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A modeling study of module arrangement and experimental investigation of single stage module for physical absorption of biogas using hollow fiber membrane contactors

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ABSTRACT

In the present work, the effect of module arrangement on the physical absorption of CO_2 and H_2S in water at high operating pressure using 2 different types of hollow fiber membrane contactor (HFMC) including polyvinylidenefluoride (PVDF) and polytetrafluoroethylene (PTFE) was studied for biogas upgrading application. The simulation was performed at various liquid velocities (0.12–2.0 m/s), pressure differences between liquid and gas phases (0.125–1.0 bar for PVDF and 1.0–5.0 bar for PTFE), operating pressures (1–30 bar) under four different module arrangement scenarios, i.e., (i) single stage module, (ii) multistage module, (iii) multistage module with splitting liquid ($\alpha = 0.2$ and 0.4) and (iv) multistage module with recycle liquid ($\varphi = 0.5$ and 1). The modeling results predicted the significant improvement of CO_2 and H_2S removal performances when the multistage module was applied for both HFMCs. To obtain the highest removal performance, the suitable liquid velocity, pressure difference between liquid and gas phases and the liquid flow pattern of the module arrangement are the key concerns. For PVDF, the multistage module with splitting liquid ($\alpha = 0.2$) provides the highest performance at the liquid velocity of 0.12 m/s, pressure difference of 0.125 bar and the operating pressure of 30 bar. For PTFE, at the liquid velocity of 2.0 m/s and the pressure difference of 1.0 bar, the multistage module with recycle liquid ($\varphi = 1$) gives the highest performance at the operating pressure of 1–20 bar while the multistage module with splitting liquid ($\alpha = 0.4$) provides the highest performance at the operating pressure of 30 bar.

1. Introduction

In recent decades, biogas production and utilization for energy generation tend to increase continuously. Generally, it primarily comprises of 30–40% CO_2, 60–70% CH_4 and the trace of H_2S depending on the source of biogas [1–4]. CO_2 and H_2S are acid gas impurities that impact seriously on health and also reduce biogas heating value [2,5,6]. Therefore, biogas upgrading is an imperative process to remove CO_2 and H_2S from the raw biogas to achieve the high quality of biogas and avoid the corrosion problems with process equipment [7,8]. Hollow fiber membrane contactor (HFMC) which integrates conventional absorption and membrane process has attracted attention as an alternative technology for CO_2 and H_2S removal by the literature [3,9–12]. Hydrophobic hollow fiber membrane, e.g., polyvinylidenefluoride (PVDF), polytetrafluoroethylene (PTFE), is typically employed in the hollow fiber membrane contacting process to divide the contact between gas and liquid phases, to avoid the dispersion of one phase into another phase [2,13–16].

To improve the performance of membrane contacting process, physical and chemical absorption of pressurized CO_2 and H_2S were previously studied [17–19]. Marzouk et al. [20] reported the experimental results of simultaneous absorption of CO_2 and H_2S from 5% CO_2, 2% H_2S in balance of CH_4 at high pressure using polytetrafluoroethylene (PTFE) and polytetrafluoroethylene-*co*-perfluorinated alkyl vinyl ether (PFA) hollow fiber membranes. They reported that the increase of inlet gas pressure enhanced the CO_2 and H_2S fluxes for both physical and chemical absorption processes. Kang et al. [9] investigated the removal of high concentration CO_2 from natural gas using activated

