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1 **COMPARISON OF THREE DIFFERENT TECHNIQUES FOR GAS-LIQUID**
2 **MASS TRANSFER VISUALIZATION**

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4
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8
9 **Abstract**

10 In this article three different techniques for visualizing gas-liquid mass transfer are
11 reviewed according to their applications in air-water systems. The three techniques
12 are: Planar Laser Induced Fluorescence (PLIF with the use of fluorescent resorufin),
13 PLIF with Inhibition (PLIF-I with the use of a ruthenium complex), and a
14 colorimetric technique (using a redox reaction with the pink-colored dye resorufin).
15 For comparison purpose, the three techniques were conducted in the same
16 experimental set-up to characterize the local oxygen mass transfer from a single air
17 bubble (with equivalent diameter ~1 mm) rising in water. This paper rigorously
18 compares these three techniques and aims to point out their advantages and
19 limitations. The comparison was divided into two parts: the visualization and the
20 quantification of the mass transfer. The discussion focused on the image quality, the
21 required equipment, and the accuracy of the quantification results including the mass
22 flux, the diffusion coefficient and the mass transfer coefficient. A guideline was
23 provided for choosing the technique that most accurately visualizes and characterizes
24 the local mass transfer in gas-liquid systems.

25 **Keywords:** Bubble, Gas-liquid, Mass transfer, Fluorescence, Colorimetric reaction,
26 Fluorescence quenching

27 1. INTRODUCTION

28 Most of the modern visualization techniques for gas-liquid mass transfer are based on
29 the addition of an indicator (fluorescent dye or colorimetric dye) into the liquid phase
30 which can be captured by optical instrument [1,2]. Compared with the probe
31 technique [3,4], the visualization techniques have the advantages of no flow
32 disturbance, quick response time and high visual resolution, and the capacity to
33 elucidate the profile near the gas-liquid interface despite its limited thickness [5].
34 They can also be easily combined with other optical methods that can provide
35 simultaneous measurements of flow velocity and temperature [6].

36 According to the principle of these techniques, the main visualization techniques can
37 be classified as the traditional fluorescence technique (i.e. PLIF: Planar Laser Induced
38 Fluorescence), the inhibited fluorescence technique (i.e. PLIF-I: Planar Laser Induced
39 Fluorescence with Inhibition), and the colorimetric technique (with a dissolved
40 gas-sensitive dye).

41 1.1 Traditional fluorescence technique

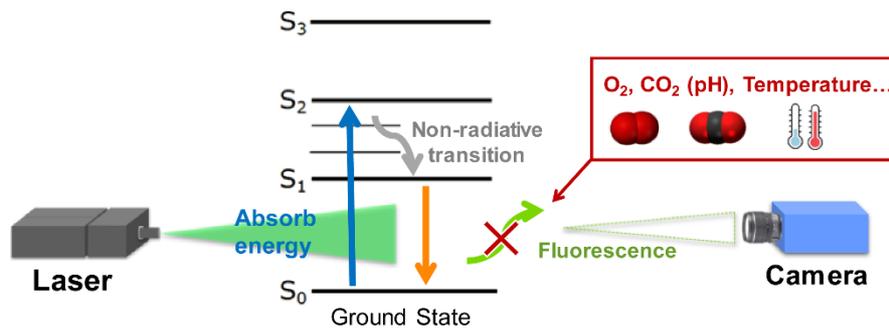
42 Planar Laser-induced fluorescence technique (PLIF) was proposed in the late 1960s
43 and was applied to gas combustion process for studying the heat transfer at early stage
44 [7]. Then PLIF has been rapidly extended to study the mixing by the dispersion of
45 fluorescent tracers in liquid and gaseous flows. As a traditional fluorescence
46 technique, the measurement is based on the linearity between the fluorescence
47 intensity and the concentration of fluorophore in the weak excitation regime [8].
48 Under weak excitation condition (excitation intensity I much smaller than the
49 saturation intensity of the dye), the relationship between fluorescence level F and
50 local concentration of the dye C_{dye} has a simplified relationship:

$$51 \quad F \propto I \cdot C_{dye} \quad (1)$$

52 where I can be assumed as constant [9,10]. Thus, using a multi-pixel light sensitive
 53 sensor (CMOS or CCD cameras), one can record the fluorescence signal and thus
 54 determine the corresponding instantaneous concentration field of the dye.

55 **1.2 Inhibited fluorescence technique**

56 The traditional PLIF requires the transported species itself play as a role of
 57 fluorophore which is difficult to realize for most gas-liquid species. Therefore, several
 58 variants of PLIF technique appears with the respective personalized name: PLIF-I
 59 (PLIF with Inhibition [11]), 2T-LIF (Two-tracer LIF [12]), LIF/HPTS [13], BPLIF
 60 (Blocked PLI, [14]), I_{PH}^R -PLIF (Ratiometric pH-sensitive-inhibited [15]), DeLIF
 61 (Dual Emission LIF, [16]), etc. Different from traditional PLIF techniques, the
 62 fluorophore is here pre-mixed homogeneously into the liquid. Fluorescence of this
 63 kind of dye under monochromatic excitation can be affected by the presence of a
 64 flow-passive scalar (e.g. concentration of a chemical species, pH value, temperature
 65 etc.) (Figure 1). The value of this scalar can be deduced from the variation of
 66 fluorescence intensity once a previous calibration is done that fluorescence intensities
 67 are measured for several preset values of the scalar.



68

69 **Figure 1 Schematic description of the principle of variant LIF techniques by**
 70 **Jablonski diagram.**

71

72 A representative example of the inhibited fluorescence technique (noted as PLIF-I
 73 afterward) is based on the quenching phenomena of the fluorescence [17]. In a

74 quenching experiment, the ability of some molecules called “quenchers” to inhibit the
75 fluorescence is considered which can reduce the fluorescence lifetime of the
76 fluorophore. Regarding the fluorescence lifetime is proportional to fluorescence
77 intensity, the dynamic quenching process is expressed as the Stern-Volmer
78 relationship:

$$79 \quad \frac{I_Q}{I_0} = \frac{1}{1+K_{SV}Q} \quad (2)$$

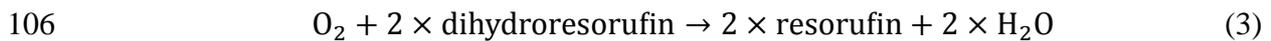
80 where K_{SV} is the Stern-Volmer constant, and I_Q and I_0 the fluorescence intensities
81 in the presence and absence of quencher, respectively. The parameters I_0 and K_{SV} of
82 the Stern-Volmer equation can be easily determined by a calibration process by
83 measuring several I_Q values under different preset concentration of quenchers.

84 The combination of fluorescence intensity measurements with the effect of
85 fluorescence quenching for gas concentration measurements in liquids was first
86 introduced by Vaughn and Weber [18]. As oxygen has been known as an excellent
87 quencher, Wolff et al. [19] introduced primarily PLIF-I technique to visualize O_2
88 concentration near a gas-liquid interface. Many following researchers [5,20–22] have
89 continued to work on the same subject by studying the absorption of O_2 .

