HOLLOW FIBRE MEMBRANE CONTACTORS IN CO₂ DESORPTION: A REVIEW

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HIGHLIGHTS

1. The state-of-art hollow fiber membrane contactors in post-combustion CO₂ desorption are provided.
2. An overview of the developments of membranes and absorbents in CO₂ desorption is presented.
3. The main issues in commercializing membrane contactors (wetting and fouling) are analyzed.
4. A clear comparison between CO₂ desorption models developed in the literature is presented.

KEYWORDS

Post-combustion carbon capture, CO₂ desorption, solvent regeneration, hollow fiber membrane contactor.

ABSTRACT

Post-combustion carbon capture (PCC) using membrane technology is a rising alternative to PCC processes, which is based on conventional equipment, such as CO₂ absorption-desorption in packed columns, to mitigate human-associated climate change. Research on CO₂ capture system has been centered on improving the CO₂ absorption process efficiency, while little attention has been devoted to the performance of the solvent regeneration technology.

In this review, research development is focused on the performance of the CO₂ desorption process using hollow fiber membrane contactors (HFMCs). The main impact factors in membrane contactor technology are presented, including solvent and membrane material selection, membrane module design, mass transfer correlations, operational issues, modeling and simulations. Each section has been discussed in terms of its current status while describing future challenges to improve the technology for its industrial implementation.

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56 LIST OF ABBREVIATIONS

AAAS	Amine Amino Acid Salts
AAS	Amino Acid Salts
2-AB	2-Amine-1H-benzimidazole
AEPD	2-Amino-2-ethyl-1,3-propanediol
AL ₂ O ₃	Aluminum Oxide
AMP	2-Amino-2-methyl-1-propanol Methane
3-AP	3-Amino-1-propanol
ATP	Ambient Temperature Process
CAMD	Computer-aided Molecular Design
CAP	Chilled Ammonia Process
CCUS	Carbon Capture, Utilization and Storage
CO ₂	Carbon Dioxide

DEA	Diethanolamine
DETA	Dietilentriamina
DI	Deionized
DMPDAH	Dimethylpropylenediamine
EDA	Ethyl Diazoacetate
GLYMAPA	Glycine blended with Amine-3-(methylamino)propylamine
HDA	Hexadecyl Acetate
He	Helium
HF	Hollow Fiber
IL	Ionic Liquid
K ₂ CO ₃	Potassium Carbonate
KLys	Potassium Lycinate
MC	Membrane Contactor
MEA	Monoethanolamine
MDEA	Methyldiethanolamine
MMM	Mixed Matrix Membrane
MMT	Montmorillonite
MVR	Membrane Vacuum Regeneration
N ₂	Nitrogen
NH ₃	Ammonium
NMP	N-Methyl-2-pyrrolidone
PA	Phosphoric Acid
PAMAM	Polyamidoamine
PCC	Post-Combustion Carbon Capture
PDA	Polydopamine
PG	Potassium Glycinate
PE	Polyethylene

PEG	Polyethylene Glycol
PEI	Polyetherimide
PILM	Poly(ionic liquid) Membranes
PIL–IL	Poly(ionic liquid)–Ionic Liquid
PP	Polypropylene
PSF	Polysulfone
PTFE	Polytetrafluoroethylene
PTMSP	Poly(1-trimethylsilyl)-1-propyne
PVDF	Polyvinylidene fluoride
PZ	Piperazine
SARMAPA	Sarcosine blended with Amine-3-(methylamino)propylamine
SILM	Supported Ionic Liquid Membranes
SiO ₂	Silicon Dioxide
TEA	Triethanolamine
TEPA	Tetraethylenepentamine
TETA	Triethylenetetramine
THAM	Tris(hydroxymethyl)aminomethane
TiO ₂	Titanium Dioxide
[emim][Ac]	1-Ethyl-3-methylimidazolium Acetate
[emim][EtSO ₄]	1-Ethyl-3-methylimidazolium Ethyl Sulfate
Aqueous [bmim][BF ₄]	1-Butyl-3-methylimidazolium Tetrafluoroborate
Aqueous [apmim][BF ₄]	1-(3-Aminopropyl)-3-methyl-imidazolium Tetrafluoroborate
[emim][MS]	1-Ethyl-3-methylimidazolium Methyl Sulfate
[emim][DCA]	1-Ethyl-3-methylimidazolium Dicyanamide

1. Introduction

The improvement and promotion of carbon capture technologies are required to reduce the globally rising CO₂ emissions, which is one of the most harmful gases associated with global warming and other negative climatic effects.¹

Common strategies rely on carbon capture and sequestration (CCS), which is focused on long-term underground CO₂ storage in aquifers or other geological formations,² and carbon capture and utilization (CCU), which produces products with a high value added, such as chemicals or fuels, from CO₂.³ These are widely considered for mitigating the ongoing rise in levels of atmospheric CO₂ attributed to increasing industrial demands of fossil fuel sources due to the increase in population and economic growth.⁴

To mitigate excess anthropogenic carbon dioxide (CO₂) emissions (approx. 4GtC·yr⁻¹) and achieve the 2°C target, a portfolio of strategies should be implemented together, with carbon capture utilization and storage (CCUS) gaining attention. CCUS includes CO₂ capture from gas mixtures, such as flue gases, and its subsequent conversion into useful and valuable products.

Although many industrial processes can be decarbonized, 25% of anthropogenic CO₂ emissions from coal plants are considered key for carbon capture technologies due to the viability of decreasing future emissions from power plants and empowering the use of a sustainable and low-cost source of energy.⁵ There are currently three sufficiently developed large-scale operation technologies for CO₂ capture from coal plants (Figure 1):⁶ i) **Post-combustion capture**, where the flue gas is directly treated after the combustion step. This strategy can be applied, in principle, to any existing plant where a CO₂-containing stream is produced (a so-called retrofit option). However, the large energy required for this operation due to the low CO₂ concentration is an important factor

to decrease.⁴ **ii) Pre-combustion capture** is related to integrated gasification combined with cycle power plants, in which coal is usually gasified to syngas (CO_2 and H_2) in a low oxygen environment at pressures of approximately 50 bar before combustion in a gas turbine. Calculations show promising process economics; however, pure oxygen from air separation, CO_2 and hydrogen separation are required, increasing the cost. Additionally, the complex nature of the process hampers its fast commercialization.⁷ **iii) Oxy-fuel combustion** uses pure oxygen rather than air to burn coal, which produces a flue gas stream mainly composed of CO_2 and steam. The CO_2 can be enriched easily by steam condensation. The O_2 -combustion considerably increases the flame temperature, and the absence of N_2 reduces the concentration of NO_x formed. The main disadvantage of oxy-fuel combustion is the extra separation process required for generating pure O_2 from the atmosphere.⁸

From these three developments, post-combustion capture has the ability to retrofit to an existing power plant. More studies on this approach have been conducted by comparing both pre-fuel and oxy-fuel combustion, reaching industrial implementation. At this stage the process design study has to explore the additional economic cost of the installation and the extra energy requirements.⁶ The CO_2 capture cost is predicted to be higher than \$60/t CO_2 captured, which is not economically viable based on carbon prices in the short term.⁸ Therefore, more research has to be done in the field of post-combustion CO_2 capture based on promising technologies, such as membrane contactors, for the operational and economic viability of power plants with CO_2 capture systems.⁹

Figure 1. Schematic process diagram of the three different technologies for carbon capture from a power plant.

1.1. Post-combustion carbon capture

Liquid absorption using monoethanolamine (MEA) is recognized as the state-of-the-art post-combustion carbon capture technology, largely due to its high CO₂ removal efficiency from flue gas (normally >80%), even at low CO₂ concentrations.¹⁰ This technology has been studied since 1991; however, further developments are required to achieve a better process performance, which reduces the energy consumption and environmental impact. Developments in the technology configuration and absorbent selection should decrease the energy consumption to 0.2 MWh/ton of CO₂.¹¹

A conventional carbon capture technology using absorbents is shown in Figure 2, where direct gas-liquid contact between the outlet flue gas from a power plant and the liquid absorbent takes place inside a scrubber or packed bed. CO₂ is then absorbed by the solvent, lowering the CO₂ concentration in the flue gas, and then the solution with

dissolved CO₂ enters a stripper column where the CO₂ is desorbed by heat. The regenerated lean solution is returned to the absorber, and compressed CO₂ is collected in the upper part of the desorber column and transported.¹²

Figure 2. Conventional configuration of post-combustion carbon capture technology based on liquid absorbents. Reprinted with permission from Zhao et al.¹⁰ Copyright (2016) Elsevier B.V.

Currently, direct gas-liquid contact using amine-based solutions is the leading post-combustion process carbon capture technology, with a market presence higher than 90% due to a high reaction rate, good absorption performance and low cost of production. However, there are some disadvantages that should be studied, such as the large energy required for the regeneration process and environmental issues related to solvent losses, thermal degradation and equipment corrosion due to the high temperature applied.^{13–15} The main concept of improving carbon capture with a minimal energy penalty and

installation cost is to maximize the liquid-gas contact area and choose an appropriate solvent for the desorption process, since energy consumption in solvent regeneration is estimated to contribute up to 80% of the overall carbon capture system.¹⁶ Moreover, efforts to reduce the regeneration temperature to significantly below those used in industrial processes should not be ignored, since low regeneration temperatures generally contribute to the enormous decrease in energy consumption and a thermal degradation loss of absorbents. Research has been conducted for the development of high-performance membrane carbon dioxide separation from gaseous mixtures, effective solvent systems and optimization of the process configuration.¹⁷

In general, for membrane carbon dioxide separation based on the membrane desorption process, two types of membranes could be used: membrane contactors with porous membranes, or nonporous/dense membranes. The carbon desorption process with dense membranes is generally accomplished via the solution-diffusion mechanism. This method consists of three steps: first, gas molecules dissolve in the membrane at the feed side, then they diffuse into and pass through the membrane, before finally desorbing from the other side of the membrane. In these membranes, a selective thin dense layer reduces the permeation flux through the membrane due to resistance to the gas passage.

Although dense membranes could bring about some advantages, such as a reduction of the wetting phenomena (explained in section 2.2 Membrane challenges), non-porous contactors are rarely used for CO₂ absorption/desorption since these fibers show lower performance than porous membranes because of their higher mass transfer resistance.¹⁸ However, due to the potential of their solution-diffusion mechanism and new fabrication opportunities, such as a thin dense film coating over the porous membranes,¹⁹ this review do not examined these novel methods, as they can be deeply found in other studies.^{19,20}

In membrane technology, the process performance is highly influenced by the configuration on the membrane contactor. Both flat sheet and cylindrical membrane modules could be employed in gas-liquid membrane contactor. A cylindrical membrane module is divided into three types of hollow fibers, capillaries, and tubes, according to their shape. Hollow fiber membrane contactors (HFMCs) are used more often than other types in gas separation because they provide the highest surface-to-volume ratio.²¹ A flat sheet membrane contactor is normally installed in a plate-and-frame configuration. Studies in the area of flat sheet membrane for CO₂ capture technology is scarce, due to the contact area in this configuration is much reduced than that in HFMCs. The major interest of HFMC is concerned with the possibility of significantly decreasing the size of the installation and lower energy consumption, while keeping the regeneration performances unchanged.

From this perspective, the solvent regeneration process using porous HFMC has been considered a promising CO₂ desorption technology in recent years; however, limited reviews of this technology are available.²³ This work proposes a state-of-the-art evaluation of studies on HFMC technology fundamentals and challenges in solvent regeneration in the post-combustion carbon capture process in recent years. In the following sections, the CO₂ desorption process in carbon capture is discussed thoroughly in the following aspects: (I) solvent classification; (II) membrane material and solvent compatibility; (III) HFMC configuration; and (IV) modeling studies.