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Nomenclature

$C_{i,in}$	inlet concentration of component i in gas phase [mol/m^3
$C_{i,membrane}$	concentration of component i in membrane [mol/m^3
$C_{i,out}$	outlet concentration of component i in gas phase [mol/m^3]
$C_{i,shell}$	concentration of component i in gas phase [mol/m^3]
$C_{i,tube}$	concentration of component i in liquid phase [mol/m^3]
$D_{i,g-membrane}$	molecular diffusivity of component i in gas phase within membrane [m^2/s]
$D_{i,l-membrane}$	molecular diffusivity of component i in liquid phase within membrane, m^2/s]
$D_{i,shell}$	molecular diffusivity of component i in gas phase [m^2/s]
$D_{i,tube}$	molecular diffusivity of component i in liquid phase [m^2/s]
d_i	inner fiber diameter [m]
$d_{i,shell}$	inner diameter of shell [m]
d_o	outer fiber diameter [m]
$d_{o,tube}$	outer diameter of tube [m]
$f(r)$	log-normal distribution function [m^{-1}]
H_i	Henry's constant of component i [dimensionless]
L	fiber length [m]
n	mole of gas [mol]
n_p	pore number [dimensionless]
P_G	pressure in gas phase [bar]
P_L	pressure in liquid phase [bar]
P_{L0}	inlet liquid pressure [bar]
$P_{L,out}$	outlet liquid pressure [bar]
ΔP_{L-G}	different or trans-membrane pressure [bar]
$\Delta P_{L-G,out}$	different or trans-membrane pressure at the outlet liquid [bar]
$\Delta P_{L-G,wetting}$	wetting pressure [bar]
$\Delta P_L(z)$	pressure drop in liquid phase along the membrane module [bar]
$Q_{G,in}$	inlet gas volumetric flow rate [m^3/s]
$Q_{G,out}$	outlet gas volumetric flow rate [m^3/s]
r	membrane pore radius [m]
r_i	critical pore radius [m]
r_m	mean or average membrane pore radius [m]
r_{max}	maximum membrane pore radius [m]
r_{min}	minimum membrane pore radius [m]
r_p	membrane pore radius [m]
$\langle r_i \rangle$	average critical pore radius [m]
R	gas constant [m^{3}atm/mol K]
T	temperature [K]
T_b	normal boiling point [K]
v_g	gas velocity [m/s]
v_l	liquid velocity [m/s]
V	volume of gas [m^3]
V_b	liquid molar volume at normal boiling point [cm^3/g mol]
V_m	total membrane pore volume [m^3]
V_w	liquid-filled pore volume [m^3]
$V_{z,shell}$	velocity of gas from the convection in shell side [m/s]
$V_{z,tube}$	velocity of water from the convection in tube side [m/s]
x^*	wetting ratio [dimensionless]
$\langle x^* \rangle$	average wetting ratio [dimensionless]
z	local length [m]
Z	compressibility factor [dimensionless]
Z^0	functions compressibility-factor correlation [dimensionless]
Z^1	generalized compressibility-factor correlation [dimensionless]

Greek symbols

ε	membrane porosity [dimensionless]
φ	packing density [dimensionless]
γ	surface tension of liquid [mN/m]
θ	contact angle [degree]
σ	geometric standard deviation [dimensionless]
τ	tortuosity of membrane [dimensionless]
ω	acentric factor [dimensionless]

methydiethanolamine (aMDEA) with piperazine as an absorbent. They found that CO_2 removal efficiency and CH_4 loss increased with the increasing pressure or reducing feed gas flow rate. It was also revealed that the overall mass transfer coefficient decreased with the increasing pressure due to the decrease of gas diffusivity. Al-Mazouqi et al. [21] presented that the high pressure long-term absorption of CO_2 and H_2S using PFA hollow fiber membrane were stable during the operation period (7 weeks) when 30 wt% K_2CO_3 + 1 wt% DEA solvent was employed as an absorbent. It has been known that the penetration of absorbents into membrane pores, called membrane wetting, is also an important problems of the membrane contactors since the membrane wetting directly increases the mass transfer resistance in membrane [22–25]. Khaisri et al. [26] developed the mathematical model to investigate the influence of partial wetting on gas absorption performance using PTFE membrane and MEA as an absorbent. They found that the increase of percent wetting decreased the absorption flux and overall mass transfer coefficient. The experimental and modeling study of CO_2 removal by single and mixed amine using hollow fiber membrane module is also reported by Iliuta et al. [27] who found that the absorption performance is significantly decreased with membrane wetting. This is supported by the study of Goyal et al. [28] who proposed the model for the absorption of CO_2 using polypropylene hollow fiber membrane considering the influence of partially-wetting. Boributh et al. [29] proposed the analytical solution for the calculation of wetting ratio. They reported that the suitable operating conditions including liquid velocity and pressure difference between gas and liquid lines should be considered to decrease the membrane wetting for the highest performance of the absorption using HFMC. Another alternative method to improve the performance of HFMC is the design of module configuration. This approach has been previously reported to be able to decrease the influence of membrane wetting [30]. The proper liquid velocity and flow pattern for the multistage module are known to be important factors. Nevertheless, until now, only a few works have reported the effect of module arrangement on the absorption of CO_2 using HFMC [30,31]. Moreover, from our knowledge, the absorption efficiency of biogas using HFMC with different module arrangement at high operating pressure has not been investigated.

The aim of this work is to simulate the physical absorption of CO_2 and H_2S from biogas at high operating pressure using two different types of HFMC including PVDF and PTFE. It should be noted that although PVDF membrane is generally known to have wetting problems, the current cost of PVDF membrane is significantly cheaper than the PTFE membrane [32]. Therefore, it is challenge to study the performance of using PVDF and PTFE membranes. The average membrane wetting at high operating was analytically considered based on log-normal distribution function. The effects of pressure difference between liquid and gas phases, liquid velocity and operating pressure on CO_2 and H_2S removal were studied. From the modeling results, the comparison of single stage and multistage module with different splitting and recycle ratios were presented.