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## Energy Efficient Process for CO<sub>2</sub> Capture from Flue gas with Novel Fixed-site-carrier Membranes

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### Abstract

CO<sub>2</sub> capture from large stationary sources is considered as one of the most promising technologies to mitigate CO<sub>2</sub> emissions in atmosphere and reduce global warming. Amine absorption is the state-of-the-art technology for CO<sub>2</sub> capture, while high energy consumption and potential environmental impacts due to solvent emission and degradation needs to develop second generation solvents with high CO<sub>2</sub> loading capacity. Seeking environmentally friendly and energy efficient process with gas separation membranes could be an alternative for this application. In order to compare process feasibility of different techniques for CO<sub>2</sub> capture, the general criteria on energy consumption and cost estimation were provided in the current work. The proposed criteria provided an effective way in techno-economic feasibility analysis for CO<sub>2</sub> capture by easily adjusting relevant parameters. HYSYS simulation was conducted on a scenario of CO<sub>2</sub> capture from a gross output 819 MWe power plant with novel fixed-site-carrier membranes. A relatively low efficiency penalty of 10% and a competitive CO<sub>2</sub> capture cost of 47.3 \$/tonne CO<sub>2</sub> captured were found to be competitive to conventional amine absorption. Membrane systems have potentials for CO<sub>2</sub> capture if such performance can be achieved on pilot scale demonstration.

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### 1. Introduction

The control of greenhouse gas emissions is the most challenging environmental issues related to the global climate change, and strong interests have been focused on the reduction of CO<sub>2</sub> emissions from the large CO<sub>2</sub> point sources such as the fossil fuel power plants and other industries (e.g., cement, steel and iron production, natural gas and refinery plants) to mitigate global warming. Different techniques such as chemical absorption (e.g., monoethanolamine (MEA), methyldiethanolamine (MDEA)) and physical absorption (e.g., Selexol, Rectisol),

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physical adsorption (e.g., molecular sieves, metal organic frameworks), cryogenics and gas separation membranes could be used for CO<sub>2</sub> capture from flue gas in power plants and off-gas from the industries [1-4]. Conventional amine absorption is the state-of-the-art technology, but high energy consumption significantly increase electricity generation cost in power plant or add some extra cost in industry. Moreover, it causes environmental pollution due to solvent emission and degradation. Although a lot of effort has been put on development of second generation advanced amine solvents such as 2-amino-2- methyl-1-propanol (AMP) to improve CO<sub>2</sub> loading capacity [5], high energy consumption still hinders their commercial applications. Ionic liquids (ILs) have been considered as one of the most promising solvents for CO<sub>2</sub> capture owing to no or less contamination on gas stream and almost negligible solvent losses [6]. However, most of ILs are still in lab-scale production and not yet commercially available, which hinders their large-scale applications in short term. Recently, solid physical adsorbents such as metal-organic-frameworks (MOFs) [2] received a great attraction for CO<sub>2</sub> capture due to their high CO<sub>2</sub> adsorption capacity and relatively low energy consumption for regeneration, but low selectivity is still a challenge related to their commercial applications. Gas separation membrane technology as an energy efficient and environmentally friendly process has already been commercially used in selected gas purification processes such as air separation and natural gas sweetening [7], which is found to be an alternative and competitive technique for CO<sub>2</sub> capture from flue gas compared to conventional chemical absorption and physical adsorption. Great effort has been recently put on development of high performance membranes for this potential application, examples are:[3, 8-15]. Different types of membrane materials such as common polymers [16], microporous organic polymers (MOPs) [14, 17-19], fixed-site-carrier (FSC) membranes [12, 13, 20], mixed matrix membranes (MMMs) [21-25], carbon molecular sieve membranes (CMSMs) [26, 27] as well as inorganic membranes [28, 29] have been reported for CO<sub>2</sub> separation. However, in order to make membranes commercially applicable for CO<sub>2</sub> capture and compete with conventional amine absorption process, membrane systems should possess relatively low energy consumption and specific capture cost together with a good stability exposure to impurities of SO<sub>2</sub> and NO<sub>x</sub> which are usually involved in flue gas.

Techno-economic evaluation is usually conducted for process feasibility analysis, and some literature reported feasibility analysis on membrane systems for CO<sub>2</sub> capture by process simulation and cost estimation, examples are: [8, 9, 27, 30]. However, comparison of energy consumption and capture cost between amine absorption and membrane technology is quite difficult due to the difference in process and system. In amine absorption system, the main energy consumption is the required heat duty for solvent regeneration which can be directly taken from the steam generated in boiler, together with a small part of power demands for blowers and solvent pumps [31, 32]. However, energy consumption in membrane systems only comes from power demands of major driving equipment (e.g., compressors and pumps) without any heat duty. Thus, in order to compare energy consumption between membrane and amine absorption systems in the same baseline, specific equivalent power consumption was used in the current work. Moreover, the simple, unique criteria on cost estimation were also provided to evaluate economic feasibility of membrane systems. A case study on CO<sub>2</sub> capture from flue gas in a post-combustion coal fired power plant using fixed-site-carrier membranes was conducted in this work. The proposed criteria were employed for estimation of energy consumption and CO<sub>2</sub> capture cost, and to document process feasibility of membrane gas separation system.

