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Local investigations on the gas-liquid mass transfer around Taylor bubbles flowing in a meandering millimetric square channel

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ABSTRACT

Gas-liquid mass transfer around Taylor bubbles moving in a meandering millimetric square channel was locally visualized and characterized in the present study. For that, the colorimetric technique proposed by Dietrich et al. (2013) was implemented. With this technique, the evolution of equivalent oxygen concentration fields in the liquid slugs passing through one and several bends was firstly described. In particular, it was observed how the flow structure (recirculation zones) inside the liquid slugs were twisted and split by the periodic bends (centrifugal effect), until reaching, after several bends, a uniform O 2 concentration inside the liquid slugs. The influence of the “turning point”, joining two “straight” sections of meandering channel was also highlighted: a slowing down of the gas-liquid mass transfer was clearly shown. Volumetric mass transfer coefficients were determined at last by fitting the experimental axial profiles of averaged oxygen concentrations in the liquid slugs (before the turning point) with the ones predicted by a classical plug-flow model.

1. Introduction

Since the last decade, process intensification opens up interesting perspectives for the fine chemical and pharmaceutical industries (Stankiewicz and Moulijn, 2000; Tochon et al., 2010; Commenge and Falk, 2014; Gourdon et al., 2015). A wide range of chemistries has been investigated through the implementation of flow reactors, showing the advantages and the improvements related to such technologies, with regards to quality, safety, competitiveness and eco-impact (Pelleter and Renaud, 2009; Nieves-Remacha et al., 2013; Darvas et al., 2014; Elgue et al., 2015). In this way, various equipments have been developed, benefiting from miniaturization techniques and micro (or milli) fluidics. Most of them are the devices where chemical reactions are performed in narrow channels involving a very high surface area to volume ratio and thus providing very efficient rates of mass and heat transfer.

Among all the process intensification technologies, heat exchanger reactors (HEX reactors), combining a reactor and a heat exchanger in only one unit, are particularly performing in terms of versatility and modularity features, but also of heat and mass transfer capabilities (Anxionnaz et al., 2008; Anxionnaz, 2009; Théron et al., 2014). They now become standard tools for process development and continuous production. In order to combine intensified heat and mass transfers, high residence time and compactness in such HEX reactors, the use of 2D-structured meandering (or tortuous or serpentine or wavy) channel structures constitutes an interesting solution. Indeed, contrary to the case in a straight channel, the occurrence of curvatures (or bends) generates flow instabilities and secondary flows (Dean vortices) in fluid motion (Dean, 1928), where the mixing between the core fluid and near-wall fluid can be improved; the boundary layers are also disrupted and thinned, thus affecting positively heat and mass

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transfer phenomena. Most of the existed literature dealing with such type of meandering channels focus on the study of single liquid phase flows, reactive or not (Xiong and Chung, 2007; Anxionnaz-Minvielle et al., 2013; Karale et al., 2013; Dai et al., 2015a). Few attention has been surprisingly paid on the implementation of gas-liquid systems in meandering channels, whereas: (i) the gas-liquid systems occupy a key place in scientific research and industrial application fields (hydrogenations, sensitized photo-oxygenations, fluorinations, biochemical reactions); (ii) there still remains many fundamental questions (coupling between transport phenomena and kinetics); (iii) studies in straight micro- or millimetric channels are the object of an abundant literature (van Baten and Krishna, 2004; Roudet et al., 2011; Yao et al., 2014; Haghnegahdar et al., 2016; Haase et al., 2016; Butler et al., 2016). In such two-phase reactive systems, the knowledge of mass transfer between gas and liquid phases is then extremely important; as managing the chemical kinetics, it can become the limiting step controlling the chemical reactions in terms of conversion and selectivity.

While the influence of the occurrence of bends in millimetric channels on the gas-liquid hydrodynamics (i.e. flow regime, mixing efficiency, interfacial area) has been highlighted by several authors (Günther et al., 2004; Fries and von Rohr, 2009; Dessimoz et al., 2010), rare are at present the studies quantifying how curvatures affect the gas-liquid mass transfer (Roudet et al., 2011; Kuhn and Jensen, 2012). Roudet et al. (2011) showed that, when compared to a straight channel of identical compactness and sectional-area, the meandering channel induced: (i) a delay in the transition from Taylor to annular-slug regimes; (ii) a rise of 10–20% in bubble lengths while conserving almost identical slug lengths; (iii) higher deformations of bubble nose and rear due to centrifugal forces (bends). They also observed that, for the Taylor flow regime, $k_\alpha$ increased coherently when increasing superficial gas velocity $U_g$, and that the meandering geometry had a small influence. On the contrary, this effect was found no more negligible for the slug-annular flow regime. At last, they demonstrated that, at identical compactness, the meandering channel was found to be the most competitive. As these authors used a global experimental method (measurements of concentrations in dissolved oxygen along the channel length by oxygen microsensors), the full understanding of the mechanism controlling the gas-liquid mass transfer in a meandering channel could not be achieved, and in particular the contribution of the curvatures. In order to fill these gaps, local measurements, such as concentration fields of the transferred gas phase around bubbles in the liquid slugs and films, are required. However, to our best knowledge, there exists no research on the local visualization and characterization of gas-liquid mass transfer in meandering channels.

