

# Analysis of hybrid membrane and chemical absorption systems for CO<sub>2</sub> capture

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**Abstract**—Amine-based absorption of CO<sub>2</sub> is currently the industry standard technology for capturing CO<sub>2</sub> emitted from power plants, refineries and other large chemical plants. However, more recently there have been a number of competing technologies under consideration, including the use of membranes for CO<sub>2</sub> separation and purification. We constructed and analyzed two different hybrid configurations combining and connecting chemical absorption with membrane separation. For a particular flue gas which is currently treated with amine-based chemical absorption at a pilot plant we considered and tested how membranes could be integrated to improve the performance of the CO<sub>2</sub> capture. In particular we looked at the CO<sub>2</sub> removal efficiency and the energy requirements. Sensitivity analysis was performed varying the size of the membranes and the solvent flow rate.

Keywords: Membrane Separation, Chemical Absorption, CO<sub>2</sub> Capture, Process Design, Hybrid Separation

## INTRODUCTION

Growing emissions of CO<sub>2</sub> are a global concern and there is great demand for solutions that can capture or reduce these emissions conveniently with minimal energy and costs involved. However, in most cases the methods for CO<sub>2</sub> capture are also energy intensive. So for a power plant this would mean using a significant fraction of the energy generated to capture the associated CO<sub>2</sub> produced.

There have been a number of studies looking at the use of amine-based solvents for CO<sub>2</sub> capture [1–5]. These include cases where the absorption has been simulated either by using stage-based equilibrium calculations [4] or by solving sets of rate based equations [3,5]. In addition, a number of authors have used process simulators such as Aspen plus [1,2].

There have recently been a number of studies published considering the capture of CO<sub>2</sub> using membranes [6–8]. These studies mostly consider the potential of membranes with a predicted CO<sub>2</sub> permeance of 1,000 GPU\* rather than considering the capability of existing membranes. However, considering that membranes are constantly being improved by developers in industry and academia, this is not an unrealistic assumption.

In many cases the simulation of membranes considers only a binary gas separation which simplifies the calculations. However, multicomponent models are also available [9,10] and can be used for design and optimization of gas separation processes.

Combining different separation technologies to form hybrid systems has been considered by a number of different authors, as reviewed by Suk and Matsuura [11]. For the case of natural gas sweetening, Bhide et al. [12] considered the benefits of hybrid membrane and chemical absorption and they show that under certain conditions the hybrid solution is economically favorable. Belaisaoui et al. [13] combine a membrane separation with a cryogenic separa-

tion process for the capture of post-combustion CO<sub>2</sub> capture. In addition, Scholz et al. [14] have considered the use of hybrid systems for the upgrading of biogas.

We investigated the benefits and any possible disadvantages associated with hybrid membrane and chemical absorption systems for post-combustion CO<sub>2</sub> capture. In this way we aim to show how energy requirements can be reduced using hybrid systems in a way similar to Belaisaoui et al. [13], who showed that membranes combined with a cryogenic process can give more energy efficient removal of CO<sub>2</sub>.

## PROCESS MODELING AND DESIGN METHODOLOGY

### 1. Membrane Separation

In this work membrane separation is simulated based on the models and equations given by Katoh et al. [10] and Coker et al. [9]. These simulations model the gas separation of hollow fiber membranes operated in a counter-current flow pattern. According to these models, the rate of transfer of component *i* across the membrane is given by Eq. (1).

$$J_i = Q_i A_m (P_r x_r - P_p y_i) \quad (1)$$

where  $Q_i$  is the permeance of component *i*,  $A_m$  is the effective surface area of the membrane,  $P_r$  and  $P_p$  refer to the retentate and permeate pressures and  $x$  and  $y$  are the retentate and permeate mole fractions. Here the most important parameters for membrane-based gas separation are the permeances of each component, which determine the relative rate of transfer of the different components across the membrane.

