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1 EXPERIMENTAL INVESTIGATION OF BUBBLE AND DROP FORMATION AT  
2 SUBMERGED ORIFICES

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18  
19  
20 **Keywords:** bubble, drop, formation, PIV measurements

21 **Abstract**

22 The aim of this study is to investigate bubble/drop formation at a single submerged orifice in  
23 stagnant Newtonian fluids and to gain a qualitative understanding of the formation  
24 mechanism. The effects of various governing parameters were studied. The formation  
25 behaviours of bubble and drop in Newtonian aqueous solutions were investigated  
26 experimentally under different operating conditions with various orifices. The results show  
27 that the volume of detached dispersed phase increases with viscosity, surface tension, orifice  
28 diameter and dispersed phase flow rate. A PIV system was employed to measure  
29 quantitatively the velocity flow field during the bubble/drop formation, giving interesting  
30 information useful for the elucidation of the fundamental formation process at the orifice. It  
31 was revealed that the orifice shape has a strong influence on formed bubble size. Furthermore,  
32 based on a simple mass balance, a global correlation was proposed to predict both bubble and  
33 drop size with good agreement.

## 36 **1. Introduction**

37 The formation of gas bubbles or liquid drops is an important fundamental phenomenon that  
38 conditions significantly the hydrodynamics in gas-liquid and liquid-liquid reactors commonly  
39 used in chemical and petrochemical processes, cosmetics, mineral processing, etc. The contact  
40 between phases is generally achieved either by bubbling gas into the liquid or by making  
41 droplets in another liquid in various applications such as absorption, distillation,  
42 emulsification, froth flotation, etc.

43

44 In most of the multiphase equipment, the knowledge of the transport and transfer processes  
45 across a gas-liquid or liquid-liquid interface is useful for the estimation of interfacial area,  
46 mass transfer coefficient and dispersed holdup. Furthermore, the physicochemical properties  
47 of the liquid phase (viscosity, surface tension, density, etc.) as well as the characteristics of  
48 the dispersed phase (bubble/drop size, bubble/drop rise velocity, etc.) govern the  
49 hydrodynamics and flow pattern in the system. The formation mechanism determines the  
50 primitive bubble or drop size in the system and has logically important consequences on the  
51 hydrodynamics.

52

53 The earliest reports on the formation of single bubbles and drops were published by Tate  
54 (1864) and Bashforth & Adams (1883). Bashforth and Adams computed the shape of liquid  
55 menisci for equilibrium conditions and published their results in the form of tables. From their  
56 data, the evolution of the bubble or drop volume can be calculated in the quasi-steady regime.  
57 Different models of varying complexity have been proposed including viscous and inertial  
58 forces. Davidson and Schüler (1960a and 1960b) presented models for dynamical bubble  
59 formation at submerged orifices for the case of viscous and inviscid liquids. A significant  
60 amount of works on the bubble formation at submerged orifices over a wide range of

61 operating parameters have been published in the literature in the last few decades. Ouz and  
62 Prosperetti (1993) investigated the dynamic bubble growth at a needle. The importance of the  
63 chamber volume under a submerged orifice was pointed out by Marmur and Rubin (1976).  
64 Kumar and Kuloor (1970) published an exhaustive report on bubble formation under a wide  
65 range of operating conditions. Tsuge (1986) reviewed the hydrodynamics of bubble formation  
66 from submerged orifices and discussed various models for the bubble formation mechanism.  
67 Rabiger and Vogelpohl (1986) briefly discussed the various parameters that affect the bubble  
68 formation. Since then, bubble formation receives continuous attention in the literature, for  
69 example, Jamialahmadi *et al.*, 2001; Li *et al.*, 2002; Kulkarni and Joshi, 2005; Buwa *et al.*,  
70 2007. Most of these studies can be classified on the basis of the operating conditions for the  
71 gas phase, such as constant flow rate or constant pressure. However, the observations of many  
72 investigators are not concordant because of variation induced by the physical properties of the  
73 gas-liquid systems, the type of nozzles and the operating parameters (gas velocity, system  
74 pressure, etc.). Hence, more observations are required to clarify the bubble formation process.

