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Onset of entrainment between immiscible liquid layers due to rising gas bubbles

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Abstract—The phenomenon of the onset of liquid entrainment by gas bubbles rising vertically across the interface between two immiscible liquid layers is addressed both analytically and experimentally. An analytical model is developed which predicts the minimum gas bubble volume necessary to initiate the onset of entrainment. This model predicts that the phenomenon is characterized by a four-region flow regime. The criterion is based upon the bubble wake dynamics and the physical and transport properties of the two liquids. When compared with the experimental data for eight immiscible liquid pairs, the model and the data exhibit excellent agreement.

INTRODUCTION

In a number of applications, a gas is bubbled up through layers of immiscible fluids. This situation is found in such diverse applications as metallurgical processing and nuclear reactor safety. In the first case, oxygen, argon, SO_2, or other gases may bubble through a pool of molten metal with an overlying pool of molten silica slag. In the latter application, nuclear safety assessments are concerned with bubbles of non-condensable gases from concrete decomposition rising through overlying pools of molten fuel and metal. In all these occurrences, one is concerned with the possibility of entraining the denser liquid from the bottom layer into the top layer of lighter liquid by the rising gas bubbles. Such entrainment can significantly increase both heat and mass transfer between the two immiscible liquid layers, a phenomenon which could be either detrimental or desirable depending upon the specific application.

Previous investigations reported in the literature indicate that entrainment between the liquid layers is clearly observed in some cases while apparently not present in others. Szekely [1] deals with the case of a bubble-stirred interface between the two immiscible liquids without entrainment. Porter *et al.* [2] report observations of aqueous solutions overlying liquid mercury with no apparent entrainment of the mercury into the aqueous phase. Poggi *et al.* [3] report experimental indications of entrainment occurring between pools of glycerine/water over mercury, fused salt over lead, and slag over molten copper. More recently, Werle [4] and Greene and co-workers [5, 6] investigated the fluid mechanics and heat transfer behavior of silicone oil and water over liquid metal pools of Wood's metal and mercury, and silicone oil over water pools, both cases with gas bubbling from below. It was found in both investigations that the rates of heat transfer between the two overlying immiscible liquid layers driven by gas bubbles rising through the interface were strongly dependent upon whether the system exhibited entrainment or stratification. The gas bubbling was able to support entrainment for the oil over water system, however the water/oil over Wood's metal/mercury systems remained stratified with no mass transfer between phases. As reported by the authors, the fluid systems which underwent entrainment exhibited rates of interlayer heat transfer that greatly exceeded those measured for the fluid systems that remained stratified. Epstein *et al.* [7] tested stratified pools of organic liquid and water with nitrogen as the bubbling gas. These authors reported that a well-mixed emulsion of the two liquid phases was formed upon exceeding some critical value of gas flux. Gonzalez and Corradini [8] and Suter and Yadigaroglu [9] also reported clear indications of mixing between stratified layers of liquids under certain operating conditions.

The accumulated evidence indicates that entrainment between the immiscible liquid layers could occur or not occur depending on the specific liquid system and operating conditions. The objective of this study was to develop a criterion for predicting the onset of interfacial entrainment between the liquid layers. A

NOMENCLATURE

g	gravitational body force	ρ	density
R	radius	ρ^*	density ratio (inequality (10))
S	shape factor for gas bubble	σ_{12}	interfacial surface tension
V	volume	ω	dimensionless gas bubble volume (equation (7)).
V_g^*	critical gas volume for penetration (equation (2)).		

Greek symbols

β	column contact angle (Fig. 4)
θ	film contact angle (Fig. 3)

Subscripts

1	lighter liquid (upper pool)
2	denser liquid (lower pool)
g	gas.

conceptual limiting criterion is developed and then compared with experimental observations.

PHYSICAL PHENOMENA

Consider a pool consisting of stratified layers of two immiscible liquids, as illustrated in Fig. 1. If gas is injected at the bottom of the pool and flows upward through the liquid layers, entrainment and mixing of the heavier liquid (density ρ_2) into the lighter liquid (density ρ_1) can occur, as reported above. Our objective is to develop a criterion that can predict the minimum gas flow needed to cause such entrainment. Clearly, the phenomenon is affected by the two-phase flow regime of gas in the lower liquid pool. As gas flux increases, the two-phase flow regime changes from bubbly flow, to churn-turbulent flow, annular flow, and eventually to dispersed flow [10]. Since our concern is with the minimum gas flow that would induce entrainment, attention is centered on the regime of discrete bubbly flow, as illustrated in Fig. 1.

