



Simultaneous absorption of carbon dioxide and nitrogen dioxide from simulated flue gas stream using gas-liquid membrane contacting system

Jalil Ghobadi^a, David Ramirez^{a,*}, Shooka Khoramfar^a, Robert Jerman^b, Michele Crane^b, Kenneth Hobbs^b

^a Department of Environmental Engineering, Texas A&M University-Kingsville, TX, USA

^b Markel Corporation, Plymouth Meeting, PA, USA

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ABSTRACT

Membrane gas contactors are a promising alternative to conventional post-combustion carbon capture technologies. However, residuals of the other acid gas compounds can exist in the flue gas streams emitted from industrial facilities, having a notable impact on the absorption performance of the membrane system. Simultaneous removal of CO₂ and NO₂ from a simulated flue gas stream was carried out in a polytetrafluoroethylene (PTFE) hollow fiber gas-liquid membrane contacting (GLMC) system using different scrubbing solutions. A series of experiments were conducted to study the effects of operating conditions such as gas and liquid cross flow velocities, concentration of feed gas, absorbent nature and concentration, and long-term performance of the GLMC system on the removal efficiencies as well as mass transfer rates of CO₂ and NO₂. Experimental results indicated that simultaneous absorption of CO₂ and NO₂ were enhanced with increasing the liquid-phase cross flow velocity, decreasing gas-phase cross flow velocity, and using chemical stripping absorbents. Moreover, it was shown the sodium hydroxide to be a superior absorbent as compared to alkanolamine solutions for the co-capture of CO₂ and NO₂ species. It was observed that low concentrations of NO₂ in the feed gas had a minimal impact on the decarbonization of GLMC system. The durability of the membrane system was also evaluated by running the simultaneous gas removal experiments over a 24-h period. The consistency of the absorption efficiency results confirmed the potential of using PTFE membrane system for the simultaneous absorption of CO₂ and NO₂ gases.

1. Introduction

The increased annual growth of energy demand has compelled power plants to produce more energy resulting in higher levels of air pollution especially in urban areas (Sun et al., 2016). The use of fossil fuels by power plants and the transportation sector generates compounds such as CO₂, SO₂, NO_x, and particulate matter during the combustion process. Among these compounds, CO₂ has the greatest impact on the climate change and contributes to approximately 55% of global warming (Lv et al., 2012). NO_x is a well-known air pollutant causing harm to the human respiratory system, photochemical smog, and global warming (Sun et al., 2016). NO_x represents a family of seven compounds (N₂O, NO, N₂O₃, NO₂, N₂O₄, NO₃, and N₂O₅). Among these compounds, the most common forms of nitrogen oxides in atmospheric air are nitrogen mono-oxide (NO) and nitrogen dioxide (NO₂), which are both products of the combustion of fossil fuels (Sun et al., 2016; Thomas and Vanderschuren, 1998). The United States Environmental

Protection Agency (US EPA) feels that NO₂ can be considered as a good surrogate for this family of compounds because NO₂ is the most prevalent form of NO_x in the atmosphere. Also NO can rapidly be converted to NO₂ in the atmosphere (Liémans and Thomas, 2013; US EPA, 2006). Therefore, it is necessary to remove these compounds from flue gas before they are released directly to the environment.

There are many available technologies to remove CO₂ and NO_x from flue gas streams. Some of these technologies are being used commercially and some are still at the research level (Park et al., 2009; Rufford et al., 2012; Sun et al., 2016). The CO₂ emissions from the combustion of fossil fuels can be controlled using a variety of carbon capture and storage (CCS) techniques such as physical and chemical absorption (Khoramfar et al., 2018; Russo et al., 2017; Yu et al., 2012), solid adsorption (Choi et al., 2009), cryogenic distillation (Aaron and Tsouris, 2005), and membrane separation (Favre, 2011). Strategies to mitigate NO_x emissions from power plants can be classified into two broad categories: combustion control techniques which aims to reduce the

* Corresponding author at: 917 West Avenue B, Room 376, Kingsville, TX, 78363, USA.

