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Optical determination of oxygen mass transfer in a helically-coiled pipe compared to a straight horizontal tube

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HIGHLIGHTS

- Measurement of mass transfer coefficients in a helical pipe.
- Locally resolved measurement through colour reaction with resazurin.
- Comparison with straight horizontal tube shows the advantages of the helix.
- First two to three turns have about one order of magnitude higher volumetric mass transfer coefficients.

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ABSTRACT

The gas-liquid mass transfer of oxygen has been examined experimentally in a helically-coiled pipe using an optical colorimetric method. Two tracer redox-reactions have been used for this purpose: (i) methylene blue and (ii) resazurin. The gas hold-up has been varied from 0.1 to 0.6, and liquid flow rate from 0.37 to 0.68 l/min, both leading to plug-flow conditions in the helix for all parameter combinations. A progress variable has been defined to measure the advancement of the reaction and thus, oxygen transfer to the liquid. The resazurin reaction turned out to be more appropriate for mass transfer studies, due to the bigger time constant difference of oxidation and reduction. Therefore, it can be adapted more easily to the specific conditions of the set-up. It also possesses more intense colours and higher fluorescence intensity, which might be useful for other mass transfer studies. As expected, the oxygen mass transfer increases with gas hold up, but liquid flow rate has little influence. A comparison with a horizontal tube of the same diameter shows the drastically increased mass transfer in the helix, due to better radial mixing and different flow pattern. Depending on the experimental conditions, oxygen concentration in the helix was up to twice that of the horizontal tube. This can be explained by the much higher liquid side mass transfer velocity, which is determined in this study from the experimental $k_l a$-values and specific bubble surface per turn of the helix. For this purpose, bubble dimensions and gas-liquid cell volumes have been determined from the experimental images. The paper thus presents a locally resolved characterization of gas-liquid mass transfer in a helically coiled reactor. The high mass transfer especially during the first turns of the helix shows its advantages compared to a straight tube.

1. Introduction

Generally, curved pipes show improved performance regarding key processes like mixing, heat transfer, and mass transfer (Abdalla, 1994; Jokiel et al., 2017; Moulin et al., 1996; Rennie and Raghavan, 2005). Indeed, the existence of counter-rotating vortices, corresponding to the secondary flow in helical pipes, is mainly responsible for the improved mixing and mass transfer characteristics. Dean (1927, 1928) showed that these vortices are arising due to the unbalanced centrifugal forces exerted on the flow. Consequently, helical pipes have improved radial mixing and residence time distributions, e.g. Klutz et al. (2015), Kurt et al. (2016), Vural Gürsel et al. (2016). However, the flow in helical pipes involves typically a higher pressure drop when compared to a straight pipe of the same length (Ito, 1959; Ju et al., 2001; Mishra and Gupta, 1979). (Vanka et al., 2004) performed a numerical study to compare the mixing performance between curved and straight channels, considering the effects of Reynolds and Schmidt numbers. They showed that the mixing in curved channels is dramatically more efficient than in a straight channel. (Kumar et al., 2006) performed another numerical investigation to compare mixing of miscible liquids in helical pipes and straight pipes. Likewise, they showed that the mixing efficiency in helical pipes is generally

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Nomenclature

a	interfacial area [1/m]
A_b	bubble surface [m^2]
b	coil pitch [mm]
c_{col}	colorant concentration [mg/l]
$c_{O2equiv}$	equivalent oxygen concentration [mg/l]
$c_{o2}(t)$	actual oxygen concentration [mg/l]
c^*_{o2}	oxygen saturation concentration [mg/l]
c_{sat}	resazurin saturation concentration [g/l]
D	inner coil diameter [mm]
d	inner pipe diameter [mm]
d_b	bubble diameter [mm]
e	eccentricity [–]
h_b	bubble height [mm]
I	local intensity [a.u.]
Io	initial, zero intensity [a.u.]
If	final, saturated intensity [a.u.]
l_b	bubble length [mm]
k_l	liquid-side mass transfer coefficient [m/s]
k_la	volumetric mass transfer coefficient [1/s]
L	coil length [m]
M_{DHR}	molar weight of DHR [g/mol]
M_{LMB}	molar weight of LMB [g/mol]
M_{O_2}	molar weight of oxygen [g/mol]
n	number of turns [–]
pv	progress variable [–]
Q_l	liquid flow rate [l/min]
Q_g	gas flow rate [l/min]
S_s	specific bubble surface per turn [1/m]
t	time [s]
v_m	liquid-side mass transfer velocity [m/s]
$v_{s,l}$	superficial liquid velocity [m/s]
$v_{s,g}$	superficial gas velocity [m/s]
V_{turn}	volume of one turn of the helix [m^3]

