

Simulated economics assessment of hollow fiber CO₂ separation modules

Yoon Ah Jang*, Young Moo Lee**, and Yeong-Koo Yeo*[†]

*Department of Chemical Engineering, Hanyang University, Seoul 133-791, Korea

**WCU Department of Energy, Hanyang University, Seoul 133-791, Korea

(Received 21 February 2011 • accepted 12 August 2011)

Abstract—Various conditions under which the hollow fiber membrane separation system would be the optimal selection are investigated in terms of cost effectiveness. Numerical simulation is carried out to examine the effects of different configurations such as single-stage, two-stage and three-stage CO₂ separation processes. In particular, the hollow fiber membrane processes for CO₂ separation with vacuum pumps, heat exchangers, coolers and compressors to provide pressurized feed streams are analyzed. Operating costs are evaluated and compared numerically for the processes with and without recycle streams to compare feasibility for commercial implementation while maintaining the purity and recovery ratio as high as possible.

Key words: Economic Analysis, Modeling of Separation Process, Carbon Dioxide, Hollow Fiber Membrane

INTRODUCTION

Electricity production from fossil fuel based power plants will be challenged by growing concerns that anthropogenic emission of green-house gases such as carbon dioxide is contributing to the global climate change. Today, worldwide, all the existing coal-fired power plants emit about 2 billion tons of CO₂ per year. The regulation of the carbon dioxide emissions implies the development of specific CO₂ capture technologies that can be retrofitted to existing power plants as well designed into new plants with the goal to achieve 90% of CO₂ capture, limiting the increase in cost of electricity to no more than 35% [1]. Therefore, the recovery of CO₂ from large emission sources is a formidable technological and scientific challenge which has received considerable attention for several years [2-4]. The cost of CO₂ separation occupies 70-80% of total cost required in CO₂ recovery and storage. Thus reduction of CO₂ separation cost makes the greatest contribution to improve the economics of the CO₂ treatment industry.

Recently, membrane processes have found increased use in environmental-based applications including water and wastewater treatment, materials recovery, separation and clarification. Membrane separation processes are very attractive because of their lower energy consumption compared to other technologies. In fact, membrane-based CO₂ separation has many advantages over other separation methods: 1) it is energy effective because phase transition is not involved; 2) operation and maintenance are simple because related facilities are compact and small; 3) adjustment of operating conditions and scale-up are relatively easy [5]. Recently, a new membrane with high permeability (about 500 times as high as that of acetate membrane and about 300 times as high as that of polyimide membrane) was developed to enhance economics of CO₂ separation by membranes [6]. Results on the design and operation of multiple-stage membrane separation processes were reported for

CO₂ separation with recovery rate of 90% and mole fraction of 99% in the power generation plant using LNG feed [7].

Although much work has been published on the physical performance of membrane processes, there is relatively little literature and experience with regard to the design and optimization of these processes from an economic point of view. This is due in part to the large number of design and operating variables that come into the selection and design of membrane systems. The economics of carbon capture and storage schemes is currently a subject of great interest, as evidenced by the many recent publications. Hendriks, Wildenborg, Blok and Floris van Wees (2000) estimated costs for CO₂ removal projects in the Netherlands under various scenarios. For a range of fuel prices and discount rates their estimated capture cost was as follows: natural gas combined cycle U.S. \$ 41-66 and furnace/combined heat and power up to U.S. \$45 [8]. Van Der Sluijs, Hendriks and Blok (2003) examined with a computer program based on the cross flow permeation model for membranes several parameters which are optimized to give the lowest specific CO₂ mitigation costs. With gas separation membranes commercially available, the minimum attainable specific mitigation costs are calculated to be U.S. \$48 per ton of CO₂ avoided (at 50% CO₂ purity, 75% CO₂ recovery). When restrictions are posed to the purity of CO₂ (95%) and the degree of CO₂ recovery (90%), this figure is much higher: US\$71 per ton of CO₂ avoided [9]. Ho, Allinson and Wiley (2008) investigated how costs for CO₂ capture using membranes can be reduced by operating under vacuum conditions [10]. The flue gas is pressurized to 1.5 bar, whereas the permeate stream is at 0.08 bar. Under these operating conditions, the capture cost is U.S. \$54/ton CO₂ avoided compared to U.S. \$82/ton CO₂ avoided using membrane processes with a pressurized feed. This is a reduction of 35% [10].

So far, various membrane separation schemes for the recovery of CO₂ have been proposed, but economic analysis of these methods has not received much attention. In this paper, we investigate the different conditions under which the hollow fiber system would be the optimal selection in terms of cost efficiency. Numerical simu-

[†]To whom correspondence should be addressed.
E-mail: ykyeo@hanyang.ac.kr

lation is used to examine the effects of different configurations such as single-stage, two-stage and three-stage CO₂ separation processes. In particular, the hollow fiber membrane separation processes is analyzed to provide pressurized feed streams with the vacuum pump and the compressor. Operating costs are evaluated and compared numerically for the processes with and without recycle streams to identify the optimal operating conditions while maintaining the maximum purity and recovery ratio.

MODELS

The cost required in the CO₂ recovery occupies 70-80% of the total cost for the CO₂ recovery and storage system. The hollow fiber module explored in the present study can be configured as the counter-current flow type or co-current flow type. In both types, we have two options: 1) the helium gas is fed in the opposite direction with the feed gas; 2) the helium stream is depressurized at the permeate outlet. If the size of the module is relatively small, there is little difference between the counter-current flow type and the co-current flow type, while the difference becomes significant as the module size increases. For the counter-current flow type, the difference in the driving force between the inlet and the outlet is nearly constant and small while, for the case of the co-current flow type, we can see the decrease in the driving force from the inlet to the outlet. In general, the counter-current flow type has some advantages over the co-current flow type in efficiency and operational flexibility. The separation module investigated in this work is perfect-mixing model as shown in Fig. 1. In the perfect-mixing model, compositions exhibit minimal variations even for low recovery rate.

