



Experimental and Computational Investigation of Gas Separation in Adsorbent-coated Microchannels



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HIGHLIGHTS

- Experimental and computational study of gas separation in adsorbent-coated microchannels.
- PLOT columns with zeolite 5A used to study sequential breakthrough of N₂ and CO₂.
- Model predictions for adsorption time and temperature rise agree well with data.
- Experiments on custom microchannels show excellent agreement with predictions.

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ABSTRACT

An experimental and computational investigation of gas separation in adsorbent-coated microchannels is performed. Experiments on porous-layer-open-tubular (PLOT) columns containing zeolite 5A using a ternary mixture of helium (He), nitrogen (N₂) and carbon dioxide (CO₂) show sequential breakthrough of N₂ and CO₂ and gradual saturation of adsorption sites with trace water. Multiple tests conducted for the adsorbent breakthrough investigation result in determination of adsorbent layer properties. The heat and mass transfer model results for a pressure drop range of 5–55 kPa and channel lengths from 1 to 4 m are found to be in reasonable agreement with the data with an average absolute deviation (AAD) of 14% for adsorption time and 41% for temperature rise (ΔT). The larger error in ΔT is thought to be due to uneven adsorbent loading of the PLOT columns. Therefore, the adsorption experiments are also conducted with custom-made adsorbent-coated microchannels with known adsorbent mass and layer thickness. The AADs for adsorption time and ΔT then decrease to 4% and 25%, respectively, indicating that the overall gas separation process is described well by the adsorption stage models, with local variation due to fabrication variability.

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1. Introduction

Adsorption-based gas separation processes utilized for natural gas purification and carbon dioxide capture from flue gas have been shown to benefit from the use of adsorbent-coated microchannels, which yields a greater process capacity, and competitive product purities and gas recoveries when compared with other conventionally used geometries (Pahinkar et al., 2017). Due to high heat and mass transfer coefficients in microchannels, the execution of the various stages of the adsorption-based gas separation cycle is faster than that of adsorbent beds, and a sharp wave

front is maintained for the adsorption and regeneration stages of the cycle.

Pressure swing adsorption (PSA) in microchannels is found to be effective because of unimpeded transmission of pressure waves along its length. In a PSA process, the depressurization in microchannels is found very effective when the stage performance is compared with the fixed bed depressurization processes in the literature. Pahinkar et al. (2015) conducted a computational investigation of depressurization in microchannels and reported up to four times greater CO₂ removal capacities, when compared with equilibrium-based CH₄-CO₂ separation studies by Kapoor and Yang (1989) using zeolite 5A and N₂-CO₂ separation studies by Shen et al. (2011) using activated carbon. They also predicted up to 25 times greater process capacity with adsorbent-coated microchannels as compared with the PSA process investigated by Krishnamurthy et al. (2014).

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Nomenclature

Symbols

A	area [m ²]
a, b	line fitting coefficients in type-B uncertainty
A_1, A_2, \dots	DSL coefficients [mol kg ⁻¹ or mol kg ⁻¹ K]
b, B	DSL coefficients [kPa ⁻¹]
C	concentration [kg m ⁻³]
C_A	adsorbed concentration [mol m ⁻³]
$C_{A,Eq}$	equilibrium concentration [mol m ⁻³]
C_P	heat capacity [J kg ⁻¹ K ⁻¹]
C_T	valve actuation constant [s ⁻¹]
C_V	valve flow coefficient [kg s ⁻¹ kPa ⁻¹]
d, D	DSL coefficients [kPa ⁻¹]
d_{pore}	macropore (void) size in adsorbent layer [m]
D_A	axial dispersion coefficient [m ² s ⁻¹]
$D_{crystal}$	intra-crystalline diffusion coefficient [m ² s ⁻¹]
D_{eff}	effective diffusion coefficient [m ² s ⁻¹]
D_h	hydraulic diameter [m]
IH	heat of adsorption [J mol ⁻¹]
E	activation energy [J mol ⁻¹]
eps	gas channel surface roughness [m]
f	friction factor [-]
h	enthalpy density [J m ⁻³]
h_{free}	free or natural convection coefficient [W m ⁻² K ⁻¹]
h_m	mass transfer coefficient [m s ⁻¹]
h_T	heat transfer coefficient [W m ⁻² K ⁻¹]
k	thermal conductivity [W m ⁻¹ K ⁻¹]
K_{LDF}	linear driving force constant [s ⁻¹]
Kn	Knudsen number [m m ⁻¹]
L	length [m]
M	mass [kg]
\dot{m}	mass flowrate [kg s ⁻¹]
\dot{M}	mass collection rate [kg kg ⁻¹ hr ⁻¹]
MF	adsorbent loading [kg kg-polymer ⁻¹]
MW	molecular weight [kg kmole ⁻¹]
N	sample size [-]
N_{nodes}	number of nodes [-]
P	pressure [kPa]
Pe	Peclet number [-]
Pe_{grid}	grid Peclet number [-]
$Peri$	channel perimeter [m]
$Q_{B/D}$	heat of adsorption in DSL equation [J mol ⁻¹]
R	radius [m]
$r_{crystal}$	adsorbent crystal size [m]
Re	Reynolds number [-]
R	overall transport resistance [m ⁻² s or m-K W ⁻¹]
Ru	Universal gas constant [J mol ⁻¹ K ⁻¹]
t	time [s]
th	adsorbent layer thickness [m]
th_{FS}	monolith wall thickness [m]

