2015 International Nuclear Atlantic Conference - INAC 2015
São Paulo, SP, Brazil, October 4-9, 2015
ASSOCIAÇÃO BRASILEIRA DE ENERGIA NUCLEAR - ABEN
***ISBN*: 978-85-99141-06-9**

EXPERIMENTAL STUDY OF SINGLE TAYLOR BUBBLES RISING IN STAGNANT LIQUID MIXTURES INSIDE OF VERTICAL TUBES

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ABSTRACT

The present work reports an experimental study of single Taylor bubbles rising in vertical tubes filled with water-glycerin mixtures by using the pulse-echo ultrasonic technique. A 2m long acrylic tube with inner diameter of 24 mm was used in the experiments. Initially, the tube was sealed at the ends and filled partially with the liquid mixtures to leave an air pocket of length L_0 at the top end. A Taylor bubble was formed by the inversion of the tube. The rising bubbles were detected by ultrasonic transducers located at the upper part of the tube. The velocity, the length and the profile of the bubbles and the thickness of the liquid film around them were obtained from the ultrasonic signals processing. The liquid film thickness in the vertical tube was also determined by a graphic method that relates the bubble length L_b with the initial length of the air pocket L_0. It was observed that the bubble velocity decreased with increasing viscosity, while the film thickness increased. It was shown that the liquid film thickness determined by the graphic method fitted well the higher viscosities data, but overestimated the lower viscosities data. Additionally, the results indicated that some correlations developed to estimate the thickness of liquid films falling down inside/outside of tubes and down a plane surface could be applied to estimate the thickness of liquid films falling around Taylor bubbles in an Inverse Viscosity Number (N_f) range different to those considered in the literature.

1. INTRODUCTION

The understanding of the mechanics of liquid films is of great importance in situations involving two-phase flow where heat and mass transfer occur, which is absolutely relevant to the nuclear, petroleum and chemical industries. Among the factors affecting these transfer rates, there are the transfer coefficient and the interfacial transfer area, that depends on the wave motion at the gas-liquid interface and on the liquid film thickness.

Slug flow is characterized by long bullet-shaped bubbles, also called Taylor bubbles or elongated bubbles, which occupy nearly the entire cross-section of the pipe and a liquid slug between successive bubbles. The liquid moves around the bubbles in a thin film and expands at the rear of the bubble, inducing a liquid wake.

Experimental data for the thicknesses of falling films around Taylor bubbles are scarce in the literature and two studies can be highlighted. Nogueira *et al.* [1] employed the particle image velocimetry (PIV) and the pulsed shadow technique (PST) to measure the elongated bubble profile and the velocity profile in the liquid film. Llewellin *et al.* [2] used a dimensionless approach to rewrite some correlations available in the literature to estimate the film thickness, in accordance with an appropriate dimensionless number. Then their experimental measurements of the film thicknesses were compared with those correlations in order to evaluate their validities. However the experimental method employed to make these measurements are only able to be applied at the experimental apparatus used in this work, as will be presented in the sequence of the present paper. Llewellin *et al.*'s [2] method cannot be applied under usual conditions of two-phase flow.

It is common to consider that when the film around a Taylor bubble is thin, the local curvature of the tube can be neglected and the behavior of this film is the same of a liquid falling down the inner or outer surface of a cylindrical tube or down a plane surface (Goldsmith and Mason [3] and Brown [4]). This assumption of thin film is applicable in several engineering situations involving the ascent of Taylor bubbles in tubes filled with water or another low-viscosity liquid. Thus, the studies about liquid films falling down surfaces are of great relevance to the study of falling liquid films around Taylor bubbles and several correlations can be found in the literature to estimate the thicknesses of these films.

Nusseldt [5] by doing an analysis of the phenomenon of heat transfer in vertically falling films presented one of the first correlations to estimate the liquid film thickness (δ). Nusseldt's [5] relationships were obtained from the balance of the forces acting on an element of the liquid film with the assumption of viscous flow, where neither shearing nor wave motion of the liquid surface occur. The expression for the averaged falling film thickness was defined by:

$$\delta = \left(\frac{3\mu_L^2}{4\rho_L^2 g} Re_f \right)^{1/3}, \tag{1}$$

where μ_L is the dynamic viscosity of the liquid, ρ_L is liquid density, g is the gravitational acceleration and Re_f is the film Reynolds number.