## 2. Membrane materials for CO<sub>2</sub> capture

Novel fixed-site-carrier (FSC) membranes were developed by coating a thin polyvinylamine (PVAm) selective layer on top of polysulfone (PSf) ultrafiltration membrane for CO<sub>2</sub>/N<sub>2</sub> separation at Memfo group of NTNU. The prepared large flat-sheet FSC membranes (30cm×30cm) showed a high separation performance both CO<sub>2</sub> permeance (up to 5 m<sup>3</sup>(STP)/(m<sup>2</sup>.h.bar) and CO<sub>2</sub>/N<sub>2</sub> selectivity based on gas permeation testing at 2 bar and 35°C [13]. The FSC membranes can be operated in water vapor saturated gas process- this reduces the pre-treatment cost on dehydration of flue gas. Although water vapor permeation through the membranes has not been fully explored, preliminary results showed a similar water vapor permeance compared to CO<sub>2</sub> at low pressure. Moreover, Membrane performance was also tested at EDP's power plant in Sines (Portugal) to document the working of the membranes (a pilot-scale membrane module with a membrane area 2 m<sup>2</sup>) in NanoGLOWA project ([www.nanoglowa.com](http://www.nanoglowa.com)). This type of membranes presented a good stability over 6 months by exposed to a side stream of real flue gas (12 % CO<sub>2</sub> - 70% N<sub>2</sub> -13% H<sub>2</sub>O- 5% O<sub>2</sub>, 200 ppm SO<sub>2</sub>, 200 ppm NO<sub>x</sub>, 20mg/Nm<sup>3</sup> fly ashes). Recently, FSC membranes have

also been tested in Norcem cement factory at Brevik (Norway) where CO<sub>2</sub> feed concentration is ca. 17-20%. The initial testing will be finished by October 2014 to document the potentials of membrane system for CO<sub>2</sub> capture in cement factory, and compare to other technologies that was also tested there such as amine absorption (Aker Clean Carbon, Norway) and The Research Triangle Institute (RTI, US) solid adsorption [33]. The developed FSC membranes were chosen for process simulation to document techno-economic feasibility of CO<sub>2</sub> capture from flue gas.

### 3. Energy consumption estimation for CO<sub>2</sub> capture

In order to estimate energy consumption for CO<sub>2</sub> capture, a power plant with a constant input fuel (coal, natural gas or oil) is chosen as reference base. Thus, total CO<sub>2</sub> produced is fixed no matter whether CO<sub>2</sub> capture unit is installed or not. However, the whole plant thermal efficiency decreased due to the loss of net power output by integration of CO<sub>2</sub> capture unit [34]- this leads to the increase of CO<sub>2</sub> intensity as indicated in Fig. 1. It is worth noting that total amount of CO<sub>2</sub> avoided ( $W_{ref,net}E_{ref} - W_{m,net}E_m$ ) is the same even though specific CO<sub>2</sub> avoided (kg CO<sub>2</sub> avoided / kWh net electricity output,  $E_{ref} - \frac{W_{m,net}E_m}{W_{ref,net}}$ ) based on reference plant (specific CO<sub>2</sub> avoided (ref)) is different from specific CO<sub>2</sub> avoided based on capture plant ( $\frac{W_{ref,net}E_{ref}}{W_{m,net}} - E_m$ ) as illustrated in Fig. 1.

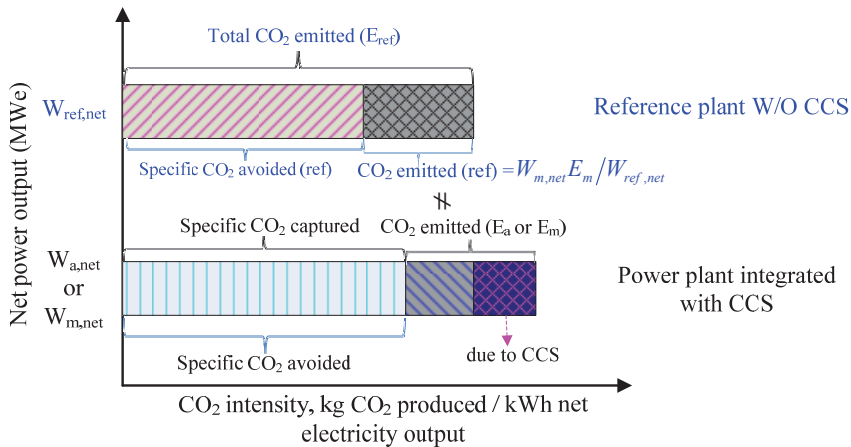


Fig. 1. Illustration of CO<sub>2</sub> intensity in power plant with and without CCS

