Post-combustion carbon capture technologies: energetic analysis and life cycle assessment

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Abstract

An integrated framework focusing on the energetic analysis and environmental impacts of a CO_2 capture and storage (CCS) system is presented, in which the process simulation method and the life cycle assessment (LCA) method are integrated and applied to the CCS value chain. Three scenarios for carbon capture from post-combustion power plant - an MEA-based system, a gas separation membrane process and a hybrid membrane-cryogenic process are studied. The energy efficiency of power plant and the specific capture energy consumption for each scenario are estimated from process simulation. The environmental impacts for each scenario and the base case without CCS are assessed with LCA method. The results show that the MEA-based capture system faces the challenges of higher energy consumption, and higher environmental impact caused by solvent degradation and emissions compared to gas membrane separation processes. The hybrid membrane-cryogenic process shows a better environmental potential for CO_2 capture from flue gases due to much lower power consumption and relatively lower environmental impacts.

Keywords: Carbon capture, Gas separation membrane, Cryogenic process, Hybrid membrane-cryogenic process, process simulation, life cycle assessment

1. Background

CO$_2$ emissions from fossil based industries have contributed to the serious global warming problems. CO$_2$ capture and storage (CCS) has been regarded as one of the most promising options to utilize fossil fuels continuously without the significant influence to the climate change. Till now, a large number of studies have focused on the assessment of energy consumption, capture cost and environmental impacts in CCS, while most of them analysed MEA-based capture systems which have been proven in chemical production industries, for example, process analysis and techno-economic assessment (Abu-Zahra et al., 2007; Huang et al., 2010; Husebye et al., 2011; IEA, 2006; Sanpasertparnich et al., 2010; Schach et al., 2010; Sipocz and Tobiesen, 2012) and environmental impact assessment(Nie et al., 2013) (Koornneef et al., 2011; Koornneef et al., 2012; Korre et al., 2010; Pehnt and Henkel, 2009; Schreiber et al., 2009; Singh et al., 2011a; Singh et al., 2011b). Research on some emerging capture technologies such as membranes, ionic liquids and metal organic frameworks (MOFs) is in progress with the aim of reducing capture energy consumption and capital investment (Favre, 2011; Figueroa et al., 2008; MacDowell et al., 2010; Zhang et al., 2012b), and shows a promising application in CO$_2$ capture. However, these processes are still at the lab-scale or small pilot-scale demonstration stage. In this paper, an integrated framework is proposed and used to assess the capture energy consumption and environmental impacts of the different CCS chains with conventional and emerging capture technologies, considering three post-combustion CO$_2$ capture technologies: an MEA-based system, a gas separation membrane process, and a hybrid membrane-cryogenic process.

Gas separation membranes have generated growing interests in recent years, as unlike MEA-based system, CO$_2$ capture using membrane needs no or little chemicals, requires no retrofitting for the existing power plants, have relative ease of scale-up and operation, (Ritter and Ebner, 2007; Zhao et al., 2010), and is flexible for separating gases when high purity gas streams are not vital (Powell and Qiao, 2006) At present, most of the studies on gas separation membranes focus on the membrane material selection and functionalization, membrane preparation and characterization (Aaron and Tsouris, 2005; Powell and Qiao, 2006; Scholes et al., 2008), and membrane process analysis(Bounaceur et al., 2006; He et al., 2009; Hussain and Hagg, 2010; Merkel et al., 2010; Van Der Sluijs et al., 1992; Zhao et al., 2010; Zhao et al., 2008). For instance, the parametric studies and the systematic energy analysis of a single stage membrane process (Bounaceur et al., 2006; Zhao et al., 2008), the energetic and

economic analyses of multi-stage membrane processes (Zhao et al., 2010), the influences of membrane parameters and process configurations on the energy consumption and cost considering a real industrial process (Merkel et al., 2010), the process feasibility of post-combustion of the real flue gas by facilitated transport membrane based on process simulation and cost estimations (Hussain and Hagg, 2010) as well as the process feasibility analysis of hollow fiber carbon membranes for CO_2 capture from flue gases (He and Hagg, 2011, 2013). These contributions are important for understanding the feasibilities of CO_2 capture with membrane technologies; however, the assessment results are not directly comparable as different authors use different membranes and process parameters, membrane models, process configuration and assumptions. To the best of our knowledge, there are no studies focusing on the environment impacts of membrane technologies, thus no quantitative results can be referred to understand their environmental benefits and concerns.

In our previous work (Zhang et al., 2013), we conducted the analysis for parametric influence, capture cost and exergy efficiency of the post-combustion membrane based carbon capture process. As for environmental issues, it has been shown in literature that a CCS system with an MEA-based capture method though can achieve a significant reduction of CO_2 emissions but have multiple environmental trade-offs(Koornneef et al., 2011; Koornneef et al., 2012; Korre et al., 2010; Pehnt and Henkel, 2009; Schreiber et al., 2009; Singh et al., 2011a; Singh et al., 2011b). Although there are no MEA degradation and solvent losses in a membrane process, the preparation and replacement of membrane might cause additional environmental impacts. Thus, assessing systematically the energy consumption and environmental impacts of membrane capture technologies is important.

In this study, we evaluate the energy consumption and environmental impacts of three different post-combustion capture technologies, i.e., an MEA-based system, a gas separation membrane process, and a hybrid membrane-cryogenic process, using energetic analysis and life cycle assessment (LCA) method. At first, the capture process for each scenario is designed and simulated on the basis of engineering heuristics and judgment to determine the relevant operating parameters, and then the process simulation and optimization are performed to obtain the detailed energy and mass flows, as well as the parameters of all unit operations involved in the whole system. Based on the simulation results and process configuration of the CCS system, the LCA method is applied to evaluate the environmental impacts of the system considering the complete life cycle from resource mining to CO_2 injection.