90 **1.3 Colorimetric technique**

91 The third category of techniques calls the colorimetric technique. It is based on the
92 oxidation and reduction reactions by which the concentration of the target species can
93 be deduced from the change in color of a dissolved dye. Among the literature about
94 colorimetric methods, methylene blue [23], resazurin [24] and leuco-indigo carmine
95 [25] were mainly applied as dyes to investigate mass transfer and concentration fields.
96 The method used in this study was the colorimetric technique proposed by Dietrich et
97 al. [24]. Resazurin (CAS 62758-13-8, molecular mass: $229.19 \text{ g}\cdot\text{mol}^{-1}$) was chosen as
98 the dye which reacts with oxygen in the presence of sodium hydroxide (NaOH) and
99 glucose solution.

100 When resazurin dissolves in water (blue and not fluorescent), it is reduced to resorufin
101 (pink and highly fluorescent). Then reversible reduction-oxidation occurs between
102 resorufin and dihydroresorufin (colorless and not fluorescent). Since the oxidation
103 reaction is quasi-instantaneous and the reduction reaction is slow (few minutes), it is
104 possible to visualize the pink colored field under a white light source. The oxidation
105 reaction can be expressed as follows:



107 When there is sufficient resazurin to react with all the transferred oxygen, the number
108 of moles of dissolved oxygen can be deduced from the number of moles of resazurin,
109 as

$$110 \quad n_{\text{O}_2, \text{transferred}} = n_{\text{O}_2, \text{reacted}} = \frac{n_{\text{resazurin}}}{2} \quad (4)$$

111 **1.4 Application of the visualization techniques to gas-liquid systems**

112 All the above three techniques have been applied to a wide variety of gas-liquid
113 contacting conditions, including bubbly flows, flows across the planar interface and
114 Taylor flows / confined flows.

115 Among several industrial processes (e.g. bubble column reactor, stirred tank, etc.), the
116 gas phase usually appears as a form of bubble swarms or isolated bubble because of
117 their high interfacial area for gas-liquid mass transfer [26]. The visualization
118 techniques provides an efficient tool to characterize the transferring process for
119 different type of gas: air [11,27–29], oxygen [20,30] and carbon dioxide [1,31,32].

120 A series of representative researches have been conducted which apply visualization
121 techniques to investigate mass transfer from bubbles in recent decades. Bork et al. [33]
122 investigated the impact of local phenomena on mass transfer from bubbles according
123 to the bubble diameters and the liquid viscosities as variation parameters by using the
124 quenching reaction with a ruthenium complex. Stöhr et al. [31] visualized the

125 gas-liquid mass transfer and wake structure of rising bubbles using acid-based PLIF.
126 The resulting PLIF image sequences revealed the evolution of characteristic patterns
127 in the near and far wake of the bubbles and further allowed the estimation of bubble
128 size and rise velocity. Kück et al. [30,34] investigated the impact of local phenomena
129 on mass transfer from single free-rising gas bubbles with and without a superimposed
130 chemical reaction using PLIF and PIV (Particle Image Velocimetry). Jimenez et al.
131 [11] worked on the mass transfer of an ellipsoidal bubble, also indicating the
132 influence of the bubble diameter and the liquid composition (glycerol, salt, glucose)
133 on the flux density by PLIF-I technique. Based on a similar configuration, Dietrich et
134 al. [35] captured pictures of the concentration field at micro-scale in the laminar
135 bubble wake, which enabled the gas-liquid diffusivity to be accurately evaluated in a
136 very short time of around one second. Saito and Toriu [36] used experimental results
137 from highly precise measurements of bubble volume to clarify the instantaneous
138 mass-transfer coefficients of a zigzag CO₂ bubble. The zigzag motion and surface
139 oscillation, as well as the CO₂ dissolution (mass transfer) process from the bubble to
140 the surrounding liquid, were simultaneously visualized by using two high-speed
141 cameras and mirrors with the PLIF with a PH sensitive dye HPTS. Based on a similar
142 configuration, Huang and Saito [13,37,38] discussed the influences of gas-liquid
143 interface contamination on the bubble motions, bubble wake patterns, and the mass
144 transfer process, through the reconstruction of three-dimensional bubble wakes in
145 purified water and contaminated water. Timmermann et al. [39] investigated the
146 influence of bubble collisions on the local concentration field at well-defined
147 conditions and that of a superimposed sodium sulfite reaction on the concentration
148 field after bouncing. Using a high-speed PIV/LIF system combined with direct
149 visualization, the local and temporal evolution of the CO₂ chemisorption in aqueous
150 NaOH solution in a cylindrical bubble column was investigated [12] and compared
151 with simulation results [40]. Focusing on the convective mass transfer contribution,
152 Kong et al. [41] used fluorescein as a dye to study the case of a millimetric CO₂

153 bubble suspended under a substrate and dissolved in pure liquid water. Based on the
154 reduction of the colorimetric indicator (resorufin) in presence of oxygen, Dietrich and
155 Hébrard [42] estimated the liquid-side mass transfer coefficient around a freely rising
156 bubble and the result proved that the colorimetric technique had the potential to
157 provide new, detailed insight into the spatiotemporal dynamics of the mass transfer of
158 rising gas bubbles. More recently, the PLIF-I technique was applied to a
159 non-Newtonian fluid to study the impact of fluid properties on the mass transfer
160 coefficient [43,44].

161 These studies from the literature show that the techniques using soluble fluorescent or
162 colorimetric dyes and optical equipment are efficient to visualize the local mass
163 transfer from a bubble with high resolution.

164 Taylor flows have been widely studied because of their performance in terms of both
165 heat and mass transfer, and their regularity, stability, and operability in a broad
166 operating domain. Dietrich et al. [24] first proposed the colorimetric technique (with
167 resorufin) to investigate mass-transfer in Taylor flow. Gray value maps were
168 converted to equivalent oxygen concentrations and a mass transfer model was
169 established, presenting the local characterization of gas-liquid mass transfer. In
170 addition, Yang et al. [45] considered the possibility of enhancing gas-liquid mass
171 transfer by this oxygen colorimetric reaction. Once the kinetic constant and diffusion
172 coefficients had been determined by colorimetric methods, the Hatta number and
173 enhancement factor were deduced, and there was no enhancement of the gas-liquid
174 mass transfer by this extremely fast oxygen reaction. As for the non-invasive and
175 instantaneous advantages, Yang et al. [46] further investigated the mass transfer
176 contribution of the bubble formation stage to the overall process. The liquid side mass
177 transfer coefficients at the moment immediately after the pinch-off stage were
178 obtained and found bigger than in the flowing stage, which demonstrated that the
179 contribution of the bubble formation stage to overall mass transfer was reasonably
180 large [47,48]. Furthermore, Yang et al. [49] studied the mass transfer intensification in

181 a square meandering channel. A comparison between methylene blue and resazurin
182 systems used in a helically-coiled tube showed better performance by the resazurin
183 system due to the larger difference of time constants in terms of oxidation and
184 reduction processes [50]. Krieger et al. [25] utilized leuco-indigo carmine to visualize
185 local mass transfer and concentration distributions, also investigating chemical
186 selectivity because of the existence of the anionic radical intermediate state. As
187 overlap of three different indigo carmine colors could result in multiple colors, the
188 isolation of each indigo carmine in the calibration procedure was achieved by an
189 imaging processing technique and UV/Vis spectrometry methods. In a more
190 complicated system for the combined mass transfer process and chemical reaction,
191 Kastens et al. [51] reported oxygen concentration fields and wake structures around a
192 Taylor bubble and compared the wake structures they observed with that obtained
193 using the PIV-LIF technique, while Butler et al. [52,53] first investigated the mass
194 transfer of a Taylor oxygen bubble by means of the PLIF-I technique and deduced the
195 oxygen concentration based on Stern-Volmer relations using the ruthenium
196 fluorescence intensities. By employing two groups of PLIF-I images, namely with O₂
197 and N₂ as the gas phase, they successfully corrected the light scattered by the gas
198 phase in O₂ images by means of N₂ images taken as references. They separated mass
199 transfer coefficients for film to bubble, film to slug and bubble cap to slug, with a
200 good precision.