75

76 Bubble and drop formation from a nozzle or an orifice have been extensively studied from  
77 theoretical and experimental perspectives as mentioned before. Many studies were devoted to  
78 drop formation in a continuous fluid (Null and Johnson, 1958; Narasinga *et al.* ;1966; Schele  
79 and Bernard, 1968; Chazal and Ryan, 1971; Hamad *et al.* 2001) and correlations were  
80 proposed based on the properties of the fluids studied (density, viscosity, surface tension) and  
81 on the operating conditions (diameter of the opening, flow of the dispersed phase) in order to  
82 predict the drop volume. The theoretical analysis has been primarily based on macroscopic  
83 force balances assuming that drop formation occurs in two stages. The first stage is the static  
84 growth of the drop, which ends when the forces are no longer in equilibrium. The second  
85 stage corresponds to the necking and breaking of the drop from the nozzle (Scheele and

86 Meister, 1968; Heertjes *et al.*, 1971). From these studies, it can be concluded that the volume  
87 of the drops depends not only on the nozzle size and liquid properties, but also on the liquid  
88 flow rate through the nozzle (unlike the pendant drop case (Michael, 1981).

89 These studies mainly differ in how the authors approach the second formation stage. The  
90 simplified model gives a good approximation for the drop size when the flow rate is small but  
91 deviations from the experimental measurements occur when the flow rate becomes greater  
92 (Clift *et al.* 1978; Kumar and Kuloor, 1970). Although the previously cited studies of drop  
93 formation provide some interesting features of the phenomenon, there is still few information  
94 on the fundamental formation process.

95  
96 To improve our understanding of the bubble and drop formation mechanism, significant  
97 information could be obtained from Particle Image Velocimetry (PIV) measurements. While a  
98 great deal of research has focused on rising bubbles in Newtonian and non-Newtonian fluids  
99 using PIV measurements (Funfschilling and Li, 2001), few experimental studies concern such  
100 measurements for the bubble formation. Scarano (1997) studied bubble formation in a  
101 Newtonian fluid with a large bubble wake masking the flow circulation, Li *et al.* (2001)  
102 studied bubble formation in non-Newtonian fluids during the expansion stage at an orifice. To  
103 our best knowledge, complete velocity flow field measurements for the bubble or drop  
104 formation have never been reported in the literature yet. A thorough knowledge of bubble and  
105 drop size evolution and break-up is necessary for complete prediction and design of practical  
106 processes that involve interfacial phenomena and flows between two phases. In this study, a  
107 PIV system was used to obtain this original information and gain new light into this subject.

108

109

## 110 2. Experimental set-up and operating conditions

111 The experimental setup used to investigate bubble and drop formation under constant flow  
112 conditions is shown in Fig. 1. The experiments were conducted in a square PMMA column  
113 (inner dimensions: 0.1 m wide, 0.5 m high) filled with aqueous phases. The gas bubbles and  
114 the organic liquid drops were generated through a submerged orifice located at the bottom of  
115 the central section of the column. Two types of orifices were used (a plate orifice of 0.01 m  
116 and a thin needle). Various orifice diameters ( $0.5$  to  $4 \times 10^{-3}$  m) were investigated in this study.  
117

118 A capillary edge of 0.2 mm was used to avoid the wetting problem at the orifice. All  
119 experiments were carried out at a constant ambient temperature of 293K. A Rheometric Fluid  
120 Spectrometer RFS II (Rheometric Scientific, USA) was employed to measure the rheological  
121 properties. The surface and interfacial tensions of the solutions were measured using a  
122 Tracker tensiometer (I.T. Concept, France) for gas-liquid and liquid-liquid systems. The  
123 physical properties of these systems used are reported in Tables 1 and 2.  
124

125 **Table 1.** Fluid properties

Fluid	density $\text{kg.m}^{-3}$	viscosity Pa.s	Tension force $\times 10^{-3} \text{ N.m}^{-1}$
Air	1.15	$1.8 \times 10^{-5}$	-
Water	998	$1 \times 10^{-3}$	72.5
HV45 50% wt	1030	0.205	37.5
HV45 65% wt	1050	0.515	38.5
Silicon oil 5	950	$6 \times 10^{-3}$	18.5
Silicon oil 10	953	$11 \times 10^{-3}$	20.2
Silicon oil 50	955	$55 \times 10^{-3}$	20.3
Silicon oil 100	960	0.11	20.4

126

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130

131 **Table 2.** *Interface properties* (Surface or interfacial tension  $\times 10^{-3}$  N.m<sup>-1</sup>)

Fluid	Silicon oil 5	Silicon oil 10	Silicon oil 50	Silicon oil 100
Water	24.2	22.1	20.2	18.5
HV45 50% wt	18.8	15.2	13.4	11.8
HV45 65% wt	18.2	14.9	13.1	11.2

132

133 Dimensionless numbers, such as Bond (buoyancy and tension force), Reynolds (inertia and

134 viscous forces), and Weber (inertia and tension forces) numbers were used to characterize the