In the first part of this study, a series of photographic records was obtained to aid in qualitative understanding of the physical phenomenon. Stratified layers of silicone oil over colored aqueous solutions of copper sulfate in a transparent 10 cm diameter column were used for these experiments. Single bubbles of air were injected at the bottom of the column and photographed as they penetrated the interface between the two liquids. Examples of the photographic records together with sketches of various stages of the phenomenon are shown in Fig. 2. It is

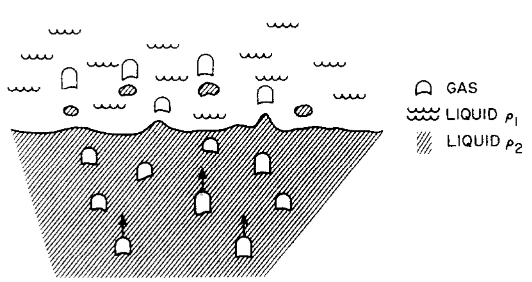

FIG. 1. Bubbly flow of gas through a pool of two immiscible liquids.

seen in Figs. 2(a)–(c) that as the gas bubble penetrates the liquid interface, it begins to drag a column of the denser fluid in its wake into the lighter fluid. As the bubble continues to rise (Figs. 2(d)–(f)), this 'wake' column elongates and then necks down and finally snaps to free a glob of the denser fluid which is then successfully entrained into the upper fluid by the rising gas bubble. In these experiments, depending on the size of the gas bubble, globules of the lower liquid would be so entrained or not entrained into the upper pool. In the latter case, a raised column (such as in Fig. 2(d)) could still occur but then the column would be pulled back into the lower pool by surface tension without necking down and breaking off as an entrained globule. It was also observed that small gas bubbles would sometimes be trapped at the two-fluid interface and be prevented from penetrating into the upper layer of lighter liquid.

The question to be addressed is, "Under what conditions does a rising bubble penetrate the interface between the two liquid pools and entrain some volume of the denser liquid into the lighter liquid." Our interpretation of the photographic records indicated that the entrainment process was caused by the rising gas bubble having sufficient buoyancy to penetrate the liquid–liquid interface and to drag in its wake a globule of the denser fluid into the upper pool, with sufficient force to overcome interfacial tension between the two liquids as well as the negative buoyancy of the denser fluid. In the following section, a criterion is developed based on this hypothesized mechanism to estimate the minimum size gas bubble able to cause entrainment for a given pair of immiscible liquids.

FORMULATION OF CRITERION

Consider a gas bubble of volume V_g as it rises through the lower liquid layer and approaches the interface. To successfully entrain fluid of density ρ_2 into the fluid of density ρ_1, the gas bubble must necessarily meet both of the following requirements:

(a) successfully penetrate the liquid–liquid interface and rise into the upper liquid layer;

