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Local visualization and investigation on gas-liquid mass transfer around Taylor bubbles flowing in an in-plane spiral-shaped millireactor

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Introduction

Microstructured reactors present several significant advantages over conventional macroscale reactors, such as smaller volumes, higher surface to volume ratios and enhanced mass and heat transfer. In particular, gas-liquid reactive flows are of interest due to numerous industrial applications, such as hydrogenations, sensitized photo-oxygenations or photoredox reactions, fluorinations, biochemical reactions. Among all of the gas-liquid flow patterns, Taylor flow is one of the most investigated flow regimes as providing stable, regular and easily tunable conditions. A large number of researches have been focused on investigations in straight channels [1], while few studies are devoted to channels with bends, such as meandering, spiral-shaped and helicoidal tubes [2], even if they enable to reach longer residence time with a more compact design. Spiral-shaped millireactors, specially developed for flow photochemistry [3,4], offer a continuous curvature along the channel that can induce transverse Dean flows, leading to enhanced mixing efficiency. However, no study reports investigations at a local scale on gas-liquid mass transfer in such devices. In keeping with this context, this work aims at locally visualizing and quantifying the gas-liquid mass transfer in an in-plane spiral-shaped millireactor under Taylor flow conditions, using the colorimetric technique proposed by Dietrich et al. (2013).

Methods

The experimental set-up was depicted in Fig. 1(a). The spiral-shaped millireactor consisted of a tube (FEP material) inserted inside a channel carved into a flat poly (methyl methacrylate) plate and wound into a spiral geometry.



Fig. 1. (a) schematic diagram of the experimental set-up. (b) typical image of gas-liquid flows

Different diameters of tubing (500 µm, 1 and 2 mm) were used, and the length of the tubing could also be modulated from a few cm and up to 10 m. The bubbles were generated using a T-junction with different inner diameters. Thanks to its spiral geometry, the developed millireactor was flat and compact (160×160 mm²), allowing for homogeneous irradiation over the whole tubing surface in photochemistry applications. Air was used as gas phase and was fed using house compressed air lines; air flow rates ware controlled by a HORIBASTEC airflow controller. Liquid flow was supplied by a neMESYS syringe pump (Model: NEM-B100-01 A) equipped with a 100mL Harvard Instruments syringe. The entire millireactor was put into a glass tank filled with water to avoid optical distortion. The shadowgraph method was applied to investigate the gas-liquid hydrodynamics and mass transfer: The channel was lighted by a LitePad HO LED backlight (Rosco[®]) and the images were recorded by a monochromatic high-speed CMOS camera (Photron[®] SA3) at 2000 frames per second. The colorimetric technique is a non-intrusive and direct method, which has been successfully used to study mass transfer for Taylor flow stage in straight and meandering milli-channels [6] and bubble formation stage in microchannels. It is based on an oxygen sensitive dye, resazurin, which can be reduced to resorufin and further reduced to dihydroresorufin. The change of color results from reversible oxidation (fast) and reduction (slow) reactions between dihydroresorufin (colorless) and resorufin (pink). This color intensity can be related to an equivalent dissolved oxygen concentration by establishing calibration curves and image postprocessing.

Results and Discussion

A typical image of Taylor flow is shown in Fig. 1(b). Depending on the gas and liquid flow rates and on the tube diameter, two-phase spatial distribution was observed and the transition between flow regimes was identified. Moreover, gas hold-up, bubble and liquid slug length and bubble velocity evolution along the curved channels were quantified. The effects of the ratio of gas superficial velocity and liquid superficial velocity and Dean's number on gas-liquid mass transfer were explored as well as the effect of the diameter of the T-junction. The effect of the tube curvature on the oxygen concentration field inside the liquid slug was also discussed.

Conclusion

By using the colorimetric technique, gas-liquid mass transfer was studied in an in-plane spiral-shape millireactor at a local scale. The influence of the different operating parameters was quantified and a first attempt of scaling law was proposed.

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