

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

Nicolas Dietrich, Gilles Hébrard

▶ **To cite this version:**

Nicolas Dietrich, Gilles Hébrard. Visualisation of gas-liquid mass transfer around a rising bubble in a quiescent liquid using an oxygen sensitive dye. Heat and Mass Transfer, 2018, 54 (7), pp.2163-2171. 10.1007/s00231-018-2297-3 . hal-01886478

HAL Id: hal-01886478
https://hal.science/hal-01886478

Submitted on 17 Jun 2019

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

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

Nicolas DIETRICH* & Gilles HEBRARD

LISBP, Université de Toulouse, CNRS, INRA, INSA, Toulouse, France

* To whom correspondence should be addressed.

Tel.: +33(0)5 61 55 97 81, Fax: +33 (0)5 61 55 97 60, E-mail: dietrich@insa-toulouse.fr

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 $g.mol^{-1}$).

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).

From this, the following formulation was selected: glucose and sodium hydroxide were both diluted at 20 $g.L^{-1}$ in deionized water, and the concentration of resazurin was fixed at 0.1 $g.L^{-1}$. The experimental set-up consisted of a transparent column fitted with a bubble injection system, and optical equipment designed to allow the rising bubble to be visualized. It is shown in Figure 2. The 100 x 100 x 300 mm^3 column had a square cross-section. It had three transparent glass sides and one PMMA side where the injection system was set. The base was made of glass and the top of the column was open. The mass transfer from the top is considered as neglectable (Jimenez et al., 2012a). The injection system was placed 10 mm from the bottom of the column. It was composed of a very thin glass capillary with an inner diameter that could range from 0.01 to 0.1 mm connected to an automatically controlled syringe (Harvard Apparatus PHD 2000). The inner diameter was changed in order to generate bubbles of several sizes. The syringe was filled with air and the flow rate was chosen to produce only a single bubble at a time. Bubbles were formed at the centre of the column in a liquid containing the colorimetric solution. A drain tap was fitted at the bottom of the column to evacuate the liquid easily after each experiment.

A CCD (Charge-Coupled-Device) colour camera (Guppy Pro, Allied Vision, Germany, 8 bits, 30 Hz, 1024 x 1024 $pixels^2$) was placed horizontally, perpendicular to the column - on the x axis - and was focused on the centre of the column. The digital camera was fitted with a 105 mm objective (Micro-Nikkor 60 mm f/2.8, Nikon) to reach a high spatial resolution of approximately 0.023 mm/pixel on the focused area. A backlight panel was placed behind the column to film the bubble by the shadowgraph technique. The colorimetric images of the moving bubbles in the column were recorded and treated by a procedure developed in Matlab software. Details of this study will be described in the next section. Experiments were performed at 295 K in an air conditioned room, and at atmospheric pressure. Fluids were air and deionized water containing D-glucose anhydrous (Fischer Scientific®, CAS 50-99-7), sodium hydroxide (VWR®, CAS 1310-73-2) and resazurin (Sigma Aldrich®, CAS 62758-13-

8). Density (ρ_L), dynamic viscosity (μ_L) and static surface tension (σ_L) were measured respectively by means of a pycnometer (ρ_L ± 0.2 kg.m^{-3}), an RM180 Rheomat Rheometric Scientific® viscometer (μ_L ± 10^{-3} mPa.s), and Digidrop GBX® or Krüss tensiometers (σ_L ± 0.5 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 (g.L^{-1})	σ_L (mN.m^{-1})	μ_L (mPa.s)	ρ_L (kg.m^{-3})
Deionized water	-	73.4	1.003	996.8
Glucose	20	75.5	1.1179	1005.2
NaOH	20			
Glucose	20			
NaOH	20	75.4	1.1179	1005.3
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 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 stoechiometry 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 $mg.L^{-1}$), it was possible to associate the observed averaged grey level with a dissolved oxygen concentration: 0, 0.075, .25, and 1 $mg.L^{-1}$) 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_0. 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_0 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 $g.L^{-1}$ (Figure 5-a) and 20 $g.L^{-1}$ (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 $g.L^{-1}$). 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 $g.L^{-1}$, σ = 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 Kalliroscopic 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).

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

A swarm of several bubbles was also injected into the column filled with the colorimetric solution. As shown in Figure 7, the wake of the bubbles was characterized by irregular, tortuous structures of dissolved O_2, indicating a turbulent velocity field in the wake.