Specific primary energy consumption for CO<sub>2</sub> avoided (SPECCA, MJ<sub>th</sub>/kg CO<sub>2</sub>) was used to estimate energy intensity of different processes reported in the literature [32], which is described as follows,

$$SPECCA = \frac{Q - Q_{Ref}}{E_{Ref} - E} = \frac{3600 \times (\frac{1}{\eta} - \frac{1}{\eta_{Ref}})}{E_{Ref} - E} \tag{1}$$

where  $Q$  and  $E$  are thermal energy / heat rate (kJ<sub>LHV</sub>/kWh) and CO<sub>2</sub> emission rate (kg CO<sub>2</sub> /kWh).  $\eta$  is power plant efficiency,  $Ref$  is reference plant without CCS. It is worth noting that  $(E_{ref} - E)$  in Eq. (1) may be negative if energy penalty (i.e., reduction in net power output) of a process is very high (e.g., >50 %) together with a low capture ratio (e.g., <50 %). Thus, Eq. (1) is modified as,

$$SPECCA = \frac{3600 \times (\frac{1}{\eta} - \frac{1}{\eta_{Ref}})}{\frac{W_{ref,net}}{W} E_{Ref} - E} \tag{2}$$

where  $W$  (MWe) is net power output in power plant. Energy consumption estimated from Eq. (2) is thermal energy which is different from membrane systems where only electricity is used without any heat duty. Thus, in

order to compare energy consumption of membrane systems with amine absorption, specific equivalent power consumption for CO<sub>2</sub> avoided (SEPCCA) is then employed. Table 1 shows the parameters of reference plant without capture and retrofitting plant integrated with CO<sub>2</sub> capture systems (amine or membrane).

Table 1 Comparison of cases with and without CO<sub>2</sub> capture

Parameter	Unit	Without capture (Reference)	Capture with amine system	Capture with membrane system
Gross power output	MWe	$W_{ref}$	$W_a$	$W_m$
Auxiliary power consumption	MWe	$W_{ref,aux}$	$W_{ref,aux} + W_{a,aux}^{\#}$	$W_{ref,aux} + W_{m,aux}^*$
Net power output	MWe	$W_{ref,net}$	$W_{a,net}$	$W_{m,net}$
Thermal efficiency	% LHV	$\eta_{ref}$	$\eta_a$	$\eta_m$
CO <sub>2</sub> emitted	Kg/MWh <sub>net</sub>	$E_{ref}$	$E_a$	$E_m$
SEPCCA	MJ <sub>c</sub> /kg CO <sub>2</sub>	-	Eq. (3)	Eq. (4) or Eq. (5)

$W_{a,aux}^{\#}$ : power consumption of blowers, solvent pumps and CO<sub>2</sub> compressors and pumps,  $W_{m,aux}^*$ : power consumption of flue gas compressors, inter-stage compressors, vacuum pumps and CO<sub>2</sub> compressors and pumps.

For benchmark technology of amine absorption, SEPCCA can be estimated by,

$$SEPCCA = \frac{3600 \times (W_{ref,net} - W_{a,net})}{W_{ref,net} E_{ref} - W_{a,net} E_a} = \frac{3600 \times (\eta_{ref} - \eta_a)}{\eta_{ref} E_{ref} - \eta_a E_a} \quad (3)$$

SEPCCA (MJ<sub>c</sub>/ kg CO<sub>2</sub>) is mainly dependent on plant thermal efficiency penalty ( $\eta_{ref} - \eta_a$ ) due to extra energy consumption from CCS units. Reduction of heat duty for solvent regeneration can significantly reduce energy consumption in amine-based absorption system.

While for membrane systems, SEPCCA can be calculated by,

$$SEPCCA = \frac{3600 \times (W_{ref,net} - W_{m,net})}{W_{ref,net} E_{ref} - W_{m,net} E_m} \quad (4)$$

or

$$SEPCCA = \frac{3600 \times W_{m,aux}}{W_{ref,net} E_{ref} \phi} \quad (5)$$

where  $\phi$  is CO<sub>2</sub> capture ratio or recovery (ratio of CO<sub>2</sub> flow rate between permeate and feed). Eq. (4) is typically used to estimate energy consumption of CO<sub>2</sub> capture in power plants. While for CO<sub>2</sub> capture from industry such as cement, refinery, and steel/iron production plants, electricity is directly bought from power plants with a specific price (this electricity cost covers CO<sub>2</sub> capture cost in power plants) to provide power demands for capture units. Thus, Eq. (5) is then employed to evaluate membrane systems for CO<sub>2</sub> capture from off-gas in industry. Energy consumption of membrane systems for CO<sub>2</sub> capture is now determined by total power demands of major driving equipment and CO<sub>2</sub> capture ratio.

