Perfectly monodisperse microbubbling by capillary flow focusing: An alternate physical description and universal scaling

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(Received 11 June 2003; published 27 February 2004)

In a recent work [Phys. Rev. Lett. **87**, 274501 (2001)], a method to produce monodisperse microbubbles was described. The physics of the phenomenon was explained in terms of the absolute instabilities of a gas microjet formed when a liquid stream which surrounds a coflowing gas stream is forced through a small orifice. Now, a much more consistent physical picture to describe the phenomenon which corrects prior assumptions is presented. Consequently, a much simpler and universal scaling law for the microbubble size is finally obtained which involves the orifice diameter and the gas/liquid flow rates ratio only. All data shown in prior works, together with newly obtained data sets, have been analyzed anew. These are in remarkable agreement with the here proposed scaling law.

DOI: 10.1103/PhysRevE.69.027301 PACS number(s): 47.55.Dz, 47.20.Cq, 47.15.Hg, 47.55.Bx

Here we report a fundamental theoretical correction to a recent publication [1] where a method to produce monodisperse microbubbles was described. In that work, we presented the original method and a theoretical model where the phenomenon was explained in terms of the absolute instabilities of a very slender gas microjet. That jet was formed when a liquid stream which surrounded a coflowing gas stream was forced through a small orifice (see Fig. 1).

Owing to some unexplained discrepancies found earlier, in addition to those data presented in Ref. [1], we have performed subsequent experiments to explore those unclear parametrical occurrences. At a certain point, the departure between our prior model predictions and the new data was too conspicuous to be dismissed. This led us to reconsider our prior model assumptions from the very beginning in our published works [1,2]. In this work, a simpler and now consistent physical picture to describe the phenomenon which corrects prior assumptions is reported here, from which a simple and universal scaling law for the microbubble diameter d_b is finally obtained. This law involves the orifice diameter D and the gas/liquid flow rates ratio only, like it was already suggested in a heuristic fashion at the end of our prior work [1]. Here, we correct our prior model and give a closed explanation to that scaling law which was in good agreement with experiments but we could not explain in our previous work. All data shown in Ref. [1], together with newly obtained data sets, have been analyzed anew. These are now in remarkable agreement with the scaling law here presented. Starting again from the comparison of inertia to viscous forces for both liquid and gas streams, we refer again to the relevant Reynolds numbers of the flow, given by Eq. (1) in Ref. [1]. It was noticed earlier that while the liquid Reynolds numbers were large, the gas ones were always about the order unity [1]. However, the profound implications of this fact were somehow overlooked and we incorrectly embraced the assumption that the liquid and gas momenta per unit volume were of the same order, which immediately implies that the gas velocity was always much larger than the coflowing liquid velocity.

In the absence of a better understanding on the phenomenon, prior model assumed a characteristic evolution time $t_c = d_j D^2/Q_l$, where d_j was the assumed gas "jet" diameter, if such jet ever existed, D is the exit orifice diameter, and Q_l is the liquid flow rate through that orifice. The model uncertainties were collected into one single function that was justified to be dependent on the gas Reynolds number only (Eq. (2), see Ref. [1]). To obtain the gas jet diameter, in the absence of any sufficient resolution measurements, it ought to be "calculated" (not measured) from the real root of the approximate equation (3) [1], assuming that both the liquid and the gas flows were accelerated in the axial direction through the orifice to reach a situation where the transversal liquid pressure gradients were negligible, and the only difference in pressure between liquid and gas was given by the surface tension (which, by the way, would require that the jet was stable up to the exit orifice).

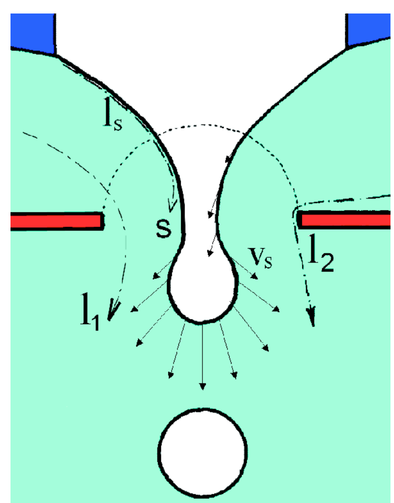

FIG. 1. (Color online) Sketch of the physical process. A liquid stream (streamlines l_1 and l_2) is forced through a small orifice of diameter D. A gas "core" stream is concentrically injected in the liquid stream through the hole, forming a row of perfectly monodisperse microbubbles. s and l_s are the curvilinear coordinate and the corresponding meridional streamline coinciding with the gas-liquid surface, and $\mathbf{v}_s$ is the velocity of liquid particles at that surface.