To simulate the separation of gases a tanks-in-series model is implemented (see Fig. 1) based on Eqs. (1)–(5):

$$\frac{dx_{i,j}}{dt} = \left( \frac{F_{r,j-1} x_{i,j-1} - J_{i,j,n} - F_{r,j} x_{i,j}}{V_r/S_r} \right) \quad (2)$$

\* 1 GPU =  $7.501 \times 10^{-12}$  Nm<sup>3</sup> m<sup>-2</sup> s<sup>-1</sup> Pa<sup>-1</sup>

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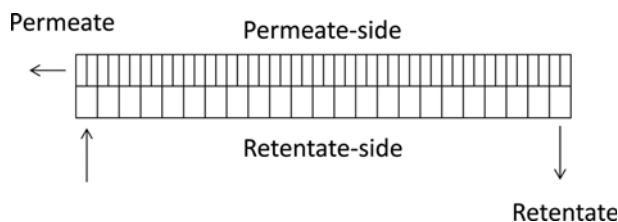


Fig. 1. Schematic diagram of a counter-current membrane.

$$\frac{dy_{i,n}}{dt} = \left( \frac{F_{p,n+1}Y_{i,n+1} + J_{i,j,n} - F_{p,n}Y_{i,n}}{V_p/S_p} \right) \quad (3)$$

$$F_{r,j} = F_{r,j-1} - \sum_{i=1}^{C_n} J_{i,j} \quad (4)$$

$$F_{p,n} = F_{p,n+1} + \sum_{i=1}^{C_n} J_{i,n} \quad (5)$$

where the retentate mole fraction  $x_{i,j}$  of component  $i$  in tank  $j$  and the permeate mole fraction  $y_{i,n}$  of component  $i$  in tank  $n$  are calculated using Eqs. (2) and (3).  $V_r$  and  $V_p$  are the volumes of the retentate and permeate sides, and  $S_r$  and  $S_p$  are the number of tanks used to model the retentate and permeate.  $F_{r,j}$  and  $F_{p,n}$  are the total flow rates out of tanks  $j$  and  $n$  into tanks  $j+1$  and  $n-1$  calculated using Eqs. (4) and (5). Also,  $J_{i,j,n}$  refers to the volumetric flow rate of component  $i$  between tanks  $j$  and  $n$ .

In this model it is assumed that there is no pressure drop on the retentate side and on the permeate side the pressure change can be calculated using the Hagen-Poiseuille equation [9,10,15].

To simulate the gas separation using this model, Eqs. (1)–(5) are solved simultaneously using the Newton-Raphson method programmed within Matlab. To obtain reasonable initial conditions, these equations are also solved in Matlab using relaxation methods similar to those described by Katoh [10].

To create the pressure difference it is necessary to use either compressors applied to the feed or vacuum pumps connected to the permeate outlet. The energy required for these pressure changing units is calculated using Eqs. (6)–(8):

$$T_2 = \frac{T_1}{\eta} \left[ \left( \frac{P_2}{P_1} \right)^{\frac{\gamma-1}{\gamma}} - 1 \right] + T_1 \quad (6)$$

$$\text{Work} = C_p(T_2 - T_1) \cdot F \quad (7)$$

$$\gamma = \frac{C_p}{C_v} \quad (8)$$

where  $T_1$  and  $T_2$  are the inlet and outlet temperatures,  $P_1$  and  $P_2$  are the inlet and outlet pressures,  $\eta$  is the adiabatic efficiency,  $C_p$

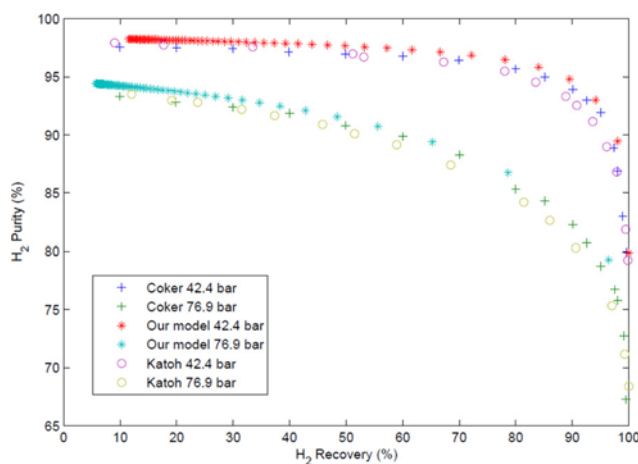


Fig. 2. Comparison of model implemented against literature models.

and  $C_v$  are the constant pressure and constant volume heat capacity,  $\gamma$  is the ratio of heat capacities and  $F$  is the gas flow rate.

