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Investigation of a Hybrid System for Post-Combustion Capture

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Abstract

This paper investigates an approach for post-combustion CO₂ capture from the pulverized coal power plant “Kleine Kopje” in North-Rheine Westphalia (NRK-NRW). The system combines a single polymer membrane and refrigeration distillation to achieve optimal capture performance. Membrane separation was chosen as an alternative to amine-based separation due to its once-through process, and the refrigeration system was introduced to eliminate the need of multi-stage membranes in order to achieve high separation degree as well as to optimize the end-product purity. Process design and simulation was carried out using Aspen Plus® to determine the energetic and economic performance of the system. The energetic performance of the hybrid system at a feed pressure of 1.5 bar showed that 337.5 kWh/t_{separated} CO₂ was required to separate 90% of CO₂ and capture processes doubled the electricity price from 33.7 €/MWh to 65.5 €/MWh due to high investments for the membrane. Amine-based absorption exhibited slightly better energetic performance at a separation degree of 90%, while the hybrid system was more energy-efficient at lower separation degrees.

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1. Introduction

CCS is expected to be the leading technology for mitigating large-scale carbon emissions. An MIT study on coal [1] concludes that CO₂ capture and sequestration (CCS) is a potential technology for significantly reducing CO₂ emissions while also allowing coal to meet the ever-increasing global energy demand. However, several issues such as substantial investment, cost and public acceptance of the technology have limited its implementation. It will be virtually impossible to halve CO₂ emission by 2050 without deploying CCS on a full commercial scale [2, 3]. Furthermore, although renewables are assumed to be growing rapidly, CCS technology can still potentially cut a large portion of GHGs emissions in the power sector.

CO₂ capture is divided into three main categories based on its processing route: oxyfuel, pre-combustion and post-combustion capture. Here we will concentrate on post-combustion capture. Post-combustion systems separate CO₂ from the flue gas after combustion in power plants. The main advantage of this method is that the power plants can be retrofitted with CO₂ capture units. In addition, the same method has been used commercially in natural gas purification. Post-combustion systems can be realized using several technology options, such as absorption with solvents, adsorption, cryogenic separation and membranes.

The most common technology for post-combustion capture is amine-based absorption. This system normally uses a liquid solvent to capture CO₂ (typically 3–15% by volume) from a flue gas stream, which mainly comprises N₂, CO₂, H₂O and O₂. For a modern pulverized-coal combustion or a natural gas combined-cycle power plant, absorption systems typically employ an organic solvent such as monoethanolamine (MEA), one of the amine compounds [4]. The obstacles of scaling up MEA absorption in power plants are the high-energy parasitic load and solvent degradation. Systems integration with power plants is also the subject of research, including heat and waste heat recovery.

Selective permeable properties of membranes can be used to separate particular components of a gas stream. They work like a filter, allowing only (or mostly) CO₂ to pass through the material. Although membrane separation finds many current commercial applications in industry (some of a large scale, like CO₂ separation from natural gas), they have not yet been installed in carbon capture systems, which represent a large-scale and demanding scenario in terms of reliability and low cost requirements [5]. In the post-combustion capture process, the use of optimized polymer membranes was found to be a competitive technology for CO₂/N₂ separation in comparison with conventional MEA absorption [6-8].

The energy required for membranes is mainly determined by the desired separation degree (% CO₂ captured from flue gas) and the CO₂ purity. In the membrane separation process, compressors and/or vacuum pumps create the driving force which enables CO₂ in the flue gas to diffuse from the feed side to the permeate side. The use of optimized polymer membranes [9, 10] for the post-combustion route was found to be a competitive technology in comparison with the popular MEA absorption [6, 7]. The integration and performance of polymer membrane separation for post-combustion capture is being studied intensively [7, 11-15].

Membrane separation is regarded as one of the contenders to the current state-of-the-art amine-based absorption for post-combustion CO₂ capture. The simplicity of membrane technology is suitable for post-combustion where power plants must be retrofitted. The installation of membrane modules is arguably easier than installing absorption-desorption columns and it is relatively straightforward. Moreover, CO₂/N₂-selective membranes have recently been developed for CCS [6, 15]. Nevertheless, capturing CO₂ still poses serious R&D challenges due to the scale of CO₂ emissions by power stations. Single membrane separation cannot achieve a satisfactory degree of CO₂ separation and purity, while multi-stage configuration has shown promising results [8].

Refrigeration technology has been utilized for over 70 years and is still used in modern technology to separate air, producing liquid nitrogen, oxygen and argon in the process. With regard to carbon capture, refrigeration separation technology can be implemented in the oxyfuel and pre-combustion routes to obtain pure oxygen. A similar approach can be applied to post-combustion capture by cooling the flue gas and separating the CO₂ by means of the boiling point difference. The process of cooling and separating a gas mixture by inducing a phase change is termed refrigeration distillation [16].

Several hybrid concepts have been developed including the combination of refrigeration and membrane [17, 18]. Currently, the US Department of Energy and American Air Liquide [19] are attempting to use a combination of hollow fiber membranes and refrigeration units to achieve the target of 90% CO₂ capture without increasing the cost

of electricity by more than 35%. Large membrane areas could cost much more than expected, and the land area required for a CCS plant itself may also be excessive. Therefore combining membranes with another separation technology in a hybrid system may be preferable for capturing CO₂. Furthermore, the possibility of reducing energy requirements is also an attractive feature of hybrid systems.

In this paper, the hybrid process of combining membranes and refrigeration separation to achieve efficient post-combustion carbon capture is investigated in a simulation study using the software Aspen Plus 7.3. The hybrid process combines CO₂ pre-concentration with a membrane unit as a first step and CO₂ refrigeration condensation as a second step. The specific energy consumption is calculated for different degrees of CO₂ separation. The hybrid system, cascaded membrane system and MEA absorption are compared. The hybrid system is also economically analyzed.

2. Process Simulation

2.1. Reference Power Plant

In the present work, a reference power plant referred to as the Reference Power Plant Nordrhein-Westfalen (RW-NRW) [20] was chosen for energetic analyses. Multi-stage polymer membranes were installed after the SCR-DeNO_x, dust removal (E-filter) and desulphurization (FGD) processes and before emission through the cooling tower, analogous to amine stripping processes [21].

Aspen Plus 7.3 software was used for the simulation. There are different thermodynamic models for the energy balance calculation available in Aspen; for the case described here, the Peng-Robinson equation of state was chosen.

A hard coal named Klein Kopje was used to simulate the flow rate and the components of the flue gas for the multi-stage membrane calculation. The element analysis data of Klein Kopje coal were: C 65.5%, H 3.5 %, O 7.4%, N 1.5%, S 0.6%, ash 14.2%, moisture 7.3%; and the heat value was 25 MJ/kg. The coefficient of air excess (air-to-fuel ratio) was assumed to be 1.15. The basic data of RW-NRW and the simulation results of the flue gas are listed in Table 1. The residue of the pollutant in the flue gas consisted of approximately 50 vppm SO₂ and approximately 200 ppm NO₂.

