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Journal of Natural Gas Science and Engineering

journal homepage: www.elsevier.com/locate/jngse

Stripping of CO₂ from different aqueous solvents using PVDF hollow fiber membrane contacting process



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A R T I C L E I N F O

Article history: Received 13 July 2014 Received in revised form 12 October 2014 Accepted 13 October 2014 Available online 30 October 2014

Keywords: Amine solutions CO₂ stripping Membrane contactors PVDF hollow fiber

ABSTRACT

The stripping of carbon dioxide from liquid absorbents is an important task in the operation of gas —liquid membrane contacting processes. In order to gain a better understanding on the role of various absorbents on CO₂ stripping, potassium glycinate (PG), monoethanolamine (MEA), diethanolamine (DEA), and 2-amino-2-methyl-1-propanol (AMP) were applied as absorbent/stripping solutions. The membrane used for the experiments was hollow fiber polyvinylidenefluoride (PVDF) membrane fabricated via thermally induced phase separation method. The performances of various amine solutions on the CO₂ stripping capability were investigated. CO₂ stripping experiments revealed that regardless of type of solvent the CO₂ stripping flux and efficiency rapidly increases with liquid temperature, pressure and initial CO₂ concentration. In addition, the gas—liquid contact time was a key factor to enhance the stripping flux at low temperature while liquid phase boundary layer thickness and associated mass transfer resistance is important at elevated temperatures. Moreover the comparison stripping experiments between shell and tube side liquid flow revealed that for low packing density tube side flow gives better stripping performance.

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1. Introduction

One of vital component of the world's supply of energy that has fulfilled economic viability and environmental sustainability is natural gas. In 2014, natural gas supplied 23.7% of the world's energy demand and the volume of natural gas consumption grew by 1.4% (BP, 2014). Although natural gas is mostly considered as a "clean" fuel as compared to other fossil fuels, the natural gas found in reservoirs deposit is not necessarily "clean" and free of impurities. Natural gas consists primarily of methane as the prevailing element but it also contains considerable amounts of contaminating compounds of CO₂, N₂, Hg, He, H₂S. Due to its acidic character, the presence of CO₂ can lead to corrosion in equipment and pipelines. Also CO₂ present in natural gas will reduce the heating value of the gas. So the growth in the global demand for natural gas has led to large scale removal of CO₂ from a gas stream as an important industrial operation. Conventionally, this sweetening i.e., the removal of acidic gases is accomplished by gas-liquid absorption-stripping processes using aqueous solutions of alkanolamines. The most commonly used amines include MEA,

DEA, MDEA (Rufford, T.E. et al., 2012; Huang, H.Y. et al., 2003; Cavenati, and Grande, 2006). Though a mature technology being applied extensively today, this gas absorption process is highly energy intensive for regeneration of the solvents and is also plagued by corrosion problems. Membrane technology is a promising method to replace the conventional absorption technology. It has a high energy efficiency, is easy to scale-up because of its modular design and it has a high area-to-volume ratio (Simons, 2010). The utilization of hollow fiber gas liquid membrane contactor modules for separation applications has attracted great interest over the past decade (Bhide and Stern, 1993; Hoff, K.A. et al., 2004). In hollow fiber gas liquid membrane contactor absorption/stripping processes the porous hydrophobic membrane acts as a fixed interface between the gas and the liquid phase without dispersing one phase into another. If the liquid phase will made to flow through the hollow fiber membrane lumen side, then the gas phase will made to flow through the other side and vice versa. In the case of CO₂/CH₄ separation, CO₂ diffuses from the feed gas side through the membrane and is then absorbed in the selective absorption liquid. The loaded liquid circulates from the absorber to the stripper, a second membrane contactor in which stripping of CO_2 occurs (Fig. 1).

Hollow fiber gas liquid membrane contactor as CO_2 absorber has been extensively studied by several researchers and exciting

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Fig. 1. Membrane gas absorption/stripper process.

experimental and theoretical results have been reported (Yan, S.-p. et al., 2007; Lin, S.-H. et al., 2008). Despite the fact that solvent regeneration is responsible for the major cost component in gas separation processes due to energy consumption (Michael, 2010), the studies on CO₂ stripping from physical and chemical liquid absorbents using membrane contactor have started recently and there are a few reports documented in the open literature (Naim et al., 2014; Koonaphapdeelert, 2009; Khaisri, S. et al., 2011; Mansourizadeh and Ismail, 2011; Simioni, et a., 2011; Portugal et al., 2007). In CO₂ membrane stripper the CO₂ loaded liquid circulates from the absorber and then CO_2 is desorbed to a sweep gas. Generally nitrogen used as sweep gas. It was found that the sort of membrane, nature of solvent liquid and process parameters such as liquid flow rate, rich solution temperature, and transmembrane pressure and module configuration are the key parameters for the stripping operation. In order to compare the stripping performance of different membranes, CO₂ stripping flux at various operating conditions is summarized in Table 1. In most of the works the removal efficiency was not mentioned. Few researchers achieved the removal efficiency, at least higher than fifty percent at elevated temperature by using surface modified fibers. As CO₂ stripping for regeneration of the liquid absorbents in gas separation industries is the most energy consuming process due to high heat requirement, further investigations to develop the technology is deemed necessary.

