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Membrane properties required for post-combustion CO₂ capture at coal-fired power plants

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ABSTRACT

This paper focuses on the identification of membrane properties required to enable cost-competitive post-combustion CO₂ capture from a coal power plant using membrane-based processes. In order to identify such properties, a numerical version of the attainable region approach proposed by Lindqvist et al., built as part of the of the iCCS tool developed by SINTEF Energy Research, is used to identify and assess the technical and cost performances of the optimal membrane process for a given set of membrane properties (selectivity and permeance). This numerical model is used to assess the cost performances of 1600 sets of membrane properties (selectivity and permeance) for post-combustion CO₂ capture from a coal power plant as defined by the European Benchmarking Task Force and compare it with the reference commercial solvent concept (MEA) to identify the membrane properties required in a base case that treats both membrane- and MEA-based processes as mature and developed. The results show that to reach this competitiveness with simple process configurations requires a permeance of at least 3 m³(STP)/(m² · h · bar) with high selectivity, or alternatively a selectivity of at least 65 with high permeances. These limits can be reduced to permeances as low as 1 m³(STP)/(m² · h · bar) with high selectivity, or selectivities as low as 30 with high permeances, when advanced membrane process configurations are being considered. The assessments of five additional cases quantify how additional costs associated with demonstration projects and higher membrane module costs can significantly increase the selectivities and permeances required to compete with MEA based capture.

In order to link the membrane development works to the results presented in this paper, the constraint introduced by Robeson's upper bound limitation, as well as data available in the literature on membrane modules and polymeric materials, are compared with the results obtained. The inclusion of the upper bound shows that the capacity to generate thin membrane film layers is important in order to avoid reducing the range of membrane properties, in which diffusion governed membrane can be interesting in term of cost performances, especially in cases that take demonstration and/or higher module costs into consideration. The comparison with literature data shows that while several membranes and polymeric materials have the potential to be cost-competitive with further properties improvements, and once membrane-based CO₂ capture becomes mature and demonstrated, financial support will be required to demonstrate and help mature the technology.

Finally, ways to use the results presented here for membrane development by membrane development experts, for membrane selection by industrial users, and for technology development and demonstration support by decision-makers are discussed.

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Abbreviations: ASC, advanced supercritical pulverised bituminous coal; CAPEX, capital expenditures; CCR, CO₂ capture ratio; CCS, carbon capture and storage; CEPCI, chemical engineering plant cost index; EBTF, European Benchmarking Task Force; EPC, Engineering, Procurement, Construction; EPCCI, European Power Capital Costs Index; FOAK, First Of A Kind; GHG, greenhouse gas; IPCC, Intergovernmental Panel on Climate Change; LCA, life-cycle assessment; MEA, mono-ethanolamine; NOAK, Nth Of A Kind; OPEX, operating expenditures; SOAK, Second Of A Kind; TDC, total direct costs

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1. Introduction

According to data from the International Energy Agency [1], mitigation efforts across the world have led to a halt at 32.3 billion tonnes in the global emissions of carbon dioxide from the energy sector in 2014. This is the first time in 40 years that a halt or a reduction in greenhouse gas emissions, not linked to an economic turnaround, has been observed. However, despite this encouraging stall, significant efforts and measures will still have to be taken in order to meet the 2 °C constraint.

Carbon capture and storage (CCS) is regarded as one of the most promising technologies for reducing man-made carbon atmospheric emissions, and is projected to provide 14% of the reduction in man-made greenhouse gas (GHG) emissions by 2050 [2]. While solvent-based CO₂ capture is the most mature and demonstrated technologies for CO₂ capture, other emerging technologies such as membrane, cryogenic separation, precipitating solvents, and adsorption have the potential to significantly reduce costs in the long run. Among these emerging technologies, membrane-based CO₂ capture is regarded as one of the most mature and promising options [3].

To compete with solvent-based systems for CO₂ capture, development of membranes with improved performances is essential. While Robeson has identified the theoretical constraints on achievable membrane material properties (selectivity and permeability) [4], Powell and Qiao [5] and Scholes et al. [6] have gathered together the properties of more than 400 materials that could be used to build polymeric membrane modules for separation of CO₂/N₂ mixtures. While several membranes with either low permeances or low to moderate selectivities have been reported [7], two distinctive approaches have been considered for the development of membranes. The first approach has been to start from membranes with moderate selectivity (30–50) and good permeances and try to improve mainly their permeances while maintaining or if possible slightly increasing their selectivity [8]. The second is to start with high selectivity (150–200) membranes, such as Mixed Matrix membranes, many of which have low permeance, and to try to improve their permeance performance [7,9,10].

However, even if membrane development experts have a good idea of what membrane properties are desired for membrane-based CO₂ separation [8], no benchmarking has been performed to quantify the membrane properties (permeance and selectivity) required for membrane process to compete with solvent-based CO₂ capture. In view of the high investments required for membrane processes and the process design's heavy dependence on membrane properties, a cost-based comparative approach as proposed here should be developed and employed to identify the range of membrane properties that are required if membrane processes are to compete with solvent-based CO₂ capture.

Although membrane processes are conceptually very simple, complicated multi-stage membrane process configurations are often employed in practice to meet product purity and capture ratio constraints. To minimize the cost of CO₂ capture of such membrane systems, multiple process design decisions regarding process configuration, operating conditions and membrane properties have to be made to ensure a suitable driving force for gas separation and determine the optimal trade-off between the separation work and membrane area requirements.

A graphical analysis called the Attainable Region Approach has been developed by Lindqvist et al. [11–13] in order to easily design a cost-optimal multi-stage membrane separation system for given membrane properties. In this study, a numerical version of the analysis proposed by Lindqvist et al. [11–13], built as part of the of the iCCS tool

[14,15] developed by SINTEF Energy Research within the BIGCCS Research Centre [16], is used to identify the membrane properties required for membrane systems to be economically competitive with the commercial MEA-based technology for post-combustion CO₂ capture from a coal power plant. In addition to the identification of the membrane properties required for a base case, five additional cases were modelled and analysed in order to quantify the impact of the membrane module cost and the additional costs associated with demonstration projects on the membrane properties required. Finally, the results are compared with literature data and the utilisation of results by membrane developers and decision-makers are discussed.

2. Methodology

2.1. Concept and case study descriptions

The aim of this study is to identify the membrane properties required to enable cost-competitive CO₂ post-combustion capture using polymeric membrane from a coal power plant compared to the reference commercially available capture technology (MEA based absorption). In this concept, the two CO₂ capture technologies are considered to capture 90% of the CO₂ from the post-combustion flue gas of a European-based coal-fired power plant, as described by the European Benchmarking Task Force (EBTF) [17].

In order to reach this objective, a numerical model of the attainable region approach presented previously [11–13] is used to optimize and evaluate several combinations of membrane properties (selectivity and permeance). The cost performances resulting of the membrane optimisation process are combined with the power plant costs assessed by the EBTF and compared with the reference power plant using MEA-based solvent capture to identify which combinations of membrane properties can lead to a capture process that will be cost-competitive with the reference technology.

2.2. Technical modelling

2.2.1. Coal-based power plant

The power plant under consideration is based on an Advanced SuperCritical (ASC) boiler and turbine as presented by Anantharaman et al. [17]. This coal-based power plant delivers a gross power of 819 MWe without carbon capture. Once auxiliary power accounted, the net power output of the plant is 754.3 MWe, giving a net plant efficiency of 45.5 %.

This power plant emits after desulphurisation a wet flue gas at the rate of 781.77 kg/s and which contains 13.7%_{wet,vol} of CO₂ (equivalent to 15.2%_{dry,vol}). The coal power plant therefore emits an average of 4.3 Mt_{CO2} per annum and produces electricity with a CO₂ emission rate of 763 kg/MWh when no capture is considered.

The power plant with CO₂ capture is expected to have the same overall organization. However, as shown in Fig. 1, the cleaned flue gas from the power plant containing CO₂ is sent to the CO₂ capture

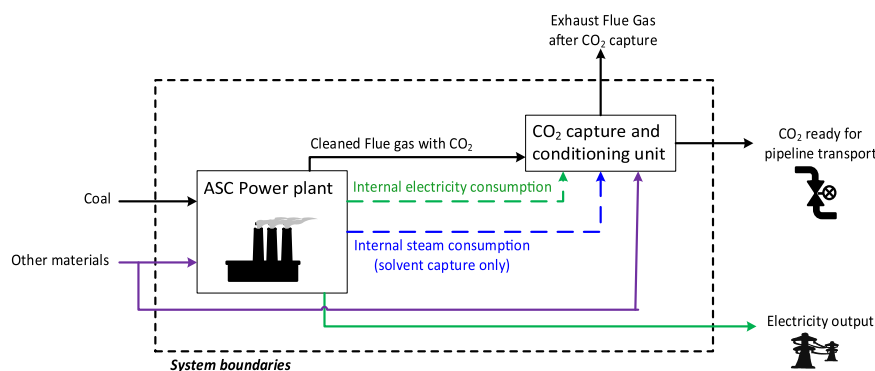


Fig. 1. Schematic view of the ASC power plant with CO₂ capture and conditioning.

and conditioning unit. During the CO₂ capture step, CO₂ is removed from the flue gas using either a membrane-based process or MEA solvent-based CO₂ capture. The thermal power necessary for the solvent-based capture and the electrical power required by both capture technologies are provided by the power plant and therefore their consumption reduces the overall performances of the power plant with CO₂ capture. The captured CO₂ is then conditioned to meet the conditions required for pipeline transport and storage, while the rest of the flue gas is vented. The CO₂ conditioning process consists of compression stages and pumping, combined with the removal of unwanted components (dehydration) to reach a CO₂ purity of at least 95% [18] and a pressure of 110 bar [17].