Table 1. RW-NRW power plant basic data [20] and simulation results of the flue gas conditions after removal of the pollutants using Klein Kopje hard coal

Gross output	600 MW
Net output	555 MW
Net efficiency	45.9 %
Steam parameters	285 bar/600°C/620°C
Operation time	6000 h/year
Fuel input	1 Mt/year
Investment cost	€ 517.1 million
O&M cost	€ 7.8 million/year
Fuel cost	€ 41/tonne
Electricity price	3.37 ¢/kWh
Flue gas condition after NO _x , particulate and SO ₂ removal	
Pressure	1.05 bar
Temperature	50°C
Flow rate	1.6 million Nm ³ /h

Main components of the flue gas	
CO ₂	13.5 mol%
N ₂	70.1 mol%
O ₂	3.7 mol%
H ₂ O	11.9 mol%
Ar	0.8 mol%

2.2. CO₂-Selective Membrane

Rubbery polymers based on PEO or PEG emerged as promising candidates [22] using the material selection guidelines proposed by Merkel et al. [15]. The feasibility of producing block copolymers such as Polyactive[®] [23-25] on a large scale is an important prerequisite for the subsequent fabrication of gas separation membranes and modules. Crosslinked PEO systems are promising for use in post-combustion carbon capture if the material design strategy can be extended to the fabrication of thin-film composite or asymmetric membranes. Based on data in the literature and from the Membrain [26] and MetPore projects [27], the membrane properties in terms of temperature variation are listed in Table 2. Certain data estimations were made.

Table 2. Membrane properties of Polyactive[®] membrane, referring to data from the literature [14, 22-27]

Polyactive [®] membrane			
Temperature	50°C	30°C	25°C
CO ₂	5	4.3	3
H ₂ O	15	43.3	53
CO ₂ /N ₂	25	36	50
O ₂ /N ₂	2.8	2.8	2.8
Ar/N ₂	2.8	2.8	2.8
SO ₂ /N ₂	270	270	270
NO/N ₂	5	5	5

2.3. Hybrid System

A process flow diagram of the hybrid system is shown in Figure 1. The concept of the hybrid process began after the common pollutants were removed. The flue gas temperature was then reduced to 25°C, in which some mole fraction of water was condensed. Subsequently, 67% of the water content in the flue gas was removed using a sulfonated poly(ether ether ketone) (SPEEK) membrane [28]. Water was not completely removed from the flue gas because a study by Low [12] concluded that the presence of water from a process standpoint has a slightly positive sweep effect on polyactive[®] membranes due to the co-permeation of water vapor, which increases the CO₂ separation degree at a fixed membrane area.

The membrane was driven by a vacuum pump. After membrane separation, the permeate flue gas stream was ready to be purified in the refrigeration section. In order to liquefy the flue gas stream, CO₂ partial pressure must be above its critical point (see CO₂ phase diagram [29] in Figure 2). Therefore, the permeate stream was compressed to a pressure of 15 bar using four-stage compressors with intercoolers to maintain the temperature at 25°C. Note that the heat exchanged in the intercoolers can be integrated with the retentate stream to heat up the retentate stream prior to the expansion process. In the interstage cooling, the pressure drop was assumed to be 0.03 bar. Furthermore, condensed water was removed from the stream in the compressor suction scrubber to avoid corrosion and other damaging effects in the compressor, represented by the flash block in Aspen.

Water proved to be a beneficial component in terms of improving membrane separation performance. However, it can lead to complications in the cold processing unit, where no water should be allowed to enter because the presence of ice is impermissible. To overcome this problem, a molecular sieve unit based on the adsorption method

was used before cooling the permeate stream. This approach is similar to liquefied natural gas (LNG) processing where extremely low concentrations of water (0.1 ppm) are essential. For water removal, the zeolite molecular sieve 3A was used because it only adsorbs H₂O and does not adsorb CO₂ in the process [16]. Zeolite 3A can remove up to 99.99% water from the inlet stream [30].

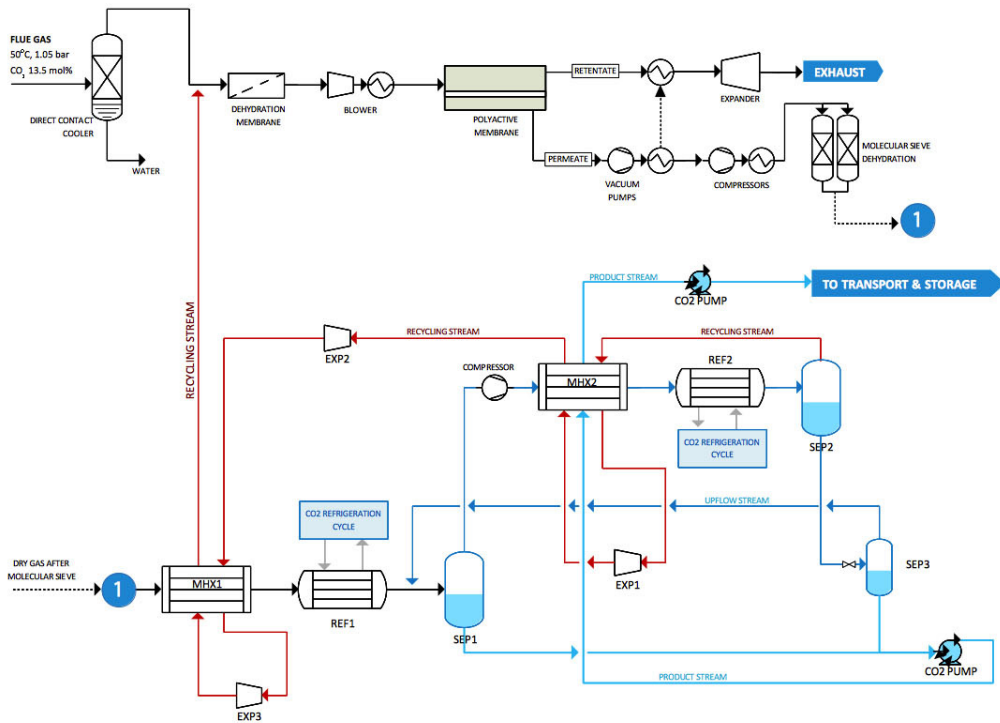


Figure 1. Process flow sheet diagram of the hybrid post-combustion system

After gas dehydration, the permeate stream contained a very small fraction of water allowing the cooling process to be carried out. Plate-fin and coil-wound heat exchangers are the most common for this application, and are well known for their ability to handle small temperature differences and withstand pressures up to 110 bar [31, 32]. In this case, the plate-fin type was chosen [30]. In the flow sheet, MHX1 and MHX2 are the two multiple heat exchangers.

