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Measurement and simulation of mass transfer and backmixing behavior in a gas-liquid helically coiled tubular reactor

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HIGHLIGHTS

- Different measurement techniques were used to study the gas-liquid mass transfer.
- Computational Fluid Dynamics (CFD) was used to model the gas-liquid mass transfer.
- The results indicate an influence of the coil curvature ratio on the mass transfer.

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ABSTRACT

Volumetric mass transfer coefficients (k_La) and Bodenstein numbers (Bo) for the elongated bubble flow regime in horizontal helically-coiled tube reactors are reported using two different measurement techniques (oxygen optodes, and optical observation of an oxygen-sensitive dye). Additionally, the gas-liquid mass transfer and the residence time behavior of the two-phase flow were described with a 3D Computational Fluid Dynamics (CFD) model, and also with a one-dimensional two-phase model. For this study, 16 cases involving different gas and liquid volumetric flow rates were employed to generate air-water flows through two helically coiled tubes with curvature ratios of $\delta_1 = 0.093$ and $\delta_2 = 0.3$, respectively. The superficial gas and liquid Reynolds numbers ($Re_{s,G}$ and $Re_{s,L}$) and the gas hold-up (ϵ_G) are varied from 494 to 2483, from 1456 to 2713, and from 0.46 to 0.81, respectively. The mass transfer measurements show an increasing gas-liquid mass transfer rate with increasing superficial velocity of the liquid-phase and $Re_{s,G}$. The Bodenstein number decreases with increasing gas-phase Reynolds number and increases with increasing superficial velocity of the liquid-phase. Correlations describing the mass transfer and backmixing behavior are proposed. The CFD results are in excellent agreement with the experimental data. With the 1D two-phase model it is possible to describe the residence time behavior of the two-phase flow through the helically coiled tube.

1. Introduction

For continuous processes, the most often used chemical reactors are stirred tank reactors, tubular reactors and variations of these types. Due to their specific characteristics, they feature different backmixing properties. These backmixing properties are very important, since they strongly influence the performance of the reactor, namely the conversion and selectivity behavior in dependence on the underlying reaction kinetics. Moreover, if the reactants are in different state of aggregation, the backmixing behavior found in such multi-phase flows is completely different from one-phasic operation, and the mass transfer between the phases becomes an additional factor, often limiting conversion rates. In such cases, mass transfer must be enhanced by modifying the reactor geometry, e.g., by adding static mixers within tubular reactors.

It is known for a long time that the axial backmixing and mass transfer behavior of tubular reactors can be enhanced by using coiled tubes instead of straight ones (Vashishth et al., 2008). Due to coiling, the centrifugal force is induced which influence the flow and a strong secondary flow forms in radial direction, leading to Dean vortices. The secondary flow reduces axial backmixing and

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Nomenclature

ΔP_{TP}	two-phasic pressure loss (Pa)
δ	curvature ratio $\delta = \frac{d}{D}$
ϵ_i	volume fraction $\epsilon_i = \frac{\dot{V}_i}{\dot{V}_G + \dot{V}_L}$
η	dynamic viscosity (Pa s)
ν	stoichiometric coefficient
ρ	density $(\mathrm{kg\,m^{-3}})$
Σ	sink and source term $(\mathrm{mol\,m^{-3}\,s^{-1}})$
σ	surface tension $(\mathrm{kg\,s^{-2}})$
τ	residence time (s)
$\overline{\tau}$	characteristic time of methylene blue oxidation (s)
τ_{Probe}	response time of the oxygen probe (s)
Θ	dimensionless time $\Theta = \frac{t}{\tau}$