2. Methodology

The concept of the integrated method and the system boundary of CCS involved in this study are shown in Fig. 1. The main motivation of this work is to investigate the performance of energy consumption and environmental impacts in a designed system framework with process simulation and life cycle assessment. The system boundary for LCA will cover the CCS value chain from resource extraction, pre-treatment, distribution and transport, infrastructure, electricity generation (advanced super critical boiler and turbine), CO_2 capture (three capture technologies: an MEA-based system, a two-stage membrane system and a hybrid membrane/cryogenic process), CO_2 transport (pipeline) and storage. The detailed descriptions are given as follows.
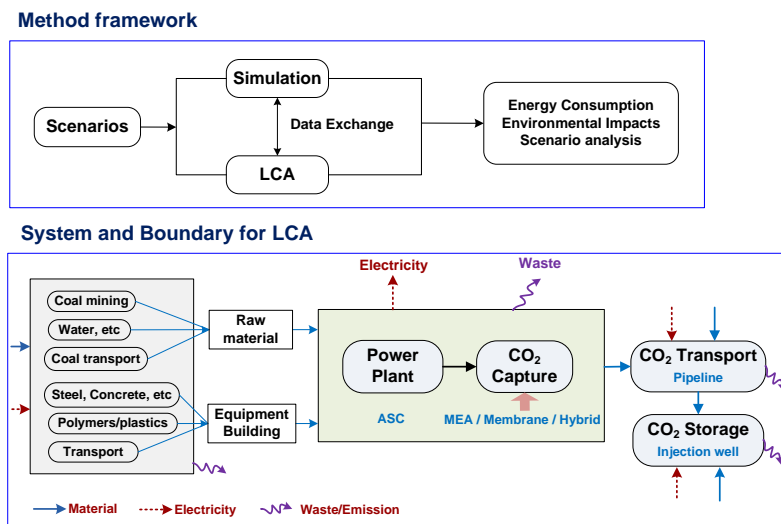
Method framework

Fig. 1. Integrated method and system boundary of LCA

2.1 Process models

The power plant is based on an advanced super critical boiler and turbine delivering 819 MWe (gross) without carbon capture. When the auxiliary power is taken into account, the net power output is 754.3 MWe, yielding a net cycle efficiency of 45.5%. The models involved in the power plant and the flue gas composition as well as the parameters are from the literature (Anantharaman et al., 2011).

For the MEA-based capture process, the mechanism of CO_2 absorption in an amine solution using an absorption column is quite complex, and there are many references about

the chemical reactions and vapor-liquid equilibrium involved in this system (Aaron and Tsouris, 2005; MacDowell et al., 2010). It is a typical electrolyte thermodynamic system and the amines property package in Aspen Plus® is often used to model the absorption and desorption processes. For the unit operations, a RadFrac model is used in both the absorber and the stripper. The absorber is a simple RadFrac column, whereas the stripper column has a condenser on the top and a reboiler at the bottom. Considering the low efficiency of the gas absorber, it is modeled with a Murphree efficiency of 25~30% (Husebye et al., 2012). In the MEA-based CO_2 capture process, another major issue is the degradation of the solvents through the irreversible side reactions with other flue gas components as SO_2, NO_x and O_2, leading to solvent losses and degradation wastes (Ho et al., 2011; Koornneef et al., 2008; Portal, 2011). We assumed the MEA losses are mainly due to emissions to air with the sweet flue gas and oxidative degradation forming heat stable salts (Uyanga and Idem, 2007; Veltman et al., 2010).

For a gas separation membrane process, the membrane model used in this work is applicable to any high flux asymmetric membranes in a cross-flow pattern which can meet the assumptions described in the literature (Pan, 1983; Zhao et al., 2008). The equations and the detailed calculation procedure can be found in the literature (Pan, 1983). Here, the simulation is performed by PRO/II® software (Simulation Science, Inc.)(Zhao et al., 2010; Zhao et al., 2008). The Flowsheet Optimizer unit operation in the software is used to optimize the parameters such as feed gas pressure and membrane area based on the minimization of the power consumption to achieve as a specific separation requirement on CO_2 capture ratio and CO_2 purity(Zhang et al., 2013). The energy consumption for gas separation membrane process is mainly from the electric energy demands by gas compressors, vacuum pumps etc, which is different from an MEA-based capture process where the energy consumption is mainly from the heat energy demand in regeneration of the CO_2-rich solvent in the stripper. The cryogenic gas separation process is simulated with Aspen Plus® software. Soave-Redlich-Kwong (SRK) model is used to calculate the thermodynamic properties, and the multi-stream heat-cold exchanger model, the flash model and expander model are used to simulate the gas-liquid separator and adiabatic expansion unit.

2.2 Life cycle assessment

Regarding CCS, one of the pendent questions is whether CCS is good for the environment, or does the reduction in climate change-related damage outweigh the increase of the other

environmental impacts? In order to answer these questions, the LCA method can give an overview of the interventions caused by or required by the processes within the boundary of the studied system. The boundary is determined by the concept called Cradle to Grave i.e from raw material production to end-of-life disposal of product. In a CCS system, the boundary covers the whole CCS value chain, from coal mining to CO_2 storage with injection as illustrated in Fig. 1.The compressed CO_2 is assumed to be transported over 200 km via pipeline and injected into a secure offshore site at 800m depth for storage.

For the LCA, the detailed unit and process level information obtained from the process simulation data for the power plant and CO_2 capture system is used in combination with the Ecoinvent v2.2 database (EcoinventCentre, 2010) to model the value chain upstream and downstream processes. The characterization factors from ReCiPe method v1.03 (ReCiPe, 2010) are used to estimate the potential environmental impacts of the emissions incurred. The environmental impacts are categorized as 10 environmental mid-point indicators: GWP (global warming) - CO_2eq; FEP (freshwater eutrophication) - P eq; MEP (marine eutrophication) - N eq; TAP (terrestrial acidification) - SO_2eq; PMFP (particulate matter formation) - PM10 eq; POFP (photochemical oxidant formation) - NMVOC; ODP (ozone depletion) - CFC-11 eq; HTP (human toxicity) - 1,4 DCB eq; FETP (freshwater ecotoxicity) - 1,4 DCB eq; TETP (terrestrial ecotoxicity) - 1,4 DCB eq.

To understand the contribution of specific value chain processes to the net GHG emissions, the CCS value chain defined in this work is divided into five sub-chains (SubCh-1 to SubCh-5). SubCh-1 refers to the infrastructures of power plant chain, capture plant, transport (pipeline) and CO_2 storage (well). SubCh-2 is the coal supply chain for the whole system. SubCh-3 is the capture specific infrastructure and supplies, for example, MEA solvent make-up input, packing in the tower, membrane material inputs and so on. SubCh-4 refers to the combustion chain, i.e., the coal burned in power plant and SubCh-5 is the CO_2 capture process.

3. Process description

3.1 Flue gas properties

The base case of the power plant is an advanced super critical boiler and turbine delivering 819 MWe (gross) without carbon capture. The feedstock coal is South African Douglas Premium 2 with the flow rate 65.8 kg/s. The composition of the flue gas from the coal power plant is shown in Table 1 (Anantharaman et al., 2011). Flue gas is pretreated to remove fly

ashes and acid gases of SO_2 and NO_x before feeding into the CO_2 capture system. For capturing 90% (capture ratio) of the total CO_2 in the flue gas, the capacity of CO_2 capture unit is estimated about 3.9 million ton per year.