T	temperature [K or °C]
u	velocity [m s ⁻¹]
U	total uncertainty
U_A	type-A uncertainty
U_B	type-B uncertainty
U_g	nodal internal energy density [J m ⁻³]
V	volume [m ³]
VI	virtual instrument
V_p	valve position [-]
x	mole fraction [-]
X	sensor reading
\bar{X}	mean sensor reading
y	mass fraction [-]
Y_i	sensor reading used for type-B calibration
z	axial position [m]

Greek

ε	adsorbent matrix void fraction [m ³ m ⁻³]
ε_r	radiation emissivity [-]
λ	mean free path [m]
μ	viscosity [kg m ⁻¹ s ⁻¹]
ρ	density [kg m ⁻³]
σ	Stefan-Boltzmann constant [W m ⁻² K ⁻⁴]
τ	tortuosity factor [m m ⁻¹]
ω	adsorbent volume fraction [m ³ m ⁻³]

Superscripts and subscripts

ads	adsorption
binder	silica binder
conv	convection
FS	fused silica
g	microchannel
h	hydraulic
Heat	related to heat transfer
i	assigned species (He, N ₂ , CO ₂)
in	inlet
ins	insulation
Knudsen	Knudsen diffusion
Mass	related to mass transfer
mid	midpoint
mix	mixture
ordinary	molecular or ordinary diffusion
out	outlet
rad	radiation
total	total (net) diffusion
v	valve
w	adsorbent layer or wall

PSA processes utilizing fixed adsorbent beds are commonly used for gas separation applications, as bed-based temperature swing adsorption (TSA) processes are deemed difficult to implement due to the low thermal conductivity of the adsorbent material as well as due to the presence of void spaces in the bed (Riemer et al., 1994). Therefore, heat duties and process times are expected to be large and heat distribution is found to be ineffective (Moate and LeVan, 2010), negating the large-scale viability of such a system. However, TSA processes in microchannels are enhanced due to high heat and mass transfer coefficients and the small thermal mass of the adsorbent. Lively et al. (2009) demonstrated the fabrication of sub-millimeter mixed matrix membrane hollow fibers (MMM), which combine the advantages of polymeric membranes

and adsorbent particles (Bernardo et al., 2009), thereby opening up potential avenues for revisiting the use of TSA processes for gas separation. These MMMs can be fabricated within frameworks already available for making conventional polymeric membranes, emphasizing the low capital costs associated with their use. Pahinkar et al. (2017) investigated a TSA process for natural gas purification using an adsorbent-polymer matrix based on the hollow fibers reported by Lively et al. (2009) with the flow of working fluids and HTFs through separate, alternating parallel channels. They found that process capacity was improved over bed-based PSA processes by up to an order of magnitude at similar values of purity and recoveries. Subsequently, by sending the process gases and the heat transfer fluid (HTF) through the same

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