Hollow Fiber Membrane Contactors for CO 2 Capture: Modeling and Up-Scaling to CO 2 Capture for an 800 MW e Coal Power Station

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D o s s i e r
Post Combustion CO2 Capture
Captage de CO2 en postcombustion

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Résumé — **Contacteurs à membrane à fibres creuses pour la capture de CO_2 : modélisation et mise à l'échelle de la capture du CO_2 d'une centrale électrique au charbon de 800 MWe** — Une analyse technico-économique a été effectuée pour comparer les modules de membrane à fibres creuses (HFMM, *Hollow Fiber Membrane Modules*) avec les colonnes à garnissage structuré, plus conventionnelles, pour une utilisation comme absorbeur dans les systèmes de capture de CO_2 à base d'amine pour centrale électrique. Afin de simuler le fonctionnement d'un système HFMM de taille industrielle, un modèle bidimensionnel a été développé et validé en se basant sur les données issues d'un HFMM de laboratoire. Après diverses expériences réussies et une validation du modèle, un HFMM à l'échelle du pilote a été construit et simulé avec le même modèle. Les résultats des simulations, pour les deux tailles de HFMM, ont été utilisés pour évaluer la faisabilité d'une mise à l'échelle vers un système HFMM à même de capturer le CO_2 d'une centrale électrique de 800 MWe. Les exigences du système – longueur totale de la membrane, surface totale de contact et volume du module – ont été déterminées à partir des simulations et utilisées pour établir une comparaison économique avec les colonnes à garnissage structuré. Les résultats indiquent qu'une réduction significative des coûts d'au moins 50% est nécessaire pour rendre les HFMM compétitifs par rapport aux colonnes à garnissage structuré. Des études approfondies restent nécessaires pour optimiser plusieurs paramètres de conception des HFMM de taille industrielle, tels que le rapport d'aspect (longueur/diamètre du module), la durée de vie de la membrane, le matériau et la forme du module, tout en abaissant le coût global. Cependant, les HFMM présentent l'avantage d'une surface de contact par unité de volume et d'une capacité de mise à l'échelle du module plus importantes, ces paramètres étant clés pour les applications nécessitant une empreinte limitée ou une configuration flexible.

Abstract — *Hollow Fiber Membrane Contactors for CO_2 Capture: Modeling and Up-Scaling to CO_2 Capture for an 800 MW$_e$ Coal Power Station* — *A techno-economic analysis was completed to compare the use of Hollow Fiber Membrane Modules (HFMM) with the more conventional structured packing columns as the absorber in amine-based CO_2 capture systems for power plants. In order to simulate the operation of industrial scale HFMM systems, a two-dimensional model was developed*

and validated based on results of a laboratory scale HFMM. After successful experiments and validation of the model, a pilot scale HFMM was constructed and simulated with the same model. The results of the simulations, from both sizes of HFMM, were used to assess the feasibility of further up-scaling to a HFMM system to capture the CO_2 from an 800 MW_e power plant. The system requirements – membrane fiber length, total contact surface area, and module volume – were determined from simulations and used for an economic comparison with structured packing columns. Results showed that a significant cost reduction of at least 50% is required to make HFMM competitive with structured packing columns. Several factors for the design of industrial scale HFMM require further investigation, such as the optimal aspect ratio (module length/diameter), membrane lifetime, and casing material and shape, in addition to the need to reduce the overall cost. However, HFMM were also shown to have the advantages of having a higher contact surface area per unit volume and modular scale-up, key factors for applications requiring limited footprints or flexibility in configuration.

INTRODUCTION

The need to reduce the amount of CO_2 that is emitted into the atmosphere is an issue that is being addressed globally. Targets for CO_2 emissions reduction have been set in several countries, such as a reduction to 17% below 2005 levels by 2020 in the United States, a reduction per unit of GDP (Gross Domestic Product) of 40-45% below 2005 levels by 2020 in China, and a reduction of 20% below 1990 levels by 2020 in the European Union [1]. In order to meet these ambitions, a multi-headed approach will be needed where less fossil-based energy is consumed, by increasing energy efficiency and the share of renewable energy, and the fossil based-energy that is consumed becomes cleaner. For the latter, Carbon Capture, Utilization, and Storage (CCUS) will be key in reducing the emissions from the 1.2×10^8 GWh of energy that is generated from fossil fuels every year [2].

This work is concerned with carbon capture, specifically post-combustion carbon capture, as it will be the most important solution for existing power plants in the near-term [3]. As an example, the flue gas from a coal-fired power plant, a dilute stream of CO_2 of about 14 vol% in N_2, was considered. The conditions of the flue gas are mild – atmospheric pressure and about 50°C – but the volumes are immense, around 600 m^3/s for an 800 MW_e coal-fired power plant. In order to separate the relatively small amount of CO_2 (10-15 vol%) from the flue gas, the gas is usually brought into contact with a chemical solvent, typically an amine, such that the CO_2 reacts with the solvent and can be later released from the solvent upon heating of the liquid or a reduction in pressure. The systems involved in such a separation are huge, upwards of 16 000 m^3 in volume and 2 million m^2 of contact area for the absorption columns alone, for the 800 MW_e case[1].