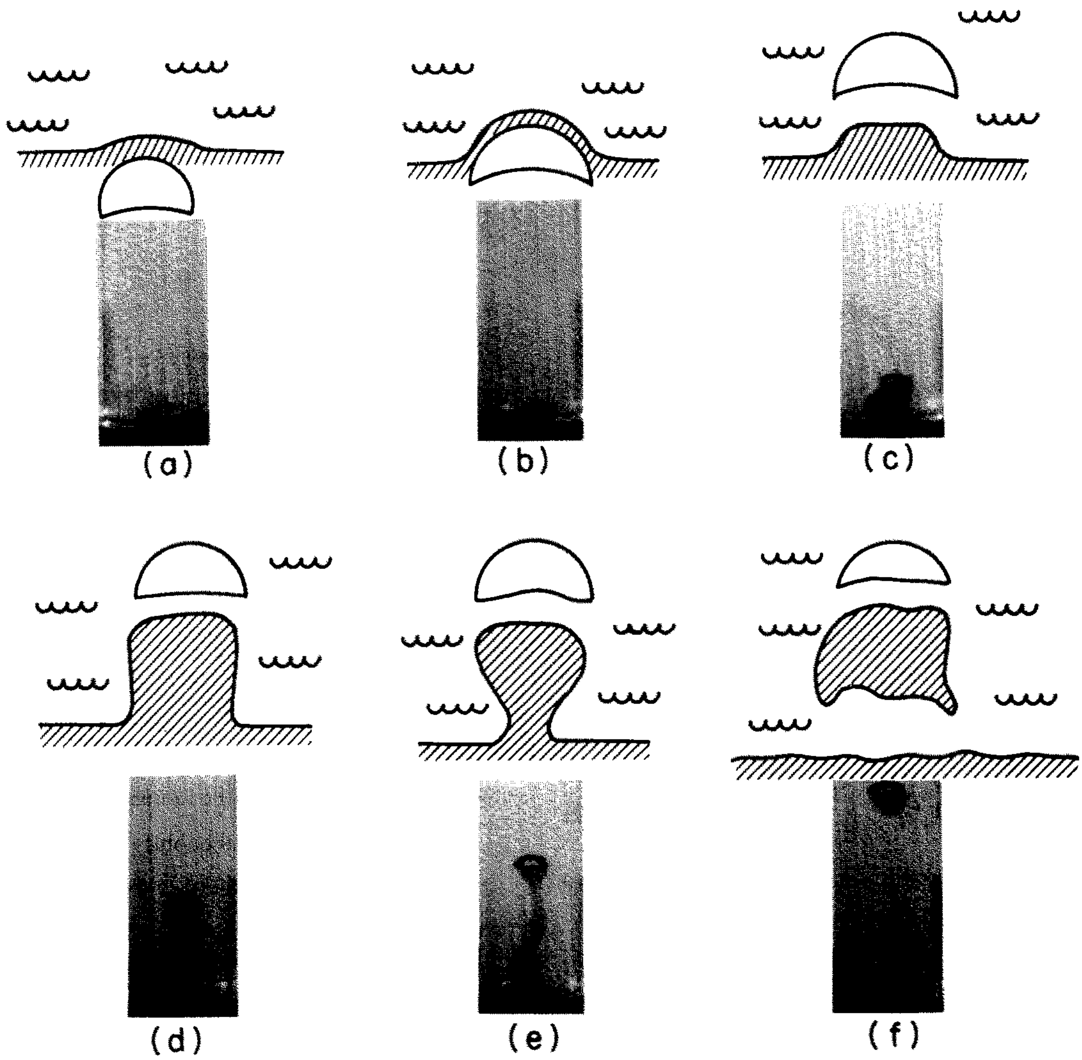

FIG. 2. Stages of the entrainment process.

(b) successfully drag a globule of the ρ_2 fluid in its wake, into the upper liquid layer, a sufficient distance so as to detach the globule from the liquid–liquid interface.

We will formulate the necessary criterion for each of these two requirements.

The following simplifying assumptions are taken as reasonable hypotheses:

(a) consider only individual bubbles, i.e. no interaction between successive or neighboring bubbles;

(b) cylindrical symmetry about the vertical axis of the rising bubble;

(c) one-dimensional motion in the vertical direction only;

(d) limit to low velocities so that inertial and viscous shear forces are negligible compared to buoyancy and surface tension forces.

Penetration

First consider a bubble of volume V_g as it passes the liquid–liquid interface. Visual observations in this investigation and elsewhere [9] indicate the tendency for a small 'film' of the ρ_2 liquid to surround the bubble as it attempts to penetrate the interface, as illustrated in Fig. 3(a). Neglecting dynamic effects, the forces acting on the bubble-film assembly are upward buoyancy and downward surface tension. The effective buoyancy force decreases as the bubble passes from the heavier ρ_2 fluid into the lighter ρ_1 fluid. At the same time, the downward component of the surface tension force increases as the angle θ changes from 0 to $\pi/2$. Therefore, the critical moment is when all of V_g is in the upper layer and the angle θ is equal to $\pi/2$, as indicated in Fig. 3(b). For successful penetration by the gas bubble, this minimum net upward force must be greater than zero. Neglecting the mass of the thin film around the bubble, and for the limiting case of a spherical bubble, the penetration criterion can be written as

$$V_g(\rho_1-\rho_g)g-3.90\sigma_{12}(V_g)^{1/3} > 0. \quad (1)$$

That is, for the gas bubble to penetrate the interface,

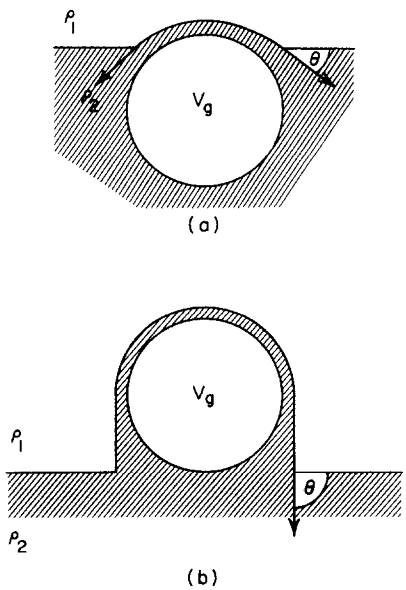

FIG. 3. Idealized concept for gas bubble passing a liquid–liquid interface.

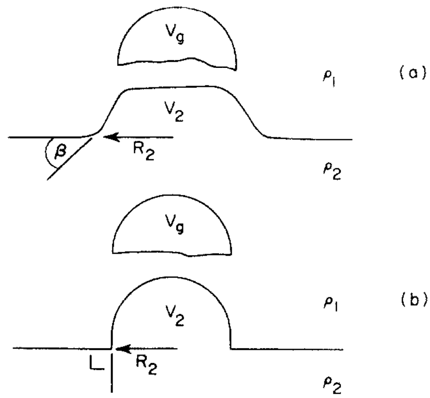

FIG. 4. Geometrical system for analysis.

its volume V_g must exceed a critical volume V_g^* given by

$$V_g^* = \left[\frac{3.9\sigma_{12}}{g(\rho_1 - \rho_g)}\right]^{3/2} \tag{2}$$

where σ_{12} is the interfacial tension between the two liquids.

