Supporting Information

A Techno-Economic Assessment of Polymer Membrane Systems for Post-combustion Carbon Capture at Coal-fired Power Plants

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This supporting information (SI) provides texts, tables and figures pertaining to: (1) introduction to the Integrated Environmental Control Model; (2) gas separation models under the cross-flow pattern; (3) gas separation models under the countercurrent flow pattern; (4) costing method for membrane systems; (5) cost of CO_2 avoided for multi-stage membrane systems; (6) revenue requirements of plants with membrane systems.

2 Figures and 7 Tables in Supporting Information

S1. Introduction to Integrated Environmental Control Model

The Integrated Environmental Control Model (IECM) is a publicly available computer simulation tool developed by Carnegie Mellon University for the U.S. Department of Energy's National Energy Technology Laboratory.[1] The IECM is developed for preliminary design and analysis of electricity generation options including pulverized coal (PC), integrated gasification combined cycle (IGCC), and natural gas combined cycle (IGCC) systems. This tool provides systematic estimates of the plant-level performance, costs, and environmental emissions of fossil fuel power plants and air pollution control systems. The IECM also offers a variety of carbon capture and storage technologies for PC, IGCC and NGCC plants.

The process performance models in the IECM are formulated based on fundamental mass and energy balances along with empirical data and are further coupled with 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 a variety of environmental control options.[1] The costing method and nomenclature employed in the IECM are based on the Electric Power Research Institute's (EPRI) Technical Assessment Guide (TAG).[2] The IECM also offers the capability to quantify key uncertainties and perform comparative analyses of current and advanced system designs.

S2. Gas Separation Models under Cross-Flow Pattern

Here we briefly summarize the analytical approach to solving the gas separation models under the cross-flow pattern. As shown in Figure S-1, the local permeation rate of a gas component in a binary (CO_2 and N_2) membrane system over a differential membrane area is described as: [3]

$$-y dq = J_{CO2} dA = \frac{P^*_{CO2}}{\delta}\left[xP_f - yP_p\right]dA \tag{S-1}$$

$$-(1-y)dq = J_{N2} dA = \frac{P^*_{N2}}{\delta}\left[(1-x)P_f - (1-y)P_p\right]dA \tag{S-2}$$

Diving two equations above leads to:

$$\frac{y}{1-y} = \frac{\alpha(1-y/\phi)}{(1-x)-(1-y)/\phi} \tag{S-3}$$

Where A is the membrane area(cm^2); J is the volumetric flux(cm^3/(cm^2.s)); P^* is the gas permeability(cm^3.cm/(s.cm^2.cmHg)); P_f and P_p are the pressures in the feed and permeate sides(cmHg); q is the gas flow rate(cm^3/s); x and y are the concentrations of CO_2 in the feed and permeate streams (%); δ is the membrane thickness (cm); α is the permeability ratio (P^*_{CO2}/P^*_{N2}) for CO_2 versus N_2 gases, and also is called membrane selectivity; ϕ is the pressure ratio (P_f/P_p) for feed versus permeate sides. Eq. S-3 relates the concentrations of CO_2 in both feed and permeate streams at a point along the pathway. Weller and Steiner applied ingenious transformations to obtain an analytical solution to the governing equations as:[3]

$$\frac{(1-\theta^*)(1-x)}{1-x_f} = \left(\frac{u_f - E/D}{u - E/D}\right)^R \left(\frac{u_f - \alpha + F}{u - \alpha + F}\right)^S \left(\frac{u_f - f}{u - f}\right)^T \tag{S-4}$$

Furthermore, the membrane area required was obtained as: [1]

$$A_m = \frac{tq_f}{P_f P^*_{N2}} \int_{i_o}^{i_f} \frac{(1-\theta^*)(1-x)}{(f_i - i)\left[\frac{1}{1+i} - \frac{1}{\phi}\left(\frac{1}{1+f_i}\right)\right]} di \tag{S-5}$$

Where:

$$\theta^* = 1 - \frac{q}{q_f}$$

$$i = \frac{x}{1-x}$$

$$u = -Di + \left(D^2 i^2 - 2Ei + F^2\right)^{0.5}$$

$$D=0.5\left[\frac{(1-\alpha)}{\phi}+\alpha\right]$$

$$E=\frac{\alpha}{2}-DF$$

$$F=-0.5\left(\frac{1-\alpha}{\phi}-1\right)$$

$$R=\frac{1}{2D-1}$$

$$S=\frac{\alpha(D-1)+F}{(2D-1)\left(\frac{\alpha}{2}-F\right)}$$

$$T=\frac{1}{1-D-E/F}$$

$$f_i=(Di-F)+(D^2i^2-2Ei+F^2)^{0.5}$$

Given feed compositions, membrane properties, feed- and permeate- side pressure designs and membrane module stage-cut, the CO_2 concentrations of permeate and residue streams and membrane area can be solved using the analytical approach above via an iterative process.