201 In confined 2D Hele-Shaw cells, bubbles were largely flattened by narrow gaps and
202 confined to move with only two degrees of freedom. Bouche et al. [27] used two
203 fluorescent dyes (i.e., Fluorescein Sodium and Rhodamine WT) to remove the light
204 reflections caused by the gas interface, and Alm eras et al. [54] further analyzed the
205 fluctuations of concentration. Roudet et al. [55] estimated the total mass transfer rate
206 using the Sherwood number and Péclet number and compared the mass transfer rate
207 in the films with total mass transfer. Felis et al. [56,57] investigated the oxygen mass
208 transfer and local oxygen concentration around a rising bubble and compared the

209 equivalent oxygen concentration obtained by resazurin with that given by Cu/O₂
210 complexes.

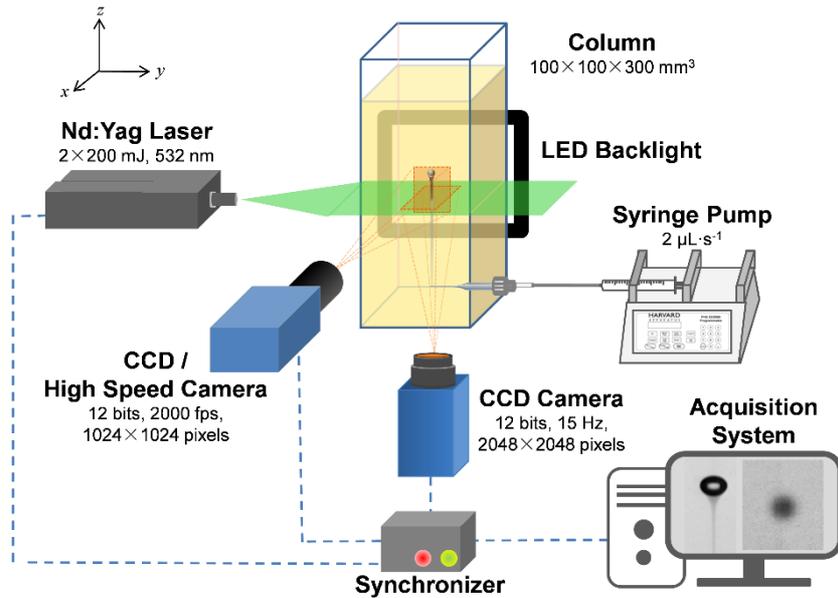
211 Several excellent reviews have been made focusing on fluorescence techniques [8,58],
212 however, they mixed the traditional PLIF techniques with inhibited PLIF techniques
213 even though they are theoretically different. For comparison purpose, the
214 experiments of the work presented here were conducted on the same experimental
215 setup to characterize the local mass transfer and the diffusion around a single air
216 bubble rising in water by using PLIF (with fluorescent resorufin), PLIF-I (with
217 ruthenium complex), and colorimetric techniques (with pink resorufin).

218 **2. MATERIALS AND METHOD**

219 **2.1 Experimental setup**

220 The experimental setup, depicted in Figure 2, was based on a dual camera system [20].
221 The glass column made of transparent PMMA (Poly (methyl methacrylate)) had
222 dimensions of 100×100×300 mm³. A stainless steel needle was inserted into the
223 column through a hole located 20 mm above the bottom of the column. The needle
224 was used for bubble injection, the bubble being pushed out of the tip of the needle
225 when the other side of the needle was connected to a syringe pump (Harvard
226 Apparatus PHD 22/2000 Programmable, USA).

227



228

229

Figure 2 Experimental setup

230

231 For laser-based techniques (i.e. PLIF and PLIF-I), a horizontal laser sheet was
 232 generated by a Nd:Yag laser (Dantec Dynamics Dualpower 200-15, France) and
 233 positioned about 100 mm above the needle. The high-intensity laser (2×200 mJ)
 234 emitted light with a wavelength of 532 nm, which enabled the dye to be excited with a
 235 frequency of 15 Hz.

236 When the bubble was rising in the column, the images of the fluorescence in the
 237 bubble wake were recorded by a Charge-Coupled Device (CCD) camera (Dantec
 238 Dynamics Flowsense CM). The camera was placed under the column and focused on
 239 the laser sheet. A microlens (105 mm f/8, Canon) with an extension tube (36 mm) was
 240 applied to increase the magnification of the investigated area. The length scale was
 241 calibrated with a ruler and image area was about $10 \times 10 \text{ mm}^2$ in real distances. A 570
 242 nm high-pass filter was placed in front of the lens to block the laser light. The CCD
 243 camera was synchronized with the laser thus 15 images were acquired every second in
 244 the forms of 12 bits and 2048×2048 pixels of resolution. For each bubble, the time
 245 was set to 0 when the first image containing the transferred mass was taken. A

246 high-speed camera (Photron SA3, Japan) was placed orthogonally to the first camera,
247 next to one lateral side of the column. It was used to capture the hydrodynamics of the
248 bubble, such as the velocity, shape, and trajectory of the bubble at a rate of 2000 fps.
249 The images were in 8 bit format with a resolution of 1024×1024 pixels. The length
250 scale was calibrated with a ruler and, in real distances, the image area was about
251 30×30 mm². The backlight was provided by a LED lighting board facing the camera.

252 The images from the two cameras were then transferred to the acquisition system,
253 which consisted of two professional software packages (DynamicStudio 4.0 and
254 Photron Fastcam Viewer 2). They can carry out simple image adjustment such as
255 color filtration, length calibration, etc. It should be noted that the above experimental
256 setup was designed to characterize the mass transfer in the bubble wake. To visualize
257 the mass transfer near the bubble interface, the laser sheet should be positioned
258 vertically and the positions of the two cameras may be switched.

259 All the experiments were carried out under the controlled conditions, i.e. with the
260 same needle (~ 0.7 mm of inner diameter) and same flow rate (2 μL·s⁻¹) for bubble
261 injection, at a temperature of 293.15 K and under atmospheric pressure (1 atm). The
262 good reproducibility of the bubble size (~ 1 mm), shape (ellipsoidal) and trajectory
263 (vertical) of the injected bubbles allowed us to assume that each of the individual
264 bubbles examined was identical. The details of the materials and properties of the
265 bubbles in the experiments are given in Table 1.