Entrainment

Next consider a gas bubble of volume V_g after it penetrates the liquid interface, as shown in Fig. 4. Under buoyancy driven motion, the gas volume rises through the upper lighter liquid and attempts to pull in its wake a column of the heavier lower liquid. Let V_2 indicate the volume of the denser fluid pulled above the liquid interface and R_2 denote the column radius, with interfacial contact angle β as indicated in Fig. 4(a). A vertical force balance on the combined $V_g + V_2$ configuration indicates an upward buoyancy force of

$$\text{Buoyancy (up)} = [V_g(\rho_1 - \rho_g) - V_2(\rho_2 - \rho_1)]g$$

and a downward force due to surface tension of

$$\text{Surface tension (down)} = 2\pi R_2 \sigma_{12} \sin\beta.$$

For the entrainment process to continue successfully, the upward force must exceed the downward force, hence

$$V_g(\rho_1 - \rho_g) - V_2(\rho_2 - \rho_1) > \frac{2\pi R_2 \sigma_{12}}{g} \sin\beta. \tag{3}$$

The surface tension force is maximum when $(\sin\beta)$ approaches unity, corresponding to the situation shown in Fig. 4(b). In this state, assuming a hemispherical volume, the wake volume V_2 corresponds to

$$V_2 \geqslant \tfrac{2}{3}\pi R_2^3 \quad \text{at } \beta = \pi/2. \tag{4}$$

The photographic records indicate that the radius of the wake column (R_2) is approximately equal to the projection radius of the leading gas bubble. Hence one can write the approximate relationship between R_2 and V_g as follows:

$$R_2 = \left(\frac{3}{4\pi} V_g\right)^{1/3} S \tag{5}$$

where S is a shape function for the gas bubble, measuring its projection radius vs the radius of a sphere

$S = 1$ for a spherical bubble

$S > 1$ for a spherical-cap bubble.

For the minimum condition, take $S = 1$, resulting in the following necessary criterion for entrainment of ρ_2 fluid into the ρ_1 layer:

$$V_g > \left[\frac{7.8\sigma_{12}}{g(3\rho_1 - \rho_2 - 2\rho_g)}\right]^{3/2}. \tag{6}$$

We can define a dimensionless bubble volume by normalizing with respect to the minimum penetration gas volume (V_g^*), as given by equation (2)

$$\omega \equiv \frac{V_g}{V_g^*} = V_g \left[\frac{g(\rho_1 - \rho_g)}{3.9\sigma_{12}}\right]^{3/2}. \tag{7}$$

The criterion for interface penetration by the rising gas bubble becomes

$$\omega > 1 \tag{8}$$

and the criterion for possible entrainment of heavier liquid into the lighter liquid is

$$\omega > \left[\frac{2(\rho_1 - \rho_g)}{(3\rho_1 - \rho_2 - 2\rho_g)}\right]^{3/2}. \tag{9}$$

For most cases, the gas density is negligible compared to the liquid densities, and inequality (9) simplifies to

$$\omega > \left[\frac{2}{3 - \rho^*}\right]^{3/2} \tag{10}$$

where $\rho^* = \rho_2/\rho_1$.

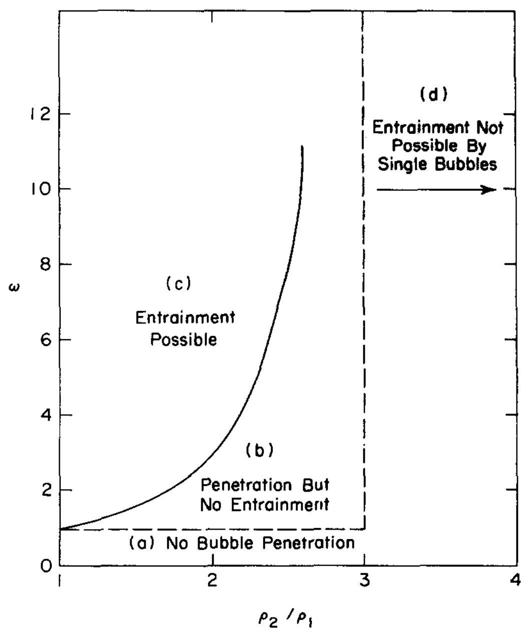

FIG. 5. Map of possible penetration and entrainment regimes.

