Environmental Science & Technology

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Techno-Economic Assessment of Polymer Membrane Systems for Postcombustion Carbon Capture at Coal-Fired Power Plants

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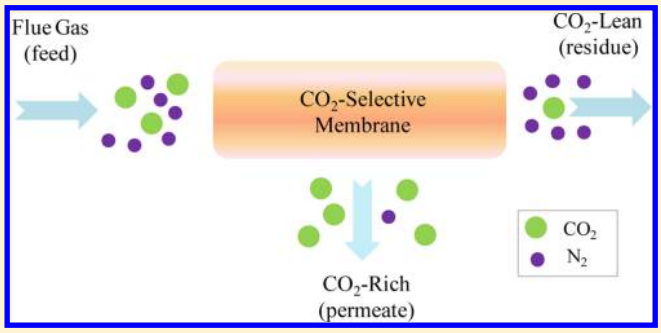
Supporting Information

ABSTRACT: This study investigates the feasibility of polymer membrane systems for postcombustion carbon dioxide (CO_2) capture at coal-fired power plants. Using newly developed performance and cost models, our analysis shows that membrane systems configured with multiple stages or steps are capable of meeting capture targets of 90% CO_2 removal efficiency and 95+% product purity. A combined driving force design using both compressors and vacuum pumps is most effective for reducing the cost of CO_2 avoided. Further reductions in the overall system energy penalty and cost can be obtained by recycling a portion of CO_2 via a two-stage, two-step membrane configuration with air sweep to increase the CO_2 partial pressure of feed flue gas. For a typical plant with carbon capture and storage, this yielded a 15% lower cost per metric ton of CO_2 avoided compared to a plant using a current amine-based capture system. A series of parametric analyses also is undertaken to identify paths for enhancing the viability of membrane-based capture technology.



INTRODUCTION

Coal-fired power plants account for nearly 50% of U.S. electricity supply and about a third of national emissions of carbon dioxide (CO_2).[1] Carbon capture and storage (CCS) could play an important role in deeply cutting power plant CO_2 emissions. However, adding current amine-based capture systems to coal-fired power plants would increase the cost of electricity by about 70−80% and incur roughly a 25−40% energy penalty.[2,3] The addition of amine-based CCS would also nearly double plant water use.[4] To address these major drawbacks, the U.S. Department of Energy is intensively supporting research and development (R&D) on advanced CCS systems.[5,6] Membranes have many industrial applications such as atmospheric gas and industrial gas purification. Potential applications for capturing CO_2 from power plant flue gases mainly involves the separation of CO_2 from nitrogen (the main constituent of flue gas) for postcombustion CCS, and CO_2 from hydrogen (the main constituent of fuel gas) for precombustion CCS. This paper focuses on polymeric membranes for postcombustion CO_2 capture, which is the most advanced of the membrane capture systems. Other membrane technologies under development for CO_2 capture include nondispersive absorption-based porous membranes, gas permeation membranes, and supported liquid membranes.[7]

Gas separation using polymeric membranes generally follows a solution−diffusion mechanism.[8] To be effective for postcombustion CO_2 capture, membrane materials should possess a number of features including high CO_2 permeability, high selectivity of CO_2 versus N_2 and other gases, thermal and chemical stability, and resistance to aging.[9] However, there is a common trade-off between membrane properties: more permeable polymers are less selective and vice versa.[10] The CO_2 versus N_2 selectivity for most polymeric membrane materials (e.g., polyimides and polyacetylenes) is less than 50−70, although CO_2 permeability also can vary substantially for a given type of material.[9,11]

Membrane systems are being actively investigated for removing CO_2 from power plant flue gases. Most of these studies have evaluated single-stage or two-stage systems designed with either a low CO_2 removal efficiency or a low CO_2 product purity.[11−22] These conditions are not comparable to amine-based postcombustion capture systems, which achieve high capture rates (90% or more) with a very high CO_2 purity (over 99%).[2] And while existing studies have broadly investigated the effects of membrane properties on system performance, they scarcely look into impacts of alternative power plant designs.

Beyond the modeling of membrane separation processes, some recent studies also evaluated energy penalties and costs for membrane-based capture systems. Using vacuum pumps in place of compressors to reduce energy penalties was proposed in some studies.[12,16,19−21] However, vacuum equipment is more capital intensive; and the assumption of vacuum pressures less than 0.1−0.2 bar in optimum process designs likely biases assessments of membrane system feasibility because it is hard to practically achieve such low vacuum pressures for a full-scale capture system.[12]

Received: December 10, 2012
Revised: February 1, 2013
Accepted: February 13, 2013
Published: February 13, 2013

In addition, the costing methods and scope for membrane systems analysis are not consistent across existing studies, and commonly result in the under-estimation of total system cost. For example, indirect capital costs and owner's costs, which can amount to more than 40% of total capital cost, are often ignored. CO_2 product compression is not included in the scope of some studies' energy and cost calculations, although it is also one of major cost components of a CCS system. Many studies also fail to report the cost of CO_2 avoided—the most widely used cost measure for inter- and intratechnology comparisons.[23] Rather, many membrane studies report only the "cost of CO_2 captured". This measure substantially underestimates the cost of avoiding CO_2 emissions to the atmosphere because it ignores the additional CO_2 emissions associated with process energy requirements, and typically employs arbitrary electricity price assumptions that vary by more than a factor of 2.[12,13,20,21,24] As noted elsewhere,[23] it is thus an inappropriate metric for comparing the cost of plants employing different CCS systems, whose purpose is to supply electricity while avoiding CO_2 emission to the atmosphere.

The fundamental objective of this paper, therefore, is to present a more complete and systematic evaluation of the technical feasibility and cost of polymeric membrane systems for CO_2 capture at pulverized coal (PC) power plants. We recognize, however, that minor gas components in real flue gases, such as sulfur oxides (SO_x) and carbon monoxide (CO), also may affect membrane separation performance,[25] although their influences still remain largely unstudied. This lack of a more complete understanding limits efforts of current system modeling (including this study) to focus only on the separation of CO_2 and N_2.[25,26] Given that focus, more specifically, we first quantify the performance of different process configurations and design parameters to identify feasible membrane systems that are able to simultaneously achieve 90% CO_2 removal efficiency and 95% or more product purity. We further estimate the costs of alternative designs based on a complete and systematic costing basis. We also compare the overall cost of a membrane-based CCS system with that of an amine-based system representing current commercial technology.