Latin symbols

b	coil pitch (m)
Bo	Bodenstein number $Bo = \frac{u\,L}{D_{ax}}$
C	dimensionless concentration $C = \frac{c}{c_{sat}}$
c	concentration $(\mathrm{mol\,m^{-3}})$
$\bar{c}$	dimensionless progress variable $\bar{c} = \frac{I - I_0}{I_f - I_0}$
$C_{L\ \mathrm{bulk}}$	bulk liquid concentration $(\mathrm{kg\,m^{-3}})$
$C_{L\ \mathrm{sur}}$	concentration at liquid surface $(\mathrm{kg\,m^{-3}})$
c_{sat}	liquid saturation concentration $c_{sat} = P\,X_{O_2}\,K_H^{\#}$ $(\mathrm{mol\,m^3})$
D	inner coil diameter (m)
d	inner tube diameter (m)
De	Dean number $De = Re\left[\left(\frac{D}{d}\right)\left(1 + \frac{b}{\pi\,D}\right)^2\right]^{-0.5}$ (Mishra and Gupta, 1979)
D_{ax}	axial dispersion coefficient $(\mathrm{m^2\,s^{-1}})$
D_L	liquid diffusion coefficient $(\mathrm{m^2\,s^{-1}})$
$E(\Theta)$	dimensionless age distribution function
Eu	Euler number $Eu = \frac{\Delta P_{TP}}{\rho_L u_{s,L}^2}$
$F(\Theta)$	dimensionless cumulative distribution function
I	local intensity (a.u.)
k	overall mass transfer coefficient $(\mathrm{m\,s^{-1}})$
K_H	dimensionless Henry coefficient $K_H = K_H^{\#} R\,T$
$K_H^{\#}$	Henry coefficient $K_H^{\#} = 1.3 \cdot 10^{-5}\ \mathrm{Pa\,m^{-3}\,mol^{-1}}$ (Sander, 2015)
$k_L a$	volumetric mass transfer coefficient $(\mathrm{s^{-1}})$
L	length of the coil (m)
$\dot{m}$	mass transfer rate per unit volume $(\mathrm{kg\,m^{-3}\,s^{-1}})$
n_0	number density of bubble seeds $(\mathrm{m^{-3}})$
P	pressure (Pa)
R	universal gas constant $R = 8.31441\ \mathrm{J\,mol^{-1}\,K^{-1}}$ (VDI, 2010)
r	average bubble radius (m)
R^2	determination coefficient $R^2 = 1 - \frac{\sum_{j=1}^{n}\left(y_j - f_j\right)^2}{\sum_{j=1}^{n}\left(y_j - \bar{y}_i\right)^2}$
$Re_{s,i}$	superficial Reynolds number of phase i $Re_{s,i} = \frac{\rho_i\,u_{s,i}\,d}{\eta_i}$
T	temperature (K)
t	time (s)
u	velocity $(\mathrm{m\,s^{-1}})$
V	volume $(\mathrm{m^3})$
$\dot{V}$	volumetric flow rate $(\mathrm{m^3\,s^{-1}})$
X_{O_2}	molar fraction of oxygen in air $X_{O_2} = 0.209$ (VDI, 2010)
Z	dimensionless length scale $Z = \frac{z}{L}$
z	length scale (m)

Subscripts

0	onset
exp.	experimental
F	fastest
f	final
G	gas
I	integral
i	gas or liquid
L	liquid
s	superficial

improves mass and energy transfer between the phases (Vashishth et al., 2008). Other advantages of the coiled configuration are its higher compactness and relatively large surface area-to-volume ratio. Additionally, the laminar flow regime in coiled tubes is observed at larger Reynolds numbers, compared to straight tubes. However, it must be kept in mind that coiling of the tube leads to an increased pressure loss (Ito, 1959; Mishra and Gupta, 1979; Ju et al., 2001).

The present study concentrates on the backmixing properties and mass transfer in two-phase (gas/liquid) systems flowing through helically coiled tubes with the helix axis being positioned horizontally. As discussed in the next section, such systems have already been studied by other groups focusing on pressure loss (Rippel et al., 1966; Mishra and Gupta, 1979; Saxena et al., 1990; Ali, 2001; Mandal and Das, 2002; Kumar et al., 2006; Vashishth et al., 2008), heat transfer (Naphon and Wongwises, 2006; Kumar et al., 2006; Jayakumar et al., 2008; Fsadni and Whitty, 2016), or gas hold-up (Rippel et al., 1966; Saxena et al., 1990; Mandal and Das, 2002; Vashishth et al., 2008). In the present work, mass transfer and axial backmixing of air-water-flows through coiled tubes was investigated by a combination of complementary techniques: inline probes measuring oxygen absorption in water, non-intrusive optical measurements, and simulations relying on Computational Fluid Dynamics (CFD). Thanks to the combination of several powerful tools, the mass transfer and axial dispersion behavior in two-phase flows in helical coils can be better understood, opening the door for a later study involving additionally chemical reactions in this two-phase system.

2. State of the art

2.1. Mass transfer

Many studies have considered mass transfer in helically coiled tubes. Banerjee et al. (1970) studied the physical and chemical gas-liquid mass transfer of an annular flow and determined $k_L a$-values between $0.1\ \mathrm{s^{-1}}$ and $0.47\ \mathrm{s^{-1}}$. The volumetric mass transfer coefficients $k_L a$ were correlated with the pressure loss of the coil, depending on the tube diameter. Similar measurements in the slug-flow regime were carried out in helically coiled tubes by Kulic and Rhodes (1974). These authors found $k_L a$-values between $0.02\ \mathrm{s^{-1}}$ and $0.25\ \mathrm{s^{-1}}$ for the physical mass transfer and values between $0.22\ \mathrm{s^{-1}}$ and $0.55\ \mathrm{s^{-1}}$ for the chemical mass transfer. A comparison of the mass transfer data from Banerjee et al. (1970) and Kulic and Rhodes (1974) revealed a dependency of the mass transfer coefficient on the flow regime. Jepsen (1970) compared the physical mass transfer in helically coiled tubes and in straight horizontal tubes. The determined $k_L a$-values in the coil were between $0.01\ \mathrm{s^{-1}}$ and $0.2\ \mathrm{s^{-1}}$. The comparison indicated a higher mass transfer rate in coiled tubes. The slightly increased mass transfer rate found in coiled tubes was confirmed by other studies (Shah and Sharma, 1973; Hameed and Muhammed, 2003; Abdel-Aziz et al., 2010; Thandlam et al., 2016) for liquid-liquid, solid-liquid and gas-liquid situations. It should be mentioned that all these studies were performed in vertically orientated coils, which is differed from the configuration used in the present investigation.