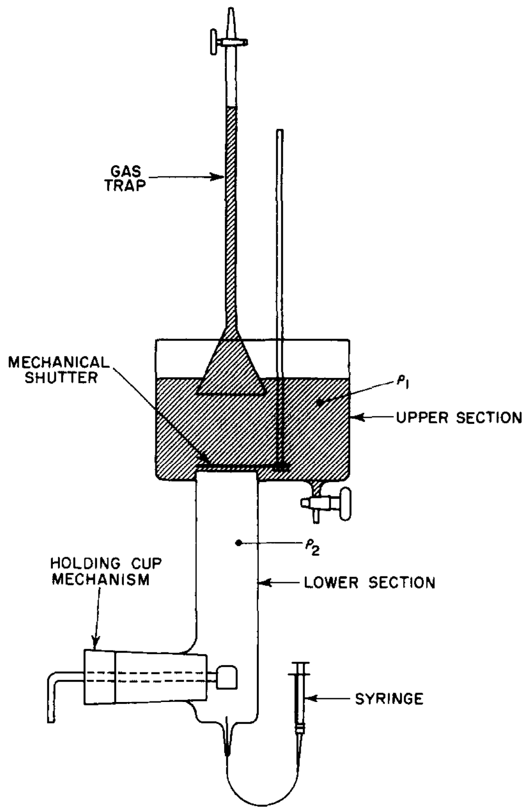

FIG. 6. Schematic diagram of experimental apparatus.

The results are plotted in Fig. 5, showing the regimes where:

(a) gas bubbles do not penetrate the liquid–liquid interface;

(b) bubbles penetrate the interface, but cannot entrain the heavier liquid into the upper layer of lighter liquid;

(c) entrainment is possible;

(d) entrainment is not possible by single bubbles by this mechanism.

Inequality (10) indicates that entrainment by this single bubble process is not possible when the ratio of liquid densities exceeds the limit

$$\rho^* > 3. \tag{11}$$

This is consistent with the observations of Porter *et al.* [2], Werle [4], and Greene and co-workers [5, 6] that no entrainment occurred for water over mercury ($\rho^* \simeq 13$) or for water over Wood's metal ($\rho^* \simeq 9.5$). It should be emphasized that this formulation treats only entrainment by single bubbles, giving a *necessary* criterion for entrainment. Thus, this deals primarily with applications at the lower range of gas flow rates, where bubbly flow exists.

EXPERIMENT

In order to test the criterion for bubble penetration and entrainment derived above, experiments were carried out with eight different pairs of immiscible liquids. For each pair of fluids, gas bubbles of varying sizes were injected to determine the regimes of entrainment and nonentrainment.

As illustrated in Figs. 6 and 7, the experimental apparatus consisted of a vertical glass column having two sections and provided with devices for injecting and measuring gas bubble volumes and for collection of entrained liquid globules. The denser and lighter liquids were contained in the lower (6.0 cm i.d. × 23.5 cm length) and upper (16 cm i.d. × 14.5 cm length) sections of the glass column, respectively. In operation, the liquid–liquid interface would be adjusted to lie just below the junction plane of the two sections. A mechanical shutter was mounted just above the junction plane to collect any volume of the denser liquid that is entrained into the lighter liquid.

Air bubbles of varying sizes could be injected at the bottom of the glass column by means of a micrometer syringe and holding-cup mechanism. After initial purging, a metered volume of gas would be injected by the syringe into the inverted holding cup. When ready, the cup would be quickly turned upward, allowing the injected gas to rise through the liquid layers as a single bubble. An inverted funnel, with attached burette mounted at the top of the test section, permitted trapping of the rising gas bubble and measuring its volume with a precision of ± 0.002 cm^3.

Table 1 lists the eight pairs of test fluids. The liquid properties listed were measured as a part of the test program. Densities were obtained by gravimetric measurements to a precision of ± 0.01 g cm^{-3} and surface tensions were measured with a Fisher Surface Tensiomat (Model 21) to a precision of

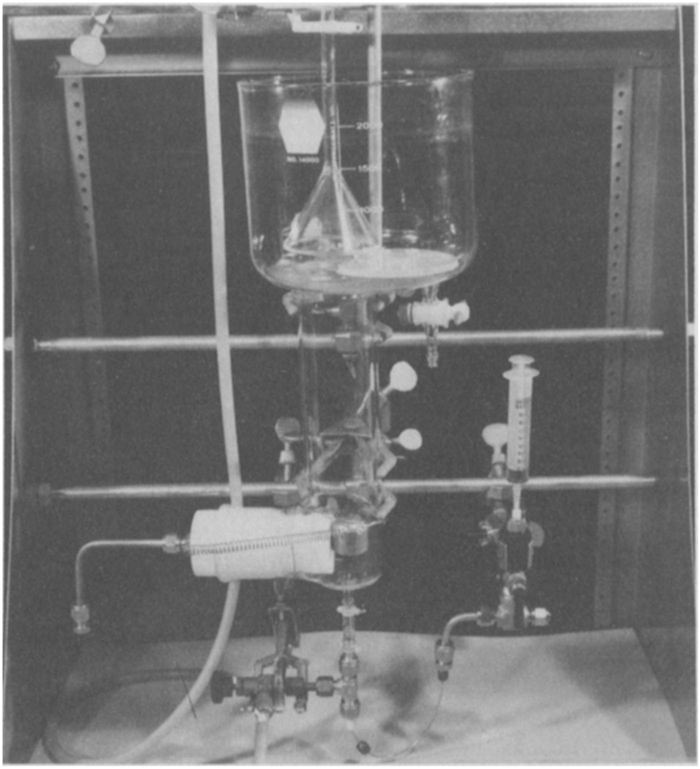

FIG. 7. Photograph of experimental apparatus.