2. HFMC in CO₂ desorption

2.1. State-of-the-art

As seen before, the development of the solvent regeneration process in carbon capture technology has centered on high-temperature CO₂ desorption, where the absorbent is regenerated by increasing the temperature in a simple reactor or scrubber.²⁴ However, since the early 21st century, the solvent regeneration process based on hollow fiber membrane contactors (HFMCs) has been considered a promising alternative to traditional heating regenerators due to the huge regeneration efficiency potential and other numerous advantages shown below. A carbon capture system based on CO₂ absorption in a scrubber column and an HFMC desorption process is illustrated in Figure 3. After CO₂ absorption, the rich solvent enters the lumen or shell side of the HFMC, where CO₂ diffuses to the gas phase through the membrane, and the solvent can be recycled into the absorption process.

Figure 3. Post-combustion carbon capture process based on the membrane desorption process. Reprinted with permission from Wang et al.²⁵ Copyright (2013) American Chemical Society.

HFCMs are, in most cases, composed of a porous membrane by adding an extra mass transfer barrier in the diffusion of CO₂ between the feed and permeate sides. The membrane provides contact between both the liquid and gas phases with no dispersion, and the selectivity for CO₂ depends on the absorbent used. HFMC equipment provides high values of the effective membrane area, separates both the liquid and gas flow rates, linearly scales up the contactors and has the potential to perform the CO₂ desorption process using temperatures lower than the vaporization point of the absorber.²⁶ The specific surface area per volume for hollow fiber membrane contactors can be as high as ~1000–9000 m²m⁻³, which is up to two orders of magnitude greater than conventional packed columns. Moreover, the working temperature when HFCMs are used is usually lower than 100°C, which allows for the use of other sources of steam (e.g., low-grade steam from turbines) for solvent regeneration, reducing the energy required for the CO₂ desorption process.²⁷

Thus, the use of a membrane contactor for the carbon dioxide desorption process instead of a conventional scrubber leads to a much smaller space requirement. This technology is well suited for new and existing power plants due to the reduced footprint requirements and a much lower visual impact, as well as by providing more options for placement in existing plants.²⁸ Moreover, the modularity and simplicity of replacing the membrane contactors brings the opportunity to retrofit the membrane contactor processes with any novel membrane material or innovation in the field of membrane carbon capture processes in the future. However, long-term experiments in pilot plant have to be studied to confirm the benefits of this promising technology.

In large-scale applications using HFMC technology, the selection of both membrane materials and solvents is limited by the membrane wetting phenomena, which is the main problem of the implementation of this technology. Therefore, a great selection of

membrane-absorbent combinations, based on the overall process performance, are important in future studies.¹⁰ Table 1 summarizes both the positive and negative aspects of HFMCs in CO₂ desorption compared with conventional high-temperature regeneration.

Table 1. Positive and negative aspects of HFMCs over conventional stripper regeneration.

Aspects Compared to the conventional system (shown in Figure 2)	
Positives	
More operational Flexibility: independent flows.	Avoids operational problems of conventional scrubbers (e.g., channeling, flooding and foaming).
More economic.	In the membrane contactor, the energy consumption is lower and the equipment is more compact. Reduction over 70% in size and ca. 66% in weight ²⁹ . Small footprint (process intensification).
Linear scale-up	The modularity of membrane contactors makes the technology easy and simple to scale-up.
High, known and constant interfacial area	Easier prediction of the performance because of the independent interfacial area. Lower operating temperatures.
Negatives	
Extra mass transfer resistance	The small diameter of fibers and the membrane pores add an extra mass transfer resistance. Has the pore sizes of most hollow fiber membranes. However, a larger pore diameter induces membrane wetting. A pore size trade-off is required.
Membrane fouling and wetting	The wetting phenomena increases the membrane mass transfer resistance. Higher membrane pores diameters increase the wetting phenomena. The pore sizes of most HFMCs used in desorption process are <200 nm.
Limited applications	Limited operating pressures and temperatures based on the breakthrough value and thermal resistance of the membrane. Limited stability of the solvent and the lifetime.
Novel technology	Lack of studies in industrial applications. More research required.

The mechanism of solvent regeneration through an HFMC is introduced as the resistance-in-series model described in Figure 4. More detailed information about this model is described in section 4: Modeling CO₂ desorption. CO₂ mass transfer across the porous membrane generally involves three successive stages: (1) both diffusion and physical and/or chemical desorption from the liquid phase to the liquid-membrane interface, k_l ; (2) diffusion from the liquid-membrane to the gas-membrane interface through the membrane pores, k_m and (3) diffusion from the gas-membrane interface to the gas phase, k_g .¹⁰ CO₂ mass transfer, produced by the difference in the CO₂ partial pressure through the membrane, creates a driving force, which allows CO₂ desorption without solvent evaporation.³⁰ The CO₂ mass transfer flux (N) for the three consecutive steps can be expressed by:

$$N = k_l(C_{lb} - C_{lm})(\pi d_i L) = k_m(P_{lm} - P_{gm})(\pi d_{ln} L) = k_g(P_{gm} - P_{gb})(\pi d_o L) \quad (1)$$

where P is the partial pressure of the gas, C is the concentration in the liquid, and d_i , d_{ln} and d_o are the inner, logarithmic and outer mean diameters, respectively. The subscripts *lb* and *lm* represent the liquid bulk and liquid-membrane interface, while *gb* and *gm* correspond to the gas bulk and gas-membrane interface. In the liquid-membrane interface, the gas partial pressure in equilibrium with the gas concentration in the liquid is described according to Henry's Law ($P_{lm} = H \cdot C_{lm}$), where *H* is the Henry's Law constant.

Figure 4. Mechanism of CO₂ mass transfer from the liquid to gas phase in the solvent regeneration process. Reprinted with permission from Zhao et al.¹⁰ Copyright (2016) Elsevier B.V.

The CO₂ mass transfer driving force could be mainly achieved by two operational modes (Figure 5a and 5b): i) vacuum operating mode, where a vacuum (pressure lower than atmospheric pressure) is used on the permeate side (lumen or shell) by a vacuum pump;^{29,31–34} and ii) sweep gas operating mode, where the gases flow over the permeate side while CO₂-rich solutions flow on the other side in counter or co-current configurations. Sweep gases are usually inert gases, such as helium or nitrogen.^{35–37} Although the sweep gas operation mode could be beneficial to enhance the CO₂ mass transfer through the membrane, further purification processes must be considered to separate the CO₂ from the inert gas with additional operational costs. Consequently, other alternatives to inert gases, such as water steam, may be an opportunity to increase the CO₂ mass transfer through the membrane using a sweep gas without further purification processes, since water steam can be split from CO₂ via condensation. Significant developments have been reached in recent years using a sweep gas operation mode with water steam. However, it should be noted that additional steam sweeping would lead to

an extra heat for regeneration, and searching for a trade-off between the energy cost and an improvement in regeneration performance is recommended.³⁸

Additionally, some recent studies developed a combination of both sweep gas and vacuum modes, where a combination of both vacuum and sweep gas operation achieves a reduction in the CO₂ partial pressure in the gas phase and improves the CO₂ transfer from the liquid, seeking a trade-off between the regeneration energy cost and an enhancement of the regeneration performance.^{39,40}

Figure 5. Solvent regeneration operational modes to allow CO₂ mass transfer from the rich solvent to the gas phase. a) Pure vacuum; b) sweep gas; c) sweep gas driven by a vacuum pump

The reported experimental and modeling studies to describe the effect on the solvent regeneration performance using HFMC cover some key operating conditions: i) sweep gas and liquid velocities;⁴¹ ii) solution temperature;⁴² iii) regeneration pressure;⁴³ iv) CO₂ loading capacity;⁴⁴ and v) fiber geometry.³⁹ The regeneration performance in HFMC can be related to the average desorption flux of CO₂ ($\text{mol_CO}_2 \cdot \text{h}^{-1} \cdot \text{m}^{-2}$) and the change in the

CO₂ loading of the solvent after the desorption process, $\Delta\alpha$ (mol_CO₂·mol_solvent⁻¹).

This parameter N_{CO_2} can be expressed as

$$N_{CO_2} = \frac{n\pi R_i^2 C_{solvent} V_Z (\alpha_{rich} - \alpha_{lean})}{2n\pi R_i Z} \quad (2)$$

where V_Z is the liquid velocity (m·h⁻¹), n is the number of fibers, R_i is the inner radius of the membrane (m), $C_{solvent}$ is the concentration of the absorbent (mol·m⁻³), α_{rich} is the rich CO₂-solvent loading in the input of the fiber and α_{lean} is the mean CO₂-lean loading of the solvent in the fiber outlet. The role of temperature and the regeneration pressure could be described by the CO₂ loading change after the regeneration process.

The following findings related to the main important variables in carbon dioxide desorption are summarized below:

Liquid flow rate: The influence of this parameter on the CO₂ loading change of the absorbent before and after the regeneration process ($\Delta\alpha$) and the CO₂ desorption flux are described by equation 2. The regeneration performance defined by ($\Delta\alpha$) decreases as a function of the liquid velocity. When the liquid flow rate decreases, the regeneration performance increases because more CO₂ could diffuse from the rich solution due to the higher residence time for the same module length. However, a higher liquid flow rate usually means a higher operation potential due to the increase in the CO₂ desorption flux. At the industrial scale, CO₂ capture technology compromises both the absorption and desorption processes. In the absorption operation, a higher $\Delta\alpha$ gives a higher CO₂ absorption rate. Therefore, a low liquid flow rate is desired for large-scale applications despite reducing N_{CO_2} and increasing the size of the membrane module. For this reason, a trade-off between $\Delta\alpha$ and N_{CO_2} is relevant when a suitable liquid flow rate is applied.^{25,31,45}

Sweep gas flow rate: In contrast to the liquid flow rate, the sweep gas flow rate has a positive influence on the regeneration performance. A higher sweep gas velocity reduces the CO₂ partial pressure in the gas phase, increasing the driving force across the membrane and avoiding the main challenges faced using HFMC in solvent regeneration, including membrane wetting, concentration polarization and degradation.^{46–48} Because more sweeping gas leads to extra heat energy consumption in CO₂ regeneration, a trade-off between CO₂ desorption performance and energy cost should be considered. However, the sweep gas flow rate had only a 5-10% effect on the overall mass transfer coefficient, since the mass transfer is mainly controlled by the liquid film resistance.^{35–37}

Temperature: The CO₂ desorption process performance improved with the absorbent temperature. For a rich-CO₂ loading liquid, a higher temperature allows for higher equilibrium partial pressures of CO₂ in the liquid, generating a greater driving force through the membrane and higher overall mass transfer coefficient values. Moreover, a higher temperature increases the chemical reaction rate and CO₂ diffusion, while the viscosity of the absorbent decreases, improving the regeneration performance. Additionally, to reduce the energy expense and avoid HFMC desorption issues, such as solvent evaporation and membrane degradation, a low-temperature process is usually suggested.^{31,49–51}

CO₂ loading: A higher CO₂ loading capacity of the absorbent promotes the CO₂ desorption flux due to the increase in the CO₂ equilibrium partial pressure in the liquid side of the membrane contactor, boosting the driving force across the membrane; it is the choice of absorbent that the CO₂ loading capacity mainly depends on.⁵² Chemical absorbents usually lead to higher CO₂ loading than physical absorbents. Nevertheless,

lower values of the loading change ($\Delta\alpha$) are often obtained with chemical absorbents since more energy is required for CO₂ separation when a chemical reaction between CO₂ and the absorbent takes place. Choosing the proper absorbent to account for the compromise between $\Delta\alpha$ and N_{CO_2} is crucial since the regeneration performance is clearly described by the loading change after the regeneration process.