With regard to this context, this paper aims at locally studying the mass transfer around Taylor bubbles flowing in a meandering millimetric square channel, identical in elementary shape to the one used by Roudet et al. (2011) but longer. For this purpose, the colorimetric technique proposed by Dietrich et al. (2013) will be used, consisting in implementing a redox reaction involving an oxygen-sensitive dye (resazurin). Its relevancy for locally visualizing and characterizing gas-liquid mass transfer at different scales has been outlined by Kherbeche et al. (2013) and Yang et al. (2016b). In addition, the conditions required to avoid any enhancement of the gas-liquid mass transfer by this reaction have been recently identified (Yang et al., 2016a); for that, the Hatta number and enhancement factor have been quantified from the determination of the characteristic time of the redox reaction kinetic and of the diffusion coefficients of the dye and of oxygen in the reactive medium. In comparison with PLIF technique (e.g. Butler et al., 2016), the colorimetric method is more convenient and user friendly as it does not need any laser excitation or inserting a physical sensor.

This paper will be structured as follows. The hydrodynamics of gas-liquid system (air/reactive medium) will be firstly characterized, in particular in terms of the gas-liquid flow map, overall gas hold-up, bubble length, shape and velocity. The colorimetric technique will be implemented in a second time, only in the case of the Taylor regime. The fields of equivalent oxygen concentration in the liquid slugs will be presented and discussed, as well as their changes according to the locations in the bend and all along the channel length; the influence of gas and liquid flow rates on these

**Nomenclature**

- **Greek letters**
  - $\mu_L$: dynamic viscosity of the dye solution, Pa s
  - $\rho_L$: density of the dye solution, kg m$^{-3}$
  - $\sigma_L$: surface tension of the dye solution, N m$^{-1}$

- **Dimensionless numbers**
  - $Ca$: Capillary number, $=\mu_L U_B/\sigma_L$, dimensionless
  - $Ce$: Centrifugal number, $=\rho_L U_B^2 \sqrt{l}/\sigma_L \times r_0$, dimensionless
  - $Re$: Reynold number, $=\rho_L U_B^2 \sqrt{l}/\sigma_L \times r_0$, dimensionless
  - $We$: Weber number, $=\mu_L U_B^2 \sqrt{l}/\sigma_L$, dimensionless

- **Subscripts**
  - B: bubble
  - bt: before turning point
  - d: drift velocity
  - G: gas phase
  - L: liquid phase
  - s: liquid slug
  - UC: unit cell
fields will be also outlined. From them, the evolution of averaged O₂ concentration with the axial location in the channel will be determined, enabling the overall volumetric mass transfer coefficients to be deduced using the classical plug-flow model. At last, these data will make possible the calculation of liquid-side mass transfer coefficients, based on an accurate determination of interfacial areas.

2. Materials and methods

2.1. General principles of the colorimetric technique

The colorimetric technique, proposed by Dietrich et al. (2013), is based on an oxygen-sensitive dye, named resazurin, that could react with oxygen in the presence of sodium hydroxide (NaOH) and glucose solution. In the reduced form, named dihydroresorufin, the solution is colorless, while in presence of oxygen, the oxidized form, named resorufin, is characterized by an intense pink color. As shown by previous works (Dietrich et al., 2013; Kherbeche et al., 2013; Yang et al., 2016b), the main advantage of this technique is the extent of the oxidation reaction which corresponds to the amount of transferred (or dissolved) oxygen, and to a concentration of resazurin fixed at 0.1 g L⁻¹. For each system, an optimal factor E was determined, enabling the overall volumetric mass transfer to be stable (Fig. 1b).

2.2. Fluid properties

All the experiments were performed at 293.15 K and atmospheric pressure. For the hydrodynamic experiments, the liquid phase was dye-free, composed only of D-glucose anhydrous (Fisher Scientific, CAS 50-99-7) and sodium hydroxide (VWR, CAS 1310-73-2), both diluted at 20 g L⁻¹ in deionized water (conductivity: 51.2 μS m⁻¹). For the mass transfer experiments, resazurin (Sigma Aldrich, CAS 62758-13-8, molecular mass: 229.19 g mol⁻¹) which concentration was fixed at 0.1 g L⁻¹, was added to the previous solution. The density ρ₁, dynamic viscosity μ₁, and static surface tension σ₁ were measured by means of a pycnometer (ρ₁ ± 0.2 kg m⁻³), a RM180 Rheomat Rheometric Scientific® viscometer (μ₁ ± 10⁻³ mPa s), and a Digidrop GBX® or Krüss tensiometer (σ₁ ± 0.5 mN m⁻¹) respectively. The oxygen saturation concentration C₀ was measured by implementing the Winkler technique (Winkler, 1888) and by means of optical oxygen probes (Hach-Lange, and Schafer Scientific, model 5850E) at 2000 frames per second. Image resolutions were 164 μm per pixel and 20 μm per pixel for the hydrodynamics and mass transfer experiments, respectively. The regions of interest were 168.1 ± 0.2 mm² and 20.1 ± 0.1 mm², respectively. For the mass transfer experiments, the gas-liquid flow was observed at 7 different axial distances X from the gas injection, namely: X = 0.02 m, 0.07 m, 0.12 m, 0.17 m, 0.22 m, 0.32 m and 0.42 m, as depicted in Fig. 1(b).