#### 4. The criteria of cost estimation

Capital cost is estimated on the basis of major equipment in a process (e.g., compressor, blower, pump, expander, heat exchanger, absorber, stripper and membrane unit), which can provide an accuracy in the range of -25% to 40%, and is typically used for the preliminary feasibility analysis of different techniques. Bare module costing ( $C_{BM}$ ) technique accounts to purchased cost ( $C_p^0$ ) of equipment in the base condition (carbon steel material and near ambient pressure), and a multiplying bare module factor ( $F_{BM}$ ) is employed to cover the influences of any specific equipment type, specific materials for construction and operating pressure. Bare module cost ( $C_{BM}$ ) for each piece of equipment is sum of direct and indirect costs,

$$C_{BM} = C_p^0 F_{BM} \quad (6)$$

An excel program of CAPCOST 2012 is used to estimate capital cost based on equipment module approach [35]. Total capital cost ( $C_{TM}$ ) including contingency and contractor fee in addition to direct and indirect cost is calculated as follows:

$$C_{TM} = 1.18 \sum_{i=1}^n C_{BM,i} \quad (7)$$

where  $n$  is the total number of individual units. A chemical engineering plant cost index (CEPCI) of 467.2 for equipment is adopted for all inflation adjustments of 2013. Annual capital related cost (CRC) is estimated to be 20 % of total capital cost ( $C_{TM}$ ). Annual operating expenditure (OPEX) only considers electricity and labor costs to simplify cost estimation (price is based on the literature [30, 36, 37]) in this work. A schematic illustration of cost estimation for CO<sub>2</sub> capture with membrane system is shown in Fig. 2. The specific CO<sub>2</sub> capture cost significantly depends on process condition (e.g., temperature, pressure) and membrane material properties (i.e., selectivity and permeance) which should be optimized in a specific application to minimize cost. Operating cost mainly depends on electricity cost, while capital cost is determined by major driving equipment and membrane unit costs. Annual productivity depends on CO<sub>2</sub> capture ratio at a given feed flow. Thus, specific CO<sub>2</sub> capture cost can be estimated based on capital cost, operating cost and capture ratio.

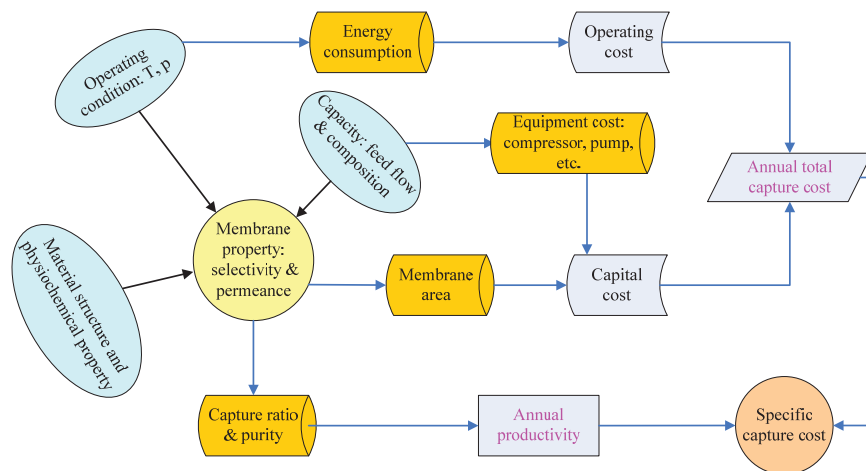


Fig. 2. Framework of CO<sub>2</sub> capture cost estimation with membrane system

In order to simplify capital cost estimation, specific equipment cost listed in Table 2 can also be employed to calculate bare module cost ( $C_{BM}$ ) of major equipment. Membrane unit cost is mainly dependent on membrane materials and production cost. Polymeric membranes cost is usually lower compared to inorganic membranes, and a membrane skid cost between 20 \$/m<sup>2</sup> Koros et al. [38] and 50 \$/m<sup>2</sup> Merkel et al. [37] were used to estimate membrane unit cost in the literature. Table 3 shows a general cost estimation model of membrane system for CO<sub>2</sub> capture from flue gas in power plant or off-gas in industry. Annual CO<sub>2</sub> capture cost is determined by annual CRC and OPEX. It is worth noting that CO<sub>2</sub> capture cost described here is different from CO<sub>2</sub> avoided cost reported by Rao et al. [31] where CO<sub>2</sub> avoided cost was estimated by,

$$CO_2 \text{ avoided cost } (\$/\text{tonne}) = \frac{(\$/\text{MWh})_{\text{capture}} - (\$/\text{MWh})_{\text{reference}}}{(\text{tonne } CO_2/\text{MWh})_{\text{reference}} - (\text{tonne } CO_2/\text{MWh})_{\text{capture}}} \quad (8)$$