APPROACH TO PERFORMANCE AND COST ESTIMATES

This section briefly describes the performance and cost models employed to evaluate membrane-based capture systems at coal-fired power plants. Further details of these models are provided in the Supporting Information (SI).

Membrane Gas Separation Models. Unlike amine-based capture systems, a membrane gas separation process has no chemical reactions and thus no need for solvent regeneration processes. Gas separation by polymeric membranes relies mainly on the permeability of membrane materials. The driving force for gas separation is the partial pressure *difference* between the feed side and the permeate side of a membrane. Transport flux through polymeric membranes is expressed as[27]

$$J = \frac{P^*}{\delta}(xP_f - yP_p) \tag{1}$$

where J is the volumetric flux ($cm^3/(cm^2 \cdot s)$); P^* is the gas permeability ($cm^3 \cdot cm/(s \cdot cm^2 \cdot cmHg)$); δ is the membrane thickness (cm); P_f and P_p are the pressures in the feed and permeate sides (cmHg); and x and y are the concentrations of CO_2 in the feed and permeate streams (vol %). Membranes can be packed in modules to operate under different flow patterns such as cross-flow, concurrent flow, and countercurrent flow. Cross-flow modules are most often used in industrial practice.

In formulating theoretical separation models, isothermal conditions, negligible pressure drop, and constant gas permeability independent of pressure are generally assumed. The local permeation rate of either CO_2 or N_2 in a binary membrane system for the cross-flow pattern over a differential membrane area (dA) is described as[27]

$$-ydq = J_{CO2}dA = \frac{P^*_{CO2}}{\delta}[xP_f - yP_p]dA \tag{2}$$

$$-(1-y)dq = J_{N2}dA = \frac{P^*_{N2}}{\delta}[(1-x)P_f - (1-y)P_p]dA \tag{3}$$

Dividing eq 2 by eq 3 yields

$$\frac{y}{1-y} = \frac{\alpha(1 - y/\phi)}{(1-x) - (1-y)/\phi} \tag{4}$$

where A is the membrane area (cm^2); q is the gas flow rate (cm^3/s); α is the membrane selectivity (P^*_{CO2}/P^*_{N2}) for CO_2 versus N_2 gases; and ϕ is the pressure ratio (P_f/P_p) for feed versus permeate sides. The CO_2 concentration of flue gas is enriched in the permeate stream through the CO_2-selective membrane. Mathematical transformations are applied to obtain an analytical solution to the governing equations,[27] which is summarized in the SI.

To increase the driving force for gas separation, a sweep gas can be used in a countercurrent module to reduce the partial pressure of CO_2 in the permeate side.[28] This, in turn, decreases the membrane area and energy requirements for gas separation. Thus, we apply the unified mathematical framework established by Pan and Habgood[28] to model the separation of CO_2 and N_2 for the countercurrent flow pattern with gas sweep. The set of model equations is detailed in the SI.

Power Use of Major Equipment. Flue gases from coal-fired power plants typically have 10−15% CO_2 by volume at atmospheric pressure, which results in a low CO_2 partial pressure. Thus, either compressing the flue gas to a high pressure at the feed side or lowering the pressure at the permeate side through vacuum pumps is needed to produce a sufficiently high partial pressure difference of CO_2 across the membrane. The energy use for either compressors or vacuum pumps is estimated as[15,17,20,29]

$$E = \frac{1}{\eta}Q\frac{\gamma RT}{\gamma - 1}[(\dot{\psi})^{(\gamma-1)/\gamma} - 1] \tag{5}$$

where E is the equipment power use or credit (W); Q is the gas flow rate through the equipment (mol/s); T is the operating temperature (K); η is the equipment efficiency (%); γ is the adiabatic expansion factor; and $\dot{\psi}$ is the pressure ratio across the compressor or vacuum pump. When a compressor is used at the feed side, the compression energy may be partly recovered from the residue stream using an expander. The recovered energy is estimated as[20]

$$E = \frac{1}{\eta}Q\frac{\gamma RT}{\gamma - 1}[1 - (\ddot{\psi})^{(\gamma-1)/\gamma}] \tag{6}$$

where $\ddot{\psi}$ is the pressure ratio across the expander.

The final CO_2 product needs to be compressed to the liquid phase for transport to a storage site. The energy requirements and cost of that compression are commonly attributed to the

capture component of the CCS chain.[30] The energy use for CO_2 product compression is estimated using the Integrated Environmental Control Model (IECM),[31] which is discussed further later.

Performance Model Summary. To briefly summarize, our analytical procedure is as follows: given the flue gas flow rate and composition, membrane properties, pressure specifications, and CO_2 removal targets, the performance model described above is used to calculate permeate and residue streamflow rates and associated CO_2 concentrations, as well as the required membrane size. The derived streamflow rates also are used to estimate the power requirements of individual pieces of equipment. Subsequently, economic calculations are driven by parameters and results of the process performance modeling.

Costing Method. The process performance model described above is linked to engineering-economic models that estimate the capital cost, annual operating and maintenance (O&M) costs, and total levelized annual cost of an overall power plant and the CO_2 capture system. The costing method and nomenclature employed in this study are based on the Electric Power Research Institute's (EPRI) Technical Assessment Guide (TAG), a widely adopted industry standard.[32]

The total capital requirement (TCR) of a membrane-based capture system takes into account the direct costs of purchasing and installing all process equipment (denoted as the process facilities capital, PFC), plus a number of indirect costs such as the general facilities cost, engineering and home office fees, contingency costs, and several categories of owner's costs. The major components of the PFC include the membrane module and frame, compressors, expanders, vacuum pumps, and heat exchangers, as well as CO_2 product compression. The indirect capital costs are commonly estimated as a percent of the PFC based on industry guidelines.[32] Fixed O&M (FOM) costs include operating labor, maintenance costs, and overhead costs associated with administrative and support labor. Variable O&M (VOM) costs include materials replacement, and electricity (or other energy) used to operate the system. Further details of TCR, FOM, and VOM are given in SI Tables S-1 and S-2.

