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1 VISUALISATION OF GAS-LIQUID MASS TRANSFER  
2 AROUND A RISING BUBBLE IN A QUIESCENT  
3 LIQUID USING AN OXYGEN SENSITIVE DYE

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20

21 **Abstract**

22 An approach for visualizing and measuring the mass transfer around a single bubble rising in a  
23 quiescent liquid is reported. A colorimetric technique, developed by (Dietrich et al., 2013) using  
24 an oxygen sensitive redox dye was implemented. It was based on the reduction of the  
25 colorimetric indicator in presence of oxygen, this reduction being catalysed by sodium  
26 hydroxide and glucose. In this study, resazurin was selected because it offered various reduced  
27 forms with colours ranging from transparent (without oxygen) to pink (in presence of oxygen).  
28 These advantages made it possible to visualize the spatio-temporal oxygen mass transfer around  
29 rising bubbles. Images were recorded by a CCD camera and, after post-processing, the shape,  
30 size, and velocity of the bubbles were measured and the colours around the bubbles mapped. A  
31 calibration, linking the level of colour with the dissolved oxygen concentration, enabled colour  
32 maps to be converted into oxygen concentration fields. A rheoscopic fluid was used to visualize  
33 the wake of the bubbles. A calculation method was also developed to determine the transferred  
34 oxygen fluxes around bubbles of two sizes ( $d = 0.82$  mm and  $d = 2.12$  mm) and the associated  
35 liquid-side mass transfer coefficients. The results compared satisfactorily with classical global  
36 measurements made by oxygen micro-sensors or from the classical models. This study thus  
37 constitutes a striking example of how this new colorimetric method could become a remarkable  
38 tool for exploring gas-liquid mass transfer in fluids.

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42 **Keywords:** Bubble, Mass transfer, Visualization, Concentration fields

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## 45           **1. Introduction**

46 Gas-liquid contactors play an important role in engineering processes, where they have many  
47 applications, including water treatment and (bio)-chemical processes (Dietrich et al., 2011;  
48 Tatin et al., 2015; Benizri et al., 2017; de Lamotte et al., 2017; Hariz et al., 2017; Kherbeche et  
49 al., 2017; Kováts et al., 2017; Wylock et al., 2017; Xie et al., 2017; Wongwailikhit et al., 2017).  
50 A good prediction of the mass transfer phenomena is necessary to achieve good reactor design  
51 leading to higher efficiency. The film model, penetration theory and the surface renewal model  
52 are three established, well-known theories describing interface mass transfer (Danckwerts,  
53 1951; Higbie, 1935; Whitman, 1923). Based on these models, film-penetration theory and the  
54 unsteady two-film model were developed (Toor and Marchello, 1958; Wang and Langemann,  
55 1994). In order to develop a further model, a good understanding of the mass transfer at  
56 interfaces is necessary. As the physical presence of sensors alters the hydrodynamics and the  
57 use of chemical compounds changes the liquid properties, the application of an invasive  
58 technique has marked limitations. As an alternative, non-invasive techniques such as  
59 visualization are a solution to estimate mass transfer accurately in bubbly systems.

60 With this in mind, optical techniques have been established based on the use of a fluorescent  
61 dye, sensitive to dissolved oxygen molecules or pH, to measure the oxygen or carbon dioxide  
62 concentration at a gas-liquid interface. When the solution containing a fluorescent dye is  
63 illuminated by a 2D-laser sheet and the local data are acquired by digital cameras, the technique  
64 is named PLIF (Planar Laser Induced Fluorescence). This technique has been adapted for the  
65 visualization of oxygen concentration by studying the fluorescence of an oxygen-sensitive dye  
66 (Munsterer and Jahne, 1998; Mühlfriedel and Baumann, 2000; Herlina and Jirka, 2004;  
67 Falkenroth et al., 2007; Walker and Peirson, 2007; Jimenez et al., 2012b, 2012a). The inhibition  
68 of the fluorescence is used to quantify the oxygen concentration. This technique is called PLIFI  
69 (Planar Laser Induced Fluorescence with Inhibition). Studies concerning mass transfer around  
70 a single bubble have also been performed to provide qualitative measurements of oxygen mass