In order to provide the high surface area needed, multiple large towers, more than 10 m in diameter and 40 m in height, are filled with a structured packing material. The flue gas is fed into the bottom of the tower and flows upwards while reacting with the liquid solvent that is fed at the top. The liquid flows over the packing material, which thus provides the high amount of contact surface needed. The large dimensions can make the retrofit of such systems to existing power plants difficult, due to space limitations, while also requiring a high initial investment. Furthermore, operational issues, such as foaming, entrainment, and channeling, can occur and are difficult to mitigate for structured packing columns [4].

An alternative option to the standard configuration, which helps to solve some of these key issues is the use of membrane contactor modules. With membrane modules, non-selective, porous, hydrophobic membranes are most often used, as investigated in this work, but composite membranes consisting of a porous support and selective dense top layer have also been considered [5]. The purpose of the membrane is to provide the contact surface required between the gas and liquid phases. The gas flows on one side of the membrane and is transported through the membrane pores in order to come into contact with the liquid, flowing on the other side of the membrane. Two membrane configurations are commonly employed for such modules: flat sheet and hollow fiber. With flat sheet membrane modules, the absorption liquid and flue gas are passed along either side of a membrane sheet, with many membranes in parallel separated by spacers. These systems are advantageous in that they are robust against particulates in the flows and are more flexible in the membrane materials that can be used, but have the disadvantages of having a relatively low specific surface area due to the inclusion of spacer material and having relatively high pressure drops [6]. The other configuration, Hollow Fiber Membrane Modules (HFMM), uses hollow fiber membranes,

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[1] Values based on internal TNO calculations completed as part of the EU FP7 project CESAR.

which have specific surface areas of more than 1 000 m^2/m^3 and are already used commercially in water treatment systems. In HFMM, hollow membrane fibers, usually a few millimeters in diameter, are used to provide the contact surface area between the gas and liquid phases. The gas usually flows usually around the outside of the fibers (shell side) and the liquid usually flows on the inside of the fiber tubes (tube side). A module is made up of thousands of fibers tightly bundled together, with several modules connected in parallel in order to achieve the required capture rate of CO$_2$. The flows are still counter current, as with the packed columns, but the flow rates can be controlled independently such that foaming, flooding, and entrainment are no longer problems. As will be shown here, the system volume is also much smaller, making HFMM well suited for retrofit applications.

While much of the work with HFMM has been focused on predicting the mass transfer of the various components within the different phases (gas, membrane, liquid) [7-10] the work presented here will discuss the impact that these systems can have on large scale post-combustion CO$_2$ capture. A techno-economic analysis was completed in order to compare the use of HFMM with more conventional structured packing columns based on the design of a CO$_2$ capture plant for an 800 MW$_e$ coal fired power plant.

The approach taken in this work was a combination of experimental, modeling, and techno-economic analysis,

as shown in Figure 1. Experimental results from a pilot scale HFMM, constructed by *Polymem* (Toulouse, France), with 10 m^2 of gas-liquid contact surface area were obtained to investigate the operation of larger scale modules. The results were then compared with a 2D HFMM mass transfer model, similar to those described in literature, which was previously validated as described in Chabanon *et al.* (this same journal issue) [11]. Fitting the model results to the experimental data allowed for the calculation of the membrane mass transfer coefficient, k_m, and other correlations for the mass transfer of components within the liquid, membrane, and gas phases. The k_m value and correlations were then used as input for a 1D HFMM model implemented in Aspen. The actual process in developing the 1D Aspen model, including the derivation of correction factors to account for the change from 2D to 1D, was not straightforward. Therefore, the work with the 1D Aspen model is outside of the scope of this paper and will be addressed in future work (the authors may be contacted for more information).

