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Axial dispersion, pressure drop and mass transfer comparison of small-scale structured reaction devices for hydrogenations

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ABSTRACT

"Designed Porous Structured Reactor" (DPSR) is porous stream-wise periodic structure that can serve both as static mixers and catalyst carriers. Two DPSRs with different inner diameters (A: 14 mm, B: 7 mm) were compared.

Residence time distributions were determined in an aqueous system at different Reynolds numbers using wire-mesh electrodes. Similar dispersion was observed in both structures, which was attributed to fully developed flow in DPSR A and B. Axial dispersion coefficients increased monotone with pore Péclet numbers and were in good agreement with literature.

DPSR A showed higher pressure drops due to lower porosity values than DPSR B. Pressure drops followed the Forchheimer equation using an Ergun model. The coating reduced the surface roughness and the porosity of the DPSRs' metal fabric.

Chemical mass transfer was evaluated for the hydrogenation of 2-methyl-3-butyn-2-ol (MBY) at various gas/liquid combinations. While gas–liquid mass transfer was determined as limiting process step, liquid–solid mass transfer was ruled out. Superior gas–liquid mass transfer coefficients were determined for DPSR B. Differences in mass transfer rates DPSR A and B were attributed to the ratios of inner diameter to the pore size and the porosities. A Stanton correlation was suggested to estimate gas–liquid mass transfer coefficients.

1. Introduction

A porous stream-wise periodic structure, that can serve both as static mixer was well as a catalyst carrier, termed "Designed Porous Structured Reactor" (DPSR) with an inner diameter of 7 mm and a length of 20 cm was suggested by Hutter et al. [1] as a viable alternate reactor for fast exothermic reactions to common packed bed reactors from a fluid dynamic point of view.

Hutter et al. [2] compared axial dispersion coefficients 20 cm DPSR elements to 20 and 30 ppi copper metal foams with lengths of 20 cm and 1 m. They used a horizontal flow setup with commercial plate electrodes (for 1 m copper foams) and self-made circular graphite electrodes in the tubing side walls (for 20 cm DPSRs and foams), without any additional flow premixing elements. While a large increase in dispersion with media length was observed, reported coefficients were in good agreement with literature for packed beds and common packing materials. As reported by Han et al. [3], it is well known from literature that axial dispersion coefficients increase asymptotically to a final value with increasing column lengths. This indicates that Hutter et al. [2] applied too short measurement sections to determine fully developed dispersion coefficients in DPSRs.

Later, Häfeli et al. [4] studied the effect of flow pre-development on axial dispersion coefficients in similar DPSRs as used by Hutter et al. [2]. For this, two identical 20 cm DPSR elements were installed in a horizontal setup. Dispersion was measured with self-made wire mesh tomographs over the second *DPSR* element, while the first one was used for premixing. Häfeli et al. [4] found significantly larger dispersion coefficients using a premixer compared to reports from Hutter et al. [2] and concluded that the difference was due to flow development and build-up of turbulence intensity over the first DPSR element.

Butscher et al. [5] demonstrated that a build-up of turbulence in DPSRs occurs as fluid passes optically with Particle Induced Velocimetry (PIV) in system of refractive index matched structure and process fluid. Butscher determined that a fully developed flow is reached after about two periodic units.

Recently, Pd coated DPSRs were tested by Elias et al. [6] in respect to their applicability as chemical reactors for the selective hydrogenation of 2-methyl-3-butyn-2-ol (MBY) to

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Nomenclature

Abbreviations

Al_2O_3	aluminum oxide
CAD	computer aided design
D	C_{10} dimers
DPSR	Designed Porous Structured Reactor
FIC	Flow Indicator Control
KCl	potassium chloride
H_2	hydrogen
H_2O	water
MBY	2-methyl-3-butyn-2-ol
MBE	2-methyl-3-buten-2-ol
MBA	2-methyl-3-butan-2-ol
Pd	palladium
PIV	Particle Induced Velocimentry
ppi	pores per inch
RTD	residence time distribution
SLS	selective laser sintering
WMS	wire-mesh sensor
ZnO	zinc oxide