To check the performance of this model we have used an example based on hydrogen separation (see parameters in Table 1) and compared our results against those generated by models in the literature [9,10] as shown in Fig. 2. These values were generated using flow rates between  $500 \text{ Nm}^3\text{hr}^{-1}$  and  $30,000 \text{ Nm}^3\text{hr}^{-1}$ .

The small differences between the three models could be explained through different correlations used to calculate properties such as gas viscosity of each component which affect the pressure drop on the permeate side of the membrane. Although, in general, this model is assumed to be working well within the assumptions.

## 2. Amine-based Chemical Absorption

To model the absorption of  $\text{CO}_2$  using amine solvents we have simulated an absorption column using Aspen Plus (see Fig. 3).

Here the flue gas and the lean solvent enter the absorber giving a rich solvent stream containing the captured  $\text{CO}_2$  and a cleaned outlet gas stream. The rich solvent stream would then be regenerated in a stripping column and returned to its initial state so it can be recycled and used again.

In this case monoethanolamine (MEA) is selected as the solvent and the unsymmetrical electrolyte NRTL model with Redlich-Kwong equation of state property package is selected. The mass transfer is represented using a two film model with correlations for interfacial area and mass transfer given by Bravo et al. [16].

The absorption chemical reaction is modelled in Aspen plus using a single irreversible reaction:



where the second-order rate constant for this reaction is given by [17]:

Table 1. Feed gas and membrane properties for hydrogen separation example [9]

Component	Feed composition (mole fraction)	Permeance (GPU)	Membrane properties	
$\text{H}_2$	0.650	100	Inner diameter	150 $\mu\text{m}$
$\text{C}_2\text{H}_4$	0.025	3.03	Outer diameter	300 $\mu\text{m}$
$\text{CH}_4$	0.210	2.86	Number of fibers	500,000
$\text{C}_2\text{H}_6$	0.080	2.00	Effective length	0.8 m
$\text{C}_3\text{H}_8$	0.035	1.89	Total membrane area	377 $\text{m}^2$

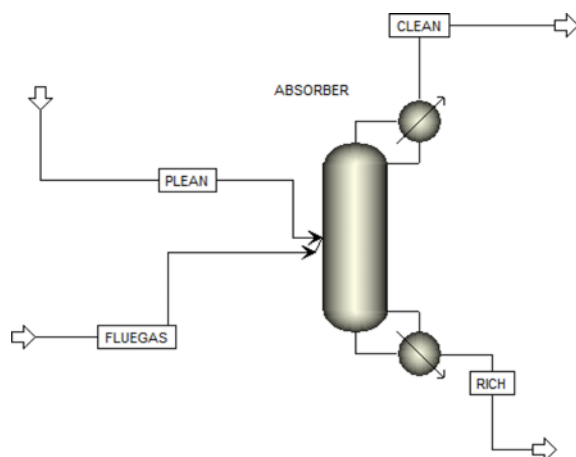


Fig. 3. Aspen plus simulation of chemical absorption using MEA.

$$k = 4.4 \times 10^8 e^{\left(\frac{-5400}{T}\right)} \quad (9)$$

A single irreversible reaction is used here to simplify the modeling which might otherwise include several equilibrium reactions. This single overall step is based on a simple zwitterion mechanism, and it is assumed irreversible here because the reverse step is not favored under typical absorption conditions (30–60 °C with low CO<sub>2</sub> content in the solvent). To check the accuracy of this absorption model we compared the results of our model against experimental data in the literature [3]. As described in the literature [3], these experiments use an absorber with diameter of 0.15 m and with 4.36 m height of Mellapak 250Y packing. Their experiments use 30%wt MEA as solvent with liquid circulation of 3 L/min and 9 L/min in cases 1 and 2 used for comparison with the current Aspen plus model (cases 1 and 2 refer to experimental runs 10 and 15 in the literature reference). Furthermore, their feed gas is 151 m<sup>3</sup> hr<sup>-1</sup> and 143 m<sup>3</sup> hr<sup>-1</sup> in cases 1 and 2, and it is assumed to be saturated with water. Based on the comparison of the outlet conditions shown in Table 2 we see that our model in Aspen plus reproduces the experimental capture rate of CO<sub>2</sub> with reasonable accuracy.