The results presented here for the industrial design are based on the calculations with the 2D mass transfer model. Two cases will be discussed – a more ideal case, assuming uniform flows of the liquid and gas for each of the fibers, and a worse case, assuming maldistribution of flows as seen with the pilot scale HFMM. The industrial 800 MW$_e$ design for each case is given followed by
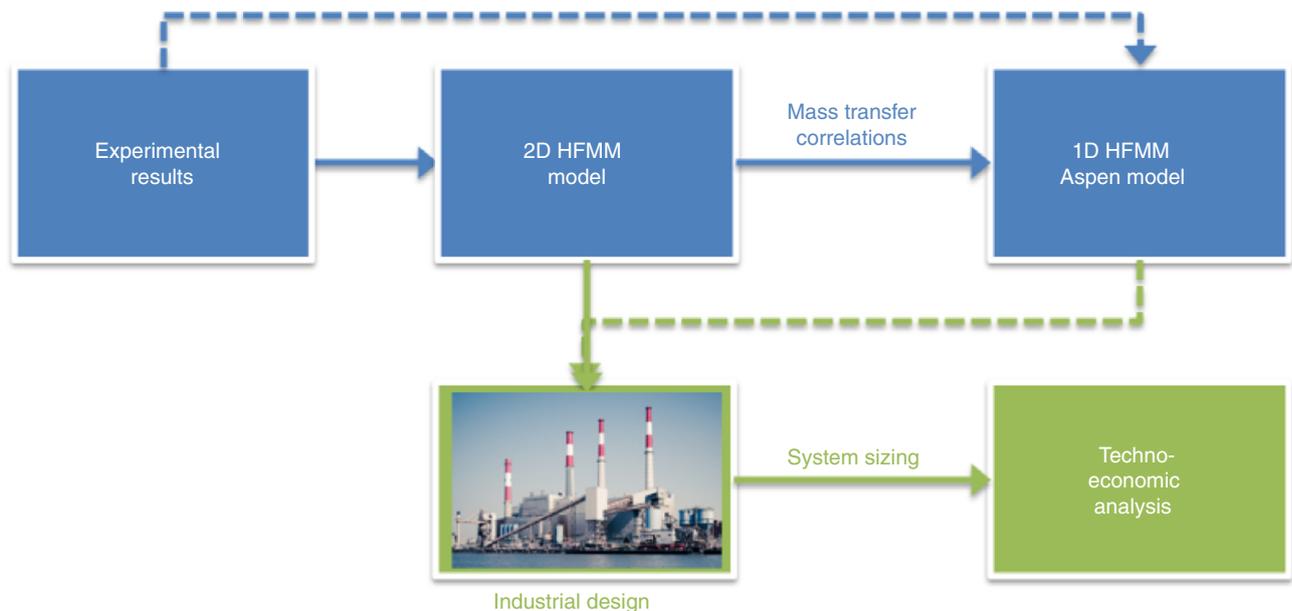

Figure 1

Schematic of the approach taken to complete the techno-economic analysis of a large scale HFMM system.

an analysis of the economics, as compared to a conventional structured packing system for the 800 MW$_e$ plant.

1 EXPERIMENTAL

The experimental work was conducted in two phases, as described in detail in Chabanon *et al.* [11] and briefly described here. The first phase concerned the operation of a small laboratory scale HFMM, made up of only 119 PTFE fibers for a total contact surface area of 0.2 m^2, with the other parameters given in Table 1. The experimental results fit very well with the 2D mass transfer model, which assumed uniform flows of both the gas and liquid, indicating that the small module represented nearly ideal conditions.

The second phase concerned the construction and operation of a pilot scale module with a total contact surface area of 10 m^2 and 8 521 fibers. The parameters of the module are also given in Table 1, where length is the actual length of the fibers and effective length is the length that is available for gas liquid contacting due to the sealant material. It should be noted that the membrane fibers were the same as those used for the laboratory scale module. The other details of the design of this module are described in [11]. In short, the gas flowed from top to bottom, counter-current to the liquid, which flowed from the top. An inline transparent section followed the liquid outlet to allow for observation of any gas bubbles and an inline filter followed the gas outlet to catch any liquid that could have percolated through the membranes. These features, along with two pressure sensors each for the gas and liquid phases, ensured that the operational parameters were set to allow for good gas-liquid separation without bubbling of the gas into the liquid phase (avoidance of bubbling was necessary for accurate measurements).

With both modules, the feed gas consisted of a mixture of 14-15% CO_2 in N_2 in each experiment while the total flow rate was on the order of liters per minute for the lab scale 100's of liters per minute for the pilot scale (10 m^3/h). The flow rates of the liquid, 30% MEA in H_2O in each case, was varied between 1×10^{-2} and 1 liters per minute for the lab and pilot scale modules, respectively. All measurements were taken after steady-state was reached.

Table 2 shows a set of the experimental results from the laboratory scale HFMM for two different liquid flow rates and several gas flow rates. The results show a smooth trend (*Fig. 2*) of increasing CO_2 at the outlet with increasing gas flow rate. This is as expected as a lower liquid to gas (Q_l/Q_g) ratio has been shown to result in a lower removal of CO_2 from the gas stream, resulting in a higher concentration of CO_2 at the outlet [12]. With an increase in liquid flow rate, the CO_2 concentration at the outlet decreases for a given gas flow rate. This is again as expected since with higher liquid flow rates, Q_l/Q_g is increased and the driving force for mass transfer of the CO_2 into the MEA solution is higher. These results were then used to validate the 2D mass transfer model.