2.2.2. CO₂ capture units

2.2.2.1. Membrane-based CO₂ capture.

In order to identify the range of membrane properties necessary for cost competitive post-combustion CO₂ capture from coal power plant using membranes, 1600 combinations of membrane properties with selectivities of up to 200 and permeances of up to 10 m³(STP)/(m²·h·bar) are evaluated. The maximum selectivity corresponds to the highest CO₂/N₂ selectivity reported in the literature [9,10] and the highest value considered by the upper bound constrain [4]. The maximum permeance considered corresponds to around twice the permeance of the membranes currently available commercially¹ [8], in order to take future membrane development in membrane materials into account, as significant development work is taking place to improve the permeance of existing membranes. It is worth noting that in practice it is not expected that a membrane with a selectivity of 200 and a permeance of 10 m³(STP)/(m²·h·bar) especially for pure diffusion-based membrane. However, there are membranes which can already reach or are planned to reach these values separately. In addition, fixed site carrier (FSC) membranes, metal-organic framework (MOF) membranes, and mixed matrix membrane (MMM) are expected to go beyond the Robeson's upper bound [4] and reach at the same time high selectivities and high permeances [19,20].

A numerical model of the attainable region approach [11] is used in order to optimise and evaluate the membrane capture process for the wide range of membrane properties under consideration. The attainable region approach principle and the numerical algorithm used for optimisation and evaluation are explained below.

2.2.2.2. The attainable region approach.

A graphical methodology for systematic and consistent design of membrane processes for post-combustion capture has previously been developed and described in detail by SINTEF Energy Research [11–13]. In this approach, the membrane separation task is divided into several stages which include a membrane unit as well as its own rotating equipment (compressors, vacuum pump and expander) and intercoolers.

The design methodology is dependent on robust models for the membrane separator and the rotating equipment. A membrane model for binary components, after Saltonstall [8], is adopted for the present study. The model assumes a membrane unit in cross-flow configuration with plug flow on the feed side and no mixing with the bulk stream on the permeate side. These assumptions permit analytical solutions of all model equations, which is favourable in terms of robustness and computational speed. However, as with other binary component models, a disadvantage of this approach is that water vapour permeation cannot be directly modelled. For this reason, the cost and/or power consumption of drying units before the membrane capture are included in our

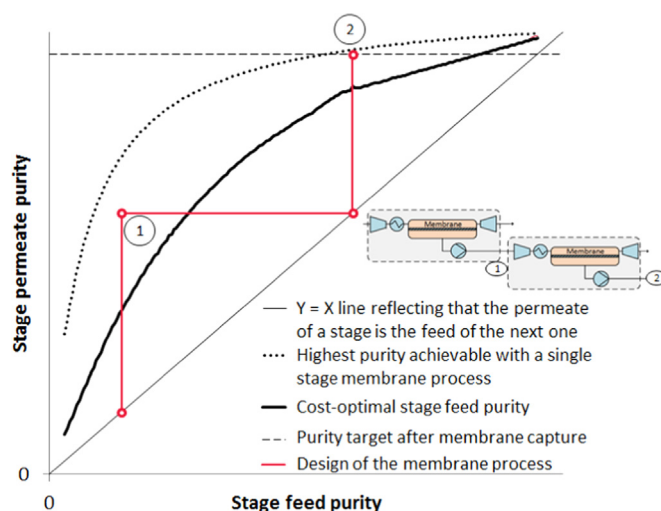


Fig. 2. Graphical representation of the attainable region approach.

analysis.

Rotating equipment is modelled as isentropic expansion or compressions of an ideal gas while the heat capacity ratio of the binary mixture is calculated from a linear regression as a function of CO₂ concentration². An isentropic efficiency of 80% is assumed, in order to take irreversibilities into account.

The membrane and rotating equipment modules, as well as a cost model that is described in Section 2.3, are used to calculate the technical and economic performance of each step. The resulting relevant permeate purities are presented as an attainable region diagram used to visualize, for a specific stage capture ratio³, the possible operating window of each membrane stage in addition to its optimal operating region, as shown in Fig. 2. In this graphical representation, the attainable region approach corresponds to the range of permeate purities between the single stage cost-optimal purity and the highest purity achievable with a single stage. The number of stages and operating points are then easily identified using a step-wise approach similar to the McCabe Thiele diagram, as shown in Fig. 2, and by comparing the costs of the various membrane configurations obtained. Once the cost-optimal design has been identified, the actual operating conditions (feed pressure, permeate pressure and area) are back-calculated from the targeted stage purity using the membrane model. An illustration of the methodology, including the results of a case design with its characteristics has previously been presented [12].

It is worth noting that this approach and the graphical solution generated is used to evaluate simple multi-stage configurations without advanced process features, such as retentate recycles or retentate heating before expansion.

2.2.2.3. The membrane process optimization.

A numerical version of the Attainable Region Approach presented above is used here in order to reduce the number of possible process designs. Reducing the number of possible process designs enables the cost-optimal membrane process configuration and design to be identified within a reasonable length of time and allows the CO₂ capture costs between the commercial MEA-based and membrane-based capture to be compared for a wide range of membrane properties.

In practice, the numerical model employing the algorithm

² Simulations in HYSYS have confirmed an accuracy of around $\pm 1\%$ for compressors, expanders and heat exchanger duties and around $\pm 5\%$ for the vacuum pump in the range of pressures evaluated.

³ This stage capture ratio is determined by considering the overall capture ratio to be attained and the approximate number of stages involved.

¹ Permeances of 5.94 m³(STP)/(m²·h·bar) have been reported for the Polaris membranes.

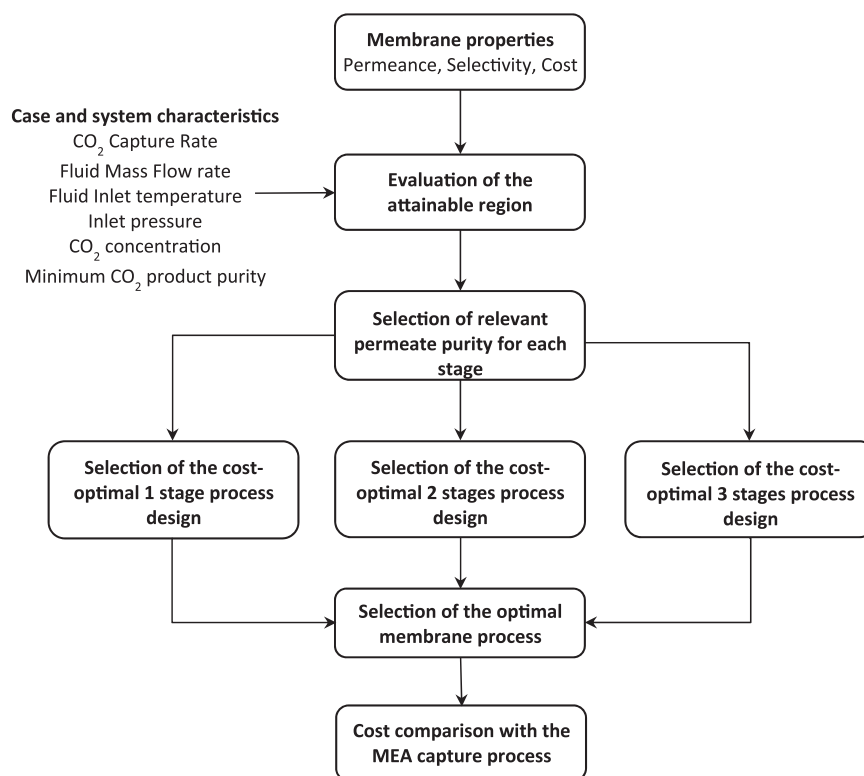


Fig. 3. Optimisation algorithm used to identify and evaluate cost-optimal membrane designs.

presented in Fig. 3 has been developed to optimise the membrane-based capture process with a configuration of up to three stages for given membrane properties. Based on the membrane properties and the system conditions being considered, the numerical model first generates the attainable region diagrams in order to select the ranges of stage feed and permeate purities relevant for each stage of the different multi-stage membrane process configurations. Based on the selected ranges of permeate purity, the cost optimal designs (membrane areas, operating pressures, permeate purities) of the one-, two- and three-stage configurations are identified and compared to select the overall cost-optimal membrane configuration and design. The overall cost-optimal membrane process and its technical and economic characteristics are recorded for comparison with the performances of the MEA-based process.

In theory, a total of around 21,000 membrane process designs are possible for a given membrane process that includes configurations of up to three stages and considering a precision of 1% on permeate and product purities of membrane systems, without taking into account the attainable region approach [21]. The attainable region as used here allows the number of possible membrane process designs to be reduced by a factor from 7 up to 1900 for the range of membrane permeance and selectivity being evaluated. This enables the numerical model to optimise and evaluate the set of membrane properties considered within a reasonable length of time.