135 gas and liquid flow:

$$136 \quad Bo = \frac{d_{int}^2 (\rho_l - \rho_g) g}{\sigma} \quad (1)$$

137

$$138 \quad Re = \frac{\rho_d \left( \frac{4Q}{\pi d_{int}^2} \right) d_{ext}}{\mu d} \quad (2)$$

139

$$140 \quad We = \frac{\rho d \left( \frac{4Q}{\pi d_{int}^2} \right)^2 d_{ext}}{\sigma} \quad (3)$$

141

142 The phenomenon was recorded with a high-speed digital video camera (CAMRECORD 600,

143 Germany) at a rate of 1000 images per second. Images contained 1280×1024 pixels

144 displaying grey levels. The lighting was provided by a 800 W halogen lamp which

145 illuminated the column in an indirect way on a white screen placed behind the column. The

146 image sequences obtained were then analysed in order to calculate the position, velocity and

147 shape of the dispersed phase. Instantaneous velocity fields around a forming bubble or a drop

148 were measured by means of a Particle Image Velocimetry technique (PIV, Dantec Dynamics,

149 Denmark) (Fig. 1). Illumination sheets were generated with two pulsed Nd-YAG LASERS

150 (SOLO-I-15 PIV New Wave Research, USA) arranged side-by-side and crossed the vertical

151 symmetry axis of the dispersed phase. The energy produced by this source was 2×15 mJ.

152 These lasers emitted green light with a wavelength of 532 nm for a duration of 8 to 10 ns. The

153 time between the flashes varied from less than one microsecond to a few milliseconds. The  
154 Nd-Yag lasers were also designed to ensure a sequence of illuminations at a frequency of 15  
155 Hz. The laser beams first crossed a cylindrical lens which gave a laser sheet of strong light  
156 intensity and small thickness (2.5 mm maximum). They were focused and superimposed on  
157 one zone of measurement. The size of the seeding particles was chosen so that the particles  
158 followed the flow correctly and produced sufficient light to make the recording possible and  
159 minimize the errors on measurements. In general, seeding particles with diameters of 10 to 50  
160 micrometers were used for liquid phases. In this study, they were composed of silvered glass  
161 microspheres of sizes ranging from 10 to 30 micrometres (average diameter of 15  
162 micrometres) and of a density of  $1400 \text{ kg.m}^{-3}$ . The size was determined to minimize both  
163 Brownian movement and seeding particle sedimentation. The camera, placed perpendicular to  
164 the laser sheets, took two successive images at the maximum intensity of the laser pulse.  
165 These images were divided into a few thousand small interrogation areas of  $16 \times 16$  pixels. A  
166 cross-correlation was then performed between the corresponding interrogation areas. When  
167 the flow was correctly inseeded, the measurement errors on the velocities were less than  
168 5%. From these experiments, the liquid velocity flow field around a forming bubble or drop  
169 were obtained at an orifice.

170



171 **3. Experimental results**

172 The experiments were chosen in the aim of obtaining quantitative information on the  
173 dynamics of bubble and drop formation.

174

175 *3.1 Bubble formation*

176 The shape evolution of bubbles with time during the process of bubble growth and  
177 detachment in aqueous solutions is shown in Fig. 2. It can be clearly seen that, in all the cases  
178 studied of bubble formation in water or HV45 solutions, bubbles expanded spherically at the  
179 beginning, then evolved from a spherical shape to an inverse teardrop shape symmetrically,  
180 with the vertical axis passing through the orifice centre. The first shape was due to the  
181 importance of surface tension at the beginning of bubble formation. Subsequently, the  
182 influence of bubble buoyancy gradually became important as the bubble grew: the interface  
183 moved upwards and the bubble was elongated to an obvious teardrop shape. Changing  
184 viscosities or physical properties modified the shape but not the global formation mechanism.

185

186 Further information about the formation mechanism could be obtained from the measurement  
187 of flow fields. The major difficulty in applying the PIV technique arose due to the light  
188 reflection at the interface. The light reflected at the bubble interface is usually much more  
189 intense than the light scattered by the tracer particles, which prevents the registration of their  
190 images on the recording media near the interface. For a big orifice, light reflection and  
191 obscurity near the bubble, due to its shadow, lead to a lack of information on the right side of  
192 the flow field. Thus only orifices of diameter smaller than  $4 \times 10^{-3}$  m were used so as to avoid  
193 this problem. The experimental flow fields obtained during the formation of a  $180 \times 10^{-9}$  m<sup>3</sup>  
194 bubble in water are shown in Fig. 3. The time between two successive frames was  $5 \times 10^{-3}$  s.  
195 The choice of this time depends on the rapidity of the physical phenomena. The time between