Because the electricity to operate the capture system is obtained from within the power plant, any fictitious "purchase price" of electricity becomes arbitrary and unnecessary. Instead, the most robust way to evaluate the cost of energy-intensive processes such as CO_2 capture systems is to compare the overall cost of electricity generation for power plants with and without CCS (the difference being the cost of CCS).[23] One of the key outputs of such an economic analysis is the total levelized cost of electricity (LCOE) for a complete power plant, including the cost of CO_2 product transport and storage (T&S) for plants with CCS.[33] In this paper, we report the cost of CCS as the difference in LCOE between a plant with membrane-based capture and a "reference plant" without capture. We also report the cost of CO_2 avoided, which quantifies the levelized cost of avoiding a ton of atmospheric CO_2 emissions while still providing a unit of electricity to customers.[23] In all cases, the reference and capture plants are the same type of PC power plant. Finally, to allow comparisons with cost results in the literature, we also report the "cost of CO_2 captured" for general cases.

Overall Plant Model. Unlike other studies that evaluate CO_2 capture processes in isolation, here, we simulate complete power plants with and without CCS. The recently enhanced IECM v 7.0-beta was used to model a supercritical PC power plant as the reference plant complying with federal New Source Performance Standards (NSPS) for air and water pollutants. The IECM is a publicly available computer model that provides systematic estimates of plant-level performance, costs, and emissions for fossil fuel power plants with and without CCS.[31] Table 1 lists the major technical and economic metrics defining the reference plant.

The performance and cost models described above for membrane-based capture were incorporated into the IECM framework. Thus, Table 1 also summarizes the "base case" performance and cost assumptions for the membrane-based

Table 1. Technical and Economic Assumptions for Baseline Pulverized Coal Power Plant and Membrane System

variable	value
power plant (w/o CCS)	
plant type	supercritical
coal type	Pittsburgh #8
environmental controls	SCR + ESP + FGD[a]
cooling system	wet tower
capacity factor (%)	75
gross/net electrical output (MW)	588/550
net plant efficiency (HHV, %)	38.9
flue gas flow rate (STP m^3/s)[b]	535
flue gas pressure (bar)	1.0
CO_2 molar concentration in flue gas (%)	12
fixed charge factor	0.113
levelized cost of electricity (constant 2010$/MWh)	58.1
membrane system	
CO_2 permeance (STP gpu)[c]	1000
CO_2/N_2 selectivity (STP)	50
system operating temperature (°C)	30
compressor/expander/vacuum pump efficiency (%)	85
product compressor energy use (kWh/mt CO_2)	93
membrane module price ($/$m^2$)	50
gas compressor installed cost ($/hp)	500
gas vacuum pump installed cost ($/hp)	1000
gas expander unit cost ($/kW)	500
heat exchanger capital cost ($/$m^2$)	300
CO_2 product compression installed cost ($/kW)	900
general facilities capital (% of PFC)	10
engineering and home office fees (% of PFC)	7
project contingency cost (% of PFC)	15
process contingency cost (% of PFC)	5
royalty fees (% of PFC)	0.5
preproduction costs	
months of fixed O&M	1
months of variable O&M	1
misc. capital cost (% of TPI[d])	2
inventory capital (% of TPC[d])	0.5
annual material replacement rate (%)	20
material replacement cost ($/$m^2$)	10
labor rate ($/h)	33
number of operating jobs (jobs/shift)	2
number of operating shifts (shifts/day)	4.75
total maintenance cost (TMC) (% of TPC)	2.5
maintenance cost allocated to labor (% of TMC)	40
administrative and support cost (% total labor)	30

[a]SCR = selective catalytic reduction; ESP = electrostatic precipitator device; FGD = flue gas desulfurization. [b]The STP indicates the standard temperature and pressure conditions (0 °C and 1 atmospheric pressure). [c]1 gas permeation unit (gpu) = 10^{-6} cm^3 (STP)/(cm^2·s·cmHg). [d]TPC = total plant cost; TPI = total plant investment.

capture system. Membrane properties and costs are based on recently reported data for polymeric membranes.[12] We assume the CO_2 product is compressed to 2000 psia using a multi-stage compressor. Cost data for other major process equipment are based on the literature and IECM estimates.[13,31,34,35] Base case assumptions regarding indirect capital and O&M costs are similar to those of an amine-based capture system.[31,35]

RESULTS

We evaluated both single- and multi-stage membrane system designs for CO_2 capture (Figure 1) considering the effects of three important factors: membrane material properties, capture process design, and power plant design. We particularly looked for designs that could reduce the energy penalties of CO_2 separation, which are a major contribution to total plant cost. The operating temperature of the capture system is maintained at 30 °C by cooling water via heat exchangers.