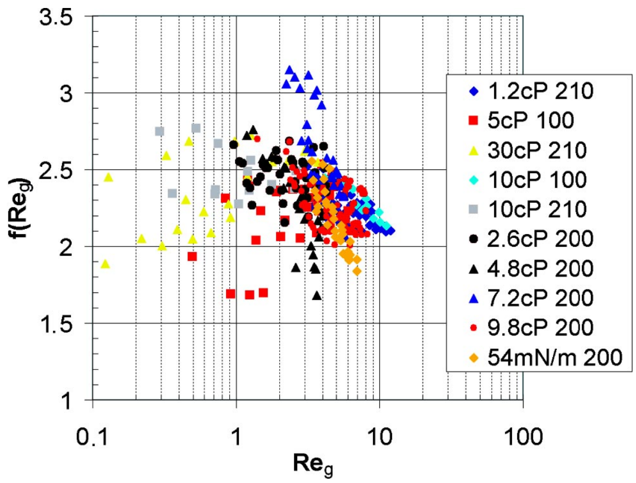

FIG. 2. (Color online) Function $f(Re_g)$. Labels indicate the following: (i) the number followed by "cP" is the liquid viscosity in cPoises, (ii) the number at the right is the orifice diameter in micrometers. The last label corresponds to a liquid with viscosity of 2.6 cP, and a surface tension of 54 mN/m.

Using that prior consideration, a collection of 416 data including old and new measurements have been replotted in Fig. 2.

This graph indicates a non-negligible, *inconsistent dispersion* of the pretended function $f(Re_g)$. In addition, the authors already noticed that the frequencies obtained from the linear analysis were rather incompatible with experiments, and the absolute instability wavelengths were extremely long, which would require the existence of a gas jet far much longer than the extremely short gas cusps (hard to be named jets) observed in reality.

The main error incurred in that previous model was the neglect of the radial pressure gradient in the liquid surrounding the expanding microbubble which forms just at the exit orifice from the attached parent bubble's cusp. This error was induced by the genuine capillary flow focusing configuration (liquid surrounded by gas, Ref. [3]), in which the imposed pressure drop through the orifice is equally applied to the liquid and the gas giving an almost entirely axial resultant (no radial pressure gradient). In that genuine configuration, the liquid jet is stable at the orifice exit because the liquid velocity is simply much smaller than the gas one. When the gas is surrounded by liquid, however, the latter prevents the former from acquiring a much larger velocity. In fact, by continuity, the gas feeding the microbubble must have a velocity at the liquid-gas interface of the same order than the liquid one (see Fig. 2). Thus, one has $O(\mathbf{v}_g)\sim O(\mathbf{v}_l)$, where $\mathbf{v}_g$ and $\mathbf{v}_l$ are the gas and the liquid velocities at the forming microbubble interface vicinity. It must be noted that, as we will see shortly, the liquid velocity at the liquid-gas interface (which moves radially at the vicinity of the expanding bubble) may not be of the order of the one close to the orifice borders, which is approximately axial and of the order of Q_l/D^2.

Contrarily to what was assumed in Ref. [1], the radial pressure gradient in the liquid is in reality the force per unit volume responsible for the radial liquid acceleration which leaves room for the expanding microbubble at the exit orifice (for an analogous phenomenon, see, for example, Ref. [6]). In the absence of this radial pressure gradient the rapid microbubble growth would be impossible. This pressure gradient comes from the pressure difference between the gas pressure inside the expanding microbubble and the surrounding liquid, approximately equal to the pressure drop through the orifice. This is so because at the exit orifice the liquid pressure decreases in lengths of the order of the orifice diameter to match the external pressure at the orifice exit. The liquid moving along streamlines close to the orifice borders, and thus far from the microbubble surface (see Fig. 1, streamline s_2), has a pressure approximately equal to the external pressure at the orifice exit, while the liquid moving along streamlines coinciding with the issuing microbubble surface (streamline l_s) must have a pressure equal to the gas pressure minus the surface tension force per unit surface, but this surface tension force is negligible since the Weber numbers $\mathrm{We}=\rho_l Q_l^2 d_b/\sigma D^4$ are large: our experimental data present a Weber number typically ranging from about 40 to about 1000. In fact, when we have now searched all our available experimental data for the minimum Weber number for which we have steady microbubbling conditions, we have never observed We less than 8, while We numbers about 500–1000 are common (this is why we could hardly make microbubbles in liquid metals using moderate pressures, i.e., below some hundred kilopascals).

Since the gas momentum in the expanding microbubble must be much smaller than the liquid one, the gas pressure must be very approximately equal to the stagnant one at the region behind the orifice, while the liquid has already experienced an expansion when it passes through the exit orifice. In other words, the gas pressure inside the expanding microbubble must be approximately equal to the gas pressure in the whole gas domain (i.e., the parent attached bubble, its cusp, and the expanding issuing microbubble, similarly to prior observations and models already established in the literature for analogous phenomena, see, for example, Ref. [6], p. 112; Refs. [7–9]). Besides, this pressure must be approximately equal to the liquid one in the stagnant region upstream of the exit orifice. This is one of the main conclusions that we want to highlight in this correction: the picture of a much faster gas stream in the gas core than the surrounding liquid is inconsistent. On the contrary, the gas pressure inside the expanding microbubble should be approximately equal to the liquid pressure at the stagnant region upstream of the orifice, neglecting the small pressure difference owing to the surface tension at the attached parent bubble.