Table 1. Test fluid properties

Fluid pair	Density ($g\ cm^{-3}$)	Surface tension ($dyn\ cm^{-1}$)
10 cs Silicone oil	0.94	10.8
Water (colored with $CuSO_4$)	1.04	
100 cs Silicone oil	0.97	31.1
Water (colored with $CuSO_4$)	1.01	
Water	1.00	24.4 (40–50°F)
R11 (refrigerant)	1.48	
Water	1.00	16.5
Bromoform	2.87	
Hexane	0.66	44.8
Water	1.00	
Glycerine	1.26	14.0
Bromoform	2.87	
Acetone	0.81	2.7
Glycerine	1.26	
100 cs Silicone oil	0.97	30.9
Glycerine	1.26	

± 0.05 dyn cm^{-1}. The tabulated values are averages obtained from ten measurements for each fluid. It should be noted that surface tension is sensitive to small amounts of impurities or additives (e.g. copper sulfate used to color the water) and direct measurements had to be obtained with the actual test fluids.

In the experiments, a pair of liquids would be charged into the apparatus and adjusted to set the liquid–liquid interface at the desired height. A specified volume of air would be injected into the holding cup, then permitted to rise as a single bubble through the two liquid layers. Visual observation and video recordings would be made during the time of bubble transit. The morphology observed was as qualitatively described above and illustrated in Fig. 2. Entrainment of the denser fluid into the upper liquid pool was discernible by eye and from the video films whenever it occurred. All experiments, with the exception of those using R11 refrigerant, were performed at room temperature. Due to the low boiling point, the R11 fluid had to be slightly refrigerated to prevent its vaporization during the tests.

RESULTS

Sample results are shown in Figs. 8 and 9 for the eight pairs of test fluids. The numerical results are listed in Table 2. Each point represents a test condition where multiple observations were made. Due to second-order perturbations, there was some stochastic variation in whether or not a specific bubble of a given size would induce entrainment. If bubbles of a given size were observed to cause entrainment in 75% (or more) of the repetitions, that condition was considered to be an 'entrainment' point. Usually, the bubble volume that satisfied this 75% threshold was also close to the minimum bubble size with observable entrainment. In Figs. 8 and 9, the cases with and

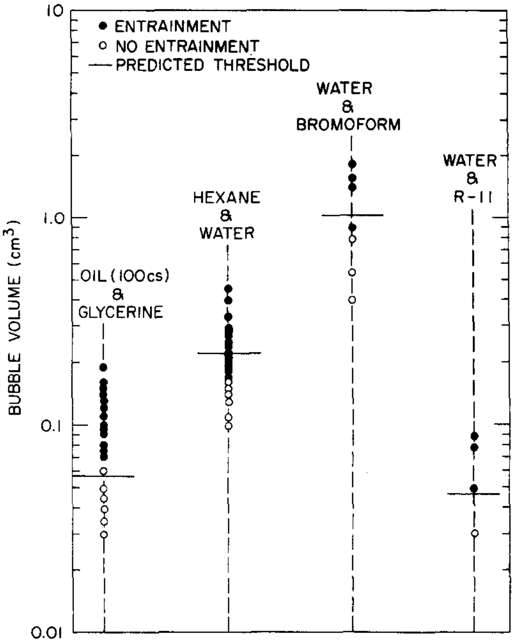

FIG. 8. Sample results: oil (100 cs)/glycerine, hexane/water, water/bromoform, water/R11.

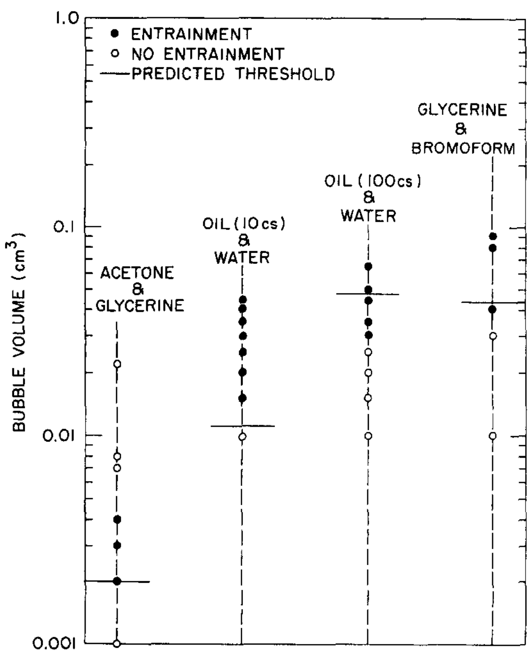

FIG. 9. Sample results: acetone/glycerine, oil (10 cs)/water, oil (100 cs)/water, glycerine/bromoform.