To complete this model of absorption it is necessary to obtain the energy required for regeneration of the solvent, and in this work we have assumed the energy required is 3.5 GJ per ton of CO<sub>2</sub> captured through desorption [13]. This allows the simple but characteristic comparison of the energy requirements in hybrid systems combining membranes with chemical absorption.

## CASE STUDY

### 1. Base Case

This case study is based on a CO<sub>2</sub> capture pilot plant from the

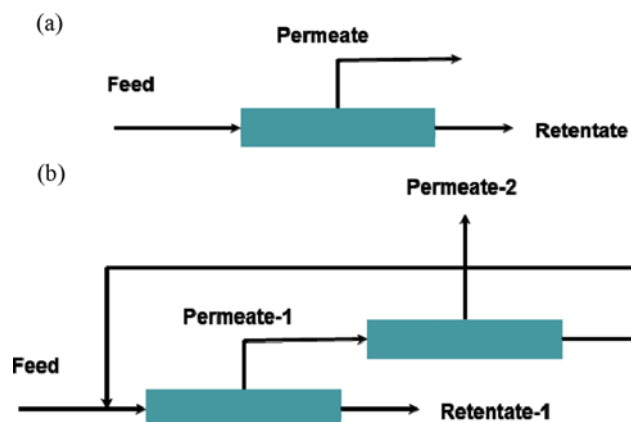


Fig. 4. Two membrane configurations considered. (a) Is a single stage membrane and (b) is a two stage membrane configuration.

literature [18], which uses an amine absorption system and captures approximately 2 tons per day of CO<sub>2</sub>.

The flue gas fed to the capture plant is specified as 574 Nm<sup>3</sup>hr<sup>-1</sup> with a volume-based composition of 10.2% CO<sub>2</sub>, 2.8% O<sub>2</sub> and 87.0% N<sub>2</sub>. In this pilot plant the absorber is 18.8 m high with a diameter of 0.46 m and the stripper is 16.7 m high with a diameter of 0.35 m. Hence, in this case the dimensions of the absorber are fixed, but in general for the design of a new CO<sub>2</sub> capture system these parameters should also be considered and modified to obtain the best performance.

Lee et al. [18] test the sensitivity of their CO<sub>2</sub> capture equipment to determine how a number of a parameter affect the CO<sub>2</sub> recovery efficiency (the percentage of CO<sub>2</sub> captured).

Here we consider the implementation of a hybrid system combining membrane separation and chemical absorption for the capture of CO<sub>2</sub> using this flue gas. Two different membrane configurations are considered (see Fig. 4). We analyze the performance of these systems in terms of CO<sub>2</sub> removal efficiency and energy requirements.

The membrane considered here for CO<sub>2</sub> capture is a hollow fiber membrane from the literature [9] operated in a counter-current arrangement. The permeances of this membrane are

$$\begin{aligned} Q_{N_2} &= 3.57 \text{ GPU} \\ Q_{O_2} &= 20 \text{ GPU} \\ Q_{CO_2} &= 60 \text{ GPU} \\ Q_{H_2O} &= 1000 \text{ GPU} \end{aligned}$$

which should be converted to Nm<sup>3</sup> m<sup>-2</sup> s<sup>-1</sup> Pa<sup>-1</sup> when used in Eqs. (1)–(5). Although the CO<sub>2</sub> permeance is relatively low in this case, the selectivity ( $P_{CO_2}/P_{N_2}$ ) is 43, which allows for significant separa-

Table 2. Absorption model compared against experimental values

Case	Inlet CO <sub>2</sub> (%vol)	Outlet CO <sub>2</sub> (%vol)		Gas outlet temperature (°C)		Lean solvent loading	Rich solvent loading	
		Expt.	Simulation	Expt.	Simulation		Expt.	Simulation
1	2.81	0.64	1.19	58.0	54.6	0.217	0.333	0.313
2	12.12	8.13	7.51	66.6	68.9	0.357	0.435	0.432

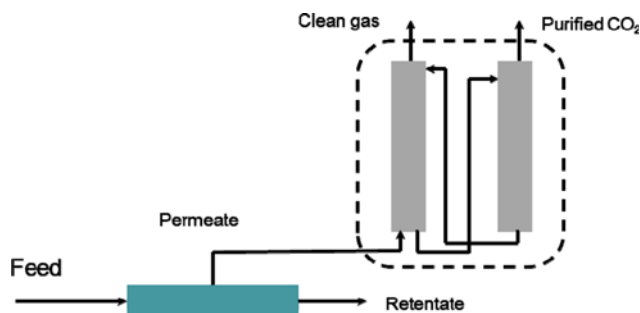


Fig. 5. Hybrid configuration 1: Absorption unit connect to the permeate of a single stage membrane.