The film Reynolds number is defined by:

$$Re_f = 4\frac{\Gamma}{\mu_L} \tag{2}$$

and

$$\Gamma = \rho_L \delta v_f, \tag{3}$$

where v_f is the velocity in the film.

Considering that this model can be applied to the case of films around Taylor bubbles, Llewellin *et al.* [2] showed that in the thin-film limit ($\delta << R$), $Re_f = Re_b$. Re_b is the bubble Reynolds number defined as $Re_b = \rho_L U_b D/\mu_L$, where U_b is the rising bubble velocity and D is the inner tube diameter.

Thus, Eq. 1 can be rewritten in the following form, according to Re_b:

$$\delta = \left(\frac{3\mu_L^2}{4\rho_L^2 g} Re_b\right)^{1/3} , \tag{4}$$

Karapanstios and Karabelas [6] and Lel *et al.* [7] summarized information generated from various studies, using different fluids and experimental devices and proposed correlations to estimate the liquid film thickness according to Re_f. In the thin film limit, Karapantsios and Karabelas [6] correlation can be written as:

$$\delta \left(\frac{\rho_L^2 g}{\mu_L^2}\right)^{1/3} = 0.214 Re_b^{0.538} , \tag{5}$$

and Lel *et al.* [7] correlation becomes:

$$\delta \left(\frac{\rho_L^2 g}{\mu_L^2}\right)^{1/3} = 1 + 0.321 Re_b^{0.47} . \tag{6}$$

Considering the definitions of the dimensionless inverse viscosity number ($N_f = \rho_L \sqrt{gD^3}/\mu_L$), Re_b and Fr and defining $\delta' = \delta/R$, where R is the inner tube radius, Eqs. 4 to 6 could be rewritten as follow (Llewellin *et al.* [2]):

- Nusseldt [5] Correlation:

$$\delta' = \left(6\frac{Re_b}{N_f^2}\right)^{1/3} , \tag{7}$$

- Karapantsios and Karabelas [6] Correlation:

$$\delta' = \frac{0.428 R_b^{0.538}}{\sqrt[3]{N_f^2}} . \tag{8}$$

- Lel *et al.* [7] Correlation:

$$\delta' = \frac{2 + 0.641 Re_b^{0.47}}{\sqrt[3]{N_f^2}}, \tag{9}$$

Furthermore, it is possible to write Re_b as a function of N_f and Fr:

$$Re_b = N_f Fr \tag{10}$$

Llewellin *et al.* [2] proposed a simplification for the universal empirical correlation presented by Viana *et al.* [8], considering the conditions where Fr does not depend on Eo ($Eo > 40$):

$$Fr = 0.34 \left[1 + \left(\frac{31.08}{N_f} \right)^{1.45} \right]^{-0.71}. \tag{11}$$

Equations 10 and 11 reveal that Fr is only a function of N_f in the cases where surface tension effects can be neglected ($Eo > 40$), allowing that δ' can be written as a fucntion of N_f only, in Eqs. 4 to 6 .

Since the optical techniques (PIV and PST) are difficult to be employed at all of the industrial applications, it is important to develop and optimize techniques able to measure the bubble profiles and the thicknesses of the liquid films around Taylor bubbles. An option for this may be the pulse-eco ultrasonic technique. Ultrasonic technique is non-intrusive, presents low costs and can be applied at high pressures and temperatures. Another advantage of this technique is that it can be applied to pipes and containers of different materials and to transparent or opaque fluids. This technique was just applied to measure the thicknesses of liquid films on surfaces (Lu *et al.* [9]).

De Azevedo *et al.* [10] studied single Taylor bubbles rising in a stagnant water column and concluded that the pulse-echo ultrasonic technique can be applied to directly measure the film profiles and the equilibrium thicknesses of the falling films around the bubbles. These authors also employed this technique to measure its rising or propagation velocities (De Azevedo *et al.* [11]).

Figure 1 presents a schematic of a Taylor bubble flowing in a liquid. A typical Taylor bubble can be divided into four parts: (1) an approximately hemispherical nose, (2) a body surrounded by a falling liquid film, (3) a tail, and (4) a wake. The body can be further subdivided: (2a) around the upper part, where the developing film is accelerating and thinning, and (2b) around the lower part, where the forces acting on the film are in equilibrium and the film's thickness (δ) and velocity profile are steady.