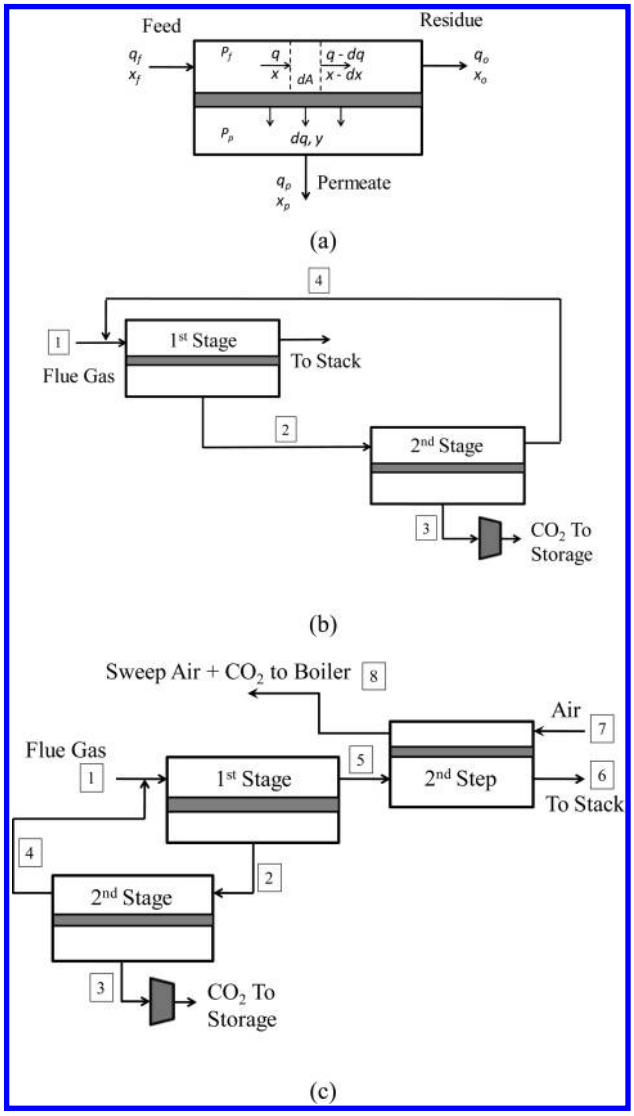


Figure 1. Configurations of membrane systems for CO_2 capture: (a) single-stage module; (b) two-stage ideal cascade; (c) two-stage and two-step system with air sweep.

Single-Stage Membrane Systems. We first estimate the purity of CO_2 product as a function of the CO_2 removal efficiency for single-stage membrane systems for the base case design with a cross-flow pattern. The CO_2 removal efficiency is defined as the percent of CO_2 captured from the flue gas. We then employ sensitivity analysis to evaluate the effects of key parameter variations including the pressure ratio, inlet CO_2 concentration, and membrane CO_2/N_2 selectivity.

Equation 4 shows that the pressure ratio and membrane selectivity are the key parameters affecting CO_2 purity. The pressure ratio was varied from 5 to 20. Beyond this range the process energy requirement became very high, as will be seen in the later energy penalty analysis of two-stage membrane systems. As shown in Figure 2a, the CO_2 product purity decreases nonlinearly with increasing CO_2 removal efficiency. Increasing

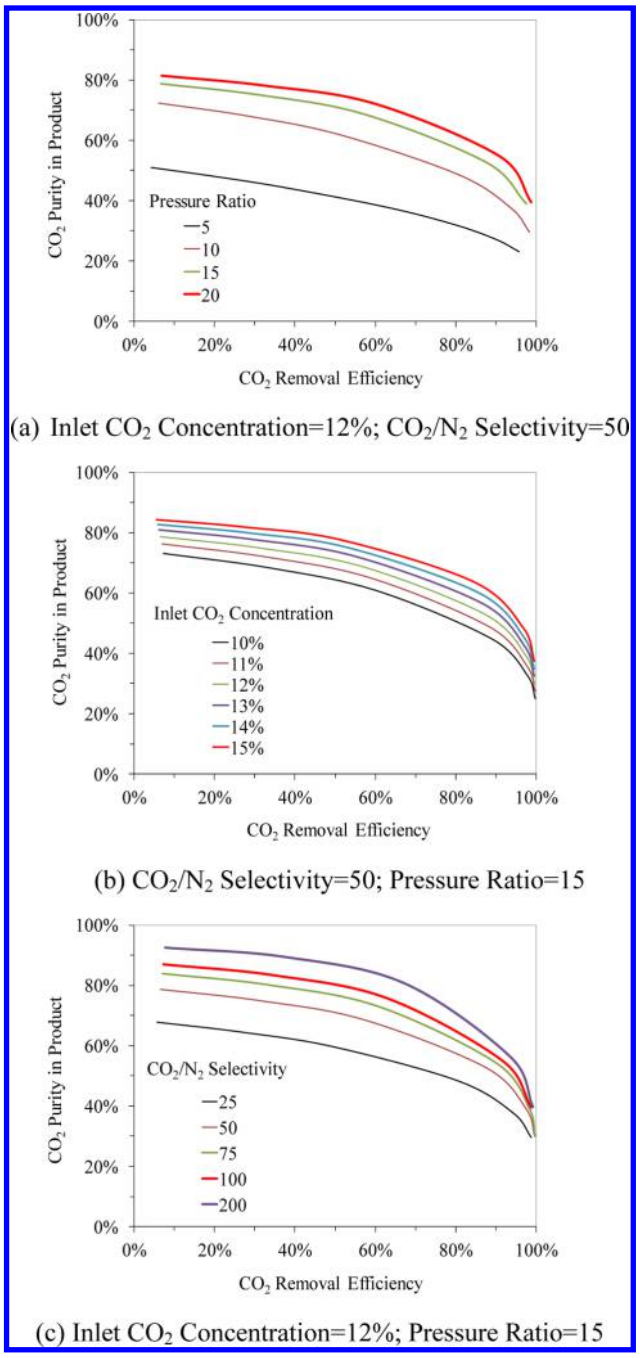


Figure 2. Parametric analysis for single-stage membrane systems: (a) pressure ratio for feed versus permeate sides; (b) inlet CO_2 concentration; (c) CO_2/N_2 selectivity.