196 two successive recordings was about  $66 \times 10^{-3}$  s and the final formation time was about 0.3 s.  
197 At  $t/t_f = 0.03$  (Fig. 3a) and  $t/t_f = 0.3$  (Fig. 3b), the expansion phase of the bubble formation can  
198 be observed. At the start of bubble expansion, an upward flow was firstly observed at the  
199 front of the bubble, which pushes the liquid (Fig 3a). As the bubble grew, the upward flow  
200 increased until the amount of liquid moved reached a critical value and the liquid fell on the  
201 sides of the bubble front (Figs. 3b and 3c). The downward flow thus created resulted in  
202 conservation of mass and caused the circulation region at the side of the bubble. In water or  
203 viscous solutions, bubble and velocity profiles were perfectly axis-symmetric, and displayed  
204 neither shape nor trajectory oscillations within the range of bubble sizes studied before  
205 detachment. Fig. 3d shows the velocity profile a few milliseconds before the gas film rupture  
206 and bubble detachment ( $t/t_f = 0.99$ ). The combination of the upward and downward flow led  
207 to the formation of axis-symmetric circulation loops. The space corresponding to the  
208 recirculation region expanded while the bubble volume increased. The recirculation was due  
209 to the elongation phase of the bubble and created a shear exerted at the neck of the elongated  
210 bubble, leading to its final rupture. At  $t/t_f = 1.15$  (Fig. 4e), the formation cycle ended with the  
211 break-up of the neck of the bubble and its release into the column. Velocity field magnitude  
212 increased with the bubble velocity, and an upward flow appeared in the wake of the bubble  
213 due to mass conservation and recirculation. The final flow field was in agreement with that  
214 reported by previous studies and PIV measurement of bubble motion in fluids (Li *et al.* 2001;  
215 Frank *et al.* 2006; Dietrich *et al.* ;2008; and Dietrich *et al.* 2010).

216

217

218

219

220

221 The detached bubble volumes were correlated as a function of flow rate for capillaries of  
222 different diameters and different systems. The effect of the gas flow rate on the bubble growth  
223 is shown in Fig.4. Obviously, the bubble volume increases gradually with the gas flow rate.  
224 The effect of the orifice diameter on the bubble volume at detachment from the orifice is  
225 plotted in Fig. 4a. It can be observed that the bubble volume changes sharply with the  
226 diameter of the orifice due to the increase of both drag and surface tension with the orifice  
227 diameter, as both forces effectively prevent quick bubble detachment from the orifice. As a  
228 result, the bubble growth time becomes longer and the bubble volume increases accordingly.

229

230 Figure 4b shows the relationship between the volume of the bubble at detachment from the  
231 orifice and the gas flow rate in solutions with different viscosities and surface tension. It is  
232 well known that the surface tension acting on the bubble tends to restrain the growth of the  
233 bubble. The increase of HV45 concentration in water led to an increment of viscosity which  
234 resulted in the increase of the viscous drag. Hence the bubble experimented more difficulty in  
235 detaching from the orifice. Consequently, bubble formation was prolonged and the volume of  
236 the bubble increased. The bubble broke finally away from the orifice because of the  
237 continuous gas input in the bubble.

238

239 Concerning the orifice shape, two configurations were tested as shown on Fig 1. Contrary to  
240 what is reported in the literature, orifice shape has a non-negligible influence. The effect of  
241 the shape of the orifice on the bubble volume at detachment is plotted in Fig. 5.

242

243 For all the diameters studied, the bubble volume was greater for bubbles formed on a thin  
244 orifice rather than a plate orifice. It can be observed that this effect was particularly stronger  
245 for small orifice diameter. Firstly, the wetting on the orifice was thought to be responsible.

246 But if this were true, the bubble should would be bigger in the case of the plate orifice.  
247 Because this was not so, wettability was discarded and local hydrodynamic effects should be  
248 envisaged.

249

250 The use of the PIV system to measure the velocity field around the two different orifice  
251 shapes could give further information to elucidate this phenomenon. The liquid velocity field  
252 around a growing bubble in stagnant water can be observed in Fig. 6 for the two injection  
253 systems. Two flow rates used were exactly the same. Clearly, the bubble was bigger in the  
254 case of the thin orifice (Fig. 6a) rather than the plate orifice (Fig 6b), confirming then the  
255 volume evolution shown in Fig 5.

256

257

258 The two velocity flow fields showed similar global tendency but, for the plate orifice,  
259 recirculation regions caused a supplementary breakup mechanism at the neck of the bubble.  
260 The circulation loops seemed to be stopped on the orifice. In contrast, , the recirculation  
261 caused mainly the bubble elongation with the thin orifice. This effect could be less marked for  
262 bigger orifices.