Regeneration pressure: A lower pressure in the gas phase of the desorbed HFMC means a higher regeneration performance for both the sweeping gas and/or vacuum operation due to the increase in the CO₂ mass transfer driving force across the membrane. In sweeping gas mode, the regeneration performance is greater than that in pure vacuum mode at the same operating pressure. However, it would also add an extra purification operation in order to perform the CO₂ separation from the inert gas. The solvent regeneration performance increases with the vacuum level. However, a higher level of vacuum increases the vacuum pump energy consumption and the risk of membrane wetting. Thus, the energy consumption of the CO₂ desorption process could be lower than that of conventional regeneration based on heat, as long as the regeneration pressure is optimized. Wang et al.⁴⁴ estimated that the energy required can be decreased within the range of 30% using a CO₂ membrane vacuum. Consequently, a relatively high regeneration ratio could be fulfilled.

Fiber geometry: The fiber dimensions of HFM contactors have an important role in regeneration performance.⁵³ On one hand, the length of the membrane improves the CO₂ desorption performance. Due to the thermodynamic limitation of the absorbent, approximately 80% of the CO₂ is desorbed through the initial 10% of the HFMC length. Therefore, the efficiency of the CO₂ desorption process will be lower through the

remaining HFMC length, since a CO₂ equilibrium between both the liquid and gas phases has been reached.

On the other hand, the CO₂ molecules are desorbed, and their diffusion from the lumen to the liquid/gas interphase takes less time when smaller fiber diameters are used, which increases the regeneration performance. Nonetheless, a smaller fiber diameter means a higher pressure drop through the HFMC, which means more power is required to drive the liquid through the fiber. The pressure drop is usually described as the difference in pressure between both the inlet and outlet of the HFMC. The effect of this variable on the regeneration performance is explained in section 4: Modeling CO₂ desorption.

2.2. Major challenges

To fulfil the state-of-the-art assessment of solvent regeneration in HFMCs, some challenges would be faced to establish this technology as a large-scale commercial alternative, including wetting the membrane, fouling and energy consumption.⁵⁴

2.2.1. Wetting phenomena

In the HFMC stripping process, the wetting phenomena are the main challenge to overcome since the reduced gas phase operation pressure increases the mass transfer resistance via solvent penetration inside the pores of the membrane, which leads to a sharp drop in the regeneration performance. The parameter used to evaluate membrane wetting by a solvent is the breakthrough pressure (Δp_b), defined as the minimum pressure implemented on the liquid phase that allows the solvent to enter the membrane pores.⁵⁵ This parameter can be calculated by the Laplace-Young equation, since the wetting

phenomena mainly depend on the liquid surface tension (γ), the contact angle between both the liquid solvent and membrane surface (θ) and $d_{p,max}$, which is the maximum pore diameter of the membrane.

$$\Delta p_b = \frac{4\gamma \cos \theta}{d_{p,max}} \quad (3)$$

After the breakthrough pressure has been reached, wetting phenomena occur before in pores with higher diameters and continue with smaller pores until the membrane turns entirely wetted, as shown in Figure 6(2). To avoid membrane wetting in practical applications, the breakthrough pressure should always be above the transmembrane pressure.

The breakthrough pressure is normally greater for membranes with smaller pore size. So far, membrane contactors with membrane pore size lower than 200 nm is normally used for CO₂ capture technology, since higher pore size could mean solvent penetration and consequently a CO₂ mass transfer reduction. Preferably, the membrane should be fabricated by a high hydrophobic surface, comprised of a thin film with small pores, on a highly porous support with a great pore size.

Figure 6. Diagram of the theoretical behaviors of a microporous membrane. (1) No membrane wetting, (2) membrane wetting, and (3) regeneration mode via a vacuum system. Adapted with permission from Xu et al.⁵¹ Copyright (2019) Elsevier B.V.

Moreover, long-term operation results in morphological changes in the membrane and may also decrease the membrane contact angle (Figure 7), which reduces the minimum breakthrough pressure required to maintain a high regeneration performance. The increase in the size of membrane pores over the operation time is a result of absorbent molecule penetration increasing the lateral forces inside the pore. Therefore, the CO_2 desorption process based on HFMC and operated without membrane wetting is a key point to guarantee long-term application.⁵⁶

Figure 7. SEM images of polypropylene membrane in a HFMC. (a) new; (b) 480 h of running. Reprinted with permission from Fang et al.³¹ Copyright (2012) Elsevier Ltd.

For this purpose, different membrane materials have been extensively studied.^{10,57} Ceramic membranes offer better chemical and thermal stabilities and can be used at elevated temperatures and/or in operations with hard chemical requirements since most polymeric HFMCs are not able to resist temperatures above 100°C because of their poor ability to withstand some chemical reactions and high temperatures.⁵¹ However, since the regeneration temperature in HFMCs is usually lower than 100°C, polymeric membrane contactors have some advantages over other materials, such as hydrophobicity and a low cost.²² Most commercially available gas separation membranes have been fabricated from polymeric materials because of the availability of numerous suitable polymers and the ease of membrane fabrication with desirable properties from different polymeric materials. Therefore, in this paper we focus on carbon dioxide separation via polymeric membrane technologies. Hydrophobic polymeric microporous membranes, especially

membranes made of polypropylene (PP), polyvinylidene fluoride (PVDF) and polytetrafluoroethylene (PTFE), are commonly used due to their low-cost fabrication and wide range of chemical and morphological tunability.³⁰ However, in recent years more research has been done in the field of membrane material type and membrane selection to avoid the main issues of CO₂ capture using membrane contactors. The main advancements and future directions of research in this field are stated in section 2.3.

2.2.2. Membrane fouling

Generally, microporous membrane processes have the potential issue of membrane fouling via the accumulation of contaminants in porous membranes, which decrease the mass transfer through the membrane.³⁰ Compared to membrane wetting, fouling phenomena receive much less attention since the majority of the research work is carried out at lab- and pilot scales, in which the CO₂-rich solution is generally without contaminants (e.g., rust or sediments) and absorbent degradation has not been taken into account. However, membrane fouling investigations for solvent regeneration under realistic conditions are required, since the contaminants in the flue gas could be transferred to the absorbent in the absorption stage. As we discussed above, the fouling phenomena in the desorption process are more serious in the liquid phase; however, reduced pressures in the gas side of the HFMC in a solvent regeneration process could also promote membrane fouling in the permeate phase. For this reason, removing fine particles and contaminants from the flue gas in a pretreatment operation is required to avoid further issues in solvent regeneration. More work in the field of membrane fouling phenomena should be done using real flue gas compositions to describe the process performance more accurately.^{10,22}

2.2.3. Energy consumption

The regeneration process in HFMC has the ability to minimize the temperature required for the solvent regeneration process; however, the additional energy cost to maintain the reduced partial pressure in the gas side by sweeping gas and/or vacuum must be considered. Some reports^{31,44,49} have compared the energy demand for solvent regeneration with MEA as the absorbent and using water steam as a sweep gas in HFMC with those via conventional high-temperature desorption in a stripper. The total energy duty is estimated with no distinction in the type of energy from two competing duties, the energy to run the vacuum pump and the energy associated with the temperature to break the chemical bond between the absorbent molecules and CO₂. Figure 8 shows the energy duty contribution of both the vacuum pump and reaction to the overall process energy duty using membranes based on a thin film of nonporous Teflon AF1600 on porous polypropylene (PP).³⁹ It is important to remark that energy duty contributions will be different depending on the membrane material and their operational limits. However, considering the limitations for each material in terms of both the temperature and transmembrane pressure (discussed in section 2.2.1. Wetting phenomena), Figure 8 could create the state-of-the-art solvent regeneration energy consumption.

Figure 8. Contribution percentage (%) of both the vacuum pump and reaction energy duty to the solvent regeneration process total energy duty using a film of nonporous Teflon AF1600 on porous polypropylene (PP) as a function of temperature. Reprinted with permission from Scholes.³⁹ Copyright (2019) Elsevier Ltd.

Scholes³⁹ studied CO₂ desorption using HFMC and reduced the energy required for solvent regeneration by approximately 30% compared with conventional high-temperature desorption operating in the range of 80-100°C and 20 kPa of vacuum. Above 100°C, the MEA solution starts to vaporize, and below that temperature range, the mass transfer coefficient decreases dramatically. The latter could be extrapolated to the CO₂ desorption process using other membrane materials. Therefore, a higher temperature requires a small contribution of the vacuum pump to desorb CO₂ from the liquid, while the energy duty of the vacuum pump increases as the temperature decreases.

However, future investigations should model other membrane materials and absorbents since the interest in and the potential of HFMC in CO₂ desorption partly depend on solvent properties.

2.3. Future directions in membrane materials

The membrane material is one of the most important variables that must be studied to increase the performance of the CO₂ desorption process by solving the issues described above.

The most common materials are based on microporous organic polymers, as discussed before. These membrane materials are largely applied in CO₂ capture technology because of their hydrophobic nature and low-cost. However, some operational issues, such as chemical degradation, low mechanical stability and swelling phenomena, can be emerge in the polymeric membrane when organic absorbents are used. Therefore, membranes based on ceramic materials such as silica or zeolites may take advantage of polymeric membranes due to their thermal and chemical resistance. However, in terms of CO₂ capture performance, ceramic membranes have lower CO₂ selectivity and permeability than polymeric membranes. In addition, ceramic HFMCs also have higher cost and lower membrane area per contactor's volume, which decrease the process efficiency by using the same module dimensions.

Different fabrication methods have been used for the development of extra-hydrophobic membranes with large mechanical, thermal and chemical resistances for long-term performance. For mixed matrix membranes (MMM) that mainly combine polymers with ceramic materials, carbon molecular sieves, nanoparticles, etc., Zhang et al.⁵⁸ incorporated nanoparticles and fluorinated silica on PEI HFM by using a triple-orifice spinneret; Naim et al.^{59,60} calculated the CO₂ desorption flux and stripping efficiency of PVDF and PEI hollow fiber membranes containing different types of

nonsolvent additives; Dashtarzhandi et al.⁶¹ improved the membrane morphology by the deposition of inorganic particles in the polymeric membrane of an HFMC; Cersosimo et al.⁶² developed thermally rearranged (TR) polymeric HFM that showed superior mass transfer gas properties over conventional polymeric membranes; and Hashemifard et al.⁶³ performed a heat treatment on a membrane contactor with hollow fibers by exposing them to temperatures of 80°C for 5 minutes under a hot air atmosphere before the desorption process, enhancing the performance of the CO₂ stripping process.

In general, mixed matrix membranes exhibit higher mechanical properties than pure polymer membranes and have a lower cost than inorganic (mineral) membranes.⁶⁴ However, this research field requires more focus in the next years since studies in promising membrane materials for CO₂ desorption in HFMCs are still scarce.

In spite of the promising results after evaluating the studies of the literature in the area of HFMC technology for carbon capture, it is still necessary an overall analysis under industrial conditions to establish this technology as a competitive alternative for the CO₂ capture technology.⁶⁵

3. Absorbent selection

Post-combustion carbon capture technology based on the absorption-desorption process using solvents to remove CO₂ from flue gas is the most studied and applied method.⁶⁶ The absorbents used for this technology present several disadvantages, such as absorbent vaporization and deterioration, which decrease the performance capacity of the solvent regeneration process.⁶⁷ On this basis, absorbent selection is an important task to reduce both the capital and operational costs, which makes CO₂ capture more effective and commercially viable. Although research studies in the field of absorbent selection from the solvent regeneration perspective are scarce,⁶⁸ some remarks from the recent literature are given as follows:

Styring⁶⁹ made comparisons between the chemical interactions of different liquid solvents and CO₂ by the percentage of carbon dioxide captured by the absorbents; Mumford et al.⁷⁰ reviewed numerous carbon capture pilot plants of physical and chemical solvents ranging from organic to inorganic absorbents; Budzianowski⁷¹ discussed the different types of commercial absorbents and developed selection criteria in large-scale carbon capture processes based on CO₂ loading capacity, chemical reaction, viscosity, volatility, deterioration, environmental effects, CO₂ desorption energy consumption and cost; Vega et al.⁷² stated that chemical absorbents are more suitable for industrial processes conducted at atmospheric pressure and flue gas with low CO₂ concentrations, while physical absorbents are limited to high-pressure flue gas processes; Papadopoulos et al.⁷³ evaluated the computer-aided molecular design method (CAMD) as an effective absorbent selection tool for carbon capture technology; Sreedhar et al.²³ provided solvent classification, synthesis protocols and a comparison of their performance in carbon capture; Mota-Martinez et al.⁷⁴ concluded that solvent viscosity, CO₂ loading and the

chemical reaction rate mainly determined the CO₂ capture cost, and Borhani and Wang⁷⁵ reviewed the different absorption-desorption mechanisms (chemisorption, physisorption and chemisorption) using different available methods, such as experimental data, predictive models, and CAMD.