Eq. (8) is only applicable when net power output is the same in reference plant and CO<sub>2</sub> capture plant. However, this is obviously impossible if fuel input is constant. It is also found that CO<sub>2</sub> avoided cost calculated from Eq. (8) can be negative if CO<sub>2</sub> capture ratio is quite low (i.e.,  $E_{\text{ref}} < E_m$  or  $E_{\text{as}}$ , see section 2) as different baselines are chosen in the numerator and denominator of Eq. (8). Thus, CO<sub>2</sub> avoided cost with amine absorption and membrane system is re-estimated by equations (9) and (10), respectively.

$$CO_2 \text{ avoided cost } \left( \frac{\$}{\text{tonne}} \right) = \frac{(\$ / MWh)_{\text{capture}} - (\$ / MWh)_{\text{reference}}}{\frac{W_{ref,net}}{W_{a,net}} E_{ref} - E_a} \times 1000 \quad (9)$$

and

$$CO_2 \text{ avoided cost } \left( \frac{\$}{\text{tonne}} \right) = \frac{(\$ / MWh)_{\text{reference}} \left( \frac{W_{ref,net} - W_{m,net}}{W_{m,net}} \right) + \left( \frac{CRC + \text{Annual LC}}{W_{m,net} \times 7500} \right)}{\frac{W_{ref,net}}{W_{m,net}} E_{ref} - E_m} \times 1000 \quad (10)$$

The first item in the right side of Eq. (10) is the increase of electricity generation cost due to loss of net power output which covers the electricity used in CO<sub>2</sub> capture unit. The second item represents extra CO<sub>2</sub> capture unit cost (excluding electricity cost in OPEX). Eq. (10) can be used to assess CO<sub>2</sub> capture cost in power plant. However, estimation of electricity generation cost in a retrofitted plant integrated with CCS could be quite complex compared to cost estimation model described in Table 3 which provides an easier way for economic feasibility analysis of CO<sub>2</sub> capture with membrane systems.

Table 2 Specific bare module cost of major equipment

Equipment	Material	Type	Specific cost, \$/kW
Flue gas compressor	Carbon steel	Centrifugal	850
Inter-stage compressor	Carbon steel	Centrifugal	850
Expander	Carbon steel	Radial	630
Vacuum pump	Carbon steel	Rotary	1300
CO <sub>2</sub> pump	Stainless steel	Centrifugal	1350
CO <sub>2</sub> compressor	Stainless steel	Centrifugal	1800

Table 3 Cost estimation model for CO<sub>2</sub> capture with membrane system

Category	Parameter	Value
Capital Expenditure (CAPEX)	Membrane unit cost (C <sub>BM, M</sub> )	20~50 \$/m <sup>2</sup>
	Membrane replacement cost	20% × C <sub>BM, M</sub>
	Compressor, vacuum pump and turbo expander cost (C <sub>BM, i</sub> )	Eq. 6 or Table 2
	Total capital cost (C <sub>TM</sub> )	Eq. 7
Annual Operating Expenditure (OPEX)	Labor cost (LC)*	15 \$/hr
	Electricity cost (PC) <sup>#</sup>	0.04 \$/kWh
	OPEX	LC + PC
Annual capital related cost (CRC)	0.2 × C <sub>TM</sub> (covering depreciation, interest and maintenance)	
CO <sub>2</sub> capture cost	(CRC+OPEX) / annual CO <sub>2</sub> captured, \$/tonne CO <sub>2</sub> captured	
Other assumptions	Membrane lifetime	5 year
	Project lifetime	25 year
	Operating time	7500 hrs/year

\*: direct labor cost per 25 MMSCFD, #: electricity price varies widely from country to country

## 5. Case study -CO<sub>2</sub> capture from coal fired power plant

### 5.1. Process description and simulation basis

An advanced supercritical (ASC) coal fired power plant with a gross output 819 MWe without CO<sub>2</sub> capture reported in CESAR project [32] was chosen as reference base. The plant thermal efficiency was estimated to be 45.5% based on a net power output of 754 MWe and a fuel input LHV 1657.1 MWth together with a 65 MWe auxiliary power consumed. CO<sub>2</sub> capture unit is located right after pre-treatment units of selective catalytic reduction (SCR) DeNO<sub>x</sub> unit, electrostatic precipitator and flue gas desulphurization (FGD), and the characteristics of flue gas are shown in Table 4. Only the main components of CO<sub>2</sub>, N<sub>2</sub>, O<sub>2</sub> and water were considered to simplify process simulation in the current work. For membrane system, condensed water is removed from flue gas before feeding into membrane unit (see Fig. 3, condenser). In order to get high membrane separation performance, ca. 2.3% water vapor (RH >90 %, at 35 °C and 2.5bar) is maintained in feed gas stream.