263 Many parameters such as flow conditions, orifice size and shape, and physical properties were  
264 tested. The results show that the bubble volume increased with the liquid viscosity, surface  
265 tension, orifice diameter and gas flow rate.

266

### 267 *3.2 Drop formation*

268 The setup used to form drops was the same as for bubble formation. But it consisted  
269 essentially of a fine capillary tube through which the oil phase was delivered at a constant  
270 volumetric flow rate. The plate orifice was not used because of serious wetting problems  
271 occurring during drop formation. A liquid drop was formed directly from the tip of the tube.

272 The capillary tubes were made of stainless steel and had tips that were polished and machined  
273 flat to allow the drop to wet the front surface completely.

274

275 In order to avoid any wetting problems with the oil phase, the drop formation was studied  
276 only at several sizes of thin orifice: 0.5, 1, 2, 3 and  $4 \times 10^{-3}$  m respectively. When a viscous oil  
277 was injected continuously through an orifice into quiescent, immiscible aqueous phase under  
278 low flow rates, oil phase broke up as discrete drops directly from the tip of the nozzle under  
279 buoyancy as shown in Fig. 7. It shows a temporary picture sequence obtained by the high-  
280 speed camera during the experiments for a drop forming in quiescent water from a tube of  
281  $4 \times 10^{-3}$  m, at a constant liquid flow rate of  $10^{-9} \text{ m}^3 \text{ s}^{-1}$ . As the drop volume increased with the  
282 continuous oil alimentation, the drop rose on the edge of the orifice. The drop shape gradually  
283 changed from quasi-spherical to teardrop during the growth as shown by Zhang (1999). When  
284 the size of the drop became sufficiently large, the buoyancy force exceeded the surface  
285 tension force (at  $t/t_f = 0.75$ ), the drop necked quickly and an important part started to detach  
286 from the orifice. During this sequence (necking), the drop elongated quickly and contracted in  
287 the middle to create a liquid thread that connected the drop and the liquid on the orifice  
288 (Zhang 1999). This thread was stretched with the fast rise of the drop and broke at its downer  
289 end, resulting in a free primary drop. After the thread breakup, the cone-shaped liquid  
290 remaining on the orifice tended to retrieve its apex and became spherical because of the  
291 capillary force. Typically this rupture at both ends of the thead lead to the creation of a  
292 satellite small droplet, having a volume less than 1% of that of the main drop.

293

294 The velocity field measured by PIV is shown in Fig. 8. Due to the lower speed of the  
295 expansion phase of the growing drop, it was very difficult to measure a velocity profile with  
296 precision at these velocities. However, it can be supposed that velocity profiles in the

297 expansion phase were the same as for bubble formation. The drop expansion should firstly  
298 create an upward flow at its front as a result of the drop pushing the liquid. As in the case of  
299 bubbles, the upward flow increased until the quantity of liquid moved reached a critical value  
300 and the liquid descended along the sides of the drop. The combination of the upward and  
301 downward flows led to the formation of axis-symmetric circulation loops as shown in Fig. 8a.  
302 As seen in Fig. 8b, the recirculation region expanded as the drop volume increased. This  
303 recirculation region was a consequence of the drop elongation leading to its rupture (Fig 8c).  
304 At the moment of rupture, a thin thread of liquid linked the drop and the orifice. The  
305 formation cycle ended with the breakup of the drop neck and its release into the continuous  
306 phase. The magnitude of the liquid velocity field increased with the drop velocity, and an  
307 upward flow appeared in the wake of the drop due to mass conservation and recirculation.  
308 This final flow field is quite comparable to the bubble formation.

309

310 Fig. 9 focuses on the velocity profile when the thread broke, leading to the formation of a  
311 secondary drop. The velocity flow fields before and after the detachment of a drop are  
312 represented for a  $250 \times 10^{-9} \text{ m}^3 \cdot \text{s}^{-1}$  drop of HS100 silicone oil in water (orifice size  $3 \times 10^{-3} \text{ m}$ ).

313

314 Before the rupture, the flow fields around the thread displayed complex features: partly  
315 upward flow behind the drop and partly directed downward. Fig. 9 illustrates the stretching  
316 effects before and after the rupture of the liquid thread behind the neck of the drop. The  
317 downward and upward flows stretched the thread in opposed directions, leading to subsequent  
318 rupture, which explains why the rupture occurs in the middle of the fluid thread and creates  
319 satellite droplets due to interfacial instability. The velocity flow fields before and after the  
320 rupture shown in Fig. 9 corroborate the shape evolution of the drop.