In general, solvents used in conventional carbon capture technology could be proper candidates for HFMC technology, while some considerations, such as low viscosity, high surface tension and absorbent-membrane compatibility, are often necessary. Available studies on solvent selection reviews in HFMC carbon capture have focused on CO₂ absorption into the solvent.⁷⁶ However, there is relatively little information in the literature on solvent selection for CO₂ stripping (desorption), although the stripping unit is the most energy-consuming process in carbon capture.⁴⁵ Rahim⁴¹ studied the ease of CO₂ stripping from liquid absorbents in HFMC by modeling and experimental tasks using alkanolamines, amino acid salts and aqueous ammonia. Li and Keener¹⁷ introduced membrane technology as a promising activity to decrease regeneration costs and reviewed different amine-based solvents.

This section of the review evaluates the main pros and cons of the most commonly used absorbents in CO₂ solvent regeneration. Table 2 summarizes the main group of absorbents with the most experimentally tested solvents of each group, as well as their merits and demerits. Since there is a potential to replace the conventional desorption scrubber in solvent regeneration with a membrane contactor, the available studies on CO₂ desorption from liquid absorbents by HFMC are also reviewed. Table 3 shows a compilation of the regeneration flux performances of various absorbents from experimental studies using HFMC. Experimental results have found that solvent selection is a very important task since the CO₂ loading in the liquid and process parameters, such as temperature, flow rate and liquid concentration, are closely related to the solvent regeneration performance using

HFMC.⁷⁷ This evaluation will facilitate the reader to recognize different types of solvents, which provides a blueprint for solvent selection in the field of CO₂ desorption using HFMC as a promising carbon capture technology.

3.1. Amine-based solvents

Alkanolamines are low-cost basic solutions that have been established as the most commonly used absorbent type for carbon capture technology.⁷⁶ Depending on the central nitrogen molecule substitution, three groups of amine-based solvents can be distinguished: single, double and triple substitution represent primary, secondary and tertiary amines, respectively.

Monoethanolamine (MEA) is the most commonly used primary amine because of its easy production and great chemical reaction rate.⁷² However, the use of MEA as an absorbent with conventional direct gas-liquid contact operations has been related to some environmental issues, such as absorbent losses due to solvent volatility.¹

Since energy consumption in the conventional heating solvent regeneration process mainly depends on solvent selection, an important effort was made using both pure and blended amine-based solvents, as remarked in the following studies: Zhu et al.⁷⁸ worked with a 20% MEA - 10% DETA blended solution, increasing the absorption performance. However, the regeneration performance was slightly lower than that of the 30% MEA solution. Bernhardsen and Knuutila⁷⁹ evaluated the regeneration energy consumption using pure amino-based absorbents by changing the operational conditions, such as liquid velocity and amine concentration; Li and Keener¹⁷ calculated the energy penalty of carbon capture technology using MEA, which was approximately 25% of the power plant output energy. Nwaoha et al.⁸⁰ reviewed blending amine solvents aimed at minimizing both the solvent deterioration and the energy required for the CO₂ desorption process; Saeed et al.¹² focused on the MEA degradation understanding by studying ways to reduce

degradation, corrosion inhibition and evaluating of environmental concerns; Shi et al.⁸¹ performed experimental work using blended amine-based solutions; Zhang et al.⁸² developed a rate-based screening method to find the optimal molar ratio of a blended amine solution, and more recently Kumar and Mondal⁸³ studied the solvent regeneration performance for aqueous blends and compared it with conventional MEA from the angle of regeneration capacities and process efficiency.

In solvent regeneration using HFMC, many studies of compatibility in amine-based membrane technology were conducted from the solvent selection point of view, since membrane chemical degradation or pore wetting can occur, thus decreasing the solvent regeneration performance.

Fang et al.,³¹ Yan et al.⁸⁴ and Wang et al.⁸⁵ experimentally evaluated the relationship between solvent composition and MVR efficiency using 23 types of alkanolamines; Khaisri et al.³⁵ and Lee et al.¹⁶ analyzed the CO₂ stripping performance in monoethanolamine (MEA) using both polymeric and ceramic hollow fiber membrane contactors; Volkov et al.⁸⁶ evaluated membrane-amine solvent compatibility using PTFE flat membranes; Malakhov and Bazhenov⁸⁷ studied the desorption of CO₂ from an aqueous methyl diethanolamine (MDEA) solution through membranes made of PTMSP; Listiyana et al.⁴² conducted experiments of CO₂ desorption in PP HFMC using blended amines to increase CO₂ regeneration efficiency and reduce the cost of solvent usage; and Mohammed et al.⁸⁸ modeled CO₂ desorption from diethanolamine (DEA) solution using PVDF HFMC.

Although many alternatives of promising solvents for carbon capture are reviewed in this work, conventional high-temperature regeneration using MEA is the benchmark for full-scale applications¹⁷.

3.2. Ammonia

Ammonia (NH₃) solution is a favorable solvent mainly because of its low-cost production, great chemical stability and high CO₂ loading (approximately three times that of MEA).⁷⁶ However, its economic and operational viability is limited due to its low CO₂ reactivity and ammonia split during CO₂ capture because of high solvent volatility (normal boiling point -33°C).⁸⁹

To overcome the problem of a large amount of ammonia loss, CO₂ capture technology using NH₃ as an absorbent can be classified into two types, depending on the absorption temperature.⁷⁵ The low-temperature process (chilled ammonia process, CAP) involves the absorption of CO₂ conducted at low temperatures (2 – 10°C) to control NH₃ slip in the absorber and increase the CO₂ concentration in the regeneration at the cost of the high cooling duty required to chill the absorbent and flue gas. The ambient temperature process (ATP) (25-40°C) is known in the literature as an aqueous ammonia process, in which the absorption process occurs at room temperature and a washing column is used to solve the NH₃ slip problem.⁹⁰ The inherent advantages of CAP due to the properties of the solvent are established.⁹¹

Some research included the comparison of CAP with conventional amine-based materials from the energy consumption perspective to develop solvent selection criteria, as follows: Darde et al.⁹² demonstrated a lower regeneration energy consumption using CAP over amine-based processes; Dave et al.⁹³ conducted an interesting study by proposing several ammonia-based scenarios and compared them with conventional MEA-based processes; Rivera-Tinico and Bouallou⁹⁴ stated that NH₃ has a 3 times higher absorption capacity than most amines, such as MEA; Shakerian et al.⁹⁵ discussed both amine- and ammonia-based processes and concluded the lower regeneration energy cost of the ammonia-based process; and Hanak et al.⁹⁶ validated the ammonia-based CO₂ capture model and scaled up to a 580 MW coal power plant. The CAP process resulted

in a 4.7–15.7% reduction in the equivalent work required in the reference amine scrubbing plant.

Recent works have studied ammonia-based CO₂ capture via HFMC, since this technology offers promising perspectives to reduce the ammonia slip limited to 1% of the solvent.^{89,97} To date, some investigations have shown the potential of HFMC in combination with ammonia-based solvents to remove CO₂ from flue gases.^{21,98–100} Villeneuve et al.⁹⁹ listed the key findings of ammonia-based CO₂ capture using membrane contactors and compared that process, in terms of solvent loss, with a conventional packed column. However, their results showed that the membrane contactors did not generate a stable CO₂ capture performance, which makes scale-up and commercial applications impossible.⁹⁰ Moreover, the desorption process in ammonia-based CO₂ capture using membrane contactors has not yet been investigated. To make this technology more competitive, more research is needed to make use of the unique properties of aqueous NH₃ and to address its specific issues.

3.3. Amino acids

Amino acid salt (AAS) solutions have gained interest as an absorption liquid substitute for amines for the solvent regeneration process in carbon capture.¹⁰¹ The ionic nature of the solvent solution makes it less volatile, avoiding one of the main disadvantages of amine-based absorbents, i.e., solvent losses due to vaporization. Additionally, AAS provides the added advantage of no environmental issues, resistance against oxidation and thermal stability.^{102,103} However, AAS has some operational issues due to the solvent precipitation phenomena that appear at high CO₂ loadings, resulting in a higher desorption energy requirement to redissolve these solvent precipitates.^{75,104}

There are two different approaches in AAS solutions. On one hand, conventional amino acid salts (AASs) are formed by neutralization with an inorganic base such as potassium.

Some representative examples could be potassium salt of glycine, potassium salt of taurine or potassium salt of sarcosine.^{105–107} On the other hand, amine amino acid salts (AAAS) are formed by neutralization with organic bases such as amines. A prominent example of these AAAS is amine-3-(methyamino)propylamine (MAPA) with sarcosine (SARMAPA)¹⁰⁸ or with glycine (GLYMAPA).¹⁰⁹

AAAS were proposed to solve the high energy requirements in the desorption process since many studies reviewed by Hu et al.¹¹⁰ have shown poor desorption potential attributed to the lower equilibrium temperature sensitivity when conventional AAS were used as an absorbent.

Assessments of amine amino acid salts for CO₂ desorption have shown that they have a performance similar to MEA. Aronu's group^{106,108,109} reported in a semiquantitative study that amine amino acid salts (AAAS) using MAPA amine had a better performance than potassium salts in terms of both kinetics and desorption efficiency. Additionally, they completed pilot plant tests using SARMAPA, and the results showed that this solvent had a lower regeneration energy, a lower corrosion and reduced stripping steam (approximately 50% lower than using MEA).¹⁰¹

Additionally, previous studies proposed inorganic amino acid salts blended with amine-based solvents to decrease the absorbent surface tension, which could be significant for avoiding the wetting phenomena in HFMC.¹¹¹ However, amine regeneration using membrane contactors has not yet been studied since a greater understanding of these absorbents is required. Suleman et al.¹¹² and Ramezani et al.¹¹³ blended potassium lycinat (KLys) and MEA to study the effect on CO₂ desorption performance. A similar desorption performance was obtained by Song et al.¹¹⁴ and Park et al.¹¹⁵, who studied the desorption performance of 3 AAAS formed from potassium AAS (K-alanine, K-serine, K-glycine, K-sarcosine, K-taurine and K- α -aminobutyric) with piperazine. The net cyclic

capacities were higher than those using MEA (approximately 25%), which resulted in a lower energy being required for the solvent regeneration process.

Recently, it was also reported that amino acids can be blended with other liquids, confirming the phase change of amino acid solvents.¹¹¹ Blends of amino acid salts with ethanol,¹¹⁶ ethylene glycol,¹¹⁷ potassium carbonate,^{118,119} ammonia¹²⁰ and ionic liquid¹¹¹ have been tested as phase change amino acid solvents.

One of the key points of these new phase change absorbents is the capacity to form CO₂-rich precipitates, such as carbonates in the absorption column, which are easy to remove before the regeneration process, decreasing the energy consumption of the CO₂ desorption process. Recent studies reported an approximately 30-40% lower energy compared to using MEAs.¹²¹ However, aqueous-based phase-change amino acid solvents are a recently emerged concept, and further investigation is still required for worldwide acceptance of this energy-efficient technology.