In the present work, the pulse-echo ultrasonic technique was used to measure the rising bubble velocities, the bubble lenghts and the thicknesses of the liquid films falling around

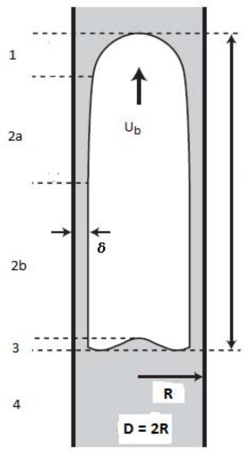

Figure 1: Schematic of a Taylor bubble flowing in a liquid.

Taylor bubbles in a stagnant liquid vertical column sealed at the ends. Water, glycerin and water-glycerin solutions were used as working fluids to evaluate the influence of the liquid viscosity in the measured parameters. Finally, these parameters were used to evaluate the correlations defined by Eqs. 7 to 9.

2. EXPERIMENTAL FACILITIES AND PROCEDURES

The experimental data were obtained from a vertical column partially filled with stagnant liquid located at the Thermo-Hydraulic Laboratory of the Nuclear Engineering Institute (LTE/IEN/CNEN). Figure 2 illustrates the vertical column that consists of an acrylic tube of 2.0 m long with inner diameter of 24 mm sealed at the ends. A Taylor bubble was formed by the invertion (t_1 to t_2) of the pipe partially filled with liquid to leave an air pocket of length L_0.

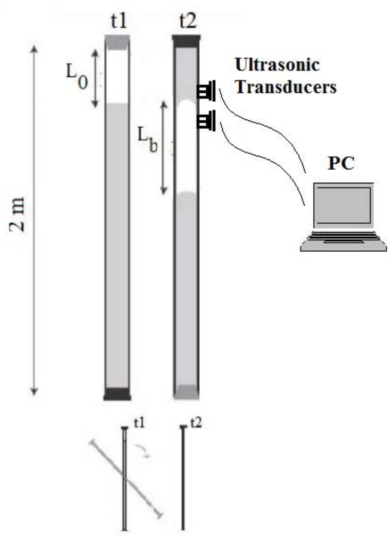

Figure 2: Schematic of the stagnant liquid vertical column used in this work.

This apparatus is similar to that used by Llewellin *et al.* [2]. These authors measured the bubble lenght L_b and plotted the relationship between the bubble lenght L_b and the inicial lenght of the air pocket L_0 for each set of data (inner tube diameter and working fluid). They found a linear relationship between L_b and L_0:

$$L_b = \alpha + \beta L_0, \tag{12}$$

where $\beta = (1-\delta^{'})^{-2}$ and α is a constant related to the lenght of the nose and tail regions. $\delta^{'}$ was defined by the authors as the ratio of the film thickness to the pipe ratio ($\delta^{'} = \delta$ / R) and was named *dimensionless film thickness* .

Using this procedure, Llewellin *et al.* [2] determined the liquid film thicknesses around Taylor bubbles. According to them, the results were in good agreement with the values measured by Nogueira *et al.* [1] employing optical techniques.

The high speed ultrasonic system used to measure the flow parameters consists of a generator/multiplexer board, transducers and a computer (PC) with a LabView software developed at the IEN to control up to four transducers in pulse-echo or transmission modes. Two ultrasonic transducers of 10 MHz and 6.35 mm diameter, Olympus piezoelectric-type transducers (Model V112), were mounted at the distance of 10 cm between them and they were located at 50 cm and 60 cm, respectively, from the top of the tube, except when glycerin was used as working fluid. In this case, the transducers were located at 70 cm and 80 cm from the top, respectively.

The pulse-echo ultrasonic technique is based on the high difference between the acoustic impedances of the gas and liquid fases, which allow that almost 99% of the incident wave be reflected by a gas-liquid interface. Thus, the location of a gas-liquid interface can be determined by measuring the transit time between the emission of a wave and its return after being reflected. The transit time can be obtained by the time interval between two consecutive reflections. Figure 3 shows a typical echogram registered by the ultrasonic system when an acrylic tube full with some liquid is analyzed.

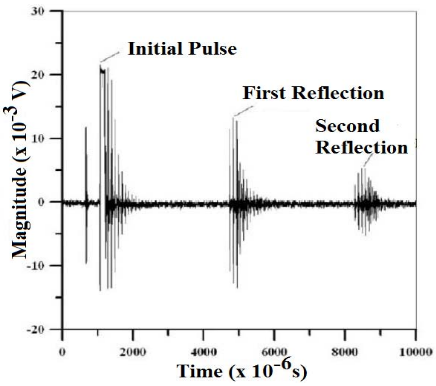

Figure 3: Typical echogram of the ultrasonic system.

