



# Visualisation of gas-liquid mass transfer around a rising bubble in a quiescent liquid using an oxygen sensitive dye

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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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## **Abstract**

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

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

## 1. Introduction

Gas-liquid contactors play an important role in engineering processes, where they have many applications, including water treatment and (bio)-chemical processes (Dietrich et al., 2011; Tatin et al., 2015; Benizri et al., 2017; de Lamotte et al., 2017; Hariz et al., 2017; Kherbeche et al., 2017; Kováts et al., 2017; Wylock et al., 2017; Xie et al., 2017; Wongwailikhit et al., 2017). A good prediction of the mass transfer phenomena is necessary to achieve good reactor design leading to higher efficiency. The film model, penetration theory and the surface renewal model are three established, well-known theories describing interface mass transfer (Danckwerts, 1951; Higbie, 1935; Whitman, 1923). Based on these models, film-penetration theory and the unsteady two-film model were developed (Toor and Marchello, 1958; Wang and Langemann, 1994). In order to develop a further model, a good understanding of the mass transfer at interfaces is necessary. As the physical presence of sensors alters the hydrodynamics and the use of chemical compounds changes the liquid properties, the application of an invasive technique has marked limitations. As an alternative, non-invasive techniques such as visualization are a solution to estimate mass transfer accurately in bubbly systems. With this in mind, optical techniques have been established based on the use of a fluorescent dye, sensitive to dissolved oxygen molecules or pH, to measure the oxygen or carbon dioxide concentration at a gas-liquid interface. When the solution containing a fluorescent dye is illuminated by a 2D-laser sheet and the local data are acquired by digital cameras, the technique is named PLIF (Planar Laser Induced Fluorescence). This technique has been adapted for the visualization of oxygen concentration by studying the fluorescence of an oxygen-sensitive dye (Munsterer and Jahne, 1998; Mühlfriedel and Baumann, 2000; Herlina and Jirka, 2004; Falkenroth et al., 2007; Walker and Peirson, 2007; Jimenez et al., 2012b, 2012a). The inhibition of the fluorescence is used to quantify the oxygen concentration. This technique is called PLIFI (Planar Laser Induced Fluorescence with Inhibition). Studies concerning mass transfer around a single bubble have also been performed to provide qualitative measurements of oxygen mass

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

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

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

## **2. Material and Methods**

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

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

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

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

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

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

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

- (quasi)-instantaneous kinetics for the oxidation of dihydroresorufin (colourless) to resorufin (pink),
- sufficiently slow kinetics (few minutes) for the reduction of resorufin (pink) to dihydroresorufin (colourless),
- 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-

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

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

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			

### 3. Implementation of the colorimetric technique

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

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

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

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

its wake remained. However, because of some temporal non-homogeneities in the lighting, the background grey level (and oxygen concentration) could differ from 0.

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

#### 4. Results

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

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

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

	<i>Diameter</i> [mm]	<i>Velocity</i> $u_B$ [m/s]	<i>Reynolds</i> <i>Number</i> [-]	<i>Mass</i> <i>transferred</i> [mg]	$k_L$ exp [m/s]	$k_L$ theory [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 
$$\overline{m} = \iiint C(x, y).dx \cdot dy \cdot dz \quad (2)$$

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

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.  $\overline{C}$  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 
$$\varphi(X') = k_L \cdot a \cdot C^* \quad (4)$$

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

## 5. Conclusion

An oxygen-sensitive dye technique for visualizing gas-liquid mass transfer (Dietrich et al., 2013) at the interface of rising gas bubbles has been implemented for a free rising bubble for the first time. Resorufin reacts quickly with oxygen without altering the mass transfer. The technique was applied to rising bubbles with diameters between 0.82 and 2.12 mm presenting rectilinear, oscillatory or irregular motions. The resulting image sequences reveal the evolution of characteristic structures in the wake of the bubbles. The technique thus proves to be an extension of the existing experimental methods with the potential of providing new and detailed insights into the spatio-temporal dynamics of mass transfer of rising gas bubbles. A calculation method developed for this experiment made it possible to estimate the mass transfer coefficient. Good agreement was observed, validating the method at this scale and in a 3D column. This 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 <sup>-1</sup>
349	C	oxygen concentration, mg.L <sup>-1</sup>
350	C*	oxygen solubility, mg.L <sup>-1</sup>
351	d	diameter, m
352	D	diffusion coefficient, m <sup>2</sup> .s <sup>-1</sup>
353	φ	flux, g.m <sup>-2</sup> .s <sup>-1</sup>
354	k <sub>L</sub>	mass transfer coefficient, m.s <sup>-1</sup>
355	m	mass, g
356	n	amount, mol
357	S	column section, m <sup>2</sup>
358	t <sub>c</sub>	bubble characteristic time = d/u <sub>B</sub> , s
359	u <sub>B</sub>	velocity, m.s <sup>-1</sup>
360	V	volume, m <sup>3</sup>
361	X'	measurement position, m
362	x, y, z	distance, m

363

364

365 *Greek letters*

366	σ	tension force, N/m
367	μ	viscosity, Pa.s
368	ρ	density, kg.m <sup>-3</sup>

369

370 *Subscripts*

371	c	characteristic time
372	B	bubble

373

374 *Superscripts*

375	—	average of the value
376	*	oxygen-saturated solution (theoretical value)

377

378

379

## Table legends

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

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

## Figure legends

Figure 1: Reduction of resazurin to resofurin by glucose and sodium hydroxide (step [1]). Reduction of resofurin to dihydroresorufin in presence of carbohydrate (step [2]). Reoxidation of dihydroresorufin to resorufin in presence of oxygen (step [3]).

Figure 2: Experimental set-up.

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

Figure 4: Calibration curve

Figure 5: Visualization of dissolved oxygen concentration around a spherical bubble rising in the colorimetric solution at rest. The composition of this solution is: 100 mg.L<sup>-1</sup> of resazurin, 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.

Figure 6: (a) Visualization of dissolved oxygen concentration around an ellipsoidal bubble 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 g.L<sup>-1</sup> of NaOH. (b) Visualization of hydrodynamics around an ellipsoidal bubble rising in a kallirotropic solution.

Figure 7: Visualization of dissolved oxygen concentration around a swarm of ellipsoidal bubbles 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 g.L<sup>-1</sup> of NaOH.

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