To better understand the chemical process between the absorbent and CO₂, and to explore the potential of further applications, characterization, such as solubility, density, viscosity, and surface tension of blended solutions, is necessary. The formulation of new AAS-based solvents and experimental tasks are needed to understand the behavior between absorbents and CO₂ for future research in the field of carbon capture technology.¹⁰²

3.4. Ionic Liquids

ILs are frequently described as organic salts, which have melting temperatures lower than 100°C. ILs are commonly composed of both asymmetric organic cations and organic or inorganic anions.¹⁰ Ionic liquids (ILs) are presented as the most promising absorbents to solve current CO₂ capture technique issues due to their high capital and operating costs

from their large equipment, solvent degradation, high solvent losses, the corrosive nature of solvents and high energy consumption in solvent regeneration.¹²²

One of the main characteristics of ILs is the flexibility in their formulation and fabrication, which gives them special properties over other absorbents, such as a low volatility and thermal stability.^{123,124} However, some issues of the majority of ILs are their high price and high viscosity due to their composition being based on ions. A higher viscosity may reduce the diffusion rate of CO₂ in the bulk ionic liquid, which decreases the CO₂ mass transfer and makes their use as an absorbent in carbon capture processes more challenging.³³ To avoid time-consuming measurements and complicated interpretation procedures due to the high flexibility in solvent design, molecular modeling and simulation software are commonly used to evaluate the solubility of CO₂ in ionic liquids estimated from such operational parameters as temperature and pressure and the properties of ionic liquids (w, T_c, P_c).^{125,126}

Recently, important studies on carbon capture performance using ILs have been discussed in some reviews,^{127,128} including interesting works with traditional ILs, as well as other new IL-based absorbents such as switchable ILs and functionalized ILs. Chen et al.,¹²⁹ Lu et al.¹³⁰ and Li et al.¹³¹ confirmed that functionalized ILs exhibit a significantly high speed for CO₂ capture, excellent absorption capacity and easy regeneration; Lu et al.¹³² and Xiao et al.¹³³ proved that IL blending solvents yield high cycling CO₂ capacity, energy savings, good regenerability and especially a low viscosity; Privalova et al.,¹³⁴ Li et al.¹³¹ and Zheng et al.¹³⁵ studied switchable ILs as promising IL-based solvents due to their low cost and ability to be recycled. Moreover, blending these ILs with polymeric membranes to make IL-based membranes could decrease the IL viscosity, which led to an increase in the solvent regeneration process efficiency. In that field, IL-based membranes were deeply reviewed.^{124,136} These have been classified into (1) supported

ionic liquid membranes (SILMs); (2) poly(ionic liquid) membranes (PILMs); (3) poly(ionic liquid)–ionic liquid (PIL–IL) composite membranes; (4) polymer-ionic liquid composite membranes; and (5) ion-gel membranes. However, information about CO₂ desorption from these recent IL-based processes is limited, and more research is needed to define the solvent selection criteria in terms of the CO₂ desorption performance.

More work has been performed in conventional ILs and their potential to increase regeneration performance compared with amino-based technology: De Riva et al.¹³⁷ studied 8 ILs in a commercial packing column and the importance of IL properties in solvent regeneration efficiency; and Shiflett et al.,¹³⁸ Khonkaen et al.¹³⁹ and Ma et al.¹⁴ reported that simulation tasks reduced the energy consumption (approximately 15-30%) of the CO₂ desorption process by using different ILs compared to MEA.

Additionally, solvent regeneration in HFMC based on conventional ILs has been documented by Mulukutla et al.³² and Bazhenov et al.¹⁴⁰ who studied IL-based membrane contactors focused on solvent-membrane compatibility. Lu et al.¹⁴¹ and Simons et al.³³ covered the design, modeling and construction of an experimental apparatus of low-temperature CO₂ desorption using different ILs to address the influence of several parameters on the steady-state carbon capture rate.

IL-based membranes and IL-based absorbents are thus presented as promising alternatives to amine-based technology, allowing for process intensification. However, such challenges as wetting of the membrane, the presence of another compound in the gas stream, limited long-term stability, solvent issues and solvent regeneration processes should be improved to make this technology more competitive on an industrial scale.

3.5. Nanofluids

The proper choice of solvent is a determinant of the CO₂ desorption process performance. Various single absorbents, such as alkanolamines, ammonia, amino acids and ionic liquids, have been studied for solvent regeneration.⁴² Recently, a mix of pure solvents has been presented by several studies to improve the performance of CO₂ separation. Some of these mixtures of solvents were reviewed in this work before; for example, IL- blended with amine solvents, the problem of high viscosity, or amino acids blended with carbonates, which decrease the surface tension of the solution and increase the cyclic capacity. Nanofluids are a new concept of solvents that involve a nanometer-scale material, such as nanoparticles dispersed into a fluid. The nature of nanoparticles could be broad. However, studies in carbon capture using nanofluids are mainly limited to SiO₂, TiO₂, and Al₂O₃ dispersed into water or MEA.¹⁴² Although these nanoparticles are dispersed in a fluid, the CO₂ adsorption mechanism could be neglected due to their reduced mass fraction. Therefore, CO₂ absorption into the nanofluid is considered.¹⁴³

The use of nanofluids as a promising solvent for carbon capture technology has been evaluated in recent years. The main advantages of these absorbents over conventional MEA solutions are an increase in the heat and mass transfer performance due to the large surface area provided by the dispersed nanoparticles. Moreover, the thermal stability and higher diffusivity have been tested by the literature in this field.¹⁴⁴

The improvement of gas solubility into the absorbent by the addition of a third dispersed phase, i.e., nanoparticles, was investigated by several researchers.^{145–147} However, a few studies have been conducted on CO₂ desorption: J.W. Lee et al.¹⁴⁸ and Lee et al.¹⁴⁹ compared the CO₂ regeneration efficiency of nanofluids (Al₂O₃ and SiO₂ nanoparticles dispersed in DI water and methanol) and pure solvents. The use of nanofluids enhanced

the desorption performance by approximately 10-20% in all cases; J.H. Lee et al.¹⁵⁰ proposed that the morphological change by the nanoparticles during the CO₂ regeneration process is the most influential mechanism of the regeneration performance improvement; Wang et al.⁴⁰ and Yu¹⁵¹ compared the CO₂ desorption efficiency using a base solution of MEA (30 wt.%) and dispersed SiO₂, TiO₂, and Al₂O₃ nanoparticles into that base solution. Later, the CO₂ desorption performance was increased by approximately 10%. Using the same nanoparticles dispersed on a tertiary amine solvent (TETA); and Jiang et al.¹⁵² studied the solvent regeneration energy consumption. Nanofluids showed a lower energy consumption than pure TETA since nanofluids cause more heat to be distributed to the desorption due to a higher heat flux.

In the research area of HFMC technology, nanofluids are considered promising absorbents for the membrane separation process because of the great results achieved in carbon capture.¹⁵³ However, studies for CO₂ capture using nanofluid absorbents coupled with HFMC are insufficient.

822 Table 2. Overview of the group of solvents used in CO₂ desorption.

Group	Merits	Demerits	solvent commonly tested in CO ₂ desorption	References
Amines	low cost, high CO ₂ reactivity	energy-intensive, degradation and corrosion in an oxidizing atmosphere, limited loading capacity, limited loading capacity	MEA (primary), DEA (secondary), MDEA (tertiary), PZ (cyclic amine); (5-30 w.t.%)	17
Ammonia	high absorption capacity, easy availability, low regeneration costs, removes SO ₂ , NO _x , used as fertilizers	low reactivity, high volatility with ammonia split, unwanted side reactions	Aqueous ammonia (5–20 w.t.%)	99
Amino Acids	non-toxic, thermal and chemical stability, low regeneration energy, easy availability low vapor pressure	solvent precipitation, high energy required in regeneration	Potassium salts of Taurine, Glycine and sarcosine (AAS); SARMAPA and GLYMAPA (AAAS)	111,154
Ionic liquids	non-inflammability, non-explosivity, high thermal and chemical stability, high reaction selectivity, low vapor pressure	high cost of new ILs, high viscosity, low mass transfer rates	imidazolium room temperature IIs (e.g., [bmim][BF ₄], [emim][Ac], [emim][EtSO ₄])	140
Nanofluids	low viscosity, high heat and mass transfer, low regeneration energy	high cost, lack of experimental data	SiO ₂ , TiO ₂ , and Al ₂ O ₃ particles in water or amines	155

823 Table 3. CO₂ Regeneration flux performances of various absorbents using hollow *fiber*
824 membrane contactors

Absorbent	Membrane material/configuration	Specific area (m ²)	Vacuum pressure (kPa)	Sweep gas flowrate (L·min ⁻¹)	Liquid flowrate (L·min ⁻¹)	Temperature (°C)	CO ₂ stripping flux (10 ⁴ mol m ⁻² s ⁻¹)	Reference
[emim][Ac]	PP/parallel	0.18	4	-	0.06	40	0.252	43
[emim][EtSO ₄]	PP/parallel	0.18	4	-	0.06	40	0.00738	
Aqueous [bmim][BF ₄]	PP/parallel	0.51	50	-	0.08-0.28	24	47.3	141
Aqueous [apmim][BF ₄]	PP/parallel	0.51	50	-	0.08-0.28	24	2.36	
Aqueous PG + AMP	PP/parallel	0.27	60	-	0.04-0.2	24	21.3	156
Aqueous PG + AMP	PP/parallel	0.27	-	2 (N ₂)	0.04-0.2	24	0.426	
MEA 20 w.t. %	PP/parallel	0.13	10	1.15 · 10 ⁻³ (steam)	0.008-0.05	65	1.8	38
MEA 30 w.t. %	PP/parallel	0.21	20		0.02	75	1.31	34
MEA 30 w.t. %	PP/parallel	0.21	20	5 · 10 ⁻⁴ (steam)	0.02	75	4	

DEA 30 w.t.%	PP/parallel	0.21	20		0.02	75	2.8
DEA 30 w.t.%	PP/parallel	0.21	20	$5 \cdot 10^{-4}$ (steam)	0.02	75	4
TEA 30 w.t.%	PP/parallel	0.21	20		0.02	75	0.57
TEA 30 w.t.%	PP/parallel	0.21	20	$5 \cdot 10^{-4}$ (steam)	0.02	75	0.74
MDEA 30 w.t.%	PP/parallel	0.21	20		0.02	75	0.93
MDEA 30 w.t.%	PP/parallel	0.21	20	$5 \cdot 10^{-4}$ (steam)	0.02	75	1
EDA 30 w.t.%	PP/parallel	0.21	20		0.02	75	2.51
EDA 30 w.t.%	PP/parallel	0.21	20	$5 \cdot 10^{-4}$ (steam)	0.02	75	4.66
DETA 30 w.t.%	PP/parallel	0.21	20		0.02	75	1.66
DETA 30 w.t.%	PP/parallel	0.21	20	$5 \cdot 10^{-4}$ (steam)	0.02	75	2.9
TETA 30 w.t.%	PP/parallel	0.21	20		0.02	75	2
TETA 30 w.t.%	PP/parallel	0.21	20	$5 \cdot 10^{-4}$ (steam)	0.02	75	2.42
TEPA 30 w.t.%	PP/parallel	0.21	20		0.02	75	1.55
TEPA 30 w.t.%	PP/parallel	0.21	20	$5 \cdot 10^{-4}$ (steam)	0.02	75	1.87
AMP 30 w.t.%	PP/parallel	0.21	20		0.02	75	3.66