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

$$\overline{m} = \iiint C(x,y).dx \cdot dy \cdot dz \qquad (2)$$

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

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

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

$$\varphi(X') = k_L.a.C^* \qquad (4)$$

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

$$\frac{\partial \overline{C}}{\partial X'} = \frac{k_L.a.C^*}{u_B} \qquad (5)$$

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

$$\overline{C} = \frac{k_L.a.X'.C^*}{u_B} \qquad (6)$$

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:

$$\overline{C} = \frac{\overline{m}}{V_{column}} = \frac{\iiint C(x,y).dx \cdot dy \cdot dz}{X.\ S} \qquad (7)$$

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:

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

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):

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

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

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

Nomenclature

$\mathrm{Re} = \frac{U_B.d_B\rho}{\mu}$	Reynolds number, dimensionless
$Sc = \frac{\mu}{\rho.D_{O_2}}$	Schmidt number, dimensionless

a	interfacial area, m^{-1}
C	oxygen concentration, $mg.L^{-1}$
C^*	oxygen solubility, $mg.L^{-1}$
d	diameter, m
D	diffusion coefficient, $m^2.s^{-1}$
ϕ	flux, $g.m^{-2}.s^{-1}$
k_L	mass transfer coefficient, $m.s^{-1}$
m	mass, g
n	amount, mol
S	column section, m^2
t_c	bubble characteristic time = d/u_B, s
u_B	velocity, $m.s^{-1}$
V	volume, m^3
X’	measurement position, m
x, y, z	distance, m

Greek letters

σ	tension force, N/m
μ	viscosity, Pa.s
ρ	density, $kg.m^{-3}$

Subscripts

c	characteristic time
B	bubble

Superscripts

$\bar{}$	average of the value
*	oxygen-saturated solution (theoretical value)

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 $mg.L^{-1}$of resazurin, 20 $g.L^{-1}$of glucose and (a) 20 $g.L^{-1}$NaOH (b) 50 $g.L^{-1}$NaOH.

Figure 6: (a) Visualization of dissolved oxygen concentration around an ellipsoidal bubble rising in the colorimetric solution at rest. 100 $mg.L^{-1}$of resasurin, 20 $g.L^{-1}$of glucose and 20 $g.L^{-1}$of NaOH. (b) Visualization of hydrodynamics around an ellipsoidal bubble rising in a kalliroscopic solution.

Figure 7: Visualization of dissolved oxygen concentration around a swarm of ellipsoidal bubbles rising in the colorimetric solution at rest. 100 $mg.L^{-1}$of resazurin, 20 $g.L^{-1}$of glucose and 20 $g.L^{-1}$of NaOH.

References

Benizri, D., Dietrich, N., Hébrard, G., 2017. Experimental characterization of multi-component absorption in complex liquid: New method and apparatus. Chemical Engineering Science, 13th International Conference on Gas-Liquid and Gas-Liquid-Solid Reactor Engineering 170, 116–121. https://doi.org/10.1016/j.ces.2017.03.024

Bork, O., Schlueter, M., Raebiger, N., 2005. The Impact of Local Phenomena on Mass Transfer in Gas-Liquid Systems. Can. J. Chem. Eng. 83, 658–666. https://doi.org/10.1002/cjce.5450830406

Butler, C., Cid, E., Billet, A.-M., 2016. Modelling of mass transfer in Taylor flow: Investigation with the PLIF-I technique. Chemical Engineering Research and Design, 10th European Congress of Chemical Engineering 115, Part B, 292–302. https://doi.org/10.1016/j.cherd.2016.09.001

Campbell, J.A., 1963. Kinetics—Early and often. J. Chem. Educ. 40, 578. https://doi.org/10.1021/ed040p578

Danckwerts, P.V., 1951. Significance of Liquid-Film Coefficients in Gas Absorption. Ind. Eng. Chem. 43, 1460–1467. https://doi.org/10.1021/ie50498a055

Dani, A., Guiraud, P., Cockx, A., 2007. Local measurement of oxygen transfer around a single bubble by planar laser-induced fluorescence. Chemical Engineering Science 62, 7245–7252. https://doi.org/10.1016/j.ces.2007.08.047

de Lamotte, A., Delafosse, A., Calvo, S., Delvigne, F., Toye, D., 2017. Investigating the effects of hydrodynamics and mixing on mass transfer through the free-surface in stirred tank bioreactors. Chemical Engineering Science 172, 125–142. https://doi.org/10.1016/j.ces.2017.06.028

