Chemical Engineering Science 165 (2017) 192–203

Contents lists available at ScienceDirect

Chemical Engineering Science

journal homepage: www.elsevier.com/locate/ces

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

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## HIGHLIGHTS

- Local investigations of the gas-liquid mass transfer in a meandering millichannel.
- The presence of bends affects significantly the gas-liquid mass transfer.
- Evolution of mass flux of $O_2$ transferred along the channel length.
- Influence of the connection part of two "straight" sections on mass transfer.

## ARTICLE INFO

*Article history:*
Received 15 December 2016
Received in revised form 27 February 2017
Accepted 2 March 2017
Available online 4 March 2017

*Keywords:*
Gas-liquid mass transfer
Taylor bubbles
Meandering channel
Colorimetric technique
Image processing

## 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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http://dx.doi.org/10.1016/j.ces.2017.03.007
0009-2509/

**Nomenclature**

| | |
|---|---|
| $a$ | the interfacial area, $m^{-1}$ |
| $A$ | $l^2$, cross-sectional area of the channel, $m^2$ |
| $C$ | equivalent oxygen concentration, kg $m^{-3}$ |
| $C$ | average equivalent oxygen concentration in the liquid slug of the unit cell, kg $m^{-3}$ |
| $C^*$ | dissolved oxygen concentration at saturation, kg $m^{-3}$ |
| $C_0$ | distribution parameter |
| $d_h$ | hydraulic diameter of the meandering channel, m |
| e | eccentricity, $e^2 = 1 - l^2/L_B^2$ |
| $j$ | superficial velocity, m $s^{-1}$ |
| $k_L$ | the liquid side mass transfer coefficient, m $s^{-1}$ |
| $k_La$ | volumetric mass transfer coefficient, $s^{-1}$ |
| $l$ | width of the channel, m |
| $L$ | length, m |
| $Q$ | flow rate, $m^3$ $s^{-1}$ |
| $t$ | flowing time, s |
| $U$ | velocity, m $s^{-1}$ |
| $V$ | volume, $m^3$ |
| $X$ | axial position, m |
| $\beta$ | volumetric quality of gas |
| $\varepsilon$ | gas hold-up |
| $\varphi$ | volumetric mass flux of the transferred $O_2$ per unit of bubble surface, kg $m^2$ $s^{-1}$ |

*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, $=\mu_L \times U_B/\sigma_L$, dimensionless |
| $Ce$ | Centrifugal number, $=\rho_L \times U_B^2 \times l^2/\sigma_L \times r_c$, dimensionless |
| $Re$ | Reynold number, $=\rho_L \times U_B \times l/\mu_L$, dimensionless |
| $We$ | Weber number, $=\rho_L \times U_B^2 \times l/\sigma_L$, dimensionless |

*Subscripts*

| | |
|---|---|
| B | bubble |
| bt | before turning point |
| d | drift velocity |
| G | gas phase |
| $L$ | liquid phase |
| s | liquid slug |
| UC | unit cell |

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 raises 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, $k_La$ increased coherently when increasing superficial gas velocity $j_g$, 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