tion of  $N_2$  and  $CO_2$ . Also, the geometric properties of the hollow fibers are:

Inner diameter=150 nm  
Outer diameter=300 nm  
Effective length=0.8 m

where the total area is given by

$$\text{Total area} = \text{Outer diameter} \cdot \pi \cdot \text{Effective length} \cdot \text{number of fibers} \quad (10)$$

To create the pressure difference in the membrane systems it is assumed that all membranes have a vacuum pump connected to their permeate outlet operated at 10 kPa. The energy required for vacuum pumps is calculated using Eqs. (6)-(8) using an adiabatic efficiency of 85% (the same value used by Zhai and Rubin [7]).

## 2. Hybrid Capture Systems : Configuration 1

In this first configuration (see Fig. 5) the permeate stream from a single membrane stage is connected to a chemical absorption system. Hence, in this case the  $CO_2$  in the feed (flue gas) must first pass through the membrane before it is passed to the absorption equipment. This also means that any  $CO_2$  remaining in the membrane retentate outlet is released to the atmosphere. Similarly, the clean gas stream coming from the absorber is also released to the atmosphere and only the purified  $CO_2$  stream coming from the desorber contains the captured  $CO_2$ .

For this configuration we investigated the potential benefits. Namely, we tested if the addition of a membrane could reduce energy requirements by reducing the solvent flow rate required inside the absorption system.

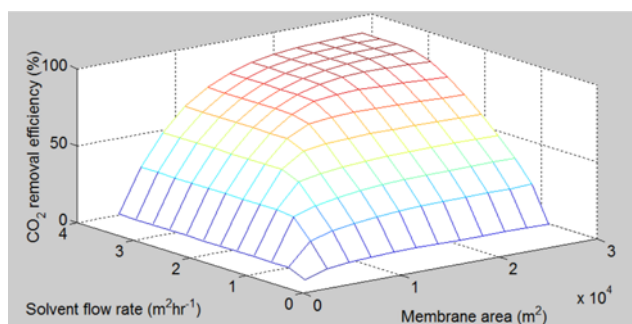


Fig. 6. Effects of membrane area and solvent flow rate on the overall  $CO_2$  removal efficiency.

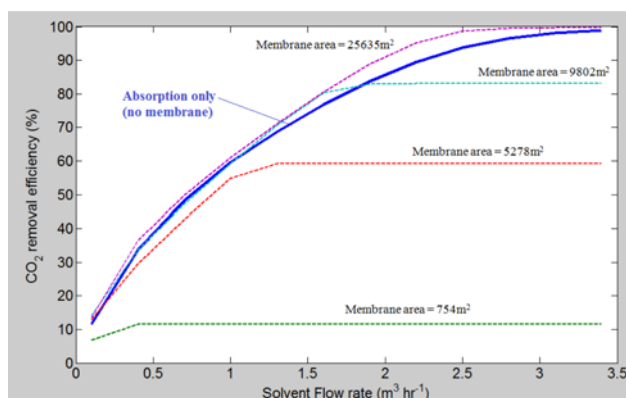


Fig. 7. Comparison of hybrid configuration 1 against an absorption system with no membrane.

To test if the addition of a membrane can reduce solvent flow rate and energy consumption, the membrane surface area and the solvent flow rate were varied to show the effects on the  $CO_2$  capture efficiency.

Figs. 6 and 7 show that high  $CO_2$  recovery for this configuration requires both high membrane area and high solvent flow rate. In cases using low membrane area only a fraction of the  $CO_2$  in the flue gas is passed to the absorption unit and subsequently only this fraction can be captured using the solvents.

Fig. 7 shows that the addition of a large membrane can increase the  $CO_2$  removal efficiency slightly compared with the base case (absorption only). However, this should be balanced against the increased energy required due to using the membrane and absorption systems together.

The energy required for  $CO_2$  capture through absorption is mainly due to the energy of desorption which is required to regenerate the solvent (see Fig. 8). This is a function of the efficiency of absorption into the solvent and the energy required per ton of  $CO_2$  captured.

