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Scale-up of PEEK hollow fiber membrane contactor for post-combustion CO_2 capture

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ABSTRACT

Poly(ether ether ketone) (PEEK) hollow fiber membrane modules were scaled up from 2 in. diameter and 16 in. long to 4 in. diameter and 58 in. long (a factor of 90 increase in membrane area) for CO_2 capture from flue gases using a membrane contactor process, which combines advantageous features of both absorption and membrane processes. Field tests of a 4-in.-diameter module with activated methyldiethanolamine (aMDEA) solvent demonstrated greater than 90% CO_2 removal. The mass transfer coefficient in the absorption step was 1.2 $(s)^{-1}$, which is over an order of magnitude greater than that of conventional column contactors. The membrane module was further scaled up from 4 in. to 8 in. diameter (a factor of 3 increase in membrane area). Single-gas permeation measurements indicated that 8-in. modules have an intrinsic CO_2 permeance as high as 2150–2670 GPU. One 8-in.-diameter module was tested in membrane contactor mode for CO_2 capture using a simulated flue gas. Carbon dioxide removal rate of 91.3% was achieved with a mass transfer coefficient of 1.5 $(s)^{-1}$. Parametric tests indicated CO_2 flux and capture rate increased with increasing feed pressure, solvent flow velocity and solvent temperature. PEEK hollow fiber membrane contactor is effective in capturing CO_2 from low CO_2-concentration feeds, showing its high potential for coal or natural gas flue gas CO_2 capture.

1. Introduction

The greatest concern to climate change is the emission of greenhouse gases, especially CO_2 from fossil-fuel power plants. The amount of CO_2 attributed to the U.S. electric power sector is expected to increase by 11% from 2012 to 2040, although electric power generation from non-carbon sources (such as nuclear and renewable fuels) is projected to increase during this period [1]. Therefore, to address concerns about global climate change, CO_2 capture and storage is one of many approaches that is critical to significantly reducing domestic and global CO_2 emissions.

The US Department of Energy (DOE) has a goal of achieving a 90% capture rate in existing plants with 95% CO_2 purity at a cost of $40/tonne of CO_2 captured by 2025 [2]. To achieve this goal by any technological means is very difficult, because flue gas is hot, near atmospheric pressure, high in volume, and often contaminated with other impurities (O_2, SO_x, NO_x, and ash). Amine absorption is the current industry and DOE benchmark technology for capture of CO_2 from power plant flue gas. Residual oxygen in the flue gas is especially troublesome for conventional amine plants because of oxidative degradation of the amine. In addition, the heat duty of the stripper places a substantial burden on the steam supply. It is estimated that for every pound of CO_2 captured, 2–3 pounds of steam is required [3,4]. DOE/NETL systems analysis studies estimated that using an aqueous monoethanolamine absorption process to capture 90% of the CO_2 from flue gas in existing plants will result in a CO_2 capture cost of $56/tonne (DOE Case 12) at the plant gate [5], which is above the 2025 DOE NETL Carbon Capture Program post-combustion capture cost goal ($40/tonne of CO_2 captured). These factors result in enormous amine circulation rates, large equipment, and large energy requirements. Therefore, it is important to develop new advanced CO_2 capture technologies in order to maintain the cost-effectiveness of U.S. coal-fired or natural-gas-fired power generation.

Through analysis and feasibility studies, Favre [6] reported that gas separation membranes are a technically and economically viable option for post-combustion CO_2 capture. Membranes are compact and can be retrofitted onto the tail end of a power-plant flue gas stream without complicated integration schemes. The conventional gas separation membrane process operates by a solution/diffusion mechanism, and the separation driving force is provided by the partial pressure

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difference of each component across the membrane. This process requires either flue gas compression, permeate sweep, application of permeate side vacuum, or a combination of these steps to provide the separation driving force required. Elaborate process design and optimization become prerequisite for conventional membrane processes in CO_2 capture from flue gases [7].

The main limitation of conventional membrane processes is the process pressure ratio (feed gas pressure/permeate gas pressure) limitation [8]. In practical gas separation applications, the pressure ratio across the membrane is usually between 5 and 15 [9]. When the membrane separation process is pressure ratio limited, the product CO_2 concentration will be limited even when the membrane selectivity is much larger than the pressure ratio. Thus, multiple membrane stages are required to generate greater than 95% pure CO_2 product (DOE's target) from flue gases using the conventional membrane process.

The membrane contactor process (also known as hybrid membrane/absorption process) combines advantageous features of both absorption and membrane processes to provide a cost-effective solution for CO_2 capture from flue gases. Hollow fiber membrane contactors for CO_2 capture, especially the absorption process, have been reported [10–14]. In this process, CO_2-containing gas passes through small-diameter membrane tubes (hollow fibers with porous walls) while a CO_2 selective solvent (typically an amine solution) flows on the shell side of the membrane. CO_2 permeates through the membrane and is absorbed in the solvent. The CO_2 rich solvent is regenerated and sent back to the membrane absorber.