1. Single-stage Module

Fig. 2 shows the model for the single-stage separation process. The overall mass balance is given by

$$q = q_o + q_p \tag{1}$$

where q is molar flow rate and subscripts f, o and p denote feed, rejected and permeate fluxes, respectively. The stage-cut, θ , which is the fraction of transmitted part from the feed, is defined as

$$\theta = \frac{q_p}{q_f} \tag{2}$$

The rate of CO₂ transmission or diffusion can be written as

$$\frac{q_A}{A_m} = \frac{q_p y_p}{A_m} = \left(\frac{P'_A}{t}\right)(p_h x_o - p_l y_p) \tag{3}$$

Similar equations can be written for N₂ as follows:

$$\frac{q_B}{A_m} = \frac{q_p(1-y_p)}{A_m} = \left(\frac{P'_B}{t}\right)[p_h(1-x_o) - p_l(1-y_p)] \tag{4}$$

where p is the pressure, A_m is the membrane area, (P'_A/t) is the per-

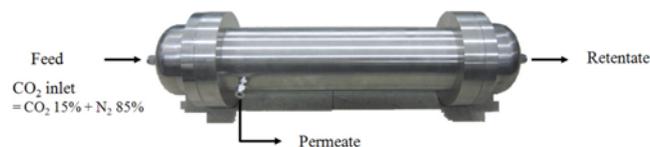
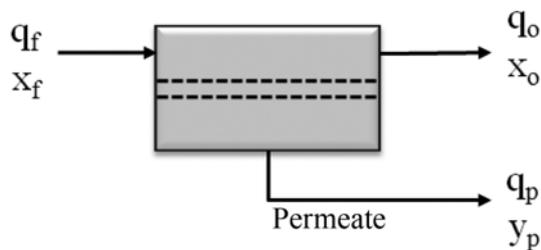
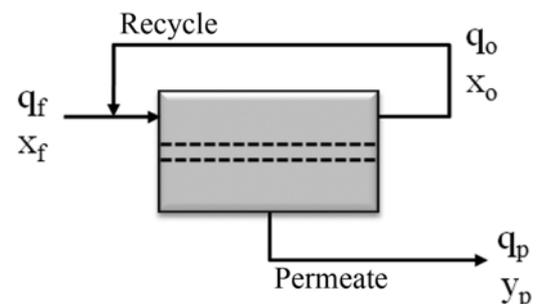


Fig. 1. Membrane separation module used in the permeation test.



(a) Single-stage without recycle



(b) Single-stage with recycle

Fig. 2. Single-stage separation module with and without recycle.

meability of CO₂ and subscripts h and l denote feed and permeate flows, respectively. Division of Eq. (3) by Eq. (4) gives

$$\frac{y_p}{1-y_p} = \frac{\alpha^* [x_o - (p_l/p_h)y_p]}{(1-x_o) - (p_l/p_h)(1-y_p)} \tag{5}$$

The above equation shows the relation between the permeate composition y_p and that of rejected solution x_o. The ideal separation factor α^* is defined as

$$\alpha^* \equiv \frac{P'_A}{P'_B} \tag{6}$$

The overall CO₂ balance is

$$q_f x_f = q_o x_o + q_p y_p \tag{7}$$

Rearrangement of Eq. (7) gives

$$x_o = \frac{x_f - \theta y_p}{(1-\theta)} \text{ or } y_p = \frac{x_f - x_o(1-\theta)}{\theta} \tag{8}$$

Substitution of q_p = θq_f (from Eq. (2)) into Eq. (3) and after some rearrangement gives

$$A_m = \frac{\theta q_f y_p}{(P'_A/t)(p_h x_o - p_l y_p)} \tag{9}$$

Usually values of x_f, θ , α^* , p_l/p_h are given (or known) and y_p, x_o and A_m are calculated. y_p can be obtained from the quadratic formula as

$$y_p = \frac{-b_1 + \sqrt{b_1^2 - 4a_1c_1}}{2a_1} \tag{10}$$

where

$$a_1 = \theta + \gamma_2 - \gamma_2 \alpha^* - \gamma_2 \theta - \alpha^* \theta + \gamma_2 \alpha^* \theta$$

$$b_1 = 1 - \theta - x_f - \gamma_2 + \gamma_2 \theta + \alpha^* \theta + \gamma_2 \alpha^* - \gamma_2 \alpha^* \theta + \alpha^* x_f$$

$$c_i = -\alpha^* x_f$$

Using y_p , values of x_o and A_m can be obtained from Eqs. (8) and (9), respectively. The recovery ratio is defined by

$$\text{recovery} = \frac{q_p y_p}{q_f x_f} \quad (11)$$

In the single-stage module with recycle shown in Fig. 2, the rejected gas is recycled to be combined with the feed stream. Thus, the total feed flow rate and the mole fraction of CO₂ in the feed are given by

$$q_{f1} = q_f + q_o \quad (12)$$

$$x_{f1} = \frac{q_f x_f + q_o x_o}{q_f + q_o} \quad (13)$$

2. Multi-stage Module

For multi-stage modules, additional mass balances are required in-between individual stage modules, which are modeled by aforementioned single stage equations. Figs. 3 and 4 show multi-stage membrane separation configurations. In the multi-stage module without recycle, the permeate flux from the previous stage becomes the feed flux of the following stage. In the multi-stage module with recycle, the rejected flux from one stage becomes part of feed flux for the previous stage. For a two-stage system, the total feed flow rate and the mole fraction of CO₂ are given by

$$q_{f1} = q_f \quad (14)$$

$$x_{f1} = x_f \quad (15)$$

$$q_{f2} = q_{o1} \quad (16)$$

$$x_{f2} = y_{p1} \quad (17)$$

For a two-stage system with recycle, the total feed flow rate and the mole fraction of CO₂ which are recycled and fed into the 1st-stage are given by

$$q_{f2} = q_f + q_{o2} \quad (18)$$

$$x_{f2} = \frac{q_f x_f + q_{o2} x_{o2}}{q_f + q_{o2}} \quad (19)$$

Total feed flow rate and the mole fraction of CO₂ at the inlet of the 2nd-stage are also represented by Eq. (16) and (17).

