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REVIEW ARTICLE

### CO<sub>2</sub> capture using membrane contactors: a systematic literature review

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**Abstract** With fossil fuel being the major source of energy, CO<sub>2</sub> emission levels need to be reduced to a minimal amount namely from anthropogenic sources. Energy consumption is expected to rise by 48% in the next 30 years, and global warming is becoming an alarming issue which needs to be addressed on a thorough technical basis. Nonetheless, exploring CO<sub>2</sub> capture using membrane contactor technology has shown great potential to be applied and utilised by industry to deal with post- and pre-combustion of CO<sub>2</sub>. A systematic review has been conducted to analyse and assess CO<sub>2</sub> removal using membrane contactors for capturing techniques in industrial processes. The review began with a total of 2,650 papers, which were obtained from three major databases, and then were excluded down to a final number of 525 papers following a defined set of criteria. The results showed that hollow fibre membranes have demonstrated popularity, as well as the use of amine solvents for CO<sub>2</sub> removal. This current systematic review in CO<sub>2</sub> removal and capture is an important milestone in the acknowledgment of up to date research with the potential to serve as a benchmark databank for

further research in similar areas of work. This study provides the first systematic enquiry in the evidence to research further sustainable methods to capture and separate CO<sub>2</sub>.

**Keywords** CO<sub>2</sub> capture, preferred reporting items for systematic reviews and meta-analyses, membrane contactor, absorbent

## 1 Introduction

The global energy consumption has doubled since the year 1970 predominated by fossil-based fuels such as oil, natural gas and coal [1]. These conventional resources have accounted for more than 80% of the global primary energy consumption in 2015 [1]. The total energy consumption is expected to increase by up to one third by 2060, and electricity consumption is projected to double as well [1]. Energy from renewable sources and nuclear power are growing at a rapid rate of 2.6% and 2.3% per year, respectively. Nevertheless, the reliance on fossil fuels will not decline as it is forecasted that fossil fuels will represent 78% of the world's energy use by 2040 [2]. Fossil fuelled power plants account for approximately 40% of the total CO<sub>2</sub> emissions, with coal fired power stations being the predominant contributor [3]. The combustion of these fossil fuels produces CO<sub>2</sub> at high rates which is recognised as the main greenhouse gas that contributes to climate change. The anthropogenic increase of atmospheric CO<sub>2</sub> concentration in the environment is projected to cause substantial fluctuations in the climate. It is estimated that approximately half of the existing CO<sub>2</sub> emissions are absorbed by the ocean and land ecosystems. However, sensitivity of climate and atmospheric CO<sub>2</sub> concentrations; create the feedback carbon loop [4]. On the other hand, CO<sub>2</sub> has a growing potential for by-product end-use in both the industrial and energy production sectors. The utilisation of CO<sub>2</sub> as a by-product would reap economic benefits as well as concurrently alleviating the concerns regarding global climate change [5].

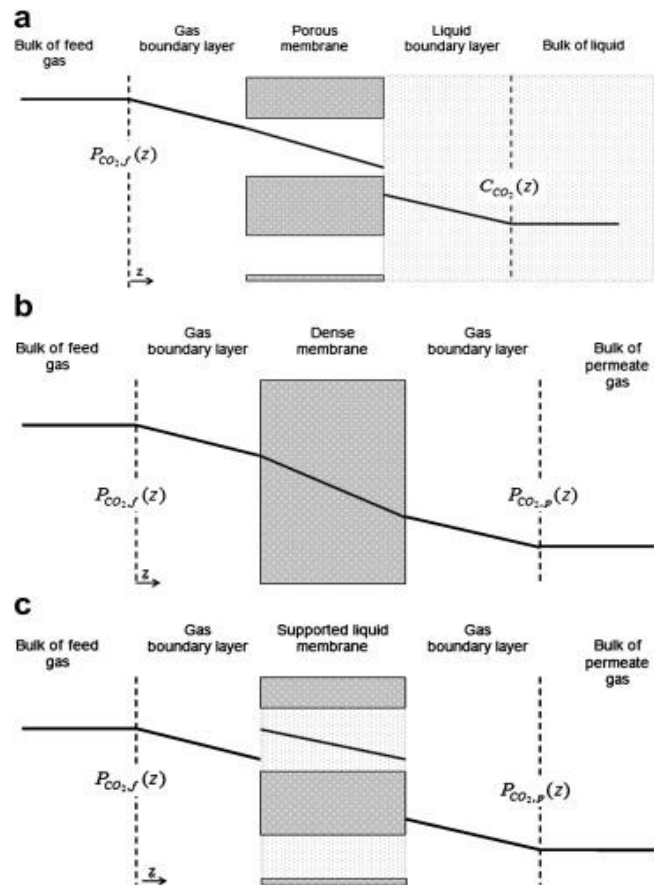
There are currently three main technologies which have been developed and implemented to capture CO<sub>2</sub> from fossil fuel combustion plants. These are pre-, post- and oxyfuel combustion [6]. The latter comprises of burning the fuel with almost pure O<sub>2</sub> as an alternative to air. In order to regulate the flame temperature, some part of the flue gas is recycled back into the furnace/boiler segment [6]. The key purpose of using this technology is to produce a flue gas with a high concentration of CO<sub>2</sub> and water vapour; and consequently, remove the CO<sub>2</sub> from the flue gas by dehydration coupled with a low temperature purification process [6]. The pre-combustion technique is a mature technology and has been used in the chemical industries for over 90 years. Here, the primary fuel is processed with steam and air/oxygen to produce synthesis gas (mixture consisting mainly of H<sub>2</sub> and CO). Excess H<sub>2</sub> and CO<sub>2</sub> are produced by reacting steam and CO in a shift reactor. The CO<sub>2</sub> is then removed, typically by a physical or chemical absorption process, subsequently in a H<sub>2</sub>-rich fuel which can be used in various applications, such as boilers, furnaces, gas turbines, engines and fuel cells [7,8]. Post-combustion is also often used to remove CO<sub>2</sub> which is produced from the flue gases generated by the combustion of the fuel in air. Normally a liquid solvent is used to obtain the small fraction of CO<sub>2</sub>