without entrainment are indicated by dark and open symbols, respectively. The threshold criterion for entrainment, given by inequality (6), is indicated by the horizontal line for each pair of test fluids. There is a 500-fold range in the predicted bubble volume for onset of entrainment for these widely different fluids. The results of Figs. 8 and 9 show generally good agreement between experiment and theory, with the onset of significant entrainment occurring close to the proposed thresholds. The acetone/glycerine fluid pair (Fig. 9) exhibited an interesting but unexplained quirk, that is an onset-of-entrainment transition followed by a subsequent return to conditions not supporting entrainment. This anomalous behavior has not been investigated further at present, and for the purposes of this paper, the first onset-of-entrainment volume was chosen for analysis.

According to the theory proposed above, the onset criterion for entrainment is affected primarily by just two physical property groups:

ratio of liquid densities, ρ^*

ratio of interfacial tension/liquid density, σ_{12}/ρ_1.

The eight pairs of liquids used in these experiments covered a 20-fold range in σ_{12}/ρ_1 and almost a 3-fold range in ρ^*. These data were examined to test the effects of physical properties on the entrainment onset criterion. Figure 10 plots the conditions corresponding to onset of entrainment determined experimentally for the eight pairs of test fluids as a function of the density ratio, ρ^*. The gas volume for onset, V_g, is normalized with respect to the penetration volume, V_g^*, to give the dimensionless onset volume, ω, as defined by equation (7). The points in Fig. 10 represent the smallest ω that caused significant entrain-

Table 2. Comparison of measured and calculated minimum bubble volume for entrainment onset

Fluid pair	Minimum bubble volume for entrainment onset† Measured‡	Measured range	Calculated
10 cs Silicone oil/water (colored with $CuSO_4$)	0.015	0.010–0.020	0.011
100 cs Silicone oil/water (colored with $CuSO_4$)	0.030	0.020–0.035	0.048
Water/R11 (refrigerant)	0.050	0.030–0.050	0.046
Water/bromoform	0.90	0.55–1.40	1.02
Hexane/water	0.17	0.15–0.21	0.22
Glycerine/bromoform	0.04	0.03–0.09	0.043
Acetone/glycerine	0.002	0.001–0.003	0.002
100 cs Silicone oil/glycerine	0.07	0.05–0.09	0.058

† Bubble volume in cm^3.
‡ Entrainment occurred in 75% of the cases at this volume.

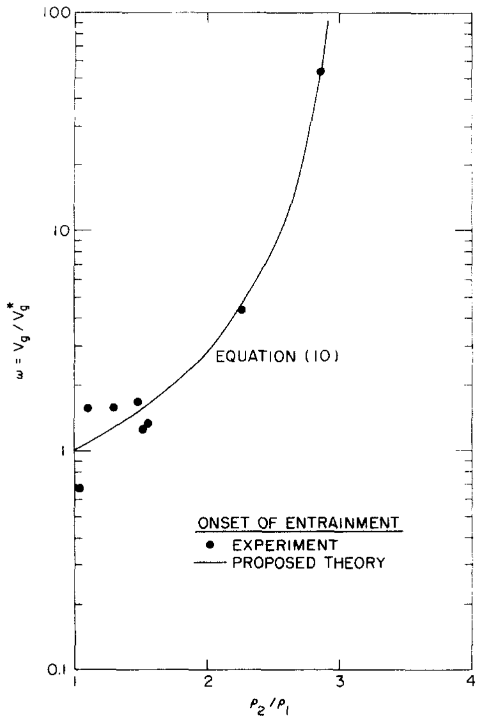

FIG. 10. Effect of liquid densities on onset of entrainment.

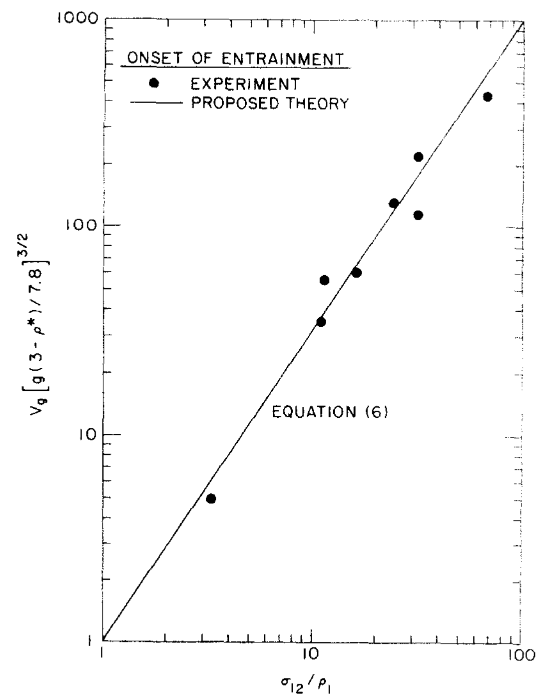

FIG. 11. Effect of interfacial tension/liquid density ratio on onset of entrainment.

ment for each pair of fluids. It is seen that the 3-fold range in ρ^* caused two orders-of-magnitude change in the onset ω. The curve on Fig. 10 represents the theoretical criterion for onset of entrainment, as given by inequality (10). Good agreement between experiment and theory was obtained; the theory not only predicted the parametric effect of ρ^* but also gave good estimates of the magnitude of ω.

