



High pressure removal of acid gases using hollow fiber membrane contactors: Further characterization and long-term operational stability



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ABSTRACT

Our previously reported high pressure hollow fibers membrane (HFM) modules were specifically developed with aim to meet the operation requirements for the industrial sweetening of natural gas. The present work presents further characterization of CO₂ and H₂S high pressure removal under different experimental conditions, which are in particular relevant to the ultimate intended industrial application. These include module packing density, feed gas/absorption liquid flow rate ratio, absorption liquid temperature as well as solvent loading. The operational stability of the HFM module, equipped with the PFA hollow fibers, was also investigated and reported. The presented results on the long-term operational stability of membrane modules based on hollow fibers, under industrially relevant conditions are believed to be the first report of its kind. The PFA-HFM modules showed excellent operational stability during the test period, i.e., ~200 operation hours over a 7-week period using pressurized feed gas pressure (50 bar) and hot absorption solvent (100 °C).

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1. Introduction

The world's energy needs are projected to rise by 48% from 2012 to 2040 (International Energy Outl, 2016). Among different energy sources, natural gas represents the largest increase in world's energy utilization from 120 trillion cubic feet (Tcf) in 2012 to 203 Tcf in 2040 (International Energy Outl, 2016), which places natural gas in the spotlight. Despite this promising trend, almost all natural gas requires pretreatment before commercial utilization. Approximately 30% of the world's gas fields are significantly contaminated with CO₂, H₂S, in addition to some other impurities such as mercaptans and organic sulfides. Industrial natural gas sweetening aims to meet strict product specifications (Kohl and Nielsen, 1997; Alcheikhhamdon and Hoorfar, 2016; Mansourizadeh and Ismail, 2009) for H₂S levels, for safety and environmental reasons, and CO₂ for the required range of gross calorific value (GCV) (Lambert et al., 2006; Selene and Chou, 2003). Membrane system processes are attractive candidates for several industrial applications (Khulbe

and Matsuura, 2016). Applications of HFM module in various gas separation processes have attracted considerable interest of several research groups (Marzouk et al., 2012, 2010a, 2010b; Al-Marzouqi et al., 2009; Simons et al., 2009; Khaisri et al., 2009; Rajabzadeh et al., 2009; Fosi-Kofal et al., 2016; Park et al., 2008; Atcharyawut et al., 2007; Kosaraju et al., 2005; Dindore and Brilman, 2004; Yeon et al., 2003; Hedayat et al., 2011; Li et al., 1998; Zhang, 2016).

The main advantages of membrane separation over other technologies include its compact size, convenience of installation, lower energy consumption and lower cost. Different solvents have been used with HFM modules for the capture of CO₂ from different gas streams (Karror and Sirkar, 1993; Rangwala, 1996; Kumar et al., 2002; Wang et al., 2005; Li and Chen, 2005). The capture of H₂S using HFM modules and different solvents was also reported, albeit at lower rate, due to the extra safety precautions requirements for experimenting with H₂S (Marzouk et al., 2010b; Li et al., 1998; Wang et al., 2002, 2004; Boucif et al., 2007). Over the past few years, the authors reported stepwise progress towards the ultimate goal of utilizing HFM modules in industrial natural gas sweetening. In summary, our previous efforts can be divided into five phases: (i) comparing the commercially available HFM modules at low feed gas pressure (Al-Marzouqi et al., 2009), (ii) designing and custom

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constructing of HFM modules suitable for high pressure treatment of gas stream up to 50 bar (Marzouk et al., 2010a, 2010b) as the natural gas is typically explored as a pressured stream of 50 bar or more (Dindore and Brilman, 2004) and the treatment process is highly preferred to be conducted at the natural pressure to avoid the cost of re-pressurizing the gas, which is normally needed in the subsequent processing (Marzouk and Al-Marzouqi, 2010), (iii) assessing two potential microporous hollow fibers, i.e., expanded polytetrafluoroethylene (ePTFE) fibers and poly(tetrafluoroethylene-co-perfluorinated alkyl vinyl ether (PFA) fibers which were selected because of their known superior chemical compatibility of with the aggressive solvents such as amine solutions and strong alkalis, (iv) evaluating of the simultaneous CO₂ and H₂S removal from synthetic feed gas stream mimicking natural gas composition under similar conditions are used in industry (Marzouk et al., 2012), and (v) developing of a comprehensive mathematical model (Al-Marzouqi et al., 2008a, 2008b; Faiz and Al-Marzouqi, 2009; Faiz and Al-Marzouqi, 2011).

The primary objective of the present work is to investigate the capture of CO₂ and H₂S from gas stream under different experimental conditions; such as loading, temperature, gas/liquid velocity ratio and long term stability; to gain more understanding of the parameters which may affect the optimum design of HFM modules for industrial applications. The parameters considered in this study were module's surface area, feed gas/absorption solvent flow ratio, gas and solvent temperatures and solvent loading. The assessment of the operational stability of a PFA membrane module at high pressure and temperature will also be reported.