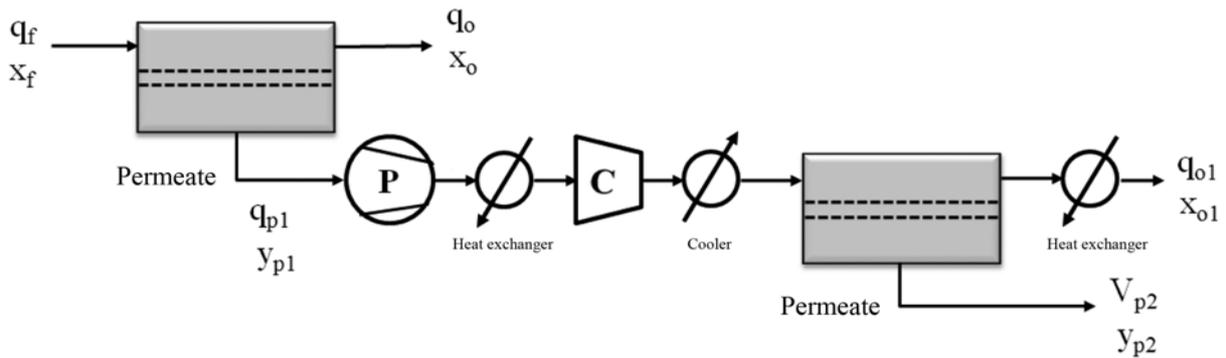
For the three-stage system, total feed flow rate and the mole fraction of CO₂ which are fed into the inlets of the 1st- and 2nd-stage are given by Eqs. (14)-(17), while those which are fed into the inlet of the 3rd-stage are given by

$$q_{f3} = q_{o2} \quad (20)$$

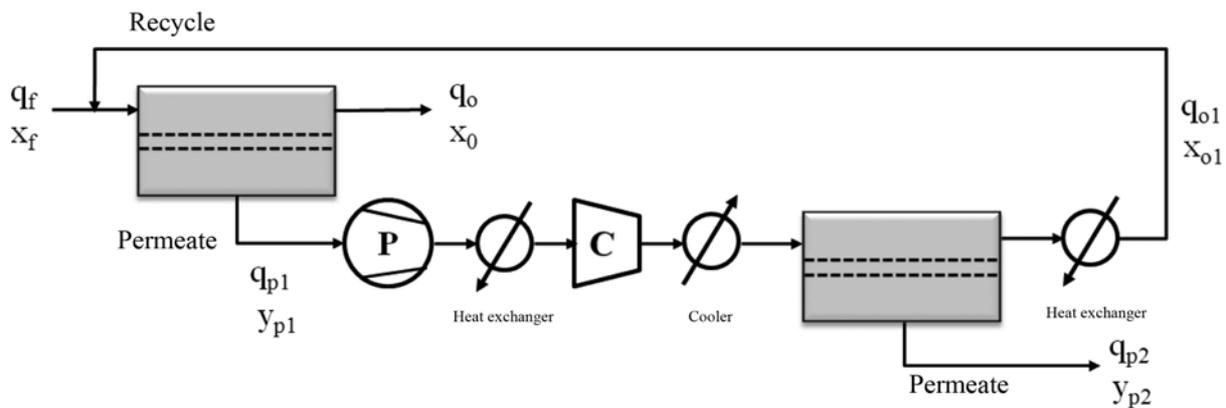
$$x_{f3} = y_{p2} \quad (21)$$

For the three-stage system with recycle, total feed flow rate and the mole fraction of CO₂, which are recycled and fed into the inlet of the 1st-stage, are given by Eq. (18) and (19), while those which are recycled and fed into the inlet of the 2nd-stage are given by

$$q_{f2} = q_{p1} + q_{o3} \quad (22)$$



(a) Two-stage without recycle



(b) Two-stage with recycle

Fig. 3. Two-stage separation module with and without recycle.

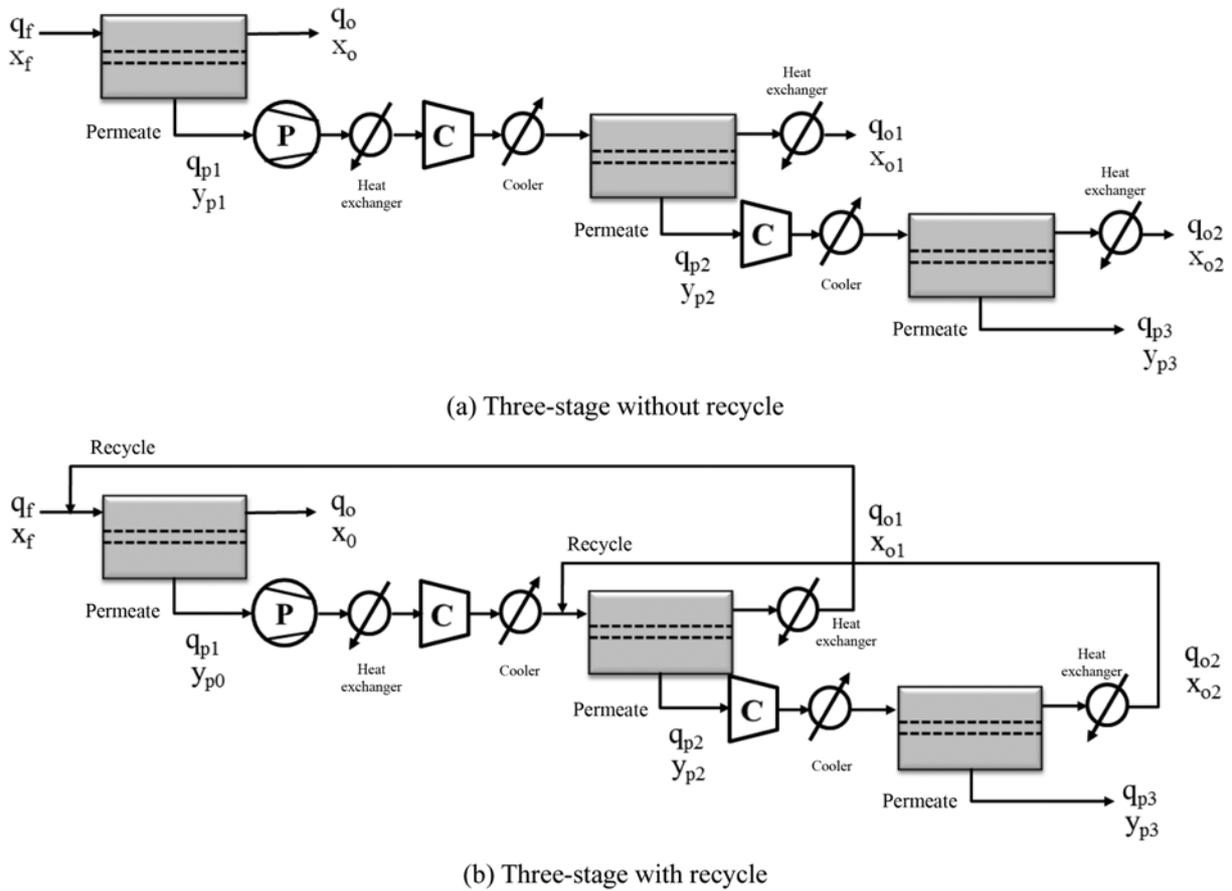


Fig. 4. Three-stage separation module with and without recycle.

$$x_{f2} = \frac{q_{p1}y_{p1} + q_{o3}x_{o3}}{q_f + q_{o3}} \quad (23)$$

Total feed flow rate and the mole fraction of CO₂ which are recycled and fed into the inlet of the 3rd-stage are given by Eq. (20) and (21).