2.3. Description of the experimental set-up

The experimental set-up was depicted in Fig. 1(a). It consisted of a meandering channel composed of a series of elementary units (named bends) (Fig. 1b). The channel, horizontally placed, had a square cross section A = l² where l = 2 mm, and the elementary bend, which geometry was identical to the one used in Roudet et al. (2011), was characterized by a radius of curvature r of 1.3 mm and a straight length between two bends of 6.94 mm (see Fig. 1c). Every about 15 bends, there was a big bend, named “turning point”, enabling to connect the two “straight” channel sections, which leads to change the flow direction in 180° and in order to have a long channel (the total developed straight length of the meandering channel L = 1.37 m), while keeping a compact device. The channel was curved in a PolyMethyl MethAcrylate (PMMA) plate (3 cm thick), and roofed over in a watertight manner by another plate (3 cm thick). The gas phase (air) was injected via a hole of diameter 1.6 mm, pierced perpendicularly to the channel; the gas injection was spaced from about eight bends from the liquid one so as to enable the liquid flow to be stable (Fig. 1b). Gas (air) was delivered from the supply pipeline regulated by a mass flow controller (Brooks® model 5890E). Before entering in the main channel, the gas flowed through a winding of a 2 m long capillary of 750 μm internal diameter, aiming at to increase the pressure drop in order to create a stable gas injection in the liquid flow. Liquid phase was pumped by a Cantoni™ jet pump with the flow rate regulated by a mass flow control system (Micro motion®). The volumetric gas flow Qg rates ranged from 0.2 to 9 L h⁻¹ and liquid flow rates Ql from 1 to 2 L h⁻¹. The corresponding superficial gas velocities jg and liquid velocities jL were: 0.014 m s⁻¹ ≤ jg ≤ Qg/r² ≤ 0.625 m s⁻¹ and 0.069 m s⁻¹ ≤ jL = Ql/2m² ≤ 0.139 m s⁻¹.

2.4. Image acquisition system

The shadowgraph method was applied to investigate the gas-liquid hydrodynamics and mass transfer inside the meandering channel. The channel was lighted by a LitePad HO LED backlight (Rosco®) and the images of the bubbles flowing along the channel were recorded by a monochromatic high-speed CMOS camera (Photron® SA3) at 2000 frames per second. Image resolutions were 164 μm per pixel and 20 μm per pixel for the hydrodynamics and mass transfer experiments, respectively. The regions of interest were 168.1 ± 0.2 mm² and 20.1 ± 0.1 mm², respectively. For the mass transfer experiments, the gas-liquid flow was observed at 7 different axial distances X from the gas injection, namely: X = 0.02 m, 0.07 m, 0.12 m, 0.17 m, 0.22 m, 0.32 m and 0.42 m, as depicted in Fig. 1(b).

2.5. Image processing

2.5.1. Hydrodynamics of the gas-liquid flow

A typical raw image of the gas-liquid flow all along the channel length (1.37 m) is shown in Fig. 2(a). In order to extract the hydrodynamics parameters (gas hold-up, bubble length and...
velocity, liquid slug length etc.) from the raw image, a background image when the channel was filled with the liquid phase was needed, shown in Fig. 2(b). Then, an image post-treatment algorithm was implemented on the software Matlab (R2015a). In this algorithm, the background division approach was used to eliminate the effect of the background, instead of the traditional method consisting in subtracting the background image to each image; this was found to be more suitable to avoid illumination fluctuations and to diminish the confusion between background and foreground in pixels (Izquierdo-Guerra and García-Reyes, 2010). The image processing made possible the accurate detection of all the bubbles and liquid slugs inside the channel, as displayed in Fig. 2 (c) and (d) respectively, as below:

- For the gas bubbles image (Fig. 2c), it was generated by the segmentation methods in image processing and analysis in Matlab. Namely, after dividing the raw image (Fig. 2a) with the background image when the channel was filled with the liquid phase (Fig. 2b), one could obtain the image where only the bubbles left (not shown here). Then the image segmentation methods were implemented to identify all the bubbles on the image (Fig. 2c).
- For the liquid slugs image (Fig. 2d), it was generated by subtracting the image where only the channel left on the image with Fig. 2(c). The image where only the channel left on the image was obtained by dividing the background image when the channel was filled with the air phase (not shown here), with the one filled with the liquid phase (Fig. 2b).

![Fig. 1. (a) Schematic diagram of the experimental set-up. (b) Picture of the meandering channel in the horizontal xy-plan defined in (a). X corresponds to the location (curvilinear coordinate) in the channel from the gas inlet. X = 0 is the location where the two phases enter in contact. The channel has a cross-sectional area \( A = 2 \times 2 \text{ mm}^2 \), and a total developed straight length \( L \) of 1.37 m. Black circles represent the axial positions where the equivalent oxygen concentration fields were measured. The sections of the channel corresponding to the so-called "turning points" are outlined by blue rectangles. The arrows indicate the flow direction. (c) Schematic of elementary units (bends) of the meandering channel.