AMP 30 w.t.%	PP/parallel	0.21	20	$5 \cdot 10^{-4}$ (steam)	0.02	75	3.95
AEPD 15 w.t.%	PP/parallel	0.21	20		0.02	75	0.696
AEPD 15 w.t.%	PP/parallel	0.21	20	$5 \cdot 10^{-4}$ (steam)	0.02	75	0.831
AEPD 30 w.t.%	PP/parallel	0.21	20		0.02	75	1.71
AEPD 30 w.t.%	PP/parallel	0.21	20	$5 \cdot 10^{-4}$ (steam)	0.02	75	1.94
THAM 30 w.t.%	PP/parallel	0.21	20		0.02	75	1.26
THAM 30 w.t.%	PP/parallel	0.21	20	$5 \cdot 10^{-4}$ (steam)	0.02	75	1.16
3-AP 30 w.t.%	PP/parallel	0.21	20		0.02	75	0.863
3-AP 30 w.t.%	PP/parallel	0.21	20	$5 \cdot 10^{-4}$ (steam)	0.02	75	2.18
2-AB 30 w.t.%	PP/parallel	0.21	20		0.02	75	2.63
2-AB 30 w.t.%	PP/parallel	0.21	20	$5 \cdot 10^{-4}$ (steam)	0.02	75	3.82
AP 30 w.t.%	PP/parallel	0.21	20		0.02	75	0.728
AP 30 w.t.%	PP/parallel	0.21	20	$5 \cdot 10^{-4}$ (steam)	0.02	75	3.98
PDA 30 w.t.%	PP/parallel	0.21	20		0.02	75	2.9
PDA 30 w.t.%	PP/parallel	0.21	20	$5 \cdot 10^{-4}$ (steam)	0.02	75	3.25

HDA 30 w.t.%	PP/parallel	0.21	20		0.02	75	8.23	
HDA 30 w.t.%	PP/parallel	0.21	20	$5 \cdot 10^{-4}$ (steam)	0.02	75	2	
[bmim][DCA]	PP/crossflow	0.05	-	$4.7 \cdot 10^{-2}$ (He)	0.0027 - 0.0044	79	0.1	32
[bmim][DCA]-20 w.t.% PAMA M Gen 0 Dendrimer	PP/crossflow	0.05	-	$2.3 \cdot 10^{-2}$ (He)	0.0027 - 0.0044	75	1.79	
[bmim][DCA]-20 w.t.% PAMA M Gen 0 Dendrimer	PP/crossflow	0.05	3.99	-	0.0027 - 0.0044	85	3.32	
Pure activated MDEA with PZ	PP/crossflow	0.05	3.38	$5 \cdot 10^{-2}$ (He)	0.0042 - 0.0052	90	24.6	157
DEA 30 w.t.% activated with K_2CO_3	PP/parallel	0.041	20	-	0.1	70	18	42
DEA 30 w.t.% activated with arginine	PP/parallel	0.041	20	-	0.1	70	17	
DMPDA H acetate 50 w.t.%	Polyolefin-Polyethylene/parallel	-	8	-	0.1-0.4	40	0.65	33

PG 5.6 w.t.%	PVD F/parallel	0.003 4	-	0.2 (N ₂)	0.04	60	750	41
DEA 5 w.t.%	PVD F/parallel	0.003 4	-	0.2 (N ₂)	0.04	60	600	
AMP 4.5 w.t.%	PVD F/parallel	0.003 4	-	0.2 (N ₂)	0.04	60	400	
MEA 3 w.t.%	PVD F/parallel	0.003 4	-	0.2 (N ₂)	0.04	60	500	
Water	MM M PVD- MMT /parallel	0.003 2	-	$1.2 \cdot 10^{-3}$ (N ₂)	0.2	27	42	61
MEA 30 w.t.%	AL ₂ O ₃ (ceramic)/ parallel	0.003	61	-	0.003- 0.009	80	45	16
MEA 30 w.t.%	PTFE /parallel	0.04	-	0.1 (N ₂)	0.027- 0.1	100	5	35
MEA 15 w.t.%	AL ₂ O ₃ (ceramic)/ parallel	0.01	-	0.5 (N ₂)	0.015- 0.075	80	24	45
MEA 18 w.t.%	PTFE /parallel	0.04	-	0.1 (N ₂)	0.027- 0.1	90	2	158
DEA 10 w.t.%	PVD F/parallel	0.003 5	-	0.1 (N ₂)	0.08	80	160	59
DEA 10 w.t.%	PVD F- PEG/	0.003 5	-	0.1 (N ₂)	0.08	80	460	

	parallel							
DEA 10 w.t.%	PVD F/parallel	0.0063	-	0.1 (N ₂)	0.2	80	3	159
Water	PSF/p parallel	0.0063	-	0.5 (N ₂)	0.2	80	0.4	
Water	PVD F/parallel	0.01	-	0.1 (N ₂)	0.05-0.2	28	0.0008	46
Water	PVD F/parallel	0.0056	-	0.1 (N ₂)	0.06-0.21	60	1.5	160
Water	PVD F-glycerol-NMP/parallel	0.0056	-	0.1 (N ₂)	0.06-0.21	50	0.45	161
DEA 10 w.t.%	PEI-PEG/parallel	0.0034	-	0.5 (N ₂)	0.2	80	350	162
DEA 10 w.t.%	PEI/p parallel	0.0034	-	0.1 (N ₂)	0.2	80	60	163
MEA 6 w.t.%	PEI-PA/p parallel	0.0034	-	0.1 (N ₂)	0.027-0.1	80	2	60
MEA 0.6 w.t.%	PEI/p parallel	0.011	-	0.1 (N ₂)	0.2	80	5.1	164
MEA 6 w.t.%	PSF/p parallel	0.045	-	0.1 (N ₂)	0.05-0.3	80	2	165
[emim][MS]	PP/p parallel	0.18	-	0.1 (N ₂)	0.06	15	0.49	36
[emim][DCA]	PP/p parallel	0.18	-	0.1 (N ₂)	0.06	15	1.13	

4. Modeling CO₂ desorption

Compared to exhaustive modeling and simulation studies on the CO₂ absorption process, investigations in the area of modeling the solvent regeneration process using HFMC and CO₂ mass transfer through the membrane are scarce.

According to the literature, equivalent to the absorption process, the resistance-in-series model can describe the resistance of the CO₂ mass transfer from the liquid phase to the gas phase through the membrane. However, the mass transfer process of CO₂ in a desorption process is from the CO₂-rich absorbent to the permeate side.

This section summarizes the mass transfer correlations and modeling approaches developed for the CO₂ desorption process using HFMC. The first task of any modeling strategy corresponds to the definition of the input variables that can be classified into the following categories, as shown in Figure 9: membrane material (porosity, packing factor, surface properties, etc.); liquid absorbent (viscosity, density, CO₂ solubility, chemical reaction, etc.), compatibility between the membrane material and solvent characteristics (surface tension, contact angle, etc.); operating conditions (temperature, vacuum pressure, flow rates, etc.) and flow configurations.¹⁰

Figure 9. A general outline of process modeling and simulation for post-combustion carbon capture technology using HFMC. Reprinted with permission from Zhao et al.¹⁰ Copyright (2016) Elsevier B.V.

As mentioned before, a broad range of liquid absorbents and membrane materials could be applied in the CO₂ solvent regeneration process using HFMC. The temperature is usually lower than 100°C, and sweep gas and/or vacuum pressure is applied on the gas permeate side to allow for the mass transfer driving force. In terms of the fluid flow configuration conditions, the liquid can flow on the tube or shell side. A detailed comparison of these two flow modes was developed in some previous reviews.^{49,76,166}

Some issues, such as canalization of the fluid (channeling and dead zones) and high shell side pressure drop, are related to when the liquid flows in the shell side, which could decrease the overall efficiency of the process. Therefore, the liquid through the tube

configuration is commonly used in the literature. However, in large-scale applications, some issues could be considered when the liquid flows through the tube, such as a higher mass transfer resistance and a higher pump cost due to the small diameter of the fibers.¹⁶⁷

Additionally, since sweep gas could be applied in the permeate side of the HFMC to increase the solvent regeneration performance, two different types of contactors are considered in the literature, depending on the flow directions: parallel and cross-flow.²⁹ In the parallel-flow module, the two-phase fluids circulate in parallel. Each liquid-phase flows in one side of the membrane contactor. The liquid and gas fluids can flow co-currently or counter-currently in relation to each of them.¹⁶⁸ The latter provides a higher driving force for CO₂ desorption, enhancing the mass transfer rate.¹⁶⁹ Parallel flow in the countercurrent configuration can be easily operated and fabricated. However, channeling issues could appear on the shell side, which leads to a lower CO₂ desorption performance compared to the cross-flow module configuration.⁷⁷ In cross-flow operation, the mass transfer is parallel to the flow and perpendicular, which is usually created by a baffle configuration inside the module (Figure 10). From an experimental perspective, cross-flow is favored since it offers many advantages compared to parallel flow, such as large mass transfer coefficients, lower pressure drop on the shell side, well-defined flow conditions on both phase fluid sides and a lower mass transfer resistance due to the remixing effect.²²

Figure 10. Parallel and crossflow configurations in HFMC. Reprinted with permission from Scholes and Shen.⁴⁹ Copyright (2018) The Chemical Industry and Engineering Society of China, and Chemical Industry Press.

Taking these considerations related to the input variables into account, the target of modeling and simulation studies is to describe the efficiency of HFMC in the solvent regeneration process and predict the experimental results, as well as the sensitivity analysis.^{33,44,84,156,170} Three modeling approaches of different complexity were considered for the modeling tasks of a membrane contactor CO₂ desorption process based on a single fiber of the membrane contactor.¹⁰ Table 4 outlines the assumptions for each model approach. In this section, we analyze the main modeling possibilities presented in the literature to find a balance between the model complexity and an experimental prediction of the solvent regeneration process via HFMC in a carbon capture system.

4.1. Constant overall mass transfer coefficient approach

The constant overall mass transfer coefficient method considering mixing cup concentrations is the simplest model of the CO₂ desorption process using membrane contactors reported in the literature.¹⁰ The overall mass transfer through the membrane could be divided into three steps: the transfer of CO₂ through the liquid side boundary layer, the transfer across the membrane and the transfer of CO₂ through the gas side boundary layer. A schematic representation of the mass transfer stages is presented in Figure 4. Each stage is related to a specific mass transfer resistance (Figure 11), and consequently, the overall mass transfer coefficient (K_{ol}) could be described by the resistance-in-series model for gas desorption in an HFMC process without wetting phenomena:⁵¹

$$\text{Liquid in the lumen side: } \frac{1}{K_{ol}} = \frac{1}{Ek_l} + \frac{Hd_i}{k_m d_{ln}} + \frac{Hd_i}{k_g d_o} \quad (4)$$

$$\text{Liquid in the shell side: } \frac{1}{K_{ol}} = \frac{1}{Ek_l} + \frac{Hd_o}{k_m d_{ln}} + \frac{Hd_o}{k_g d_i} \quad (5)$$

where (k_g), (k_m) and (k_l) are the mass transfer coefficients of each gas, membrane and liquid side, respectively; (d_o), (d_i) and (d_{ln}) are the outer, inner and logarithmic mean diameters of each membrane contactor fiber; (H) is the dimensionless Henry's Law constant, which can be estimated from the experimental data of CO₂ solubility in the absorbent; and (E) is the enhancement factor, which defined the improvement in the mass transfer coefficient due to the chemical reaction between CO₂ and the absorbent selected in the liquid-side diffusion boundary layer.^{171,172}

Figure 11. Sketch of the resistance- in-series model.

Additionally, the calculation of the overall mass transfer coefficient can be simply estimated using equation 6. The ideal plug flow consideration could be assumed due to the constant surface area of the membrane fiber.

$$K_{ol} = \frac{Q_l}{A} \frac{C_{l,out} - C_{l,in}}{\Delta C_{l,m}} \quad (6)$$

In this expression, Q_l is the liquid flow rate ($\text{m}^3 \cdot \text{s}^{-1}$); $C_{l,out}$ and $C_{l,in}$ are the gas concentrations of the liquid in both the outlet and inlet ($\text{mol} \cdot \text{m}^{-3}$); $\Delta C_{l,m}$ is the liquid concentration expressed as the logarithmic mean driving force ($\text{mol} \cdot \text{m}^{-3}$); and A is the gas-liquid contact area (m^2).

