

Modeling and economic analysis of CO₂ separation process with hollow fiber membrane modules

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(Received 31 August 2006 • accepted 6 November 2006)

Abstract—The main purpose of the study was to develop a model using ASPEN and Excel simulation method to establish optimum CO₂ separation process utilizing hollow fiber membrane modules to treat exhaust gas from LNG combustion. During the simulation, optimum conditions of each CO₂ separation scenario were determined while operating parameters of CO₂ separation process were varied. The characteristics of hollow fibers membrane were assigned as 60 GPU of permeability and 25 of selectivity for the simulation. The simulation results illustrated that 4 stage connection of membrane module is required in order to achieve over 99% of CO₂ purity and 90% of recovery rate. The resulted optimum design and operation parameters throughout the simulation were also correlated with the experimental data from the actual CO₂ separation facility which has a capacity of 1,000 Nm³/day located in the Korea Research Institute of Chemical Technology. Throughout the simulation, the operating parameters of minimum energy consumption were evaluated. Economic analysis of pilot scale of CO₂ separation plant was done with the comparison of energy cost of CO₂ recovery and equipment cost of the plant based on the simulation model.

Key words: CO₂ Separation, Membrane, Simulation, Economic Analysis

INTRODUCTION

In accordance with a series of international agreements concerning global climate change, Korea must prepare for the duty of reducing CO₂ emission. Most of the CO₂ separation processes require a large amount of parasitic energy consumption so that a method of optimum arrangement of individual components of a CO₂ separation system is required. Furthermore, energy in the separation process can be minimized through finding the operating parameters of CO₂ separation process with minimal energy consumption. One of the methods for finding the optimum option is system simulation to develop low energy-consumption CO₂ separation process. In the present investigation, membrane separation process with hollow fiber is simulated to find optimum design and operation parameters for CO₂ separation process. The membrane separation of CO₂ needs relatively low process requirements compared to absorption or distillation type separation. The membrane method is operated easily and inexpensively because of its physical separation mechanism and recognized as a suitable CO₂ separation process for application in a small-to-medium scale facility [1].

SIMULATION

1. Theory

To determine the separation behavior of CO₂ from the exhaust gas, governing equations of separation in hollow fiber membranes are utilized during the simulation, which is shown in Eq. (1). The

area of membrane module is set with the design data of actual pilot plant as in Eq. (2) [2]. With these assumptions, the performance of CO₂ separation is determined with simulation for the various cases of different CO₂ concentration of every stage and recovery ratio. The amount of required membrane module, which is proportional to the cost of the facility, is then determined by Eq. (3).

$$(a-1)y_i^2 + \left(1-a - \frac{1}{R} - \frac{x(a-1)}{R}\right)y_i + \frac{ax}{R} = 0 \quad (1)$$

$$A = \frac{(Vy)_{out}}{(J_A)_{out}} = \frac{(Vy)_{out}}{Q_A(P_1x - P_2y)_{ave}} = \frac{(Vy)_{out}}{\text{Permeate flux} \times (x - Ry)_{ave}} \quad (2)$$

$$M = \frac{\text{effective membrane area (A)}}{\text{hollow fiber membrane area (a)}} \quad (3)$$

2. Modeling Methods

Several parameters of hollow fiber membrane are also incorporated into the simulation code for the simulation of the CO₂ membrane separation process, which is shown in Table 1 [3]. The hollow fiber membrane module is assumed such that each membrane

Table 1. Parameters of hollow fiber membrane used during simulation

Parameter	Parameter		
Inlet pressure, P ₁ [atm]	6	Num of HFM per module [Ea]	16,000 (25,000)
Outlet pressure, P ₂ [atm]	0.1	Feed rate [m ³ /day]	1,000 (100,000)
Selectivity [CO ₂ /N ₂]	25	Length of HF [cm]	50 (300)
Permeability [GPU]	60	Pressure ratio [P ₂ /P ₁]	0.0167

* (): commercial condition.

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‡This work was presented at the 6th Korea-China Workshop on Clean Energy Technology held at Busan, Korea, July 4-7, 2006.