E-mail addresses: amirghobadi1988@yahoo.com (J. Ghobadi), david.ramirez@tamuk.edu (D. Ramirez).

generation of NO_x during fossil fuel combustion process (Komiya and Inoue, 1980; Liémans and Thomas, 2013), and post-combustion control techniques focus on the removal of NO_x from the flue gas (Chang et al., 2004; Park et al., 2009).

Post-combustion strategies are commonly used in existing power plants (Steenvelde et al., 2006). Post-combustion control methods commonly used for NO_x removal include catalytic reduction (selective catalytic reduction (SCR) and non-selective catalytic reduction (SNCR)) (Kurooka, 2011; Thomas and Vanderschuren, 1998), adsorption (Takeuchi et al., 1997), chemical absorption (Chambers and Sherwood, 1937; Lefers and van den Berg, 1982; Weisweiler et al., 1990), and membrane separation (Park et al., 2009). Application of SCR and SNCR methods are limited because of their requirement for high reaction temperature (about 1173–1273 K), high capital cost and large footprint (Park et al., 2009).

Chemical absorption methods are the most common and economical way of controlling various acidic gas compounds and particulate matter at the same time (Yang et al., 1996). They are widely used in conventional chemical scrubbing systems (e.g. packed towers, venture scrubbers, spray towers and wet scrubbers) where the flue gas is in direct contact with the scrubbing liquid. Chemical absorption methods do however have important drawbacks such as high capital cost, corrosion, and large space requirements. Moreover, treating a large amount of flue gas in a conventional gas-liquid scrubbing system requires a column with a large cross-sectional area and excess liquid absorbent to prevent foaming and channeling. The use of excess amounts of scrubbing liquids also increases the regeneration expenses and operating costs. As an alternative, gas absorption using membrane contactors with a high packing density is the most efficient way of providing the required contacting surface area between gas and liquid phase at low scrubbing liquid flow rates (Nii and Takeuchi, 1994). The membrane contactor technology uses a non-dispersive microporous membrane arranged in a modular pattern that selectively separates gases from gas streams. Gas selectivity is achieved using membranes with low permeability (Li and Chen, 2005). With the use of membrane contactors, any profile of fluid-fluid interfaces can be achieved; however, shape of contact in conventional gas-liquid separation equipment is completely accidental (Ghobadi et al., 2018).

Hollow fibers, one of the common membrane configurations, have been used in the majority of research studies on fluid separations (Brunetti et al., 2010; Li and Chen, 2005; Lv et al., 2010; Vaseghi et al., 2016; Wang et al., 2005). Hollow fiber membrane contactors (HFMCs), enable a gas mixture to travel through the shell-side or lumen-side of the hollow fibers where it can flow counter-currently or co-currently with respect to the scrubbing liquid. Membranes can be classified as hydrophobic and hydrophilic (Chabanon et al., 2011; Li and Chen, 2005; Lv et al., 2010). Membrane contactors can operate under overall-wetted or non-wetted (dry) mode depending on the pressure and flow rate of the gas and scrubbing liquid. The driving force for the gas separation in hollow fiber membrane contactors is a concentration and/or pressure gradient between the gas and liquid phases (Ghobadi et al., 2018; Mansourizadeh and Ismail, 2011). HFMCs separate the gas and liquid phases through a microporous membrane. The membrane acts as a barrier between the liquid and gas phase and provides contact surface area for the two phases. The open volume within the membrane wall, the membrane pores, generally remains filled with a gas through which the chemical species diffuse. The gas removal process occurs when the gas flows through the lumen-side or shell-side of the hollow fibers while the liquid flows through the other side. Acidic gas compounds in the gas stream diffuse through the microporous membrane which is absorbed by the flowing liquid absorbent on the opposite side. The separation of acid gas elements is a result of the concentration gradient between the absorbent and the gas stream (Zhang et al., 2014). The interaction between the selective liquid absorbent and the selected gas solute determine the removal performance. Some advantages of using the hollow fiber membrane contactors (HFMCs) for acidic gas removal processes