In typical circumstances, some correlations are used to estimate each local mass transfer coefficient (k_g , k_m and k_l). For (k_g) and (k_l), equations based on the Sherwood number, as described in equation 7, have been reported in the literature, where d is the diameter (m) and D_g and D_l are the diffusion coefficients of CO_2 in both the gas and liquid phases, respectively ($\text{m}^2 \cdot \text{s}^{-1}$).¹⁷³ However, it is important to note that (k_g) is usually negligible

compared to the other terms since the mass transfer resistance in the liquid phase is the main resistance in the process.³⁵

$$k_g = Sh_g \frac{D_g}{d} ; k_l = Sh_l \frac{D_l}{d} \quad (7)$$

For liquid flow in the tube side of the HFMC, mass transfer across the solvent boundary layer ($k_{l,t}$) is described through three different Sherwood correlations developed by Yang, Kreulen, and Sieder.^{174–176} The different correlations were compared by Scholes and Shen⁴⁹, who indicated that for the solvent regeneration process, the correlation developed by Yang and Cussler based on Reynolds and Schmidt dimensionless numbers, with two options available dependent on the Graetz Number (a measurement of where laminar flow has been established in the fiber) (equation 8), is expected to be the most robust model.¹⁷⁴

$$Sh_t = \frac{k_{l,t} d_i}{D_l} = 0.5 \left(\frac{d_i}{L} Re Sc \right); Gz = \frac{d_i}{L} Re Sc < 6$$

$$Sh_t = \frac{k_{l,t} d_i}{D_l} = 1.62 \left(\frac{d_i}{L} Re Sc \right)^{\frac{1}{3}}; Gz > 6 \quad (8)$$

For solvent on the shell side of the HFMC, the mass transfer evaluation is complex since the geometry and the distribution of each fiber in the HFMC could influence the liquid flow profile. The mass transfer coefficient of the shell side ($k_{l,s}$) can be calculated from the Sherwood number by equation 9, where (dh) and (L) are the contactor hydraulic diameter and length, respectively, (A) is the specific contact area, (ϕ) is the packing factor density, and (Re) and (Sc) are Reynolds and Schmidt dimensionless numbers, respectively:

$$Sh_s = \frac{k_{ls}d_h}{D_l} = Af(\varphi) \left(\frac{d_h}{L}\right)^\alpha Re^\beta Sc^\gamma \quad (9)$$

A deep evaluation of the mass transfer correlations for both the parallel flow and cross-flow was conducted by Scholes and Shen.⁴⁹ The comparison stated that the Kreith and Black correlation is acceptable for CO₂ desorption process modeling for cross-flow behavior, while Yang and Cussler's correlation is in good agreement for parallel flow.

Kreith and Black:

$$Sh_s = 0.39Re^{0.59}Sc^{0.33}; Re < 2100$$

$$Sh_s = 0.116(Re^{0.66} - 125) \left[1 + \left(\frac{d_h}{L}\right)^{0.66}\right]; 2100 < Re < 10000 \quad (10)$$

Yang and Cussler:

$$Sh_s = 1.25 \left(\frac{d_h}{L} Re\right)^{0.93} Sc^{0.33} \quad (11)$$

The membrane mass transfer coefficient (k_m) can be estimated following Knudsen diffusion in nonwetting conditions (equation 12), where D_m is the CO₂ diffusion coefficient in the membrane (m² s⁻¹), and ε , τ , δ are the membrane porosity (-), tortuosity (-) and thickness (m), respectively.

$$\frac{1}{k_m} = \frac{\tau\delta}{D_m\varepsilon} \quad (12)$$

Since the membranes are hydrophobic, the membrane pores filled with gas are usually assumed to be long as the pressure difference between both liquid and gas phases does not exceed the membrane wetting (breakthrough) pressure. The wetting phenomena produced mainly by liquid breakthrough increase the membrane mass transfer resistance.

However, to model the partial membrane wetting phenomena, equation 13 is usually proposed in the reviewed literature.

$$\frac{1}{k_m} = \frac{(1-\phi)\tau\delta}{D_m\varepsilon} + \frac{\phi\tau\delta}{D_l\varepsilon} \quad (13)$$

ϕ is considered as the membrane pore wetting ratio, which could be described by the breakthrough pressure,²⁵ as defined in section 2.2.1: Wetting phenomena. Although these models can be valid for a wide estimation or particular conditions, such as a high excess CO₂ on the liquid side of the process, they are not very useful to determine the main parameters of the CO₂ desorption model, since the regeneration performance mainly depends on the CO₂ desorption flux variation through the fiber length.¹⁷⁷ This model is valuable for identifying the phase acting as a bottleneck for mass transfer and understanding the desorption process from a theoretical point of view. However, more rigorous model approaches are required to describe large-scale applications.¹⁷³

4.2. One-dimensional modeling (1D)

In the one-dimensional model of CO₂ desorption in HFMC, the mass transfer of CO₂ across the membrane considering mixing cup concentrations is described by the boundary layer theory. As Figure 12 shows, each membrane fiber of the HFMC is divided into many small elements (ΔZ) along the length of the desorption module, and the mass balance equations of CO₂ in each element are calculated by the membrane flux (N) from their local values of the driving forces. The mass balance of CO₂ desorbed from the liquid phase of a small segment in the z-direction of the membrane contactor can be written as:

$$\frac{dF_{g \text{ (or l)}}}{dz} = n\pi d_o N \quad (14)$$

999 in which either the CO₂ molar flow rate in the gas phase or the dissolved gas CO₂ in the
1000 liquid phase would be F (mol s⁻¹), and n refers to the number of fibers.

1001 Some important considerations must be considered. On one hand, both liquid and gas
1002 flow are considered laminar and constant with the established axial velocity profiles. On
1003 the other hand, the entire HFMC is usually operated in isothermal conditions by
1004 submerging it in a water bath. To our knowledge, two studies of the one-dimensional
1005 model of CO₂ desorption using HFMC under adiabatic conditions were found in the
1006 literature.^{39,178}

1007 The local mass transfer flux of dissolved gas (N , mol m⁻² s⁻¹) from the liquid to gas bulk
1008 can be commonly expressed using the overall mass transfer coefficient (K_{ol}) and CO₂
1009 concentration difference as the local driving force, where C_l (mol m⁻³) is the CO₂
1010 concentration in the bulk liquid phase and C^* is the CO₂ concentration in the liquid phase
1011 in equilibrium with the CO₂ partial pressure in the gas phase. The latter can be predicted
1012 from the vapor-liquid equilibrium (VLE) data calculated by different thermodynamic
1013 models reviewed by Sreedhar et al.²³ In the mass transfer driving force calculation, C^* is
1014 usually reported in the form of a correlation between the partial pressure of CO₂ on the
1015 gas side (p) and Henry's constant (H):

$$1016 \quad N = K_{ol}(C_l - C^*) = K_{ol}(C_l - p \cdot H) \quad (15)$$

1017 Based on this information, the differential equation for each phase (gas and liquid)
1018 presented in equation 14 can be written as:

$$1019 \quad \frac{dF_{g(or\ l)}}{dz} = n\pi d_o N K_{ol}(C_l - p \cdot H) \quad (16)$$

In the constant mass transfer coefficient approach, the chemical reaction is conventionally modeled using the enhancement factor (E), which reduces the liquid mass transfer resistance in the overall mass transfer calculation. This approach is also assumed in the 1D model. The chemical desorption reaction only occurs in the diffusion liquid-gas boundary layer inside the membrane pores, increasing the liquid mass transfer by the enhancement factor.¹⁷⁸ The calculation of E mainly depends on the reaction regime and the CO₂ diffusion limitation in the gas-liquid interface.¹⁷⁹

Additionally, the axial profile (from z=0 to z=L) of the CO₂ concentration is affected by the pressure drop ΔP (kPa). The latter is usually fixed to ensure a good distribution of both liquid and gas phase flows.¹⁷⁷ When the liquid flows in the lumen of the fiber with laminar flow, it can be estimated using the classic Poiseuille equation.

$$\Delta P = \frac{0.128\eta L Q_l}{\pi d_i^4} \quad (17)$$

where L is the membrane length (m); η is the liquid dynamic viscosity (kPa s); Q_l is the volumetric flow rate (m³ s⁻¹); and d_i refers to the membrane fiber's inside diameter (m). The control of the pressure drop when the liquid flows in the shell is more complex to obtain since the assumption of laminar flow could not give reliable results. Moreover, studies by Albarracin-Zaida et al.¹⁷⁸ show that the shell side pressure drop is approximately 50% greater than that of the lumen. Additionally, rising the liquid pressure drop increases the likelihood of membrane wetting and fouling, as shown in section 2: Model issues.

The 1D approach, through the use of a mixed-cup concentration, is well established because the reaction film is very thin, and the concentration in the liquid phase of both reactants and products is nearly flat.^{25,45,180} However, a slower reaction CO_2 -solvent reaction or higher fiber diameter of the HFMC could very well lead to substantial radial gradients to be introduced in the 2D approach.

Figure 12. Mass balance of species i in a small segment in the z -direction of the hollow fiber. Reprinted with permission from Rongwong et al.¹⁸⁰ Copyright (2017) Elsevier B.V.

4.3. Two-dimensional modeling (2D)

Both the radial and axial profiles of CO₂ concentration over the fiber length can be modeled by a 2D approach. A 2D model is formulated from the consideration of the gas- and liquid-side convective diffusion expressions paired with the chemical reaction equations through the use of cylindrical geometry coordinates in terms of partial differential equations. Not only is the desorption module divided into small elements (ΔZ) along the length, but the radius of the fiber is also divided from the center to the shell boundary layer (ΔR).^{181,182} A common process representation of the CO₂ desorption process is described in Figure 13, where the rich CO₂ solvent is introduced into the lumen, while a sweep gas and/or vacuum is applied on the shell side.

Figure 13. A representation of the HPMC in the CO₂ desorption process. Reprinted with permission from Mehdipourghazi et al.¹⁸³ Copyright (2015) Elsevier B.V.