71 transfer around bubbles using oxygen as a fluorescence quencher (Roy and Duke, 2004; Bork  
72 et al., 2005; Dani et al., 2007; Francois et al., 2011a, 2011b, Jimenez et al., 2013a, 2013b, 2014;  
73 Dietrich et al., 2015; Butler et al., 2016; Roudet et al., 2017; Xu et al., 2017) and to obtain local  
74 measurements of concentration distributions around carbon dioxide bubbles using pH-sensitive  
75 dye (Someya et al., 2005; Hanyu and Saito, 2010; Stöhr et al., 2009; Saito and Toriu, 2015;  
76 Huang and Saito, 2015, 2017). These works demonstrate the relevance of using the PLIFI  
77 technique to observe mass transfer phenomena. However, the technique has limitations for  
78 quantifying mass transfer around a bubble. In the presence of a bubble, the fluorescence  
79 tracking is disturbed by light reflections and refractions at its surface. A white ring is then  
80 systematically observed around the bubble as well as the bubble shadow, resulting in an  
81 intensity contrast between the solution to the left of the bubble and the solution to the right. The  
82 optical issues require heavy image processing leading to less precise information. Furthermore,  
83 the mass transfer is only observed in a 2D-plane parallel to the bubble trajectory. Other  
84 techniques such as digital holographic interferometry (Klein and Wanders, 1987; Kutepov et  
85 al., 2001; Wylock et al., 2011) are used to visualize the in-depth profile of density which is  
86 proportional to the dissolved gas concentration. But these techniques, very precise, should be  
87 applied in systems with an important density gradient generated by gas-liquid mass transfer,  
88 which is not the case of air-water systems.

89 In order to avoid these problems, another optical solution was developed, using an oxygen-  
90 sensitive dye. This technique is based on fast redox reaction between dihydroresorufin  
91 (colourless) and oxygen, creating resorufin (pink). As the diffusion coefficient of the dye is  
92 weak in comparison with that of oxygen in these liquids, it is possible to visualize the presence  
93 of oxygen around these interfaces. Kherbeche et al., (2013) used this technique to study the  
94 mass transfer around a bubble in a 2D-packed column, Dietrich et al., (2013) around bubbles  
95 flowing in a square milli-channel and (Yang et al., 2017b, 2016, 2017a; Krieger et al., 2017.)  
96 around bubbles at forming stage in a micro-channel. This paper proposes to apply this recent

97 technique to a bubble rising in a quiescent liquid, which has previously been visualized only by  
98 the PLIFI technique. The main advantage is that the mass transfer can be visualized without  
99 any laser excitation, making this technique easy and user friendly. In a first part, the general  
100 principles of the colorimetric technique will be presented, together with the experimental set-  
101 up and the methods used (image acquisition). Secondly, the way to implement this technique  
102 will be described, in particular the calibration procedure and the different image processing  
103 steps. Results will be expressed in terms of a dissolved oxygen field around the bubble, mass  
104 flux of transferred oxygen and a liquid-side mass transfer coefficient.

105

## 106 **2. Material and Methods**

107 The colorimetric technique is based on the use of an oxygen-sensitive dye. The main advantage  
108 is that it is non-intrusive, as the measurements are carried out without disturbing the flow or the  
109 physicochemical properties and without inserting a physical sensor. A large number of organic  
110 chemical compounds can undergo oxidation/reduction reactions while presenting the properties  
111 of:

- 112 - being intensely coloured in the oxidized or reduced state,
- 113 - and being either colourless or differently coloured in the conjugate (opposite) state.

114 For example, methylene blue is blue in the oxidized state but colourless in the reduced  
115 condition. The well-known “blue bottle” experiments use this property (Cook *et al.*, 1994;  
116 Engerer and Cook, 1999; Wellman and Noble, 2003). The principle is that an alkaline solution  
117 of glucose (reducing agent) reduces the methylene blue from a blue to a colourless form, then  
118 shaking the solution raises the concentration of dissolved oxygen in the mixture, which, in turn,  
119 oxidizes the methylene blue back to the blue form. When the dissolved oxygen is consumed by  
120 the reaction, the methylene blue is slowly reduced by the remaining glucose and becomes  
121 colourless again. This cycle can be repeated many times by alternating periods with and without  
122 shaking, these reactions being reversible until the reactants are used up.