Table 2. Compositions of LNG exhaust gas

Composition	wt%	vol%
N ₂	72.2	71.3
O ₂	2.0	1.7
H ₂ O	11.0	18.2
CO ₂	13.9	8.8
Total	100.0	100.0

module has 16,000 strings of hollow fiber. The input values of selectivity and permeability are chosen as 25 and 60 GPU, respectively. A flue gas flow rate of 41.7 m³/h and initial concentration of 12% CO₂ are used for the entire simulation, the values of which are the same as those of the pilot plant. Initial compositions of input gas utilized in the simulation are shown in Table 2. During the simulation, the target of the separation process is set at 90% of recovery rate and 99% of CO₂ purity [4].

A model of the entire process is developed step by step. First, a simulation for the CO₂ separation process composed with 1 stage of module is modeled with Excel. The algorithm for the membrane module is based on the actual design of a pilot CO₂ separation plant, which links together the relationships of Eqs. (1), (2) and (3). Furthermore, characteristics of hollow fiber membrane are integrated into the simulation code with the basic concept of the CO₂ recovery process. With the 1 stage membrane module code, CO₂ recovery rate, permeated CO₂ flow rate and the amount of required membrane modules are calculated. Simulation is then carried out with fixing a CO₂ concentration of retentate (X_r) as 1%, and the required membrane area, number of required modules, flow rate and CO₂ concentration of permeate gas are determined throughout the simulation [5].

After that, using the code of a 1 stage membrane module, 4 stage membrane modules are composed like a cascade with return flow for each stage. This process is designed for obtaining a higher CO₂ recovery rate than 1 stage membrane module and considered for recycling of retentate gas. Flow rates assigned in the simulation are equalized as in Eq. (4) for 3 stages with return and in Eq. (5) for 1 stage without return.

$$L(I, N) = L(I, N-1) + V(I, N-1) + V(I+1, N-1) \quad (4)$$

$$L(I, N) = L(I, N-1) \quad (5)$$

For the convenience of simulation, calculation is iterated up to 100 times and the recycled concentration of retentate gas is assumed to equal the concentration of the feed gas. With the above recirculation concept, simulation using the entire code is completed, and the simulation results are determined as required membrane area, number of module, permeated CO₂ concentration and the amount of permeated gas for each initial operation condition. The simulation process is initially processed without recycling of retentate gas, and then the process with recycling of retentate gas.

Sensitivity analysis is carried out with changing the inlet and outlet pressures of the CO₂ separation process. To evaluate the process behavior with changing pressure, the inlet pressure of each stage is changed up to 7 atm, while other conditions are fixed. For the variation of outlet pressure, only the pressure of 1 stage module is changed

Table 3. Assumptions made for the economic analysis

Assumption for economic analysis
1) Equipment costs were provided by vendor.
2) Volume efficiency of compressor: 58% (pilot scale), 85% (commercial scale)
3) Mechanical efficiency & power factor of motor: 87%, 85%
4) Operating lifetime of plant: 20 year, operating ratio of plant: 100%
5) Cost of electricity: 5.2 cent/kW (convert to dollar, \$1=980 won)
6) Neglect a working cost.
7) Discount rate: 7%

to 0.1, 0.5 and 1 atm. A sensitivity study with pressure is important, since the equipment, such as a compressor or pump, consumes a great deal of energy for separating CO₂ gas. As a result, pressure is the important parameter for operating a membrane separation process. Sensitivity analysis is performed for minimizing consumed energy in the process with changing pressure.

3. Economic Analysis of CO₂ Separation Process

Economic analysis for the CO₂ separation process is based on the optimum equipment combination from the simulation results. Both energy cost and capital cost for CO₂ separation processes are evaluated from the prices and specifications of major equipment in CO₂ recovery plants. The net present value (NPV) is calculated through the economic analysis for a CO₂ recovery plant. The lifetime of the plant is assigned as 20 years, and discount rate is 7%. Several restrictions are applied in the evaluation of economical efficiency of a CO₂ recovery plant. The assumptions used in the economic analysis are shown in Table 3.

