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Gas–liquid mass transfer in a falling film microreactor: Effect of reactor orientation on liquid-side mass transfer coefficient

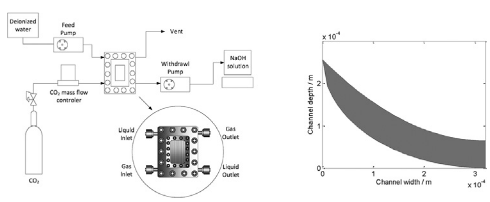
David Lokhat*, Ashveer Krishen Domah, Kuveshan Padayachee, Aman Baboolal, Deresh Ramjugernath

Reactor Technology Research Group, School of Engineering, University of KwaZulu-Natal, Durban 4041, South Africa

HIGHLIGHTS

- Lower mass transfer rates in inclined microchannels.
- Curved interface approximation gives more realistic mass transfer coefficients.
- Cellular convection in microchannels responsible for enhanced mass transfer.

GRAPHICAL ABSTRACT



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ABSTRACT

Microreactors offer a unique platform for chemical syntheses and have been applied to numerous reaction types including nitrations, fluorinations and hydrogenations. A key feature of falling film microreactors is the comparably large specific surface area they afford compared to conventional reactors. The enhanced heat and mass transfer characteristics can be exploited for rapid and exothermic reactions. Adequate understanding of the mass transfer processes occurring within microchannels is necessary for proper reactor design and optimization. In the current study the influence of reaction plate orientation and gas flowrate on liquid-side mass transfer coefficient was investigated via CO_2 absorption experiments. Lower plate angles resulted in lower liquid-side mass transfer coefficients. At higher film velocities the rate of mass transfer was greater. The experimentally determined mass transfer coefficients were at least twice as high as those predicted either by film or penetration theory. The enhancement in mass transfer is suggested to be due to cellular convection in the microchannels. For inclined reaction plates, increasing the gas flowrate had a positive effect on the mass transfer characteristics due to induced fluctuations of the gas–liquid interface.

1. Introduction

Microreactors are reactors with reaction channels of the order of micrometers, at which diffusion is the dominant mixing mechanism rather than turbulent eddies. The interest in this technology has been focused on fine and speciality chemical manufacture (Fletcher et al., 2002). The falling film microreactor (FFMR) is the most popular type of gas–liquid contactor in this class of reactors. Under normal operating conditions, the liquid flows down a microstructured reaction plate under the action of gravity and spreads to form an expanded thin film in each microchannel. The gas flows over the liquid film contained within the microchannels. This design allows for specific gas–liquid interfacial areas of up 20,000 m^2/m^3 to be achieved, which is noticeably higher than other gas–liquid contacting reactors (Hessel et al., 2000; Yue et al., 2007; Ziegenbalg et al., 2010). The high specific areas made available by the reactor design allow for high heat and mass transfer rates to be achieved which make FFMRs an efficient tool for studying the kinetics of fast and highly exothermic gas–liquid reactions. Researchers have exploited the high heat and mass transfer rates of FFMRs for reactions such as catalysed

* Corresponding author.
E-mail address: lokhat@ukzn.ac.za (D. Lokhat).

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Nomenclature

A_L	liquid cross sectional area in the microchannel, m^2
C_{out}	liquid phase concentration of CO_2 in the reactor outlet, $mol \cdot dm^{-3}$
C^*	saturated concentration of CO_2, $mol \cdot dm^{-3}$
C_{HCl}	concentration of HCl, $mol \cdot dm^{-3}$
D	CO_2 diffusivity in the liquid phase, $m^2 \cdot s^{-1}$
Fo	Fourier number
j_L	mean liquid velocity, $m \cdot s^{-1}$
k_L	liquid-side mass transfer coefficient, $m \cdot s^{-1}$
$k_{L,\ pen}$	penetration model liquid-side mass transfer coefficient, $m \cdot s^{-1}$
$k_{L,\ film}$	film model liquid-side mass transfer coefficient, $m \cdot s^{-1}$
L_{arc}	arc length of parabola, m
L_{ch}	length of microchannel, m
N_{ch}	number of channels
Q_L	liquid volumetric flowrate, $m^3 \cdot s^{-1}$
Re	Reynold number
t	collection time, s
W_{ch}	width of microchannel, m
We	Weber number
V_{HCl}	volume of HCl dispensed between titration endpoints, dm^3
x	width dimension of microchannel
y	depth dimension of microchannel

Greek symbols

δ_L	thickness of falling film, m
ν_L	kinematic viscosity of liquid, $m^2 \cdot s^{-1}$
θ	angle of reaction plate, °
σ_L	surface tension of liquid, $N \cdot m^{-1}$
ρ_L	density of liquid, $kg \cdot m^{-3}$

hydrogenation (Yeong et al., 2003), direct fluorination of aromatics (Jähnisch et al., 2000) and direct synthesis of hydrogen peroxide (Inoue et al., 2007).