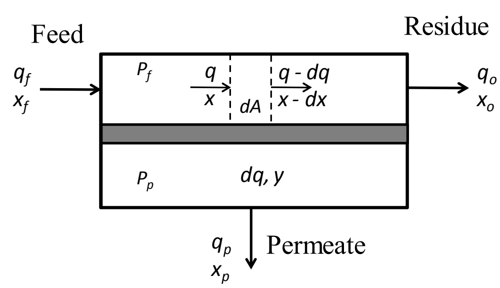

Figure S-1 Cross-flow Membrane Module

S3. Gas Separation Models under Countercurrent Flow Pattern

This study adopted a widely-used mathematical framework developed by Pan and Habgood to model binary gas separation under the countercurrent gas flow pattern with a sweep gas. The modeling framework was derived from mass balances with major assumptions:[4] (a) two permeable components; (b) constant gas permeability independent of pressure, which is the same as that of the pure gas; (c) negligible pressure drop; and (d) negligible diffusion along the flow path, and plug flow appearing in both the feed and permeate sides. Here are the governing equations: [4]

$$\frac{q_f}{q_w} = \frac{y - x_w + F_w(y - y_w)}{y - x} \tag{S-6}$$

$$\frac{q_p}{q_w} = \frac{x_w - x + F_w(y_w - x)}{y - x} \tag{S-7}$$

$$u = \frac{u_w(y - x)}{y - x_w + F_w(y - y_w)} \tag{S-8}$$

$$v = \frac{v_w F_w(y - x)}{x_w - x + F_w(y_w - x)} \tag{S-9}$$

$$\frac{dy}{dx} = \frac{y - x_w + F_w(y - y_w)}{x - x_w + F_w(x - y_w)} \times \left\{\frac{\alpha(1 - y)(x - \gamma y) - y[(1 - x - u) - \gamma(1 - y - v)]}{\alpha(1 - x)(x - \gamma y) - x[(1 - x - u) - \gamma(1 - y - v)]}\right\} \tag{s-10}$$

The dimensionless membrane area is defined as ($\frac{\left(P^*_{N2}/_{\delta}\right)P_f A}{q_w}$), and is estimated as:

$$\frac{dR^w}{dx} = \frac{y - x_w + F_w(y - y_w)}{(x - y)\{\alpha(1 - x)(x - \gamma y) - x[(1 - x - u) - \gamma(1 - y - v)]\}} \tag{S-11}$$

As shown in Figure S-2, the boundary conditions are:

$q_f = q_f^o$, x=x_f, μ=u_f at R^w=R_f^w (negative, since the feed and permeate streams flow in opposite directions); q_p=q_{p_w} (negative), y=y_w, v=v_w at R^w=0.

Here: F_w is the ratio of sweep gas- versus residue-flow rate in the countercurrent flow pattern; R^w is the dimensionless membrane area with a reference point at the residue end; q_f is the feed-side flow rate (cm^3/s); q_p is the permeate-side flow rate(cm^3/s); q_w is the residue flow rate(cm^3/s); u is the molar concentration of nonpermeable components in the feed-side stream; v is the molar concentration of nonpermeable components in the permeate-side stream; x and y are the concentrations of CO_2 in the feed and permeate streams (%); γ is the ratio of permeate-versus feed-pressure; α is the CO_2/N_2 selectivity. In addition, the subscript w refers to conditions at the residue end, and the subscript f refers to conditions at the feed inlet end.

There are seven variables (q_w, x_w, u_w, q_{p_f}, y_f, v_f, R_f^w) together to satisfy the six governing equations. We may treat one desired variable such as x_w, as a known variable in terms of specific design conditions or requirements, and then use the trial-and-error method to determine the rest unknown variables.