Figure 11 examines the parametric effect of the property group, σ_{12}/ρ_1. According to the proposed theory, the group

$$V_g[g(3-\rho^*)/7.8]^{3/2}$$

should be equal to $(\sigma_{12}/\rho_1)^{3/2}$, where V_g is the threshold volume of the gas bubble to cause entrainment. The experimental measurements for the test fluids are plotted in Fig. 11 and confirm this prediction over a 20-fold range of σ_{12}/ρ_1. The theoretical prediction, inequality (6), is also plotted and shows very good agreement with the experimental results over the 100-fold range of the threshold volume. The axes in this figure are in the units expressed in Table 1 and Figs. 8 and 9. It is worth noting that the proposed theory, based on first principles, obtained this good agreement with measurements without the use of any empirical parameters.

SUMMARY AND CONCLUSION

The problem of entrainment between stratified layers of immiscible liquids caused by rising gas bubbles was examined to develop a criterion for onset of entrainment. Visualization experiments led to a hypothesis that entrainment by single bubbles is caused by the levitation of a small column of the denser fluid in the wake of the bubble as the bubble passes across the liquid–liquid interface. A first-principle analysis led to a theoretical criterion for the threshold volume of gas bubble necessary to cause entrainment, indicating a strong dependence on the liquid–liquid density ratio (ρ^*) and the interfacial surface tension/liquid density ratio (σ_{12}/ρ_1).

Experiments were conducted to measure actual onset of entrainment for eight different pairs of fluids. The experiments covered a 3-fold range in ρ^*, a 20-fold range in σ_{12}/ρ_1, and a 2000-fold range in the gas bubble volume. The proposed theoretical criterion, with no empirical parameters, was able to predict the experimental measurements with good agreement over the entire test range.

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ENTRAINEMENT ENTRE COUCHES DE LIQUIDES NON MISCIBLES DU A LA MONTEE DE BULLES GAZEUSES

Résumé—On étudie analytiquement et expérimentalement le phénomène d'entraînement de liquide par des bulles de gaz s'élevant verticalement à travers l'interface entre deux couches de liquides non miscibles. Un modèle analytique prédit le volume minimal de bulle de gaz nécessaire pour mettre en place l'entraînement. Ce modèle prévoit un régime d'écoulement à quatre régions. Le critère est basé sur la dynamique de sillage de bulle et les propriétés physiques et de transport des deux liquides. Le modèle et les données expérimentales s'accordent de façon excellente pour huit paires de liquides non miscibles.

DAS EINSETZEN DES ENTRAINMENTS ZWISCHEN ZWEI NICHT MISCHBAREN FLÜSSIGKEITSSCHICHTEN DURCH AUFSTEIGENDE GASBLASEN

Zusammenfassung—Es wird das Einsetzen des Entrainments analytisch und experimentell für den Fall untersucht, daß Gasblasen senkrecht durch die Grenzfläche zwischen zwei nicht mischbaren Flüssigkeiten aufsteigen. Es wird ein analytisches Modell zur Berechnung des minimalen Gasblasenvolumens entwickelt, bei dem das Entrainment einsetzt. Dieses Modell besagt, daß das Phänomen durch vier verschiedene Strömungsformen charakterisiert wird. Das Kriterium basiert auf der Blasennachlaufdynamik und den physikalischen Eigenschaften und den Transportgrößen der beiden Flüssigkeiten. Der Vergleich mit Meßwerten für acht nicht mischbare Flüssigkeitspaare zeigt hervorragende Übereinstimmung von Modell und Versuchsergebnissen.

УВЛЕЧЕНИЕ ЧАСТИЦ МЕЖДУ СЛОЯМИ НЕСМЕШИВАЮЩЕЙСЯ ЖИДКОСТИ В РЕЗУЛЬТАТЕ ПОДЪЕМА ГАЗОВЫХ ПУЗЫРЬКОВ

Аннотация—Аналитически и численно изучается увлечение частиц жидкости пузырьками газа, всплывающими вертикально через границу раздела между слоями двух несмешивающихся жидкостей. Разработана аналитическая модель, по которой рассчитывается минимальный объем пузырьков газа, необходимый для увлечения частиц. Показано, что явление характеризуется четырьмя режимами течения. Критерий основан на динамике следа пузырька и физических и транспортных свойствах двух жидкостей. При сравнении с экспериментальными данными для восьми пар несмешивающихся жидкостей показано соответствие между данными расчета и эксперимента.