Table 1. Flue gas composition from post-combustion coal power plant (Anantharaman et al., 2011)

Parameters	Unit	Raw flue gas
Flue gas flow rate	kg/s	781.8
Temperature	°C	50
Pressure	bar	1.016
Composition		
O_2	Vol. % Wet	3.65
CO_2	Vol. % Wet	13.73
SO_2	mg/Nm^3	85
NOx	mg/Nm^3	120
H_2O	Vol. % Wet	9.73
Ar	Vol. % Wet	0.005
N_2	Vol. % Wet	72.86
Particulate	mg/Nm^3	8

3.2 Process design

Fig.2 shows the process flow diagrams of three different CCS scenarios. In the MEA-base capture process (Fig. 2(a)), the flue gas from power plant passes through an existing flue gas desulphurization (FGD) to remove SO_2 and selective catalytic reduction (SCR) unit to remove NO_x. In order to reduce the temperature, a water washing tower is added after a gas blower. The pretreated flue gas is then contacted with the MEA solvent in a counter-current pattern inside the packed absorber. The treated gas is then released from the top of the absorber along with some traces of MEA solvent which is recovered by a water washing tower. The CO_2-rich solvent is taken out from the bottom of the absorber and pumped (~2 bar) into a hot-cold exchanger to be preheated by the regenerated lean solvent from the stripper and delivered to the CO_2 stripper. CO_2 is released from the top of the stripper, and then the water in the captured CO_2 stream is condensed and recycled back to the stripper. The dry CO_2 is compressed and pumped to the pipeline for the transportation. The water balance is important in capture and regeneration process, thus part of the water from the CO_2 condenser is recycled back to the capture system.
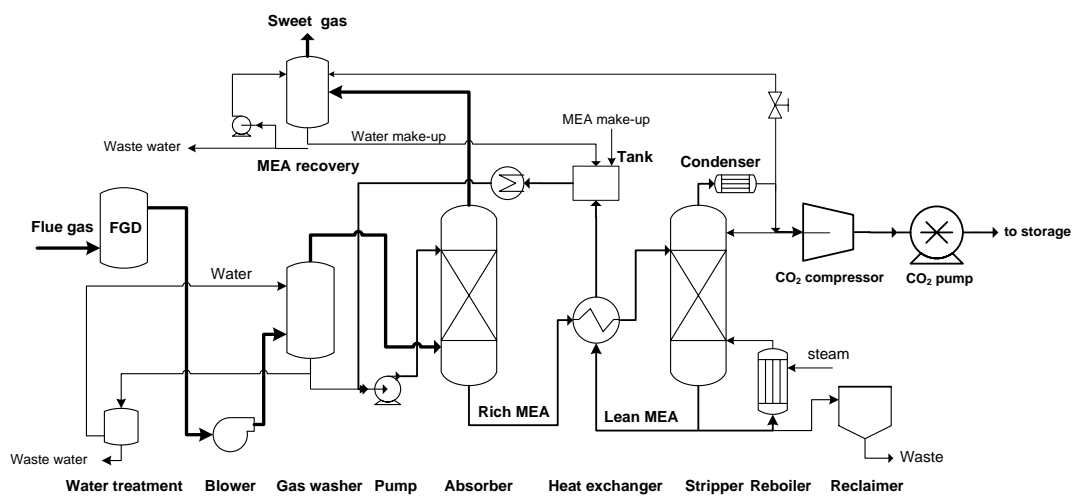
Fig.2(b) presents a two-stage gas separation membrane process for post-combustion carbon capture. In this study we consider a composite polyvinylamine (PVAm)/Poly(phenylene

oxide)(PPO) hollow fiber membrane as it has very high permeance and selectivity, which approach to industrialization (Sandru et al., 2010). Considering the low CO_2 concentration in the flue gas and the limitation of the membrane materials reported, since using one stage membrane process cannot reach the assigned capture ratio (90%) and CO_2 purity (95mol%), therefore a two-stage membrane system is preferred. The flue gas pre-treatment is the same as the MEA-based process in Fig. 2(a). The pretreated flue gas is compressed and fed into the 1st stage membrane unit at a higher pressure compared to the permeate side. A proportion of CO_2 can easily pass through the membranes, while the other gases (such as O_2, N_2) will be mostly retained due to their lower permeabilities. In order to improve the driving force of the gas transportation, vacuum pump is used in the permeate side. The retentate stream is subsequently sent into the expander to recover the power, which can reduce the net power consumption. A similar process is designed in the 2nd stage membrane unit. The process operating parameters such as pressure ratio, membrane area, and the capture load distribution between two stages are optimized based on the specific capture requirements.

Separating CO_2 from the mixed gas with low temperature liquefaction method, especially for high CO_2 concentration gases, is quite attractive. It has been proved that the cryogenic separation technology has advantages to separate CO_2 from higher CO_2 concentration gas mixture, such as an oxy-combustion power plant system (Pipitone and Bolland, 2009), but is not considered economically competitive due to its extremely high power consumption while treating low CO_2 concentration flue gases (~1.7 MJe/kg CO_2 for 30% fed gas, only with 35% capture ratio) (Berstad et al., 2012). Similar to the MEA-based systems, the specific capture power consumption deceases rapidly with the increase of CO_2 concentration of the feed gas. The gas from the 1st stage membrane can reach a relatively high CO_2 concentration, indicating that the combination of membrane system with cryogenic separation process may significantly reduce the capture energy consumption with great feasibility. Belaissaoui et al. also proved the advantage of low energy consumption for the hybrid membrane-cryogenic carbon capture process by process modeling and simulation (Belaissaoui et al., 2012).