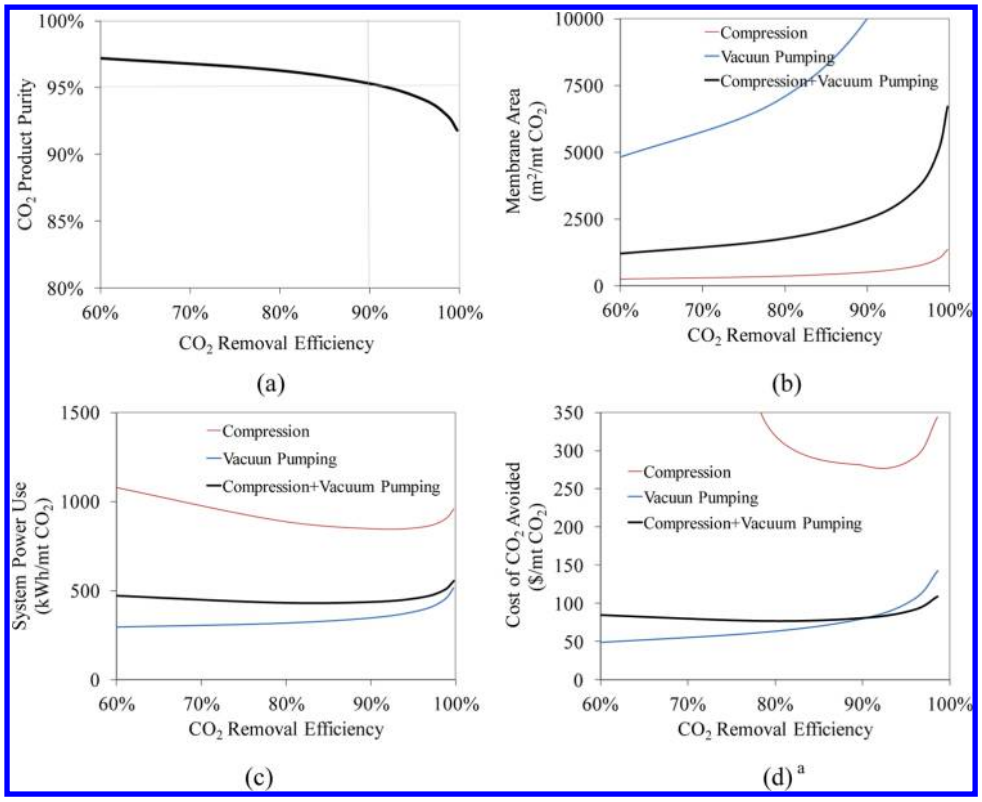


Figure 3. Performance and cost of a two-stage membrane system with three different driving force designs for membrane gas separation at a pressure ratio of 20. In estimating the cost of CO_2 avoided, the reference plant has the same net electrical output (372 MW) as the base capture plant with the two-stage membrane system. The reference plant LCOE is \$64.0/MWh, and the reference CO_2 emission rate is 0.82 kg/kWh. The CO_2 permeance is 1000 gpu for all cases.

the pressure ratio improves the CO_2 purity for any given removal efficiency. The CO_2 purity also increases with higher CO_2 concentrations in the flue gas, as exemplified in Figure 2b, and with increasing CO_2/N_2 selectivity as seen in Figure 2c. However, the results of all parameter variations shown in Figure 2 clearly reveal that single-stage membrane systems are not able to fulfill the desired separation criteria. Thus, we next look at multi-stage membrane configurations.

Two-Stage Membrane Systems. We next investigate the ideal two-stage cascade configuration shown in Figure 1b, where the residue stream (Stream 4) from the second stage is recycled to mix with the feed gas into the first stage. The recycled stream also has the same composition as the feed gas (Stream 1). The CO_2 concentration is enriched in the permeate stream of the second-stage module (Stream 3), and is subsequently compressed to a liquid state for pipeline transport to a storage site. We again estimate the CO_2 product purity (Stream 3) as a function of the overall CO_2 removal efficiency, assuming the same pressure ratios and membrane properties for the two stages under the cross-flow pattern.

A series of driving force designs is analyzed for the base case given in Table 1. Figure 3a shows that the two-stage configuration can indeed fulfill the CO_2 removal and concentration targets at a pressure ratio of about twenty. To provide this required pressure ratio, we looked at three strategies including compression of the feed gas, vacuum pumping at the permeate side, and a combination of those two methods. For the design using compressors alone, the feed-side pressure is 20 bar and the permeate-side pressure is 1 bar; for the design using vacuum pumps alone, the permeate-side pressure is 0.05 bar and the feed-side pressure is still kept at 1 bar; and for the hybrid design, the pressure is 4.0 bar at the feed side and 0.20 bar at the permeate side. Figure 3b and 3c illustrate the trade-offs between membrane area and power use for the three driving force designs. The feed-compression design leads to the smallest membrane area but the highest power use over the range of removal efficiencies. In contrast, the design also using only vacuum pumping has the smallest power use but requires the largest membrane areas. This design assumes that the technical challenges of maintaining a low vacuum pressure are successfully overcome. The hybrid design yields intermediate results for membrane area and power use. For 90% CO_2 capture, the power requirement for the hybrid design corresponds to about 30% of the gross power plant electrical output. Using compressors alone would nearly double that requirement, making it far too large to be affordable, even when an expander is used to recover a significant part of the energy as reported elsewhere.[12] Figure 3d shows that the hybrid design also has the lowest cost of CO_2 avoided (which includes final CO_2 compression, T&S costs) for capture efficiencies at or above 90%. As shown later, however, this system is still more costly than an amine-based capture system.

To improve on the 2-stage design, a recent study by Merkel et al. (2010) suggested using combustion air as a sweep gas in a membrane module to recycle the permeated CO_2 and thus increase the feed concentration of CO_2 into a hybrid membrane capture system.[12] Here, we further investigate a multi-stage, multi-step configuration with CO_2 recycling.

Two-stage, Two-step Membrane Systems with CO_2 Recycling. A two-stage, two-step (TSTS) configuration with sweep gas is shown in Figure 1c. In comparison with the previous two-stage configuration, the residue stream out of the first stage is

subject to further CO_2 removal in the second-step module using a countercurrent flow pattern. The feed air for combustion (Stream 7) is used as a sweep gas in the second-step module, carrying the permeated CO_2 (Stream 8) back to the boiler along with the feed air. The decarbonized flue gas is vented at Stream 6, while the enriched CO_2 product at Stream 3 is compressed for transport and storage.

To achieve CO_2 removal targets, the TSTS membrane system utilizes the hybrid driving force design described above, with the feed-side pressure of the first two stages at 2.0 bar and the permeate-side pressure at 0.20 bar. In the second-step module there is no vacuum pump at the permeate side where the sweep stream is at atmospheric pressure and the sweep air has the same flow rate as the feed stream.[36] As a result, the inlet CO_2 concentration of the flue gas feed stream (Stream 1) increases from the original 12% to about 18%. The permeate stream with sweep air (Stream 8) has about 7% CO_2, while the vented flue gas contains about 1.5% CO_2 (Stream 6), and the product stream has 95+% CO_2 (Stream 3). The membrane properties for this design are given in Table 1 for the base case.