Block MHX2 cooled the permeate stream to -33°C before it subsequently flowed to EXP2 for further expansion. In this process, the cold sources were the pressurized liquid CO₂ (product stream) and recycle stream from SEP2 block. After the expansion process in EXP2 from 22 bar to 6 bar, the recycle stream entered block MHX1 as a cold source. The outlet of the cool stream was again expanded by EXP3 and re-entered MHX1, where the stream pressure decreased to 1.5 bar. Subsequently, the stream was recycled back to membrane separation unit.

In order to achieve a high CO₂ separation degree, it is essential to cool the permeate stream as much as possible, while remaining above the CO₂ triple point (-56.6°C and 5.11 bar). In this case, an additional cold source was necessary to lower the product stream temperature even further to -52°C. After MHX1 and MHX2, external CO₂ refrigeration cycle blocks coupled with heat exchangers REF1 and REF2 (see Figure 1) were introduced to achieve the desired permeate temperature of -52°C. This created the condition where part of the CO₂ was liquefied and

subsequently ready for separation.

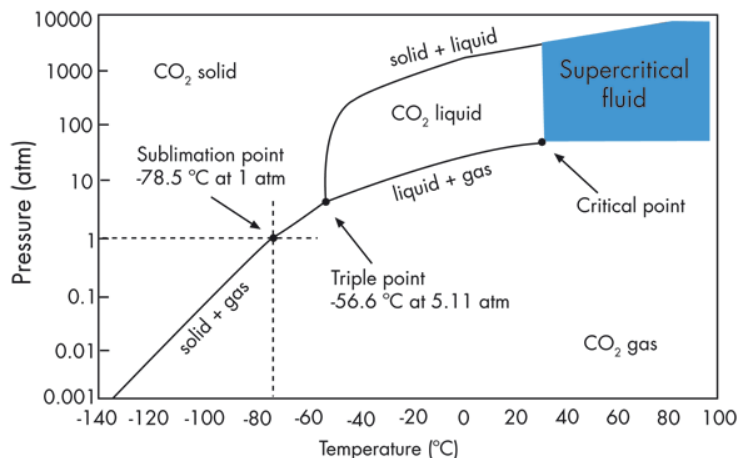


Figure 2. CO₂ phase diagram [29]

During the process, some of the CO₂ in the permeate stream experienced a phase change to liquid allowing the liquefied CO₂ to be separated in unit operation SEP1. The remaining gas stream from SEP1 was not directly recycled to the membrane feed. Rather, it was re-compressed to 55 bar and subsequently cooled in the MHX2 block and the second external CO₂ refrigeration cycle REF2. The stream temperature was -52 °C. Again, this additional stage separated liquid CO₂ from the main stream in unit operation SEP2. After separation was complete, liquid CO₂ streams from SEP1 and SEP3 were then mixed and pumped to 110 bar. Since the cold liquid CO₂ was used as a cold source for the heat exchange process, the pressure was initially increased to 60 bar by PUMP1 to avoid excessive pressure in the plate-fin heat exchanger. Finally, after passing through MHX2, the pressure of the product stream was increased by PUMP2 to 110 bar for CO₂ transport.

2.4. Energy Consumption

In the membrane separation part, there were two major power consumers: blower or compressor (depending on the pressure ratio) at the feed side and vacuum pumps at the permeate side. Energy was recovered via an expander of the retentate stream (refer to Figure 1). The main energy consumers in the refrigeration section were the multi-stage compressors. Additionally, a small fraction of energy was needed for liquid CO₂ pumps. The energy consumed in each separation stage is shown in Table 2. For the compression, CO₂ pumps consumed a very small fraction of energy because the CO₂ was already in the liquid phase after the refrigeration separation unit. Therefore, the compression energy requirement of the hybrid system is much lower than that of the multi-stage membrane system and MEA absorption.

Figures 3-5 show the energy performance of all separation degree targets as percentage shares. In this separation degree mode, the refrigeration unit requires slightly more energy than the membrane separation unit since it consists of more compressors and needs relatively high pressures to produce a CO₂ purity of 98%. From the pie charts, it is obvious that vacuum pumps and compressors account for the biggest share of the overall energy consumption. The dominant factors include high separation degree demand, large gas flow rate in the membrane separation, and high operating pressure in the refrigeration section. Furthermore, the power required for CO₂ refrigeration also comes from the compression process. Minimizing the energy consumption in this part is tricky since it depends on the flow rate of the permeate stream, which is dictated by the separation degree target.

A CO₂ separation degree of 90% means that more impurities (in this case, N₂) are passed through the membrane. This means that more compression energy is consumed by the refrigeration unit and higher pressure is needed to

purify the stream. Lower compression pressure is theoretically possible, but it will be less effective for purifying CO₂, consequently increasing the flow rate of the recycle stream and energy consumption of the membrane unit. For CO₂ separation degrees of 70% and 50%, the energy consumption is similar to that for 90%, although the consumption of the refrigeration unit is slightly higher than that of the membrane unit for a separation degree of around 57-59%. Due to the lower separation degree target, the energy consumption of vacuum pumps and compressors is reduced significantly.

Table 2. Energy consumption of the hybrid system for CO₂ separation degrees of 50%, 70% and 90%

	Unit	90% separation degree	70% separation degree	50% separation degree
Membrane	Blower	20535	20535	20535
	Expander retentate	-13616	-13926	-14231
	Vacuum pump	45424	26533	17094
	Area [km ²]	1.92	0.758	0.379
Refrigeration	Gas dehydration sieve	2776	833	529
	Multi-stage compressors	44692	25310	15981
	Expander 1	-3877	-1399	-759
	Expander 2	-5479	-1991	-1218
	Expander 3	-6093	-2345	-756
	Second stage compressor	13265	5015	2406
	Refrigeration cycle 1	16734	23058	18893
	Refrigeration cycle 2	11775	1625	0
Compression	CO ₂ pump	1270	954	684
	Intercooler	184	122	74
Total energy requirement [kW]		127591	84326	59232
CO ₂ captured [kmol/hr]		8592	6682	4773
Specific energy requirement [kWh/tCO ₂]		337.5	286.8	282.0
Efficiency penalty [%]		10.6	7.0	4.9