Roman symbols

a_p	viscous factor in Eq. (10) [Pa s m^{-2}]
a	specific gas–liquid interface area [m^2 m^{-3}]
b_p	inertial factor in Eq. (10) [Pa s^2 m^{-3}]
A	specific surface area [m^2 m^{-3}]
A_s	specific surface area of structure of fluid per volume of solid [m^2 m^{-3}]
A_{solid}	surface area obtained from CAD [m^2]
c	molar concentration [mol m^{-3}]
$\overline{c^*_{H_2}}$	mean logarithmic concentration of dissolved hydrogen [mol m^{-3}]
$c^*_{H_2}$	concentration of dissolved hydrogen [mol m^{-3}]
Ca	Carberry number [–]
C_f	friction factor [–]
Da_{II}	second Damköhler number [–]
D_{ax}	axial dispersion coefficient [m^2 s^{-1}]
D, D_m	molecular diffusion coefficient [m^2 s^{-1}]
d_b	bubble diameter [m]
d_i	inner diameter [m]
d_o	out diameter [m]
d_p	pore diameter [m]
Eu	Euler number [–]
$E(t)$	residence time distribution [s]
$F(t)$	cumulative residence time distribution [s]
F	volumetric flow rate [m^3 s^{-1}]
H_{H_2}	Henry's law constant [Pa m^3 mol^{-1}]
$k_{r,1}$	first order reaction rate constant [m^3 MBY mol^{-1} Pd s^{-1}]
k_L	gas–liquid mass transfer coefficient [m s^{-1}]
k_La	volumetric gas–liquid mass transfer coefficient [s^{-1}]
k_{ls}	liquid–solid mass transfer coefficient [m s^{-1}]
L	length [m]
n_{CAD}	number of parallel channels to approximate DPSRs
P	reactor pressure [bar]
ΔP	reactor pressure drop [Pa]
$\Delta P/L$	pressure drop [Pa m^{-1}]
Pe_p	pore Péclet number
r_1	first order reaction rate [mol m^{-3} s^{-1}]
Re_p	pore Reynolds number [–]
R_{exp}	experimentally determined volumetric reaction rate [mol m^{-3} s^{-1}]
S	selectivity [–]
Sc	Schmidt number [–]
Sh	Sherwood number [–]
St	Stanton number [–]
T	reactor temperature [°C]
t	time [s]
V	reactor volume [m^3]
v	interstitial flow velocity [m s^{-1}]
v_b	interstitial bulk velocity [m s^{-1}]
v_s	superficial bulk velocity [m s^{-1}]
X	conversion [–]
x	spatial coordinate [m]

Additional sub- and superscripts

0	reactor inlet
1	reactor outlet
A	DPSR A
B	DPSR B
device	referring to a component consisting of a DPSR and two tube connectors
exp	experimentally determined
G	gas phase
in	reactor inlet
L	liquid phase
out	reactor outlet
p	pore
pulse	referring to a concentration pulse
s	surface
step	concentration step increase in Eq. (3)
theo	theoretically determined

Greek letters

α	Ergun parameter in Eq. (11)
β	Ergun parameter in Eq. (12)
ϵ_s	reactor porosity [–]
ν	kinetic viscosity [m^2 s^{-1}]
μ	viscosity of the reaction mixture [Pa s]
τ	mean residence time [s]
θ	catalyst loading [mol Pd m^3 MBY]
ρ	density [kg m^{-3}]
Π	dimensionless group

2-methyl-3-butyn-2-ol (MBE), as shown in Fig. 1. This reaction was chosen for its industrial relevance as selective hydrogenations, especially of C—C triple bonds, are often mentioned as key steps in the large scale production chemicals such as vitamin intermediates [7]. The reaction itself is well known and was previously studied by various authors, e.g. Semagina et al. [8] and Grasemann et al. [9]. DPSRs were found to be viable tools for process intensification compared to batch reactors, especially in terms of selectivity and yield. However, large mass transfer limitations were observed.

Depending on the specific application and economic scale of the analyzed industrial process, a single mili scale reaction devices can reach the boundaries of its production capacities. Increases in production capacity of such devices can easily achieved by repetition of identical process equipment commonly known as "numbering up", since single channel setups are relatively straight forward in their characterization and operation. To avoid unnecessary repetition of utility devices, like pumps for instance, flow splitting devices are often incorporated to ascertain even flow supply to each separate reaction device. Examples of such concepts are presented by Riekert [10], Kashid et al. [11] and Müller et al. [12]. Alternatively to