ECONOMIC ASSESSMENT

Due to the relatively low concentration of CO₂ in the stack gas from power plants driven by fossil fuels (4(LNG)-16 (coal) vol%), the CO₂ recovery process requires high cost. Among various CO₂ recovery processes, the membrane separation scheme is widely accepted as one of the most cost-effective process. Table 1 shows the characteristic properties of the membrane used in the present study.

Table 1. Parameters used in numerical simulations

| Parameter | Value | Unit |
|---------------|--------------------|----------------------------|
| x_f | 0.15 | - |
| q_f | 0.0129 | mol/sec |
| α^* | 30 | - |
| T | 298.15 | K |
| R | 8.314 | Pa·m ³ /mol·K |
| $P'_{CO_2/t}$ | 2×10^{-7} | mol/m ² ·sec·Pa |
| p_l | 1 | bar |
| p_h | 5 | bar |

The separation model to be used in the economic assessment is based on the existing plant models (Li et al. [11]; Stropnik et al. [12]) which are modified to include 13 equations. Annual recovery and separation cost for 1 ton of CO₂ are estimated by using the model. Table 2 lists related unit costs and process parameters. The relation representing separation cost of the membrane module employs constant values and the membrane cost used in the present study is 44.33 dollar/m² [11]. The cost of the membrane module with feed rate of

Table 2. Values of parameters used in the economic assessment computation [11]

| Parameter | Value | Unit |
|-----------|--------|-----------------------|
| a | 0.064 | - |
| a_m | 0.225 | - |
| c | 1 | - |
| C_{he} | 4.48 | million dollar |
| F_h | 1.8 | - |
| K_m | 44.33 | dollar/m ² |
| K_{mf} | 0.3225 | million dollar |
| K_{c1} | 3.84 | million dollar |
| K_{c2} | 30.84 | million dollar |
| K_{vp} | | million dollar |
| K_{el} | 0.06 | cent/kWh |
| γ | 1.4 | - |

0.0129 mol/s is estimated and the cost of the membrane frame including casing, valves and tubing is calculated by using Eq. (25). The depreciation period for the compressor, the vacuum pump and membrane modules is assumed to be 25 years, while the lifetime of the membrane is assumed to be 5 years. It is assumed that the maintenance costs for the compressor, the vacuum pump and heat exchangers are 3.6% of the investment cost and those for the membrane and the membrane frame are 1% of the investment cost. Eqs. (30) and (31) are based on some ideal assumptions concerning energy consumption by compressors and vacuum pumps [13]. The efficiency of the vacuum pump, the motor and the compressor is assumed to be 50%, 90% and 85%, respectively. The market price of electricity is set to 70 won/kWh. Evaluation of economics is performed for single-stage and multi-stage modules with and without recycles. The permeation pressure for the single-stage module is set to 0.2 bar and those for 2 and 3-stage modules are set to 1 bar.

1. Estimated Investment

Basic costs for the membrane, the membrane frame, the compressor, the vacuum pump and the heat exchanger are estimated by Eqs. (24)-(28). Costs for the membrane are given by

$$I_m = A_m K_m \tag{24}$$

$$I_{mf} = (A_m / 2000)^{0.7} K_{mf} \tag{25}$$

where the membrane area A_m is the sum of individual membrane areas for multi-stage module, K_m is the membrane cost per square meter and K_{mf} is the basic cost for the membrane frame. Costs of other facilities can be obtained from the following relations.

$$I_c = K_{c1} \cdot F_h + K_{c2} \cdot F_h \tag{26}$$

$$I_{vp} = K_{vp} \cdot F_h \tag{27}$$

$$I_{he} = C_{he} \tag{28}$$

where K_{c1} and K_{c2} are cost parameters for the compressor and can be defined as the capture cost and CO₂ compression cost, respectively. K_{vp} is the basic cost for the vacuum pump, which is assumed to be four times of that for the compressor. C_{he} is the basic cost of the heat exchanger and the cooler. The dimensionless parameter F_h is the design cost factor of individual equipment.

2. Energy Consumption in Compression Equipment

The permeate pressure of the first stage is reduced up to 0.2 bar to achieve higher purity and recovery rate. For this purpose a vacuum pump is employed to suck in permeate gases and to exhaust them at atmospheric pressure. In the case of multi-stage module, the feed to the 2nd-stage is pressurized up to 6 bar by using a compressor. A heat exchanger and a cooler are added to prevent deformation of the membrane due to temperature increase caused by pressurization. Representations of energy consumption at individual equipment are given by Eqs. (30) and (31). Total energy consumption is given by Eq. (29).

$$P_{tot} = \sum P_c + \sum P_{vp} \tag{29}$$

$$P_c = \mu (\gamma / (\gamma - 1)) RT [(P_2 / P_1)^{\frac{(\gamma-1)}{\gamma}} - 1] / [1 - (c(P_2 / P_2)^\gamma - 1)] \tag{30}$$

$$P_{vp} = \mu (\gamma / (\gamma - 1)) RT [(P_2 / P_1)^{\frac{(\gamma-1)}{\gamma}} - 1] \tag{31}$$

where μ denotes molar flow rate to the vacuum pump and γ is defined as $\gamma = (C_p / C_v)$. For ideal system, the value of γ is taken as 1.4. R is the gas constant ($R = 8.314 \text{ J/mol}\cdot\text{K}$) and P_1 and P_2 are inlet and throughout pressure at the corresponding equipment, respectively. c is a factor taking a value between 0.05 and 1. In this work, change in volume is ignored and c is set to 1.

3. Annual Costs

Based on preceding relations, we can compute annual costs for investment, maintenance, energy and total membrane separation. In the computation, the lifetime of the membrane area is set to 5 years and that of other facilities is set to 25 years. Again, the maintenance cost for the compressor, the vacuum pump and heat exchangers is assumed to be 3.6% of the investment cost, while that for the membrane and the membrane frame is assumed to be 1% of the investment cost.

$$C_{cap} = (\sum I_c + \sum I_{vp} + \sum I_{he} + I_m) a + I_m a_m \tag{32}$$

$$C_{O\&M} = 0.036 (\sum I_c + \sum I_{vp} + \sum I_{he}) + 0.01 (I_m \cdot I_{mf}) \tag{33}$$

$$C_{en} = t_{op} \cdot P_{tot} \cdot C_{O\&M} \tag{34}$$

$$C_{tot} = C_{cap} + C_{en} + C_{O\&M} \tag{35}$$

where a and a_m denote depreciation rates. The annual cost (won/ton/year) required in the separation of CO₂ is given by

$$\dot{C}_{CO_2} = C_{tot} / M_{CO_2, \text{ann, separated}} \tag{36}$$

where M represents the annual amount of separated CO₂ (ton/year).

