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High Temperature Polybenzimidazole Hollow Fiber Membranes for Hydrogen Separation and Carbon Dioxide Capture from Synthesis Gas

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Abstract

Sustainable reliance on hydrocarbon feedstocks for energy generation requires CO_2 separation technology development for energy efficient carbon capture from industrial mixed gas streams. High temperature H_2 selective glassy polymer membranes are an attractive option for energy efficient H_2/CO_2 separations in advanced power production schemes with integrated carbon capture. They enable high overall process efficiencies by providing energy efficient CO_2 separations at process relevant operating conditions and correspondingly, minimized parasitic energy losses. Polybenzimidazole (PBI)-based materials have demonstrated commercially attractive H_2/CO_2 separation characteristics and exceptional tolerance to hydrocarbon fuel derived synthesis (syngas) gas operating conditions and chemical environments. To realize a commercially attractive carbon capture technology based on these PBI materials, development of high performance, robust PBI hollow fiber membranes (HFMs) is required. In this work, we discuss outcomes of our recent efforts to demonstrate and optimize the fabrication and performance of PBI HFMs for use in pre-combustion carbon capture schemes. These efforts have resulted in PBI HFMs with commercially attractive fabrication protocols, defect minimized structures, and commercially attractive permselectivity characteristics at IGCC syngas process relevant conditions. The H_2/CO_2 separation performance of these PBI HFMs presented here in realistic process conditions is greater than that of any other polymeric system reported to-date.

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Keywords: Polybenzimidazole; pre-combustion; carbon capture; H_2/CO_2 separations; hollow fiber membrane

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1. Introduction

Carbon capture from power generation processes is essential for mitigation of the harmful effects of rising CO_2 levels in our atmosphere.[1] To be technically and economically viable, a successful separation method must be applicable to industrially relevant gas streams at realistic temperatures, and be compatible with large gas volumes. While the separation of CO_2 from process streams can be accomplished via standard separation techniques such as solvent scrubbing and pressure-swing adsorption, the effectiveness and efficiency of these current technologies for separating CO_2 is limited, especially when considering pre-combustion carbon capture process schemes. These aforementioned separation techniques typically require low temperatures and produce a low-pressure CO_2 stream, resulting in significant energy penalties for separating CO_2. In contrast, membrane separations offer the possibility of reduced footprint, lower parasitic load, CO_2 production at higher pressure, process temperature matching for warm fuel gas processing, decreased capital costs, continuous facile operation due to their passive platform, and low-maintenance operations. Thus, pre-combustion H_2 separation and carbon dioxide capture from synthesis (syn) gas in an integrated gasification combined cycle (IGCC) power production scheme provides significant opportunity for membrane-based technologies. The high temperature (>200 °C) and presence of H_2S and steam in syngas derived from solid fuels such as coal and biomass present a very challenging operating environment for any separation system. Therefore, a proposed membrane material must have thermo-chemical stability characteristics that address these challenges. The high pressure of IGCC syngas and high partial pressure of H_2 in that stream provide the driving force necessary for energy efficient membrane separation. As a result, if a proposed material and membrane comprised of that material possesses adequate permselectivity characteristics at process conditions (chemical, thermal, pressure), sufficient driving force exists to realize the target separations via optimized process integration pathways and correspondingly, with minimized parasitic losses.

Polymer membranes have been used successfully in a number of industrial applications, including the production of high-purity nitrogen, gas dehydration, removal of acid gases, and recovery of hydrogen from process streams for recycle. However, successful use of a polymer membrane for syngas separations requires a membrane that is thermally, chemically, and mechanically stable at high temperature and high pressure in the presence of the chemically challenging syngas components. Unfortunately, the commercially available polymeric materials currently employed in separation applications are not stable in these demanding environments to the degree required. Current membrane materials are often subject to chemical degradation by minor components in the process stream, a problem that is exacerbated by elevated temperature. Additionally, as the glass transition temperature (T_g) of the polymer is approached, membrane selectivity reductions are commonly observed due to increased polymer mobility and its influences on free-volume and flux declines often occur due to membrane compaction. Consequently, there is a compelling need for membrane materials and subsequent capture systems based on those materials that can operate under more extreme environmental conditions for extended periods of time while providing a level of performance that is economically sustainable by the end user.

Development and demonstration of high T_g materials to address the aforementioned limitations of the current state of the art and the corresponding separations needs of industry including the utility sector is a focus of this work. Polybenzimidazole (PBI)-based membranes have demonstrated commercially-attractive H_2/CO_2 selectivity, exceptional thermal stability (T_g > 400 °C), and exceptional tolerance to H_2S.[2] Systems and economic analyses established the techno-economic viability and advantages of these materials over state of the art CO_2 separation technology (Selexol^{TM}) [3, 4] and indicate the strong potential for PBI membrane based capture technology to meet and exceed the U.S. Department of Energy (DOE) Office of Fossil Energy (FE)/National Energy Technology Laboratory (NETL) Strategic Center for Coal – Carbon Capture Program goals. These system analyses also make clear that a high area density membrane module design is necessary to realize the desired step-change in both performance and cost of CO_2 capture associated with the use of this membrane based capture technology. One promising option for achieving a substantial increase in active membrane area density is the use of a hollow fiber membrane platform. A hollow fiber module is the membrane configuration with the highest achievable packing density, i.e., the highest membrane selective area density. Hollow fiber modules have been fabricated to obtain as