The measurement of gas–liquid mass transfer in a falling film micro reactor has formed the basis for a number of published studies (Zanfir et al., 2005; Zhang et al., 2009). The reactive absorption of carbon dioxide (CO_2) and sulphur dioxide (SO_2) in aqueous sodium hydroxide have both been used effectively to characterize mass transfer efficiency in microreactors. For the measurement of liquid phase mass transfer coefficients, CO_2 absorption is preferred. The CO_2 reacts rapidly with the NaOH at the gas–liquid interface and forms sodium carbonate. The analysis of liquid reaction products can be done simply by titration and the reactants are inexpensive and non-toxic. For the measurement of gas phase mass transfer coefficient, the SO_2 method is preferred. Very low concentrations of the reactive gas-phase species must be used for measurement of the gas-phase mass transfer coefficient, to ensure that the process is limited by the gas phase mass transfer (Commenge et al., 2011). An SO_2 gas analyser is much more sensitive and selective at this concentration range compared to CO_2 sensors which may encounter interference from the nitrogen diluent.

Applications of these methods are illustrated in the literature. Zanfir et al. (2005) carried out an experimental and modelling study of the reactive absorption of carbon dioxide in a falling film microreactor. A two-dimensional model was formulated to represent the flow through the reactor and the experimental results were compared to the model prediction in terms of carbon dioxide conversion. The model gave good agreement with the experimental data at low inlet NaOH concentration. Moreover, the parametric analysis of the model indicated that the major rate limitation was on the liquid side and that the CO_2 was consumed within a short distance from the gas–liquid interface. Zhang et al. (2009) studied the hydrodynamics and mass transfer of gas–liquid flow in a FFMR. Once again the CO_2 absorption method was employed, with titration of the liquid phase product to determine the efficiency of the process. Al-Rawashdeh et al. (2008) developed a three-dimensional computational fluid dynamic model of a falling film microreactor based on realistic channel geometry profiles. The model was validated experimentally by the absorption of CO_2 in NaOH aqueous solution. The model allowed the authors to investigate the effects of channel fabrication precision, liquid flow distribution, gas chamber height, and hydrophilic-hydrophobic plate material on the conversion of CO_2. It was found that fabrication imprecisions and liquid maldistribution lowered the reaction conversion by approximately 2%. However, none of these previous studies have considered the effect of reactor orientation on the rate of mass transfer. The reactor orientation is considered an additional operating parameter that can be used to adjust the performance of a FFMR. In particular it has been suggested that changes to the reactor orientation can be used to modify liquid residence times for slower reactions (Hessel et al., 2008). There is a need therefore to understand the effect of reactor orientation on other operational characteristics of the apparatus, particularly the rate of mass transfer.

In this work CO_2 absorption measurements were used to quantify the effect of the degree of inclination of the reaction plate and gas flowrate in a FFMR on the liquid-side mass transfer coefficient. For the specific geometry of the microchannels used in this study, approximations for the shape and length of the liquid menisci were used to compute the mass transfer coefficients. These results were compared to the case of a flat interface and the relative differences were analysed.

2. Experimental

The FFMR used in this study is a well-known and well-studied microstructured reactor which was invented by the Institut für Mikrotechnik Mainz GmbH and has successfully shown its ability to maintain thin liquid films of up to 100 μm in thickness (Al-Rawashdeh et al., 2008). The FFMR consisted of a copper cooling plate, a stainless steel reaction plate with etched microchannels and a viewing window, fabricated from glass, which enclosed the gas headspace. The device was fabricated from 316Ti stainless steel and was equipped with a microstructured panel containing 32 microchannels of dimensions $641\ \mu m \times 257\ \mu m \times 66.4$ mm. The experimental system is shown in Fig. 1.

Carbon dioxide gas (Afrox, > 99%) was supplied in a standard cylinder. The feed gas flowrate was controlled using a thermal mass flow controller (Alicat). Inlet gas flowrates were fixed at either $50\ cm^3 \cdot min^{-1}$ and $80\ cm^3 \cdot min^{-1}$. Deionized water (conductivity 0.07 μS) was mixed with ethylene glycol (Riedel-de Haen, > 99%) to produce a 12 wt% solution and was used as the liquid feed. This ensured satisfactory wetting of the channel surface due to a lower surface tension of the liquid. In all experiments the liquid flow into the microreactor was controlled using a HPLC pump. The feed pump supplies liquid to a header. The liquid