RESULTS AND DISCUSSION

1. Characteristics of CO₂ Separation Modules

Evaluation of cost requires the knowledge on the behavior of CO₂ separation modules. We can investigate characteristics of each separation module by numerical simulations based on modeling equations. Basic data used in the numerical simulations are listed in Table 1.

1-1. Single-stage Module

In general, a large active area is required to achieve the desired

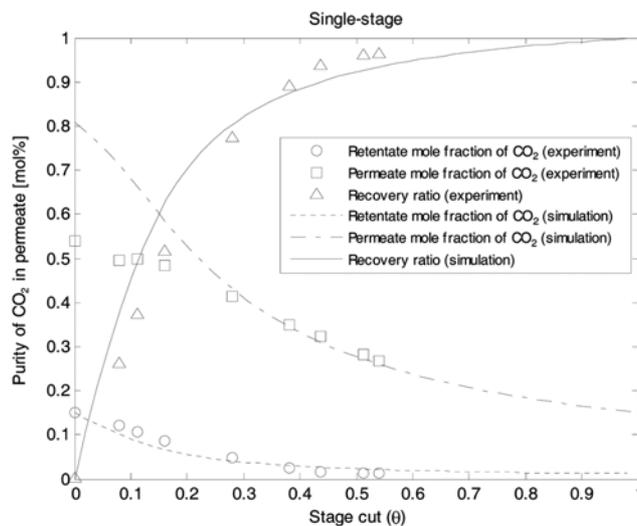


Fig. 5. Results of numerical simulations compared with those of experiments.

recovery rate from feed gas mixtures by using polymer membranes. It is well known that hollow fiber separation membranes provide larger active area per unit volume compared to other separation membranes. Thus we can expect improved economics by the use of hollow fiber membranes to separate CO₂ from gas mixtures. Fig. 5 shows results of numerical simulations compared with experimental results concerning changes in recovery and purity according to stage-cut (0.1-0.9) for the single-stage module. Here purity is denoted by the concentration of CO₂ in the permeate stream. The experimental results shown in Fig. 5 are obtained at the atmospheric condition without recycle. The membrane used in the experiment was polybenzoxazole (PBO) based hollow fiber membrane which gives rise to high free volume elements with high gas permeability. Details on the experiment can be found elsewhere [14]. As can be seen, purity does not grow over 90% in the single-stage module. Results of simulations exhibit excellent tracking performance to experimental results, which demonstrates the effectiveness of the proposed model in the assessment of economics. Higher purity and recovery ratio can be achieved by lowering the permeate pressure to 0.2 bar because purity and recovery ratio increase as the difference between the feed pressure and the permeate pressure increase.

Effects of recycles for the single-stage module are shown in Fig. 6 with the permeate pressure being set to 0.2 atm. For the single-stage module without recycle, we can see that the purity decreases from 68.10 mol% to 16.52 mol% and the recovery ratio increases from 45.40% to 99.14% when the stage-cut increases from 0.1 to 0.9. The most favorable operating points can be regarded as those providing higher purity and recovery ratio. In this sense, we can take the intersection of two curves (curves representing the purity and the recovery ratio in Fig. 6) as the optimal operating point. At this point, the permeate pressure is 0.2 bar, the feed pressure is 5 bar and the stage-cut is 0.12. For the selectivity of 30, the purity and the recycle ratio are 64.96 mol% and 51.97%, respectively, and the membrane area is 360 cm². For the single-stage module with recycle, the purity decreases from 60.76 mol% to 15.24 mol% and the recovery ratio increases from 49.78% to 99.74% when the stage-cut increases from 0.1 to 0.9. As in the case with recycle, the stage-

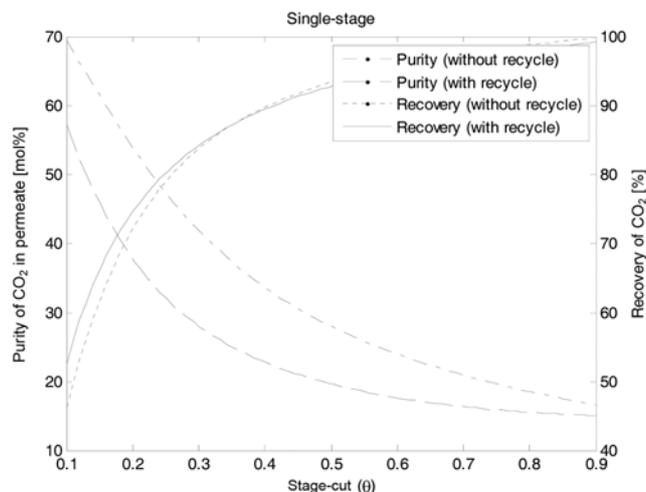


Fig. 6. Results of numerical simulations for the single-stage module.

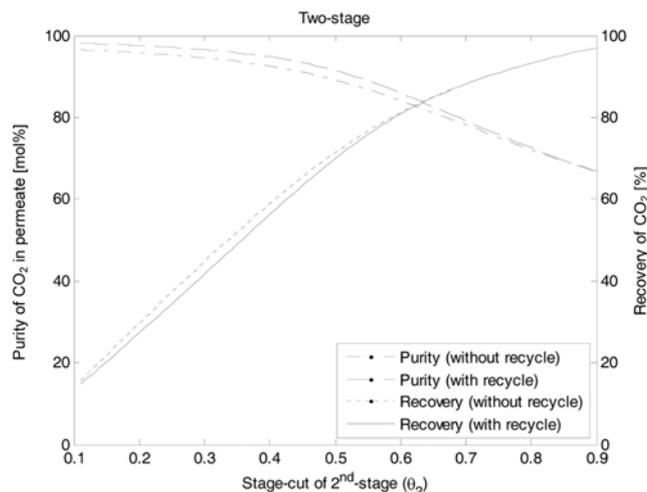


Fig. 7. Results of numerical simulations for the two-stage module: $\theta_1=0.14$.

cut is 0.12 at the intersection. At this value of the stage-cut, the purity and the recycle ratio are 55.84 mol% and 56.73%, respectively, and the membrane area is 909 cm². We can see that the purity increases, while the recovery ratio decreases at the fixed stage-cut of 0.12, which demonstrates the improvement in the recovery by the recycle.