Table 4 Characteristics of flue gas in reference plant

Parameter	Value
Flue gas flow rate, kmol/h	9.58E+4
Temperature, °C	50
Pressure, bar	1.016
Master component mole fraction (mol. %) <sup>*</sup>	
CO <sub>2</sub>	13.74
N <sub>2</sub>	72.88
O <sub>2</sub>	3.65
H <sub>2</sub> O	9.73

<sup>\*</sup>: the impurities of SO<sub>2</sub>, NO<sub>x</sub> and fly ashes are not included here.

CO<sub>2</sub> capture ratio was found to significantly influence energy consumption and capture cost in membrane system, and increasing capture ratio will evidently increase energy consumption [36]. It was also reported that pursuing an excessively high capture ratio may lead to a much higher capture cost at the same CO<sub>2</sub> purity [36]. Thus, a CO<sub>2</sub> capture ratio of 90 % was set in simulation. Moreover, CO<sub>2</sub> purity of 90 % and 95 % were reported in the literature [9, 27, 30, 36, 39]. A 95 % CO<sub>2</sub> purity was set as separation requirement for membrane system in this work.

In order to document process and economic feasibility of membrane systems and compare to conventional amine absorption system, a two-stage membrane system is designed for CO<sub>2</sub> capture from the above mentioned power plant using high performance polyvinylamine (PVAm) / polyvinylalcohol (PVA) blend FSC membranes (developed by Memfo group at NTNU) based on the following assumptions, and simulation basis is listed in Table 5.

- A moderate CO<sub>2</sub> permeance of 2 m<sup>3</sup> (STP) / (m<sup>2</sup>.h.bar) at a feed and permeate pressure of 2.5 bar and 250 mbar (optimal pressure reported by He et al. [40]) was employed. This performance is relatively lower (considering the influences of real flue gas and operating condition) compared to experimental data reported by Kim et al. [13].
- A CO<sub>2</sub> / N<sub>2</sub> and CO<sub>2</sub> / O<sub>2</sub> selectivity of 135 and 30 were set considering a high stage-cut (10-20 %) that should be achieved in the real process. Selectivity of CO<sub>2</sub>/H<sub>2</sub>O is assumed to 1.
- The efficiency of compressor, expander and pump is assumed to be 85 %.
- A counter-current configuration is used for the membrane transport model
- The captured CO<sub>2</sub> was compressed to 75 bar and pumped to 110 bar for pipeline transportation.

Table 5 Simulation basis of membrane system for CO<sub>2</sub> capture

Parameter	1st & 2nd stage
Feed pressure, bar	2.5
Permeate pressure, bar	0.25
Temperature, °C	35

CO <sub>2</sub> permeance, m <sup>2</sup> (STP)/(m <sup>2</sup> .h.bar)	2
CO <sub>2</sub> /N <sub>2</sub> selectivity	135
CO <sub>2</sub> /O <sub>2</sub> selectivity	30
CO <sub>2</sub> /H <sub>2</sub> O selectivity	1
CO <sub>2</sub> capture ratio, %	90
CO <sub>2</sub> purity, %	95

### 5.2. Process simulation

A 30% MEA solution was used for CO<sub>2</sub> capture in CESAR project where absorber and stripper were operated at 40-60 °C and 120 °C, respectively. A CO<sub>2</sub> captured flow rate of 518.8 tonne/h was achieved at a capture ratio of 89 % [32]. Thermal efficiency penalty was found to be 12.1 %, and the SPECCA and SEPCCA were estimated to be 3.0 MJ<sub>th</sub> (calculated by eq. (2)) and 1.42 MJ<sub>e</sub> (estimated by Eq. (3)) per kg CO<sub>2</sub> avoided as shown in Table 6. In order to compare energy efficiency and techno-economic feasibility of a membrane system with a conventional amine system, HYSYS simulation integrated with ChemBrane unit (developed by Memfo group at NTNU) was conducted for CO<sub>2</sub> capture from flue gas with membrane system. It has already been reported that single stage membrane unit cannot accomplish the specific separation requirement for both high CO<sub>2</sub> capture ratio (> 80%) and CO<sub>2</sub> purity (> 95%) simultaneously in our previous work [8, 9]. In addition, energy efficiency could be significantly improved using a multiple-stage membrane system to reduce the irreversibility of the whole process as reported by Zhang et al. [36]. Thus, a two-stage cascade membrane system related to permeate stream was designed for process simulation and feasibility analysis. The schematic process flow diagram (PFD) is shown in Fig. 3. The first stage membrane unit is used for pre-concentration of CO<sub>2</sub> and controlling CO<sub>2</sub> capture ratio, while second stage membrane unit is employed for final CO<sub>2</sub> purification up to 95 %. Flue gas was initially compressed to a given pressure (2.5bar), condensed water was removed using condenser. Compressed flue gas with high relative humidity (RH>90%) is then fed into the first stage membrane unit. Permeate stream is re-compressed to 2.5bar and fed into 2<sup>nd</sup> stage membrane for final purification to reach CO<sub>2</sub> purity >95%. N<sub>2</sub> concentrated retentate are re-heated with compression heat to recover more work from expander. The captured CO<sub>2</sub> was compressed to 75bar using multi-stage compressors with intercooling and further pumped to 110bar for pipeline transportation. It is worth noting that vacuum pump is not standard equipment in HYSYS, and its power consumption is estimated by compression with compressors,