123 The main drawback when visualizing mass transfer using methylene blue is the slow kinetics  
124 of the reactions (Wellman and Noble, 2003). However, other redox dyes are available, such as  
125 indigo carmine or resazurin that are respectively red and green, and blue and pink in their  
126 different forms. The choice of dye mainly depends on two factors:

- 127 - the speed of the kinetics of the reaction between dissolved oxygen and dye,
- 128 - and the intensity of the colour generated.

129 In this study, three dyes were tested: methylene blue, indigo carmine and resazurin. The latter  
130 dye was chosen as enabling a good compromise to be achieved between kinetics and colour. In  
131 addition, resazurin is well-known as its reduction has been used for about 50 years to monitor  
132 bacterial and yeast contamination of milk, and also for assessing semen quality (Campbell,  
133 1963). As shown in Figure 1, resazurin (blue and not fluorescent) is reduced to resorufin (pink  
134 and highly fluorescent) which is itself reduced to dihydroresorufin (colourless and not  
135 fluorescent). These reactions are catalysed by the presence of glucose and sodium hydroxide.  
136 In alkaline solutions, glucose is oxidized to D-gluconic acid or alpha-D-gluconolactone. The  
137 change of colour results from the reversible oxidation-reduction reaction between resorufin  
138 (pink) and dihydroresorufin (colourless). Note that the complete chemical formula of resazurin  
139 is 7-hydroxy-3H-phenoxazin-3-one-10-oxide (molecular mass  $229.19 \text{ g}\cdot\text{mol}^{-1}$ ).

140 A preliminary study (not detailed here) was carried out to define the best formulation of the dye  
141 solution, namely the one leading to:

- 142 - (quasi)-instantaneous kinetics for the oxidation of dihydroresorufin (colourless) to  
143 resorufin (pink),
- 144 - sufficiently slow kinetics (few minutes) for the reduction of resorufin (pink) to  
145 dihydroresorufin (colourless),
- 146 - a high colorimetric yield (*i.e.* the colour intensity).

147 From this, the following formulation was selected: glucose and sodium hydroxide were both  
148 diluted at  $20 \text{ g.L}^{-1}$  in deionized water, and the concentration of resazurin was fixed at  $0.1 \text{ g.L}^{-1}$ .  
149 The experimental set-up consisted of a transparent column fitted with a bubble injection system,  
150 and optical equipment designed to allow the rising bubble to be visualized. It is shown in Figure  
151 2. The  $100 \times 100 \times 300 \text{ mm}^3$  column had a square cross-section. It had three transparent glass  
152 sides and one PMMA side where the injection system was set. The base was made of glass and  
153 the top of the column was open. The mass transfer from the top is considered as neglectable  
154 (Jimenez et al., 2012a). The injection system was placed 10 mm from the bottom of the column.  
155 It was composed of a very thin glass capillary with an inner diameter that could range from 0.01  
156 to 0.1 mm connected to an automatically controlled syringe (Harvard Apparatus PHD 2000).  
157 The inner diameter was changed in order to generate bubbles of several sizes. The syringe was  
158 filled with air and the flow rate was chosen to produce only a single bubble at a time. Bubbles  
159 were formed at the centre of the column in a liquid containing the colorimetric solution. A drain  
160 tap was fitted at the bottom of the column to evacuate the liquid easily after each experiment.  
161  
162 A CCD (Charge-Coupled-Device) colour camera (Guppy Pro, Allied Vision, Germany, 8 bits,  
163  $30 \text{ Hz}$ ,  $1024 \times 1024 \text{ pixels}^2$ ) was placed horizontally, perpendicular to the column - on the x  
164 axis - and was focused on the centre of the column. The digital camera was fitted with a 105  
165 mm objective (Micro-Nikkor 60 mm f/2.8, Nikon) to reach a high spatial resolution of  
166 approximately  $0.023 \text{ mm/pixel}$  on the focused area. A backlight panel was placed behind the  
167 column to film the bubble by the shadowgraph technique. The colorimetric images of the  
168 moving bubbles in the column were recorded and treated by a procedure developed in Matlab  
169 software. Details of this study will be described in the next section. Experiments were  
170 performed at  $295 \text{ K}$  in an air conditioned room, and at atmospheric pressure. Fluids were air  
171 and deionized water containing D-glucose anhydrous (Fischer Scientific®, CAS 50-99-7),  
172 sodium hydroxide (VWR®, CAS 1310-73-2) and resazurin (Sigma Aldrich®, CAS 62758-13-