1-2. Two-stage Module

Fig. 7 shows results of computations for the two-stage module where the stage-cut of the 1st-stage is fixed as 0.14, while that of the 2nd-stage varies from 0.1 to 0.9. Here the purity is represented by the concentration of CO₂ in the permeate stream from the 2nd-stage. For the two-stage module without recycle, the purity decreases from 96.55 mol% to 66.62 mol% and the recovery ratio increases from 15.63% to 97.04% when the stage-cut of the 2nd-stage increases from 0.1 to 0.9. When the stage-cut of the 2nd-stage is 0.66, the purity and the recycle ratio are 80.39 mol% and 85.86%, respectively, and the membrane areas of the 1st-stage and the 2nd-stage are 454 cm² and 199 cm², respectively. For the two-stage module with recycle, the purity decreases from 98.15 mol% to 66.77 mol% and the recovery ratio increases from 13.35% to 97.03% when the stage-cut of the 2nd-stage increases from 0.1 to 0.9. When the stage-cut of the 2nd-stage is 0.66, the purity and the recycle ratio are 81.89 mol% and 85.55%, respectively, and the membrane areas of the 1st-stage and the 2nd-stage are 460 cm² and 195 cm², respectively. For the two-stage module, we could achieve more than 80% both in the purity and the recovery ratio and the required membrane area of the 2nd-stage is reduced.

1-3. Three-stage Module

Results of computations for the three-stage module are shown in Fig. 8. Here the stage-cut of the 1st-stage is fixed as 0.23, that of the 2nd-stage is set to 0.66 while that of the 3rd-stage varies from 0.1 to 0.9. In this case, the purity is given by the concentration of CO₂ in the permeate stream from the 3rd-stage. For the three-stage module without recycle, the purity decreases from 94.70 mol% to 64.99 mol% and the recovery ratio increases from 15.66% to 96.73% when the stage-cut of the 3rd-stage increases from 0.1 to 0.9. When the stage-cut of the 3rd-stage is 0.6, the purity and the recycle ratio are 80.76 mol% and 80.13%, respectively, and the membrane areas of

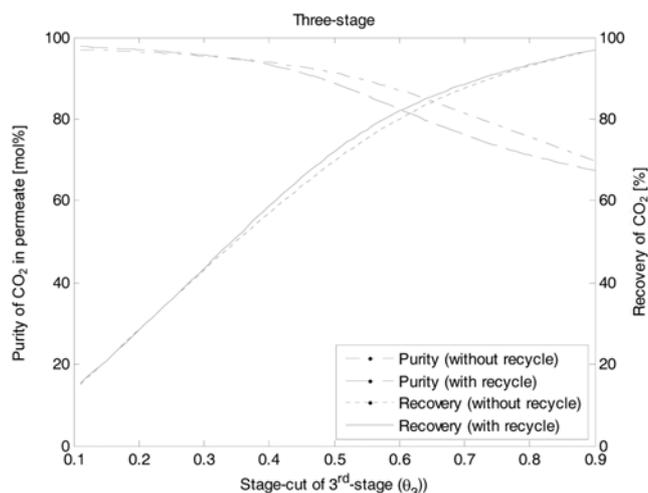


Fig. 8. Results of numerical simulations for the three-stage module: $\theta_1=0.23$, $\theta_2=0.66$.

the 1st-stage, the 2nd-stage and the 3rd-stage are 696 cm², 417 cm² and 137 cm², respectively. For the three-stage module with recycle, the purity decreases from 97.83 mol% to 67.45 mol% and the recovery ratio increases from 13.85% to 97.02% when the stage-cut of the 2nd-stage increases from 0.1 to 0.9. At the optimal condition, the purity and the recycle ratio are 82.40 mol% and 81.92%, respectively, and the membrane areas of the 1st-stage, the 2nd-stage and the 3rd-stage are 15,400 cm², 773 cm² and 227 cm², respectively.

2. Economics Assessment of Separation Modules

The general approach in the assessment of economics is to figure out the most economic separation module while maintaining maximal purity and recovery ratio for modules with and without recycle stream. In the cost calculation, the unit membrane cost was set to 44.33 dollar/m² and the unit electricity cost was set to 70 won/kWh.

2-1. Single-stage Module

For the single-stage separation module (Fig. 2), separation cost was calculated without considering operating cost of additional equipment. Note that commercial implementation can hardly be considered for the single-stage module because of relatively low purity and recovery (<80%). However, for the purpose of comparison, separation costs were computed for the module with and without recycle stream. The membrane area required in the single-stage without recycle is 360 cm² while that in the single-stage with recycle is

909 cm², which is 2.5 times larger than the previous one. The cost of the membrane and the frame for the single-stage module without recycle amounts to 1.5959 dollar/yr and 153.8936 dollar/a, respectively. For the case with recycle, the corresponding cost is 4.0296 dollar/yr and 294.36089 dollar/yr, respectively. The capital cost and the maintenance cost for the single-stage module without recycle was found to be 10.2083 dollar/yr and 1.5549 dollar/yr, respectively, and the total separation cost was 11.7632 dollar/yr. For the single-stage module with recycle, the capital cost and the maintenance cost was 19.7424 dollar/yr and 2.9834 dollar/a, respectively, and the total separation cost was 22.7258 dollar/yr. The annual separation cost for 1 kg of CO₂ separated was 0.0093 dollar without recycle and 0.0053 dollar with recycle. We can see that, for the single-stage module with recycle, the total cost increases due to increased flow rates while the annual CO₂ separation cost decreases. As expected, separation with recycle provides lower separation cost while maintaining the desired purity level.