The economic analysis begins with the case of a pilot plant for the first time, and then is expanded to the commercial case, which has the capacity of treating 100,000 Nm³/day of exhaust gas. The amount of recovered CO₂ is calculated with Eq. (6) and the energy cost for operating a CO₂ recovery plant is shown as in Eq. (7).

$$CR = V \times 8760(h) \times OR \times 0.00178 \quad (6)$$

$$EC = \frac{PW \times 8760(h) \times OR \times PC}{CR} \quad (7)$$

The CO₂ recovery cost of the CO₂ separation plant is also calculated as shown in Eq. (8) [6].

$$RC = \frac{\sum_{i=1}^n \frac{T_i + S_i}{(1+r)^i}}{\sum_{i=1}^n \frac{CR_i}{(1+r)^i}} \quad (8)$$

Total onsite cost (T_i) is defined as sum of the total equipment cost, installation cost, piping cost, instrument control cost and electrical equipment cost. Startup cost (S_i) was defined as the power consumption of the operating equipment. The operated equipment is mostly composed of a compressor, vacuum pump, chiller etc. The discount rate (r) is commonly assumed as 7%. The economic analysis for CO₂ recovery plant is performed through a calculation of NPV of capital cost for a plant divided by the NPV of recovered CO₂ cost with 7% discount rate for 20 years. The economic analy-

ses for a commercial CO₂ recovery plant were evaluated as sensitivity analysis by various operating parameters such as initial CO₂ concentration, selectivity and pressure. For the variation of initial CO₂ concentration, two cases were applied as 12% for LNG exhaust gas and 30% for the flue gas in a cement manufacturing plant, respectively. An economic analysis of a CO₂ separation plant was also carried out with changing selectivity from 25 to 100.

RESULTS

The simulation of a single stage module was carried out with a change of inlet pressure from 3 to 15 atm. The profiles of flow rate and CO₂ concentration of permeated gas are shown in Fig. 1, which illustrates that CO₂ concentration increased and flow rate of permeate gas decreased with the increase of inlet pressure as shown in Fig. 1. Since the actual separation process requires a large quantity of both CO₂ concentration and flow rate, the optimum inlet pressure of the membrane module is selected at the intersection of both curves, which is about 4-7 atm. The area of the membrane, i.e., amount of module, decreased with the increase of inlet pressure as shown in Fig. 2. While the concentration of retentate (X_r) was fixed at 1%, the simulation results of a single stage module were determined as

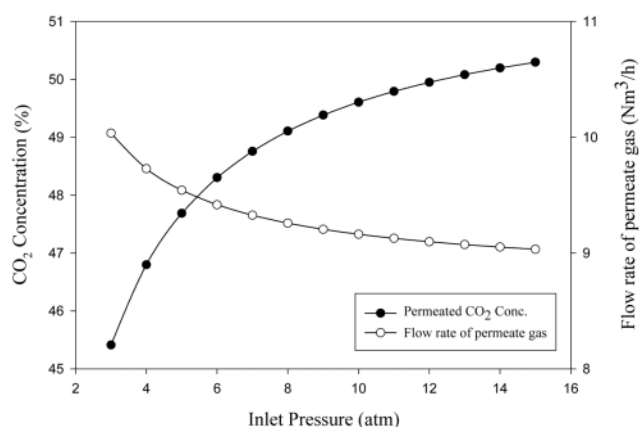


Fig. 1. Flow rate and CO₂ concentration profile with change of inlet pressure.

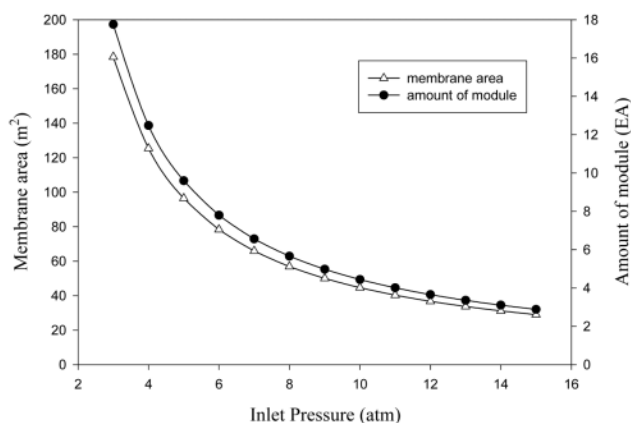
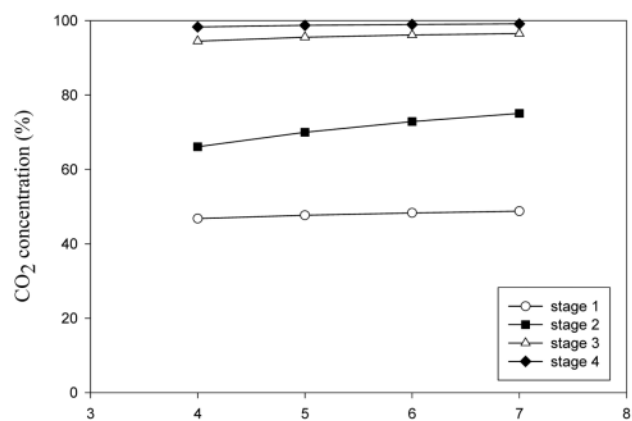


Fig. 2. Required membrane of area and number of modules with change of inlet pressure.