266

267 **Table 1 Composition of the fluids and properties of the bubbles for comparison**
268 **of three techniques**

	PLIF-I	PLIF	Colorimetric technique
Liquid-Gas	Deionized water + Air bubble		
Dye	Ruthenium Complex	Resazurin 100 mg·L ⁻¹ (+ NaOH 20 g·L ⁻¹)	

	75 mg·L ⁻¹	+ Glucose 20 g·L ⁻¹)
σ [mN·m ⁻¹]	72	75
ρ [kg·m ⁻³]	1000	1004
μ [mPa·s]	1.003	1.100
Diameter of bubble	1.25 ± 0.04 mm	
Aspect ratio of bubble	1.13 ± 0.02	
Velocity of bubble	295 ± 10 mm·s ⁻¹	
<i>Re</i>	368 ± 20	

269

270 2.2 Oxygen indicator (dye)

271 In the mass transfer visualization experiments, the oxygen concentration is indicated
 272 by different dissolved dyes: ruthenium complex for PLIF-I and resorufin for the
 273 colorimetric technique and the PLIF technique.

274 Ruthenium complex

275 In this study, a type of ruthenium complex (C₃₆H₂₄Cl₂N₆Ru·xH₂O, Sigma-Aldrich,
 276 USA, CAS: 207802-45-7) is adopted as a fluorescence dye. This dye is easy to
 277 dissolve in water and has a large separation between absorption and emission spectra
 278 (Stock shift) [30,34]. The fluorescence efficiency has been tested for different
 279 concentrations of dye [59] by comparing the difference of the gray levels when
 280 dissolved oxygen concentration varies from 0 mg·L⁻¹ to 3 mg·L⁻¹. It was found that
 281 when the concentration of the dye passes a critical value (~75 mg·L⁻¹), the
 282 fluorescence efficiency no longer increases. Thus, the concentration of dye was set at
 283 75 mg·L⁻¹ to guarantee fluorescence efficiency and economy. Before each experiment,
 284 the dye was added into the water and stirred together for 1 hour for well mixing.

285 **Resazurin (resorufin)**

286 Another dye used in this research is resazurin ($C_{12}H_6NNaO_4$, Sigma-Aldrich, USA,
287 CAS: 62758-13-8). Resazurin is a phenoxazin-3-one dye which has been widely
288 employed for testing bacterial or yeast contamination in biological fluids and milk,
289 and also identifying the semen quality by colorimetry since the 1950s [60].

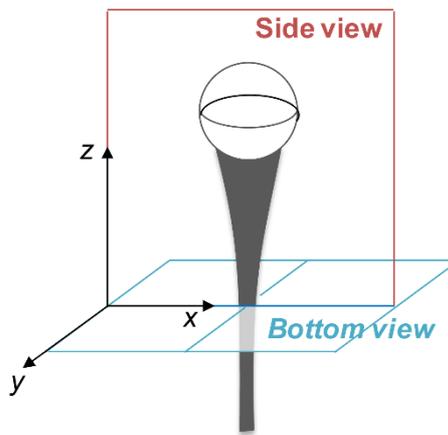
290 The dye solution is prepared with $100 \text{ mg}\cdot\text{L}^{-1}$ in the presence of sodium hydroxide
291 (VWR, USA, CAS: 1310-73-2) and D-glucose anhydrous (Fischer Scientific, USA,
292 CAS: 50-99-7), both diluted at $20 \text{ g}\cdot\text{L}^{-1}$. According to Anderson et al. [61], the reaction
293 kinetics could be generally assumed to be the first-order dependence on the
294 concentrations of glucose and resazurin, which means one mole of resorufin needs
295 one mole of resazurin to be reduced with one mole of glucose. In our cases, the
296 concentration of resazurin equals to $100 \text{ mg}\cdot\text{L}^{-1}$, namely $4.36\times 10^{-4} \text{ mol}\cdot\text{L}^{-1}$, thus the
297 minimum concentration of glucose needs to be $7.86\times 10^{-2} \text{ g}\cdot\text{L}^{-1}$. For the minimum
298 concentration of NaOH required, in fact, the first step of colorimetric reaction is the
299 enolization of glucose by $[\text{OH}^-]$, then the glucose enediolate reduces resazurin to
300 resorufin. However, unfortunately it is difficult to obtain the actual value of $[\text{OH}^-]$ by
301 simply considering the quantity of NaOH used to prepare the solution. Therefore, we
302 suggest the optimal employed concentrations of glucose and NaOH to be both 20
303 $\text{g}\cdot\text{L}^{-1}$, and the concentration of resazurin to be $0.1 \text{ g}\cdot\text{L}^{-1}$ to ensure the visualization and
304 the post-treatment of the colored fields with a balance between the reaction kinetic
305 rates and the requirement of adequate color intensity levels.

306 Under the applied concentration of the dye (with NaOH and Glucose), the
307 corresponding enhancement factor equals to 1.03, which means that the enhancement
308 effect of the colorimetric reaction on the mass transfer is negligible [45]. It should be
309 noted that the presence of the dye had a small impact on the fluid property. Especially
310 for cases with resazurin, the extra chemicals (NaOH and Glucose) slightly increased
311 the viscosity of the liquid compared with the result measured in pure water.

312 As mentioned in section 1.3, the reduced resazurin, namely resorufin (pink), is highly
313 fluorescent, which means it can play a role as an oxygen indicator. This feature makes it
314 can be also used as a dye for PLIF technique.

315 3. VISUALIZATION RESULTS

316 With the experimental configuration, the mass transferred in the wake of the bubble
317 could be visualized from two directions: side view and bottom view (see Figure 3).



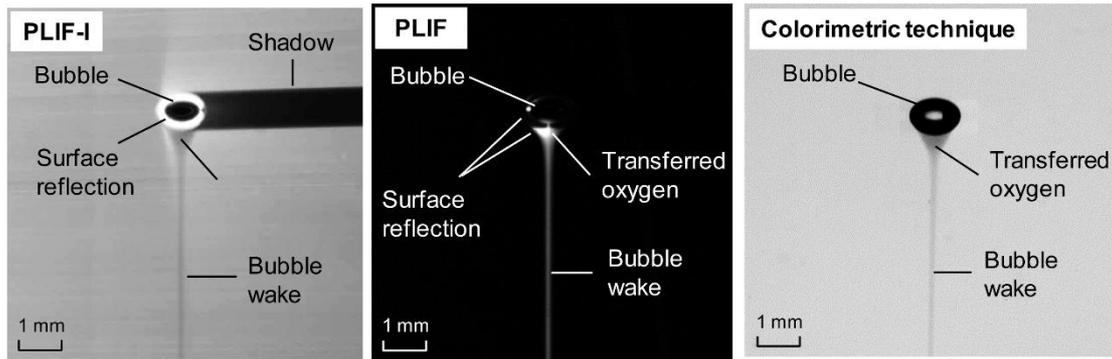
318

319 **Figure 3 Schematic description of side view and bottom view for the mass**
320 **transfer from the bubble**

321

322 An example of images recorded by the three different visualization techniques from
323 the side view are shown in Figure 4.