When there is no sweep gas used in the permeate side, the permeate concentration at the residue end is determined as: [4]

$$\frac{y_w}{1-y_w} = \frac{\alpha(x_w - \gamma y_w)}{1 - x_w - u_w - \gamma(1-y_w)} \tag{S-12}$$

The value of $\left(\frac{dy}{dx}\right)$ appears to be indeterminate at the residue end, thus its value is determined in terms of L'Hopital's rule via differentiating its numerator and denominator with respect to x, respectively. Then, there is:

$$\left(\frac{dy}{dx}\right)_w = \frac{(y_w - x_w)[\alpha - (\alpha-1)y_w] - u_w y_w}{\alpha(1-x_w)(x_w - \gamma y_w) - x_w[(1-x_w-u_w) - \gamma(1-y_w)] - (y_w - x_w)[(\alpha-1)(2\gamma y_w - x_w - \gamma) - 1 + u_w]} \tag{S-13}$$

After y_w and Equation S-13 are obtained, Equation S-10 can be solved. The toolbox of Ordinary Differential Equation (ODE) in MATLAB is used to solve the differential equations for modeling membrane CO_2/N_2 separation under the countercurrent flow pattern.

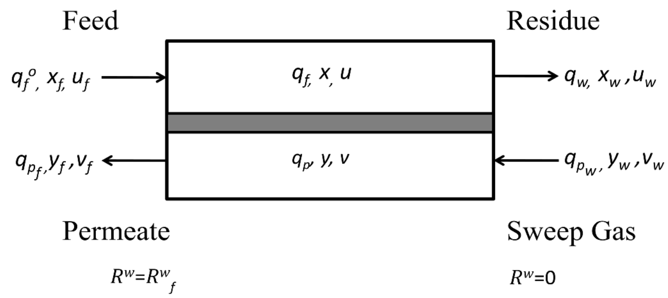

Figure S-2 Countercurrent Membrane Module

S4. Costing Method

The costing framework of this study is based on the EPRI's TAG.[2] This session presents more details of estimating individual cost categories discussed in the main paper, including the capital, operating and maintenance (O&M) costs. The direct capital cost estimation for the major components is referred to previous studies.[5] Tables S-1 and S-2 summarize the approaches to capital, fixed and variable O&M cost estimates for membrane systems, respectively. The nomenclature is explained in detail in the EPRI's TAG.

Table S-1 Capital Cost Estimation for Membrane Systems

Process Area[4]	**Method**[a]	**Plant Costs**	**Method**
Membrane module (1)	$A_m \cdot c_m$	Process facilities capital (8)	
Membrane frame (2)	$\left(\frac{A_m}{2000}\right)^{0.7} \cdot c_{mf}$	General facilities capital (9)	10% of PFC
Compressors (3)	$e_{cpr} \cdot c_{cpr}$	Eng. & home office fees (10)	7% of PFC
Expander (4)	$e_{exp} \cdot k_{exp} \cdot F_h$	Project contingency cost (11)	15% of PFC
Vacuum pumps (5)	$e_{vp} \cdot c_{vp}$	Process contingency cost (12)	5% of PFC
Heat exchangers (6)	$A_{HeEx} \cdot c_{HeEx}$	Interest Charges (13)	
CO_2 product compression (7)	$e_{cmp} \cdot c_{cmp}$	Royalty fees (14)	0.5% of PFC
		Preproduction (startup) cost (15)	
		Inventory capital (16)	0.5% of TPC[b]
Process facilities capital (PFC) (8)	**(1) +(2)+….+ (7)**	**Total capital requirement (TCR)**	**(8) + (9) +…. + (16)**

[a] Notation: A_m: membrane area (m^2); c_m: unit cost of membrane module ($\$/m^2$); c_{mf}: referred frame cost (M\$ 0.238)[5]; e_{cpr}: compressor power use (kW); c_{cpr}: installed unit cost (\$/kW); e_{exp}: expander power use (kW); k_{exp}: unit cost (\$/kW); F_h: equipment cost factor for housing, installation, etc (1.8)[5]; e_{vp}: vacuum pump power use (kW); c_{vp}: installed unit cost of vacuum pump (\$/kW); A_{HeEx}: heat exchanger area (m^2); c_{HeEx}: installed unit cost of heat exchanger ($\$/m^2$); e_{cmp}: CO_2 product compression power use (kW); and c_{cmp}: installed unit cost of CO_2 product compression (\$/kW).

[b] TPC= total plant cost, which is the sum of (8)+(9)+(10)+(11)+(12).