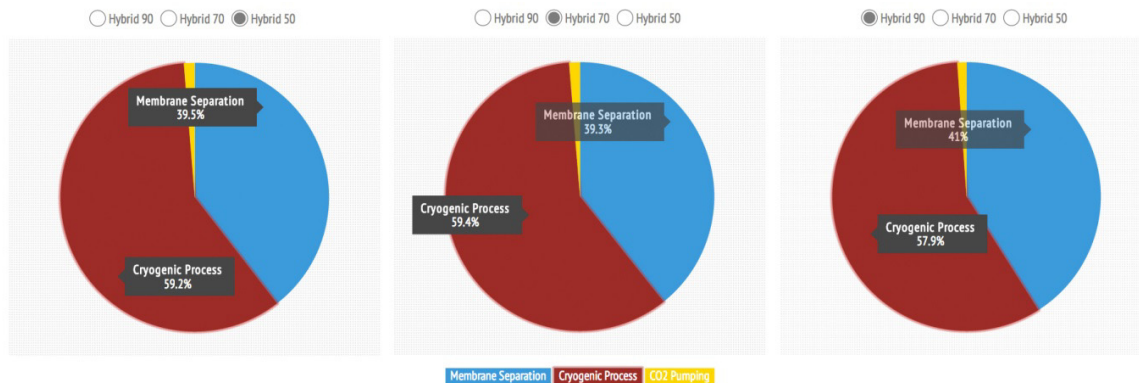


Figure 3. Energy consumption share of the hybrid system

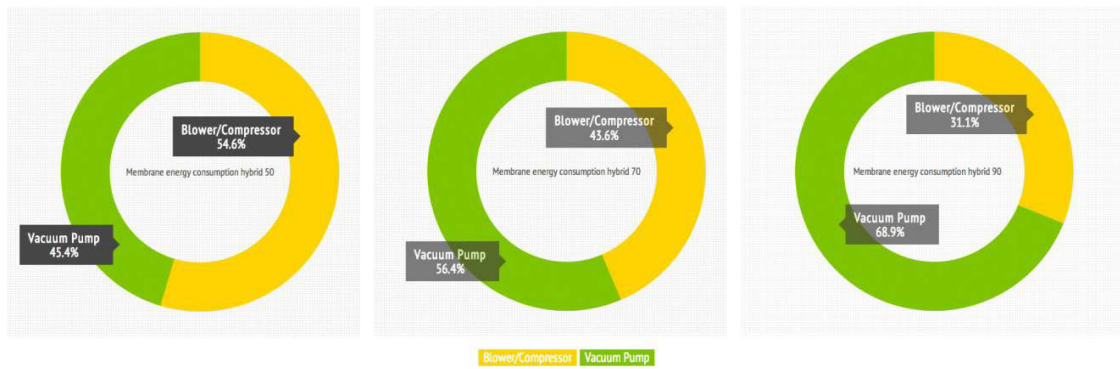


Figure 4. Energy consumption share of the membrane separation unit

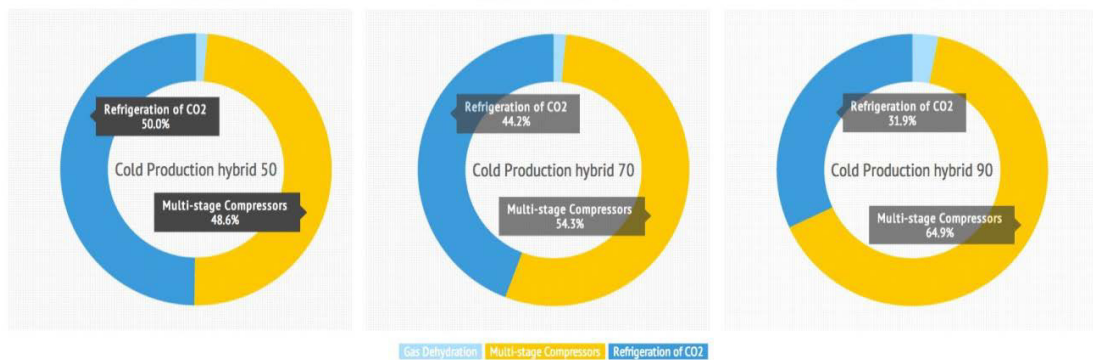


Figure 5. Energy consumption share of the refrigeration unit

As the current state-of-the-art CCS technology, it is interesting to see the comparison between the MEA absorption and the hybrid system. Although full-scale MEA absorption for power plants does not yet exist, several publications have reported the performance of the system [33-35]. From the energy consumption standpoint, the amine-based absorption process with monoethanolamine (MEA) as the most common solvent is normally taken as the current benchmark of the CO₂ capture energetic performance. Several publications have compared the MEA process with the membrane separation system [11, 36, 37].

A MEA absorption process needs mainly thermal energy for the regeneration process. This thermal energy is taken from the IP/LP steam turbine. Therefore, the energy requirement of MEA adsorption is normally expressed in thermal energy per tonne CO₂ captured. For the hybrid system, most of the energy required is for the vacuum pumps which create a membrane driving force and for the compressors which produce a cold temperature. The hybrid system, cascaded membrane system (Cascaded A) [8] and MEA absorption are compared in Figure 6.

At low separation degrees, the hybrid system had a similar energetic performance to the cascaded membrane system and a better performance than MEA absorption. The hybrid system had efficiency penalties of 7%-pts. and 4.9%-pts. for CO₂ separation degrees of 70% and 50 %, respectively. One advantage of the hybrid system is CO₂ purity, which reached 98.3 mol% in three cases. For the hybrid system with a separation degree of 90%, the efficiency penalty was 10.6%-pts, approaching the 10.3%-pts. of MEA absorption but much better than the cascaded membrane system at 11.4%-pts.

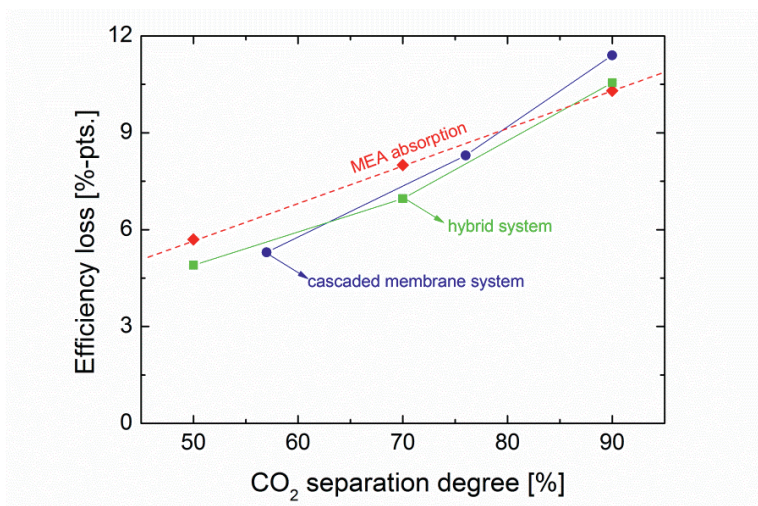


Figure 6. Comparison of the hybrid system, cascaded membrane system and MEA absorption

3. Cost Estimation and Economic Analysis

3.1. Cost Estimation

This section discusses the economic analysis based on the Aspen thermodynamic simulation. It scrutinizes the share of equipment cost, which affects the levelized cost of electricity and the cost of CO₂ captured and avoided, which is the common economic term in the CCS field. The calculation method is based on a number of references [38-40], most notably on the work by Zhao [41] and Zhai [42]. The cost estimation is based on present conditions and neglects future technology improvement. The equipment cost is referred to as the process equipment cost (PEC) and is calculated according to the assumptions in Table 3. The purchase cost for all separation degree targets (hybrid 90, hybrid 70 and hybrid 50) are presented in Table 4. Table 5 summarizes the results of levelized investment and operation and maintenance costs.