324



325

326 **Figure 4 Recorded images from different visualization techniques (Side view)**

327

328 On the image by PLIF-I and the colorimetric technique, the mass transfer zone is dark
 329 against a bright background, whereas the opposite is true for the images by PLIF.
 330 Some observations can be made:

331 (1) For the side view, the images using the laser techniques (PLIF or PLIF-I)
 332 suffer from a problem of reflection around the bubble surface.

333 (2) Regarding the bubble shadow on the PLIF-I image, a white ring appears
 334 around the bubble interface, making the bubble boundary difficult to recognize.
 335 Moreover, the reflection also has an impact on the mass transfer area near the bubble
 336 surface, especially for the part facing the laser (left part on the image). The effect
 337 diminishes along the laser direction but still cannot be neglected.

338 (3) For the PLIF image, the fluorescence effect is quite strong and the light
 339 around the bubble surface is relatively weaker than that in the bubble wake.
 340 Reflection occurs at the transfer interface, making the gray level recorded in this area
 341 much higher than the actually induced value.

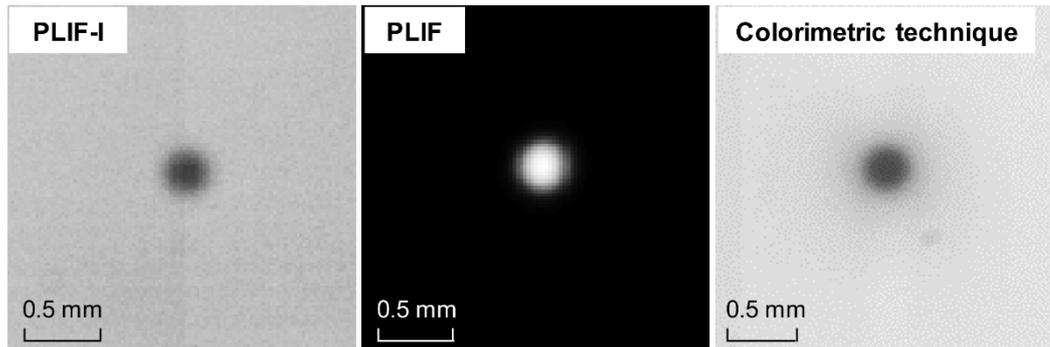
342 (4) On the other hand, the local mass transfer by the colorimetric technique is
 343 quite clear, without the reflection issue. This property makes the colorimetric method
 344 suitable for tracking the mass transfer even near the gas-liquid interface.

345 (5) Far from the bubble surface, all three methods can visualize the mass transfer

346 properly.

347 Due to the reflection problem of the laser techniques, there is a lack of information
348 near the bubble surface, so it is difficult to precisely quantify the mass transfer with
349 side view images. By contrast, the absence of a reflection problem means that the
350 colorimetric technique can be used to calculate the mass transfer coefficient [42].

351



352

353 **Figure 5 Images recorded by different visualization techniques (bottom view)**

354

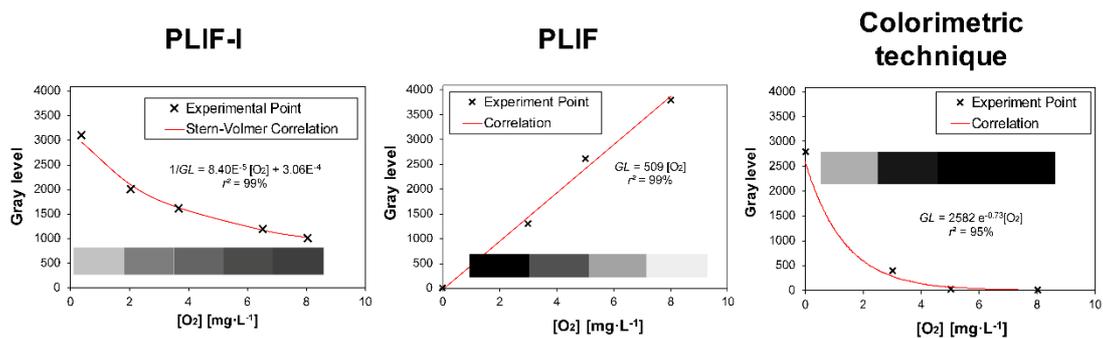
355 To avoid the reflection around the bubble caused by the laser flash, the camera is
356 placed under the column to record the mass transfer in one cross-section of the bubble
357 wake far from the passed bubble. From the bottom view, as shown in Figure 5, there
358 is good contrast between the mass transfer 'spot' and the background. The transferred
359 mass (dissolved oxygen) is concentrated at the spot center and becomes dispersed
360 near the boundary of the spot. Among the three images, the one by PLIF seems to
361 have the best quality, with the clear background (totally black) due to the different
362 color indicating mass transfer (white). The mass transfer can be quantified with
363 bottom view images for all three techniques.

364 **4. QUANTIFICATION OF MASS TRANSFER**

365 The relationship between the real oxygen concentration and the gray level recorded

366 on the images was established by the calibration process. Examples of the calibration
 367 results for the three techniques are shown in Figure 6. It can be seen that all three
 368 curves represent monotonic functions leading to a reasonable calculation of the
 369 oxygen concentration. The curve for PLIF is a straight line, showing that the gray
 370 level increase is directly proportional to higher oxygen concentration. For the other
 371 two curves (PLIF-I and colorimetric technique), the gray levels drop sharply,
 372 indicating that the dye is sensitive to low oxygen concentration ($[O_2] < 4 \text{ mg}\cdot\text{L}^{-1}$) but,
 373 when the oxygen concentration rises, the variation of the gray level is relatively small.
 374 For most of our cases, the oxygen concentration in the bubble wake was small within
 375 a reasonable calibration range.

376



377

378 **Figure 6 Examples of calibration result for three visualization techniques**
 379 **(PLIF-I, PLIF, and colorimetric technique).**

380

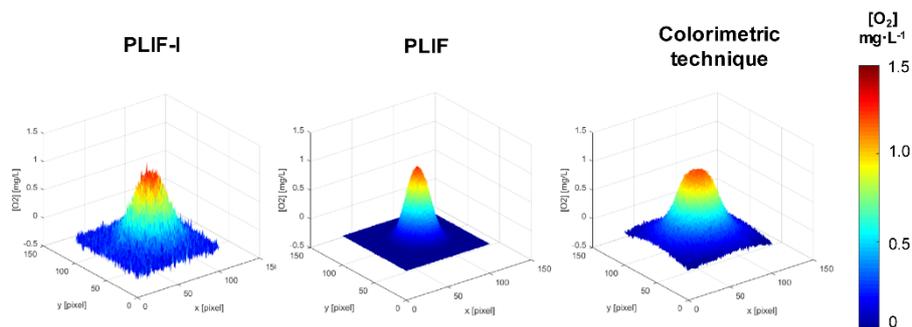
381 Regarding the sensitivity of calibration, when dissolved oxygen concentration varies
 382 from $0 \text{ mg}\cdot\text{L}^{-1}$ to $8 \text{ mg}\cdot\text{L}^{-1}$, the change of gray levels for PLIF-I, PLIF, and the
 383 colorimetric technique is 2000, 4000 and 2500 respectively. Under the configuration
 384 investigated here, resorufin is more sensitive to the oxygen concentration when used
 385 as a fluorescent dye than when used as a colorimetric dye and is also better than the
 386 ruthenium complex. This property leads to better contrast between the transferred
 387 mass and the background, which will be discussed in the following sections.