2-2. Two-stage Module

In the two-stage separation module (Fig. 3), a vacuum pump, a compressor, a couple of heat exchangers and a cooler are added to maintain the pressure level of the feed stream to the 2nd-stage. Energy costs required in this equipment are calculated by Eqs. (30) and (31). The exit pressure from the vacuum pump and from the compressor was set to 1.095 bar and 5 bar, respectively. The heat exchanger and the cooler are required to maintain the temperature level of the exit stream. For the two-stage module, the area of the 1st-stage with and without recycle stream is 460 cm² and 454 cm², respectively, and the area of the 2nd-stage with and without recycle stream is 195 cm² and 199 cm², respectively. The total separation cost with recycle stream was found to be 10.3512 million dollars, which was the same both without recycle stream and with recycle stream. The annual separation cost for 1 kg of CO₂ separated was 8.2464 × 10³ dollars without recycle and 8.0953 × 10³ dollars with recycle. As in the case of the single-stage module with recycle, the total cost increases due to increased flow rates and increased membrane area while the annual CO₂ separation cost decreases. Both the recovery ratio and the purity were greater than 80%, which couldn't be achieved in the single-stage module. This fact demonstrates that the multi-stage module is more advantageous than the single-stage module. Results of cost computations for the two-stage module with and without recycle are summarized in Table 4.

2-3. Three-stage Module

In the three-stage separation module (Fig. 4), a vacuum pump, compressors, heat exchangers and coolers are added to maintain

Table 3. Annual capture costs for single-stage with and without recycle

| Type | Cost | Value | Unit |
|------------------------------|--|---------|--|
| Single-stage without recycle | Specific CO ₂ separation cost | 0.0093 | dollar/kg _{separatedCO₂} /yr |
| | Total cost | 11.7632 | dollar/yr |
| | Capital cost | 10.2083 | dollar/yr |
| | O&M cost | 1.5549 | dollar/yr |
| Single-stage with recycle | Specific CO ₂ separation cost | 0.0053 | dollar/kg _{separatedCO₂} /yr |
| | Total cost | 22.7258 | dollar/yr |
| | Capital cost | 19.7424 | dollar/yr |
| | O&M cost | 2.9834 | dollar/yr |

Table 4. Annual capture costs for two-stage with and without recycle

| Type | Cost | Value | Unit |
|---------------------------|--|---------|--|
| Two-stage without recycle | Specific CO ₂ separation cost | 8.2464 | dollar/kg _{separatedCO₂} /yr |
| | Total cost | 10.3512 | million dollar/yr |
| | Capital cost | 6.6248 | million dollar/yr |
| | O&M cost | 3.7264 | million dollar/yr |
| | Energy cost | 0.4452 | dollar/yr |
| Two-stage with recycle | Specific CO ₂ separation cost | 8.0953 | dollar/kg _{separatedCO₂} /yr |
| | Total cost | 10.3512 | million dollar/yr |
| | Capital cost | 6.6248 | million dollar/yr |
| | O&M cost | 3.7264 | million dollar/yr |
| | Energy cost | 0.4699 | dollar/ yr |

Table 5. Annual capture costs for three-stage with and without recycle

| Type | Cost | Value | Unit |
|-----------------------------|--|----------------------|--|
| Three-stage without recycle | Specific CO ₂ separation cost | 1.1329×10^3 | dollar/kg _{separatedCO₂} /yr |
| | Total cost | 14.2866 | million dollar/yr |
| | Capital cost | 7.9903 | million dollar/yr |
| | O&M cost | 6.2963 | million dollar/yr |
| | Energy cost | 27.3773 | dollar/yr |
| Three-stage with recycle | Specific CO ₂ separation cost | 8.8832×10^3 | dollar/kg _{separatedCO₂} /yr |
| | Total cost | 14.2866 | million dollar/yr |
| | Capital cost | 7.9903 | million dollar/yr |
| | O&M cost | 6.2963 | million dollar/yr |
| | Energy cost | 173.5205 | dollar/yr |

the pressure level of the feed stream between the 2nd-stage and the 3rd-stage. Again, the increased energy costs required in this equipment are calculated by Eqs. (30) and (31). The total separation cost without recycle stream was found to be 14.2866 million dollars, which was the same without a recycle stream. The annual separation cost for 1 kg of CO₂ separated was 1.1329×10^3 dollars with-

out recycle and 8.8832×10^3 dollars with recycle. We can see that the CO₂ separation cost increases compared to the one- and two-stage separation modules even though we can achieve higher purity and recovery ratio than other modules. This fact suggests that the two-stage separation module is a more profitable candidate for commercial implementation. Table 5 shows results of cost computations for the three-stage module.

Figs. 9 and 10 display comparison of membrane separation costs between the single- and multi-stage separation modules. Fig. 9 shows changes in separation costs for the single-stage module without any

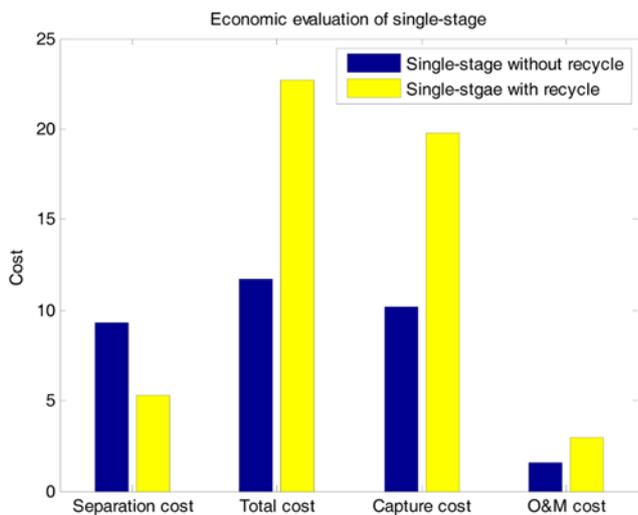


Fig. 9. Economic evaluation of the single-stage module: Separation cost [dollar/g_{separatedCO₂}/yr], Total cost, Capture cost, O&M cost [dollar].

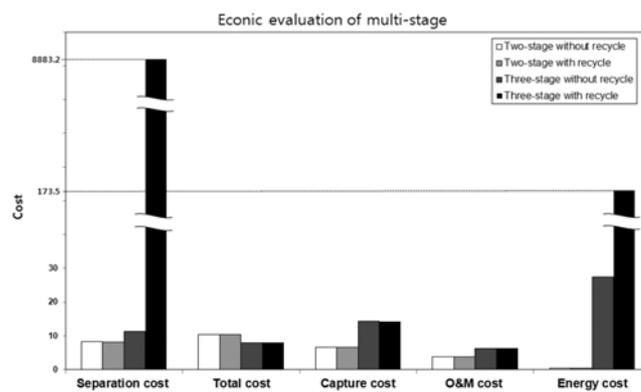


Fig. 10. Economic evaluation of the two- and three-stage modules: Separation cost [dollar/g_{separatedCO₂}/yr], Total cost, Capture cost, O&M cost [million dollar], Energy cost [dollar].

additional equipment. Although the single-stage module with recycle stream exhibits higher total separation cost, the capital cost as well as the maintenance cost, it gives lower annual CO₂ separation cost, which demonstrates the effectiveness of the use of a recycle stream. Fig. 10 shows a comparison of membrane separation costs between the two- and three-stage separation modules with and without recycle streams. As can be seen, the two-stage module with recycle exhibits the lowest annual CO₂ separation cost.