For the pilot scale module, the results of several of the experiments are given in Table 3, for a range of gas and liquid flow rates. The inlet CO_2 concentration in

TABLE 1

Details of the laboratory scale and pilot scale HFMM

		Lab scale	Pilot scale
Module	Inner diameter (m)	1.24×10^{-2}	0.105
	Length (m)	0.35	1
	Effective length (m)	0.30	0.88
	Number of fibers (-)	119	8521
	Packing ratio (-)	0.59	0.648
	Specific interfacial area (m^2/m^3)	1 331	1 329
Fiber	Inner diameter (m)	4.3×10^{-4}	4.3×10^{-4}
	Outer diameter (m)	8.7×10^{-4}	8.7×10^{-4}
	Porosity (-)	0.336	0.336
	Material (-)	PTFE	PTFE

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TABLE 2

Experimental data from the laboratory scale HFMM

| | Experimental data | | | | | | CO₂ removal | |
| | Liquid | | | Gas | | | | |
No.	Q_l (mL/min)	Temp. at inlet (°C)	CO₂ lean loading (mol/mol)	Q_g (L/min)	CO₂ conc. at inlet (%)	Q_l/Q_g (kg/kg)	CO₂ conc. at outlet (%)	Experiment efficiency (%)
1	10	Room temp.	0	0.1	15.2	3.29	0.1	99.3
3	10	Room temp.	0	0.2	15.2	1.64	0.1	99.3
39	10	Room temp.	0	1.5	15.7	0.22	4.8	69.4
41	10	Room temp.	0	3.0	14.9	0.11	9.0	39.6
43	10	Room temp.	0	4.5	14.9	0.07	9.2	38.3
38	10	Room temp.	0	6.0	15.0	0.05	10.7	28.7
25	50	Room temp.	0	0.6	14.5	2.75	< 0.05	99.9
13	50	Room temp.	0	0.2	14.9	8.23	0.1	99.5
27	50	Room temp.	0	1.5	15.0	1.10	2.0	86.7
57	50	Room temp.	0	3.0	15.6	0.55	4.4	71.8
30	50	Room temp.	0	4.5	14.7	0.37	6.9	53.1
58	50	Room temp.	0	6.0	14.8	0.27	8.6	41.9
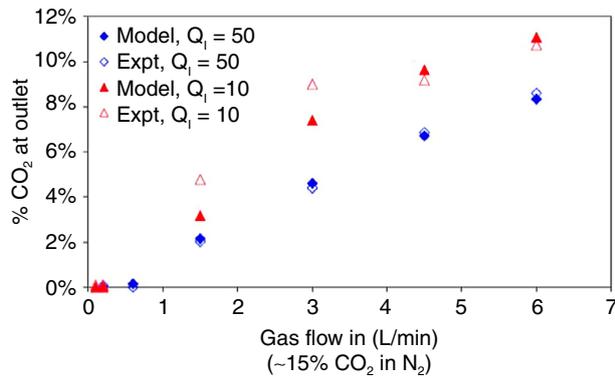
Figure 2

Comparison of simulation results and experimental data for the laboratory scale HFMM of the CO_2 outlet concentration for gas inlet flow rates of $50 \times 10^{-3} - 6.00$ L/min and two liquid flow rates, $Q_l = 10 \times 10^{-3}$ and 50×10^{-3} L/min. Taken from [11].

the CO_2 capture efficiency was Q_l/Q_g (7th column in Tab. 3). For larger ratios of 12 or more, the capture efficiency was above 70%, while for smaller ratios of less than 2, the capture efficiency was usually less than 50% (except for experiment No. 64). These experiments were then simulated with the 2D model, as discussed in the following section.

2 MODELING

The 2D mass transfer model is described in detail in [11]. Briefly, the model is based on mass and energy balances written for each phase – liquid, membrane, and gas – and each relevant component – CO_2, MEA species, and water. Between the gas and membrane phases, continuity in the concentrations and temperatures are stipulated. Between the membrane and liquid phases, equilibrium relations are used for the concentrations and continuity is used for the temperature. The model was validated based on the data from the laboratory scale HFMM, as given in Table 2. The fit between the model and experimental data, shown in Figure 2, was very good and allowed for the calculation of the membrane mass transfer coefficient. The best fit resulted in

the MEA can be seen to increase slightly with each experiment, but this was not thought to influence the results significantly. Similar trends were observed as with the lab scale module and, again, the major influence on

TABLE 3

Experimental data from the pilot scale HFMM

| No. | Experimental data | | | | | | CO$_2$ removal | |
| | Liquid | | | Gas | | | | |
	Q_l (L/h)	Temp. at inlet (°C)	CO$_2$ lean loading (mol/mol)	Q_g (Nm^3/h)	CO$_2$ conc. at inlet (%)	Q_l/Q_g (kg/kg)	CO$_2$ conc. at outlet (%)	Experiment efficiency (%)
64	30.7	16.0	0.046	10.2	14.9	1.88	6.5	56.5
67	30.6	16.0	0.084	30.2	15.0	0.53	11.8	21.5
68	100.8	16.0	0.088	30.1	14.8	1.77	8.8	40.8
69	151.0	16.0	0.107	30.3	14.4	2.67	8.2	42.8
70	50.9	16.0	0.133	10.2	12.8	3.27	4.7	63.4
72	200.9	16.0	0.157	10.1	13.8	12.94	3.8	70.5
75	200.8	16.0	0.183	30.2	14.5	3.66	9.6	33.6
76	15.7	16.0	0.179	5.5	14.6	1.90	8.4	42.5
80	100.9	16.0	0.195	5.5	15.5	12.10	4.1	73.7

a value for k_m of 2.58×10^{-4} m/s, as also reported in [11]. Given the assumptions of ideality in the model and the good fit, this k_m value was taken as the "ideal case" k_m.