388 In addition, unlike the results obtained in the microchannel [46], the calibration curve

389 of the colorimetric technique is not linear. That is because the thickness of the image
390 area is no longer negligible when the camera takes images from the bottom of the
391 column and the calibration is actually implemented within a volume instead of a thin
392 plane. Thus, the Beer-Lambert absorption decreases the gray level, especially for the
393 solutions with high concentrations of the dye.

394 With the calibration curve, the gray level expresses the oxygen concentration, and the
395 concentration field of the dissolved oxygen in the bubble wake is plotted in Figure 7.
396 It can be seen that the background noise on the PLIF image is less significant than in
397 the other two images, which is consistent with the result for the side view in Figure 4.
398 The background noise on the PLIF-I image is relatively significant, so a larger
399 threshold is needed for PLIF-I to eliminate the impact of background noise.

400



401

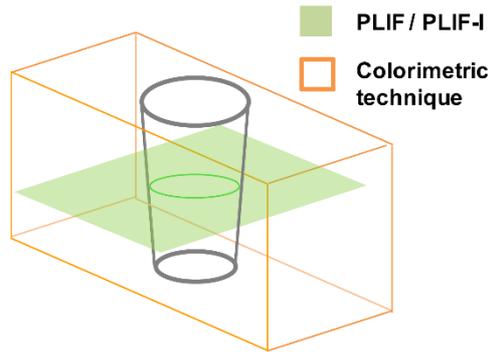
402 **Figure 7 Examples of the dissolved oxygen concentration field in the bubble**
403 **wake at the same moment (bottom view).**

404

405 Unlike PLIF and PLIF-I techniques, the colorimetric technique gives a background
406 concentration much higher than zero. It should be noted that the gray levels (mass
407 transfer) recorded on the images by these three techniques are not exactly the same
408 (see Figure 8). Since only a plane slice is lit by the laser sheet, the results by PLIF and
409 PLIF-I display exactly the mass transfer in the cross-section concerned. For the
410 colorimetric technique, the whole column is illuminated by the backlight and the

411 camera thus records the information not only in the focal plane but also in the front
412 and the rear ones due to the depth of field. Its results refer to a superposition of the
413 mass transfer in a series of continuous cross sections. This was probably the source of
414 the deviation on the mass transfer estimation.

415



416

417 **Figure 8 Image composition of mass transfer in bubble wake**

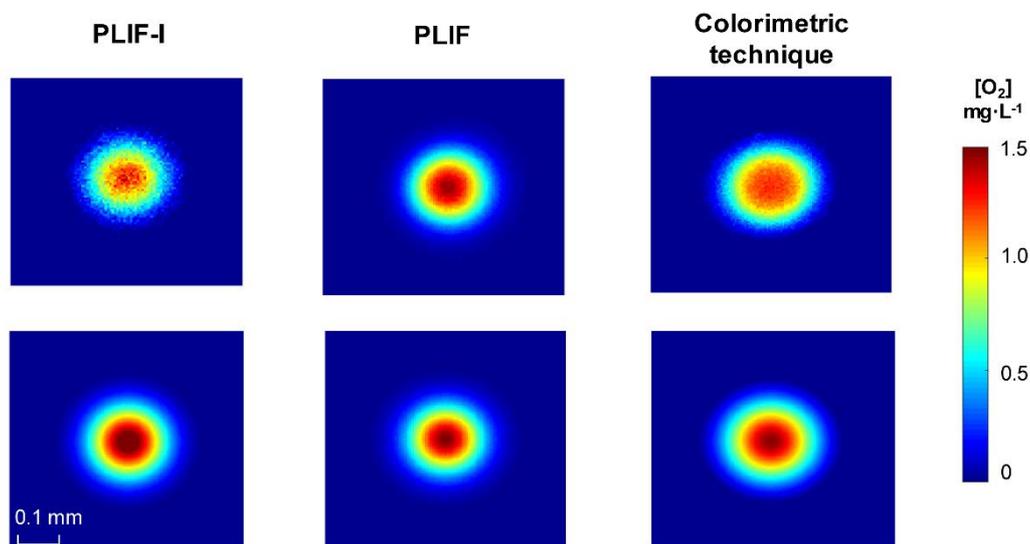
418

419 In order to obtain the transferred mass flux, the image quality should be improved. An
420 image processing procedure was implemented using Matlab (2017b) software. Firstly,
421 the background impact was removed by subtracting the raw image from the average
422 of 50 consecutive images just before the bubble went across the image plane. Then a
423 threshold is applied to remove the remaining noise at the image background. A central
424 region with resolution of 150×150 pixels was extracted which contained the mass
425 transfer spot (see Figure 9 top). Since the spot had a circular distribution of the
426 oxygen concentration that the oxygen diffused from the spot center to the surrounding,
427 the extracted images of the oxygen field were then fitted with the 2D Gaussian
428 diffusing model (see [35]), in which the oxygen concentration $[O_2]$ on the pixel (x, y)
429 was estimated by:

430
$$[O_2](x, y) = A \exp \frac{-(x-X)^2 + (y-Y)^2}{B} \quad (5)$$

431 where A, B are the parameters representing the properties of a Gaussian distribution,

432 and (X,Y) is the center of the spot. These four parameters were determined by
 433 minimizing the error between the measured value $[O_2]$ and the value from the fitting
 434 model (Eq. (5)). The fitting images are shown in Figure 9 bottom.



435

436 **Figure 9 Corrected images (top) and fitted images (bottom) of the concentration**
 437 **field of dissolved oxygen in the bubble wake.**

438

439 The fitting errors for all the three techniques are within 5%, validating the fitting
 440 Gaussian model. Summing the oxygen concentration in each pixel of the image area
 441 (see [62]) gives the fluxes before and after the fitting, as shown in Table 2. The PLIF
 442 technique has the best fitting result thanks to the good quality of its raw images.