Table 2 summarizes the major performance and cost results. The capture system energy use accounts for 19% of the gross electrical output of the power plant, compared to 30% for the simpler two-stage system. This is a major factor in the overall cost reduction seen for the TSTS system. Overall, the plant LCOE increases by about 61%, from \$61.5/MWh for the reference plant to \$99.1/MWh for the CCS plant with the same net electrical output. Approximately 90% of the total CCS cost (\$37.7/MWh for the TSTS case) represents the cost of capture system; the remaining 10% is the cost of T&S. A lower equipment efficiency of 75% for compressors, expander, and vacuum pumps increases the energy penalty of the capture system to 22% and the LCOE of the capture plant by \$4.8/MWh.

Table 2. Key Process Design Parameters, Performance, and Cost Results for Base Multi-Stage Membrane Systems with 90% CO_2 Capture

variable	two-stage system	two-stage and two-step system with air sweep
process design parameters		
CO_2 concentration into capture system	12%	18%
feed-side pressure (bars)	4.0	2.0
permeate side pressure (bar)		
1st and 2nd stages	0.2	0.2
2nd step	n/a	1.0
CO_2 product purity	95%	95+%
transport and storage cost (\$/mt CO_2)	5.0	5.0
performance and cost results		
membrane area (m^2/mt/h CO_2)	2521	4297
system electrical use (kWh/mt CO_2)	436	275
system energy penalty (% of MWg)	30	19
total cost of CO_2 captured (\$/mt CO_2)[a]	46.8	35.1
cost of CO_2 avoided (\$/mt CO_2)[b]		
(constant net electrical output)[c]	80	52
(constant gross electrical output)[d]	88	56
added cost for CCS (\$/MWh)[c]	56.5	37.7

[a]Calculated as difference in LCOE of plants with and without capture divided by tons CO_2 captured, not including CO_2 transport and storage cost. [b]See Supporting Information for the definition and calculation details. [c]The reference plant has the same net electric output as the plant with CCS. [d]The reference plant has the same gross electric output as the plant with CCS.

A sensitivity analysis was carried out to further examine how different feed-side pressure and pressure ratio designs affect the system performance and cost. In estimating the cost of CO_2 avoided, the reference plant (without CCS) has the same net electrical output as the base CCS plant. The pressure ratio is varied from 6 to 12, whereas the feed-side pressure is elevated from 2.0 bar to 4.0 bar. Figure 4 shows that for any given pressure ratio, using higher feed-side pressures and pressure ratios significantly reduces the required membrane area but increases the system power requirements. As a result of these offsetting effects, the change in the pressure ratio does not significantly affect the cost of CO_2 avoided, although costs are more sensitive to the feed-side pressure than the pressure ratio.

Additional sensitivity analyses were undertaken to characterize the effect of membrane properties on process cost. Here the CO_2/N_2 selectivity is varied from 25 to 75, while the CO_2 permeance is evaluated at 1000, 2500, and 5000 gpu. In this analysis, the feed-side pressure is kept at 2.0 bar, while the permeate-side pressure is held at 0.20 bar for selectivities of more than 30 and at 0.15 bar for the lower selectivity (in order to achieve the target product purity). Figure 5a shows that for a given permeance, the cost of CO_2 avoided decreases slightly up to a selectivity of 35, then remains roughly constant. In contrast, increasing the CO_2 permeance more dramatically reduces the cost of CO_2 avoided by decreasing the required membrane area. For example, for membranes with a selectivity of 50 the cost of CO_2 avoided decreases by approximately 20% when the CO_2 permeance is increased from 1000 to 5000 gpu.

In addition to membrane properties, the assumed membrane price obviously affects cost estimates for membrane systems. Figure 5b shows that effect for three CO_2 permeances. The membrane facilities price accounts for both the membrane module and frame cost components. Lower membrane prices result in lower costs of CO_2 avoided. For example, for a permeance of 1000 gpu the cost of CO_2 avoided decreases from \$67/mt to \$42/mt as the unit price falls from \$150 to \$10 per square meter. At higher values of CO_2 permeance the avoidance cost is less sensitive to the membrane price because the required membrane area decreases with increasing permeance.

Effect of Power Plant Design on Membrane System Cost. Prior membrane studies typically assume a single type of power plant or flue gas stream. However, both the plant type and coal quality are important factors affecting the performance and cost of a PC power plant and the cost of CO_2 capture.[37] Few studies, however, have evaluated their impacts on membrane-based capture systems. Here, we evaluate three power plant types and three coal ranks widely used in U.S. power plant studies. All plants with and without CCS (12 cases altogether) have a net capacity of 550 MW. To reduce the amount of sulfur dioxide (SO_2) emissions entering the capture unit, the flue gas desulfurization systems (FGD) for plants with capture are designed for a removal efficiency of 98%, which exceeds the NSPS requirements assumed for the reference plant. The membrane systems all employ the sweep-based configuration with CO_2 recycling for 90% CO_2 capture.