The specific surface area per volume for hollow fiber membrane contactors can be as high as ~1000–9000 m^2/m^3, which is up to two orders of magnitude greater than conventional contactors (free dispersion columns: ~3.0–35 m^2/m^3, packed and tray columns: 30–300 m^2/m^3, and mechanically agitated columns: ~160–500 m^2/m^3) [15,16]. Thus, the use of a membrane contactor instead of a conventional amine scrubber leads to a much smaller space requirement. This technology is well suited for new and existing pulverized coal-fired or natural-fired power plants due to the reduced footprint requirement and a much lower visual impact as well as providing more options for placement in the confines of existing plants.

In the hybrid membrane/absorption process, the permeate side partial pressure of CO_2 can be considered close to zero due to the chemical reaction of CO_2 with the absorption solvent, and thus overcomes the pressure ratio problem encountered by the conventional gas membrane process. Feed compression or permeate vacuum is not required to create the separation driving force for gas molecules to be transported through the membrane. The process selectivity for the hybrid membrane/absorption process is determined by the chemical affinity of the absorption solvent to CO_2. Therefore, high purity CO_2 product can be realized in a single stage.

In our previous study [17], the feasibility of utilizing a PEEK membrane based hollow fiber contactor in combination with chemical solvents to separate and capture CO_2 from simulated flue gases with high efficiency (at least 90% removal) was successfully established via testing of 2-in.-diameter modules. Advanced hindered amines and promoted carbonate solvent have been used to minimize solvent degradation and lower regeneration energy requirements. Greater than 90% CO_2 capture with greater than 95% CO_2 purity has been achieved in one stage with activated methyldiethanolamine (aMDEA) solvent. The measured volumetric mass transfer coefficient for the 2-in.-diameter module was as high as 1.7 $(s)^{-1}$, which is more than 20 times greater than the mass transfer coefficient of a packed column. The lab tests also indicated that the CO_2 capture performance was stable and not affected by flue gas contaminants, including O_2, NO_2, and SO_2, with aMDEA solvent. The PEEK hollow fiber membrane contactor provides a unique platform to maximize the benefits of new and advanced solvents including those currently supported the DOE.

In the current study, the PEEK membrane modules were scaled up from 2 in. diameter and 16 in. long to 4 in. diameter and 58 in. long (a factor of 90 increase in membrane area) for field testing, then further scaled to commercial-sized 8-in. diameter (another factor of 3 increase in membrane area). High CO_2 capture performances were observed for these modules, indicting scalability of the PEEK hollow fiber membrane contactor process.

2. Experimental methods

2.1. Hollow fiber membrane fabrication and surface modification

The hollow fiber membranes are manufactured from the best in class commercial engineered plastic, PEEK. Porous PEEK hollow fibers used in preparation of super-hydrophobic membranes were manufactured by a high temperature melt extrusion process. In this process, a mixture containing PEEK and PEI (polyether imide), was melt extruded to form a hollow fiber, which was then cooled in air and sent to reagent bath, where PEI was removed. The resulting product was porous PEEK fiber. The details of the process were described previously [18].

The super-hydrophobicity of the porous PEEK membrane was generated by surface modification with a functional perfluoro oligomer. Prior to grafting with the perfluoro oligomer, the surface of the porous PEEK was functionalized with -OH groups by reacting ketone groups in the PEEK polymer backbone with monoethanolamine. The functionalized porous PEEK was prepared in a single step Reactive Porogen Removal process during porous PEEK fiber preparation according to US Patent 7,176,273 [19].

2.2. Membrane module fabrication and characterization

Module design and construction have significant impacts on the overall gas mass transfer coefficient by minimizing liquid side resistance and maximizing the driving force. The following key design elements and development work have been carried out:

1) Four-port counter-current flow design, enabling optimum driving force for the acid gas absorption;
2) Computerized structural packing minimizing the absorption liquid malflow;
3) Optimum fiber packing density to minimize the liquid pressure drop and optimize the liquid flow turbulence;
4) Optimized winding patterns to promote the liquid side gas mass transport; and
5) Curved hollow fiber with enhanced gas phase mass transport.

The cartridge was potted by epoxy and sealed with O-rings and housed in a stainless steel pressure vessel. The non-leaking characteristics of membrane modules were determined by pressurizing a water/hexane mixture in the shell side of the module and observing if there was any liquid collection in the tube side. Any liquid collected on the tube side would indicate leakage of the membrane module.

The membrane's intrinsic gas permeation property for CO_2 was measured in a flow system shown in Fig. 1. The CO_2 was fed to the tube side of the module. Flux was measured by a flow meter. The pressure normalized flux, permeance (P), is:

$$P = \frac{J}{\Delta p} \tag{1}$$

where J is the total steady state flux through the membrane and Δp is pressure differential between the tube and shell sides. Since the module has a cross-flow design, a log-mean pressure drop (Δp) was used to calculate the driving force from the partial pressures

$$\Delta p = \frac{(P_{f,i} - P_p) - (P_{f,o} - P_p)}{Ln[(P_{f,i} - P_p)/(P_{f,o} - P_p)]} \tag{2}$$

where $P_{f,i}$, $P_{f,o}$, and P_p are the pressures of gas feed inlet, gas feed outlet

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