Fig. 2(c) presents a hybrid membrane-cryogenic capture process. The membrane part is similar to the 1st stage in membrane process (Fig. 2 (b)). The cryogenic part is based on Joule-Thompson effect, to produce power by the adiabatic expansion of a high pressure stream. After sensitivity analysis of key parameters and also based on the industrial data and engineering heuristics, the suitable parameters are determined. Generally, the CO_2-rich gas from the membrane unit is compressed about 33bar in Compressor-2, and fed into a heat

exchanger (Ex-1) to achieve a cryogenic temperature being cooled down by the expander outlet gas, separating the liquid CO_2 in Flash-1 based on the vapor-liquid separation principle. In order to get the target purity of captured CO_2 to 95%, it is designed to go through two flash drums (Flash-1 and Flahs-2). The high purity CO_2 is compressed to liquid state and pumped to 110 bar for transport. The decarbonized gas is pre-heated to about 80°C by the heat produced in the inter-cooling of gas compressors before being fed into Expander-2. The cold stream from Expander-1 can also be used to pre-cool the gas from Compressor-1. The advantage of designed process is that it requires no external compression refrigeration cycles or refrigerants, such as propane, which makes the process more simple and low cost.

(a) Typical process of CO_2 capture with MEA-based solvent
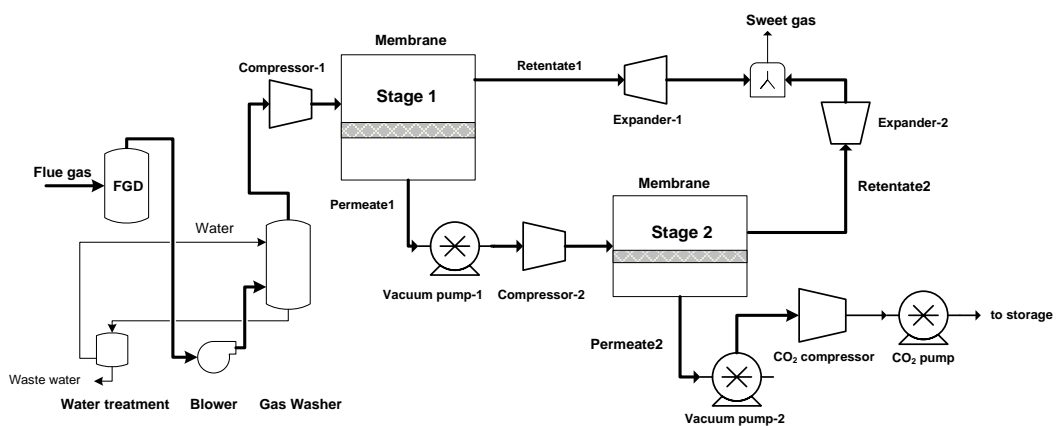
(b) Two-stage gas separation membrane capture process

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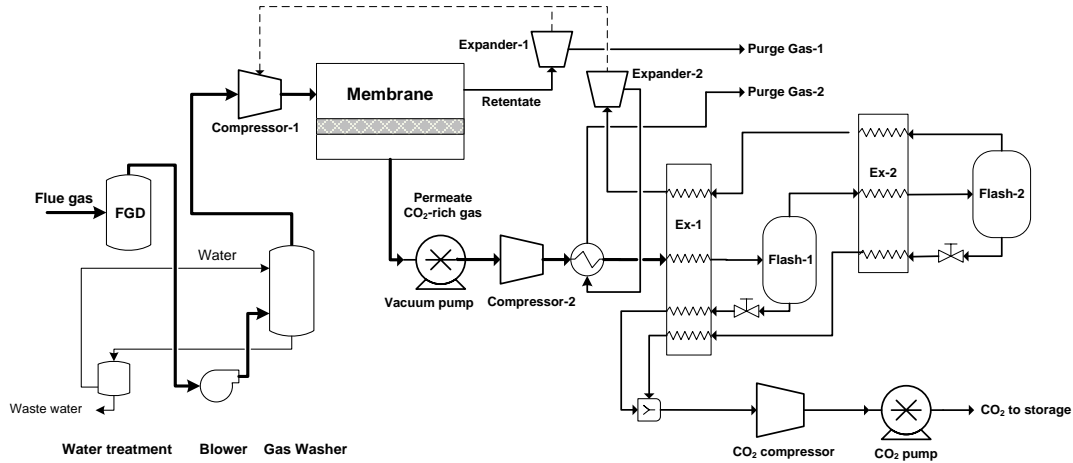
(c) Hybrid membrane-cryogenic capture process

Fig.2. Flow sheet diagrams of three capture processes

Regarding the energy consumption for carbon capture, two parts of energy consumption are involved in an MEA process, one is the steam extraction from the power plant IP/LP steam turbine which is used for stripping CO_2 from the rich solvent, i.e., thermal energy; the other part is the electric energy consumption for gas blower, solvent pump, CO_2 compression, as well as the other driving machines. Unlike the MEA process, in the gas separation membrane process and hybrid membrane-cryogenic process, mainly electric energy is needed to drive the compressors and vacuum pumps. For ease of comparison, the energy consumption in a MEA-based technology is also converted to be an equivalent electric energy consumption based on the electricity penalty caused by CO_2 capture and compression in a power plant system. In this work, we use 'MJe' represents the energy in electric energy form, while 'MJth' represents energy in thermal form.

3.3 Key parameters and assumptions

The main parameters for modeling these three scenarios are listed in Table 2. The final capture ratio and CO_2 purity in each scenario are assigned to be 90% and 95% (mol), respectively. The parameters are mainly from literature, industrial data and engineering heuristics (Anantharaman et al., 2010; Favre, 2011; Husebye et al., 2012; Hussain and Hagg, 2010; Merkel et al., 2010; Stern, 1968; Zhang et al., 2012a), and also based on the sensitivity analysis in this work.

LCA for MEA-based systems have been adequately studied (Koornneef et al., 2011; Koornneef et al., 2012; Korre et al., 2010; Pehnt and Henkel, 2009; Schreiber et al., 2009; Singh et al., 2011a; Singh et al., 2011b, 2012), which possesses relatively abundant and reliable data for LCA analysis, while gas separation membranes for CO_2 capture have relatively sparse data for LCA analysis in the published literature. In this work, the membrane is modeled based on the experimental data from literature (Sandru et al., 2010), and is assumed to be prepared by coating a PVAm selective layer on the outer surface of PPO hollow fibers. The thickness of PVAm selective layer is estimated to be 1.1 μm, and the diameters of porous support of (PPO) hollow fibers are estimated as 0.5/0.34 (outer/inner) mm. The wall thickness of the support is 0.08 mm. PVAm is produced with base hydrolysis of Poly (N-vinylformamide) (PNVF) process and PNVF is produced by the reaction of acetaldehyde and formamide (Gu et al., 2001; Witek et al., 2007). Thus, the chemicals used for membrane preparation can be estimated based on the total membrane area obtained from the process simulation. Loss of active compounds such as PVAm is also ignored to simplify the investigation. Data for power plant and capture infrastructure are based on Ecoinvent v2.2. dataset (EcoinventCentre, 2010) and Kleijn et al.'s work (Kleijn et al., 2011).