2. Experimental

2.1. Materials

Nitrogen (99.99%) and gas mixtures cylinders (5% CO₂– 2.0% H₂S–in CH₄ balance) for the simultaneous H₂S and CO₂ removal and (5% CO₂ and 95% CH₄) for CO₂ removal were acquired from Air Products (UAE). PFA fibers (0.25 mm ID, 0.65 mm OD) were purchased from Entegris (Germany). A low viscosity epoxy was acquired from Buehler (Resin No. 20-8140-128, Hardener No. 20-8142-064). Sodium Hydroxide and Diethanolamine (99%) were received from Sigma-Aldrich. The sulfide abatement solution was a sodium hypochlorite stock solution (12% w/v) and was purchased from the local stores. Deionized water was used throughout in preparing all solutions.

2.2. Experimental setup

The experimental setup used in the present work is shown in Fig. 1. A pneumatic high-pressure pump (Knauer) was used to deliver the absorption solvent, in a counter flow arrangement, to the tube side of the module. Mass flow controller (Parker, Porter model 201) was used to set the required flow rate of the feed gas mixture, which was let to pass through the shell side of the module. The upstream gas and solvent pressures were manually controlled using two back-pressure regulators (Tescom), respectively, in such a way that the solvent pressure was kept at slightly higher pressure (0.5 bar) than that of the feed gas stream. The initial concentration readings of CO₂ and H₂S which corresponds to the inlet gas composition were established at the beginning of each experiment by sending the gas stream directly to the gas analyzers. Steady state concentration levels for CO₂ and H₂S in the exit gas stream were used to calculate the flux and percent removal of these gases, respectively, for various sets of experimental conditions. The temperature of both the feed gas and absorption solvents were controlled using heating tapes. For ambient temperature

experiments (i.e., 22 ± 1 °C), the control units of the heating tapes were turned off.

2.3. Modules with different membrane surface areas

Two different modules were fabricated using 300 and 500 PFA fibers in stainless steel shell, respectively, using our previous procedure (Marzouk et al., 2010b, 2012). These two modules are labelled M300 and M500, respectively, and used throughout the present work. The specifications of the modules are presented in Table 1.

3. Results and discussions

3.1. Effect of the module packing density and membrane surface area

The simultaneous H₂S and CO₂ removal using the M300 and M500 modules was studied at ambient temperature and at different feed gas pressures. The obtained results were presented in Fig. 2a and 2b, respectively. In this investigation, the feed gas (2% H₂S, 5% CO₂ and 93% CH₄) was fed at 1500 mL min⁻¹ through the shell side of the module. Whereas the absorption sodium hydroxide solution (0.5 mol/L) was pumped, counter flow, through the tube side of the module at 10 mL min⁻¹. The results showed that the percent removal of both H₂S and CO₂ were enhanced by increasing the feed gas pressure as a result of the enhanced solubility at higher pressures for both modules. However, M500 showed better H₂S and CO₂ removal which was attributed to the higher surface area as compared with M300. This can be further explained by the more favorable lower gas/liquid velocity ratio obtained with the M500 compared to that obtained with M300. The calculated gas/liquid velocity ratios were 24 and 10.6 for M300 and M500, respectively. For each stream, the velocity (m/s) was calculated by dividing the stream flow rate (m³/s) by the corresponding cross sectional area (m²) available for flow. Lower gas/liquid velocity ratio implies higher contact time and enhanced absorption.

The theoretical calculation of overall gas mass transfer coefficient, $K_{OGj,cal}$, was obtained by using resistances in series approach, as described in our previous report (Marzouk et al., 2012). Equation (1) is used to calculate overall mass transfer coefficient from the experimental data on the basis of the overall gas phase molar concentration difference, K_{OG} :

$$K_{OGj,exp} = \frac{Q_G}{A_o} \ln \left(\frac{C_{Gj,in}}{C_{Gj,out}} \right) \quad (1)$$

where Q_G is the volumetric feed gas flow rate at the operation pressure and temperature, $C_{G,in}$ and $C_{G,out}$ are the concentrations in the gas phase at the inlet and outlet of the module, respectively, and A is the total outer membrane area.

The overall mass transfer coefficient based on the theoretical predictions and experimental data were shown in Fig. 3a and 3b, respectively. As expected, the mass transfer coefficient decreases with increasing pressure due to the decreased gas phase diffusivities (Marzouk et al., 2012). The agreement between the model predictions and the experimental results was better for M300 module than that of M500 module. The appreciable difference between the experimental results and model predictions for M500 at pressures less than 30 Bar is not fully understood at this stage and requires further investigation.

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