321

322 Fig. 10 represents the volume evolution of formed drops for several systems used. Firstly, the  
323 influence of the orifice size was investigated for a silicone oil drop in water. The results for  
324 different orifice sizes tested are reported in Fig. 10a. As for bubble formation, the bigger the  
325 orifice, the greater was the drop volume. However, the volume evolution with the flow rate of  
326 dispersed phase differed from that for the bubble. For drop formation with a small orifice, the  
327 volume increased with the flow rate until a maximum value. Thereafter, the volume  
328 decreased. This maximum was observed in other systems using different silicone oil (HS50 or  
329 HS10) as shown in Fig. 10b. The tendency was the same for these three systems. This  
330 maximum volume was observed by Narasinga (1964). The analysis by means of the high-  
331 speed camera could explain this phenomenon. In Fig. 10b, three formation sequences were  
332 plotted with the increase of the flow rate. The higher the flow rate, the longer was the neck  
333 behind the drop. The formation time was thus shorter at high flow rate than at low flow rate.  
334 PIV measurements provided quantitative information about this phenomenon. The velocity  
335 fields around a forming drop were quite different according to a low or a high flow rate (Fig.  
336 11).

337

338 The wake created by the preceding drop exerts a strong suction effect on the early stages of  
339 the forming drop at orifice (Fig. 11b), and cannot be neglected as in many theoretical models  
340 developed previously. This effect becomes smaller for the lower flow rate (Fig. 11a). The  
341 wake effect leads to a shorter drop formation time and, indeed, a smaller volume of the drop  
342 at detachment.

343

### 344 *3.3 Global correlation*

345 Even if the shape evolution is not exactly the same for drop and bubble formation, the  
346 formation mechanism is quite similar for the systems studied. This study was carried out

347 under large ranges of experimental conditions with a total of 250 data points: bubble formation  
 348 in water and viscous fluids with several orifice sizes ( $0.5$  to  $4 \times 10^{-3}$  m) and several flow rates;  
 349 drop formation for several silicon oils (HS100, HS50 and HS10) in water and viscous fluids,  
 350 with several orifice sizes ( $0.5$  to  $4 \times 10^{-3}$  m) and several flow rates. To establish a comparison,  
 351 it is interesting to propose a correlation for the dispersed phase volume at detachment. If  
 352 inertia and viscous force are neglected (static model hypothesis) in the formation process, the  
 353 only two forces acting on forming bubbles or drops at orifice are the buoyancy  
 354  $P_A = (\rho_l - \rho_g) \cdot g \cdot V$  and the capillary force  $F_c = \sigma \cdot \pi \cdot d_{int} \cdot \cos\theta$ . The global force balance leads to  
 355 Equation (4):

$$356 \quad \frac{d}{d_{int}} = \left( \frac{6\sigma}{d_{int}^2 (\rho_l - \rho_g) g} \right)^{1/3} \quad (4)$$

357  
 358 The number of bubbles or drops  $N_{b,d}$  is defined on the basis of the Bond number, equivalent  
 359 diameter of dispersed phase and orifice internal diameter as follows:

$$360 \quad N_{b,d} = \frac{d}{d_{int}} Bo^{\frac{1}{3}} \quad (5)$$

361  
 362 As shown in Eq. 4, this number is equal to  $N_0 = 6^{1/3} \approx 1.81$  under static conditions. But this  
 363 value is obtained for a system where inertia and viscous drag are neglected, which is not  
 364 really the case for some of our experimental systems, e.g. formation in HV45 50% or viscous  
 365 silicone oil. In order to take into account these forces, a correction factor could be added,  
 366 using Reynolds and Weber numbers and viscosity ratio between two phases, and interpreting  
 367 the deviation between the static model and experimental data. The obtained correlation in  
 368 Equ. 6 gives a good prediction of the bubble and drop volume.

$$369 \quad \frac{N_{b,d}}{N_0} = 1 + \frac{We^{0.4}}{Re^{0.43}} \left( \frac{\mu_c}{\mu_d} \right)^{0.38} \quad (6)$$

370 Fig. 12 represents the experimental  $N_{b,d}$  versus the correlated one. The average error between  
 371 experiment data and the correlation is less than 11%. Thus, the correlation can be used for the



372 prediction of the formed volume of a dispersed phase under various conditions: Reynolds  
373 number ranging from  $10^{-3}$  to  $10^3$ , orifice sizes between  $0.5$  to  $4 \times 10^{-3}$  m, stagnant phase  
374 viscosity from  $1$  to  $500$  mPa.s, and dispersed phase viscosity from  $1.8 \times 10^{-5}$  to  $10^{-1}$  Pa.s.  
375 Nevertheless, this correlation is only valid for the thin orifice when no wetting phenomenon is  
376 observed on the edge of the orifice. When wetting and spreading are observed, especially for  
377 drop formation, the formation process and measured volume are not reproducible. The  
378 influence of the orifice material and shape could lead to large errors in volume estimation and  
379 require still more investigation.