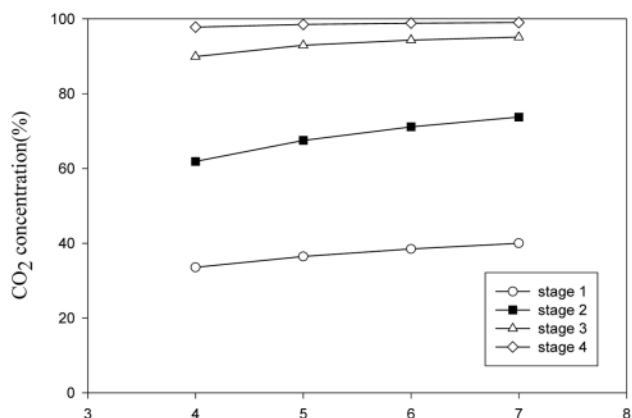
Table 4. Results of 1st stage membrane process at fixing retentate concentration at 1%

Parameter	Result
Required membrane area [m ²]	77.2
Amount of required modules [EA]	8 (≅ 7.68)
Flow rate of permeate gas [m ³ /h]	9.22
CO ₂ concentration of permeate [%]	48.9
*CO ₂ recovery [%]	90.3

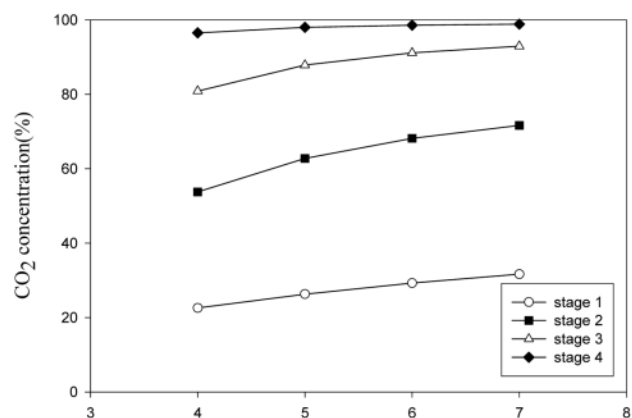
$$* \text{CO}_2 \text{ recovery (\%)} = \frac{Q_{\text{perm}} \times [\text{CO}_2]_{\text{perm}}}{Q_{\text{feed}} \times [\text{CO}_2]_{\text{feed}}}$$



(a) Inlet Pressure (atm), outlet pressure = 0.1 atm @ 1st stage



(b) Inlet Pressure(atm), outlet pressure = 0.5 atm @ 1st stage



(c) Inlet Pressure(atm), outlet pressure = 1 atm @ 1st stage

Fig. 3. CO₂ concentration in the 1st stage by change of inlet pressure & outlet pressure.

in Table 4. With 6 and 0.1 atm of inlet and outlet pressure, the concentration of permeated CO₂ gas of 48.9%, CO₂ recovery rate of 90.3%, permeated gas flow rate of 9.22 m³/h, amounts of required module of 8 (≈ 7.68), and required membrane area of 77.2 m² were determined.

To evaluate the energy requirement of the each stage for a 4 stage membrane module, the CO₂ concentration of permeate in every stage was calculated with changing the inlet pressure from 4 to 7 atm. Since the pressure of the 1st stage has a major effect on pressure distribution in the entire module, a sensitivity analysis was performed with changing outlet pressure of 1st stage to 0.1, 0.5, and 1 atm. While changing inlet pressure of 1 stage in the range of 4–7 atm, profiles of CO₂ concentration were obtained as shown in Fig. 3. The higher the pressure ratio between inlet and outlet, the higher the CO₂ concentration that is obtained, and a higher pressure ratio minimizes the effect of inlet pressure on CO₂ concentration of the permeated gas. When the outlet pressure is below 6 atm, CO₂ separation process should be operated with 0.1 atm in order to obtain 99% CO₂ concentration with 4 stage modules, as in Fig. 3.