173 8). Density ( $\rho_L$ ), dynamic viscosity ( $\mu_L$ ) and static surface tension ( $\sigma_L$ ) were measured  
 174 respectively by means of a pycnometer ( $\rho_L \pm 0.2 \text{ kg.m}^{-3}$ ), an RM180 Rheomat Rheometric  
 175 Scientific® viscometer ( $\mu_L \pm 10^{-3} \text{ mPa.s}$ ), and Digidrop GBX® or Krüss tensiometers ( $\sigma_L \pm 0.5$   
 176  $\text{mN.m}^{-1}$ ). These physico-chemical properties are reported in Table 1.

177

178

179 *Table 1. Properties of clean water and the synthetic liquid media at 20°C.*

180

| Liquid phase    | $C \text{ (g.L}^{-1}\text{)}$ | $\sigma_L \text{ (mN.m}^{-1}\text{)}$ | $\mu_L \text{ (mPa.s)}$ | $\rho_L \text{ (kg.m}^{-3}\text{)}$ |
|-----------------|-------------------------------|---------------------------------------|-------------------------|-------------------------------------|
| Deionized water | -                             | 73.4                                  | 1.003                   | 996.8                               |
| Glucose         | 20                            | 75.5                                  | 1.1179                  | 1005.2                              |
| NaOH            | 20                            |                                       |                         |                                     |
| Glucose         | 20                            | 75.4                                  | 1.1179                  | 1005.3                              |
| NaOH            | 20                            |                                       |                         |                                     |
| Resazurin       | 0.1                           |                                       |                         |                                     |

181

182

### 183 3. Implementation of the colorimetric technique

184 Based on experimentally acquired images (Figure 3-a), a specific approach was developed to  
 185 quantify the mass transferred in the bubble wake. To achieve this quantification, the first step  
 186 was to establish a calibration curve between the “grey level” observed on the recorded image  
 187 and the corresponding oxygen concentration. For this, several colorimetric solutions were  
 188 prepared at different concentrations of resazurin (1, 5, and 10  $\text{mg.L}^{-1}$ ). They were saturated in  
 189 oxygen from the air in order to reach their highest intensity of pink (or their highest value of  
 190 grey level on the acquired image) then added in a 10 mm glass tube inside the column, in the  
 191 bubble rising axis. As the mass transferred by the bubble is located in its wake, it is then possible  
 192 to realize a pseudo 2D calibration in a 3D column. For each colorimetric solution about 100  
 193 pictures were recorded and averaged without bubbles. In addition, images were acquired when

194 no oxygen was present in the colorimetric solution. The stoichiometry of the reaction between  
195 oxygen and dihydroresorufin is given by Equation 1. When there is a sufficient amount of  
196 resazurin to react with all the oxygen transferred, the number of moles of dissolved oxygen can  
197 easily be deduced from the number of moles of resazurin, as

$$198 \quad n_{O_2,transferred} = n_{O_2,reacted} = \frac{n_{dihydroresorufin}}{2} = \frac{n_{resazurin}}{2} \quad (1)$$