Dietrich, N., Francois, J., Jimenez, M., Cockx, A., Guiraud, P., Hébrard, G., 2015. Fast Measurements of the Gas-Liquid Diffusion Coefficient in the Gaussian Wake of a Spherical Bubble. Chem. Eng. Technol. 38, 941–946. https://doi.org/10.1002/ceat.201400471

Dietrich, N., Loubière, K., Jimenez, M., Hébrard, G., Gourdon, C., 2013. A new direct technique for visualizing and measuring gas–liquid mass transfer around bubbles moving in a straight millimetric square channel. Chemical Engineering Science 100, 172–182. https://doi.org/10.1016/j.ces.2013.03.041

Dietrich, N., Poncin, S., Li, H.Z., 2011. Dynamical deformation of a flat liquid–liquid interface. Exp Fluids. https://doi.org/10.1007/s00348-010-0989-7

Dietrich, N., Poncin, S., Pheulpin, S., Huai, Z.L., 2008. Passage of a bubble through a liquid-liquid interface. AIChE Journal 54, 594–600.

Falkenroth, A., Degreif, K., Jähne, B., 2007. Visualisation of Oxygen Concentration Fields in the Mass Boundary Layer by Fluorescence Quenching, in: Garbe, P.-D.D.C.S., Handler, D.R.A., Jähne, P.D.B. (Eds.), Transport at the Air-Sea Interface, Environmental Science and Engineering. Springer Berlin Heidelberg, pp. 59–72.

Francois, J., Dietrich, N., Cockx, A., 2011a. A Novel Methodology to Measure Mass Transfer Around a Bubble. Mod. Phys. Lett. B 25, 1993–2000. https://doi.org/10.1142/S0217984911027236

Francois, J., Dietrich, N., Guiraud, P., Cockx, A., 2011b. Direct measurement of mass transfer around a single bubble by micro-PLIFI. Chemical Engineering Science In Press, Corrected Proof. https://doi.org/10.1016/j.ces.2011.01.049

Hanyu, K., Saito, T., 2010. Dynamical mass-transfer process of a CO2 bubble measured by using LIF/HPTS visualisation and photoelectric probing. Can. J. Chem. Eng. 88, 551–560. https://doi.org/10.1002/cjce.20319

Hariz, R., del Rio Sanz, J.I., Mercier, C., Valentin, R., Dietrich, N., Mouloungui, Z., Hébrard, G., 2017. Absorption of toluene by vegetable oil–water emulsion in scrubbing tower:

Experiments and modeling. Chemical Engineering Science, 12th International Conference on Gas-Liquid and Gas-Liquid-Solid Reactor Engineering 157, 264–271. https://doi.org/10.1016/j.ces.2016.06.008
Herlina, G., Jirka, G.H., 2004. Application of LIF to investigate gas transfer near the air-water interface in a grid-stirred tank. Experiments in Fluids 37, 341–34.
Higbie, R., 1935. The rate of absorption of a pure gas into a still liquid during short periods of exposure. Transactions of American Institute of Chemical Engineers.
Huang, J., Saito, T., 2017. Discussion about the differences in mass transfer, bubble motion and surrounding liquid motion between a contaminated system and a clean system based on consideration of three-dimensional wake structure obtained from LIF visualization. Chemical Engineering Science, 13th International Conference on Gas-Liquid and Gas-Liquid-Solid Reactor Engineering 170, 105–115. https://doi.org/10.1016/j.ces.2017.03.030
Huang, J., Saito, T., 2015. Influence of Bubble-Surface Contamination on Instantaneous Mass Transfer. Chem. Eng. Technol. 38, 1947–1954. https://doi.org/10.1002/ceat.201500056
Jimenez, M., Dietrich, N., Cockx, A., Hébrard, and G., 2012a. Experimental study of O2 diffusion coefficient measurement at a planar gas–liquid interface by planar laser-induced fluorescence with inhibition. AIChE J 59, 325–333. https://doi.org/10.1002/aic.13805
Jimenez, M., Dietrich, N., Cockx, A., Hébrard, G., 2013a. Experimental study of O2 diffusion coefficient measurement at a planar gas–liquid interface by planar laser-induced fluorescence with inhibition. AIChE J 59, 325–333. https://doi.org/10.1002/aic.13805
Jimenez, M., Dietrich, N., Grace, J.R., Hébrard, G., 2014. Oxygen mass transfer and hydrodynamic behaviour in wastewater: determination of local impact of surfactants by visualization techniques. Water Res. 58, 111–121. https://doi.org/10.1016/j.watres.2014.03.065
Jimenez, M., Dietrich, N., Hébrard, G., 2013b. Mass transfer in the wake of non-spherical air bubbles quantified by quenching of fluorescence. Chemical Engineering Science, 11th International Conference on Gas-Liquid and Gas-Liquid-Solid Reactor Engineering 100, 160–171. https://doi.org/10.1016/j.ces.2013.01.036
Jimenez, M., Dietrich, N., Hebrard, G., 2012b. A New Method for Measuring Diffusion Coefficient of Gases in Liquids by Plif. Mod. Phys. Lett. B 26, 1150034. https://doi.org/10.1142/S0217984911500345
Kastens, S., Meyer, C., Hoffmann, M., Schlüter, M., 2017. Experimental Investigation and Modelling of Local Mass Transfer Rates in Pure and Contaminated Taylor Flows, in: Transport Processes at Fluidic Interfaces, Advances in Mathematical Fluid Mechanics. Birkhäuser, Cham, pp. 609–637. https://doi.org/10.1007/978-3-319-56602-3_21
Kherbeche, A., Milnes, J., Jimenez, M., Dietrich, N., Hébrard, G., Lekhlif, B., 2013. Multi-scale analysis of the influence of physicochemical parameters on the hydrodynamic and gas–liquid mass transfer in gas/liquid/solid reactors. Chemical Engineering Science 100, 515–528. https://doi.org/10.1016/j.ces.2013.06.025
Kherbeche, A., Nsakou, S.N., Lekhlif, B., Hébrard, G., Dietrich, N., 2017. Study of the initial glycerol concentration effects upon bacterial cells adaptation and biodegradation kinetics on a submerged aerated fixed bed reactor using biocell® packing. Journal of Materials and Environmental Sciences.
Klein, H., Wanders, K., 1987. Congress of the International Astronautical FederationHolographic interferometry near gas/liquid critical points: Results of spacelab D-1. Acta Astronautica 15, 463–465. https://doi.org/10.1016/0094-5765(87)90183-4

Kováts, P., Thévenin, D., Zähringer, K., 2017. Characterizing fluid dynamics in a bubble column aimed for the determination of reactive mass transfer. Heat Mass Transfer 1–9. https://doi.org/10.1007/s00231-017-2142-0

Krieger, W., Lamsfuß, J., Zhang, W., Kockmann, N., n.d. Local Mass Transfer Phenomena and Chemical Selectivity of Gas-Liquid Reactions in Capillaries. Chem. Eng. Technol. n/a-n/a. https://doi.org/10.1002/ceat.201700420

Kutepov, A.M., Pokusaev, B.G., Kazenin, D.A., Karlov, S.P., Vyaz'min, A.V., 2001. Interfacial Mass Transfer in the Liquid–Gas System: An Optical Study. Theoretical Foundations of Chemical Engineering 35, 213–216. https://doi.org/10.1023/A:1010409322333

Mühlfriedel, K., Baumann, K.-H., 2000. Concentration measurements during mass transfer across liquid-phase boundaries using planar laser induced fluorescence (PLIF). Experiments in Fluids 28, 279–281. https://doi.org/10.1007/s003480050388

Munsterer, T., Jahne, B., 1998. LIF measurements of concentration profiles in the aqueous mass boundary layer. Exp. Fluids 25, 190–196. https://doi.org/10.1007/s003480050223

Roudet, M., Billet, A.-M., Cazin, S., Risso, F., Roig, V., 2017. Experimental investigation of interfacial mass transfer mechanisms for a confined high-reynolds-number bubble rising in a thin gap. AIChE J. 63, 2394–2408. https://doi.org/10.1002/aic.15562

Roustan, M., 2003. Transferts gaz-liquide dans les procédés de traitement des eaux et des effluents gazeux [WWW Document]. URL http://cat.inist.fr/?aModele=afficheN&cpsidt=15123114 (accessed 1.9.14).