The simulation results are shown in Table 6 and Table 7. Specific power consumption was estimated to be 316.06 kWh/tonne CO<sub>2</sub> captured. The efficiency penalty of 10% for membrane systems was found to be relatively lower compared to a typical MEA system (~12.4%) reported in CESAR project [32]. The SEPCCA of 1.09 MJ<sub>e</sub>/kg CO<sub>2</sub> avoided estimated by Eq. (4) was also lower than MEA absorption system (1.42 MJ<sub>e</sub>/kg CO<sub>2</sub> avoided [36]) as indicated in Table 6. Moreover, energy consumption could be further brought down by integration of compression heat which will be further investigated in future work.

Table 6 Comparison of energy consumption between MEA and membrane systems

Parameter	Unit	Without capture (Reference)	Capture with MEA system [32]	Capture with membrane system
Gross power output	MWe	819	684.2	819
Auxiliary power consumption	MWe	65	135	65+165
Net power output	MWe	754	549.2	589
Efficiency	% LHV	45.5	33.1	35.5
CO <sub>2</sub> emitted	kg/MWh <sub>net</sub>	763	104.7	98.2
CO <sub>2</sub> captured	tonne/h	-	518.8	521.1
SPECCA	MJ <sub>th</sub> /kg CO <sub>2</sub>	-	3.0#	-



SEPCCA	MJe/kg CO <sub>2</sub> -	1.42*	1.09
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#: estimated by Eq. (2), \*: calculated by Eq. (3)

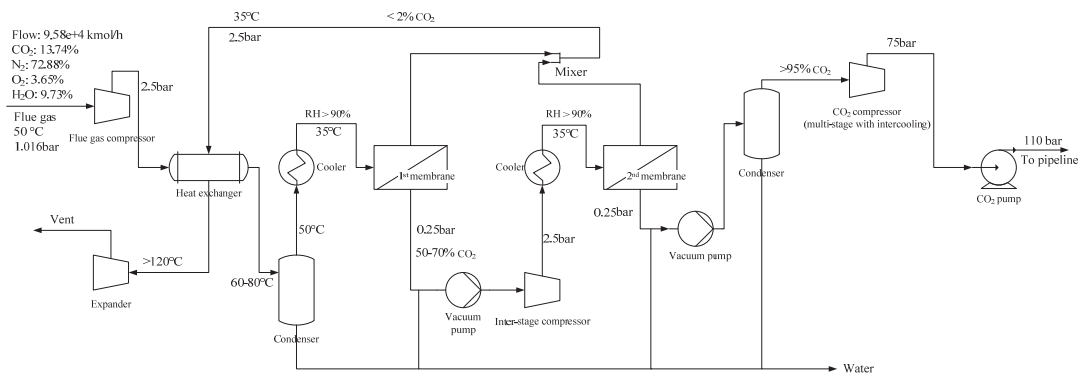


Fig. 3. Schematic PFD of a two-stage membrane system for CO<sub>2</sub> capture from flue gas

Table 7 Simulation results of membrane system for CO<sub>2</sub> capture

Parameter	Simulation results
Flue gas compressor, kw	8.55E+04
Vacuum pump <sup>#</sup> , kw	5.13E+04
Inter-stage compressor, kw	2.10E+04
CO <sub>2</sub> compressor, kw	4.86E+04
CO <sub>2</sub> Pump, kw	2.14E+03
Expander, kw	-4.38E+04 <sup>†</sup>
Total net power consumption, kw	1.65E+05
CO <sub>2</sub> capture rate, tonne/h	521.10
Specific power consumption, kWh /tonne CO <sub>2</sub> captured	316.06
CO <sub>2</sub> capture ratio, %	90.01
CO <sub>2</sub> purity, %	95.67
Total membrane area, m <sup>2</sup>	4.12E+06
Heat transfer surface area <sup>§</sup> , m <sup>2</sup>	2.07E+04

