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Reaction modelling of a microstructured falling film reactor incorporating staggered herringbone structures using eddy diffusivity concepts

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HIGHLIGHTS

- Studies of falling film microreactor with herringbone structure geometries for mass transfer enhancement.
- Suggested geometries show higher conversion for high Peclet numbers.
- Development of a new theoretical approach based on eddy diffusivity concepts.

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ABSTRACT

Falling film microreactors are ideally suited for fast exothermic reactions due to their large surface to volume ratio (up to 20,000 m^2/m^3) which greatly intensifies mass transfer. Despite the enhanced mass transfer characteristics of these reactors, mass transfer between the phases can still be the rate limiting step. To improve mass transfer, staggered herringbone structures were incorporated on the microchannel floor of falling film microreactors [1]. It was shown experimentally that reactors with herringbone structures increased CO_2 absorption in 1 M NaOH solution by up to 42%. Modelling of this system can be computationally prohibitive. This is due to its three-dimensional nature and the complexity of incorporating gas/liquid absorption and reaction with chaotic flow. For the CO_2 chemical absorption system, a pseudo 3D approach has been implemented to account for such a modelling complexity [2]. In this work, the complexity of modelling the effect of herringbone structures was simplified using an effective diffusion coefficient calculated via an eddy diffusivity approach. Good agreement between the experimental data from Ziegenbalg et al. [1] and the simulations was obtained. The simplification suggested opens the possibility to model complicated systems with minimum computational expenditure.

1. Introduction

The miniaturisation of chemical processes is becoming a popular trend in the chemical engineering field. Due to the small dimensions of microchannels, the surface to volume ratio is several orders of magnitude higher compared to conventional equipment. For this reason the rates of heat and mass transfer are typically high, leading to improvement in reaction yield and selectivity. However, for applications where the Peclet number $Pe = \frac{vd}{D}$ is large, $Pe > 10^2$, mass transfer by diffusion is slow. In this case, mass transfer can be improved by stirring the fluid transversely, disturbing the typical 1-D parabolic laminar flow profile.

The staggered herringbone micromixer proposed by Stroock et al. [3] has been shown to be effective for mixing applications [4–11]. For characterising the quality of mixing, residence time distributions (RTDs) have been studied numerically [6,12] and experimentally [13]. It has been found that RTDs for the staggered herringbone channel are narrower compared to a standard rectangular channel and that the difference reduces as the Peclet number is decreased. Another advantage of staggered herringbone channels is their efficiency in increasing mass transfer to boundaries (microchannel walls). Kirtland et al. [14] simulated mass transfer on the top wall of a channel with floor staggered herringbone structures. The simulation was performed by tracking passive tracers over a range of Peclet number with an instantaneous reaction occurring at the top wall. They found that the staggered herringbone had a higher rate of mass transfer compared to a standard

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Nomenclature

A_l	cross section of channel where liquid flows
c	liquid concentration, mol/m^3
C	herringbone cycle length
d	characteristic dimension, m
D	diffusion coefficient, m^2/s
D_{eff}	effective diffusion coefficient, m^2/s
E_D	pseudo-eddy diffusivity, m^2/s
F	flow rate, ml/min
g	acceleration due to gravity, m/s^2
I	solution ionic strength
J	mass flux across a boundary, kg/m^2 s
k	reaction constant
l	diffusion length for enhanced diffusivity calculations, m
h	parameters for gas solubilities calculations
H	Henrýs constant, mol/I atm
n	moles of species
N_{CH}	number of channels
P	pressure, Pa
Pe	peclet number
r	reaction rate, kmol/m^3 s
R	correlation of the velocity of particles
t	measured time, s
T	time period, s, or temperature, K
v	velocity, m/s
$\bar{v}$	average velocity, m/s
x	position coordinate, m
X	channel width, m
y	position coordinate, m
y_i	molar fraction
Y	liquid film thickness, m
z	position coordinate, m
z_i	valence for component i

Greek symbols

ξ	time period, s
μ	viscosity, Pa s
ρ	density, kg/m^3

rectangular channel. Additionally, other geometries (symmetric herringbone and diagonal grooves), which do not produce a chaotic flow, had mass transfer rates comparable to the herringbone channel.