![Fig. 2. Sequence of image processing. (a) Typical raw image of gas-liquid flows. (b) Raw image of the channel filled with liquid phase. (c) Detection of the bubbles (binary image). (d) Detection of the liquid slugs (binary image). Operating condition: \( \dot{j}_L = 0.139 \text{ m s}^{-1}, \dot{j}_G = 0.069 \text{ m s}^{-1} \).]
Once detected, the areas of the channel in the xy-plane (noted as $A_C$), all the 2D bubbles ($A_B$) and all the 2D liquid slugs ($A_L$) appearing along the whole length of the channel, could be obtained from the final binary images. Thus, the gas hold-up could be calculated as according to:

$$
\varepsilon = \frac{A_B}{A_C} = \frac{A_B}{\sum A_{B_i} \sum A_{L_i}}
$$

where $i= 1\ldots N$, $N$ is the number of bubbles/liquid slugs. It was observed that the difference between the summation of the bubble areas and liquid slug areas ($\sum A_{B_i}$ + $\sum A_{L_i}$) and the channel area $A_C$ is smaller than 2%, which could be attributed due to the area occupied by the liquid film.

Bubbles characteristics (i.e. bubble length $L_B$, liquid slug length $L_S$, and bubble velocity $U_B$) were determined only in Taylor flow regime (defined in Section 3.1.1). The lengths of bubble and of liquid slug (noted $L_B$ and $L_S$, respectively, illustrated in Fig. 3a) were extracted from the major axis length of the bubbles which was an automatic procedure based on a pre-defined function. The averaged values of $L_B$ and $L_S$ were calculated by considering 20 bubbles on one image and 20 images were used. The bubble velocity $U_B$ was deduced from the averaged times required by bubbles to flow along two bend periods. For a given operation condition, the standard deviations of the bubble characteristics ($L_B$, $L_S$ and $U_B$) of all the bubbles inside the channel have found to be smaller than 6% of the averaged value.

### 2.5.2. Gas-liquid mass transfer

The gas-liquid mass transfer characteristics were determined by using the same image post-treatment algorithm (implemented on the software Matlab R2015a), than the one used by Dietrich et al. (2013). It consisted of two main steps: (i) the determination of the calibration curve, and (ii) the image processing which enabled to transform pixel-by-pixel the acquired grey-level images into the equivalent oxygen concentration fields. Note that the term “equivalent” was used because in reality the oxygen concentration into the equivalent oxygen concentration fields. Note that the term “equivalent” was used because in reality the oxygen concentration was unfortunately measured in the liquid film zone close to the walls.

For the calibration process, different concentrations of resazurin solution were prepared: 0, 0.025 g L$^{-1}$, 0.05 g L$^{-1}$, 0.075 g L$^{-1}$ and 0.1 g L$^{-1}$. An example of calibration curve (at $X = 0.02$ m) is depicted in Fig. 3(b), showing the linearity between the grey levels and the equivalent amounts of oxygen transferred per unit of liquid volume. It is important to mention that the calibration process was re-implemented each time when the observation position (camera, light source, reactor) was changed. After applying the calibration curve in the image processing, the equivalent concentration field of oxygen could be obtained, and the oxygen transferred in the liquid slugs could be directly visualized, depicted in Fig. 3(c). Note that under the milli-scale conditions investigated, any oxygen concentration could be unfortunately measured in the liquid film zone close to the walls.

### 3. Results and discussion

#### 3.1. Gas-liquid hydrodynamics

**3.1.1. Flow regimes, overall gas hold-up and bubble velocity**

Depending on the gas and liquid flow rates, different spatial distributions of the two phases were observed and various flow regimes were distinguished. The cartography of the gas-liquid flow regimes is displayed in Fig. 4. Under the present operating conditions (0.014 m s$^{-1}$ ≤ $j_G$ ≤ 0.625 m s$^{-1}$; 0.069 m s$^{-1}$ ≤ $j_L$ ≤ 0.139 m s$^{-1}$), three regimes can be identified:

- The Taylor flow regime occurs at low and moderate gas flow rates, for which the lengths of bubbles and liquid slugs are constant along the meandering channel and regularly spaced. By visualization, one can observe that (i) the bubble length $L_B$ increases as the gas flow rate increases under a given liquid flow rate; (ii) $L_B$ decreases as the liquid flow rate increases under a given gas flow rate; (iii) the liquid slug length (separating two consecutive bubbles) significantly decreases as gas flow rate increases, whereas it slightly varies as liquid flow rate changes under a given gas flow rate.

- The second regime, called slug-annular flow regime, occurs at relatively high gas flow rates. Unstable slug flows appear, characterized by the occurrence of long bubbles. Two consecutive bubbles can be so close to each other that gas-liquid flow exhibits significant randomness. The rupture of an extremely long bubble could be seen under some conditions.

- The last regime is the annular flow and occurs at high gas flow rates. A continuous gas core in the channel center seems to be form.