2.2.3. MEA based CO₂ capture

The MEA solvent-based capture technology, as described in the EBTF report [22] and shown in Fig. 4, is here treated as the reference technology for CO₂ capture from the coal-fired power plant. In this process, the flue gas is fed to the absorber after being cooled and pumped, using blowers to overcome the pressure drops in the columns. In the absorber, the flue gas is put in contact with an MEA-based solution containing 30 wt% of MEA. After

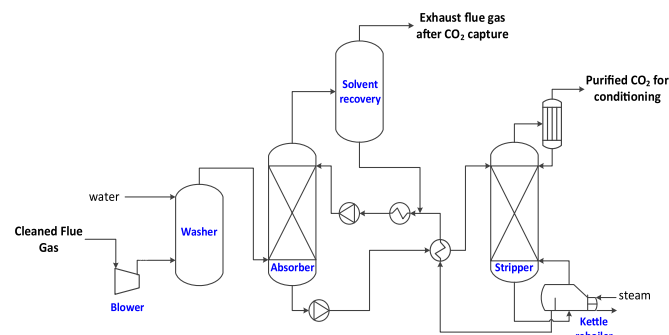


Fig. 4. Schematic process flow diagram of the MEA-based capture process [23,24].

absorption, the CO₂ is recovered at the bottom of the column, chemically bound to the solvent, while the flue gas passes through a wash section to balance water and recover solvent carried out as droplets or vapour. The "CO₂-rich" solvent is removed from the bottom of the absorber, pumped and enters a hot-cold heat exchanger to be preheated (to 120 °C) by the regenerated lean solvent, before entering the top of the stripper. Significant quantities of heat are required at the stripper reboiler to break the chemical bond between CO₂ and the solvent, and maintain regeneration conditions in the column. The vaporised water from the top of the column is recovered in the condenser and fed back to the column, while the purified CO₂ is sent through the conditioning process to reach the requirements for pipeline transport. The "lean" solvent recovered at the bottom of the column is pumped back to the top of the absorber through the hot-cold heat exchanger and a cooler used to reach lower solvent temperatures which enhance the absorption process.

It is worth noting that the heat required by this process is assumed to be extracted from the stream flow between the

intermediary and low pressure levels of the power plant, thus significantly affecting the overall performance of the plant.

2.3. Cost evaluation

Most studies evaluate and compare CO₂ capture technologies based on a Nth Of A Kind (NOAK) basis considering therefore the various technologies as mature and proven [13,17,25–28]. However these technologies are at different levels of both maturity and demonstration [3]. Moreover, although technologies such as the solvent based capture are already at an advanced level of development and at the first stages of demonstration, especially compared to other technologies, they are still not yet at an Nth Of A Kind level of cost.

As the MEA- and membrane-based CO₂ capture technologies are at different levels of maturity and as uncertainties remain regarding membrane cost, six cases detailed in Table 1 are considered in this work. These cases cover various combinations of maturity-level scenarios and membrane-module costs.

Two maturity-level scenarios are considered:

- The first scenario regards both technologies as mature and demonstrated thus to be evaluated on an Nth Of A Kind basis;
- The second aims to include costs that are representative of demonstration projects, and therefore treats both technologies at their current level of maturity, which is First Of A Kind (FOAK) for the membrane-based process and Second Of A Kind (SOAK) for the MEA-based process.

Regarding membrane module costs, the literature often considers a unitary cost of 50 \$/m² independently of the membrane's properties, and does not take the initial development cost into account. However the unitary membrane cost can be expected to be linked to the properties and performance of the specific membrane involved. Therefore, in addition to cases reflecting the additional costs associated with FOAK processes, a set of cases (from case 3 to 6) evaluates the impact of the membrane module cost on the competitiveness of the membrane compared to the MEA process. In this set of cases, the unit membrane cost is increased by a factor up to two to represent the cost of high performance membranes.

The specific data considered in each case are presented in the cost modelling section.

As each of these cases involves specific cost data, Section 2.3.1 presents the methodology used for the first case, treating this as a base case, in which both technologies are regarded as mature and demonstrated, and the membrane module cost is assumed to be 50 \$/m², while Section 2.3.2 explains how the cost data are modified to represent cases in which the membrane-based capture is treated as First Of A Kind and the MEA-based capture as Second Of A Kind.

2.3.1. Cost data of the base case

The base case assumes costs of a NOAK (Nth Of A Kind) plant to be

Table 1
Characteristics of the cases considered.

Case characteristics	Case 1	Case 2	Case 3	Case 4	Case 5	Case 6
Membrane maturity level considered	NOAK	FOAK	NOAK	FOAK	NOAK	FOAK
MEA maturity level considered	NOAK	SOAK	NOAK	SOAK	NOAK	SOAK
Membrane module cost (\$/m ²)	50	50	75	75	100	100

built at some time in the future, when all technologies are mature. Such estimates reflect the expected benefits of technological learning, but they do not adequately take into account the greater costs that typically occur in the early stages of commercialisation [29].

While the costs of the power plant with and without MEA-based CO₂ capture are extracted from the EBTF report, a cost methodology has been developed to optimize and assess the membrane-based CO₂ capture process. In order to enable a fair comparison of both capture technologies to be made, the costing methodology adopts a Bottom Up Approach developed to be consistent with the EBTF as presented below.

Investment and operating costs are given in 2014 Euro prices. As the costs available in the EBTF report are for 2008 price levels, the investments of the power plant have been updated according to the European Power Capital Costs Index (EPCCI), excluding nuclear power⁴ [30], while the investment costs of the MEA capture plant are updated according to the Chemical Engineering Plant Cost Index (CEPCI) [31]. The utilities costs are corrected according to an average yearly inflation of 1.7% [32].

2.3.1.1. Investment costs. A factor estimation method is used to estimate investment costs of the process equipment, where the direct costs estimated for each equipment are multiplied by indirect⁵ cost factors to obtain the investment costs.

While the costs of the power plant with and without MEA capture as well as the conditioning process are extracted from the EBTF and reported in Table 2, a direct costs function (€₂₀₁₄) of carbon steel equipment has been regressed using the Aspen Process Economic Analyzer[®] v7.2 (see Table 3), based on simulations performed using the membrane numerical model. However, due to their specificity, the CO₂ membrane module and framework are estimated in a different way. The membrane module is estimated considering a membrane unitary cost of 40 €₂₀₁₄/m² based on the cost update and currency conversion of the 50 \$₂₀₁₀/m² cost adopted by Zhai and Rubin [28]⁶. The membrane framework is based on the cost function suggested by van der Sluijs et al. [33] for the framework of the membrane separation system in an ammonia plant of DSM and modified by Roussanaly et al. [13] to take the influence of the design pressure of the module into account, as shown in Eq. (1) and Table 4⁷.

$$\text{Direct cost}_{\text{membrane framework}} = \left(\frac{\text{Membrane module area}}{2000} \right)^{0.7}$$

$$\text{Reference module cost} = \left(\frac{\text{Membrane module pressure}}{55} \right)^{0.875} \quad (1)$$

The investment cost of a given item of equipment is then calculated by multiplying the component's specific direct cost by the indirect cost factor (see Table 5). The total investment cost in €₂₀₁₄ is then determined by summing the estimated investment cost for all components within defined system boundaries (Eq. (2)).

$$\text{Total investment cost} = \sum (\text{Direct cost} \cdot \text{Indirect cost factor}) \quad (2)$$

⁴ The EPCCI tracks and forecasts the costs associated with the construction of a portfolio of power generation plants in Europe, and as such, is an indicator of the market price of the power plants.

⁵ Which includes the costs associated with engineering, commissioning, administration, and contingencies.

⁶ As there is no membrane module commercially available on a large scale basis, it is not possible to obtain an industrial based reference for the membrane module cost. In addition, the membrane module cost can be expected can be linked to the membrane module characteristics (selectivity, permeance, lifetime...).

⁷ It is worth noting that a limit of 25,000 m² of membrane area per module is considered in order to avoid having unrealistically large modules.

Table 2
Cost of the power plant without and with MEA capture [17].

Type of cost	Power plant without capture	Power plant with MEA capture
Plant direct cost (M€ ₂₀₁₄)	1312	1504
Plant indirect cost (M€ ₂₀₁₄)	1509	1730
Fuel cost (M€ ₂₀₁₄ /y)	147.5	147.5
Fixed operating costs (M€ ₂₀₁₄ /y)	28	47.7
Variable operating costs (M€ ₂₀₁₄ /y)	10	22.2
Levelized cost of electricity (€/MWh)	63.3	100.8
CO ₂ capture cost (€ ₂₀₁₄ /t _{CO2} avoided)	–	56.9

Table 3
Direct cost of rotating equipment and heat exchanger equipment costs based on regressions of Aspen Process Economic Analyzer^{®a}.

Type of equipment	Unitary cost	Unit
Compressor (First stage)	920	€/kW
Compressor (Second stage)	510	€/kW
Compressor (Third stage)	370	€/kW
Expander	570	€/kW
Vacuum pump	800	€/kW
Cooler	370	€/m ²

^a The direct costs of rotating equipment and heat exchanger include a 5% process contingency.

Table 4
Direct cost of the membrane framework.