Falling film microreactors are ideally suited for fast exothermic and mass transfer limited reactions. The falling film microreactor has demonstrated its successful implementation for several gas–liquid reactions. Ozonolysis [15], fluorination [16], sulfonation [17], chlorination [18], and hydrogenation [19] are just a few of these applications, some of which have been brought to pilot scale. There have also been efforts at understanding the hydrodynamic characteristics of the falling film microreactor. Zhang et al. [20] studied the flow pattern and developed an empirical correlation for the mass transfer coefficient. Commenge et al. [21] studied experimentally gas-phase residence time distributions. The RTDs were fitted to a series of continuous stirred tank reactors whose number was a function of the Reynolds number. Al-Rawashdeh et al. [22] studied the effect of the shape of the channel and its cross section on the absorption of CO_2 in sodium hydroxide solution. The falling film microreactor used in this work was developed by the Institut für Mikrotechnik Mainz GmbH (IMM). Fig. 1 shows a schematic view of its main components.

The performance of the falling film microreactor has been evaluated using the absorption of CO_2 in aqueous NaOH solution as a model reaction [2,23,24]. It was found that CO_2 is consumed within a few micrometres beyond the gas liquid interface, while OH^- shows a sharp concentration profile along the axial direction, see for example [22]. Mixing in the liquid phase occurs only by molecular diffusion. The flow in the microchannel is laminar; turbulent flows that tend to homogenise the solution are absent and molecular diffusion across the channels is dominating. To enhance transverse mixing Ziegenbalg et al. [1] reported the integration of passive micromixer structures experimentally, while Al-Rawashdeh et al. [22] calculated numerically the residence time distributions when herringbone structures were placed on the channel floors. Despite the low Reynolds numbers and hence laminar flow (Re = 3–11), the herringbone structures induced secondary transverse flows leading to chaotic flow even for an unconstrained channel with a free surface like that of a falling film. This was concluded in [22] by obtaining narrower residence time distributions and higher penetration of fluid from the free surface to the bulk for the herringbone containing channel. These results indicate strong transverse velocity components that homogenise the flow.

To model the falling film microreactor with herringbone structures incorporated in the liquid side, a multiphase 3D model is needed which can be computationally expensive. Cantu–Perez et al. [25] analysed the performance of a consecutive reaction in a layered herringbone channel where it was found that the stirring capabilities of the herringbone structures could be lumped into an effective diffusion coefficient that depends on position. A similar approach is considered in this work to simplify the numerical calculations. First, the hydrodynamics of one herringbone cycle of the liquid side is solved in three dimensions. The velocity field is then used to calculate an effective diffusion coefficient following an eddy diffusivity approach. The position-dependent effective diffusion coefficient is used to account for the enhanced transverse mass transfer induced by the herringbone structures. The reactor is modelled in two dimensions using the pseudo-3D model described in [2]. The eddy diffusivity approach described here could potentially be used to simulate complicated reaction schemes with multiple phases faster than a full 3D model. Even though this approach simplifies the modelling, it provides more detailed information than traditional residence time based models; for example in terms of concentrations as a function of position.

2. Methods and materials

2.1. Reaction system

The reaction system considered here is the absorption of carbon dioxide from a mixture of CO_2–N_2 into an aqueous solution of NaOH. This reaction was chosen since reliable kinetic data are available and extensive studies on the CO_2–NaOH reaction published so far allow us to compare reactor performance of different reactor types. The same reaction system has been investigated by Zanfir et al. [24] using a 2-D model and by Al-Rawashdeh et al. [2] using a pseudo 3-D model. The reaction kinetics and physical properties are taken from these studies. The reaction steps for CO_2 absorption in aqueous solutions of sodium hydroxide are as follows:

$$CO_{2(g)} \leftrightarrow CO_{2(l)} \quad (1)$$