Table 3. Calculation method and assumptions of the economic analysis

Aspen ID	Description	Unit	Value
KM	Polyactive® membrane price	(€/m ²)	50
KDM	Dehydration membrane price	(€/m ²)	30
KMHX	Plate-fin heat exchanger cost, installed	(€/m ²)	360
KSKID	Membrane skid	-	250,000
KBLOW	Blower cost, installed	(€/hp)	500
KEXP	Expander cost	(€/kW)	300
KVAC	Vacuum pump cost, installed	(€/hp)	1000
KHX	Heat exchanger cost, installed	(€/m ²)	300
F	Civil, piping, electrical cost factor, etc.	-	1.8
CFUEL	Coal cost	\$/t	41

FUELINPUT	Fuel input	tonne/year	1000000
IR	Interest rate	%	7.50%
POL	Plant operating life	years	35
MOL	Membrane operating life	years	5
FLH	Annual operating hours	hour/year	6000
EFFIN	Initial efficiency	%	45.90%
COE	Cost of electricity	€/kWh	0.0337
REFINV	Total annual cost of the ref. power plant	€/year	112,299,858

Table 4. Total capital requirement of the hybrid system

Aspen ID	Description	References/Equations	Separation degree target		
			90%	70%	50%
CTOW	Cooling tower	[40, 43]	€ 96,852	€ 96,852	€ 96,852
CMEM	Polyactive membrane	MEMAREA*KM	€ 95,781,785	€ 37,923,946	€ 18,933,204
CSKID1	Membrane skid polyactive	(MEMAREA/2000) ^{0.7} *KSKID	€ 30,537,822	€ 15,965,388	€ 9,817,409
CDM	Dehydration membrane	ADM*KDM	€ 292,607	€ 292,607	€ 292,607
CSKID2	Membrane skid dehydration	(ADM/2000) ^{0.7} *KSKID	€ 757,937	€ 757,937	€ 757,937
CBLOW	Blower	WBLOW*KBLOW (hp)	€ 13,769,109	€ 13,769,109	€ 13,769,109
CCOM	Compressors	WCOM*KBLOW (hp)	€ 38,861,053	€ 20,321,390	€ 12,328,484
CEXP	Expander	WEXP*KEXP*F	€ 15,695,312	€ 10,730,041	€ 9,160,510
CVAC	Vacuum pumps	WVAC*KVAC	€ 60,915,117	€ 35,582,049	€ 22,923,560
CHX	Heat exchangers (S&T)	AHX*KHX	€ 21,120,187	€ 20,023,856	€ 10,596,850
CADS	Zeolite 3A adsorption*	[43, 44]	€ 691,796	€ 201,894	€ 124,172
CMHX	Heat exchangers (plate-fin)	AMHX*KMHX	€ 23,775,462	€ 22,460,792	€ 9,192,309
CVES	Separation vessels	[40, 43]	€ 344,443	€ 201,833	€ 125,553
CPUMP	Pumps	[40, 43]	€ 251,250	€ 170,202	€ 110,766
PEC	Process equipment cost (PEC)		€ 302,890,730	€ 178,497,895	€ 108,229,322
CGF	General facilities cost	10% of PEC	€ 30,289,073	€ 17,849,790	€ 10,822,932
CENG	Engineering and home office fees	7% of PEC	€ 21,202,351	€ 12,494,853	€ 7,576,053
CCONTG	Project contingency cost	15% of PEC	€ 45,433,610	€ 26,774,684	€ 16,234,398
TPC	Total plant cost (TPC)		€ 399,815,764	€ 235,617,222	€ 142,862,705
CROYALTI	Royalty fees	0.5% of PEC	€ 1,514,454	€ 892,489	€ 541,147
INVCAP	Inventory (start-up) cost	0.5% of TPC	€ 1,999,079	€ 1,178,086	€ 714,314
CTINV	Total capital requirement (TCR)		€ 403,329,296	€ 237,687,798	€ 144,118,165

*Zeolites 3A price is taken from www.industrialfilterstore.com

Table 5. Levelized investment and O&M cost of the hybrid system

Aspen ID	Description	Equation	Separation degree target		
			90%	70%	50%
CRF	Capital recovery factor		0.081	0.081	0.081
CRFmem	Capital recovery factor* _{membrane}		0.247	0.247	0.247
CLEV	Levelized investment cost	CRF*(CTINV-CMEM-CDM)	€ 25,036,025	€ 16,253,498	€ 10,176,593
CLEVM	Levelized investment cost _{membrane}	CRFmem*(CMEM+CDM)	€ 23,746,200	€ 9,445,783	€ 4,751,942
FOM	Fixed O&M cost				
MC	Maintenance cost	1.5% of (TPC-MEMBRANES)	€ 4,086,684	€ 2,710,160	€ 1,695,923
MCMEM	Maintenance cost membrane	1% of CMEM+CDM+CSKID1+CSKID2	€ 1,273,701	€ 549,399	€ 298,012
TLC	Total O&M labor cost	30% of MC	€ 1,226,005	€ 813,048	€ 508,777
OMADM	Admin and support cost	20% of TLC	€ 245,201	€ 162,610	€ 101,755
OMSUP	O&M supplies	5% of TLC	€ 61,300	€ 40,652	€ 25,439
TMC	Total maintenance cost	MC+MCMEM+TLC+OMADM+OMSUP	€ 6,892,892	€ 4,275,869	€ 2,629,906
VOM	Variable O&M cost				
ENERCOST	Energy cost	Total energy requirement*COE*FLH	€ 25,798,820	€ 17,050,618	€ 11,976,626

There are two approaches for integrating the CCS cost structure with that of the reference power plant: first using constant gross electrical output and second using constant net electrical output. In the latter case, net output is maintained; hence, there is additional energy input. In both cases, the efficiency penalty plays the biggest role in increasing the electricity cost and the cost of CO₂ captured.

In the constant net output approach, a new variable called “avoidance ratio” is calculated as the ratio of power plant net efficiency with and without CCS. The avoidance ratio means that the scalability ratio has to be included to keep the net electrical power output constant. It is useful to scale up the CCS cost variables and calculate the avoidance rate as:

$$\text{Avoidance rate} = \frac{\eta_{\text{reference}}}{\eta_{\text{new}}}$$

3.2. Levelized Cost of Electricity

The integration of the cost of the hybrid capture system and the reference power plant enables the calculation of the new electricity cost (LCOE_{cc}), cost of CO₂ captured and cost of CO₂ avoided. Based on the total annual cost, LCOE_{cc} is calculated with the following equation:

$$LCOE_{cc}(\text{€/MWh}) = CLEV + CLEV_{mem} + TMC + ENCOST + REFCOST$$

where *CLEV* is the levelized investment cost of the hybrid system (excluding membrane), *CLEV_{mem}* is the levelized investment cost of the membranes, *TMC* is the total maintenance cost, *ENCOST* is the energy cost incurred by hybrid system operation, and *REFCOST* is the total annual cost of the reference power plant, which consists of the levelized investment, operation and maintenance, fuel cost and other additional variables from reference [20].