Dimensionless numbers

Re_{2ph}	two-phase Reynolds number, $Re_{2ph} = (v_{s,l} + v_{s,g}) \cdot d/\upsilon_1$ [–]
De_{2ph}	two-phase Dean number, $De_{2ph} = Re_{2ph}\sqrt{\delta}$ [–]
Re_l	liquid Reynolds number, $Re_l = v_{s,l} \cdot d/\upsilon_l$ [–]
De_l	liquid Dean number, $De = Re_l\sqrt{\delta}$ [–]
Ha	Hatta number [–]
E	enhancement factor [–]

Greek symbols

δ	curvature ratio of the coil, $\delta = d/D$ [–]
ε	gas hold-up [–]
ν_l	liquid kinematic viscosity [m^2/s]

Indices

i	i-th turn of the helix

Abbreviations

DHR	dihydroresorufin
LMB	leuco-methylene blue
MB	methylene blue
RF	resorufin
RZ	resazurin

much better than in straight pipes. (Mansour et al., 2017) numerically examined liquid-liquid mixing for various Reynolds and Schmidt numbers, as well as for different inlet conditions. They determined two optimal Reynolds numbers for mixing, Re = 50 and Re = 1000.

In contrast to other two-phase systems such as Taylor-flows or bubble columns, studies of mass transfer from a gas to a liquid phase are scarce for the case of helix reactors. Also, optical methods, which are routinely used in the aforementioned systems (e.g. (Böhm et al., 2016; Bouche et al., 2013; Butler et al., 2016; Butler et al., 2018; Dietrich et al., 2013; Jimenez et al., 2013; Kastens et al., 2015; Kováts et al., 2017; Merker et al., 2017; Rzehak et al., 2017; Yang et al., 2016), have seldom been applied to helical tubes. The intensification of liquid-liquid reactions and mass transfer from gasses to the liquid phase have been examined recently, e.g., by the groups of Nigam and Kockmann (Kurt et al., 2015; Kurt et al., 2017; López-Guajardo et al., 2017). These authors first used intrusive probing techniques, and more recently leuco-indigo carmine (Krieger et al., 2017a; Krieger et al., 2017b), for the determination of concentrations inside and at the outlet of the helix. (Kaufhold et al., 2013) studied the absorption of CO_2-gas in hollow helical fibres by measuring the pressure change. The flow structure of gas-liquid flows in helical tubes have been investigated by camera visualization in (Murai et al., 2006b), but these authors did not study mass transfer. A first optical study of mass transfer in helical tubes has been presented by the authors in comparison with numerical simulations and probe measurements (Jokiel et al., 2017). However, no systematic evaluation of the influencing parameters was presented there.

In this study, oxygen mass transfer from gas bubbles to an oxygen-free aqueous reaction solution is studied systematically by an experimental, optical method in a helically-coiled tube for a wide range of gas hold-up and liquid superficial velocity. Two colorimetric redox-reactions are used for that purpose, utilizing methylene blue or resazurin. The results are compared to measurements done in the same manner in a horizontal tube of same inner diameter. Bubble sizes are calculated from the obtained images. Volumetric mass transfer coefficients k_la can be determined from the measured concentrations. These allow finally for the calculation of a liquid-side mass transfer velocity. This extensive study shows the advantages of coiled reactors compared to a straight tube. In the helix, mass transfer is especially effective in its first 3 coils.

2. Helix geometry and flow conditions

A helical coil, made of glass, with an inner pipe diameter of d = 6 mm, an inner coil diameter of D = 20 mm, a pitch of b = 13.4 mm, and a total number of turns of n = 15 was considered. With the present dimensions, the coil curvature ratio is $\delta = d/D = 0.3$ and the total coil length is $L = n\sqrt{\pi^2(D+d)^2 + b^2}$ = 1.24 m. It is the same geometry as already used, as case G2, in (Jokiel et al., 2017). A drawing of the helical pipe geometry is shown in Fig. 1.

The flow conditions used in these systematic measurements are summarized in Table 1, where the gas hold-up $\varepsilon = \frac{Q_g}{Q_g+Q_l}$, the superficial liquid $v_{s,l}$ and gas velocities $v_{s,g}$ are given together with the corresponding liquid and gas flow rates (Q_l, Q_g), residence times t_{res}, two-phase Reynolds and Dean numbers ($Re_{2ph} = (v_{s,l} + v_{s,g}) \cdot d/\upsilon_1$, $De_{2ph} = Re_{2ph}\sqrt{\delta}$) and liquid Reynolds- and Dean-numbers (Re_l, De_l), for all 14 measuring points (MP01 to MP14 in Table 1). They are all situated in the plug-flow regime as shown in Fig. 2.