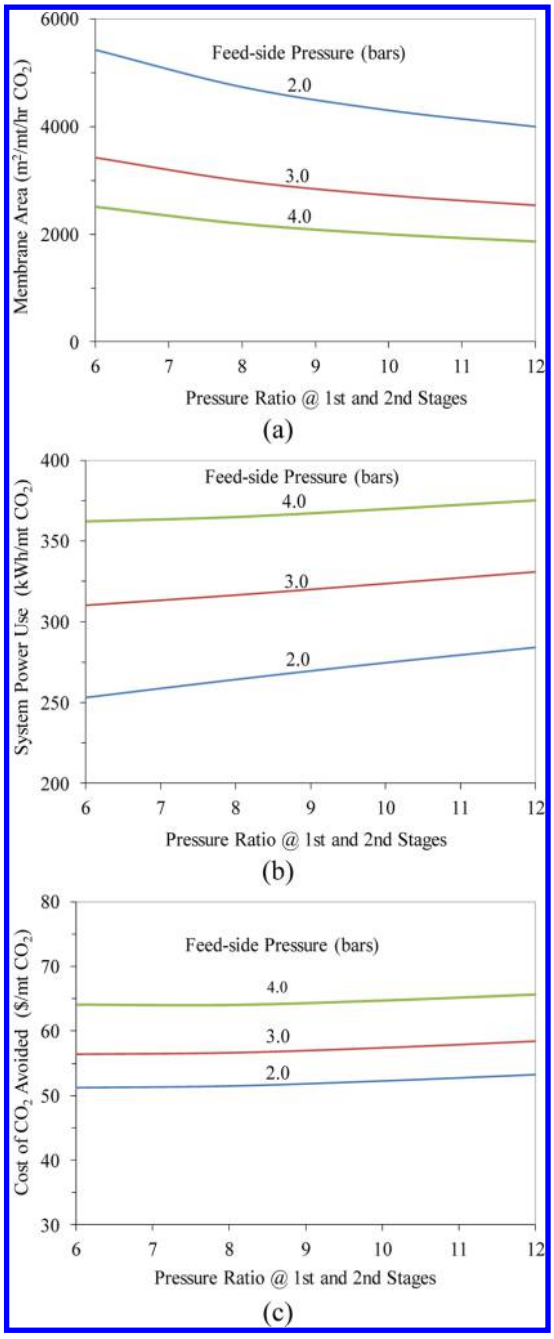


Figure 4. Effects of feed-side pressure and pressure ratio on performance and cost of CO_2 avoided by two-stage and two-step membrane systems with air sweep. The pressure ratio is determined based on whether the membrane system can meet the CO_2 separation targets.

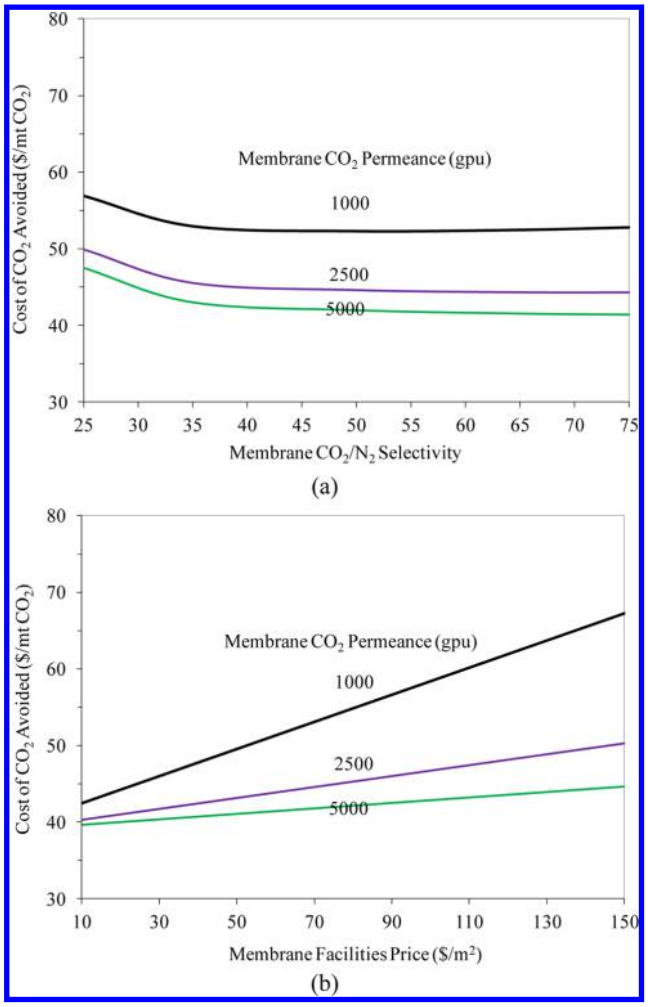


Figure 5. Effects of membrane properties and price on the cost of CO_2 avoided by two-stage and two-step membrane systems with air sweep. All costs are in constant 2010 US dollars. In estimating the cost of CO_2 avoided, the reference plant has the same net electrical output as the base capture plant with the sweep-based TSTS system. The reference plant LCOE is \$61.5/MWh, and the reference CO_2 emission rate is 0.82 kg/kWh. For the cost of CO_2 avoided, approximately \$6−7/mt CO_2 is due to transport and storage costs. The purity of CO_2 product is 95% or more for all scenarios. The membrane CO_2/N_2 selectivity is 50 for the assessments shown in (b).

The three plant types considered include subcritical, supercritical, and ultrasupercritical (USC) units using Pittsburgh #8 coal, the net plant efficiencies without CCS are 36.5%, 38.9%, and 42.9% (HHV basis), respectively.[31] When CCS is employed, the flue gas streams from the three plants using Pittsburgh #8 coal have flow rates of 735, 679, and 599 m^3/s (STP), respectively. Figure 6a further shows the effect of plant type on the added cost for CCS (see SI Table S-6 for details). The inlet flue gas volume strongly affects the capture system size and energy requirements. For the highest-efficiency USC PC plant, the added cost for CCS is approximately \$8/MWh less than the cost at the lower-efficiency subcritical PC plant.

Coal rank and composition similarly affect the flue gas volume and resulting capture costs. The three coal ranks studied here include Pittsburgh #8 bituminous coal, a Wyoming Power River Basin (PRB) sub-bituminous coal, and a North Dakota lignite coal. The HHVs for three coals are 30 840, 19 400, and 14 000 kJ/kg, respectively. For the CCS plants, the flue gas volumes for a supercritical unit fired by each of the three coals are 679, 764, and 855 m^3/s (STP), respectively, along with small differences in CO_2 concentrations. Figure 6b shows the resulting effect on the added cost for CCS. Compared to the base case plant with Pittsburgh #8 coal, the added cost for CCS increases by approximately \$10/MWh at the plant fired by North Dakota lignite.