The 2D model was then used to simulate the operation of the pilot scale module. Due to non-uniformities in the liquid and, especially, the gas flows, the same k_m value could not be used to fit the data. The k_m that was used to fit the pilot scale module data was 5.31×10^{-5} m/s, an order of magnitude smaller than the ideal case k_m (for details, see [11]). As the membrane fibers were the same in both of the modules, the decrease in the k_m value needed to fit the pilot scale data was not caused by a change in the actual k_m, but instead in the overall mass transfer coefficient, K_{ov}, incorporating the gas, membrane, and liquid phase mass transfer. Most of the decrease in the mass transfer was thought to be caused by maldistribution of the fibers, as observed by X-ray imaging of the fiber bundle, and bypassing of the gas flow around the fibers, but inconsistent liquid flow could have also been a factor. Still, since k_m was used as the fitting parameter, these non-idealities were lumped into the calculation of a modified membrane mass transfer coefficient, k_m'. As this value represented a case with much lower CO$_2$ capture efficiency than the ideal case, it was taken as the "worse case" k_m'.

Given k_m and k_m', the model was then used to study how an HFMM system could be designed for the capture of 90% of the CO$_2$ from an 800 MW$_e$ coal-fired power plant. The results are described in the following section.

3 EVALUATION AND DISCUSSION OF THE 800 MW$_e$ STUDY

3.1 Case Descriptions

Two different cases were defined for this part of the study for an 800 MW$_e$ coal-fired power plant, a base case with a gas flow temperature of 50°C, and a cooled case, with a gas flow temperature of 30°C after a Direct Contact Cooler (DCC). The cooled case was also analyzed as it resulted in a lower flue gas volume and a slightly higher CO$_2$ concentration, and thus required less membrane surface area and a smaller (cheaper) design. The characteristics of flue gases are detailed in Table 4. Also shown are the specifications for the absorption solvent conditions. These were chosen to be equivalent to those for the packed column reference case and were adjusted accordingly in the model.

For the CO$_2$ absorption, the solvent used was based on MEA, 30 wt%, with a CO$_2$ lean loading of 0.271 mol CO$_2$/mol MEA, and a rich loading of 0.468 mol CO$_2$/mol MEA. This implies that the total amine molar flow rate was calculated to be equal to 504 000 kmol/h (11 500 m^3/h) to reach 90% CO$_2$ capture.

3.2 Results for Module Requirements

The number of membrane modules was fixed at 100 and, for technical and feasibility considerations. The diameter

TABLE 4

Specifications of the flue gas conditions, solvent conditions, and HFMM designs for the two cases considered: with and without a direct contact cooler

	Base case (no DCC)	Cooled case (w/ DCC)
Flue gas		
Temperature (°C)	50	30
Pressure (atm)	1	1
Total gas flow rate (m^3/s)	680	586.5
Molar composition:		
CO_2 (%)	13.4	14.6
N_2 (%)	71.1	77.4
O_2 (%)	3.6	3.9
H_2O (%)	11.9	4.2
Solvent		
Temperature (°C)	40	40
Total liquid flow rate (m^3/s)	3.48	3.48
MEA (wt%)	30	30
Inlet loading (mol/mol)	0.271	0.271

of the modules was chosen to allow for a superficial gas velocity of 1.8 m/s, which resulted in a pressure drop of about 100 mbar along the length of a 2 m fiber, given the same packing density as in the pilot scale HFMM. The number of fibers in each module was based on the calculated module diameter and a value of 1 250 m^2/m^3 as a feasible specific membrane module contact surface area, following the manufacturing recommendation of *Polymem*, the module provider involved in this work.

The membranes for this part of the study were considered to be the same as those used in the laboratory and pilot scale modules described above with the parameters as given in Table 1. Regarding the fibers themselves, details of their characterization are given in [11]. Even for longer membranes, the structure of the membranes and loss in length due to the sealant around the fibers were assumed to not change given discussions with the membrane manufacturer, *Polymem*. The construction of such modules and support of the membrane bundle for modules longer than 1 meter is a subject of ongoing work at *Polymem*. With longer fibers, the bundle must be

supported somehow and the liquid pressure at the inlet of the fibers must not be too high as to cause wetting of the pores. In the worst case, for a large scale system, a few modules in series would be required to achieve the required CO_2 capture.