Table 2. Summary of the parameters

Unit / Process	Parameters
MEA-based system	Lean MEA solvent: MEA 28.3% (wt), 40°C, 1.113 bar
	Lean and rich MEA loading: 0.27 and 0.46 molCO_2 / mol MEA
	Murphree efficiency of absorber: 30%
	Absorber: stage number: 10
	Stripper: stage number: 15
	CO_2 condenser: 40°C, 1.8 bar
Membrane process	Composite PVAm/PPO hollow fiber membranes
	CO_2 permeance: 1 Nm^3/m^2·bar·hr
	N_2 permeance: 0.005 Nm^3/m^2·bar·hr
	O_2 permeance: 0.02 Nm^3/m^2·bar·hr;
	H_2O permeance: 2.25e-9 Nm^3/m^2·bar·hr
	CO_2/N_2 selectivity: 200
	Vacuum pressure: 0.2 bar
	Pressure of second membrane: 2 bar
Hybrid process	*Membrane section:*
	Membrane parameters are same as above
	Capture ratio: 95%
	CO_2 purity: 70% (mol)
	Vacuum pressure: 0.2 bar
	Cryogenic section:

	Pressure of fed gas compressor: 30~35 bar
	Pressure of throttle expansion valve: 3~4 bar
	Capture ratio: 90%
	CO_2 purity: 95% (mol)
	Two-stage flash and throttle expansion
CO_2 compression	Compressor: ~80 bar, pump: 110 bar
Isentropic efficiency	Compression: 80 %
	Gas blower: 80%
	Vacuum: 80%
	Pump: 80 %
	Expander: 80%
Life cycle assessment	10 impact categories:
	GWP, FEP, MEP, TAP, PMFP, POFP, ODP, HTP, TETP, FETP
Capture specifications	CO_2 Capture ratio: 90%; CO_2 purity: 95%
	CO_2 Captured: ~3.9 Million ton/y
Others	Cooling water: 15°C, 1.113 bar
	Steam: 130°C, 2.7bar
	Membrane life time: 5 years
	Lifetime of packing in the packing towers: 10 years
	Plant life time of power plant and absorption process: 40 years
	Operational time: 7500 hr/y

4. Results and discussion

4.1 Process simulation and analysis

4.1.1 MEA-based capture process

The simulation results of the capture process with MEA-based system are shown in Table 3. The energy consumption are mainly consists of five parts, i.e., sensible heat, reaction heat, stripping or vaporization heat, electric energy consumption for gas blower and solvent pumps. Among them, the heat demands to release CO_2 from the rich-solvent in the stripper accounts for the major energy consumption. Thus, the most notable feature for this process is that it consumes mainly the thermal energy instead of electric energy, and the steam is extracted from the steam turbines in the power plant which additionally requires retrofitting the existed power plant, and thus may also influence the operation of the steam turbine. The net electricity production with CCS is 559 MW, while the net electricity output without CCS is 754 MW (Anantharaman et al., 2010), hence, the electricity efficiency of the power plant reduces about 12% and the specific equivalent electric energy consumption in this scenario is estimated to be 1.35 MJe/kg CO_2. Comparing to the previous work (Anantharaman et al.,

2010), the relatively lower energy consumption in this work is owing to the lower specific separation requirement of a capture ratio 90% and a CO_2 purity of 95% (mol).

Table 3. Thermal and electric energy consumption for the studied MEA-based CO_2 capture system

Thermal (MJth/s)	
Reboiler heat duty	484.6
Stripper condenser cooling	188.5
Lean liquid cooling	234.0
Compressor cooling	78.3
Electric energy consumption (MJe/s)	
Gas blower	8.2
Pumps	10.0
CO_2 compression	45.8
Total	64.0

Based on the simulation results, the circulation of MEA solvent with 30% MEA aqueous is about 13 kton/hr and the total loading amount of MEA is about 5.8 kton. MEA loss is estimated as 1.5 kg MEA/ton CO_2 captured (Uyanga and Idem, 2007; Veltman et al., 2010), including the losses from MEA emission to air with the sweet flue gas, in waste water, oxidative degradation and heat stable salt formation. Cooling water is also needed to cool the flue gas, the compressed gases and the stripped CO_2. In this system, about 71 ton/hr waste water containing MEA and other chemicals is produced and causes the water pollution. Besides materials consumed in the capture process, the infrastructure for building the capture process is also important for LCA. Moreover, the packing used for the absorber and stripper are the Sulzer Mellapak 250X, and the total weight of packing is about 1525 ton with the life time of 10 years.

4.1.2 Gas separation membrane capture process

As describe in section 3.2, different from the amine absorption processes, the driving force of a gas separation membrane process is the partial pressure difference of the gas component across the membrane, and the energy consumptions which is mainly caused by the gas compressors and vacuum pumps. For a two-stage membrane process, the capture load distribution between the 1st and 2nd stage should be optimized. Some literature has reported different CO_2 concentration in the 1st stage permeate stream, example are: 50% (Merkel et al., 2010), 42~55% (Hussain and Hagg, 2010) and 61% (Zhao et al., 2010). Fig. 3 presents the energy consumption and membrane area in a two-stage membrane process and shows that