All levelized costs related to the hybrid system are scaled up and integrated with the power plant costs and illustrated in Figure 7. It is clear that the cost incurred by the hybrid system with a separation degree of 90% almost doubles the annual power plant cost. Note that membrane investment already comprises 13.5% of the total annual

cost, similar to the investment in the capture plant (14.7%) itself. Furthermore, additional energy accounts for a considerable share at 14.9% of the total annual cost.

Conversely, for the hybrid system with a separation degree of 70%, the cost of energy and investment in the capture plant (excluding the membrane) dominate at 12% and 11.4% of the total annual cost, respectively. A lower separation degree reduces the share of membrane investment. Furthermore, for the hybrid system with a separation degree of 50%, the additional cost of carbon capture is only 22.7%, where additional energy accounts for 9.2%, and the membrane investment cost becomes even more insignificant, accounting for merely 3.7%.

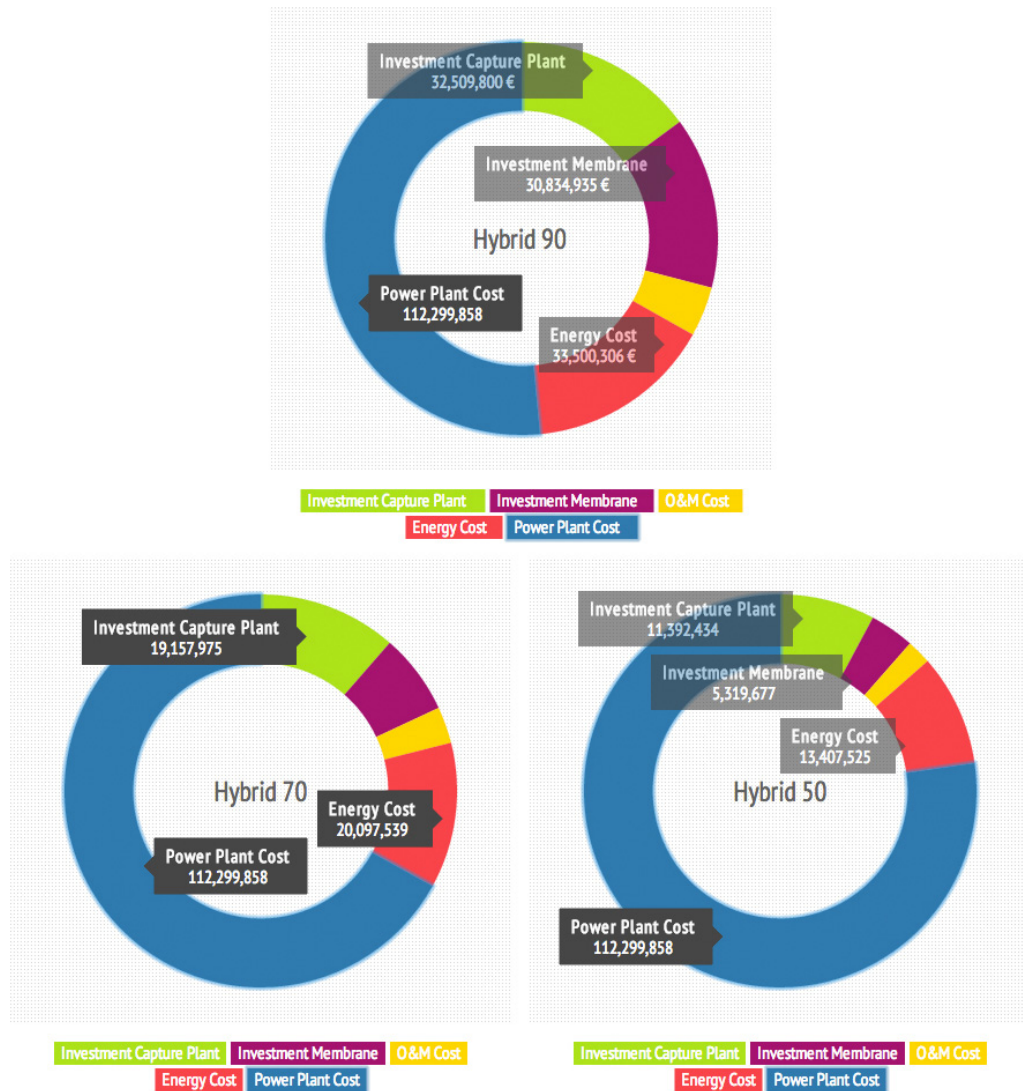


Figure 7. Total annual cost for all separation degree targets

3.3. Captured and Avoided Cost of CO₂

Both LCOE_{ref} and LCOE_{cc} serve as the basis to calculate the cost of CO₂ captured and avoided. This measure is used to assess the economic viability of a CO₂ capture system relative to the market price of CO₂ as an industrial commodity [45].

$$\text{Cost of CO}_2 \text{ captured } (\$/\text{tCO}_2) = \frac{\text{LCOE}_{cc} - \text{LCOE}_{ref}}{(\text{tCO}_2/\text{MWh})_{\text{captured}}}$$

$$\text{Cost of CO}_2 \text{ avoided } (\$/\text{tCO}_2) = \frac{\text{LCOE}_{cc} - \text{LCOE}_{ref}}{(\text{tCO}_2/\text{MWh})_{\text{avoided}}}$$

Furthermore, the term “cost of CO₂ avoided” is the most common measure of the cost of CCS [16, 45, 46]. It is used to define the cost of avoiding a tonne of CO₂ emissions while keeping the net power plant output constant. The plant annual cost, LCOE_{cc}, and cost of CO₂ avoided are summarized in Table 6.

