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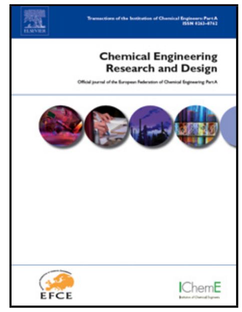

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Author: Ven Chian Quek Nilay Shah Benoît Chachuat

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Modeling for Design and Operation of High-Pressure Membrane Contactors in Natural Gas Sweetening

Ven Chian Quek[a,b], Nilay Shah[a], and Benoît Chachuat[a,1]

[a] Centre for Process Systems Engineering, Department of Chemical Engineering, Imperial College London, South Kensington Campus, London SW7 2AZ, UK

[b] Group Research & Technology, PETRONAS, 50088 Kuala Lumpur, Malaysia

Abstract

Over the past decade, membrane contactors (MBC) for CO_2 absorption have been widely recognized for their large intensification potential compared to conventional absorption towers. MBC technology uses microporous hollow-fiber membranes to enable effective gas and liquid mass transfer, without the two phases dispersing into each other. The main contribution of this paper is the development and verification of a predictive mathematical model of high-pressure MBC for natural gas sweetening applications, based on which model-based parametric analysis and optimization can be conducted. The model builds upon insight from previous modeling studies by combining 1-d and 2-d mass-balance equations to predict the CO_2 absorption flux, whereby the degree of membrane wetting itself is calculated from the knowledge of the membrane pore-size distribution. The predictive capability of the model is tested for both lab-scale and pilot-scale MBC modules, showing a close agreement of the predictions with measured CO_2 absorption fluxes at various gas and liquid flowrates, subject to a temperature correction to account for the heat of reaction in the liquid phase. The results of a model-based analysis confirm the advantages of pressurized MBC operation in terms of CO_2 removal efficiency. Finally, a comparison between vertical and horizontal modes of operation shows that the CO_2 removal efficiency in the latter can be vastly superior as it is not subject to the liquid static head and remediation strategies are discussed.

Keywords: Membrane contactor; Natural gas sweetening; CO_2 absorption; High-pressure operation; Pore size distribution; Membrane partial wetting; Experimental validation; Model-based analysis

1. Introduction

Natural Gas (NG) is presently the third most-utilized form of fossil fuel energy and is widely used for both electricity production and transportation. In the reference case of the latest International Energy Outlook [IEA, 2017], the world's NG consumption is expected to increase by 69% between 2012 and 2040, accounting for 29% of the energy consuming market, and surpassing coal as the second most utilized fuel by 2030. NG consists of a mixture of combustible hydrocarbon gases typically from methane (CH_4) to pentane (C_5H_{12}), with impurities such as carbon dioxide (CO_2). Removal of CO_2 from NG is important for various reasons. The sales gas specification for NG typically imposes a CO_2 content lower than 2-3% [TransCanada, 2016]. In liquefied natural gas (LNG) plants, CO_2 should be removed further to meet the tight specification of <50 ppmv, so as to avoid freezing in low-temperature chillers (liquefaction process); and in ammonia plants likewise, CO_2 concentrations of <100 ppmv are needed to avoid catalyst poisoning [Boucif et al., 2012; Hoff, 2003]. As far as pipeline transport is concerned, CO_2 removal avoids pumping any extra volume of gas and reduces the risk of corrosion when moisture is present in process equipment and pipeline. Regarding NG utilization lastly, the presence of CO_2 reduces the heating value of NG.

Available CO_2 removal techniques based on physical and chemical principles include absorption, adsorption, cryogenic, carbonate looping, oxyfuel combustion, and membrane separation. Chemical solvent absorption remains the most widely adopted technology to capture CO_2, using conventional packed, spray or bubble column absorption towers. About 90% of the acid gas treating processes in operation use alkanoamines solvents, such as methylethanolamine (MEA), diethanolamine (DEA), and methyldiethanolamine (MDEA), due to their versatility and ability to remove acid gases to ppm levels [Paul et al., 2007]. Nonetheless, conventional absorption towers have a high capital cost and a large physical footprint, and they are subject to operational problems such as flooding, channeling, foaming and liquid entrainment [Gabelman & Hwang, 1999]. Membrane

[1] Corresponding author. Tel.: +44 (0)207 594 5594. E-mail: b.chachuat@imperial.ac.uk