In Fig. 3, one can observe the initial pulse that corresponds to the excitation of a transducer positioned at the outer wall of the tube. After crossing the tube wall thickness, it can be observed the signal related to the reflection at the tube inner wall (first reflection). Then, the pulse crosses all the internal tube diameter and is reflected by the inner tube wall on the other side of the tube (second reflection). Thus the best signal of the passage of the bubble can be detected between the first and the second reflections and the bubble profile can be obtained by the difference of the maximum intensity of the first reflection and the maximum intensity points at the gas-liquid interface of the bubble. Note that the system requires two reference points to perform the measurement of the transit time between two reflections.

It is important to note that in the pulse-echo ultrasonic technique, the transit time between two points corresponds to the time interval that the pulse is emitted, travels some distance, is reflected in an interface and travels back the same distance until being detected by the same transducer. Thus, the distance between the emission point and the interface where the pulse is reflected corresponds to a half of the total path of the ultrasonic wave.

By using a method based on the transit time, it is possible to measure the equilibrium thickness of the falling film around a Taylor bubble. The film thicknesses were determined by the relation: $\delta = T_f c_L/2$, where T_f is the transit time at the lower part of the film region (2b in Fig. 1) and c_L is the sound velocity in the liquid phase (De Azevedo *et al.* [10]).

In order to determine the liquid film thickness, the sound velocity through the water and glycerin-water solutions should be known. The sound velocities in the working liquids were measured by the relation: $2D/T_{r12}$, where D is the inner tube diameter and T_{r12} is the measured transit time between the first and second reflections (Fig. 3). As commented previously, these reflections correspond to diametrically opposed inner walls of the tube.

The bubble rising velocities were determined by the relation: $U_b = \Delta Z/\Delta t$, where ΔZ is the distance between two transducers and Δt is the time interval between the moments that a reference point of the bubble is detected by each of these two transducers (De Azevedo *et al.* [11]).

The lengths of the bubbles were determined by the relation: $L_b = U_b(t_t - t_n)$, where U_b is the measured bubble velocity and $(t_t - t_n)$ is the time interval between the moments that the tail and the nose of the bubble are detected by the same transducer.

For this work, 250 bubbles (50 bubbles for each air pocket length L_0) were studied using distilled water as working fluid. For the other fluids, 50 bubbles (10 bubbles per L_0) were studied for each of them. The percentages given in the compositions of the mixtures are by volume. During the experiments, the temperature remained between 24 and 26^{o}C.

The fluid properties were calculated by correlations given by Cheng [12] that calculate the density (ρ_L) and the viscosity (μ_L) of the glycerin-water mixtures in the range of 0-100% and temperatures varying from 0 to 100^{o}C.

3. EXPERIMENTAL RESULTS

The experimental conditions, including the liquid densities (ρ_L), the liquid viscosities (μ_L), and the dimensionless inverse viscosity numbers (N_f), defined as $N_f = \rho_L \sqrt{gD^3}/\mu_L$, for each of the liquids used, are summarized in Tab.1.

Table 1: Experimental Conditions.

Liquid	Water	Glycerin	ρ_L (kg/m^3)	μ_L (N.s/m^2)	N_f
1	100%	0%	997	0.0009	12900
2	80%	20%	1060	0.0017	7321
3	50%	50%	1144	0.0068	1952
4	20%	80%	1217	0.0629	224
5	0%	100%	1261	0.9875	15

$D = 0.024$m; $g = 9.81$ m/s^2

Figures 4 to 6 present the curves δ' vs. N_f relative to the models of Nusseldt [5], Lel *et al.* [7] and Karapantsios and Karabelas [6], respectivelly, together with the values obtained in this work by means of the ultrasonic technique.

Figure 4 reveals a good agreement between the experimental values and those estimated by the Nusseldt [5] correlation, with higher relative differences for the case of the highest viscosity liquid used in this work (glycerin). This fact can be attributed to the thin film assumption assumed for the use of the correlation.