The desorption process model considers the solvent CO₂ concentration along the lumen of the membrane contactor to be influenced by the chemical reaction between CO₂ and

the absorbent of the contactor and the mass transfer that takes place in the gas-liquid interface. The concentration gradient of the absorbent in the liquid phase could also be considered for solvents with low initial concentrations or high volatilities, such as MEA. However, a constant solvent concentration is usually assumed in the 2D desorption model. In addition, liquid viscosity changes because CO₂ loading is neglected and isothermal conditions are usually considered in the process. Therefore, the CO₂ mass balance over each membrane contactor fiber can be expressed as equation 18, where CO₂ diffusion and a reversible chemical reaction take place simultaneously:

$$V_{z-T} \frac{\partial C_l}{\partial z} = D_l \left(\frac{\partial^2 C_l}{\partial r^2} + \frac{1}{r} \frac{\partial C_l}{\partial r} \right) - r \quad (18)$$

where C_l (mol·m⁻³) is the free CO₂ concentration on the liquid side; D_l (m²·s) is the CO₂ diffusion coefficient in the liquid; and r (mol·m⁻³·s⁻¹) is the CO₂ reaction rate, which was calculated using equation 19 (from the assumption of pseudo-first-order kinetics).

$$r = k_0 C_l = B e^{\left(\frac{-E_a}{RT}\right)} C_l \quad (19)$$

where E_a is the activation energy (KJ·mol⁻¹) and B is the pre-exponential factor of the chemical reaction equation (s⁻¹). The liquid velocity in the axial direction V_{z-T} (m·s⁻¹) on the tube side can be determined by the integration of the Hagen-Poiseuille equation, where V_{zm-T} is the mean velocity of the liquid and R_1 (m) represents the inner radius of the membrane contactor fiber.

$$V_{z-T} = 2V_{zm-T} \left[1 - \left(\frac{r}{R_1} \right)^2 \right] \quad (20)$$

Additionally, the CO₂ shell-side mass transfer is progressively governed by convection and molecular diffusion. Therefore, the continuity equations for the transport of CO₂ on the shell side can be described as:

$$D_g \left(\frac{\partial^2 C_s}{\partial r^2} + \frac{1}{r} \frac{\partial C_s}{\partial r} \right) = V_{Z-s} \frac{\partial C_s}{\partial r} \quad (21)$$

The shell side velocity profile V_{Z-s} of the HFMC is usually modeled by Happel's free surface model, although the real flow profile in the HFMC is more complex. The laminar parabolic velocity profile on the shell side of the HFMC is calculated from equation 22, where V_{Zm-s} (ms^{-1}) is the mean velocity on the shell side; r_1 and r_2 are the inner and outer fiber radii, respectively; r_3 (m) is the arbitrary radius of the shell around the fiber; and $r_{c,i}$ is the inner radius of the contactor.

$$V_{Z-s} = 2V_{Zm-s} \left[1 - \left(\frac{r_2}{r_3} \right)^2 \right] \frac{(r/r_3)^2 - (r_2/r_3)^2 + 2 \ln(r_2/r)}{3 + (r_2/r_3)^4 - 4(r_2/r_3)^4 + 4 \ln(r_2/r_3)} \quad (22)$$

$$r_3 = r_2 \sqrt{\frac{1}{1-\phi}}; 1 - \phi = \frac{Nr_2^2}{r_{c,i}^2} \quad (23)$$

The 2D modeling of the CO_2 transported from the liquid to the gas phase across the membrane required physical and chemical properties, such as CO_2 solubility in the absorbent and diffusion coefficients, for the gas and liquid phases. The finite element method is adopted for a numerical solution for a set of differential equations of the 2D model approach. Software such as Aspen Custom Modeler (ACM), MATLAB, COMSOL Multiphysics and PARDISO are commonly used.^{25,36,88,179,183–185}

The complex solution of the 2D model could be time consuming. Therefore, the development of a new model procedure based on the combination of accurate methods without high complexity must be addressed in this field.¹⁷⁹

To evaluate which model approaches should be applied to describe the CO_2 regeneration process in different situations, the following statement could be considered in future research. In our opinion, based on previous literature analyses, with rapid

reversible chemical reactions between CO₂ and the absorbent resulting in thin reaction films, the 2D model approach is not needed for the overall absorption flux estimation, since the 1D approach provides almost the same results with a lower complexity. This is important since industrially relevant operation conditions for CO₂ capture with most of the previously presented absorbents seem to respond to these conditions.

Industrial desorption conditions tend to be severe because of their characteristic pressure fluctuations, high concentration of reactants and changes in temperature along the membrane contactor. However, both modeling and experimental studies conducted using rigorous conditions are not common. Hence, there is a need for more investigations to evaluate CO₂ regeneration process viability under both realistic and industrial conditions. In broad terms, as far as we are concerned the path for achieving HFMC technology is the control of membrane wetting, which has a huge influence on the overall process performance.

Studies in membrane wetting phenomena simulations have stated that they may be simulated by using a main equivalent membrane mass-transfer coefficient instead of local values. However, to design simple and accurate membrane wetting models to describe this phenomenon in the overall reactor model, more investigation of wetting mechanisms in real applications is required.

Table 4. Overview of model assumptions and methods reported in the literature for post-combustion CO₂ capture using HFMC technology.

General model assumptions, compilation from the literature^{158,159,186}	
<i>Hydrodynamics</i>	
1. Steady-state	
2. Laminar flow with fully developed velocity profiles in both the liquid and gas phase.	
3. Plug flow in both liquid and gas phases	
<i>Thermodynamics</i>	
4. Henry's Law to describe the gas-liquid equilibrium	
5. Only CO ₂ dissolved in the absorbent	
6. Ideal gas behavior	
7. Constant physical properties, such as viscosity and density.	
<i>Reaction and mass and heat transfer</i>	
8. Kinetics including reaction reversibility	
9. Constant mass transfer coefficient	
10. Negligible Condensation or evaporation of water	
11. Isothermal behavior	
12. Axial dispersion neglected in both gas- and liquid phase.	
<i>One-dimensional:</i>^{45,180}	
13. Axial convection in both phases	
14. The reaction, which use liquid mixing-cup concentrations at chemical equilibrium, is described by the enhancement factor.	
<i>Two-dimensional:</i>^{44,183,187}	
13. Axial convection and radial diffusion for the gas, liquid and membrane side is considered.	
14. Diffusion and reaction contribution to mass balances calculated using local concentrations.	

4.4. A note for unexplored issues

Various issues should be addressed to establish this technology as a commercial alternative on the industrial scale, regardless of all the hard effort put into the modeling and simulations of HFMC in the CO₂ desorption process. The wetting of the membrane, the presence of varied dissolved gases in the solvent acting as competitors of CO₂, the degradation of the membrane from solvent interaction limiting the long-term stability, the local variations of mass transfer mainly by pore distribution variances, and the solvent evaporation losses due to the high temperature and the non-isothermal conditions are among the main challenges for accurate mass transfer calculations.^{10,65} The performance under adiabatic conditions has recently been modeled through the use of a 1D approach by Scholes,³⁹ demonstrating that the heat associated with CO₂ desorption chemical reaction generated an important temperature difference in the HFMC. However, more research in the field of adiabatic conditions and temperature profile tracking should be conducted to better understand the temperature variations in the HFMC.

Additionally, dynamic modeling is another challenge that should be addressed due to the mass transfer coefficient variations over time reported in the literature. Most investigations using HFMC technology are conducted on limited small time scales, usually hours. Experiments over longer periods, either days or months, are few and demonstrate negative effects on mass transfer performance over time, which could be an issue in membrane contactor simulations.^{33,48,87,140}

Moreover, scale-up is a key point in HFMC simulations (Figure 9) from the carbon capture perspective due to the large volume of solvent to be regenerated, which requires a higher contact area by using more HFMC or increasing its size. However, pilot-plant

studies with HFMC for CO₂ solvent regeneration are scarce.^{28,188} Effective process performance prediction at the industrial scale is greatly unexplored by existing models due to the additional complexity of predicting fluid distribution patterns and such issues as fouling and wetting effects.

5. Conclusion and outlook

HFMC is a promising technology for applications in carbon capture systems, mainly due to advantages from an operational point of view, such as compactness and modularity, and specific absorbent properties, such as CO₂ selectivity. The majority of studies on HFMC are still centered on CO₂ absorption. Solvent regeneration process investigations in the field of carbon capture technology being scarce, although HFMC has demonstrated great faculty in solvent regeneration applications.

This manuscript reviewed the main results and key points of the literature on the CO₂ desorption process from liquid solvents by HFMCs and offered a perspective about the success of this research on PCC. CO₂ desorption was reviewed regarding various aspects, such as the physicochemical characteristics of solvents and their performance analysis in carbon capture; the membrane materials and their thermal and chemical stability; the contactor configurations employed; the main operating conditions influence; and the modeling studies conducted to describe and validate the process.

The conditions that make CO₂ desorption using HFMCs more attractive than conventional high-temperature desorption technology thus far indicate their technological viability. However, the majority of research on CO₂ desorption processes using HFMC are still operated at the laboratory scale. To achieve further commercial applications, some issues that need to be addressed include the following:

- 1187 i. The membrane wetting phenomena increase the mass transfer resistance and limit the
1188 long-term process stability.
- 1189 ii. The presence of impurities decreases the selectivity of the solvent for CO₂ and CO₂
1190 mass transfer, and the solvent issues, from an economic and environmental
1191 perspective.
- 1192 iii. The modeling and simulation issues include the need to develop a time-dependent
1193 mathematical model to simulate changes in some time-dependent parameters, such as
1194 the diameter of membrane pores, and thus the prediction of the system performance
1195 under different operational conditions within the CCUS strategies.

1196

1197 After the analysis of this paper, future lab-scale experiments and pilot plant
1198 demonstrations must be made to secure the technical and economic viability of the CO₂
1199 desorption process using HFMC. Three research activities are suggested for further
1200 research directions:

- 1201 i. Research on different membrane materials that fit in the framework of low-cost
1202 processes and long membrane lifetimes is needed to avoid frequent maintenance and
1203 replacement. Mechanically robust membranes with high thermal and chemical
1204 stability are required for the CO₂ desorption process. Recent lab-scale studies in
1205 membrane fabrication, has been focused in the integration of new materials (e.g.,
1206 enzymes and nanoparticles) either on or in membranes.
- 1207 ii. New absorbents with specific physico-chemical properties (e.g., low vapor pressure,
1208 low viscosity and fast reaction kinetics) for CO₂ desorption using HFMC should be
1209 explored. Environmental issues have to be prevented by “green” solvents.
1210 Furthermore, a membrane material and absorbent compatibility are important to
1211 secure long-term desorption performance.

1212 iii. Process performance should be investigated under industrial conditions (e.g. real
1213 composition of flue gas, temperature and pressure) to develop a technology with real
1214 application in the CO₂ capture industry.

1215 iv. To further evaluate the feasibility of membranes and membrane contactors,
1216 simulation models should be developed to predict the behavior and operations of the
1217 entire system.

1218

1219 In conclusion, the CO₂ desorption process by membrane contactor technology has many
1220 advantages. Although there has been no implementation in CO₂ capture plants at a large
1221 operation scale with this technology, it has the potential to be commercially viable if the
1222 issues can be solved through successes in the proposed research activities.

1223

1224

1225 NOMENCLATURE

1226 A = gas-liquid contact area, m²

1227 C = molar concentration, mol·m⁻³

1228 D = diffusion coefficient, m²·s⁻¹

1229 d = diameter, m

1230 d_h = hydraulic diameter, m

1231 E = enhancement factor

1232 Ea = activation energy, KJ·mol⁻¹

1233 F = molar flow rate, mol s⁻¹

1234 H = Henry's constant, mol m⁻³ Pa⁻¹

1235 k_o = first order reaction rate constant, s⁻¹

1236 K_{ol} = overall mass transfer coefficient, m·s⁻¹

1237	k = mass transfer coefficient, $\text{m}\cdot\text{s}^{-1}$
1238	L = length, m
1239	n = number of fibers
1240	N = desorption molar flux, $\text{mol}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$
1241	p = partial pressure, Pa
1242	Q = volumetric flow rate, $\text{m}^3\text{ s}^{-1}$
1243	r = radial coordinate
1244	r_{CO_2} = CO_2 reaction rate, $\text{mol}\cdot\text{m}^{-3}\cdot\text{s}^{-1}$
1245	T = temperature, K
1246	V_z = velocity, $\text{m}\cdot\text{s}^{-1}$
1247	V_{zm} = liquid mean velocity, $\text{m}\cdot\text{s}^{-1}$
1248	z = axial coordinate
1249	Re = Reynolds number
1250	Sc = Schmidt number
1251	Sh = Sherwood number
1252	Subscripts
1253	g = gas
1254	i = inner
1255	in = inlet
1256	l = liquid
1257	ln = logarithmic
1258	m = membrane
1259	o = outer
1260	out = outlet

1261 s = shell

1262 t = tube

1263 **Greek Letters**

1264 ε = porosity

1265 τ = tortuosity

1266 δ = membrane thickness, m

1267 $\emptyset$ = membrane pore wetting ratio

1268 η = dynamic viscosity, kPa s

1269

1270 **NOTES**

1271 The authors declare that they have no known competing financial interests or personal
1272 relationships that could have appeared to influence the work reported in this paper.

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1988 **Table Of Contents (TOC) graphic.**

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