Roy, S., Duke, S.R., 2004. Visualization of oxygen concentration fields and measurement of concentration gradients at bubble surfaces in surfactant-contaminated water. Experiments in Fluids 36, 654–662. https://doi.org/10.1007/s00348-003-0771-1

Saito, T., Toriu, M., 2015. Effects of a bubble and the surrounding liquid motions on the instantaneous mass transfer across the gas–liquid interface. Chemical Engineering Journal 265, 164–175. https://doi.org/10.1016/j.cej.2014.12.039

Someya, S., Bando, S., Song, Y., Chen, B., Nishio, M., 2005. DeLIF measurement of pH distribution around dissolving CO2 droplet in high pressure vessel. International Journal of Heat and Mass Transfer 48, 2508–2515. https://doi.org/10.1016/j.ijheatmasstransfer.2004.12.042

Stöhr, M., Schanze, J., Khalili, A., 2009. Visualization of gas–liquid mass transfer and wake structure of rising bubbles using pH-sensitive PLIF. Exp Fluids 47, 135–143. https://doi.org/10.1007/s00348-009-0633-6

Tatin, R., Moura, L., Dietrich, N., Baig, S., Hébrard, G., 2015. Physical absorption of volatile organic compounds by spraying emulsion in a spray tower: Experiments and modelling. Chemical Engineering Research and Design 104, 409–415. https://doi.org/10.1016/j.cherd.2015.08.030

Toor, H.L., Marchello, J.M., 1958. Film-penetration model for mass and heat transfer. AIChE J. 4, 97–101. https://doi.org/10.1002/aic.690040118

Walker, J.W., Peirson, W.L., 2007. Measurement of gas transfer across wind-forced wavy air–water interfaces using laser-induced fluorescence. Exp Fluids 44, 249–259. https://doi.org/10.1007/s00348-007-0398-8

Wang, J., Langemann, H., 1994. Unsteady two-film model for mass transfer. Chem. Eng. Technol. 17, 280–284. https://doi.org/10.1002/ceat.270170410

Whitman, W.G., 1923. Preliminary experimental confirmation of the two-film theory of gas absorption. Chemical and Metallurgical Engineering 146–8.

Wongwailikhit, K., Warunyuwong, P., Chawaloesphonsiya, N., Dietrich, N., Hébrard, G., Painmanakul, P., 2017. Gas Sparger Orifice Sizes and Solid Particle Characteristics in

a Bubble Column – Relative Effect on Hydrodynamics and Mass Transfer. Chem. Eng. Technol. n/a-n/a. https://doi.org/10.1002/ceat.201700293

Wylock, C., Dehaeck, S., Cartage, T., Colinet, P., Haut, B., 2011. Experimental study of gas-liquid mass transfer coupled with chemical reactions by digital holographic interferometry, in: Chemical Engineering Science. Presented at the International Conference on Gas-Liquid and Gas-Liquid-Solid Reactor Engineering, Elsevier, pp. 3400–3412.

Wylock, C., Rednikov, A., Colinet, P., Haut, B., 2017. Experimental and numerical analysis of buoyancy-induced instability during CO2 absorption in NaHCO3–Na2CO3 aqueous solutions. Chemical Engineering Science, 12th International Conference on Gas-Liquid and Gas-Liquid-Solid Reactor Engineering 157, 232–246. https://doi.org/10.1016/j.ces.2016.04.061

Xie, X., Le Men, C., Dietrich, N., Schmitz, P., Fillaudeau, L., 2017. Local hydrodynamic investigation by PIV and CFD within a Dynamic filtration unit under laminar flow. Separation and Purification Technology. https://doi.org/10.1016/j.seppur.2017.04.009

Xu, F., Jimenez, M., Dietrich, N., Hébrard, G., 2017. Fast determination of gas-liquid diffusion coefficient by an innovative double approach. Chemical Engineering Science. https://doi.org/10.1016/j.ces.2017.02.043

Yang, L., Dietrich, N., Hébrard, G., Loubière, K., Gourdon, C., 2017a. Optical methods to investigate the enhancement factor of an oxygen-sensitive colorimetric reaction using microreactors. AIChE J. 63, 2272–2284. https://doi.org/10.1002/aic.15547

Yang, L., Dietrich, N., Loubière, K., Gourdon, C., Hébrard, G., 2016. Visualization And Characterization Of Gas-Liquid Mass Transfer Around A Taylor Bubble Right After The Formation Stage In Microreactors. Chemical Engineering Science. https://doi.org/10.1016/j.ces.2016.01.013

Yang, L., Loubière, K., Dietrich, N., Le Men, C., Gourdon, C., Hébrard, G., 2017b. Local investigations on the gas-liquid mass transfer around Taylor bubbles flowing in a meandering millimetric square channel. Chemical Engineering Science 165, 192–203. https://doi.org/10.1016/j.ces.2017.03.007