For the economic analysis, the performance and recovery cost for the pilot CO₂ separation plant are obtained with change of inlet pressure and fixed value of outlet pressure in the 1st stage as 0.1 atm. The operating conditions of each case are illustrated in Table 5 and the results of economic analysis for 5 pilot plant cases are shown in Table 6. To analyze a commercial CO₂ recovery plant, PC-5 in Table 6 was selected as base plant, in which CO₂ concentration after 4 stages of module could reach 99% purity.

Results of the economic analysis for a commercial plant with 2 different initial CO₂ concentrations are shown in Fig. 4. The separation process with a CO₂ concentration of 30% required \$48/tCO₂ for the separation, while 12% needed \$98/tCO₂, i.e., the higher the CO₂ concentration of exhaust gas, the lower the recovery cost required. A sensitivity analysis with varying the selectivity of mem-

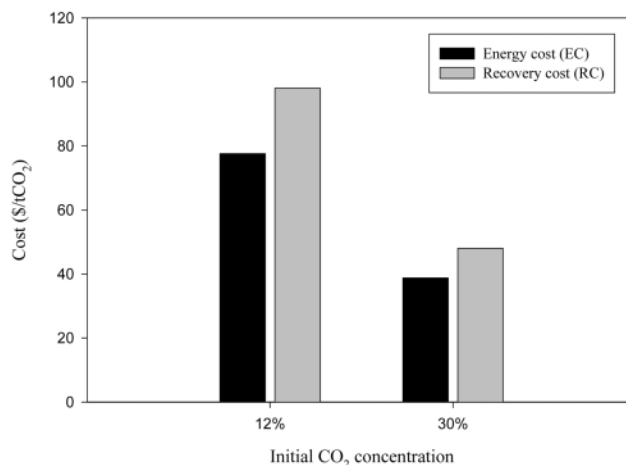


Fig. 4. Recovery cost of commercial CO₂ separation plant with initial CO₂ concentration.

brane did not represent much difference of recovery cost as in Fig. 5. The recovery cost almost appeared to constant while the selectivity was changed.

To minimize CO₂ recovery cost, the simulation continued in order to minimize the compression energy of the exhaust gas while maintaining the CO₂ recovery performance. Four different cases of commercial plant were analyzed with energy and recovery cost. During the analysis, CC-3 was set as the base case, then the pressure was varied as in Table 5. CC-1 was more economical than CC-3; however, CC-1 could obtain 98.7% CO₂ purity below that of CC-3. Furthermore, if the inlet pressure of a 1 stage membrane module was operated at 2 atm for reducing power consumption, the recovery cost of CC-4 could be reduced to \$87.8/tCO₂ and energy cost to \$56/tCO₂. The costs with different operating pressures are illus-

Table 5. Operating conditions of various scenarios in the economic analysis

Case		Operating condition (P ₁ /P ₂)	
Pilot plant	PC-1	1 stage: 2/0.1, 2 stage: 2/1, 3 stage: 2/1, 4 stage: 2/1	
	PC-2	1 stage: 3/0.1, 2 stage: 3/1, 3 stage: 3/1, 4 stage: 3/1	
	PC-3	1 stage: 4/0.1, 2 stage: 4/1, 3 stage: 4/1, 4 stage: 4/1	
	PC-4	1 stage: 5/0.1, 2 stage: 5/1, 3 stage: 5/1, 4 stage: 5/1	
	*PC-5	1 stage: 6/0.1, 2 stage: 6/1, 3 stage: 6/1, 4 stage: 6/1	
Commercial plant	CC-1	1 stage: 5/0.1, 2 stage: 5/1, 3 stage: 5/1, 4 stage: 5/1	
	CC-2	1 stage: 2/0.1, 2 stage: 5/1, 3 stage: 5/1, 4 stage: 5/1	
	*CC-3	1 stage: 6/0.1, 2 stage: 6/1, 3 stage: 6/1, 4 stage: 6/1	
	CC-4	1 stage: 2/0.1, 2 stage: 6/1, 3 stage: 6/1, 4 stage: 6/1	

*: base plant.