capture load distribution can significantly influence the specific energy consumption and the required membrane area. In Fig 3(a), the total energy consumption excludes the energy consumption used for CO_2 compression from 1 bar to 110 bar, while in Fig 3(b) such compression energy consumption includes. For a constant capture ratio of 90%, a higher CO_2 flow rate is needed at a lower CO_2 purity, resulting in a larger required membrane area. With the increase of the pressure ratio in the 1st stage, the CO_2 concentration in the 1st stage permeate increases and the 1st stage energy consumption increases as well, while correspondingly the energy consumption in the 2nd stage decreases. Thus, the synergistic effect is that the total energy consumption decreases at first and then increases as shown in Fig.3(a). Due to the typical inverse relationship between the energy consumption and the required membrane area, the total membrane area decreases continuously with the increase of 1st stage permeate CO_2 concentration, and stabilizes at higher CO_2 concentration (e.g., 65%) (Fig. 3(b)). After trading off between the specific energy consumption and the total required membrane area as shown in Fig. 3(b) and considering the possible effect of pressure ratio across the membrane (higher pressure ratio might destroy the thin membrane), 65% is chosen as the optimal CO_2 concentration out from the 1st stage, and the total required membrane area is calculated to be 17.27 million m^2.
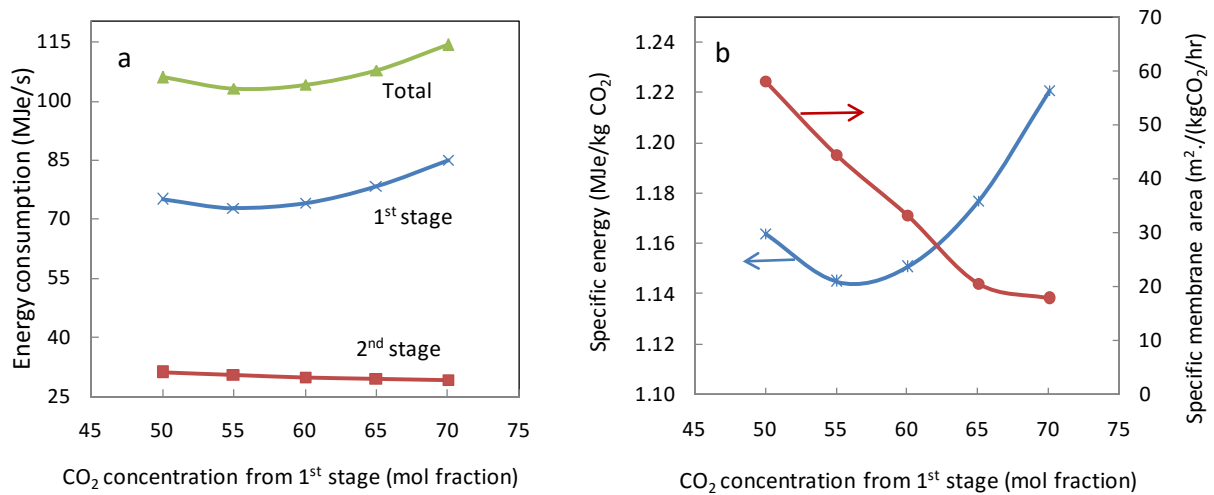

Fig. 3. Energy consumption and membrane area in a two-stage process

Table 4 shows the simulation results of a two-stage gas separation membrane process based on the simulation basis of Table 2. In this scenario, the total energy consumption is 166.4 MJe/s and the specific equivalent energy consumption is about 1.151 MJe/kg CO_2 which is over lower than that of a MEA-based system and one of the most important reasons

is we selected to use the membranes with almost highest CO_2 selectivity reported by present (Luis et al., 2012). The energy consumption for membrane is the requirement of high pressure ratio across the membrane, and the repeated recompression and suction of a large volume gas to achieve the separation requirement. It is noted that improvement in CO_2/N_2 selectivity would reduce the pressure ratio and hence the energy consumption but will need more membrane area. Thus, the trade-off between the energy consumption and the required membrane area is the main challenge for membrane process, not only in process design, but also for the membrane material development. Further, the energy consumption for CO_2 separation is 166.4 MJe/s, while only 62.5 MJe/s is needed for CO_2 compression and transport (compressing CO_2 to 110 bar). Thus, more than 62% of the total power consumption is in the CO_2 separation process.

Based on the method described in section 2.2, the total amount of PVAm demand for the coating on the PPO hollow fibers for two membrane units (two-stage process) is about 20.5 ton. With the membrane life time of 5 years and the requirement of single-time regeneration assuming 50% loss in membrane performance during the whole membrane's lifetime, the total amount of PVAm required over 40 years (expected lifetime of power plant) is 246.2 ton. In addition to the PVAm, the demand of PPO hollow fiber support is about 1230 ton for two membrane units (9843 ton over the 40 year lifetime).

Table 4. Simulation results of the two-stage membrane process for CO_2 capture

Parameters	Unit	Value
Membrane area		
Stage 1	million m^2	15.3
Stage 2	million m^2	2.0
Membrane area demand	million m^2	17.27
PVAm demand	ton	20.5
1st stage membrane		
Gas compressor-1	MJe/s	72.61
Vacuum pump-1	MJe/s	33.94
Expander-1	MJe/s	-32.54
Net energy consumption	MJe/s	74.01
2nd stage membrane		
Gas compressor-2	MJe/s	12.46
Vacuum pump-2	MJe/s	20.57
Expander-2	MJe/s	-3.1
Net energy consumption	MJe/s	29.93
CO_2 compression		

Net energy consumption	MJe/s	62.5
Total energy consumption	MJe/s	166.4
Specific energy consumption	MJe/kg CO_2	1.151

4.1.3 Hybrid membrane-cryogenic process

As described above, a hybrid membrane-cryogenic process has the potential to further reduce the capture energy consumption. Table 5 shows the simulation results of the hybrid process. Here, we take the CO_2 concentration from the membrane section as 70% after trading off the energy consumption in membrane section and cryogenic section. The specific energy consumption is lower slightly than that of the all-membrane process. The proportions of energy consumption in membrane, cryogenic and CO_2 compression sections are 52%, 29% and 19%, respectively. The low energy consumption proportion for CO_2 compression is due to the higher pressure of CO_2 product from cryogenic separation section (~4 bar), compared to ~1.8 bar in the MEA-based system, and 1.0 bar in the all-membrane process. This also confirms another possible advantage of energy saving with cryogenic method. Moreover, the required membrane area in the hybrid process is reduced to 1/3 of the membrane area in the two-stage membrane process as a higher gas pressure ratio and one stage membrane process is used. Thus, the total amount of PVAm coating on the PPO hollow fiber for the hybrid process is 6.7 ton, and the demand of PPO hollow fiber as support is about 403 ton, for a single cycle of membrane throughput without regeneration. Clearly, it can be concluded that the hybrid membrane-cryogenic process has relatively higher energy saving and low membrane area requirements.

Table 5. Simulation results of the hybrid membrane-cryogenic process for CO_2 capture

Parameters	Unit	Value
Membrane section		
Gas compressor-1	MJe/s	99.06
Vacuum pump	MJe/s	28.51
Expander-1	MJe/s	-42.71
Net energy consumption	MJe/s	84.86
Membrane area demand	million m^2	5.66
PVAm demand	Ton	6.7
Cryogenic section		
Gas compressor-2	MJe/s	56.45
Expander-2	MJe/s	-10.40
Net energy consumption	MJe/s	46.05
CO_2 compression		
Net energy consumption	MJe/s	30.57

Total energy consumption	MJe/s	161.5
Specific energy consumption	MJe/kg CO_2	1.11

4.2 Comparison and analysis

Table 6 gives an overall comparison of different cases with or without CO_2 capture and compression. Comparing the net electrical efficiencies, the hybrid system shows the highest advantage, than the all-membrane process and MEA-based system. Such results partly attributed to the very high CO_2/N_2 selectivity of the membrane used in this work.

