



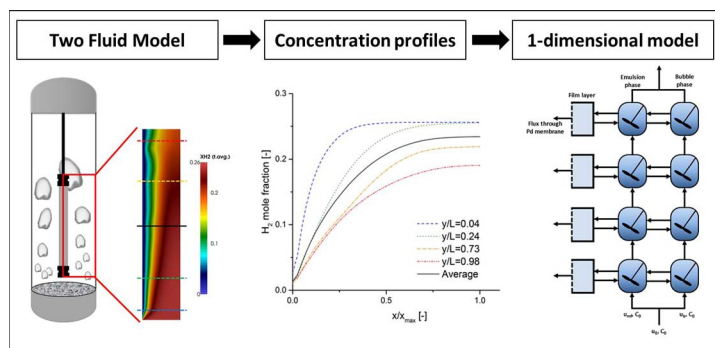
On concentration polarization in fluidized bed membrane reactors



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GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:

Fluidized bed
Pd membrane
Concentration polarization
TFM

ABSTRACT

Palladium-based membrane-assisted fluidized bed reactors have been proposed for the production of ultra-pure hydrogen at small scales. Due to the improved heat and mass transfer characteristics inside such reactors, it is commonly believed that they can outperform packed bed membrane reactor configurations. It has been widely shown that the performance of packed bed membrane reactors can suffer from serious mass transfer limitations from the bulk of the catalyst bed to the surface of the membranes (concentration polarization) when using modern highly permeable membranes. The extent of concentration polarization in fluidized bed membrane reactors has not yet been researched in detail. In this work, we have quantified the concentration polarization effect inside fluidized bed membrane reactors with immersed vertical membranes with high hydrogen fluxes. A Two-Fluid Model (TFM) was used to quantify the extent of concentration polarization and to visualize the concentration profiles near the membrane. The concentration profiles were simplified to a mass transfer boundary layer (typically 1 cm in thickness), which was implemented in a 1D fluidized bed membrane reactor model to account for the concentration polarization effects. Predictions by the TFM and the extended 1D model showed very good agreement with experimental hydrogen flux data. The experiments and models show that concentration polarization can reduce the hydrogen flux by a factor of 3 even at low H_2 concentrations in the feed (10%), which confirms that concentration polarization can also significantly affect the performance of fluidized bed membrane reactors when integrating highly permeable membranes, but to a somewhat lesser extent than packed bed membrane reactors. The extraction of hydrogen also affects the gas velocity and solids hold-up profiles in the fluidized bed.

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<http://dx.doi.org/10.1016/j.cej.2017.09.045>

Received 3 January 2017; Received in revised form 11 August 2017; Accepted 7 September 2017

Available online 11 September 2017

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Nomenclature

A	area (m ²)
c_{1,c_2}	constants in frictional stress model (-)
B	exchange of fluctuation energy (kg m ⁻¹ s ⁻³)
C	concentration (mol m ⁻³)
C_d	drag coefficient (-)
D	diffusion/dispersion coefficient (m ² s ⁻¹)
d	diameter (m)
E_a	activation energy (J mol ⁻¹)
e	coefficient of restitution (-)
f	fraction (-)
Fr	constant in frictional stress model (N m ⁻²)
g	gravitational acceleration (m s ⁻²)
g_0	radial distribution function (-)
H	height (m)
\bar{I}	unit tensor (-)
J	membrane flux (mol m ⁻² s ⁻¹)
K	mass transfer coefficient (m s ⁻¹)
k_d	mass transfer coefficient bulk to membrane (m s ⁻¹)
M_w	Molecular weight (kg mol ⁻¹)
N	flux (mol m ⁻² s ⁻¹)
P	partial pressure (Pa)
P_m	permeability (mol m ⁻¹ s ⁻¹ Pa ^{-0.5})
$P_{m,0}$	permeation constant (mol m ⁻¹ s ⁻¹ Pa ^{-0.5})
p	pressure (Pa)
Q_{pd}	permeance (mol m ⁻² s ⁻¹ Pa ^{-0.5})
R	universal gas constant (J mol ⁻¹ K ⁻¹)
r	radial position (m)
Re	Reynolds number (-)
S	strain rate (s ⁻¹)
S	source term (kg m ⁻³ s ⁻¹)
Sh	Sherwood number (-)
t	time (s)
t_m	membrane thickness (m)
T	temperature (K)
u	velocity (m s ⁻¹)
V	volume (m ³)
X	molar fraction (-)
Y	mass fraction (-)
z	axial position (m)

Greek letters

α	volume fraction (-)
β	interphase drag coefficient (kg m ⁻³ s ⁻¹)
γ	dissipation of granular energy (kg m ⁻¹ s ⁻³)
δ	film layer thickness (m)
θ	granular temperature (m ² s ⁻²)

κ	conductivity of granular energy (kg m ⁻¹ s ⁻¹)
λ	bulk viscosity (kg m ⁻¹ s ⁻¹)
μ	Shear viscosity (kg m ⁻¹ s ⁻¹)
ρ	density (kg m ⁻³)
τ	shear stress tensor (N m ⁻²)
ϕ^{fric}	angle of internal friction (°)

Subscripts & superscripts

avg	average
b	bubble
bc	bubble to cloud
be	bubble to emulsion
bulk	bulk
ce	cloud to emulsion
cell	cell(s)
e	emulsion
fric	frictional
g	gas
h	hydraulic
m	membrane
max	maximum
mf	minimum fluidization
min fr.	minimum friction
mol	molecular
n	number of CSTRs
p	particle
pp	particle-particle
pw	particle-wall
perm	permeate
r	radial
reac	reactor
rise	rise
s	solid
sim	simulation
T	transposed
tot	total

Abbreviations

CFD	computational fluid dynamics
CSTR	continuous stirred tank reactor
FBMR	fluidized bed membrane reactor
KTGF	kinetic theory of granular flow
PSA	pressure swing adsorption
SMR	steam methane reforming
TFM	two-fluid model
WGS	water gas shift

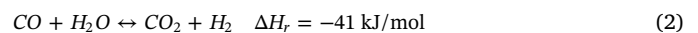
1. Introduction

Currently, hydrogen is mainly produced on large scale via steam reforming of methane (SMR) [1]. In this process, methane is first reformed with steam (Eq. (1)) in high temperature multi-tubular packed bed reactors. In a second step the carbon monoxide is converted via the water gas shift (WGS) reaction (Eq. (2)) in packed bed reactors. Typically, a two stage WGS is used to take advantage of fast reaction rates at high temperatures (450 °C) and higher equilibrium conversions at lower temperatures (200 °C). Finally, the hydrogen produced is further purified using pressure swing adsorption (PSA).

Steam methane reforming reaction (SMR):



Water gas shift reaction (WGS):



The equivalent hydrogen efficiency of the whole process is approximately 80% thanks to steam/electricity export [2]. The heat integration between the different stages becomes more complicated at smaller scales, while heat export cannot be realized in distributed hydrogen production applications. For this reason the system becomes inefficient and uneconomical at smaller scales. The cost of the hydrogen produced at large scale is around 0.2 €/Nm³ while it increases up to 0.4–0.5 €/Nm³ at smaller scales [2].

The efficiency of the hydrogen production via methane reforming can be increased by integrating hydrogen production and separation in a single multifunctional reactor. This can be achieved by using perm-

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