Table 6. Performance for pilot CO₂ recovery plant by operation conditions (recovery rate: 90%)

Case	Purity (%)	Module (EA)	CO ₂ recovery (ton/y)	EC (\$/tCO ₂)	RC (\$/tCO ₂)
PC-1	90.1	255	83.3	46.7	441.8
PC-2	97.3	47	77.1	55.1	308.2
PC-3	98.3	27	76.3	64.3	302
PC-4	98.8	19	76.2	72.4	303.1
PC-5	99	15	76	79.6	306.1

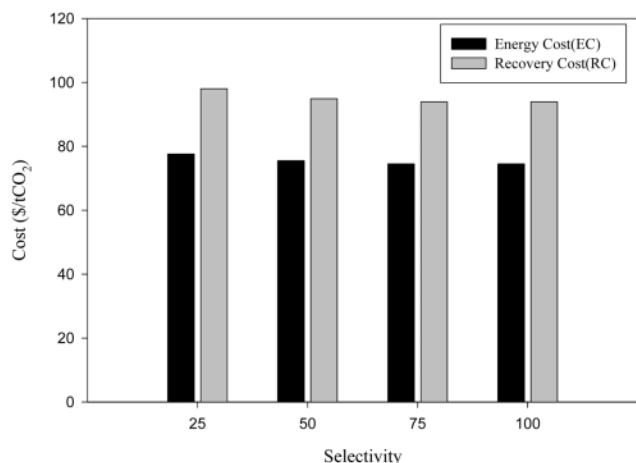


Fig. 5. Recovery cost of commercial CO₂ separation plant with selectivity of membrane.

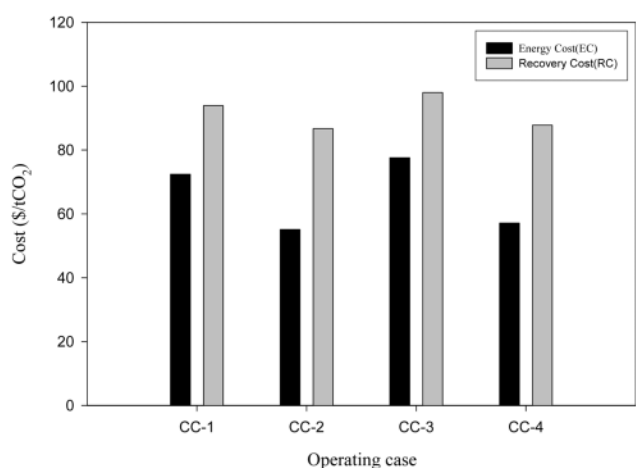


Fig. 6. Recovery cost of commercial CO₂ separation plant with operating pressure.

trated in Fig. 6. The recovery cost could be cut down with decreasing inlet pressure; however, CO₂ purity could not approach 99% of purity.

CONCLUSIONS

System simulation was carried for the separation CO₂ from flue gas with membrane by Excel and ASPEN code. To obtain the appropriate CO₂ concentration and flow rate of the permeate gas, the pressure of the membrane module was changed between 4 and 7 atm during simulation. The higher the ratio of inlet and outlet pressures of the membrane module, the higher the CO₂ concentration that could be obtained. To minimize energy consumption for a compressor and vacuum pump in the process with high pressure ratio, an economic analysis was carried out. It was found that reasonable operating conditions could be found through economic analysis for

a CO₂ recovery plant. For competing CO₂ separation with membrane among other separation methods, the membrane process should be operated at appropriate conditions which are considered both CO₂ recovery and energy consumption.

ACKNOWLEDGMENT

The research assistance received from Korea Energy Management Corporation and Ministry of Commerce, Industry and Energy (MOCIE) for this study is gratefully acknowledged.

NOMENCLATURE

- A : effective membrane area [m²]
- a : membrane area [m²]
- α : selectivity [CO₂/N₂]
- CR : annual amount of recovered CO₂ [ton]
- EC : energy cost [\$tCO₂]
- I : i-th module
- J_A : molar flux of component A [mol/m² h]
- L : flow rate of feed gas [m³/h]
- M : amount of membrane module [EA]
- N : iteration number
- n : operating time
- OR : operating ratio [%]
- PC : power cost [cent/kW]
- PW : power [kWh]
- P_1 : inlet pressure [atm]
- P_2 : outlet pressure [atm]
- Q_A : permeability of component A [L/m² h atm]
- R : pressure ratio (P_2/P_1)
- r : discount rate [%]
- RC : recovery cost [\$tCO₂]
- S_i : startup cost [\$]
- T_i : total onsite cost [\$]
- V : flow rate of permeate gas [m³/h]
- x : mole fraction of feed composition
- y_i : mole fraction of permeable species in permeate at i-th station

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