199 Thus, for each resazurin concentration (0, 1, 5, and 10 mg.L<sup>-1</sup>), it was possible to associate the  
200 observed averaged grey level with a dissolved oxygen concentration: 0, 0.075, .25, and 1 mg.L<sup>-1</sup>  
201 <sup>1</sup>) for each pixel. The calibration curve obtained is plotted in Figure 4. The linearity observed  
202 between grey level and dissolved oxygen concentration is remarkable and in agreement with  
203 the Beer-Lambert law as the grey level of the recordings GL are close to the grey level of the  
204 reference without oxygen GL<sub>0</sub>. These results are in agreement with other studies (Dietrich et  
205 al., 2013; Kherbeche et al., 2013; Yang et al., 2016) and is an undeniable advantage of this  
206 method. It is also important to note that this calibration curve is specific to the experimental  
207 set-up and conditions used. In other words, if a change was made in the light or camera  
208 parameters, the calibration would have to be performed again. Based on this calibration curve,  
209 grey levels recorded by the camera could be converted into oxygen concentration. In a first  
210 approximation for quantification, the layer thickness within the Beer-Lambert-law is neglected,  
211 so that further efforts were necessary before the quantification could be considered accurate. As  
212 an example, the grey level of the picture, without oxygen, GV<sub>0</sub> has been found very close with  
213 or without the glass tube (deviation < 1 %). Specific image processing, of which only the basic  
214 principles are presented in this paper, was thus developed. An example of a raw image to be  
215 corrected is given in Figure 3-a. As a first step, a reference image was subtracted from the raw  
216 image. This reference image was an image without any bubbles or mass transfer. By subtracting  
217 the reference image, impurities in the image (in the liquid or on the optical device) were  
218 removed (see Figure 3-b). On the resulting image, only the bubble and the mass transferred in

219 its wake remained. However, because of some temporal non-homogeneities in the lighting, the  
220 background grey level (and oxygen concentration) could differ from 0.  
221 The background grey level had thus to be set to 0 to avoid altering the mass transfer calculations  
222 by a noise removal procedure. This procedure was also performed for calibration pictures. For  
223 information, the difference between Figure 3-b and the image after this correction can be seen  
224 in Figure 3-c, which shows the noise in the picture. Bubble edges were detected using specific  
225 algorithms (Canny edge detector, extracted from the Image Processing Toolbox in Matlab®).  
226 Bubble edges were thus detected (Figure 3-d) and removed (Figure 3-e) from the corresponding  
227 image. Finally, the previous calibration was applied in order to convert grey intensity into  
228 dissolved oxygen concentration. Figure 3-f is an example of a corrected image where non null  
229 pixels represent a certain amount of oxygen transferred by the bubble. The total amount of mass  
230 transferred could easily be determined by summing the estimated concentration pixel by pixel  
231 by integrating all the visualized volume.

232

#### 233 **4. Results**

234 The technique presented above was applied to the visualization of oxygen concentration in the  
235 wake of an air bubble in a liquid at rest. The first experiments conducted for this work were  
236 based on a 0.82 mm bubble rising rectilinearly with a velocity of  $u_B = 110$  mm/s ( $Re = 90$ ).  
237 Figure 5 shows the oxygen concentration field in the liquid phase for the bubble in two different  
238 solutions. It was observed that bubbles followed a rectilinear path. NaOH concentrations of 50  
239  $g.L^{-1}$  (Figure 5-a) and 20  $g.L^{-1}$  (Figure 5-b) were tested. It was observed that resarufin (revealing  
240 the presence of oxygen) accumulated in the wake of the bubble, due to recirculation that  
241 appeared in this zone. When the concentration of glucose was high, it caused reaction n°3 to  
242 accelerate, which transformed resorufin into dihydroresorufin, inducing a decrease in the  
243 amount of coloured molecules in the bubble wake. From this comparison, a reasonable  
244 concentration of glucose was chosen (20  $g.L^{-1}$ ). On Figure 5-b, in the bubble wake, a zone of

245 higher dissolved oxygen concentration can be clearly observed, which actually corresponds to  
246 oxygen transfer from the bubble into the liquid. As depicted in literature (Dietrich et al., 2008),  
247 the bubble rose, oxygen transferred from the bubble was advected in the wake, so that the  
248 oxygen concentration above the bubble remained unchanged.