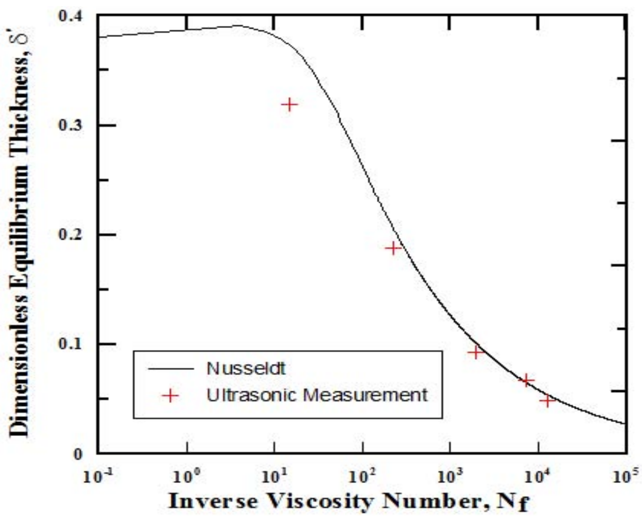

Figure 4: Relationship $\delta' vs. N_f$ for values measured by ultrasound technique and for the Nusseldt [5] correlation.

In Fig. 5, it can be observed, for the Lel *et al.* [7] correlation, similar results to those obtained for the case of the Nusseldt [5] correlation, with a high relative difference to the measured values, only for the case of glycerin. It is important to consider that the thickness increases greatly with viscosity increasing.

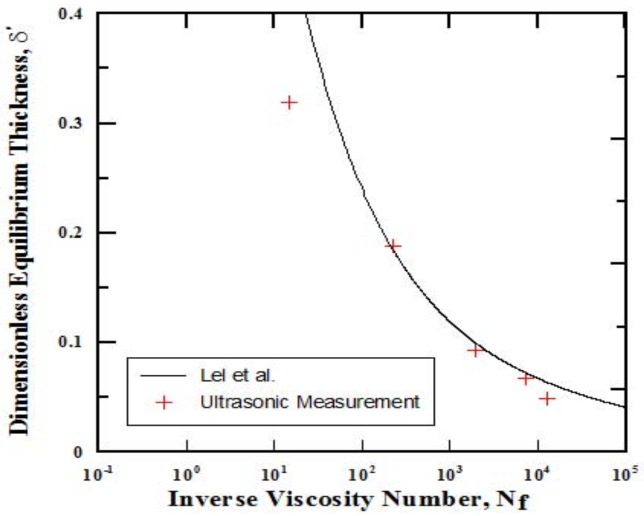

Figure 5: Relationship $\delta' vs. N_f$ for values measured by ultrasound technique and for the Lel *et al.* [7] Correlation.

For the Karapantsios and Karabelas [6] correlation, Fig. 6 shows a good agreement between the correlation values and the measured ones, in the range $1952 < N_f < 13000$ and a high relative difference between them to $N_f < 224$.

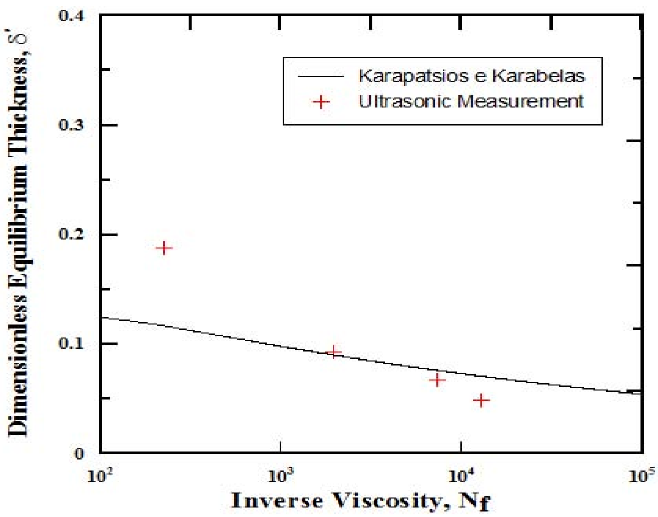

Figure 6: Relationship $\delta' vs. N_f$ for values measured by ultrasound technique and for the Karapantsios and Karabelas [6] Correlation.