Table S-2 Operating and Maintenance (O&M) Cost Estimation for Membrane Systems

Variable Cost Component	**Method**[a]	**Fixed Cost Component**	**Method**
Material replacement (1)	$(A_m \cdot \vartheta) \cdot c_{rm}$	Operating labor (4)	
Electricity (2)	MWh $\cdot$ COE	Maintenance labor (5)	40 % of TMC
CO_2 transport & storage (when considered) (3)	$m_{CO2} \cdot c_{T\&S}$	Maintenance material (6)	60 % of TMC
		Admin. & support labor (7)	30 % of Total labor
Variable O&M Costs	**(1)+(2)+(3)**	**Fixed O&M Costs**	**(4)+(5)+(6)+(7)**

[a] Notation: A_m: membrane area (m^2);c_{rm}: material replacement cost ($\$/m^2$); ϑ: annual material replacement rate (%); MWh: annual system power use (MWh); COE: cost of electricity (\$/MWh); m_{CO2}: annual CO_2 captured (mt/yr); $c_{T\&S}$: CO_2 product transport and storage cost (\$/mt CO_2).

S5. Cost of CO_2 Avoided for Multi-stage Membrane and Amine Systems

The levelized cost of electricity (LCOE) for a power plant is calculated in constant dollars as:

$$LCOE = \frac{FCF \cdot TCR + FOM}{(CF \cdot 365 \cdot 24) \cdot \mathrm{MWh_{net}}} + VOM \quad \text{(S-14)}$$

Where $LCOE$ is the revenue requirement (\$/MWh); CF is the levelized capacity factor (fraction); FCF is the fixed charge factor (fraction); FOM is the annual fixed O&M cost (\$/yr); MWh_{net} is the net plant electrical output (MWh); TCR is the total capital requirement (\$); VOM is the annual variable O&M cost (\$/MWh).

The cost of CO_2 avoided (\$/mt) is a relative measure quantifying the cost of carbon capture and storage (CCS), and is defined as:

$$Cost\ of\ CO_2\ Avoided = \frac{COE_{ccs} - COE_{ref}}{ER_{ref} - ER_{ccs}} \quad \text{(S-15)}$$

Where *COE* is the cost of electricity for the reference and CCS plants (\$/MWh); and *ER* is the CO_2 emission intensity for the reference and CCS plants (mt/MWh). Table S-3 presents the estimation details for the two membrane systems.

For comparisons between amine and membrane technologies applied to supercritical coal-fired power plants, the cost of CO_2 avoided is also estimated for the amine-based capture system using the IECM. The amine-based capture plant has the same net power output as the base capture plant using the sweep-based two-stage, two-step membrane configuration (given in Table 2 of main paper). Table S-4 gives the major technical and economic assumptions of amine-based capture system for the IECM modeling. The cost of CO_2 avoided by the amine-based capture system is presented in Table S-5.

Table S-3 Estimation of Costs of CO_2 Avoided for Multi-stage Membrane Systems

Variable	**Reference PC Plant**[a]	**PC Plants with CO_2 Capture**	
		Two-stage system	Two-stage and two-step system with air sweep
Gross electrical output (MWg)	588	588	588
Net electrical output (MW)	550	372	438
Capacity factor	75%	75%	75%
Electricity price ($/MWh)	43.2	43.2	43.2
Total annual levelized cost (2010M$/yr)	211.2	294.7	285.5
Cooling tower	11.2	11.2	11.2
Base plant	159.1	159.1	159.1
Selective catalytic reduction	5.3	5.3	5.3
In-furnance control	1.5	1.5	1.5
Electrostatic precipitator device	4.6	4.6	4.6
Flue gas desulfurization	29.4	29.4	29.4
<u>CO_2 Capture and Storage</u>			
CO_2 capture system		120.3	92.5
CO_2 product T&S[b]		13.4	13.4
Internal electrical cost assigned to base plant[c]		-50.6	-31.8
Plant cost of electricity ($/MWh)[c]	58.4	120.5	99.1
Plant CO_2 emission rate (mt/MWh)	0.82	0.11	0.10
Cost of CO_2 avoided ($/mt)		**88**	**56**

[a] The reference pulverized coal (PC) power plant is developed using the Integrated Environmental Control Model (IECM) Version 7.

[b] It is assumed that the cost of CO_2 transport and storage (T&S) included is $5/mt CO_2.

[c] The cost model charges each technology for the internal use of electricity and treats the charge as a credit for the base plant. When reporting O&M costs for individual components of the plant, these energy costs are taken into consideration. However, for the total plant they balance out and have no net effect on the overall plant O&M costs.