The evolution of gas hold-up $\varepsilon$ (measured over the whole length of the channel, see Eq. 1) is plotted in Fig. 5 as a function of the volumetric quality (fraction) of gas $\beta = (Q_G/(Q_C + Q_L))$. A deviation
between $e$ and $\beta$ could be observed leading to values of $e$ smaller than $\beta$. It can be explained by writing the mass balance of the gas phase, as:

$$A \cdot (j_c + j_l) \cdot \beta = A \cdot U_B \cdot e$$

Due to the fact that the bubble travels slightly faster than predicted by the superficial two-phase velocity, $e$ should be logically smaller than $\beta$ to verify this mass balance. The degree of deviation depends on the flow regime:

- relatively large for the Taylor flow (ranging from 1.17% to 15.95% with a mean deviation of 8.86%), confirming the occurrence of bubble slip velocity;
- moderate for the slug-annular flow, varying from 0.49% to 9.03% with a mean deviation of 4.46%;
- small for the annular flow, varying from 0.45% to 0.88% with a mean deviation of 0.67%.

Meanwhile, the Taylor flow regime is characterized by relatively low and moderate volumetric quality of gas $\beta$ (between 0.17 and 0.67). Higher $\beta$ lead to non-uniform bubble size distribution and irregular gas-liquid flow, like slug-annular flow, until reaching the annular flow regime for which $\beta$ is up to 0.90.

The evolution of bubble velocity $U_B$ as a function of the total superficial velocity $j$ (equal to $j_c + j_l$) is plotted in Fig. 5(b). It can be observed that the values of $U_B$ are always larger than those of $j_c$. This is due to the interaction between the bubbles and the liquid, resulting in a higher velocity for the bubble phase.

Fig. 4. Cartography of gas-liquid flow regimes. The flow regimes in the frame with solid lines correspond to the Taylor regime, with dashed lines the slug-annular regime, and with dash dotted lines the annular regime. The numbers in the brackets correspond to ($Ca$, $We$, $Ce$).

Fig. 5. (a) Evolution of gas hold-up as a function of the volumetric quality of gas in the meandering channel. The points inside the red dashed ellipse belong to Taylor flow. (b) Bubble velocity versus two-phase total superficial velocity. The solid line represents $U_B = j_c + j_l$, the dashed line $U_B = 1.1(j_c + j_l)$, the dash dotted line $U_B = 1.45(j_c + j_l)$. 
which considers the effect of the local relative velocity. In our case, the distribution parameter, which takes into account the effect of non-uniform flow and concentration profiles, and $U_d$ the drift velocity, considers the effect of the local relative velocity. In our case, the distribution parameter $C_o$ equals to 1.27, which is consistent with the values encountered in the literature that vary between 0.85 and 1.38 (Tsoligkas et al., 2007); the drift velocity $U_d$ equals to $-0.0043 \text{ m s}^{-1}$.

3.1.2. Bubble length and slug length

For the operating conditions corresponding to the Taylor regime ($0.014 \text{ m s}^{-1} \leq \overline{u}_c \leq 0.208 \text{ m s}^{-1}$ and $0.069 \text{ m s}^{-1} \leq \overline{u}_L \leq 0.139 \text{ m s}^{-1}$), the dimensionless Bond number $Bo = \rho_L \cdot g \cdot F / \sigma$ equals to 0.52, the Weber numbers $We = \rho_L \cdot \overline{u}_L^2 \cdot l / \sigma$, range from 0.29 to 5.32, the capillary numbers $Ca = \rho_L \cdot \overline{u}_L \cdot R / \sigma$, from 0.0015 to 0.0066 and the Reynolds numbers $Re = \rho_L \cdot \overline{u}_L \cdot l / \mu_L$ from 188 to 806. For meandering channels, one should also consider the dimensionless number comparing the centrifugal forces and capillary effects, defined as $Ce = \rho_L \cdot \overline{u}_L^2 \cdot F / \sigma \cdot r$; it ranges from 0.39 to 7.19.

The relationship between the normalized bubble length $L_b / l$ and the ratio of gas superficial velocity and liquid superficial velocity, $\alpha = \overline{u}_g / \overline{u}_l$, is plotted in Fig. 6(a). Typically, the smallest and largest bubble lengths are 2.53 mm and 8.22 mm respectively, corresponding to the normalized bubble length by the width of channel $L_b / l$ from 1.27 to 4.11. It is important to precise that no obvious decrease of $L_b$ was observed along the axial distance $X$ (not shown here), indicating that the compressibility effects of the gas phase are negligible in the channel. Garstecki et al. (2006) observed that for T-junctions with aspect ratio greater than or equal to 1, the squeezing mechanism was expected to dominate the bubble formation for $Ca < 0.01$. Therefore, a simple scaling law was proposed to predict the bubble length, as below:

$$L_b / T = 1 + \lambda \frac{\overline{u}_c}{\overline{u}_L}$$

(4)

where the value of $\lambda$ depends on the geometry of the T-junction. It can be seen from Fig. 6(a) that the linear evolution between $L_b / l$ and $J_c / J_l$ is experimentally verified, which would indicate that the squeezing mechanism is suitable to predict the bubble length. In the present study, by using the linear least square regression method, it can be obtained:

$$L_b / T = 1 + 1.69 \frac{J_c}{J_l}, \text{ mean deviation : } 6.40\%$$

(5)

The present value of $\lambda$ is larger in comparison with the one obtained by Roudet et al. (2011) (equals 1.13). This could be explained by the fact that in Roudet et al. (2011), the experiments were not carried out in aqueous solutions of glucose and sodium hydroxide, but in water, and also that the channel’s wettability could have changed.