Table 6. Summary of three scenarios and comparison

Parameters	Unit	Withoutcapture	With CO_2 capture and storage		
			MEA	Membrane	Hybrid
Gross electricity output	MWe	819.0	690.0	819.0	819.0
Auxiliary electricity consumption	MWe	65.0	131.0	231.5	226.5
Net electricity output	MWe	754.0	559.0	587.6	592.5
Efficiency	% LHV	45.5	33.7	35.5	35.8
CO_2 emitted	Kg/MWh	767.6	103.5	98.5	97.7
SEPC[*]	MJe/Kg CO_2	N/A	1.348	1.151	1.110
Waste water in capture	ton/hr	N/A	71.0	52.2	52.2

*: Specific equivalent power consumption (SEPC)

Fig. 4 illustrates the profiles of the equivalent electric energy consumption for CO_2 capture and compression of the three scenarios. In the MEA-based scenario, blowers and pumps accounts for a very small proportion of the total power consumption. The equivalent electric energy consumption for CO_2 separation with MEA-based process is the lowest, while the hybrid process is highest (membrane section + cryogenic section). However, due to a higher pressure of CO_2 product of 4 bar in the hybrid process compared to 1.02 bar in all-membrane process, the power for CO_2 compression/pump in all-membrane process is much higher than the hybrid membrane-cryogenic process, leading to the slightly reduced total energy consumption in the hybrid process. The relatively lower eenrgy consumption for CO_2 compression in the MEA-based process is owing to the moderate pressure of 1.8 bar for CO_2 coming out from stripping tower. Based on the analysis, it can be concluded that the hybrid membrane-cryogenic process shows a promising application for CO_2 capture from flue gases in post-combustion power plants due to the relatively lower energy consumption.
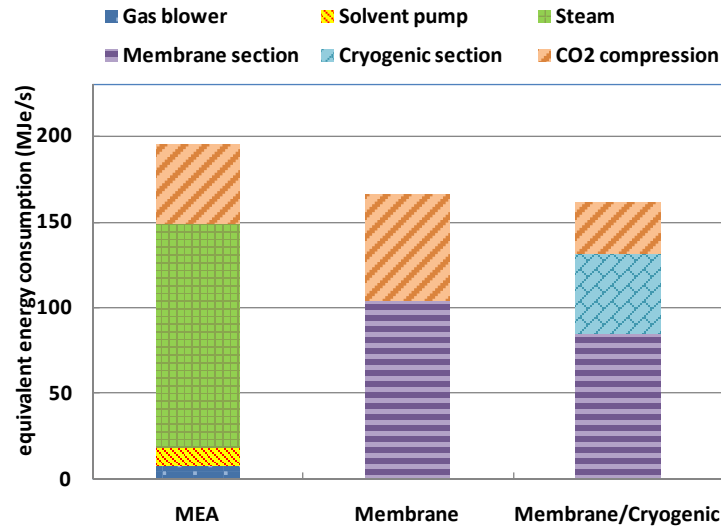
Fig. 4. Energy consumption profiles in three capture scenarios

4.3 Life cycle analysis

Fig. 5 shows the environmental impacts of the electricity generation with different capture systems relative to without CCS based on the system defined in Fig. 1. All the environmental impacts of the system without CCS are assumed as 100%. Fig. 5 confirms the significant reductions in GWP impact, but also shows the increase in environmental impacts in almost all other impact categories compared to the base case without CCS.
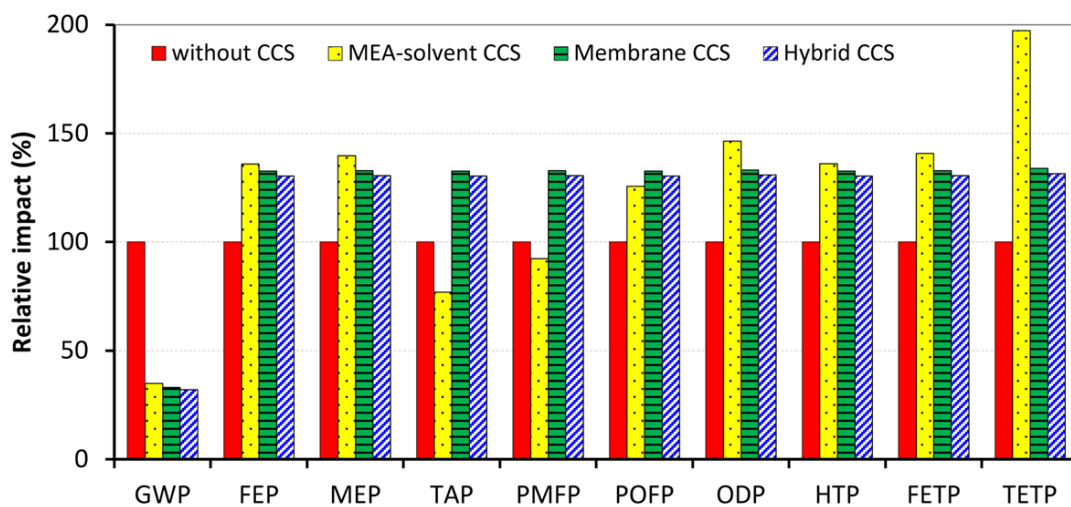
GWP (global warming): CO_2eq   FEP (freshwater eutrophication): P eq

MEP (marine eutrophication): N eq   TAP (terrestrial acidification): SO_2eq

PMFP (particulate matter formation): PM10 eq   POFP (photochemical oxidant formation): NMVOC

ODP (ozone depletion): CFC-11 eq   HTP (human toxicity): 1,4 DCB eq

FETP (freshwater ecotoxicity): 1,4 DCB eq   TETP (terrestrial ecotoxicity): 1,4 DCB eq

Fig. 5. Environmental impacts for the electricity generation using different capture systems relative to without CCS case

4.3.1 Global warming potential

For Global Warming Potential (GWP) impact, the 90% capture ratio results in a net reduction of 65%, 67% and 68% GWP for the MEA-based system, membrane system and hybrid membrane-cryogenic system respectively. This means, besides the rest of CO_2 in the decarbonized flue gas, greenhouse gas (GHG) emission caused in the infrastructure development of power plant, the coal mining and supplies, capture specific infrastructure and supplies etc. still accounts for a significant share of the total GHG produced. Hence, even as the absolute CO_2 capture ratio is fixed as 90%, the relative GWP impacts of the whole life cycle are still as high as 35% for the MEA-based system and the membrane system, and 32% for hybrid system with respect to the base case without CCS. The relative lower GWP impact of the hybrid system is mainly owing to the less additional power consumption as shown in Fig. 4.