249 Figure 6 presents a picture that was recorded for a bigger bubble ( $d = 2.12$  mm,  $u_B = 182$  mm/s,  
250  $Re = 400$ ). It exhibits deformations and oscillates periodically in the horizontal plane. The  
251 dissolved oxygen field structure suggests that the mass transfer was associated with the trailing  
252 vortices in the wake of the bubble. An aqueous rheoscopic solution made of mica powder (1  
253 g.L<sup>-1</sup>,  $\sigma = 67$  mN/m) was used to validate this hydrodynamic structure (Fig 6.b). Oscillating  
254 rising was also observed and this movement was also present in the bubble wake; the oxygen  
255 concentration field followed the streamline. It can be observed there is a good agreement  
256 directly behind the bubble but there is small differences in the far away wake. This could be  
257 explained by the interface modification, due to the mica, generating instabilities (Jimenez et al.,  
258 2014). The frequent renewal of liquid in the near wake suggested a high rate of mass transfer.  
259 It was observed that a 2 mm bubble tended to oscillate strongly (Fig 6-a) in comparison with  
260 the 1 mm bubble (Fig 5-b). These perturbations in the wake of the bubbles were visible in the  
261 raw images provided by the colorimetric technique. The observations on the mass transferred  
262 by the bubble confirmed the experiment on hydrodynamics using the Kallirosopic particles  
263 mentioned above. Table 2. Hydrodynamic and mass transfer properties of bubbles. Each  
264 condition was reproduced three times and averaged (deviation ~ 3%) \* Frossling model. \*\*  
265 Higbie model (Roustan, 2003).

266

|          | <i>Diameter</i><br>[mm] | <i>Velocity</i><br>$u_B$<br>[m/s] | <i>Reynolds</i><br><i>Number</i><br>[-] | <i>Mass</i><br><i>transferred</i><br>[mg] | $k_L$ <i>exp</i><br>[m/s] | $k_L$ <i>theory</i><br>[m/s] |
|----------|-------------------------|-----------------------------------|---|---|---------------------------|------------------------------|
| Bubble 1 | 0.82                    | 0.111                             | 90                                      | $3.48 \times 10^{-9}$                     | $9.55 \times 10^{-5}$     | $1.13 \times 10^{-4}$ *      |
| Bubble 2 | 2.12                    | 0.189                             | 400                                     | $3.13 \times 10^{-8}$                     | $6.62 \times 10^{-4}$     | $4.76 \times 10^{-4}$ **     |

267

268 A swarm of several bubbles was also injected into the column filled with the colorimetric  
269 solution. As shown in Figure 7, the wake of the bubbles was characterized by irregular, tortuous  
270 structures of dissolved O<sub>2</sub>, indicating a turbulent velocity field in the wake.

271

272 Thanks to the image processing developed in part 3, the total amount of oxygen dissolved in  
273 the wake of the bubble could be calculated as follows:

274 
$$\bar{m} = \iiint C(x, y) \cdot dx \cdot dy \cdot dz \quad (2)$$

275 These values and the bubble properties (size, velocity) are reported in Table 2. More than one  
276 hundred times more oxygen was transferred for the 2.12 mm bubble ( $3.18 \times 10^{-8}$  mg) than for  
277 the 0.82 mm bubble ( $3.48 \times 10^{-9}$  mg). This difference could be explained by the smaller bubble  
278 velocity and by the agitation in the bubble wake, which renewed the bubble surface. The  
279 purpose of this final section is to evaluate the liquid-side mass transfer coefficient ( $k_L$ ) from the  
280 dissolved oxygen concentration fields presented above. As established by Dietrich et al. (2013),  
281 the volumetric mass flux of oxygen from gas to liquid along a channel can be adapted to this  
282 system as:

283 
$$\varphi(X') = u_B \cdot \frac{\partial \bar{C}}{\partial X'} \quad (3)$$

284 where  $X'$  is the axial position of the bubble in the column such that  $X' = 0$  is the location where  
285 the bubbles are generated and  $X' = 0.2$  m the recorded position.  $\bar{C}$  is the average dissolved  
286 oxygen concentration accumulated in the column (mainly in the bubble wake) at the axial  
287 position  $X'$  along the vertical channel. Assuming that the concentration of dissolved oxygen in  
288 the liquid at the scale of the unit cell is zero due to its consumption by the chemical reaction,  
289 the mass flux of oxygen (per unit of liquid volume) can also be expressed by:

290 
$$\varphi(X') = k_L \cdot a \cdot C^* \quad (4)$$

291 where  $k_L$  is the liquid-side mass transfer coefficient (m/s),  $a$  the interfacial area between gas  
 292 and liquid phases ( $\text{m}^2/\text{m}^3$ ) and  $C^*$  the dissolved oxygen saturation concentration ( $C \sim 9 \text{ mgL}^{-1}$ ).  
 293 By combining Equations 3 and 4, relationship 5 is found:

$$294 \quad \frac{\partial \bar{C}}{\partial X'} = \frac{k_L \cdot a \cdot C^*}{u_B} \quad (5)$$

295 If the oxygen transferred at the bubble formation is neglected, integrating Equation 5 over the  
 296 entire column gives:

$$297 \quad \bar{C} = \frac{k_L \cdot a \cdot X' \cdot C^*}{u_B} \quad (6)$$

298 In order to estimate the interfacial area,  $a$ , between the gas and liquid phases, a spherical shape  
 299 is assumed for the 0.82 mm diameter bubble and ellipsoidal shape for the 2.12 mm diameter  
 300 bubbles.

301

302 The oxygen transferred by a single bubble can then be tracked by the dissolved oxygen  
 303 accumulation in the cell, as:

$$304 \quad \bar{C} = \frac{\bar{m}}{V_{column}} = \frac{\iiint C(x, y) \cdot dx \cdot dy \cdot dz}{X \cdot S} \quad (7)$$

305

306 Equation 7 does not take the dimension  $z$ , related to the channel width, into account when  
 307 integrating  $C$ . This is because the present colorimetric technique is not able to discriminate the  
 308 visualizations at different planes along the channel width. As resorufin is fluorescent, it would  
 309 be possible to use a laser sheet to illuminate the column. Consequently, for this study, we should  
 310 keep in mind that the oxygen concentration fields visualized are the result of all the different  
 311 fields existing in all the vertical locations.

312 Finally, from Equations 6 and 7, the liquid-side mass transfer coefficient can be calculated as:

$$313 \quad k_L = \frac{\bar{C} \times u_B}{X' \times a \times C^*} \quad (8)$$

314 The values of  $k_L$  are reported in Table 2. A decrease in the mass transfer coefficient is observed  
315 with decreasing bubble size. This decrease is attributable to the fall in the bubble slip velocity.  
316 To validate the colorimetric technique, the liquid-side mass transfer coefficients should be  
317 compared with those found in the literature and especially with the liquid-side mass transfer  
318 coefficients from Higbie's penetration theory (bubble diameter  $> 2$  mm) as:

$$319 \quad k_L = 2 \sqrt{\frac{D}{\pi \cdot t_C}} \quad (9)$$

320 and the Frossling theory (bubble diameter  $< 2$  mm):

$$321 \quad k_L = \frac{D_{O_2}}{d_B} \cdot (2 + 0.6 \cdot \text{Re}^{0.5} \cdot \text{Sc}^{0.33}) \quad (10)$$

322 Rather good agreement is observed with Higbie's model and with the Frossling model,  
323 demonstrating how this new visualization technique (easy to implement as without a laser  
324 source) can bring new insights for investigating mass transfer processes in a 3D **column**.