443

444 **Table 2 Result of fluxes before and after fitting for the three techniques**

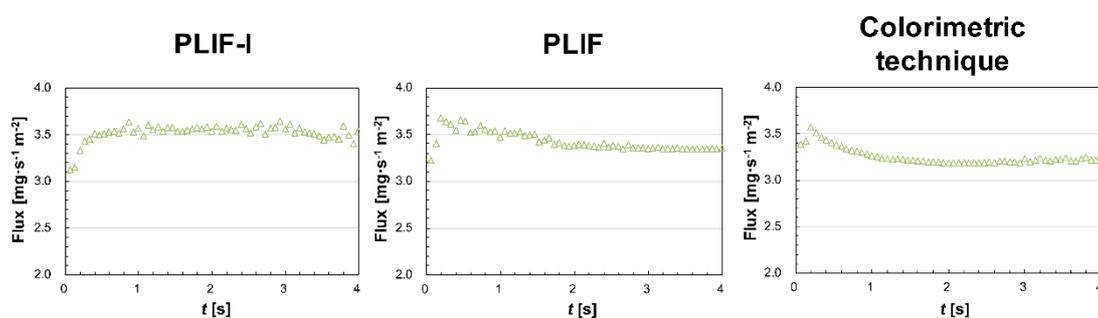
	Flux before fitting $[\text{mg}\cdot\text{s}^{-1}\cdot\text{m}^{-2}]$	Flux after fitting $[\text{mg}\cdot\text{s}^{-1}\cdot\text{m}^{-2}]$	Fitting error [%]
PLIF-I	3.53 ± 0.10	3.70	< 5%
PLIF	3.43 ± 0.03	3.45	< 1%
Colorimetric technique	3.35 ± 0.08	3.45	< 3%

445

446 The oxygen concentration field in the bubble wake was found for each recording
447 moment (every 1/15 second) and the temporal evolution of the mass flux thus
448 obtained is shown in Figure 10. For a small bubble with a linear trajectory, the mass
449 transfer in the bubble wake far from the passed bubble is dominated by diffusion
450 since the vertical convection induced by the bubble can be neglected. The mass
451 transfer takes place by two-dimensional diffusion, which means that the total flux on
452 the image should be conservative as a function of time.

453 It can be seen that, after the variation of the first several images due to the convection
454 of the bubble, the fluxes measured by the three techniques tend to be constant versus
455 time. For the PLIF and colorimetric cases, the fluxes are basically stable, but the total
456 flux is slightly smaller than that found by PLIF-I. This flux decrease may be related to
457 the presence of extra chemicals (NaOH and Glucose) impacting the liquid properties.

458

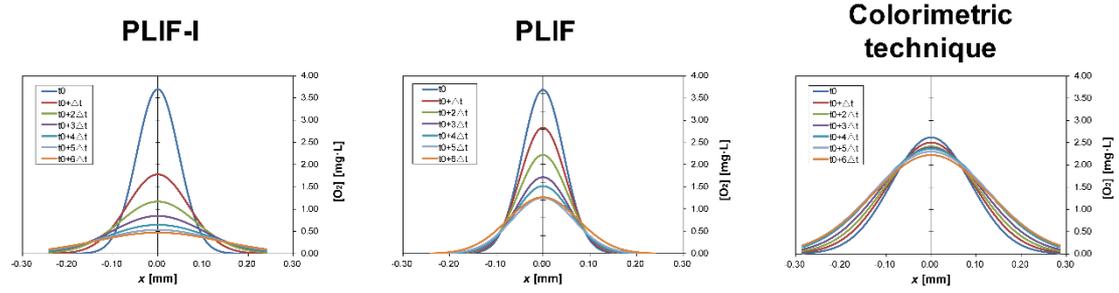


459

460 **Figure 10 Mass flux versus time measured by different techniques (PLIF-I,**
461 **PLIF, Colorimetric).**

462

463 Plotting the fitted oxygen concentration along a line passing the spot center reveals
464 that the distribution of the oxygen concentration has a Gaussian profile. Based on this
465 temporal evaluation of the oxygen profile in the bubble wake, it is possible to
466 characterize the diffusion process (see Figure 11). It can be seen that the dissolved
467 oxygen is concentrated at the spot center and then diffuses gradually to the
468 surroundings, making the concentration over the area more homogeneous.



469

470 **Figure 11** Temporal evolution of the O₂ concentration field at the cross section of
 471 the bubble wake (left to right, results by PLIF-I, PLIF, and colorimetric
 472 technique. $\Delta t = 0.67$ s)

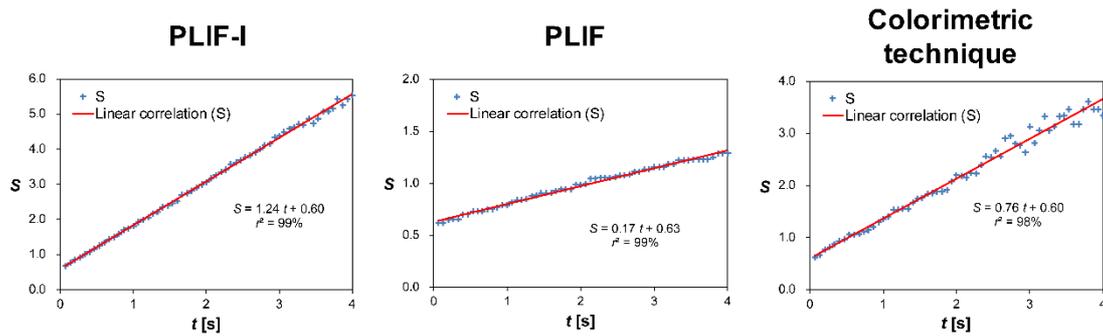
473

474 With the method introduced by Xu et al. [63], the area of the mass transfer spot
 475 expands as a function of time at a rate related to the diffusion coefficient:

476
$$S_{spot} = \pi R^2 = 2\pi Dt \tag{6}$$

477 The evolution of the spot area for the three techniques is plotted in Figure 12. For all
 478 three techniques, the curves of S_{spot} have good linearity versus time so the diffusion
 479 coefficient can be calculated from the slope of the curve (see Table 3).

480



481

482 **Figure 12** Evolution of the spot area as a function of time

483

484 **Table 3** Diffusion coefficient results for the three techniques

Slope of $S_{spot} - t$	r^2	D [$\times 10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$]
-------------------------	-------	---

PLIF-I	1.24	99%	1.97
PLIF	0.17	99%	0.27
Colorimetric	0.76	98%	1.20

485

486 The diffusion coefficients for the PLIF-I, PLIF and colorimetric techniques are 2×10^{-9}
487 $\text{m}^2 \cdot \text{s}^{-1}$, $0.27 \times 10^{-9} \text{m}^2 \cdot \text{s}^{-1}$, and $1.20 \times 10^{-9} \text{m}^2 \cdot \text{s}^{-1}$, respectively. In view of the technical
488 problem mentioned above, the estimates of the diffusion coefficient using PLIF with
489 resazurin and colorimetric technique are distorted. The reason is that these two
490 techniques are based on the tracking of a different molecule (resorufin) rather than
491 oxygen. The mathematical approach is based on the conservative flux, which may
492 also contribute to the error for the result by PLIF with resazurin and the colorimetric
493 technique. In contrast, the result by PLIF-I is consistent with the literature [30],
494 proving more accurate for the quantification of the diffusion.

495 With the fluxes obtained from the images, the mass transfer coefficient can be
496 estimated by dividing the flux J_{O_2} by the driving force. In our cases, the saturated
497 concentration $[O_2]^*$ is close to $9 \text{mg} \cdot \text{L}^{-1}$. For the PLIF-I technique, the bulk
498 concentration $[O_2]_0$, measured after the deoxygenation of the liquid, is $0.2 \text{mg} \cdot \text{L}^{-1}$.
499 For the techniques with resazurin (PLIF and colorimetric technique), the oxygen is
500 totally consumed or reacted before the experiment, so $[O_2]_0$ is assumed to be 0
501 $\text{mg} \cdot \text{L}^{-1}$.