Table 6. Power plant cost of electricity, capture and CO₂ avoidance

Aspen ID	Description	90%		70%		50%	
		Constant gross output	Constant net output	Constant gross output	Constant net output	Constant gross output	Constant net output
LEVELIZED INVESTMENT COST (€/year)							
CLEV	Levelized investment cost	25,036,025	32,509,800	16,253,498	19,157,975	10,176,593	11,392,434
CLEVM	Levelized investment cost, membrane	23,746,200	30,834,935	9,445,783	11,133,731	4,751,942	5,319,677
OPERATION AND MAINTENANCE COST (€/year)							
TMC	Total maintenance cost	6,892,892	8,950,564	4,275,869	5,039,961	2,629,906	2,944,112
ENERCOST	Additional energy cost		33,500,306		20,097,539		13,407,525
REFERENCE POWER PLANT (€/year)							
REFINV	Total annual cost reference	112,299,858	112,299,858	112,299,858	112,299,858	112,299,858	112,299,858
toNEW COST OF ELECTRICITY							
LCOecc	Electricity cost in €/MWh	65.5	65.5	50.3	50.3	43.6	43.6
CCAPTURE	Capture cost in €/tCO ₂	35.9	35.9	26.7	26.7	23.4	23.4
CAVOID	Avoidance cost in €/tCO ₂	-	48.24	-	34.02		30

For the constant net output, the CO₂ captured was scaled up based on the avoidance rate. The CO₂ avoided variable applies only to the constant net output integration.

3.4. Carbon Price

The carbon price sensitivity was calculated and compared with the $LCOE_{cc}$, as shown in Figure 8. The intersection point refers to the CO_2 permits value breakeven point in €/t CO_2 which drives the economic viability of a hybrid CCS system. Below the breakeven point, it is more favorable from an economic perspective not to implement a CCS system and power plant companies prefer to pay for CO_2 emissions by purchasing carbon credits or paying carbon tax.

Implementing a hybrid system with a targeted separation degree of 90% would only be favorable if the carbon price reached € 49/t CO_2 . A lower separation degree of 70% or 50% could be implemented at lower CO_2 permit prices of €34/ t CO_2 and € 30/t CO_2 , respectively.

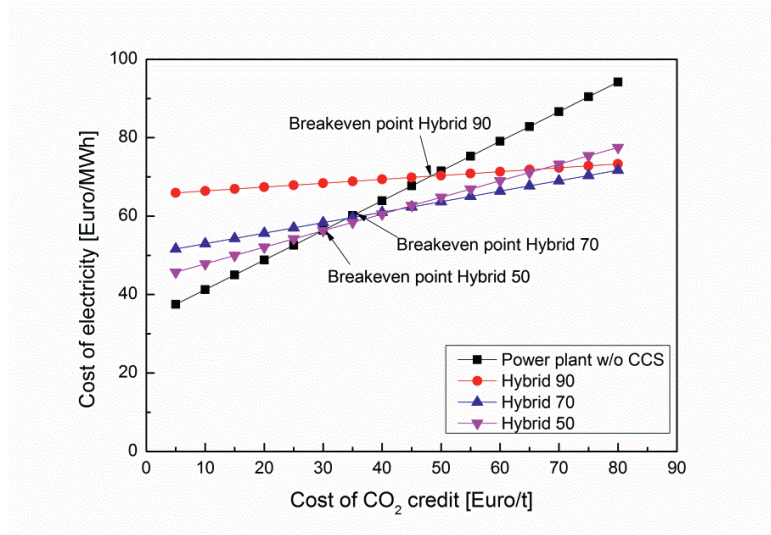


Figure 8. Breakeven point comparison for hybrid CCS system with different CO_2 separation degree

4. Conclusions

As a current state-of-the-art carbon capture technology, MEA absorption has several drawbacks including solvent degradation, high-energy consumption for the regeneration process, and additional environmental concerns regarding the solvent and its disposal. Membrane separation is expected to add to carbon capture technology, particularly to the post-combustion route due to the simplicity and practicability of retrofitting power plants. To achieve high separation degrees on a large scale, such as in pulverized-coal power plants, a multi-stage membrane with a large area is required. However, the investment cost for such a system is so high that it could hinder the implementation of the technology. A hybrid system combining membrane separation and cryogenic technology improves the membrane's ability to capture CO_2 from the flue gas, improves the purity of the final product (>98%), and reduces the membrane area required, thus leading to a lower investment cost.

- Membrane properties are the key to the hybrid system as a whole. Improving CO_2 permeability and CO_2/N_2 selectivity will enhance the separation process and reduce the work of the cryogenic unit. Better membrane properties in the future would mean less work and thus lower energy requirements.
- The membrane separation unit decides the degree of CO_2 captured, whereas the cryogenic unit controls the purity of the end-product. A higher driving force facilitates better separation although it increases the energy requirement. On the other hand, the operating pressure of cryogenic separation consists of two-stage separation vessels and dictates the desired purity.

- With an efficiency penalty of 10.6% for hybrid systems with a 90% degree of separation, the energy requirement of the hybrid system is slightly higher than MEA absorption (10.3%). However, lower separation degree targets such 70% and 50% result in efficiency penalties of 7% and 4.9%, respectively. These are lower than for MEA absorption with penalties of 8.2% and 5.8% for the same separation degree target.
- The cost structure of the hybrid system is dominated by membrane investment and the cost of energy. In a base case result for a hybrid system with a 90% separation degree with P_{feed} of 1.5 bar, the membrane area requirement is 1.92 million m^2 . Lower separation degree targets of 70% and 50% reduce the area required to 758,479 m^2 and 378,664 m^2 , respectively.
- A breakpoint condition for the hybrid system with a 90% degree of separation would be achieved if the CO_2 permit price reached € 49/ tCO_2 . At lower carbon prices of € 30/ tCO_2 and € 34/ tCO_2 , respectively, hybrid systems with CO_2 separation degrees of 70% and 50% would be more favourable for implementing for post-combustion capture.

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References

1. *The Future of Coal*, 2007, Massachusetts Institute of Technology (MIT), Massachusetts, http://web.mit.edu/coal/The_Future_of_Coal.pdf (last accessed on 14. July, 2014).
2. World Coal Institute, *SECURING THE FUTURE: Financing carbon capture and storage in post-2012 World*, 2009, World Coal Institute: London.
3. IEA, *Energy Technology Perspective*, 2010, International Energy Agency: Paris.
4. Rubin, E.S., et al., *The outlook for improved carbon capture technology*, 2012, Progress in Energy and Combustion Science, doi:10.1016/j.pecs.2012.03.003.
5. *The Global Status of CCS*, 2012: Canberra, Australia.
6. Czaperek, M., et al., *Gas separation membranes for zero-emission fossil power plants: MEM-BRAIN*. Journal of Membrane Science, 2010. **359**(1-2): p. 149-159.
7. Zhao, L., et al., *How gas separation membrane competes with chemical absorption in postcombustion capture*. Energy Procedia, 2011. **4**: p. 629-636.
8. Zhao, L., et al., *Cascaded Membrane Processes for Post-Combustion CO_2 Capture*. Chemical Engineering & Technology, 2012. **35**(3): p. 489-496.
9. Car, A., et al., *PEG modified poly(amide-b-ethylene oxide) membranes for CO_2 separation*. Journal of Membrane Science, 2008. **307**(1): p. 88-95.
10. Car, A., et al., *Tailor-made polymeric membranes based on segmented block copolymers for CO_2 separation*. Advanced Functional Materials, 2008. **18**: p. 2815-2823.
11. Zhao, L., et al., *A parametric study of CO_2/N_2 gas separation membrane processes for post-combustion capture*. Journal of Membrane Science, 2008. **325**(1): p. 284-294.
12. Low, B.T., et al., *A parametric study of the impact of membrane materials and process operating conditions on carbon capture from humidified flue gas*. Journal of Membrane Science, 2013. **431**: p. 139-155.
13. Hussain, A. and M.-B. Hägg, *A feasibility study of CO_2 capture from flue gas by a facilitated transport membrane*. Journal of Membrane Science, 2010. **359**: p. 140-148.
14. Brinkmann, T., T. Wolff, and J.-R. Pauls. *Post-Combustion Processes Employing Polymeric Membranes*. in *2nd International Conference on Energy Process Engineering Efficient Carbon Capture for Coal Power Plants*. 2011. 20th-22nd, 2011, Frankfurt am Main, Germany.
15. Merkel, T.C., et al., *Power plant post-combustion carbon dioxide capture: An opportunity for membranes*.