325

## 326 **5. Conclusion**

327 An oxygen-sensitive dye technique for visualizing gas-liquid mass transfer (Dietrich et al.,  
328 2013) at the interface of rising gas bubbles has been implemented for a free rising bubble for  
329 the first time. Resorufin reacts quickly with oxygen without altering the mass transfer. The  
330 technique was applied to rising bubbles with diameters between 0.82 and 2.12 mm presenting  
331 rectilinear, oscillatory or irregular motions. The resulting image sequences reveal the evolution  
332 of characteristic structures in the wake of the bubbles. The technique thus proves to be an  
333 extension of the existing experimental methods with the potential of providing new and detailed  
334 insights into the spatio-temporal dynamics of mass transfer of rising gas bubbles. A calculation  
335 method developed for this experiment made it possible to estimate the mass transfer coefficient.  
336 Good agreement was observed, validating the method at this scale and in a 3D column. This  
337 study thus constitutes a striking example showing that this colorimetric method could be an

338 interesting tool for investigating gas-liquid mass transfer in transparent fluids, with a view to  
339 reactor design. Moreover, this information gives new information into the complex mechanism  
340 of bubble mass transfer and could contribute to the development of rigorous theoretical models  
341 and numerical simulations.

342

343

344 **Nomenclature**

345  $Re = \frac{U_B \cdot d_B \rho}{\mu}$  Reynolds number, dimensionless

346  $Sc = \frac{\mu}{\rho \cdot D_{O_2}}$  Schmidt number, dimensionless

347

348 a interfacial area,  $m^{-1}$

349 C oxygen concentration,  $mg.L^{-1}$

350 C\* oxygen solubility,  $mg.L^{-1}$

351 d diameter, m

352 D diffusion coefficient,  $m^2.s^{-1}$

353  $\phi$  flux,  $g.m^{-2}.s^{-1}$

354  $k_L$  mass transfer coefficient,  $m.s^{-1}$

355 m mass, g

356 n amount, mol

357 S column section,  $m^2$

358  $t_c$  bubble characteristic time =  $d/u_B$ , s

359  $u_B$  velocity,  $m.s^{-1}$

360 V volume,  $m^3$

361 X' measurement position, m

362 x, y, z distance, m

363

364

365 *Greek letters*

366  $\sigma$  tension force, N/m

367  $\mu$  viscosity, Pa.s

368  $\rho$  density,  $kg.m^{-3}$

369

370 *Subscripts*

371 c characteristic time

372 B bubble

373

374 *Superscripts*

375  $\bar{\quad}$  average of the value

376 \* oxygen-saturated solution (theoretical value)

377

378

379

380 **Table legends**

381 Table 1. Properties of clean water and of the synthetic liquid media at 20°C.

382 Table 2. Hydrodynamic and mass transfer properties of bubbles. Each condition was  
383 reproduced three times and averaged (deviation ~ 3%) \* Frossling model. \*\* Higbie model.

384

385 **Figure legends**

386 Figure 1: Reduction of resazurin to resofurin by glucose and sodium hydroxide (step [1]).

387 Reduction of resofurin to dihydroresorufin in presence of carbohydrate (step [2]). Reoxidation  
388 of dihydroresorufin to resorufin in presence of oxygen (step [3]).

389 Figure 2: Experimental set-up.

390 Figure 3: Image processing procedure. (a) Raw image (b) Background removal (c) Removed  
391 noise visualization (d) Bubble edge detection (e) Bubble removal (f) Oxygen concentration  
392 field.

393 Figure 4: Calibration curve

394 Figure 5: Visualization of dissolved oxygen concentration around a spherical bubble rising in  
395 the colorimetric solution at rest. The composition of this solution is: 100 mg.L<sup>-1</sup>of resazurin,  
396 20 g.L<sup>-1</sup>of glucose and (a) 20 g.L<sup>-1</sup>NaOH (b) 50 g.L<sup>-1</sup>NaOH.

397 Figure 6: (a) Visualization of dissolved oxygen concentration around an ellipsoidal bubble  
398 rising in the colorimetric solution at rest. 100 mg.L<sup>-1</sup>of resazurin, 20 g.L<sup>-1</sup>of glucose and 20  
399 g.L<sup>-1</sup>of NaOH. (b) Visualization of hydrodynamics around an ellipsoidal bubble rising in a  
400 kallirosopic solution.

401 Figure 7: Visualization of dissolved oxygen concentration around a swarm of ellipsoidal  
402 bubbles rising in the colorimetric solution at rest. 100 mg.L<sup>-1</sup>of resazurin, 20 g.L<sup>-1</sup>of glucose  
403 and 20 g.L<sup>-1</sup>of NaOH.

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