Fig. 6(b) plots the variation of normalized slug length $L_s / l$ as a function of $J_c / J_l$. It can be observed that for a given $J_l$, $L_s / l$ first decreases significantly with the increase of $J_c / J_l$, and then changes slightly, but remain slightly dependent on $J_l$. This is consistent with the literature (Qian and Lawal, 2006; Leclerc et al., 2010; Roudet et al., 2011; Abadie et al., 2012). Regression of the experimental data provides the following correlation:

$$L_s / l = 1.87 \left( \frac{J_c}{J_l} \right)^{-0.5}, \text{ mean deviation : } 8.6\%$$

(6)

The value of the power of $J_c / J_l$ underlines the predominant contribution of the flow rate ratio to $L_s$.

Table 2 summarizes all the measured bubble characteristics.

3.1.3. Bubble shape

In the Taylor flow regime, the cartography of bubble shape displayed in Fig. 4 shows that depending on the dimensionless numbers ($Ca, We, Ce$), the bubble shape changes. It can be observed that: when $Ca < 0.003$, the bubble shape is slightly deformed, which is in agreement with what was observed in the creeping flow ($Ca < 0.01$) by Giavedoni and Saita (1999). However, in the present study ($0.0015 < Ca < 0.0066$), not only the viscous force could be attributed to the bubble shape, but also the inertial force ($0.29 < We < 5.32$).

When $We > 1.40$, the nose and rear menisci of the bubble could be well described as two hemispherical caps; however, when $We > 1.40$, the front meniscus goes flat, while the rear meniscus turns slender. This phenomena is consistent with the one observed by Roudet et al. (2011), which the bubble changes in shape were observed when $We > 1.5$. 

![Fig. 6. (a) Normalized bubble length and (b) normalized slug length versus gas liquid flow rate ratio for varying liquid phase velocities in Taylor flow regime in the meandering channel.](image-url)
When the bubble passes the bends, the centrifugal effect becomes essential. Compared with the inertial forces, the centrifugal forces have the same order of magnitude (0.39 < Ce < 7.19), thus the bubble shape is distorted by the contributions from both two forces. When Ce < 1.90, the extent of distortion of the bubble is slight; by increasing Ce, We and Ce, the bubble deformation appear to be more pronounced.

3.1.4. Interfacial area

The Taylor bubbles flowing in the meandering channel were not only affected by inertial effects, but also by the centrifugal effects induced by their passage in the periodic bends (see Figs. 3 and 4). These shape distortions imply that the bubbles cannot be described, as classically done, by two hemispherical caps for the bubble nose and rear, with a cylindrical shape for the bubble body. As a consequence, the bubbles were considered as prolate ellipsoid, leading to

$$S_B = \frac{\pi \times l^2}{2} \times \left[ 1 + \frac{L_B}{l - e} \arcsin(e) \right]$$

$$V_B = \frac{\pi \times l^2 \times L_B}{6}$$

where $S_B$ and $V_B$ represent the surface and the volume of the bubble, respectively, and $e$ is the eccentricity of the ellipsoidal bubble, $e^2 = 1 - \frac{l^2}{L_B}$. For the conventional gas-liquid contactors, the definition of interfacial area $a$ can be as follow:

$$a = \frac{S_B}{V_T}$$

where $S_C$ is the total surface of the bubbles in the contactor, and $V_T$ the whole volume of gas and liquid phases. In our case, due to the fact that one unit cell, which is constituted of one bubble and one liquid slug, appears repeatedly along the channel for Taylor flow regime, it can define $a$ as below:

$$a = \frac{S_B}{V_{UC}} = \frac{S_B}{V_L + V_B}$$

where $V_{UC}$ is the volume of one unit cell, and $V_I$ the volume of the liquid phase, which includes the volume of liquid slug and liquid film. As the gas fraction $\beta = Q_G/(Q_G + Q_L) = V_B/(V_B + V_I)$, Eq. (10) can be rewritten as:

$$\beta = \frac{\beta \times S_B}{V_B} = \frac{3 \times \frac{L_B}{l - e} \arcsin(e)}{1 + \frac{L_B}{l - e} \arcsin(e)}$$

Fig. 7 plots the evolution of $a$ as a function of $\beta$. It shows that a perfect linear relationship exists between $a$ and $\beta$. From Eq. (11), it can be known that the slope represents the ratio of $S_B$ and $V_B$.
Several filaments of higher O\textsubscript{2} concentration can be distinguished in the liquid slug. Two non-symmetrical recirculation zones with respect to the channel centerline could be identified, whereas in straight channels, two recirculation loops symmetrical and equal in size take place.

