Fig. (3e) the \(D_R\) curve shows a peak with a local maximum near \(R_{\text{min}}\), this behavior is associated with the sudden rise of liquid density predicted by present model. Fig. (3d) shows the liquid density behavior, this resembles a pulse with a maximum \((\sim 2000 \text{ Kg/m}^3\) that even suggests transient solidification of water around the bubble wall, i.e., ice particles formation [14]. \(A_2(t)\) pulse shape curve for case III, solid line in Fig. (3f), is similar to that found for case II, but with less amplitude. As before, this implies suppression of the RTI.

It is important to note that the change in behavior of \(A_2(t)\) due to gas and/or liquid density variations are only true at the end of a violent collapse; in fact the RTI will occur during the subsequent afterbounces, i.e., after the first collapse [11].

This is true for the three study cases, Fig. (4) depicts how the RTI amplifies an initial distortion over the first bounce until reaching a maximum when bubble collapses for a second time. But at the same time, Fig. (4) shows that the maximal deformation \(\max |a_2|\) is attenuated by the increment in both gas and liquid densities near the collapse.

Figure (4a) indicates that without taking into account gas density changes \(\max |a_2|\approx1.12 \mu\text{m}\), now if gas density variations are considered \(\max |a_2|\approx 98 \text{ nm}\), this is a reduction \(\approx 91\%\). By including liquid density changes the predicted \(\max |a_2|\approx 40 \text{ nm}\); this is a reduction \(\approx 96\%\) with respect to case I.

Figure (5) shows a comparison between the RTI boundaries as computed with two different models of radial oscillations. Dash-dotted line, calculated by considering gas density changes and using a simplified RPE with an isothermal assumption \(\gamma=1\) [7], is compared with line for case II.

Clearly the RTI line calculated via RPE has a vertical slope and is located to the left of line for case II, implying stronger collapses and more RT stable bubbles. This result is explained by Gilmore equation, which contains correction terms for liquid compressibility (with a damping effect on the radial oscillations) that make it a better description than the

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**Figure 4.** At the top, \(R(t)\) versus time during collapse stage and the radial bounces following the collapse, for a \(R_0=4 \mu\text{m}\) bubble driven at \(P_a=0.132\) MPa and \(f=32.8\) kHz. Zero time corresponds to the moment of minimum bubble radius. The corresponding time history of the distortion amplitude \(a_2(t)\), for: (a) case I, (b) case II and (c) case III.

**Figure 5.** Influence of the modeling of the radial bubble dynamics on the determination of the Rayleigh-Taylor instability borderlines. Lines for cases II and III were computed from Gilmore model. Curves labeled as: isothermal R-P and reduced R-P were calculated from Rayleigh-Plesset model [7]. Symbols indicate stability thresholds from experiments [13] as explained in Fig. (2).
Figure 6. Comparison between the Rayleigh-Taylor instability borderlines as computed with different values of the new overwhelming criterion defined by (15). Symbols indicate stability thresholds from experiments [13] as explained in Fig. (2).

RPE. Dashed borderline was calculated with a RPE but including radial damping mechanisms as heat transfer. Now, the line is less conservative and shows a positive slope in agreement with the experimental trend at least for bubbles with \( R_0 \leq 3.5 \mu m \).

According to Fig. (5), the present model for case III, predicts a RTI line with positive slope for bubbles with \( R_0 \geq 3 \mu m \). However, the experimental RTI threshold suggests the contrary behavior, that is to say a negative slope curve especially for bubbles in the range \( 3 \leq R_0 \leq 5.5 \mu m \).

As stated before, the RTI borderlines determined by criterion (14) imply the rupture of the bubbles. Now, we propose an overwhelming criterion defined as

\[
\max \left\{ t_{R_{\text{max}}} < t < t_{R_{\text{min}}} + t_{RT} \right\} \left( \frac{|a_n(t)|}{R(t)} \right) = \alpha
\]

\[
0 < \alpha \leq 1,
\]

which, in principle for \( \alpha < 1 \), it would allow calculation of RTI threshold lines that imply shape deformed bubbles displaying radial oscillations over many acoustic cycles and even emitting light pulses. In other words, the bubbles would remain after collapse with no break-up and dissolution.

Figure (6) illustrates RTI lines calculated from three values of the new overwhelming criterion, i.e., \( \alpha = \{1, 0.5, \text{ and } 0.3\} \). When \( \alpha=1 \) the RTI line corresponds to that of case III in Fig. (2), for \( \alpha = 0.5 \) a RTI line slightly different from that of case III is obtained.

Nevertheless, for \( \alpha = 0.3 \) the prediction is a RTI line whose slope has changed from positive to negative matching well the experimental data.

4. Conclusions

During the last instants of a micrometric bubble implosion, the presented simulations have revealed a sharp rise in gas (inside) and liquid (around the surface) densities suppressing significantly the Rayleigh-Taylor instability.

In recent years researchers have focused their attention on producing SBSL in acid–water binary liquid mixtures. The main reason is that bubbles in these liquid media emit light pulses more intense than those produced in water. However, only few theoretical works on spherical stability of these bubbles have been published and these are based on simplified RPE models [15-17].

It is expected that the stabilizing effect of gas and liquid densities on shape distortions of typical sonoluminescing bubbles found here, appear and behave similarly in SBSL acid-water systems.

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