Using only absorption for  $CO_2$  capture gives an energy requirement of 3.5 GJ per ton of  $CO_2$  [13]. However, with the addition of

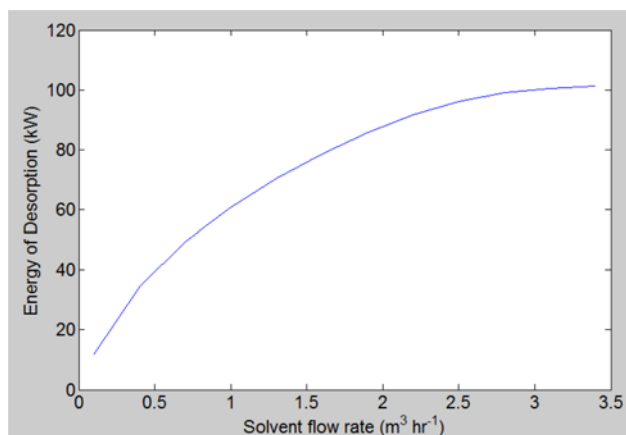


Fig. 8. Energy of desorption resulting from the absorption efficiency and a fixed energy consumption per ton of  $CO_2$  recovered.

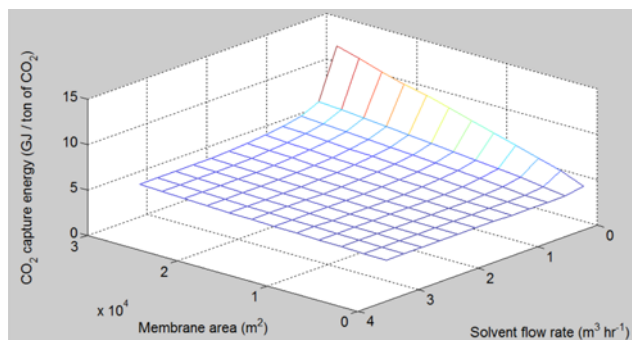


Fig. 9. CO<sub>2</sub> capture energy using hybrid configuration 1 from the case study flue gas [18].

a membrane in this configuration the energy requirements increase (as shown in Fig. 9) and the lowest energy requirement is now around 3.97 GJ per ton. This is due to the energy consumption of the vacuum pump, which increases as the size of the membrane increases leading to increased flow rates. Also, at low solvent flow rates the CO<sub>2</sub> capture energy required is increased further due to the lower CO<sub>2</sub> removal efficiency (see also Fig. 7). Hence, this configuration is quite inefficient because the CO<sub>2</sub> which is separated by the membrane still requires the same amount of energy within the absorption-desorption process.

Also, although the CO<sub>2</sub> recovery efficiency may be slightly higher with a large membrane, the overall energy required is increased, making the capture less energy efficient.

### 3. Hybrid Capture Systems : Configuration 2

In the second configuration considered, the absorption unit is connected to the retentate stream outlet from the membrane. Hence, in this configuration any CO<sub>2</sub> which does not pass through the membrane is sent to the absorber. If a single membrane is used here, then the typical purity of CO<sub>2</sub> recovered from the membrane permeate outlet would be approximately 40-50%. So for this reason a 2-stage membrane is implemented, which allows for purities between approximately 50-90% depending on the conditions. For simplicity the second membrane (connected to the permeate outlet from the first membrane) is given an area which is 10% of the size of the first membrane. This is sufficient for the analysis in the present study, but this ratio is an important parameter which should be considered when carrying out a more detailed design. Additionally, when considering the application of a membrane system it may be favorable to consider designs with three or more membranes. However, any enhanced performance obtained must be weighed against the added complexity introduced due to the additional equipment required for each membrane (e.g., compressors and vacuum pumps). Hence, for simplicity we limit our study to a two-membrane configuration.

This configuration is shown in Fig. 10 where there are three outlets for the system. The clean gas stream is the one with reduced CO<sub>2</sub> content which is released to the atmosphere. The purified CO<sub>2</sub> and the permeate-2 streams are both CO<sub>2</sub>-rich, and these are combined to give the overall CO<sub>2</sub> capture rate and an overall purity.