Type of equipment	Unitary cost	Unit	Reference
Reference module area	2000	m ²	[33]
Reference pressure	55	bar	[33]
Reference module cost	286	k€ ₂₀₁₄	[33]

2.3.1.2. Maintenance and operating costs. The fixed operating costs depend on the investment cost, and cover replacement of materials, maintenance, insurance, and labour costs. To be consistent with the EBTF report, the annual fixed operating cost is set to 28 M €₂₀₁₄ for the power plant and 7% of the Engineering, Procurement, Construction (EPC) costs for the CO₂ capture processes [17]. In addition, an annual replacement of the membrane modules of 20% is also included [34,35] with a replacement cost of 10 \$₂₀₁₀/m² cost suggested by Zhai and Rubin [28]⁸.

The variable operating costs are a function of the amount of electricity produced, and covers consumption of utilities: bituminous coal, process and clean waters, ash disposal, and limestone. While the variable operating costs of the power plant and the MEA capture unit are extracted from the EBTF report and updated in accordance with inflation, the annual variable operating costs of the membrane system are estimated using the consumption of utilities obtained from the process design. The list of utility and consumable unit costs used in the evaluation of the power plant with and without CO₂ capture is shown in Table 6.

2.3.2. Cost data representative of demonstration projects

Estimating cost data that are representative of demonstration projects has been and is still a challenging task. The increased

⁸ Zhai and Rubin suggested a replacement cost five times lower than the module investment cost.

Table 5
Indirect cost factors [17].

Indirect cost factor item	Percentage of direct cost (%)
Yard improvement	1.5
Service facilities	2
Engineering/consistency cost	4.5
Building	4
Miscellaneous	2
Owner costs	7
Project contingencies	10
Total Indirect Cost Factor	31%

Table 6
Utility costs [17].

Utilities	Reference costs	Cost Units
Bituminous coal	3.4	€ ₂₀₁₄ /GJ
Clean water	6.8	€ ₂₀₁₄ /m ³
Sea water cooling	0.39	€ ₂₀₁₄ /m ³
Ash disposal	36	€ ₂₀₁₄ /t
Limestone	40.5	€ ₂₀₁₄ /t

investment costs associated with demonstration project are here performed here in accordance with the National Energy Technology Laboratory (NETL) cost estimation guidelines [36,37]. In this approach the process and project contingencies of the CO₂ capture system are updated on the basis of Technology Readiness Level (TRL) and the demonstration level of the technology involved. Membrane-based CO₂ capture is regarded as being at TRL6 [3] and as not yet demonstrated on a large scale basis, although a few small pilot plants have been tested [38,39]. This leads to process and project contingencies of respectively 25 and 30% TDC (Total Discounted Cost) for the membrane process. The MEA-based CO₂ capture is reckoned to be at TRL9 [3], and that only one large-scale demonstration project, the Boundary Dam project, has been built and is operating [40]. The process and project contingencies associated with the MEA-based process are estimated at 15 and 15% TDC. In addition to these increased contingency costs, a lower availability of the power plant can be expected in retrofit cases due to the integration of the capture unit with the steam cycle as seen in the Boundary dam project [41]. However as no detailed data on availability decrease has been reported, this effect is not considered in the current work.

Regarding maintenance and operating costs, annual process maintenance is not regarded as significantly different from to the NOAK case. However, the membrane modules of the first membrane-based CO₂ capture demonstration project can be expected to have a shorter lifetime towards the beginning of the project than when the technology is mature and demonstrated. A membrane lifetime of three years is therefore considered for the first 10 years of a demonstration project⁹, while a five years lifetime estimate, as in the NOAK case, is subsequently employed. Finally, additional operating costs can also be expected in the first years of operation, especially for a demonstration project due to learning and training time, inefficiency, and so on. Utility consumption is therefore assumed to be 15% higher than the basis during the first three years of operation of the demonstration project for both membrane- and MEA-based CO₂ capture.

A summary of the reference cost data for the NOAK case (mature and demonstration costs) and the updated cost data

⁹ This hypothesis is selected to reflect the lower lifetime which can be expected in demonstration projects, however it is important to note that this value is hard to estimate in practice and can be expected to be specific to the membrane considered.

Table 7
Summary of cost-data updates for demonstration projects and reference data for NOAK cases [36,37].

	Mature and demonstrated costs	Demonstration costs
Membrane capture process contingency (%TDC)	5	25
Membrane capture project contingency (%TDC)	10	30
Membrane capture annual fixed OPEX (%TDC/y)	7	8.5
Annual membrane replacement during the first 10 years (%)	20	33
Annual membrane replacement after 10 years (%)	20	20
Membrane capture increased utilities consumption (%)	–	15
Membrane capture increased utilities consumption period (y)	–	3
MEA capture process contingency (%TDC)	–	15
MEA capture project contingency (%TDC)	10	15
MEA capture annual fixed OPEX (%TDC/y)	7	8.5
MEA capture increased utilities consumption (%)	–	15
MEA capture increased utilities consumption period (y)	–	3

considered for demonstration projects is presented in Table 7.

2.4. Capture technology benchmarking

Two Key Performance Indicators (KPI) are assessed here and used to compare the two capture technologies: the Levelised Cost Of Electricity (LCOE) [17] and the CO₂ avoided cost [42].

The levelised cost of electricity [€/MWh] measures the unit cost of electricity generation of a plant with and without CO₂ capture, and approximates the average discounted electricity price over the project duration that would be required as income to match the net present value of the capital and operating costs of the project. It is equal to the annualised costs divided by the annualised net electricity production, as shown in Eq. (2). The LCOE is calculated assuming a real discount rate of 8%¹⁰ and an economic lifetime of 25 years [17]. In addition, investment costs consider that construction is shared over a four-year construction period [17].

Levelised cost of electricity

$$= \frac{\text{Annualized investment} + \text{Annual OPEX}}{\text{Annual net power output}} \quad (2)$$

A second important KPI is the CO₂ avoided cost [€/tCO₂], which is obtained by comparing the levelised cost and the CO₂ emission rate to the atmosphere of the plant with and without CO₂ capture, as shown in Eq. (3). The CO₂ avoided cost approximates the average discounted CO₂ tax or quota over the duration of the project that would be required as income to match the net present value of additional capital and operating costs due to the CCS infrastructure. The CO₂ avoided cost is used as cost performance indicator to compare the membrane- and MEA-based captures.

$$\text{CO}_2 \text{ avoided cost} = \frac{(\text{LCOE})_{\text{CCS}} - (\text{LCOE})_{\text{ref}}}{(t_{\text{CO}_2}/\text{MWh})_{\text{ref}} - (t_{\text{CO}_2}/\text{MWh})_{\text{CCS}}} \quad (3)$$

where

- (LCOE)_{CCS} is the levelised cost of electricity of produced by the plant with CCS [€/MWh].
- (LCOE)_{ref} is the levelised cost of electricity of the reference plant without CCS [€/MWh].
- (t_{CO₂}/MWh)_{CCS} is the CO₂ emission rate to the atmosphere of the plant with CCS [t_{CO₂}/MWh].}}
- (t_{CO₂}/MWh)_{ref} is the CO₂ emission rate to the atmosphere of the reference plant without CCS [t_{CO₂}/MWh].}}

¹⁰ This real discount rate of 8 % corresponds to a nominal discount rate around 10% if an inflation rate of 2% is considered.

3. Results

3.1. Membrane properties required for post-combustion CO₂ capture at coal-fired power plants

The following sections discuss the evaluation of the different cases: to identify the membrane properties required to ensure cost-competitive membrane-based capture when considering both capture technologies as mature and demonstrated, to illustrate the influence of demonstration costs and membrane module cost on the competitiveness of membrane-based CO₂ capture, and finally to identify the optimal range of membrane properties.

3.1.1. The Nth Of A Kind case

The results of the cost comparison of the membrane- and MEA-based post-combustion CO₂ captures from a coal-fired power plant are presented in Fig. 5 for the base case, in which both capture technologies are considered to be mature and demonstrated, and a membrane module cost of 50 \$/m² is assumed.

In order for the results to be easily understandable by the reader and considering the high number of cases, a graphical representation of each case is used to visualise the results of the performance comparison. In this representation, the membrane selectivity and permeance are used as X- and Y-axes, as shown in Fig. 5. The relative cost efficiency of the membrane process compared to the MEA-based process for CO₂ capture from the power plant flue gas is represented by differently coloured areas.

The green area corresponds to the range of membrane properties that would lead to a membrane process with up to three stages that is cheaper than MEA-based capture. The blue areas correspond to the conditions in which more advanced configurations (with for example retentate recycle, sweep, counter-current flow pattern, etc.), which could lower the CO₂ capture costs by up to 25% [8,28]¹¹ compared to simple configurations, are required to compete with MEA-based CO₂ capture. The light blue colour represents conditions in which advanced membrane processes with CO₂ avoided costs that are between 0% and 12.5% lower than simple membrane processes, which could be achieved with the use of retentate recycles [43], are required to compete with the reference capture technology. On the other hand, the dark blue

¹¹ Merkel et al. and Zhai and Rubin evaluated the benefits of advanced configurations to a CO₂ avoided cost decrease of 30–35%, however similarly, more advanced configuration of the MEA-based capture process (EGR, flexible solvent capture) can also be used to lower the cost of MEA capture. In addition, these cost evaluations did not include the cost associated with the modification of the pulverised coal-fired boiler of the power plant which can significantly impair the cost benefit of an air sweep configuration in a retrofit case. It is therefore likely that considering a cost benefit from advanced configurations 25% higher for membrane-based capture than for MEA-based capture is a rather optimistic hypothesis.

