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The effect of operating conditions on the performance of hollow fiber membrane modules for CO_2/N_2 separation

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ABSTRACT

A commercialized polysulfone (PSf) hollow-fiber membrane module was tested for CO_2/N_2 separation performance for application in post-combustion capture. Cost efficiency, easy module manufacturing, and efficiency in gas separation are the main advantages of using PSf hollow-fiber modules for CO_2 separation. The effects of operating conditions such as temperature, pressure, and feed composition on separation performance were examined at various stage cuts. A 2-stage system including concentration of feed composition at stage 1 and production of high-purity CO_2 at stage 2 was constructed to improve separation efficiency. Higher operating temperature and pressure increased CO_2 permeance, but the loss of selectivity and higher energy consumption are a concern. Modules with various membrane areas were also used to test the effect of area on CO_2 separation.

1. Introduction

It is well known that the emission of CO_2 greenhouse gas is regarded as the most serious cause of global warming [1–3]. Several techniques are available for the separation of CO_2 from flue gas streams at present, such as chemical and physical absorption, solid adsorption, carbon molecular sieve adsorption, cryogenic distillation, membrane separation, and other novel methods. In particular, membrane-based gas separation offers great opportunities for bulk separation due to its low energy consumption, easy operation, and low maintenance compared with chemical absorption or adsorption methods that require regeneration of sorbent. Moreover, the membrane process has the advantages of modular design and lightweight, making it particularly suitable for retrofit into existing processes [4–7]. Generally, the use of polymer materials for hollow-fiber membrane preparation provides attractive properties such as high packing density, self-support, and flexibility in operation.

PSf has been widely used as a membrane material due to its satisfactory performance, mechanical strength, thermo-chemical stability, and low economic cost. For these reasons, it is already applied in industrial gas separation processes [8,9]. Other materials which show high selectivity close to 100 with low permeability, such as polyimides, facilitated transport, mixed-matrix, and carbon molecular sieve have also been studied, but high production and material costs along with expensive modules have prevented their intended commercial scale use [10–12]. Application in industrial- and utility-scale processes, including boilers, cement manufacturing, steel and aluminum production, and chemical refining can be possibly achieved by the development of a cost-effective membrane process for CO_2 separation [13,14].

Optimization of module configuration in membrane gas separation was studied by Lababidi. Three systems including single-stage, two-stage, and continuous membrane column (CMC) permeators were compared in terms of optimum cost and specific operating and design characteristics [15]. Yeom et al. also investigated coupling effects to understand the difference between permeation and separation behavior in the permeation of pure and mixed gases [16]. Many other studies on CO_2/N_2 separation have been widely carried out in respect to high permselective polymeric materials [17,18] as well as systems analysis and design to understand and arrange the membrane process effectively [19,20].

The object of this work is to examine the performance of PSf membrane modules that have already been commercially used for oxygen/nitrogen separation. Developing our understanding of the CO_2 separation performance of commercial gas separation membranes can be useful in searching for new materials, optimizing module systems, and establishing operating conditions. Experiments on pure and mixture gases with various

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Table 1
Information on membrane modules.

		Meritair 1008P	Meritair 1507P
Strands		400	3700
Effective membrane surface area (cm^2)		830	6400
Module size	Diameter (mm)	42	60
	Length (mm)	240	220
Module shape	Cross-section		
	Housing		

compositions were carried out over a wide range of stage cuts. Other effects of operating temperature, pressure, and composition were investigated through commercial asymmetric PSf membranes. A 2-stage module set up was used to enhance CO_2 purity by concentrating CO_2 gas at stage 1 and various membrane areas for stage 2 were used to confirm the effect of membrane area on separation performance.

2. Experimental

2.1. Materials

The PSf hollow-fiber membrane modules used in this study were kindly supplied by Airrane Co. (Daejeon, Korea). The membranes were prepared from a dope solution composed of PSf (Udel P-1800, Mw 24,000 g/mol, Solvay Advanced Polymers), N-methyl-2-pyrrolidone (NMP), teterahydrofuran (THF), and alcohol with the appropriate composition. The polymer was dried at 80 °C in a vacuum oven before use, and other solvents were used as received. Deionized water and tap water were used as a bore solution and coagulating medium, respectively. The coating solution was prepared by mixing polydimethylsiloxane (Sylgard 184, Dow Corning) and a curing agent in n-hexane. Membrane modules of two different sizes were used for the gas separation test, and details of the modules are given in Table 1. Pure test gases with high purity of 99.8–99.99% and mixture gases of 50% CO_2 and 50% N_2 were supplied by Praxair.

2.2. Membrane and membrane module preparation

Microporous PSf hollow-fiber support membranes were spun from a homogenously mixed dope solution composed of PSf, NMP, THF, and alcohol. After degassing overnight, the dope solution was extruded through a spinneret with an outer diameter of 0.4 mm and an inner diameter of 0.2 mm. Phase separation occurred both inside and outside of a membrane by coagulation water. Membranes were passed through an external coagulation bath and forwarded to a take-up bobbin. The fibers were washed in running water for 3 days to complete solvent–nonsolvent exchange and dried at 60 °C for 2 days in a convection oven. The dried fibers were automatically twirled by an unwinding machine and immersed in a coating solution containing 2 wt% of Sylgard 184 in n-hexane. After dip-coating, they were continuously passed through a hot-air drying machine for 20 s and rolled up in a bobbin. A bundle of hollow fiber membranes were cut to a constant length and encased in the housing, with both ends of the fiber bundle potted with epoxy resin. When the epoxy resin became solid, the resin with the embedded fiber ends was cut straight to make a gas transfer path (i.e. feed and permeate side). The final module shape is shown in Table 1. The module size and the membrane effective area depending on the potted fiber number strongly related to their gas separation performance.

2.3. Characterization

The membrane cross-section was observed by a field emission scanning electron microscope system (FE-SEM, Philips XL30 S FEG, Netherland). Several fibers were wetted in water and then directly immersed in liquid nitrogen. Frozen water on the membranes makes cutting easy, and clear cross-sectional fractures were obtained. The membranes were mounted on a metal holder and gold-sputtered for 30 s.

The traditional bubble test method was adapted to measure gas permeance for pure and mixture gases. Fig. 1 shows the experimental set-up of the bore-side feed configuration and the counter-current flow used in this study. The feed gas at controlled pressure was passed through the bore side and the permeate flowed concurrently through the cell-side of the fibers. The permeate and residual gas flux was measured by a precise flowmeter. The permeate stream was also connected to gas chromatography (GC DS6200, Donam instruments, Korea) to confirm its composition. Pure gas permeance is the pressure normalized flux of gas A, and it is expressed in GPU (gas permeance unit) (10^{-6} cm^3(STP)/cm^2·cmHg·s). The ideal selectivity of a gas pair A/B is defined as the permeance of gas A divided by the permeance of gas B. For a mixture gas test, the stage cut directly effects the permeate gas flux and selectivity. The stage cut is defined as the fractional feed flow permeated through the membrane, and it can be calculated from the flow ratio of permeate against feed. The fractional quantities of CO_2 permeated through the membrane which is designate as CO_2 recovery (ψ) is given by

$$\psi(CO_2) = \frac{Q_{pemeate} \times [CO_2]_{permeate}}{Q_{feed} \times [CO_2]_{feed}}, \tag{1}$$