When the liquid slug moves in the straight section between two bends (t = 5 ms and t = 10 ms), the amount of oxygen accumulated just in the front of the bubble tends to decrease as progressively transported towards the liquid slug core; the recirculation zones are also growing, until reaching to the first bend (t = 15 ms). At this location, they are twisted and split up. Then, the passage through a bend affects significantly the shape and locations of these recirculation zones, and more generally the structure of the flow in the liquid slug. These distortions of the filaments of higher oxygen concentrations can be explained by the centrifugal effects induced by the channel curvature and the occurrence of the Dean vortices (Anxionnaz-Minvielle et al., 2013; Dai et al., 2015b). Herein, the mixing in the liquid slugs can be enhanced, the change in equivalent oxygen concentration fields in the liquid slugs as far as the bubbles moves is an indirect measure of this phenomenon (t = 20–40 ms).

The corresponding temporal evolution of the amount of transferred O\textsubscript{2} in the liquid slug is displayed in Fig. 8b. A kind of critical point seems to exist corresponding to the time where the recirculation zones are passing the bend (t = 20 ms). Before this point, the slope of the curve is higher than the one after the point. This would highlight first the contribution of the bubble formation stage to the mass transfer, and then a slowed-down mass transfer due to the mixing effect induced by the bend that could decreases the average driving force in the liquid slugs. Deeper investigations would be required to confirm this first trend.

Fig. 9 shows the evolution of O\textsubscript{2} concentration fields in the liquid slug obtained at different axial positions (X = 0.02, 0.07, 0.12, 0.17, 0.22, 0.32 and 0.42 m) along the meandering channel (j\textsubscript{L} = 0.139 m s\textsuperscript{-1}, j\textsubscript{G} = 0.069 m s\textsuperscript{-1}). It can be seen that the equivalent O\textsubscript{2} concentration level in the liquid slugs is getting greater and greater, until reaching an almost saturated state at X = 0.42 m. The apparent recirculation zones inside the liquid slugs also tend to disappear as far as flowing in the channel and thus the concentration fields become more and more uniform in the liquid slugs. This indicates that after several bends (about 20 bends), the gradient of O\textsubscript{2} concentration within the liquid slug no longer exists.

The accumulation of O\textsubscript{2} at the front and at the rear of the bubble can be observed along the first bends, and tend to disappear, as the recirculation zones, for the higher axial locations, namely when the liquid slugs become almost perfectly mixed.

For different hydrodynamic conditions, a comparison of the equivalent O\textsubscript{2} concentration fields, observed at the axial position X = 0.07 m is proposed in Fig. 10. It can be logically observed that increasing the gas flow rate (and thus the bubble length) under a given liquid flow rate (i.e. Fig. 10a and b, or Fig. 10c and d, or e and f) leads to a rise in the amount of oxygen transferred in the liquid slugs. The opposite effect is observed for increasing the liquid flow rate (and thus the liquid slug length) at a given gas flow rate.

3.2.2. Overall gas-liquid mass transfer coefficient

The equivalent O\textsubscript{2} concentration at the point (x, y) in the liquid slug could be extracted from the image processing, noted as C(x, y). It is important to remind that the equivalent O\textsubscript{2} concentration field obtained at a given location (x, y) is a mean value along the z-axis.
(Dietrich et al., 2013), no discrimination of the equivalent oxygen concentration fields related to each xy-plane being possible along the channel height. Therefore the mass flux of O\(_2\) transferred per unit of bubble surface at a given axial position \(X\), noted as \(\varphi_{O_2}(X)\), can be calculated as below:

\[
\varphi_{O_2}(X) = \frac{\int \int C(x, y) \times dx \times dy \times dz}{S_b \times t_b}
\]  

(12)

where \(t_b\) is the time for a bubble for flowing to the axial position \(X\) after the pinch-off, which is defined according to \(X/U_B\), \(S_b\) the bubble surface, which can be calculated from Eq. (7).

Fig. 11 (a) illustrates the evolution of \(\varphi_{O_2}\) in the liquid slug as a function of \(X\) under various \(j_l\) at a given \(j_c = 0.042\) m s\(^{-1}\). It clearly appears that \(\varphi_{O_2}\) is not constant at all along the channel length: higher values are observed for the axial positions close to the gas injection (\(X < 0.02\) m). Such result is consistent with the observations made in our previous work (Yang et al., 2016b), where it was outlined how the bubble formation stage could accelerate the mass transfer, when compared to the flowing stage. Then, the value of \(\varphi_{O_2}\) decreases significantly as the bubble moves along the meandering channel, and tends to reach a plateau after \(X = 0.3\) m.

The averaged equivalent O\(_2\) concentration, \(\overline{C}\), in the liquid slug of the unit cell (see Fig. 3a) can be also calculated, at the given axial position \(X\), from the integration of the equivalent oxygen concentration field in a unit cell, or more precisely in the liquid slug as no information could be acquired in the liquid film close to the walls. This can be expressed as below:

\[
\overline{C} = \frac{\int \int C(x, y) \times dx \times dy \times dz}{L_{uc} \times L^2}
\]  

(13)

Note that as the visualization of the bubble areas of the nose and rear is also not available due to their ellipsoidal shape, the liquid volume linked to these bubble areas cannot be taken into
account for the calculation. It has been estimated that this bias could lead to a maximum error of 13% on the volume estimation of liquid phase in a unit cell.