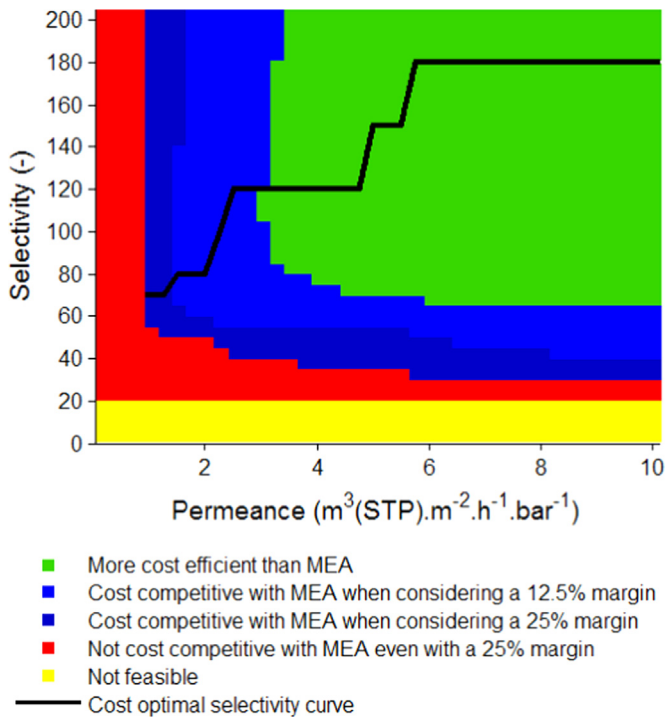


Fig. 5. Membrane properties required for cost-competitive membrane CO₂ capture considering both capture technologies as mature and demonstrated, and a membrane module cost of 50 \$/m².

area corresponds to conditions in which membrane processes need to be between 12.5% and 25% cheaper than simple configurations, requiring the use of retentate recycles and additional features (sweep, counter-current flow pattern [8], exhaust gas recirculation [44], etc.), to compete with the MEA based concept. Finally, the red area corresponds to the conditions in which membrane processes, even with advanced configurations, cannot compete with MEA capture. Finally, the yellow area corresponds to the condition in which the membrane processes cannot reach the CO₂ purity and capture ratio requirements.

The results show that when the cost of mature and developed membrane technology and a membrane cost of 50 \$/m² are considered, CO₂ capture based on simple multi-stage membrane processes can directly compete with MEA-based capture for a wide range of membrane properties. However, to reach this competitiveness with simple configurations, a range of combinations of membrane selectivity and permeance with especially a "vertical" lower limit and a "horizontal" lower limit is required as shown in Fig. 5. The membrane permeance and selectivity need to be at least superior to 3 m³(STP)/(m²·h·bar) with high selectivities (higher than 105) for the "vertical" limit or superior to 65 with high permeance (higher than 6 m³(STP)/(m²·h·bar)) for the "horizontal" limit. The range of membrane properties which can compete with MEA-based capture can however be increased by considering more advanced membrane process configurations (with for example recycle, sweep, counter-current flow pattern, etc.) as shown by the blue areas in Fig. 5. Indeed with these more advanced configurations, membrane processes with permeances as low as 1 m³(STP)/(m²·h·bar) with high selectivities (higher than 55), or selectivities as low as 30 with high permeances (higher than 5.75 m³(STP)/(m²·h·bar)) could compete with MEA-based capture. However it is important to note that by using advanced process configurations, membranes in the green area would also be able to lower their CO₂ capture cost and be even more cost competitive than MEA-based CO₂ capture.

It is worth noting that the "vertical" lines limiting the blue and

green areas are inclined clockwise. This means, as shown by Zhai and Rubin [45], that a higher selectivity does not always lead to a lower CO₂ capture cost of the membrane process. The reason for this trend is that after a certain point, for a given permeance, an increase in selectivity leads to an increase in compression costs of the process which defeat the cost benefit of a lower membrane area associated with higher selectivity membrane. This demonstrates the existence of a cost-optimal membrane selectivity for each permeance value, as represented by a black line in Fig. 5. Furthermore, the proximity of the horizontal lines limiting the blue and green areas shows that an increase in selectivity leads to a significant decrease in process costs, especially for low permeance, when the selectivities considered are below the optimal selectivity curve. However, for selectivity above the optimum, an increase in selectivity appears to have a more limited negative impact on the cost performance of the membrane process. However, the permeance appears to follow an opposite trend. Indeed, below the optimal selectivity curve, an increase in permeance seems to have a rather limited cost benefit for the membrane process, especially for low selectivity. On the other hand, the relative proximity of the vertical lines limiting the blue and green areas above the optimal selectivity curve shows that an increase in selectivity has a significant positive impact on the membrane process cost.

When considering the results of the base case, it is however to keep in mind that by the time membrane-based CO₂ capture becomes mature and demonstrated, improved solvents that are more energetic and cost-effective than MEA will probably be available and demonstrated. Solvent capture based on these improved solvent will therefore be regarded as the reference for solvent-based CO₂ capture technology. This would mean in practice that at least the left and lower parts of the blue area in Fig. 5 will not be able to compete with improved solvent-based capture, shifting the blue and green areas higher up and further right on the chart.

3.1.2. The influence of technology maturity and membrane module cost

The results of the cost comparison of the membrane- and MEA-based post-combustion CO₂ capture processes are presented in Fig. 6(a) to (f) for all the six cases described in Section 2.3 to quantify the influence of technology maturity and membrane module cost on the membrane properties required.

As discussed previously, while the unitary membrane cost can be expected to be linked to the properties and performances of the individual membranes, the literature often take into account a unitary cost of 50 \$/m² independently of the membrane properties and does not include the initial development cost. The influence of the membrane module cost on the competitiveness of the membrane-based capture process has been evaluated and is presented in Fig. 6(a) to (c). The assessment shows that higher module costs reduce the competitiveness of the membrane process, especially for demonstration projects. Higher membrane module costs in particular reduce the size of the green area, indicating that a simple membrane process configuration will probably not be efficient enough to compete in the case of higher module costs, and that more advanced configurations (with for example recycle, sweep, counter-current flow pattern, etc.) will be required. Indeed, to reach competitiveness with a simple configurations with a membrane module cost of 100 \$/m², the membrane permeance and selectivity will need to be at least superior to 6 m³(STP)/(m²·h·bar) with high selectivities (higher than 95) or superior to 70 with high permeance (higher than 8.5 m³(STP)/(m²·h·bar)), while membranes with permeances of at least 1.75 m³(STP)/(m²·h·bar) with high selectivities (higher than 55), or selectivities as low as 35 with high permeances (higher than 7 m³(STP)/(m²·h·bar)) could compete in advanced membrane configurations. In addition, the assessment also shows that higher