CONCLUSION

We performed an economic assessment for the hollow fiber CO₂ separation processes based on mathematical models. Numerical simulations were carried out to examine the effects of different configurations such as single-stage, two-stage and three-stage CO₂ separation processes. In particular, the hollow fiber membrane processes for CO₂ separation were analyzed to provide pressurized feed streams with vacuum pumps, heat exchangers, coolers and compressors. Operating costs were evaluated and compared numerically for the processes with and without recycle streams to examine the feasibility for commercial implementation while maintaining the purity and recovery ratio as high as possible. As the number of the stage increases, the purity and the recovery ratio increase and the CO₂ separation cost also increases due to the increased membrane area. It was found that the two-stage module with recycle exhibits the lowest annual CO₂ separation cost.

ACKNOWLEDGEMENTS

This work was supported by a grant (CDRS II-03-1) from the Carbon Dioxide Reduction & Sequestration Research Center, one of the 21st century frontier programs funded by the Ministry of Science and Technology of the Korean Government.

NOMENCLATURE

| | |
|------------------|--|
| a | : depreciation factor (25 year) |
| a_m | : depreciation factor (5 year), real interest rate 5% |
| A_m | : membrane area [cm ²] |
| c | : clearance factor |
| C_{cap} | : capital cost [dollar/a] |
| \dot{C}_{CO_2} | : specific CO ₂ separation cost [dollar/kg _{separatedCO₂} /a] |
| C_{en} | : energy cost per year [dollar/a] |
| C_{he} | : heat exchangers and cooling facility cost [million dollar] |
| $C_{O\&M}$ | : O&M cost [dollar/a] |
| C_{tot} | : total capture cost [dollar/a] |
| F_h | : cost factor for housing, installation, etc. |
| I_c | : investments for compressor [million dollar] |
| I_{he} | : investment for heat exchanger [million dollar] |
| I_m | : investment for membrane [dollar] |
| I_{mf} | : investment for membrane frame [million dollar] |
| I_{vp} | : investment for vacuum pump [million dollar] |
| K_{c1} | : compressor cost (for capture) [million dollar] |
| K_{c2} | : compressor cost (for CO ₂ compression) [million dollar] |
| K_{el} | : electricity cost (hard coal) [dollar/kWh] |
| K_m | : membrane unit cost [dollar/m ²] |
| K_{mf} | : permanent membrane frame cost [million dollar] |

| | |
|----------------------------|---|
| K_{vp} | : vacuum pump cost [million dollar] |
| $M_{CO_2, ann, separated}$ | : captured CO ₂ per year [ton] |
| P_1 | : inside pressure of compression equipment [bar] |
| P_2 | : outside pressure of compression equipment [bar] |
| P'_A/t | : specific permeability of species CO ₂ |
| P'_B/t | : specific permeability of species N ₂ |
| P_c | : energy used for compressors [kW] |
| p_h | : higher pressure [bar] |
| p_l | : lower pressure [bar] |
| p_{tot} | : total energy consumption [KW] |
| P_{vp} | : energy used for vacuum pump [kW] |
| q_A | : flow rate of species CO ₂ in permeate [mol/s] |
| q_B | : flow rate of species N ₂ in permeate [mol/s] |
| q_f | : flow rate of the feed side [mol/s] |
| q_o | : flow rate of the reject side [mol/s] |
| q_p | : flow rate of the permeate side [mol/s] |
| R | : gas constants [J/mol·K] |
| T | : absolute temperature [K] |
| t_{op} | : operation time [h] |
| x_f | : mole fraction of CO ₂ at the feed side [-] |
| x_o | : mole fraction of CO ₂ at the reject side [-] |
| y_p | : mole fraction of CO ₂ at the permeate side [-] |
| α^* | : ideal selectivity defined in Eq. (6) [-] |
| γ | : C_p/C_v [-] |
| θ | : stage-cut defined in Eq. (2) [-] |
| μ | : molar flow rate into the compression equipment [mol/s] |

REFERENCES

1. J. P. Ciferno, T. E. Fout, A. P. Jones and J. T. Murphy, *Chem. Eng. Progress.*, **33** (2009).
2. H. Herzog, *Environ. Sci. Technol.*, **35**(7), 148 (2001).
3. C. M. White, *Journal of Air Waste Management Association*, **53**, 645 (2003).
4. E. Favre, *J. Membr. Sci.*, **294**, 50 (2007).
5. S. H. Choi, J. H. Kim, B. S. Kim, S. B. Lee and Y. T. Lee, *Theories and Applications of Chem. Eng.*, **14**(1), 298 (2008).
6. H. B. Park, C. H. Jung, Y. M. Lee, A. J. Hill, S. J. Pas, S. T. Mudie, E. V. Wagner, B. D. Freeman and D. J. Cookson, *Science*, **318**, 254 (2007).
7. S. H. Choi, J. H. Kim, B. S. Kim and S. B. Lee, *Membr. J.*, **15**(4), 310 (2005).
8. C. A. Hendriks, A. F. B. Wildenborg, K. Blok, F. Floris and J. D. van Wees, In paper presented at the 5th International Conference on Greenhouse Gas Control Technologies (GHGT-5) (2000).
9. J. P. van der Sluijs, C. A. Hendriks and K. Blok., *Energy Convers. Manage.*, **33**, 429 (1992).
10. T. H. Minh, Guy W. Allinson and Dianne E. Wiley, *Ind. Eng. Chem. Res.*, **47**(5), 1562 (2008).
11. Z. Li, R. Ernst, B. Ludger and S. Detlef, *J. Membr. Sci.*, **359**, 160 (2010).
12. Stropnik, W. Yave and K. V. Peinemann, *Sep. Purif. Technol.*, **62**, 110 (2008).
13. T. L. Biegler, E. I. Grossmann and W. A. Westerberg, *Systematic Methods of Chemical Process Design*, PrenticeHallPTR (1997).
14. H. G. Jin, S. H. Han, Y. M. Lee and Y. K. Yeo, *Korean J. Chem. Eng.*, **28**(1), 6 (2011).