include independent liquid and gas flow, operational flexibility, cost effectiveness, high surface area to volume ratio, linear scale-up, and easier performance prediction due to constant interfacial area (deMontigny et al., 2006; Li and Chen, 2005). Some hollow fiber membranes have higher wettability which gives an additional level of resistance to mass transfer. For this reason, various performance studies have been conducted to evaluate the level of gas removal performance of polypropylene (PP), polyethylene (PE), polyvinylidene fluoride (PVDF) and polytetrafluoroethylene (PTFE) hollow fibers with different alkaline solutions (Chen et al., 2011; deMontigny et al., 2006; Gomez-Coma et al., 2016; Iliuta et al., 2015; Li and Chen, 2005; Mansourizadeh et al., 2010; Zhang et al., 2014). Among the different types of polymeric hollow fiber membranes, microporous membranes constructed from fluoropolymers such as PVDF and PTFE give the highest level of resistance to chemicals and wetting (Hoff and Svendsen, 2013; Marzouk et al., 2010). Therefore, in present study commercially available PTFE hollow fiber membranes with superior level of chemical compatibility and hydrophobicity were used for the acidic gas removal process.

HFMCs have been used for various applications including the removal of carbon dioxide (Hoff et al., 2004; Iliuta et al., 2015; Li and Zhang, 2018; Li and Chen, 2005), sulfur dioxide (Kim et al., 2015; Park et al., 2007; Zhang et al., 2018), hydrogen sulfide (Jin et al., 2017), volatile organic compounds (Everaert et al., 2003), nitrogen dioxide (Park et al., 2009), and mercury vapors (van der Vaart et al., 2001). Qi and Cussler (1985a, 1985b) were the first to propose the idea of using PP hollow fiber membrane contactors for removal of CO₂ and sodium hydroxide as the scrubbing absorbent (Qi and Cussler, 1985a, 1985b). Removal of CO₂ as one of the major greenhouse gas compounds from various gas mixtures such as CO₂/air (Rangwala, 1996), CO₂/O₂ (Simons-Fischbein, 2010), and CO₂/N₂ (Hoff et al., 2004; Rufford et al., 2012) using hollow fiber membrane contactors has been an active area of research ever since.

In addition to CO₂, a variety of theoretical and experimental studies have been conducted to evaluate the performance of various HFMCs for the removal of acidic gas compounds using different kinds of scrubbing solutions (Ghobadi et al., 2018; Marzouk et al., 2010; Rahim et al., 2015). Li et al. (1998) investigated the removal of H₂S from gas streams using asymmetric HFMC and alkanolamine solutions. Ogundiran et al. (1998) experimentally performed an SO₂ elimination process using hydrophobic hollow fiber membranes (HFMs) and proved that this technology can be successfully implemented as an alternative for conventional flue gas desulfurization scrubbers. Zhang et al. (2018) numerically investigated the influence of the gas and liquid flow patterns and the module configuration on SO₂ absorption using gas-liquid membrane module. Sun et al. (2008) also found that HFMCs coupled with seawater absorption is a reliable technology for the flue gas desulfurization process compared with conventional packed towers for seaside areas. Park et al. (2007) examined the effectiveness of using PVDF hollow fiber membranes for SO₂ and NO₂ removal using various alkaline solutions. Hollow fiber membrane contactors, on average, can remove up to 65% of SO₂ and 50% of NO₂ gas and have been proven to be a promising alternative for conventional scrubbing units.

Studies on the simultaneous absorption of acidic gas compounds using gas-liquid membrane contactors (GLMCs) have been conducted by researchers around the world. The application of GLMC modules for co-capture of acidic gas compounds can reduce the capital investment and operational costs of the gas scrubbing processes. Faiz and Al-Marzouqi (2009, 2011) conducted a series of experimental and mathematical studies on simultaneous control of CO₂ and H₂S using GLMCs and various types of alkaline and alkanolamine solutions. Keshavarz et al. (2008) mathematically developed a model to study the simultaneous absorption of CO₂ and H₂S under non-wetted conditions using HFMCs and aqueous solution of diethanolamine (DEA). Moreover, simultaneous removal of CO₂ and SO₂ as the two major pollutants of flue gas streams has been recently studied by various researchers. Lv et al. (2012) studied the co-absorption of CO₂ and SO₂ from coal-fired flue

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