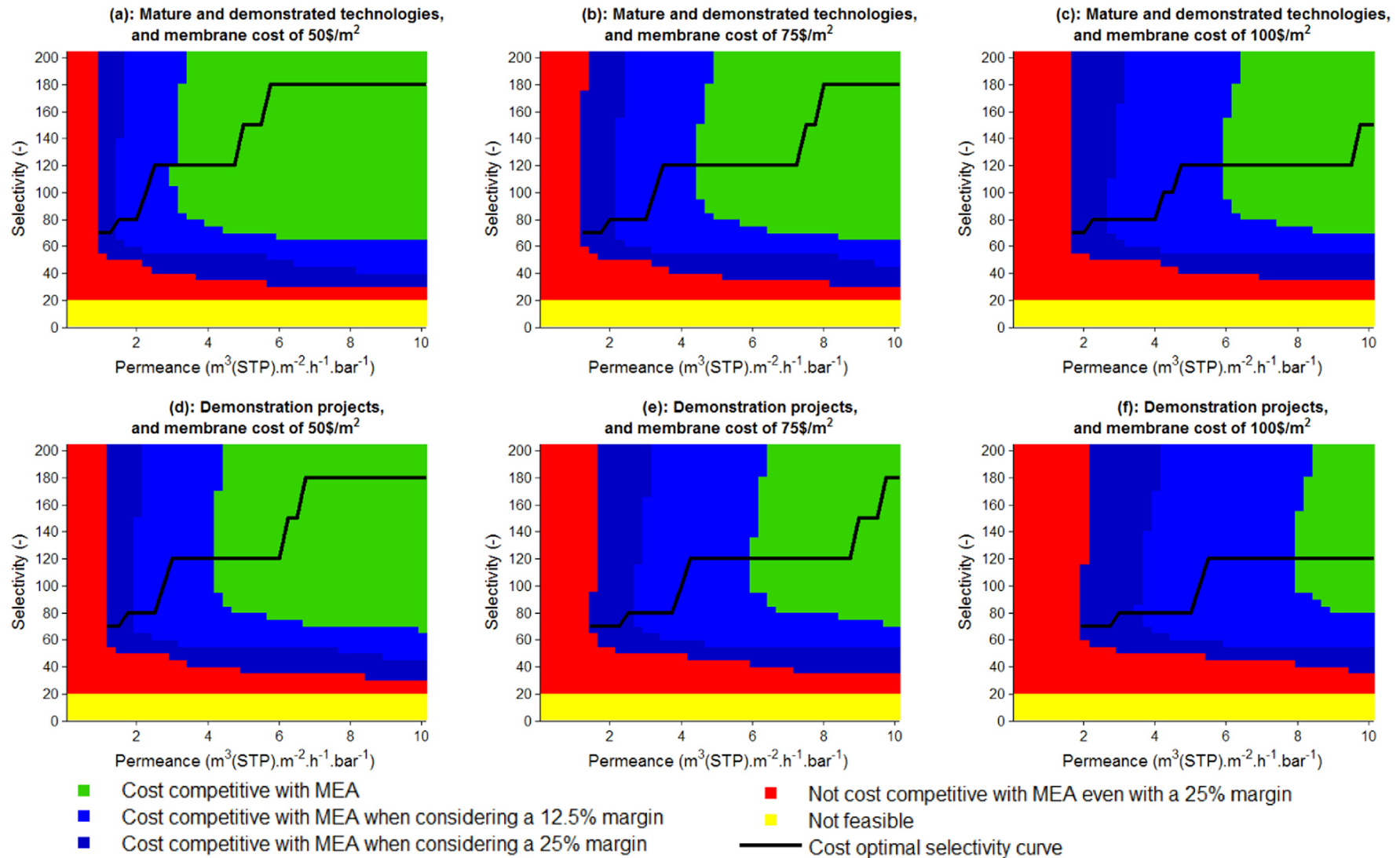


Fig. 6. Membrane properties required for cost-competitive membrane CO₂ capture considering (a) both capture technologies as mature and demonstrated and a membrane module cost of 50 \$/m² (b) both capture technologies as mature and demonstrated and a membrane module cost of 75 \$/m² (c) both capture technologies as mature and demonstrated and a membrane module cost of 100 \$/m² (d) a demonstration project with different levels of maturity and a membrane module cost of 50 \$/m² (e) demonstration project with different levels of maturity and a membrane module cost of 75 \$/m² (f) demonstration project with different levels of maturity and a membrane module cost of 100 \$/m².

membrane module cost will lower the selectivity values of the cost-optimal selectivity curve. Finally, higher membrane module costs also reduce the potential cost benefit of using a membrane-based capture process compared to a MEA-based capture.

As discussed above, membrane- and MEA-based CO₂ capture lie at different levels of maturity and demonstration, and the competitiveness of the membrane process can therefore be significantly affected at the demonstration project stage. Comparisons of Fig. 6(a–c) to (d–f) show that when the additional costs of demonstration projects presented in Section 2.3.2 are included, the range of membrane properties which can be cost-competitive with MEA capture is reduced, due to the lower maturity and demonstration levels of membrane-based CO₂ capture, which significantly increase investment and operating costs. Indeed, when including the cost of demonstration projects, the limits between blue and red areas shift in the direction of higher permeances and selectivities. Even though all blue and green areas shift to higher permeances and selectivities, it is important to note that the demonstration costs do not influence all areas in the same way. While both dark and light blue areas often extend over a wider range of membrane selectivities and permeances, the green area shrinks significantly. Indeed, to reach competitiveness with simple configurations for demonstration costs, membrane permeance and selectivity need to be at least superior to 4.25 m³(STP)/(m² · h · bar) with high selectivities (higher than 95) or 65 with high permeance (higher than 10 m³(STP)/(m² · h · bar)), while membranes process with permeances of 1.25 m³(STP)/(m² · h · bar) and high selectivities (higher than 55), or with selectivities of 30 and high permeances (higher than 8.5 m³(STP)/(m² · h · bar)) could compete in the case of advanced membrane configurations. This especially emphasizes the necessity of advanced membrane configurations in the case of demonstration projects to decrease the cost of membrane-based processes to attain cost-competitive capture despite the lower maturity and demonstration levels of these processes. As for the membrane module cost, the additional costs associated with demonstration projects also decrease the value of the cost-optimal selectivity curve.

Nevertheless, it is still important to bear in mind that the values presented in Fig. 6(a) to (f) do not take into account potential financial support from public funding bodies to help the development and demonstration of membrane-based CO₂ capture. Indeed, in order to support the development of a technology which can be expected to lower the cost of CO₂ capture and clean electricity, financial support for demonstration project may be expected to lower the additional costs of demonstration. In practice, this would help to limit the viability shifts observed in demonstration projects and in the case of higher membrane module costs, and would therefore enable a wider range of membrane properties to be cost-competitive once financial support is included, as well as increasing the benefits of using the membrane-based process for CO₂ capture in both the short and long run.

4. Discussions

4.1. The upper bound and suitability of existing membranes, membranes under development, and polymeric materials

While the results presented in Section 3 identify the membrane properties required for membrane-based process to compete with MEA-based technology for post-combustion CO₂ capture from a coal power plant in different maturity and module cost cases, it is important to put these results in the context of constraints on material properties, the properties of existing and under development membranes properties, as well as in the context of polymeric materials which could be used to build membrane modules.

Table 8

Example of characteristics of membrane module existing or under development.

Material of the membrane module	Selectivity (dimensionless)	Permeance (m ³ (STP)/(m ² · h · bar))	Reference
Polaris	50	5.94	[8]
PAAM-PVA/PS	80	0.14	[7]
PVAm/PVA	145	1.26	[7]
PDMA/PS	53	0.18	[7]
PDMAMA	80	0.03	[7]
PVAm/on PSF support	200	1	[10]
PVAm/PVA blend	174	0.58	[9]
Fixed Site Carrier	135	2	[20]

For example, the range of membrane properties used in the evaluation comprises selectivities of up to 200 and permeances up to 10 m³(STP)/(m² · h · bar) but does not consider whether a membrane module with both a selectivity of 200 and a permeance of 10 m³(STP)/(m² · h · bar) can be developed. Indeed, Robeson has demonstrated via the upper bound approach that, for diffusion-based membranes, there is a maximum selectivity which can be obtained for a given membrane CO₂ permeability [4]. Therefore, while membranes with a selectivity of 200 and low-medium permeances and membranes with a permeance of 10 m³(STP)/(m² · h · bar) and low-medium selectivities can be theoretically be achieved, the CO₂/N₂ upper-bound limitation shows that a membrane with both a selectivity of 200 and a permeance of 10 m³(STP)/(m² · h · bar) cannot be obtained without unreasonable considerations on the membrane film thickness. In order to take into account this limitation in the results of this paper, the Robeson upper bound curve is included in the assessment results presented in Fig. 7(a) to (f). However, as the upper-bound limitation is based on permeability which is a material characteristic, rather than the permeance¹², which is a membrane module characteristic, four film thicknesses ranging from 50 nm to 1 μm (50 nm, 100 nm, 500 nm and 1 μm) are used to generate Robeson upper bound linking selectivity and permeance [46,47]¹³. The inclusion of the upper bound in the graphical visualization of the results show that the range of membrane properties capable of competing with MEA-based capture, with or without advanced membrane processes, can be significantly impaired by the upper-bound limitation if very thin membrane cannot be used. The upper bound in particular narrows down the achievable green area, which represent conditions in which simple membrane configuration are cost-competitive with MEA-based capture and conditions in which advanced membrane configurations would be significantly more cost-effective than the reference solvent-based CO₂ capture. Therefore, the capacity to generate thin membrane film layers in the membrane module will be important to avoid reducing, especially in cases that involve demonstration and/or higher module costs, the range of membrane properties which are at the same time achievable and interesting in term of cost performances.

In addition to the upper-bound limitation, it is important to look at the suitability of membrane modules currently under development as well as existing materials that could be used for the development of polymeric membranes. Therefore, the characteristics of eight membrane modules at different stages of development, presented in Table 8 [7–9], and 276 polymeric materials that can be used for membrane development [5,6], are also plotted in

¹² Defined as the permeability over the film thickness.

¹³ While a membrane thickness of 1 μm can be considered as a rather easy achievable thickness target, a membrane thickness of 50 nm corresponds to the targeted thickness by Research and Development.