Comparing the validity range of N_f for the use of these three correlations, obtained in the present work and those obtained by Llewellin *et al.* [2] , it was verified that there was a high discrepancy between them, especially for lower viscosity liquids or high N_f. Figure 7 presents the curve δ' vs. N_f relative to the empiric model of Llewellin *et al.* [2] and the values measured in this work, where it can be observed that there is no agreement between the values estimated by this correlation and those measured by using the ultrasonic technique, for high N_f. This correlation can be defined as:

$$\delta' = 0.204 + 0.123\tanh(2.66 - 1.15\log N_f) \quad (13)$$

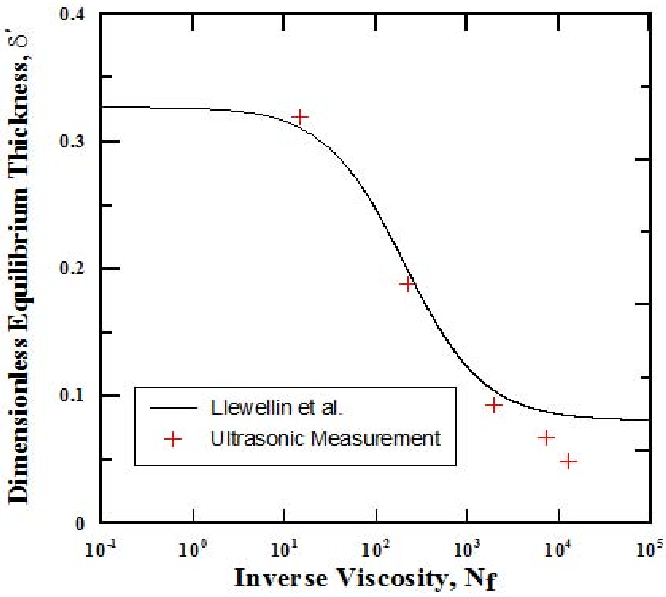

Figure 7: Relationship $\delta' vs. N_f$ for values measured by ultrasound technique and for the Llewellin *et al.* [2] correlation.

As commented earlier, Llewellin *et al.* [2] employed an apparatus similar to that used in the present work and determined the equilibrium thicknesses of the liquid films around Taylor bubbles by relating the measured bubble lengths L_b and the air pocket lenght L_0 that generated the bubbles, for each set of data (Eq. 12). Based on an analysis made by Nicklin *et al.* [13], these authors considered that the linearity observed in the L_b vs. L_0 relationship and that the fact that all bubbles studied by them had bubble lengths higher than six inner tube diameter ($L_b > 6D$) would guarantee that the liquid film has reached its equilibrium.

De Azevedo *et al.* [14] determined the equilibrium thicknesses of liquid films around elongated bubbles rising in different liquids by employing the same methodology used by Llewellin *et al.* [2] and by using the pulse-echo ultrasonic technique. All bubbles studied in that work had lengths greater than 6D and all set of data presented linear relationships for L_b vs. L_0. Nevertheless, a comparison between the values measured by the two methods for the same conditions indicated a good agreement for the cases of intermediate and high viscosity liquids (intermediate and low N_f) and a significant relative difference for the cases of low viscosity liquids or high N_f.

In order to better understand the differences between the values of the equilibrium film thicknesses measured by the ultrasonic technique and those determined by the L_b vs. L_0 relationship, an experiment was conducted, in which bubbles were generated in distilled water, from a larger number of air pocket lengths L_0. It was used 18 values of L_0 in the range of $1cm < L_0 < 40cm$. Figure 8 presents the L_b vs. L_0 relationship, for bubbles rising in water and generated from L_0 in the range described above. In case (a), the results was divided in values for $L_b < 6D$ and those for $L_b > 6D$, while in case (b) all results were presented as a single data set, covering a range of $0.78D < L_b < 19D$.