The overall CO<sub>2</sub> removal efficiencies for this second hybrid configuration are shown in Fig. 11 for a range of conditions. In com-

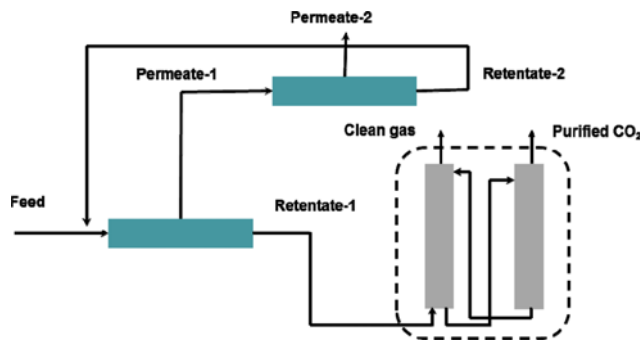


Fig. 10. Hybrid configuration 2: Absorption unit connect to the retentate of a two stage membrane.

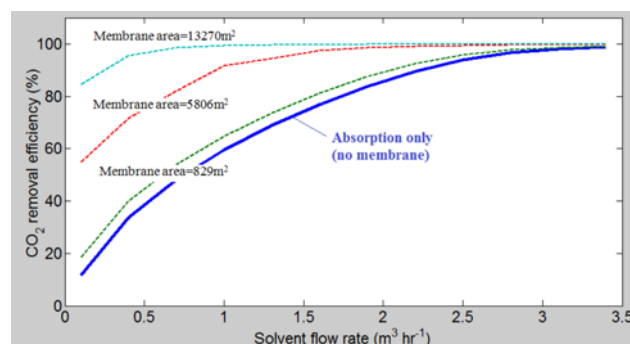


Fig. 11. Comparison of hybrid configuration 2 against an absorption system with no membrane.

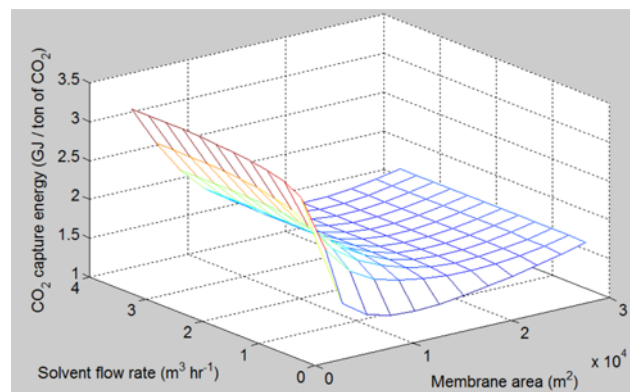


Fig. 12. CO<sub>2</sub> capture energy (kWh/ton of CO<sub>2</sub>) using hybrid configuration 2 from the case study flue gas [18].

parison with hybrid configuration 1 (see Fig. 7) it is clear that the addition of a membrane can significantly increase the CO<sub>2</sub> removal efficiency and reduce the required flow rate of solvent needed to achieve high removal efficiencies.

Considering the energy required for this second hybrid configuration, it can be seen in Fig. 12 that solutions with higher membrane area lead to lower energy requirements for CO<sub>2</sub> removal. The lowest energy solution was found by using a membrane area of 18,246 m<sup>2</sup> and only 0.1 m<sup>3</sup> hr<sup>-1</sup> of solvent requiring only 1.36 GJ per ton of CO<sub>2</sub> (significantly lower than the 3.5 GJ per ton required for only absorption). However, this low energy solution only removes 77.4%

**Table 3. Comparison of options giving approximately 90% CO<sub>2</sub> recovery**

	Hybrid 1	Hybrid 2	Only absorption	Only membrane
Membrane area (m <sup>2</sup> )	21112	10782	0	19490
Solvent flow rate (m <sup>3</sup> hr <sup>-1</sup> )	1.90	0.40	2.24	0
CO <sub>2</sub> capture energy (GJ/ton of CO <sub>2</sub> )	4.62	1.68	3.5	1.39
CO <sub>2</sub> removal efficiency (%)	90.0	90.6	90.2	90.5
Purity of CO <sub>2</sub> removed (%)	>99%	84.7	>99%	74.2

of the CO<sub>2</sub> with a CO<sub>2</sub> purity of only 82.6%. If a higher percentage of CO<sub>2</sub> recovery or purity is required, then a more energy intensive solution may be necessary.