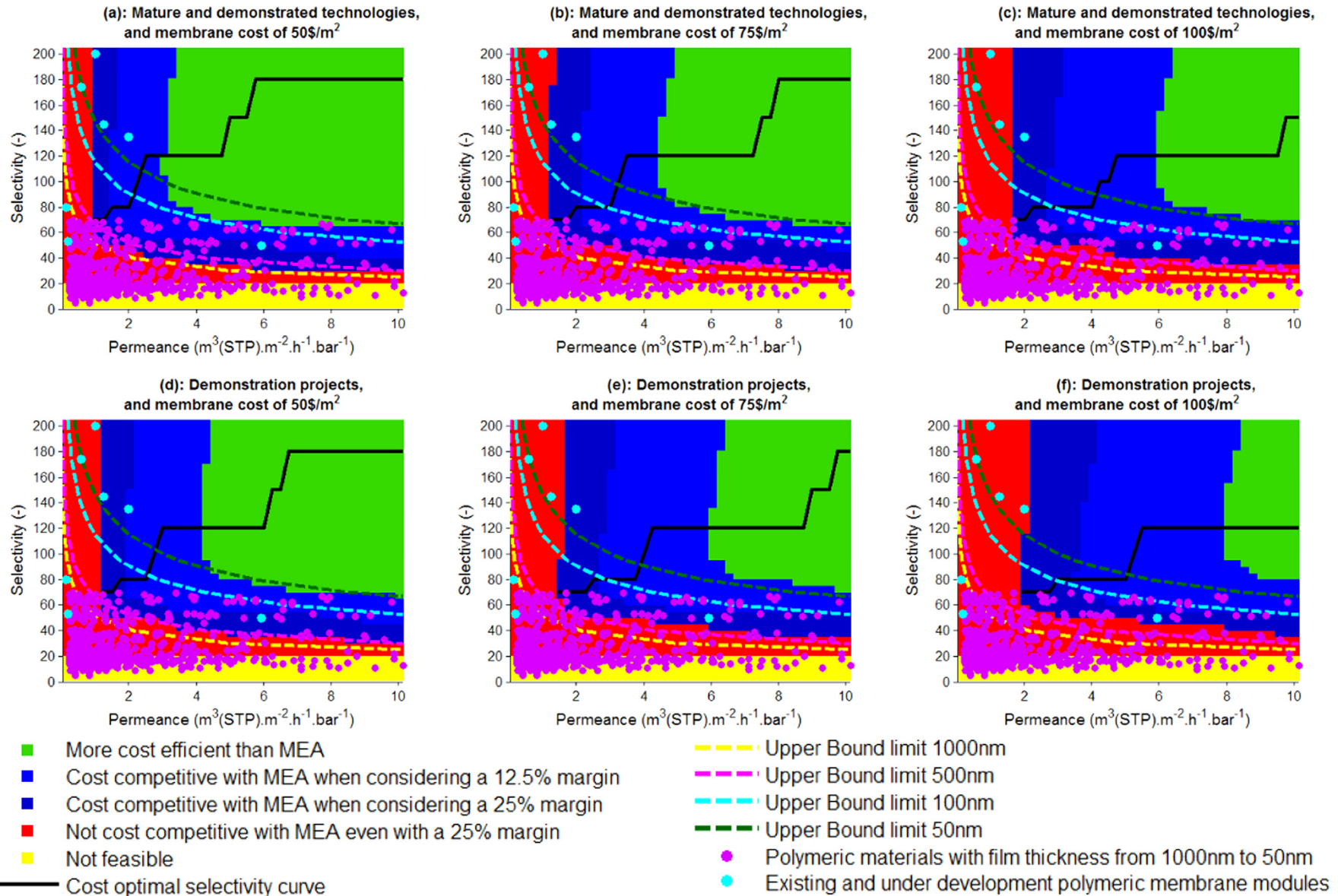


Fig. 7. Membrane properties required for cost-competitive membrane CO₂ capture, including the upper bound limitation and membrane data considering (a) both capture technologies as mature and demonstrated and a membrane module cost of 50 \$/m² (b) both capture technologies as mature and demonstrated and a membrane module cost of 75 \$/m² (c) both capture technologies as mature and demonstrated and a membrane module cost of 100 \$/m² (d) a demonstration project with different levels of maturity and a membrane module cost of 50 \$/m² (e) demonstration project with different levels of maturity and a membrane module cost of 75 \$/m² (f) demonstration project with different levels of maturity and a membrane module cost of 100 \$/m².

Fig. 7(a) to (f). As in the case of the upper bound, membrane film thicknesses of 50 nm to 1 μm are evaluated in order to obtain the range and membrane permeance which could theoretically be obtained from these polymeric materials.

The comparison shows that while most membrane modules both existing or under development need to boost their permeances, the Polaris membrane can, even in demonstration projects and at higher module costs, be a cost-competitive option using advanced processes. However, the cost performance of a process based on the Polaris membrane could be improved if further material development could increase its permeance and especially its selectivity. Furthermore, the comparison also shows that the Fixed Site Carrier membrane developed by NTNU [20] also seems to be capable of competing with MEA-based capture when used as part of advanced membrane configurations even in demonstration projects and at higher module costs. However, it is important to note that the membrane developed by NTNU considers rather conservative permeance values compared to what can be achieved with the material used [48] and if the module permeance can be improved to reach $3.25 \text{ m}^3(\text{STP})/(\text{m}^2 \cdot \text{h} \cdot \text{bar})$, even simple configurations could be competitive in the case of demonstrated technologies and membrane costs of $50 \text{ \$/m}^2$. However, in any case, demonstration will require financial support to reduce the additional costs associated with demonstration of the technology, and enable cost-competitive membrane CO_2 capture in the long run.

Finally, among the 276 membrane materials that can be used for membrane development [5,6], the comparisons show that while most of these materials will not lead to a membrane module that would be competitive with MEA-based capture, 56 of them can do so using advanced membrane processes and under specific conditions, as shown in Fig. 7(a) to (f). The materials which could lead to competitive membrane modules are shown in Table 9. As discussed above, the material thickness is also an important parameter to obtain high performances for a given material, Table 9 therefore also includes the material thickness needed to reach the dark blue, light blue, and green areas in Fig. 7(a). The calculations show that a wide range of these polymeric materials lead to reasonable thickness requirement (from 50 nm to 1410 nm) to reach the dark blue area, in which advanced membrane process configurations could lead to a capture cost up to 12.5% lower than MEA-based capture, and that therefore at least a few of them could be implemented in practice. However, only 30 of these polymeric materials can, with thicknesses ranging from 50 nm to 420 nm, reach the light blue area, therefore achieving cost-efficiency up to 25% better than MEA-based capture in an advanced membrane process configurations. Finally, only four polymeric materials seem to be able to reach the green area, in which simple membrane configurations would be able to compete with MEA, while advanced membrane configurations could reduce the cost of CO_2 capture by at least 25% compared with MEA. However as the material layers would need to be at least thinner than 90 nm to reach the membrane properties required, this target will be rather difficult to reach in practice.

In addition, as the Polaris membrane is usually regarded as the state-of-the-art membrane module for post-combustion CO_2 capture, Table 9 also includes information on whether and under which conditions these materials can lead to a module which could outperform the Polaris membrane. The results shows that, at material thicknesses above 50 nm, 29 of the 276 polymeric materials considered could theoretically compete with the Polaris membrane. However, it is important to note that all the thicknesses considered might not be achievable by all membrane materials, for example, due to material strength considerations.

Therefore, if only thicknesses greater than 100 nm, 200 nm or 300 nm can be reached with such materials, only 19, 12 and 5 of these materials could respectively outperform the Polaris membrane, while none would be able to outperform it if a thickness below 400 nm cannot be obtained.

Finally, the comparison shows that a more limited number of these polymeric materials could lead to membrane-based processes that could compete with the reference capture technology in the case of demonstration projects or higher membrane module cost, emphasizing yet again the need for financial support for the first of demonstration projects.

4.2. Providing support for membrane development and decision-makers

The ranges of membrane properties presented in the results section identify the membrane properties that could lead to a membrane-based process that could be cost-competitive with MEA-based post-combustion CO_2 capture from a coal power plant as defined by the EBTF [17]. These results can be used by different actors to support the development of cost-competitive membrane-based CO_2 capture.

First of all, the results can be used by materials experts and membrane developers to identify membranes that could be used to develop processes which can be cost competitive with MEA-based capture. For example, the results can be used by membrane developers to identify materials which might directly be good options, but also materials capable of being used as a good starting point for further improvement. The results can also be used as a guide to identify the best ways of improving a specific material in order to reach higher process performance, for example by focusing on improving selectivity rather than permeance in certain cases. Finally, the results can help to determine whether better shaping of a specific membrane material, e.g. reduced material thickness, can reasonably lead to an improved and competitive process.

The results presented here can also be used by industry to identify and select membrane module properties which can be cost-competitive with MEA-based post-combustion CO_2 capture from a coal power plant both in the longer run, when the membrane based CO_2 capture technology is mature and has been demonstrated, and in a demonstration project case in which financial support will probably be required. Industrial players can also use the graphical results to identify the different trade-offs between membrane properties and membrane module cost, in order to select the most cost-effective of the available options.

Finally, these results can be used by funding bodies to help them to enable cost-competitive membrane-based CO_2 capture. Indeed, funding agencies could use the results to identify membrane materials and modules which have, in the long run, the potential to cut the cost of CO_2 capture compared to solvent-based capture but need financial support for the first stages of development and demonstration.

However, it is important to note that the results presented in this paper are specific to the application considered and that results would vary depending on the application characteristics, such as CO_2 content in the flue gas, presence and type of impurities, membrane performances degradation, capture ratio, maturity of the technology. The results therefore cannot simply be extrapolated to other applications than post-combustion CO_2 capture from a coal-fired power plant.

Table 9
Polymeric materials that could be used to build membrane modules capable of competing with MEA-based capture and outperforming Polaris membranes.