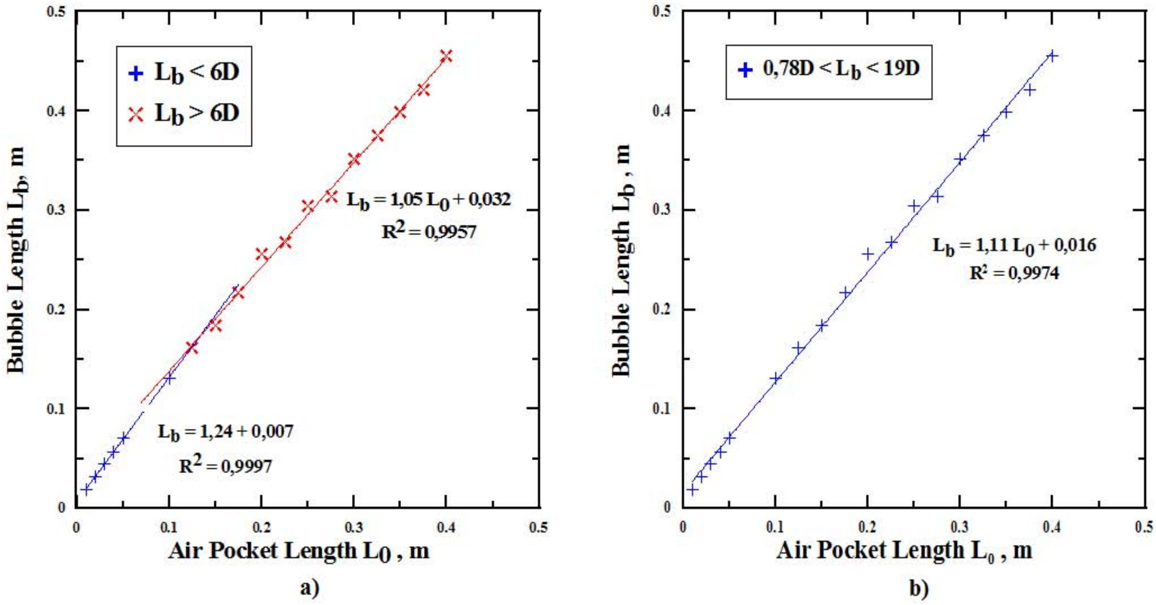

Figure 8: Relationship L_b vs. L_0 for different of air pocket lengths L_0: (a) Separated in $L_b < 6D$ and $L_b > 6D$; (b) Without separation.

The L_b vs. L_0 relationship observed in Fig. 8a presents a qualitative behavior similar to that described by Nicklin *et al.* [13], where can be detected a deviation from the linear relationship for $L_b < 6D$. This behavior induced Nicklin *et al.* [13] to consider that, for $L_b > 6D$, the bodies of the Taylor bubbles would have a perfectly cilindrical shape and, consequently, the liquid films had reached their equilibrium conditions.

However, in the present work it was verified that, for bubbles rising in distilled water, the liquid films do not reach their equilibrium for values of $L_b \approx 6D$. Figure 9 shows the perfect superposition of the film profiles (gas-liquid interface) for Taylor bubbles rising vertically in pure water and generated from different L_0. In this figure, it can be observed that the liquid films did not reach the equilibrium condition even for bubbles with lengths close to 19D.

In Fig. 8b, one can observe that it is possible to obtain a linear relationship for L_b vs. L_0, even including the points in which the films did not reach the equilibrium condition. The inclusion of this points have an effect to increase the angular coefficient of the linear relationship, which implies in the increasing of the value of δ' calculated by Eq. 12.

Thus, it was showed that the two premisses used by Llewellin *et al.* [2], in their study, as a warranty that the films have reached the equilibrium condition fail for bubbles rising in low viscosity liquids or high N_f. This can explain the disagreement of the values measured by the ultrasonic technique and those estimated by Llewellin *et al.* [2] correlation. Moreover, these results can explain the fact that this correlation overestimates δ' for the cases of high N_f.

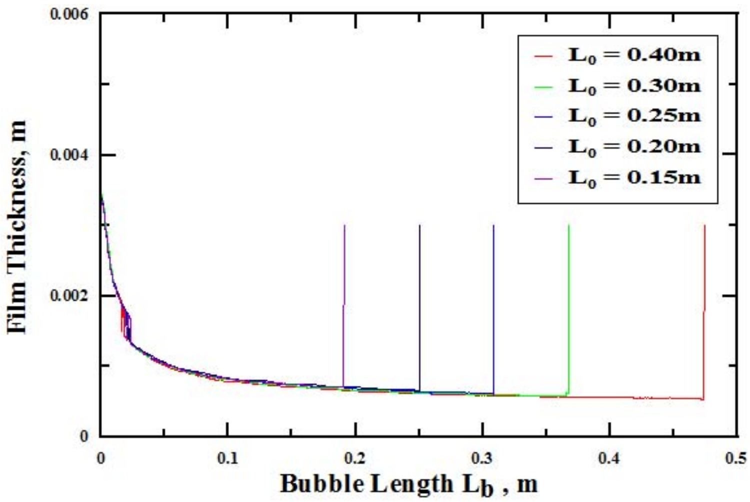

Figure 9: Superposition of the film profiles of Taylor bubbles rising vertically in pure water and generated from different L_0.