In the present study the home made 28% PVDF hollow fiber gas liquid membrane contactor modules were used to compare the CO_2 stripping performance of primary, secondary, hindered amines and

Comparison of CO₂ stripping flux of hollow fiber membranes.

F able 2 Spinning conditions.	
Dope composition	28% PVDF
Bore fluid and solvent	Triacetin
Dope solution flow rate	5 g/min
Bore fluid flow rate	7 g/min
Take up winder	10 rpm
Spinneret, OD/ID	1.5/1.1 mm
Extrusion temperature	170 °C
Quenching bath temperature	25 °C
Air gan	5 cm

amine salts. Also the effect of vital operating conditions such as liquid and gas phase flow rates, transmembrane pressure, liquid phase temperature and initial CO₂ concentrations on stripping flux and efficiency was investigated.

2. Experimental work

2.1. Reagents and materials

Polyvinylidene fluoride (PVDF) (Solef[®]6020/1001) was purchased from Solvay Company, France. Glycerol triacetate (triacetin), ethanol, monoethanolamine (MEA), diethanolamine (DEA), 2amino-2methyl-1-propanol (AMP), potassium hydroxide and glycine were purchased from Sigma Aldrich, Germany. All materials were with purity more than 99%. All chemicals were used as received without further purification. Nitrogen (99.99%) and CO₂ (99.99%) gas cylinders were purchased from Sharjah Oxygen Company, UAE. Different types of epoxy were used: Araldite 5 min rapid, Fevicol 5 min rapid and Devcon[®] 5 Minute Epoxy and Devcon[®] 5 Minute Epoxy Gel.

2.2. Preparation of hollow fiber membrane module

Hollow fiber membranes were fabricated using 1000 g of dope solution consist of 28% PVDF polymer and 72% triacetin liquid solvent. Thermally induced phase separation (TIPS) method was used for this purpose (Rahbari-Sisakht, M. et al., 2014; Ghasem et al., 2011, 2012, 2013). The dope solution was prepared and extruded at temperature of 170 °C. The spinning conditions are given in Table 2. The properties of the fabricated hollow fiber membranes are shown in Table 3. Shell and tube type module was

Table 1

Membrane	Operating conditions	CO ₂ stripping flux (mol/m ² s)	Removal %	Ref
PVDF + PEG 400	CO_2 loaded in distilled water; liquid velocity 0.4 m/s; gas flow rate 6 l/min; transmembrane pressure 0.5 \times $10^5 Pa$; temperature 50 $^\circ C$	4.5×10^{-5}	Not mentioned	Rahbari-Sishakht et al. (2014)
PVDF + glycerol + NMP	CO_2 loaded in distilled water; liquid velocity 0.4 m/s; gas velocity 0.02 m/s; transmembrane pressure 0.2 \times 10 ⁵ Pa; temperature 60 °C	1.15×10^{-4}	Not mentioned	Chen et al. (2014)
PVDF + SMM	CO ₂ loaded in DEA; liquid flow rate 200 ml/min; gas velocity 0.02 m/s; transmembrane pressure 0.2 \times 10 ⁵ Pa; temperature 80 °C	1.2×10^{-3}	82	Ghasem et al. (2012)
PSF	CO_2 loaded in distilled water; liquid flow rate 200 ml/min; gas velocity 0.02 m/s; transmembrane pressure 0.2 \times 10 ⁵ Pa; temperature 90 °C.	1.57×10^{-6}	66	Ghasem et al. (2011)
PVDF – PEG, PEI - PEG	CO_2 loaded in DEA; liquid velocity 0.8 m/s; gas flow rate 500 ml/min; transmembrane pressure 0.2 \times 10^5 Pa; temperature 80 °C.	$\begin{array}{l} \text{PVDF}-\text{PEG flux} \\ 4.0\times10^{-2}~\text{PEI}-\text{PEG} \\ 3.5\times10^{-2} \end{array}$	Not mentioned	Ghasem et al. (2012)
PTFE	CO_2 loaded in MEA; liquid velocity 0.04 m/s; gas velocity 0.01 m/s; Transmembrane pressure 0.2 \times 10^5 Pa; temperature 100 $^\circ\text{C}$	7×10^{-4}	Not mentioned	Cavenati et al. (2006)
PVDF + LiCl	CO_2 loaded in DEA; liquid velocity 0.45 m/s; transmembrane pressure 0.5×10^5 Pa; temperature 80 °C.	1.61×10^{-2}	70	Hoff et al. (2004)
PEI	CO_2 loaded in DEA; liquid velocity 0.9 m/s; gas flow rate 500 ml/min; transmembrane pressure 0.2 \times 10 ⁵ Pa; temperature 100 °C.	2.7×10^{-2}	40	Seo et al. (1996)
PTFE (flat sheet)	CO_2 loaded in K ₂ CO ₃ ; liquid flow rate 60 ml/min; gas flow rate 30 ml/min; transmembrane pressure 0.2 × 10 ⁵ Pa; temperature 100 °C.	1.25×10^{-2}	Not mentioned	Park et al. (2002)
PVDF + non solvent additives	CO_2 loaded in DEA; liquid velocity 0.72 m/s; transmembrane pressure 0.2 \times 10 5 Pa; temperature 80 $^\circ C.$	PEG 400 higher stripping flux 4.03 $\times ~10^{-2}$	60	Portugal et al. (2009)

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