#### 4. Comparison of Configurations

To make a fair comparison between the different configurations, the lowest energy solutions which give greater than 90% CO<sub>2</sub> removal have been identified based on the sensitivity analysis carried out. These results are listed in Table 3.

Using chemical absorption it is possible to obtain a very high purity stream of CO<sub>2</sub> (excluding any water which can be removed through condensation). However, this gives a relatively high energy requirement, due to the high energy needed for chemical absorption. Hybrid configuration 1 is shown to be less energy efficient because the CO<sub>2</sub> which is separated by the membrane must then also be captured using the solvent, and so energy costs are still high in this case. The introduction of a membrane in hybrid configuration 2 or using only the membrane can give reduced energy requirements, but also gives a lower purity of CO<sub>2</sub> captured.

### DISCUSSION AND CONCLUSIONS

For a particular case study of post-combustion CO<sub>2</sub> capture, the implications of combining membrane and chemical absorption for a combined hybrid separation have been investigated.

When the chemical absorption is connected to the membrane permeate stream (hybrid configuration 1), the resulting separation process was found to be more energy intensive in terms of the energy required per ton of CO<sub>2</sub> captured. This is similar to the results found by Scholz et al. [14], where they show that connecting an absorption system to a membrane permeate leads to higher costs when applied to the upgrading of biogas.

However, when chemical absorption is connected to the membrane retentate stream (hybrid configuration 2), the energy required for CO<sub>2</sub> capture can be reduced, depending on the required removal efficiency and purity.

A comparison of the different configurations for CO<sub>2</sub> capture (see Table 2) with a specified 90% CO<sub>2</sub> capture efficiency shows that a membrane system can give the lowest energy requirements (1.39 GJ/ton of CO<sub>2</sub>), but also produces a lower purity stream of CO<sub>2</sub> (74.2%). If higher purity is required, then the hybrid configuration 2 gives the second lowest energy requirements (1.68 GJ/ton of CO<sub>2</sub>) while giving a higher purity of captured CO<sub>2</sub> (84.7%). The highest purity can be obtained using either only absorption or in the hybrid configuration 1, and here the hybrid configuration was found to require around 32% more energy.

Hence, depending on the CO<sub>2</sub> recovery and purity requirements

a hybrid configuration (i.e., configuration 2) may offer the greatest reduction of energy requirements.

In practice there will be a number of additional factors which must be taken into account when considering the energy requirements over the life of the plant. For example, the performance of the membrane will degrade over time, which can be compensated for typically by changing the operating pressures and increasing the corresponding compressor and vacuum pump energies to maintain the overall CO<sub>2</sub> recovery and purity. Additionally, the CO<sub>2</sub> flow rate and composition of the feed gas will likely change with time, and hence the energy requirements may also change with time as the system is tuned to track these changes.

### ACKNOWLEDGEMENTS

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### NOMENCLATURE

$A_m$	: membrane effective surface area [m <sup>2</sup> ]
$C_n$	: number of components [dimensionless]
$C_p$	: constant pressure heat capacity [J mol <sup>-1</sup> K <sup>-1</sup> ]
$C_v$	: constant volume heat capacity [J mol <sup>-1</sup> K <sup>-1</sup> ]
$F$	: total volumetric flow rate to adjacent tank on the same side of the membrane [Nm <sup>3</sup> s <sup>-1</sup> ]
$J$	: volumetric flow rate across the membrane [Nm <sup>3</sup> s <sup>-1</sup> ]
$P$	: pressure
$Q_i$	: permeance of component $i$ [Nm <sup>3</sup> m <sup>-2</sup> s <sup>-1</sup> Pa <sup>-1</sup> ]
$S$	: number of tanks used for modelling the retentate/permeate [dimensionless]
$t$	: time [s <sup>-1</sup> ]
$T$	: temperature [K]
$V$	: volume of the retentate/permeate [m <sup>3</sup> ]
$x_i$	: retentate mole fraction of component $i$ [dimensionless]
$y_i$	: permeate mole fraction of component $i$ [dimensionless]

### Greek Characters

$\eta$	: adiabatic efficiency [dimensionless]
$\gamma$	: ratio of heat capacities [dimensionless]

### Subscripts

$i$	: $i$ -th component
$j$	: $j$ -th permeate tank



n : n-th retentate tank  
 p : permeate  
 r : retentate

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