To reinforce these assertions, Fig. 10 presents the film profiles of bubbles rising vertically in liquids with different viscosities and generated from an air pocket length $L_0 = 15$ cm . It can be observed that, for the higher viscosity liquids, the equilibrium condition of the liquid film was reached for bubble lengths much smaller than 6D, which can explain the good agreement of the values measured in the present work and those estimated by the Llewellin *et al.* [2] correlation. Moreover, this fact explains the agreement between the values of the equilibrium film thicknesses obtained by De Azevedo *et al.* [14] by using the methodology employed by Llewellin *et al.* [2] and the ultrasonic technique, as commented earlier.

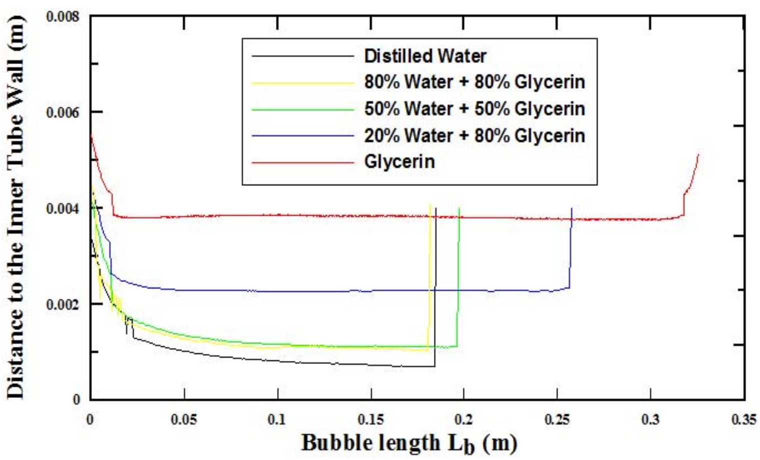

Figure 10: Liquid film profiles of bubbles rising vertically in liquids with different viscosities and generated from an air pocket length $L_0 = 15$ cm .

Table 2 summarizes the N_f validity ranges for the models and correlations discussed earlier. It can be verified that the results obtained in the present study extend the applicability of the models of Nusseldt [5] and Lel *et al.* [7], while restricted the model of Karapantsios and Karabelas [6] when compared with the results obtained by Llewellin *et* *al.* [2].

Table 2: Summary of the correlations and their validity ranges.

Correlation	Previous Literature	Llewellin *et al.*	Present Work
Nusseldt	$N_f < 3000$	$N_f < 3000$	$224 < N_f < 13000$
Lel *et al.*	$40 < N_f < 9000$	$40 < N_f < 9000$	$224 < N_f < 13000$
Karapantsios & Karabelas	$9000 < N_f < 44000$	$2000 < N_f < 20000$	$1952 < N_f < 13000$
Llewellin *et al.*	X	$0.1 < N_f < 100000$	$15 < N_f < 1952$

Previous Literature corresponds to review realized by Llewellin *et al.*.

4. CONCLUSIONS

This work used the pulse-echo ultrasonic technique to measure the rising bubble velocities, the bubble lenghts and the thicknesses of the liquid films falling around Taylor bubbles in a stagnant liquid vertical column sealed at the ends. Water, glycerin and water-glycerin solutions were used as working fluids to evaluate the influence of the liquid viscosity in the measured parameters. The liquid film thicknesses was also determined by a graphic method that relates the bubble lengths L_b with the initial lengths of the air pockets L_0.

It was showed that the graphic method employed by Llewellin *et al.* [2] overestimates δ' for low viscosity liquids or high N_f, but presents good agreement with the values measured by the pulse-echo ultrasonic technique, for intermediate and high viscosity liquids or intermediate and low N_f.

The results indicated that some models developed to estimate the thickness of liquid films falling down inside/outside of tubes and down a plane surface could be applied to estimate the thickness of liquid films falling around Taylor bubbles in an inverse viscosity number (N_f) range different to those considered in the literature. The measured values obtained in the present study extend the applicability of the models of Nusseldt [5] and Lel *et al.* [7], while restricted it for the model of Karapantsios and Karabelas [6], especially when compared with the results obtained by Llewellin *et al.* [2].

ACKNOWLEDGMENTS

The authors are grateful to CNPq, FINEP and FAPERJ for the financial support.

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