Type of membrane dense film	Membrane material	Selectivity (dimensionless)	Permeability ^a (Barrer)	Maximum thickness requirement (nm) to reach the different colored area and outperform Polaris membrane			
				Dark blue area	Light blue area	Green area	Polaris membrane
Poly(ethylene oxide)	EO/EM/AGE (80/20/2)	46	773	990	330	–	300
Poly(ethylene oxide)	EO/EM/AGE (77/23/2.3)	44	680	870	290	–	260
Poly(ethylene oxide)	EO/EM/AGE (96/4/2.5)	48	580	1410	280	–	260
Incorporating polyimide	DMeCat–durene	63	31	90	60	–	50
Incorporating polyimide	TMeCat–MDA	30	110	50	–	–	–
Copolymers and polymer blend	MDI–BPA/PEG(80)	47	48	60	–	–	–
Copolymers and polymer blend	MDI–BPA/PEG(85)	49	59	140	–	–	–
Copolymers and polymer blend	L/TDI(20)–BPA/PEG(90)	51	47	110	–	–	–
Copolymers and polymer blend	L/TDI(40)–BPA/PEG(85)	48	35	80	–	–	–
Copolymers and polymer blend	IPA–ODA/PEO3(80)	53	58	180	70	–	60
Copolymers and polymer blend	BPDA–ODA/DABA/PEO2(80)	56	36	110	–	–	–
Copolymers and polymer blend	BPDA–ODA/PEO3(75)	52	75	180	–	–	–
Copolymers and polymer blend	BPDA–mDDS/PEO3(75)	53	72	220	90	–	80
Copolymers and polymer blend	BPDA–mPD/PEO4(80)	54	81	250	100	–	90
Copolymers and polymer blend	BPDA–ODA/PEO4(80)	51	117	280	50	–	50
Copolymers and polymer blend	PMDA–ODA/PEO2(75)	54	40	120	50	–	–
Copolymers and polymer blend	PMDA–mPD/PEO3(80)	50	99	240	–	–	–
Copolymers and polymer blend	PMDA–APPS/PEO3(80)	51	159	380	70	–	70
Copolymers and polymer blend	PMDA–APPS/PEO4(70)	53	136	420	170	–	150
Copolymers and polymer blend	PMDA–mPD/PEO4(80)	52	151	360	70	–	60
Copolymers and polymer blend	PMDA–ODA/PEO4(80)	52	167	400	80	–	70
Copolymers and polymer blend	PMDA–pDDS/PEO4(80)	49	238	570	110	–	100
Copolymers and polymer blend	PMDA/BTDA–BAFL (90:10)	34	130	90	–	–	–
Copolymers and polymer blend	NTDA–BDSA(30)/CARDO/ODA	41	70	80	–	–	–
Copolymers and polymer blend	NTDA–BDSA(30)/CARDO	36	164	120	–	–	–
Cross-linked membranes	DM14/MM9 (100/0)	68	45	140	80	–	80
Cross-linked membranes	DM14/MM9 (100/0)	38	107	120	–	–	–
Cross-linked membranes	DM14/MM9 (90/10)	69	62	190	120	–	110
Cross-linked membranes	DM14/MM9 (90/10)	39	133	150	–	–	–
Cross-linked membranes	DM14/MM9 (70/30)	66	96	300	190	–	160
Cross-linked membranes	DM14/MM9 (70/30)	36	195	140	–	–	–
Cross-linked membranes	DM14/MM9 (50/50)	64	144	450	280	60	240
Cross-linked membranes	DM14/MM9 (50/50)	36	260	190	–	–	–
Cross-linked membranes	DM14/MM9 (30/70)	63	210	650	410	90	350
Cross-linked membranes	DM14/MM9 (30/70)	33	350	260	–	–	–
Cross-linked membranes	DB30/MM9 (100/0)	63	93	290	180	–	150
Cross-linked membranes	DB30/MM9 (100/0)	35	200	150	–	–	–
Cross-linked membranes	DB30/MM9 (90/10)	64	105	320	200	–	170
Cross-linked membranes	DB30/MM9 (90/10)	36	210	150	–	–	–
Cross-linked membranes	DB30/MM9 (70/30)	67	141	440	280	60	230
Cross-linked membranes	DB30/MM9 (70/30)	35	270	200	–	–	–
Cross-linked membranes	DB30/MM9 (50/50)	62	179	560	300	–	260
Cross-linked membranes	DB30/MM9 (50/50)	34	330	240	–	–	–
Cross-linked membranes	DB30/MM9 (30/70)	60	250	780	420	–	360
Cross-linked membranes	DB30/MM9 (30/70)	33	410	300	–	–	–
Cross-linked membranes	DM9/MM9 (90/10)	68	18.3	50	–	–	–
Cross-linked membranes	DM9/MM9 (90/10)	38	51	50	–	–	–
Cross-linked membranes	DM23/MM9 (90/10)	66	145	450	280	60	240
Cross-linked membranes	DM23/MM9 (90/10)	38	290	330	90	–	80
Cross-linked membranes	DB69/MM9 (90/10) (cooling)	56	240	750	300	–	260
Cross-linked membranes	DB69/MM9 (90/10) (cooling)	36	510	380	–	–	–
Cross-linked membranes	DB69/MM9 (90/10) (heating)	62	98	300	160	–	140
Cross-linked membranes	DB69/MM9 (90/10) (heating)	35	400	300	–	–	–
Cross-linked membranes	DM14/MM23 (30/70) (cooling)	62	240	750	400	–	350
Cross-linked membranes	DM14/MM23 (30/70) (cooling)	35	420	310	–	–	–
Cross-linked membranes	DM14/MM23 (30/70) (heating)	62	250	780	420	–	360

^a In order to evaluate the corresponding permeance which is a module characteristic, the permeability which is a material property must be divided by the corresponding membrane film thickness.

5. Conclusions

This paper focuses on the identification of membrane properties required to enable cost-competitive post-combustion CO₂ capture from a coal-fired power plant using membrane-based processes. To this end, a numerical version of the attainable region approach proposed by Lindqvist et al. [11–13], built as part of the of the iCCS tool [14,15] developed by SINTEF Energy Research, was

used to identify and assess the technical and cost performances of the optimal membrane process for a given set of membrane properties (selectivity and permeance).

Based on this numerical model, the cost performances of 1600 sets of membrane properties (selectivity and permeance) for CO₂ capture from an ASC power plant as defined by the EBTF were evaluated and compared with the reference commercial solvent concept (MEA) to identify the membrane properties required in a

base case, in which both membrane- and MEA-based processes are regarded as mature and developed. The results show that in order to be competitive with simple membrane process configurations, the membrane permeance and selectivity have to be at least superior to $3 \text{ m}^3(\text{STP})/(\text{m}^2 \cdot \text{h} \cdot \text{bar})$ and 65 when high selectivities and high permeances are considered, respectively. However, with more advanced configurations (for example with recycle, sweep, counter-current flow pattern, etc.), membranes processes with permeances as low as $1 \text{ m}^3(\text{STP})/(\text{m}^2 \cdot \text{h} \cdot \text{bar})$ with high selectivities, and selectivities as low as 30 with high permeances could compete with MEA-based capture. The base case also shows the existence of an optimal selectivity curve for the range of membrane properties evaluated, as previously published by Zhai and Rubin [45].

However, as membrane-based CO_2 capture is less mature and demonstrated than the reference MEA-based process, and as there are uncertainties regarding membrane module costs, a further five case studies were assessed in order to quantify the influence of the additional costs associated with demonstration projects and higher module costs. The results show that these additional costs can significantly increase the selectivities and permeances required, and therefore narrow the range of properties possible if no financial support is provided to offset these costs.

In order to link membrane development efforts to the results presented here, the constraints brought by Robeson's upper-bound limitation were included considering different membrane film thicknesses. The inclusion of the upper bound shows that the capacity to generate thin membrane film layers for the membrane module will be important to avoid reducing the range of membrane properties which are at the same time achievable and interesting in term of cost performance, especially in cases that involve demonstration and/or higher module costs. Furthermore, properties of eight existing and under development membranes and the membrane properties which can theoretically be obtained from 276 polymeric materials presented in the literature are used to provide guidance for membrane development. The results show that while several membranes have the potential to be cost-competitive after improvement and once membrane-based CO_2 capture is mature and demonstrated, financial support will be required to demonstrate and help mature the technology. They also show that a number of polymeric materials can be used to develop membrane modules which could be cost-competitive with MEA-based CO_2 capture, as well as with the Polaris membrane.

Finally, ways to use the results presented here for membrane development by membrane development experts, for membrane selection by the industry, and for technology development and demonstration support by decision-makers were discussed.

However, it is important to note that the results presented in paper are specific to the application considered and that these conclusions would vary, depending on the application characteristics such as the CO_2 content of the flue gas, impurities, capture ratio, maturity of the technology. The results presented here cannot therefore be extrapolated to other applications than post-combustion CO_2 capture from a coal-fired power plant. To further identify the potential of membranes to compete with solvent-based CO_2 capture, the membrane properties required for membrane-based processes to be a cost-competitive option for post-combustion and pre-combustion CO_2 capture from different industrial applications (refinery, syngas, cement and steel) will be investigated in the future. In addition, the opportunities of hybrid processes such as membrane separation combined with low-temperature CO_2 capture will also be investigated [49,50]. Finally, in order to further assess the full potential of membrane-based CO_2 capture, the numerical model described here will be used to assess the impact of the CO_2 capture ratio on the CO_2 avoided cost

and identify the optimal capture ratio for different membrane properties and applications [51].

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