The length of the fibers in each case was adjusted in the model in order to achieve a simulation result that met the 90% CO_2 capture criteria. This then allowed for the calculation of the total required membrane contact surface area and module volume. The results are shown below in Table 5 for both cases of the flue gas conditions and for both values of k_m – the low worse case value, k_m' and the higher ideal case value, k_m. Also included in Table 5 are the comparisons of the contact surface area and volume required for each k_m value relative to that required for a structured packing column (the numbers for the structured packed column were developed within a different part of the same EU FP7 project, CESAR; surface area = 2.01 × 10^6 m^2, volume = 16 500 m^3).

The key points to be noted from Table 5 concern the dramatic impact of the k_m value on the sizing of the system. A decrease in k_m by a factor of 5 results in an increase in required membrane area and module volume of 325% for the base case and 335% for the cooled case. When these values are compared to a structured packing column (designed for the base case), the influence of the k_m is again apparent. With the higher k_m value from the ideal case, the required surface area for the HFMM and the packed column are essentially the same (row 10 in *Tab. 5*), indicating that the mass transfer is not limited by the membrane in the HFMM, but instead is dominated by the transfer of the gas into the liquid phase. With the lower k_m' from the worse case, the extra mass transfer resistance from the non-idealities in the flows result in the situation where the surface area required for gas-liquid contact is three times higher with the HFMM than with the packed column. Still, in both cases there is a substantial reduction in volume with the HFMM relative to that required for the packed column – about 70% with the worse case k_m' value and as much as 90% with the ideal case k_m.

3.3 Configuration Design Options

Several factors must be considered in the design of HFMM for CO_2 capture from power plants, for example the casing material, the shape (round or square), the total number of modules, the number of fiber bundles within each module, and the aspect ratio (length/diameter). Large membrane modules would be difficult to install and maintain; furthermore, the aspect ratio

TABLE 5

Results for the HFMM requirements for each case and for the two k_m values discussed previously

	Base case (no DCC)	Cooled case (w/DCC)
Number of modules (-)	100	100
Packing density (m^3 fibers/m^3 module)	0.709	0.709
Specific surface area (m^2/m^3)	1 250	1 250
Module diameter (m)	4.07	3.78
# fibers/module (-)	1.20×10^7	1.04×10^7
Ineffective membrane length (m)	0.10	0.10
k_m from ideal case		
Length of fibers (m)	1.23	1.24
Total surface area (m^2)	2.00×10^6	1.74×10^6
Total volume (m^3)	1 597	1 389
Comparison with structured packing (ratio HFMM/packed column)		
Surface area (-)	0.99	0.86
Volume (-)	0.097	0.084
k_m' from worse case		
Length of fibers (m)	4.00	4.16
Total surface area (m^2)	6.49×10^6	5.82×10^6
Total volume (m^3)	5 194	4 659
Comparison with structured packing (ratio HFMM/packed column)		
Surface area (-)	3.23	2.90
Volume (-)	0.315	0.283

would make it difficult to ensure good gas flow penetration to each element. Hence, a smaller diameter module design should be considered for efficient absorption. The implications of smaller modules on capital cost would be a matter best addressed by manufacturers; however, the other factors must be considered at the same time to achieve the optimal design. To keep the absorber footprint (floor size) to a minimum, square modules would provide a better solution. The difficulty in this case would be the design of the liquid system header box which operates above atmospheric pressure, for which purpose a circular design would be preferable.

From Figure 3, it can be seen that the overall module design could be very much like a 'shell-and-tube' heat exchanger, except that each tube is a bundle of membrane fibers. If required, the whole module or individual bundles could be lifted and removed from the gas stream, for which purpose the header plate would have a top and bottom clamp for each bundle. Each module may also contain several loose-fitting baffle plates, providing further gas distribution. However, the benefit of baffle plates may not be substantial enough to justify the incurred pressure drop. This would be the case for a module design with a good aspect ratio (small diameter, large length). Another consideration of this module design may also be the possibility to design the membrane potting in such a way as to direct the gas path more effectively at the inlet and outlet of the module (shown as "sloped potting" in *Fig. 3*). This may also help overcome the pressure drop incurred by any baffles.
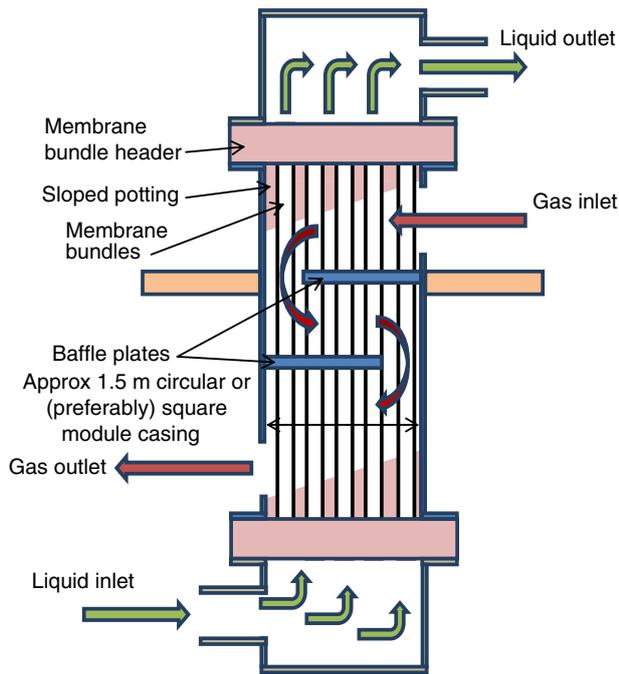
Figure 3

Detailed schematic of a possible design of a HFMM and internals.