Table S-4 Major Technical and Economic Assumptions of Amine-based Capture System

Variable	Value
FG+ sorbent concentration (wt %)	30
Liquid-to-gas ratio (ratio)	3.09
Regeneration heat requirement(kJ/kg CO_2)	3533
Heat-to-electricity efficiency (%)	18.7
Nominal sorbent loss (kg/mt CO_2)	0.3
Solvent pumping head(MPa)	0.21
CO_2 product purity (%)	99.5
CO_2 product pressure (MPa)	13.79
CO_2 Compressor Efficiency (%)	80
General facilities capital (% of PFC)	10
Engineering & home office fees (% of PFC)	7
Project contingency cost (% of PFC)	15
Process contingency cost (% of PFC)	5
Royalty fees (% of PFC)	0.5
Pre-production costs	
Months of fixed O&M	1
Months of variable O&M	1
Misc. capital cost (% of TPI)	2
Inventory capital (% of TPC)	0.5
Sorbent cost ($/mt)	2476
Number of operating jobs (jobs/shift)	2
Number of operating shifts (shifts/day)	4.75
Total maintenance cost (TMC) (% of TPC)	2.5
Maint. cost allocated to labor (% of TMC)	40
Administrative & support cost (% total labor)	30
CO_2 transport and storage ($/mt)	5

Table S-5 Estimation of Cost of CO_2 Avoided by Amine-based Capture System

Variable	Reference Plant (no CCS)	CCS Plant
Plant Capacity Factor (%)	75	75
Fixed Charge Factor	0.113	0.113
Gross electrical output (MW)	469	539
Net electrical output (MW)	438	438
CO_2 emission rate (kg/kWh-net)	0.82	0.11
Plant cost of electricity (COE) (2010$/MWh-net)	61.5	104.8
Plant COE Increase with CCS (%)		**70.4**
Cost of CO_2 avoided ($/mt)		**61.3**

S6. Levelized Cost of Electricity of A Membrane System

Tables S-6 and S-7 present the plant levelized cost of electricity (COE) for the plants with and without CCS, and the added cost for membrane-based CCS as a function of plant type and coal type, respectively.

Table S-6 Plant Cost of Electricity and Added Cost for Two-stage and Two-step Membrane System with Air Sweep as a Function of Plant Type

Case	Variable	Pulverized Coal Plant Type		
		Subcritical	Supercritical	USC
Reference Plant	Coal type	Pitt #8	Pitt #8	Pitt #8
	Steam cycle heat rate (Btu/kWh)	7790	7359	6705
	Gross plant power output (MW)	591.3	588.0	584.5
	Net plant power output (MW)	550.0	550.0	550.0
	Net plant efficiency (HHV, %)	36.5	38.9	42.9
	Plant capacity factor	75%	75%	75%
	Flue gas flow rate (S.T.P. m^3/s)	569	535	484
	Flue gas CO_2 concentration (mole-%)	11.9	11.9	11.9
	Plant levelized COE (2010$/MWh)	57.8	58.1	57.6
Capture Plant	Gross plant power output (MW)	763.8	746.5	723.6
	Net plant power output (MW)	550	550	550
	Flue gas flow rate (S.T.P. m^3/s)	735	679	599
	Capture system power use (MW)	154	142	126
	System membrane area ($10^6 \times m^2$)	2.41	2.24	1.96
	Plant levelized COE (2010 $/MWh)	99.3	96.6	91.5
	Added cost for CCS (2010 $/MWh)	**41.5**	**38.5**	**33.9**

Table S-7 Plant Cost of Electricity and Added Cost for Two-stage and Two-step Membrane System with Air Sweep as a Function of Coal Type

Case	Variable	Coal Type: Pitt #8	Coal Type: Wyoming PRB	Coal Type: North Dakota Lignite
Reference Plant	Coal heating value (Btu/lb)	13260	8340	6020
	Gross plant power output (MW)	588.0	596.0	604.7
	Net plant power output (MW)	550.0	550.0	550.0
	Plant capacity factor	75%	75%	75%
	Flue gas flow rate (S.T.P. m^3/s)	535	589	642
	Flue gas CO_2 concentration (mole-%)	11.9	11.8	11.5
	Plant levelized COE (2010$/MWh)	58.1	49.4	62.2
Capture Plant	Gross plant power output (MW)	746.5	773.5	805.3
	Net plant power output (MW)	550.0	550.0	550.0
	Flue gas flow rate (S.T.P. m^3/s)	679	764	855
	Capture system power use (MW)	142	158	174
	System membrane area ($10^6 \times m^2$)	2.24	2.53	2.86
	Plant levelized COE (2010 $/MWh)	96.6	89.2	110.4
	Added cost for CCS (2010 $/MWh)	**38.5**	**39.9**	**48.2**

References

(1) *Carnegie Mellon University's Integrated Environmental Control Model (IECM) Website*. http://www.cmu.edu/epp/iecm/index.html (accessed May 2012).

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