■ DISCUSSION

This study examined the ability of alternative membrane system configurations to achieve high CO_2 removal efficiency together with high product purity for CO_2 capture at coal-fired power

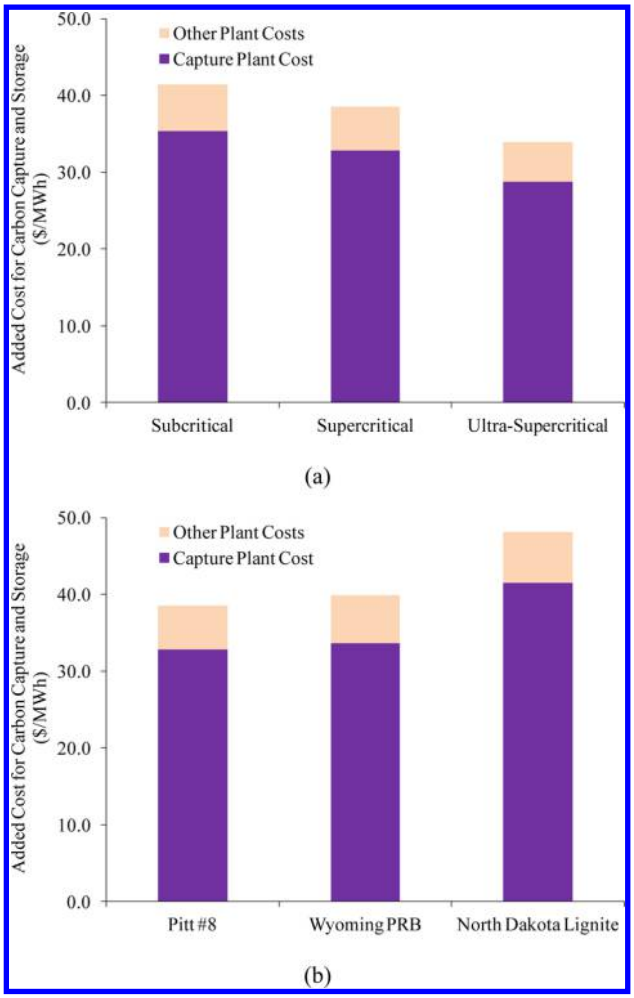


Figure 6. Effects of power plant and coal types on added cost for CO_2 capture and storage by two-stage and two-step membrane systems with air sweep. All costs are in constant 2010 US dollars. The plant size for all cases is 550 MWnet. For the added cost of electricity for CCS, approximately \$5/MWh is due to CO_2 transport and storage costs, and \$1/MWh is due to the added cost of the FGD system relative to the reference plant FGD.

plants. A single-stage process was not able to achieve that level of performance. However, a two-stage membrane system can meet those design goals, albeit with an energy penalty of about 30% of gross plant output. A more promising two-stage, two-step design employs a sweep gas that increases the CO_2 partial pressure of flue gas into the capture system by about six percentage points. This higher concentration reduced the system energy penalty and the cost of CO_2 avoided by roughly one-third.

To further assess this technology, we compared it to a current amine-based postcombustion CCS system modeled using the IECM. Relevant performance and cost details are presented in SI Tables S-4 and S-5. The estimated cost of CO_2 avoided for the sweep-based membrane system with CO_2 recycling was \$9 per metric ton lower than that of the amine-based system for 90% CO_2 capture for the assumptions employed in this analysis. While not a dramatic improvement, this result nonetheless suggests that a membrane-based capture system could be a viable alternative to current liquid solvent processes.

Several paths are available for enhancing the technology's viability. As discussed earlier, increasing the inlet CO_2 partial pressure via use of a sweep gas can reduce the energy required to produce the separation driving force. As suggested in Figure 5, efforts to produce a highly permeable membrane appear to be more economically effective than increasing the CO_2/N_2 selectivity. Lowering the cost of producing highly permeable membranes, together with further improvements in power plant efficiency and the use of high-rank coals, can further reduce the overall cost of membrane-based CCS systems.

A number of important caveats accompany this assessment. First, as noted earlier, the influences of other gas species are not accounted for in this analysis. For example, the presence of CO in real flue gases may reduce the permeability of CO_2 by competitive sorption.[38] Water might adversely impact membrane performance via competitive sorption, plasticization, and aging effects.[25] Moisture levels higher than 20% relative humidity lead to an irreversible flux loss in cellulose acetate materials and then decrease gas permeability.[25] Thus, the common assumption of constant gas permeability identical to that of a pure gas may not apply to all membrane materials and designs. On the other hand, reducing the water content in flue gases may also reduce the energy consumption for vacuum pumps used in membrane capture processes.[39] A pretreatment process using hydrophilic polymer membranes could be considered for flue gas dehydration prior to the CO_2 separation.[39,40] Additional data are thus needed to illuminate the influence of trace gases on membrane system performance and cost.

In addition, the impacts of recycled CO_2 on conventional boiler performance and other unit operations also remain unclear. A computational analysis for boiler combustion indicates the feasibility of retrofitting existing boilers to handle CO_2-enriched air, although for a fixed stoichiometry design, the higher flue gas flow rates produced about 6% higher heat absorption in superheater tubes.[41] Further sensitivity analysis shows that an additional capital cost of 25% for retrofitting the steam generator for CO_2-enriched air would increase the overall LCOE of the base capture plant by \$4.1/MWh. Again, there is a need for experimental or pilot studies to investigate these influences. Pending the availability of more comprehensive data, future analyses can present a more comprehensive picture of membrane system viability via probabilistic analyses[33] that more fully reflect the many uncertain or variable factors that influence techno-economic assessments of emerging technologies for CO_2 capture and storage.

■ ASSOCIATED CONTENT

Supporting Information

Additional text, tables, and figures regarding the IECM, mathematical models, costing method, and cost estimates. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

This research was funded by the U.S. DOE's National Energy Technology Laboratory through support contract 24905.913.ER.1041723. However, the views, findings, and recommendations expressed herein are those of the authors

alone and do not necessarily state or reflect those of the United States Government or any agency thereof.

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