Fig. 11(b) shows the evolution of the normalized averaged $O_2$ concentration $\bar{C}/C_0$ ($C_0$ being the $O_2$ concentration at saturation) in the liquid slug as a function of the axial position $X$ in the channel, under various gas superficial velocities $j_g$ and at given liquid superficial velocity $j_l = 0.069$ m s$^{-1}$. It can be observed that $\bar{C}/C_0$ increases as the gas superficial velocity increases under the given liquid superficial velocity, which is consistent with the visualization presented in Fig. 10.

For Taylor flow, the liquid phase consists in a series of mixed slugs, and then the plug-flow model is often used for mass transfer purposes. Uniform velocity and concentration over the channel cross section are then assumed, without taking into account the real structure of the flow in the liquid slug, for example, recirculation loops (Beričić and Pintar, 1997; Shao et al., 2010). Based on an oxygen mass balance on the liquid slug, it can be written (Roudet et al., 2011):

$$\phi_{O_2}(X) = j_l \cdot \frac{dC}{dX} = k_a \cdot a \cdot \frac{C - \bar{C}(X)}{L}$$  \hspace{1cm} (14)

In the present case, the concentration of oxygen, $\bar{C}(X)$ is null as fully consumed by the reaction in reality, leading to

$$\frac{\bar{C}}{C_0} = \frac{k_a \cdot a \cdot X}{j_l}$$  \hspace{1cm} (15)

where $k_a$ represents the volumetric mass transfer coefficient, s$^{-1}$.

There exists then a linear relationship between $\bar{C}/C_0$ and $X$, and the slope stands for $k_a j_l$. Once applied, it has been found that Eq. (15) could predict well the experimental values of $\bar{C}/C_0$ only for $Re < 300$; for $Re > 300$, the deviation becomes higher than 18%. Typical examples of the comparison between the experimental and predicted evolutions of $\bar{C}/C_0$ are displayed in Fig. 11(b). Note that, when a fitting of the experimental values of $\bar{C}/C_0$ with $X$ is applied (taking $X = 0.02$ m as a reference), different values are obtained at $X = 0$; this could be explained by the contribution of the bubble formation stage to the overall mass transfer. Fig. 11(b) also reveals that, for $Re > 300$, the prediction of Eq. (15) are relevant only before the axial positions below $X = 0.22$ m. As shown in Fig. 1(b), this specific location corresponds to the big bend (called “turning point”), namely to the location where the flow direction is changed in 180°. After $X = 0.22$ m, $\bar{C}/C_0$ is underestimated by the model, indicating that the big bend leads to slow down the flux of oxygen transferred (this is observed for all the operating conditions when $Re > 300$). More in-depth investigation of the velocity field structure of the two-phase flow when crossing the turning point would be required to explain this phenomenon.

The overall volumetric mass transfer coefficients deduced from the fitting between Eq. (15) with the experimental points just before the turning point, $k_a$, are reported in Table 2. They are ranged from 0.1 to 1 s$^{-1}$, which the order of magnitude is in agreement with the values reported by Roudet et al. (2011). This can verify the feasibility of the colorimetric technique. As already observed in the literature (Nieves-Remacha et al., 2013; Yao et al., 2014), $k_a$ increases when the gas superficial velocity $j_g$ increases and when the liquid superficial velocity $j_l$ increases. It is important to keep in mind that these overall volumetric mass transfer coefficients are just global parameters representing the inverse of characteristic time of mass transfer. The values here estimated remind approximate due to the assumptions made in the modelling (plug-flow) and to the fact that the contribution of the film in not taken into account.

At last, as the interfacial area $a$ can be calculated by the assumption of the bubbles as prolate ellipsoid (see Section 3.1.4), the liquid-side mass transfer coefficient before the turning point $k_i$ can be calculated by dividing $k_a j_l$ with $a$. As shown in Table 2, for a given $j_l$, $k_i$ increases with increasing $j_g$, and is less sensitive to the gas superficial velocity.

4. Conclusion

The colorimetric technique proposed by Dietrich et al. (2013), which was based on an oxygen-sensitive dye, was implemented to locally visualize and characterize the gas-liquid mass transfer in a square meandering millimetric channel. The hydrodynamics characteristics was first investigated in the channel, in terms of gas-liquid flow regime, gas hold-up, bubble length and velocity, liquid slug length and interfacial area. The oxygen concentration field inside the liquid slugs was then visualized at different axial positions. The non-symmetrical recirculation zones inside the liquid slug was expressed by the periodic bends, and tended to disappear as the bubble moving within the channel; the $O_2$ concentration inside the liquid slug became uni-
form after passing through several bends (around 20 bends). The occurrence of the “turning point” (i.e. the big bend enabling the flow direction in 180°) seemed to induce a slowing down of the mass transfer, especially when Re > 300 and Ca > 0.002. The mass flux of O₂ transferred per unit of bubble surface was significantly higher close to the gas injection due to the bubble formation stage, then it decreases significantly as the bubble flowing along the meandering channel, and becomes smaller after X = 0.3 m due to the decreasing driving force for the mass transfer. At last, by coupling this local information with a plug-flow model, overall volumetric mass transfer coefficients could be estimated. All these findings give important information to understand the gas-liquid mass transfer complex mechanism occurring in the meandering channel, which would serve as basis for implementing gas-liquid reaction in HEX reactors.

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References