The footprint of the gas absorption plant will depend mostly upon the total number of modules required for the desired CO_2 capture rate, and further work should be carried out with regards to available options for duct-work sizing. Another consideration will be the very low gas velocities in the HFMM. This may result in ash drop out, calling for an improved absorber design which can collect and remove the ash before entering the modules. Lower gas velocities will provide the advantage of less erosion; however, the impact of increased pressure drop must be further studied.

The design considerations of industrial scale-up, as well as the large differences in the two experimental systems discussed earlier suggest that an accurate prediction of the overall mass transfer coefficient is required in order to provide a more appropriate fiber length estimation. Accurately approximating the fiber length would provide a solid basis for design and industrial scale-up; however, ensuring a good distribution of gas and liquid flows (through baffles, for example), minimizing bending of fibers, and minimizing pressure drop will all be key aspects in the final design of an industrial scale system. Still, with the configurations as calculated with the model (*Tab. 5*), it can be concluded that scale-up to some degree should be feasible.

4 ECONOMIC COMPARISON WITH STRUCTURED PACKING COLUMNS

The economic feasibility of the CO_2 capture system incorporating the HFMM was also studied based on the ideal case k_m and worse case k_m' values and the system parameters, as given in Table 5. The values for the conventional structured packing columns capture system were based on calculations completed as part of the same EU FP7 CESAR project and based on the EBTF guidelines [13]. For CO_2 capture with HFMM, an equivalent system was assumed with only the absorption columns replaced by the membrane contactor modules. The capital and operating costs of the other components – stripper, pumps, blowers, etc. – remained the same (in future work, this will need to be investigated in more detail as the membrane modules cause more gas-side pressure drop and will result in higher operating costs for the blowers or even the need for additional blowers). The calculations for the structured packing absorption column were based on state-of-the-art designs with the total values given in Table 6, along with the other assumptions used in the economic calculations. The results for the packed column configuration were only available when no DCC was added to the system. Also given in Table 6 are the cost of the DCC, as calculated for the specified flue gas inlet and outlet conditions, and the cost of the membrane modules, based on past experience of the authors.

The total capital costs (not including installation costs) of the absorber for each of the cases considered are compared in Figure 4. With the lower k_m' value from the worse case, the absorber system is exceedingly more expensive than the conventional structured packing

TABLE 6

Values used in the economic analysis. Costs do not include installation costs

Description	Value
Capital costs exclusive of absorber (M€)	155.9
Total operating costs (M€/yr)	30
Annuity factor (1/yr)	0.08
Project life (yr)	40
Working hours (h/yr)	7 500
Amount CO_2 produced (ton/yr)	3.90×10^6
Cost of DCC (M€)	5.39
Cost of membrane modules (€/m²)	20

column and even with the k_m value from the ideal case, requiring the same contact surface area as the packed column, the absorber capital investment is still more than twice as high. A benefit of adding a DCC to the system is seen for the worse case k_m' and results in a capital savings of 8 M€. With the ideal case k_m, however, there is almost no difference between the two configurations.

A more detailed analysis was completed to compare the base case (no DCC) with the analysis already completed for the structured packing column. The operating
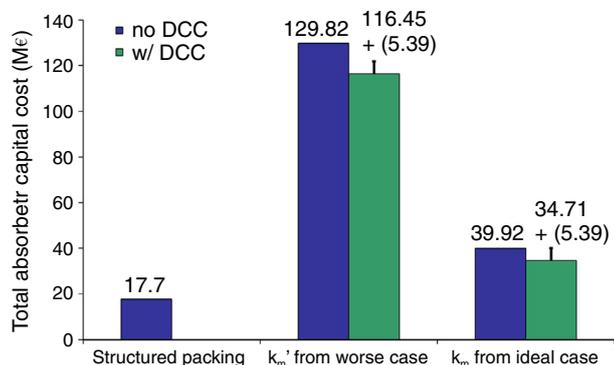

Figure 4

Comparison of total absorber capital costs for the structured packing column and each of the HFMM cases. The error bars shown for the case with the DCC indicate the cost of the DCC itself.