- Journal of Membrane Science, 2010. **359**: p. 126-139.
16. Wilcox, J., *Carbon Capture* 2012, New York: Springer.
 17. Belaisaoui, B., et al., *Hybrid membrane cryogenic process for post-combustion CO₂ capture*. Journal of Membrane Science, 2012. **415-416**: p. 424-434.
 18. Scholes, C.A., et al., *Cost competitive membrane—cryogenic post-combustion carbon capture*. International Journal of Greenhouse Gas Control, 2013. **17**: p. 341-348.
 19. Hasse, D., et al., *CO₂ capture by sub-ambient membrane operation*. Energy Procedia, 2013. **37**: p. 993-1003.
 20. *Konzeptstudie: Referenzkraftwerk Nordrhein-Westfalen (RW NRW), February 2004*, VGB Power Tech e.V., Essen, Germany.
 21. *CO₂ capture ready plants*, 2007/4, May 2007, IEA Greenhouse Gas R&D Programme (IEA GHG).
 22. Low, B.T., et al., *A parametric study of the impact of membrane materials and process operating conditions on carbon capture from humidified flue gas*. Journal of Membrane Science, 2012, accepted.
 23. Yave, W., et al., *CO₂-Phylic Polymer Membrane with Extremely High Separation Performance*. Macromolecules, 2010. **43**(1): p. 326–333.
 24. Yave, W., A. Car, and K.V. Peinemann, *Nanostructured membrane material designed for carbon dioxide separation*. Journal of Membrane Science, 2010. **350**: p. 124.
 25. Yave, W., et al., *Design, synthesis, characterization and optimization of PTT-b-PEO copolymers: A new membrane material for CO₂ separation*. Journal of Membrane Science, 2010. **362**(1-2): p. 407-416.
 26. *MEM-BRAIN Alliance (Gas separation membranes for zero-emission fossil power plants)*, Oct. 2007 - Jun. 2011, 18 research institutions and 5 industrial partners Coordinator: IEF-1, Forschungszentrum Jülich GmbH.
 27. *METPORE (Nano-structured Ceramic and Metal Supported Membranes for Gas Separation)*, Jan. 2007 - May. 2014, funded by the Federal Ministry of Economics and Technology (BMWi), Germany.
 28. Sijbesma, H., et al., *Flue gas dehydration using polymer membranes*. Journal of Membrane Science, 2008. **313**(1): p. 263-276.
 29. NEL, T., *A Study of Measurement Issues for Carbon Capture and Storage (CCS)*, http://www.tuvnel.com/tuvnel/Measurement_Issues_Carbon_Capture_and_Storage_CCS/ (last access on 18 July, 2014).
 30. Primabudi, E., *Investigation of Hybrid System for Post-combustion CO₂ Capture*, in *Master thesis* 2013, TU-Berlin.
 31. AG, L., *Aluminium Plate-Fin Heat Exchangers*, http://www.linde-engineering.com/internet.global.lindeengineering.global/en/images/P_3_2_e_12_150dpi19_5772.pdf (last access on 18 July, 2014).
 32. AG, L., *Coil-Wound Heat Exchangers*, http://www.linde-engineering.com/internet.global.lindeengineering.global/en/images/P_3_1_e_12_150dpi19_5793.pdf (last access on 18 July, 2014).
 33. Finkenrath, M., *Cost and Performance of Carbon Dioxide Capture from Power Generation*, 2011, International Energy Agency: Paris, France.
 34. Abu-Zahra, M.R.M., J.P.M. Niederer, and P.H.M. Feron, *CO₂ capture from power plants Part II. A parametric study of the economical performance based on mono-ethanolamine*. International Journal of Greenhouse Gas Control 2007. **1**: p. 135-142.
 35. Galindo-Cifre, P., et al., *Integration of a chemical process model in a power plant modelling tool for the simulation of an amine based CO₂ scrubber*. Fuel, 2009. **88**(12): p. 2481-2488.
 36. Favre, E., *Carbon dioxide recovery from post-combustion processes: Can gas permeation membranes compete with absorption?* Journal of Membrane Science, 2007. **294**(1-2): p. 50-59.
 37. Belaisaoui, B., D. Willson, and E. Favre, *Membrane gas separations and post-combustion carbon dioxide capture: Parametric sensitivity and process integration strategies*. Chemical Engineering Journal, 2012. **211-212**: p. 122-132.
 38. Centeno, T.A. and A.B. Fuertes, *Carbon molecular sieve membranes derived from a phenolic resin supported on porous ceramic tubes*. Separation and Purification Technology, 2001. **25**(1-3): p. 379-384.
 39. Santos, S. *CO₂ Capture, processing and transport*. IEA Greenhouse Gas R&D Programme 24th May, 2007.
 40. Loh, H.P., J. Lyons, and C.W. White III, *Process Equipment Cost Estimation*, 2002, NETL US Department of Energy: Pittsburgh.
 41. Zhao, L., et al., *Multi-stage gas separation membrane processes with post-combustion capture: energetic and economic analyses*. Journal of Membrane Science, 2010. **359**: p. 160-172.

42. Zhai, H. and E.S. Rubin, *Techno-economic assessment of polymer membrane systems for postcombustion carbon capture at coal-fired power plants*. Environmental Science & Technology, 2013. **47**(6): p. 3006-14.
43. Sinnott, R.K. and G. Towler, *Chemical Engineering Design: SI Edition*2009: Access Online via Elsevier.
44. Richards, J.R. and A.C.T. P.C., *Control of Gaseous Emissions*, in *APTI Course 415 Third Edition*2000, US Environmental Protection Agency: North Carolina.
45. Rubin, E.S., *Understanding the pitfalls of CCS cost estimates*. International Journal of Greenhouse Gas Control, 2012. **10**: p. 181-190.
46. Finckenrath, M., *Cost and performance of carbon dioxide capture from power generation*, 2011, OECD Publishing.