In order to well understand the GHG emission contributions to the whole chain, five sub-chains (SubCh) are analyzed as discussed in section 2.2 and the results from different sub-chains are presented in Table 7. The total GHG produced refers to the summation fromSubCh-1 to SubCh-4, while the net GWP impact (net emission) is the summation of the GHG produced and SubCh-5.The results show that the primary infrastructure (SubCh-1) of power plant, capture unit, transport (pipeline) and CO_2 storage (well) account for relatively small contribution on GWP impact, about 0.09 to 0.13% of the total GHG produced in the whole chain. Further, the MEA-based CO_2 capture system has lower energy efficiency compared to both the hybrid and membrane based systems, therefore the MEA-based system needs more coal supplies (SubCh-2) and coal combustion (SubCh-4) per unit output. The GWP contribution from capture specific supplies chain (SubCh-3) is about 0.5 % of total GHG produced in MEA-base system, however, for membrane process and hybrid process, this contribution is much smaller (< 0.02%) due to the lower demands of chemical supplies compared to the MEA-based process. Owing to these relatively higher GHG emissions in the indirect sub-chains of coal and supplies (SubCh-2), capture specific infrastructure and supplies (SubCh-3) and direct emission sub-chain of coal combustion (SubCh-4), the MEA-based CCS system has higher net GHG emissions compared to hybrid and membrane based

CCS systems. The GWP from the coal supply chain (SubCh-2) is about 15% of the total GHG produced for all cases. The absolute value of GHG produced with each CCS scenario is 30% to 35% higher than the base scenario without CCS as more energy (i.e., combustion of more coal) is needed for CCS. Considering the contribution of GHG produced to the GWP in each sub-chain, the highest GWP is located in the power plant (SubCh-4), as expected, where it accounts for about 85% of the total GHG produced.

Table 7. Absolute impacts scores in g CO_2 eq for 1kWh of electricity generation from different scenarios

	SubCh-1	SubCh-2	SubCh-3	SubCh-4	SubCh-5	GHG produced	Net emission
W/O CCS	0.8	141.2	0.0	778.2	0.0	920.2	920.2
MEA	1.5	190.6	5.9	1050.7	-926.7	1248.8	322.1
Membrane	1.5	187.3	0.1	1032.3	-916.3	1221.3	305.0
Hybrid	1.5	184.1	0.1	1014.5	-905.0	1200.2	295.2

4.3.2 Other environmental impacts

The relative impacts of three toxicity categories - HTP, FETP and TETP varies from 130 to 197% with respect to the case without CCS. The main contributions to toxicity is generally associated with coal mining, direct emission form coal combustion, the infrastructure requirements and heavy metal emission associated with the material production (as well as the emission and degradation of MEA in the capture process for TETP). The relative impacts of TETP (terrestrial ecotoxicity) are 197%, 134% and 132% for MEA-based, membrane and hybrid CCS scenarios, respectively. The high TETP impact of MEA-based CCS is mainly caused by the MEA emission and degradation which contributes > 28% to TETP. For the membrane and hybrid processes, the contributions of the coal mining and supply, and coal combustion to TETP are about 32% and 66%, respectively. The infrastructure requirement contributes only 1% to 2% to TETP, which is much lower than the coal supply chain and coal combustion. The analysis finds that coal supply chain in different scenarios makes major contributions to FETP (freshwater ecotoxicity) and human toxicity of over 85%, and is mainly caused by the landfill disposal of spoils from the coal mining. Thus, cleaner technologies should be developed in a coal supply chain to prevent the toxicity impacts on human and water organisms, as well as adverse effects on the local ecology.

Ozone depletion (ODP) indicates the potential of chemicals, containing chlorine and/or bromine to destroy the stratospheric ozone. Results in Fig. 5 show the increases of 131

to147% in ODP for three CCS scenarios, and are mainly associated with the coal supply chain (70 to 80%) of the total impact.

The MEA-based solvent shows an overall reduction of 23% in TAP (terrestrial acidification) and 8% in PMFP (particulate matter formation) due to co-capture of SO_2, NO_x and particulate matter with solvent absorption method. All the three CCS systems show significant increase in FEP (freshwater eutrophication) and MEP (marine eutrophication), i.e., an increase of about 30% to 35% in FEP and 30% to 40% in MEP. FEP is caused by the emissions of phosphorus and phosphate to water and is again associated with the disposal of coal mining spoils. MEP (Marine eutrophication) is mainly caused by the emissions of the nitrogenous compounds (NO_x, NH_3, organic bound nitrogen, etc.). The coal supply chain is the main contributor to MEP for the cases with or without CCS. The second major contribution is the direct emissions from the power plant (coal combustion) accounting for about 45% of the total MEP impact.

Except global warming potential, most of the environmental impacts increase with CCS compared to the base without CCS, irrespective of the capture technology used. It is found that the coal supply chain and coal combustion at power plant contribute significantly to the most of the impacts, and the additional coal consumption for CCS inevitable increases the impact. Thus, decrease in the energy consumption of the CO_2 capture process will provide eventual solution to reduce e the environmental stresses from CCS. Certainly, using much lower volatile and more stable solvent or non-solvent capture technology can reduce the pollution caused by the emission of the chemicals and waste by-products produced in the capture process.

5. Conclusion

Assessing an emerging CCS technology inherently faces many challenges and uncertainties because of the lack of industrial data and thorough understanding of the process. This study demonstrates the importance of technological assessment based on the energy consumption and environmental impacts of CCS with life cycle perspective.

The results show that the MEA-based capture system has the challenges of solvent degradation and emissions, the need of retrofitting the existing power plants, as well as the higher energy consumption; by using higher performance membrane, the gas separation membrane technology has the significant advantages of lower energy consumption, reduced

terrestrial ecotoxicity as compared to the MEA-based system. The hybrid membrane-cryogenic process demands minimum energy consumption among three scenarios, and moreover, produces less environmental impacts than the MEA-based system, which means good feasibilities for industrialization in the near future.

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