costs and capital costs were accounted for annually with the formula:

$$Annual\ cost = Annuity\ factor\ \times capital\ costs \\ + operating\ costs$$

The total yearly costs and cost per ton CO_2 avoided are shown in Table 7. The results show that the process is dominated by the operating costs, set constant at 30 M€/yr (*Tab. 6*). Given this and the magnitude of the amount of CO_2 that would be captured each year, the cost per ton of CO_2 avoided does not vary greatly – an increase of only 2.05 €/ton CO_2 with the ideal k_m value relative to the packed column. With the worse case k_m' value, the difference is more significant and results in a rise of 9.13 €/ton CO_2. With the assumption that the operating costs were equal between the packed column and HFMM systems and given the large influence of the operating costs, a more detailed analysis may result in significantly different results. However, when looking at the initial investment, the HFMM system is always much more costly. Furthermore, back calculating to determine the price of the membrane modules required to make HFMM competitive with packed columns shows a dramatic cost reduction is necessary –56% for the ideal case with the higher k_m value and up to 86% for the worse case with the lower k_m' value, as shown in Table 8.

TABLE 7

Results of economic analysis for the structured packing case and two cases with HFMM (no DCC)

	Structured packing	Membrane contactor (no DCC)	
		k_m', worse case	k_m, ideal case
Absorber/module capital (M€)	17.7	130	40
Total capital costs (M€)	173	285	195
Total annual cost (M€/yr)	43.24	52.81	45.62
Total cost per ton CO_2 avoided (€/ton)	51.62	60.75	53.67

TABLE 8

Required values to make HFMM cases competitive with the structured packing case

	Membrane contactor (no DCC)	
	k_m', worse case	k_m, ideal case
% Reduction in module price required (%)	86	56
Required specific module price (€/m^2)	2.75	8.85

TABLE 9
Results of economic analysis assuming a membrane lifetime of 5 years and a replacement cost of 50%

	Structured packing	k_m', worse case	k_m, ideal case
Total annual cost (M€/yr)	43.24	65.79	49.61
Additional annual cost (M€/yr)	n/a	12.98	3.99
Total cost per ton CO₂ avoided (€/ton)	51.62	65.53	55.15
Required module price for equivalent cost per ton CO₂ captured (€/m²)	n/a	1.23	3.95

Another key assumption of the analysis above is that no degradation of the membranes occurs. This is not very realistic, so the analysis was repeated assuming a membrane lifetime of 5 years and a replacement cost of 50% of the module cost, or 10 €/m² for the base case, to account for replacement of only the membrane bundles and not the entire module. The economic feasibility is much worse, as shown in Table 9, and the price of the modules must be reduced by 80% to 3.95 €/m² (assuming a constant replacement cost of 50%) even in the ideal case.

CONCLUSIONS

A case study for an industrial scale CO₂ capture system was completed using the 2D mass transfer model described in Chabanon *et al.* [11]. Based on data from two experimental systems – a laboratory scale and a pilot scale – two membrane mass transfer coefficients resulted from the fit of the simulation results and the experimental data – the "ideal case" k_m and the "worse case" k_m', respectively. Four cases were simulated for comparison utilizing the two different membrane mass transfer coefficient values (k_m and k_m'), and two different system configurations, with and without a direct contact cooler. The results showed an increase in membrane area (and thus module volume) of about 330% for the worse case relative to the ideal case.

Further economic calculations based on the design calculations showed that with the worse case k_m' value, the system is exceedingly more expensive than the conventional structured packing column, and even with the ideal case k_m value, requiring the same contact surface area as the packed column, the total capital investment is still more than twice as high. In this case, a minimum reduction of 56% of the cost of the membrane modules is required to be competitive. In the more realistic case,

including degradation of the membranes, the required reduction in capital cost was a minimum of 80%.

The design considerations of industrial scale-up, as well as the large differences in the studied cases suggest that an accurate overall mass transfer coefficient value is required in order to provide a more appropriate fiber length estimation. Accurately approximating the fiber length would provide a solid basis for design and industrial scale-up; however, with the current configuration, it can be concluded that scale-up to some degree should be feasible.

This preliminary study shows enormous challenges for HFMM technology to become feasible and that it may only be attractive where substantial benefits are gained by the much smaller volume and high flexibility of hollow fiber membrane module systems relative to packed columns. Several key parameters – overall mass transfer coefficient, membrane lifetime, and aspect ratio – must be carefully determined to ensure accurate calculations of the economic feasibility. Improvements should be made not only in the membrane materials, with increased transport across the membrane, but also with the module construction and ensuring a uniform distribution of the gas and liquid flows. Substantial decreases in the costs are required, especially in those of the membranes and module casings, in order for hollow fiber membrane module contactors to be competitive with structured packing absorption columns. Still, if trends such as those observed with reverse osmosis membrane-based system can be realized, including an 86% reduction in membrane cost between 1990 and 2005 [14], hollow fiber membrane modules for CO₂ capture may one day become competitive. Furthermore, it is important to note that any comparison between HFMM and packed columns must consider the fact that a new technology undeveloped for power plant applications is being compared with a technology that is relatively well established.

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