#: estimated with compressors; <sup>†</sup>: Expander produce work; <sup>§</sup>: Heat exchanger design based on Aspen Exchanger Design and Rating V8.0

### 5.3. Economic feasibility analysis

Cost estimation was conducted to evaluate economic feasibility of CO<sub>2</sub> capture with membrane system. Bare module costs of major equipment (coolers and condensers are not included) are estimated on the basis of specific equipment cost and power consumption listed in Table 2 and Table 7, respectively. A membrane price of 35 \$/m<sup>2</sup> is used to assess membrane unit cost considering a cheaper commercially available material (PVAm and PVA) for large-scale production of FSC membranes. The total capital cost (C<sub>TM</sub>) is estimated by cost of major driving equipment and heat exchanger together with membrane unit. A 20 % of total capital cost is then employed to

estimate annual CRC covering depreciation, interest and maintenance, while annual OPEX is estimated by labor and electricity cost. Table 8 shows economic analysis results of CO<sub>2</sub> capture with membrane systems. The specific CO<sub>2</sub> capture cost is estimated to be 47.3 \$/tonne CO<sub>2</sub> captured based on annual CRC and OPEX (almost the same with 47.6 \$/tonne avoided estimated by Eq. (10)), which indicates that membrane system is competitive to conventional amine absorption system of 59.1 \$/tonne CO<sub>2</sub> avoided [31] and 33 € / tonne CO<sub>2</sub> avoided [41]. However, water vapor in flue gas should be further investigated to document the feasibility of FSC membranes for CO<sub>2</sub> capture in a more realistic way. Moreover, it was found that membrane unit cost covers >50 % of annual CRC which can be possibly reduced by improving membrane performance (especially gas permeance). The latest high performance membranes has been reported with a CO<sub>2</sub> permeance up to 5 m<sup>3</sup> (STP) / (m<sup>2</sup>.h.bar) of a FSC membrane (developed by Memfo group at NTNU) [13] and 1500 GPU (4.1 m<sup>3</sup> (STP) / (m<sup>2</sup>.h.bar)) of Polaris™ membrane (MTR Inc.) [37] and 4.3 m<sup>3</sup> (STP) / (m<sup>2</sup>.h.bar) of Polyactive® composite membrane (Helmholtz-Zentrum Geesthacht) [42]. Their contributions could significantly promote to bring environmentally friendly membrane technology into commercial application of CO<sub>2</sub> capture from flue gases in the near future.

Table 8 Economic feasibility analysis of CO<sub>2</sub> capture from flue gas

Parameter	Unit	Value
Bare module cost*, CBM, i	M\$	280.0
Membrane unit cost, CBM, M	M\$	144.2
Membrane replace cost	M\$	115.4
Total capital cost, CTM	M\$	636.8
Annual CRC	M\$	127.4
Annual OPEX	M\$	57.6
Annual CO <sub>2</sub> captured	MMTPA#	3.9
Specific CO <sub>2</sub> capture cost	\$/ tonne CO <sub>2</sub> captured	47.3
Specific CO <sub>2</sub> avoided cost§	\$/ tonne CO <sub>2</sub> avoided	47.6

\*: including the major equipment of compressor, vacuum pump, CO<sub>2</sub> pump and expander and heat exchanger, #: Million Metric Tonne Per Annum, §: calculated based on Eq. (10), and the electricity generation cost of reference plant is assumed to be 40 \$/MWh.

## 6. Conclusions

The proposed criteria on energy consumption and cost estimation could be well used for techno-economic feasibility analysis of different membrane systems for CO<sub>2</sub> capture by easily adjusting membrane performance and/or membrane price. Specific equivalent power consumption for CO<sub>2</sub> avoided was used to evaluate energy consumption in different CO<sub>2</sub> capture processes, which provides the unique criteria for process feasibility analysis. The SEPCCA of membrane system was compared to conventional amine absorption process based on the case study of CO<sub>2</sub> capture from a gross power output 819 MWe coal fired power plant. HYSYS simulation results showed that membrane systems had relatively lower efficiency penalty 10% and energy consumption 1.09 MJ<sub>e</sub>/kg CO<sub>2</sub> avoided compared to conventional MEA absorption system ca. 12.4% and 1.42 MJ<sub>e</sub>/kg CO<sub>2</sub> avoided, respectively. The case study indicated that membrane system is one of the most costly units which can be further brought down by improving membrane performance and process optimization. The investigated FSC membrane system shows a nice potential for CO<sub>2</sub> capture from flue gas based on a CO<sub>2</sub> capture cost of 47.3 \$ per tonne CO<sub>2</sub> captured, but water vapor influence in flue gas should be further investigated.

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