380

#### 381 **4. Conclusion**

382 The present work allows a better understanding of the formation mechanism of bubble or  
383 drop. Flow field measurements obtained by PIV contribute to the flow quantification of the  
384 different steps in continuous phase encountered during the formation and provide new light  
385 into the interactions between successive drops or the influence of the orifice shape on the flow  
386 pattern around a forming bubble or drop. The visualisation of the bubble/drop shape and  
387 volume by the high-speed camera along with the flow fields by PIV would be useful for  
388 modelling the formation mechanism and validating numerical approaches of the formation  
389 process, in particular the thread rupture in the presence of interfacial instability. Finally, a  
390 global correlation for bubble and drop formation was proposed to estimate the volume at  
391 detachment from an orifice. However, this correlation is only valid in the absence of wetting  
392 problems at orifice.

393

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397

398 **Notation**

399

400  $N_{b,d}$  bubble/drop number

401  $Re$  Reynolds number

402  $Bo$  Bond number

403  $We$  Weber number

404  $V$  bubble/drop volume,  $m^3$

405  $d$  bubble/drop equivalent diameter,  $m$

406  $\mu$  fluid viscosity,  $Pa.s$

407  $g$  gravity,  $m.s^{-2}$

408  $\sigma$  surface or interfacial tension,  $N.m^{-1}$

409

410 *Subscripts*

411  $c$  continuous phase

412  $d$  dispersed phase

413  $int$  inlet diameter

414  $o$  equilibrium

415

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417 **Literature Cited**

- 418 Badam V. K., Buwa, V. and Durst F., 2007, "Experimental investigations on regimes of  
419 bubble formation on submerged orifices under constant flow conditions", Canadian Journal of  
420 Chemical Engineering, 85, 257-267.
- 421 Chao-Tai Chen, Jer-Ru Maa, Yu-Min Yang and Chien-Hsiang Chang (2001), "Drop  
422 formation from flat tip nozzles in liquid-liquid system", Int. Comm. in Heat and Mass  
423 Transfer, 28, 5, 681-692
- 424 De Chazal, L.E.M. and J.T.Ryan, (1971). "Formation of organic drops in water". AIChE J.,  
425 17,1226-1229.
- 426 Dietrich N., Poncin S. and LI H.Z, (2010). Dynamical deformation of a liquid-liquid interface,  
427 Experiments in fluids, 2010, accepted
- 428 Dietrich N., Poncin S., Pheulpin S. and LI H.Z, (2008). Passage of a bubble through a liquid-  
429 liquid interface. AIChE J. 2008, 54, 3, 594-600.
- 430 Frank X., Funfschilling D., Midoux N. & Li H.Z. (2006). Bubbles in a viscous liquid: Lattice  
431 Boltzmann simulation and experimental validation. J. Fluid Mech., 546, 113-122
- 432 Funfschilling D. and Li H.Z., Flow of non-Newtonian fluids around bubbles: PIV  
433 measurements and birefringence visualisation, (2001). Chemical Engineering Science, 56,  
434 1137-1141
- 435 Gaddis E. et Vogelpohl A. (1986). Bubble formation in quiescent liquids under constant flow  
436 conditions. *Chem. Eng. Sci.*, 41(1), 97-104.
- 437 Gerlach D., G. Biswas, F. Durst and V. Kolobaric (2005), "Quasi-static bubble formation on  
438 submerged orifices", Int. J. Heat Mass Transfer, 48, 425-438.
- 439 Harkins W.D. and F.E. Brown (1919), "Determination of surface tension and the weight of  
440 falling drops: The surface tension of water and benzene by the capillary height method", J.  
441 Am. Chem. Soc., 41, 499.
- 442 Hayworth, C.B. and R.E. Treybal (1950). "Drop formation in two liquid phase systems".
- 443 Hibiki T. and M. Ishii (2007), " Lift force in bubbly flow systems", Chem. Eng. Sci., 62,  
444 6457-6474
- 445 Horvath M., L. Steiner and S. Hartland (1978), "Prediction of drop diameter, hold-up and  
446 backmixing coefficients in liquid-liquid spray columns." Can. J. Chem. Eng., 56, 9-15.
- 447 Jamialahmadi, M., M.R. Zehtaban, H.M. Steinhagen, A. Sarrafi and J.M. Smith (2001),  
448 "Study of Bubble Formation under Constant Flow Conditions" , Trans IChemE, 79 A, 523-  
449 532