Based on the above and since the liquid flow Reynolds numbers at the exit orifice are very large (typically from about 10^2 to about 10^3), one can write at the vicinity of the liquid-gas interface

$$\frac{\partial \mathbf{v}_l}{\partial t}+\mathbf{v}_l\cdot\nabla\mathbf{v}_l=\frac{D\mathbf{v}_l}{Dt}\simeq\nabla p_l, \tag{1}$$

where the unsteady term, reflecting the radial liquid motion at the expanding bubble surface, must be of the order

$$O\left(\frac{\partial \mathbf{v}_l}{\partial t}\right) \sim O\left(\frac{Q_g}{d_b^2}\frac{Q_g}{d_b^3}\right), \tag{2}$$

In contrast, the convective term, reflecting the liquid motion in the axial direction, must be of the same order across the whole orifice section, i.e.,

$$O(\mathbf{v}_l \cdot \nabla \mathbf{v}_l) \sim O\left[\left(\frac{Q_l}{D^2}\right)^2 \bigg/ D\right]. \tag{3}$$

The three terms of Eq. (1) must be of the same order given that (i) the strong oscillatory nature of the flow at the microbubble vicinity should make the unsteady term dominant there, (ii) the radial decrease of the oscillations should make the inertial term dominant close to the orifice borders, and (iii) the pressure gradient is the driving term. Consequently, one must have

$$O(Q_g^2/d_b^5) \simeq O(Q_l^2/D^5) \Rightarrow d_b/D = \eta(Q_g/Q_l)^{0.4}, \tag{4}$$

where η must be a universal constant. Our experiments collapse around $\eta=1.1$. It is worth noting how close this scaling is to the approximate heuristic one suggested at the end of our prior work [1], and how the latest scaling agrees with experiments (see Fig. 3).

A final clarification on the imposed gas flow rate Q_g is needed. To provide a smooth, continuous, and pulseless gas feed, in our experiments we have forced the gas through very small capillaries (capillary inner diameters about 20–40 μm and lengths from about 10 to 40 mm) connected to the gas feeding tube in our flow focusing device (Fig. 1). To calculate the gas mass flow rate through the capillary, we needed to know the liquid pressure surrounding the gas feeding tube (Fig. 1). This pressure was calculated from the imposed liquid flow rate (forced with a syringe pump or from a pressurized container whose weight was controlled with time), the orifice diameter, the Reynolds number, and Dagan's correlation [10]. Thus, the gas flow rate used in the scaling and the plots was calculated from the final gas pressure and density inside the observed and measured bubble, considering the liquid temperature, surface tension, and the surrounding liquid pressure. In practice, for liquids with moderate to small surface tension, the errors incurred in assuming a volumetric gas flow rate at atmospheric pressure are relatively small and thus one can use equation

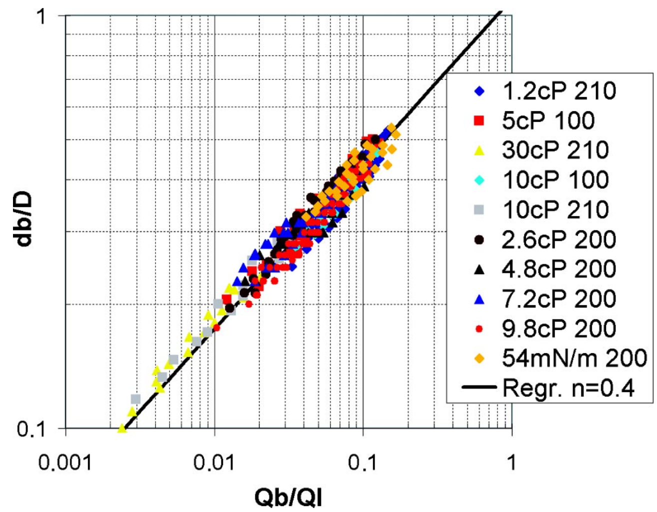

FIG. 3. (Color online) New scaling compared to experiments. Same notation as in Fig. 1.

$$d_b/D = 1.1(Q_g/Q_l)^{0.4} \tag{5}$$

with sufficient accuracy (data scatter in Fig. 3, about ±8%, should be mainly due to the gas flow rate errors). For liquid metals and small microbubbles, though, one must consider the pressure increment inside the bubble owing to surface tension to calculate the appropriate gas flow rate as it appears in Eq. (5).

This work was supported by the Spanish Ministry of Science and Technology, and by Kraft Foods Inc. Thanks must be given to J.L. Sampedro-Fernández, M. Hoc, N. Ouarty, T. Prevost, V. Marandat, and S. Vidal for their highly valuable assistance in experiments, and to Dr. M. Pérez-Saborid, Dr. J.M. López-Herrera, and Dr. J.M. Gordillo for useful suggestions.

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