502
$$k_L = \frac{J_{O_2}}{[O_2]^* - [O_2]_0} \quad (7)$$

503 The results for the mass transfer coefficients are given in Table 4. Regarding the
504 underestimated flux, the value determined by the PLIF and colorimetric technique
505 with resazurin is slightly smaller than that by PLIF-I. Nevertheless, all the values are
506 consistent with the literature [64] indicating that all three techniques can be used for
507 quantifying the mass transfer.

508

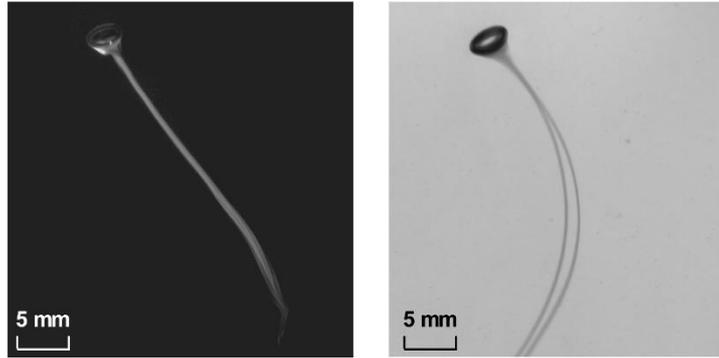
509

Table 4 Result of mass transfer coefficient by the three techniques

	k_L [$\times 10^{-4} \text{ m}\cdot\text{s}^{-1}$]
PLIF-I	4.01 ± 0.010
PLIF	3.81 ± 0.005
Colorimetric technique	3.61 ± 0.008

510 **5. DISCUSSION**

511 The advantages and limitations of each technique are summarized in Table 5. All three
512 techniques can be used for tracking the mass transfer to some extent. When resorufin
513 (from resazurin) is used as a pink dye, the colorimetric technique can even visualize
514 the mass transfer near the bubble surface. When it is used as a fluorescent dye, the
515 image by PLIF technique shows good contrast between the mass transfer area and the
516 pure black background. This property makes resorufin useful for the visualization of
517 the mass transfer in a more complex structure induced by bigger bubbles (Figure 13).
518 Since the resorufin has both pink color and fluorescence property, we can integrate
519 the colorimetric and PLIF techniques by using the current experimental set-up (Figure
520 2). Based on the dual camera system, the mass transfer in both directions (side view
521 and bottom view) can be visualized simultaneously. This would be a promising
522 technique for the following study of local mass transfer characterization.



523

524 **Figure 13 Visualization result with resorufin as a dye for bigger bubble ($D_{eq} \sim 3$**
525 **mm). Left: image by PLIF; Right: image by colorimetric technique.**

526

527 For the quantification of the mass transfer, the PLIF-I technique can give a reasonable
528 result for the diffusion coefficient and mass transfer coefficient, while the other two
529 techniques with resazurin do not correctly characterize the diffusion process due to
530 the reaction mechanism. Another limitation for the PLIF and colorimetric techniques
531 is that the oxygen indicator resazurin needs to be used together with extra chemicals
532 (NaOH and glucose), which will have an impact on the fluid under investigation. The
533 mass transfer coefficients measured by PLIF and coulometric techniques (with NaOH
534 and glucose) remain in a reasonable range ($3.6\text{-}3.8 \times 10^{-4} \text{ m}\cdot\text{s}^{-1}$), regardless a slight
535 decrease compared to the PLIF-I technique (without NaOH and glucose). Thus, for
536 our cases as oxygen mass transfer in deionized water, it has been proven that NaOH
537 and glucose have no evident effect on mass transfer performance. However, it should
538 be very careful when implement the colorimetric technique involving specific
539 solute/solvent. The reaction possibility between the colorimetric solution and
540 investigated solution should be carefully taken into account, which means NaOH and
541 glucose, even resorufin could act as contaminants which affect mass transfer.

542

543
544

Table 5 Advantages and limitations of three techniques (PLIF-I, PLIF, colorimetric technique)

	Advantage <input checked="" type="checkbox"/> or limitation <input checked="" type="checkbox"/>
PLIF-I	<ul style="list-style-type: none"><input checked="" type="checkbox"/> Tracks mass transfer in the wake far from the surface<input checked="" type="checkbox"/> Accurate quantification of flow flux<input checked="" type="checkbox"/> Characterizes the diffusion<input checked="" type="checkbox"/> Reflection at the bubble surface<input checked="" type="checkbox"/> Significant background noise<input checked="" type="checkbox"/> Shadow of the bubble
PLIF	<ul style="list-style-type: none"><input checked="" type="checkbox"/> Tracks mass transfer in the wake far from the surface<input checked="" type="checkbox"/> Less background noises<input checked="" type="checkbox"/> Cannot characterize the diffusion of oxygen<input checked="" type="checkbox"/> Reflection at the bubble surface<input checked="" type="checkbox"/> Extra chemicals
Colorimetric technique	<ul style="list-style-type: none"><input checked="" type="checkbox"/> Tracks mass transfer even near the bubble<input checked="" type="checkbox"/> No reflection near the bubble<input checked="" type="checkbox"/> Cannot characterize the diffusion of oxygen<input checked="" type="checkbox"/> Complex composition of the recorded image<input checked="" type="checkbox"/> Extra chemicals

545

546 CONCLUSIONS

547 Three different visualization techniques PLIF-I (with ruthenium complex), (PLIF
548 (with fluorescent resorufin), and a colorimetric technique (with pink resorufin)) have
549 been compared, with particular focus on their application to characterize the mass
550 transfer around a single bubble rising in water. The PLIF and colorimetric technique
551 with resazurin are both effective for tracking the local mass transfer. However, under
552 our investigation conditions, there was a small distortion in the quantification of the

553 transferred oxygen fluxes by these two techniques, due to the presence of the extra
554 chemicals (NaOH and Glucose). Moreover, they cannot be used to quantify the oxygen
555 diffusive process as oxygen is consumed. Actually, these two techniques are based on
556 the tracking of a different molecule (resorufin) instead of the oxygen molecule.
557 Concerning the colorimetric technique, it is also less precise because its image contains
558 the mass transfer not only on the target cross-section but also on those below and above
559 it. Although the processing to remove the noise in the image background is relatively
560 rigorous, PLIF-I is more accurate for the quantification of the diffusion coefficient and
561 mass transfer coefficient.

562

563 **NOMENCLATURE**

Latin Symbols

C_{dye}	concentration of the dye
D	diffusion coefficient
F	fluorescence level
I	laser intensity
I_0	fluorescence intensities without quencher
I_Q	fluorescence intensities with quencher
J	Flux density
k_L	liquid side mass transfer coefficient
K_{SV}	Stern-Volmer constant
Q	concentration of the quencher molecule
r^2	determination coefficient
S_{spot}	area of mass transfer spot
t	time

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