450 Kagan S.Z., Y.N. Kovalev and A.P. Zakharychev (1973), *Theor. Osn. Chem. Eng. (Moscow)*,  
451 7, 514.

452 Klee A.J. and R.E. Treybal (1956), "Rate of rise or fall of liquid drops". *A.I.Ch.E. J.* 1.2/4 ,  
453 444-447.

454 Krevelen van D.W. and P.J. Hoftijzer (1950), "Studies of gas bubbles formation", *Chem Eng.*  
455 *Prog.*, 46, 1, 1950.

456 Kulkarni A.A. and J. B. Joshi (2005), "Bubble Formation and Bubble Rise Velocity in Gas-  
457 Liquid Systems: A Review", *Ind. Eng. Chem. Res.*, 44, 5873-5931

458 Kumar R. and N.R. Kuloor (1970), "The formation of bubbles and drops", *Adv. Chem. Eng.*,  
459 Academic Press, N.Y.

460 Li H. Z., Frank, X. et Funfschilling, D. and Mouline Y. (2001). Towards the understanding of  
461 bubble interactions and coalescence in non-Newtonian fluids: a cognitive approach. *Chemical*  
462 *Engineering Science*, 56, 6419-6425.

463 Li H.Z., Mouline Y. and Midoux N., (2002). Modelling the bubble formation dynamics in  
464 non-Newtonian fluids, *Chem. Eng. Sci.* 57, 339-346

465 Miyahara, T., and T. Takahashi, "Bubble Volume in Single Bubbling Regime with Weeping  
466 at a Submerged Orifice," *J. Chem. Eng. Jpn.*, 17, 597-1984.

467 Nahra H.K. and Y. Kamotani (2003), "Prediction of bubble diameter at detachment from a  
468 wall orifice in liquid cross-flow under reduced and normal gravity conditions", *Chem. Eng.*  
469 *Sci.*, 58, 55-69.

470 Narasinga Rao E.V., R. Kumar and N.R. Kuloor (1966), "Drop Formation Studies in Liquid-  
471 Liquid Systems", *Chem. Eng. Sci.*, 21, 867-880.

472 Null H.R. and H.F. Johnson (1958), "Drop Formation in Liquid-Liquid Systems from Single  
473 Nozzles", *AIChE J.*, 4, 273-281.

474 Oguz H.N. and A. Prosperetti (1993), "Dynamics of bubble growth and detachment from a  
475 needle", *J. Fluid Mech.*, 257, 111-145.

476 Richards J.R., A.N. Beris and A.M. Lenhoff (1995), " Drop formation in liquid-liquid systems  
477 before and after jetting", *Phys. Fluids* 7, 2617-2630.

478 Scarano, F.: Improvements in PIV image processing application to a backward facing step;  
479 VKI PR 1997-01

480 Scheele G. and B. Meister (1968a), "Drop formation at low velocities in liquid-liquid  
481 systems", Part I: "Prediction of drop volume", *AIChE J.*, 14, 9-15.

482 Scheele G. and B. Meister (1968b), Part II: "Prediction of jetting velocity", *AIChE J.*, 14, 16-

483 19.  
484 Steiner L. and S. Hartland (1983), "Hydrodynamics of Liquid-liquid Spray Columns" in  
485 "Handbook of Fluids in Motion", ch. 40, N. Cheremisinoff and R. Gupta Ed., Ann Arbor  
486 Science.  
487 Tomotika S. (1935), "On the instability of a cylindrical thread of a viscous liquid surrounded  
488 by another viscous fluid." Proc. R. Sot. London Ser. A 150, 322.  
489 Tsuge, H. (1986). Hydrodynamics of Bubble Formation from Submerged Orifices.  
490 Encyclopedia of Fluid Mechanics, Vol. 3, Gulf Publishing, Houston, p. 191.  
491 Tyler E. (1933), Instability of liquid jets, Philos. Mag. 16, 504-516.  
492 Wong H., D. Rumschitzki and C. Maldarelli (1998), "Theory and experiment on the low-  
493 Reynolds-number expansion and contraction of a bubble pinned at a submerged tube tip", J.  
494 Fluid Mech., 356, 93-124.  
495 Wraith A.E. (1971), "Two-stage bubble growth at a submerged plate orifice", Chem. Eng.  
496 Sci., 26, 1659-1709.  
497 Zhang, X.. "Dynamics of drop formation in viscous flows", Chemical Engineering Science,  
498 199906  
499  
500  
501  
502