(3%–15% by volume) which is present in the flue gas consisting mainly of N<sub>2</sub>. Current post-combustion systems will often use an organic solvent, such as monoethanolamine (MEA), in a modern pulverised coal or natural gas combined cycle power plant [8,9].

Membrane contactor technology refers to tubular reactors that possess both chemical reaction and product separation units. This type of technology is widely applied in industries due to lower capital costs and facilitates the reaction in reaching equilibrium for the desired reaction. These reactors are most applied in dehydrogenation reactions. H<sub>2</sub> molecules can permeate through the membrane and increase conversion and making the process more economically efficient. Due to this application, membrane contactor technology has gained great popularity in recent years to be applied for CO<sub>2</sub> capture, which has been demonstrated by the studies discussed in this paper. There are two common types of membrane contactors, inert and catalytic. The former reacts as a barrier, whereas in the latter the membrane is coated or compiled from catalyst material so that can facilitate the reaction [10]. On the other hand, membrane adsorption refers to the phenomena of species separation within the membrane contactor due to the presence of functional groups of the membrane, or the sorbent utilised for the system. These are often applied in spent metal recovery and water treatment methods [11]. Membrane contactors promote contact between two phases through hydrophobic membranes and are most applied for industrial degassing of liquids and can also be categorised in hollow fibre membrane (HFM) contactors due to their arrangement and functionality.

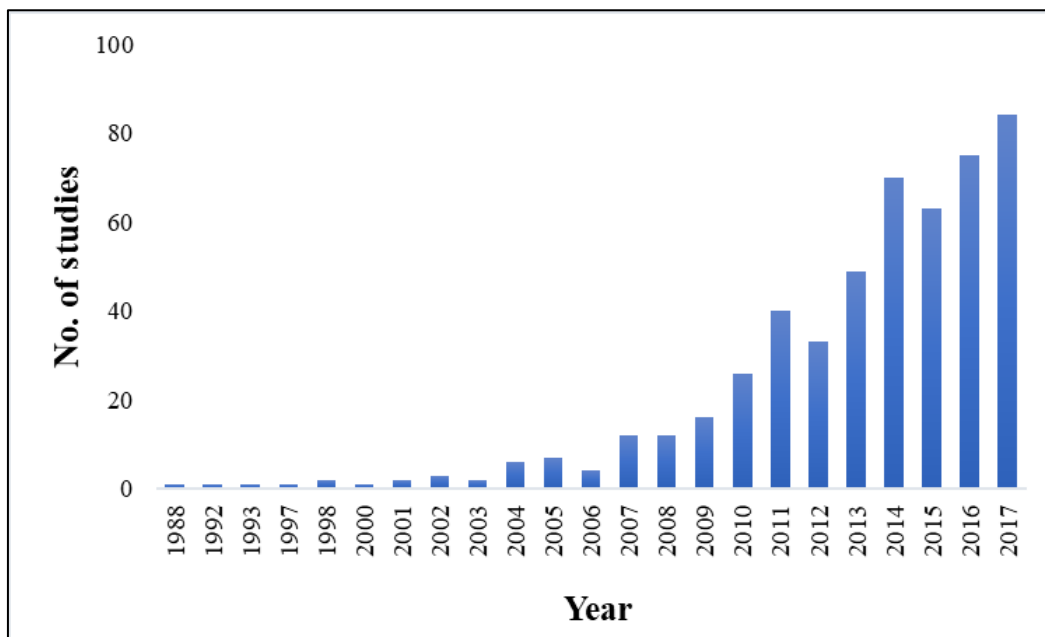
Membrane-based systems for the removal of CO<sub>2</sub> have demonstrated great superiority over conventional ones, and it has become imperative that they overcome existing issues of CO<sub>2</sub> separation and removal in pre-combustion and post-combustion systems [12]. One of the noticeable advantages of a membrane contactor system is that the reaction and separation units of the process are combined to give a single unit. As a result, the need for additional separation units is eliminated, thus making the process greener and environmentally sustainable [13]. There are three main systems which are often used as membrane processes for CO<sub>2</sub> removal (Fig. 1). These are (a) non-dispersive contact via a microporous membrane; (b) gas permeation (using dense membranes); (c) supported liquid membranes. Non-dispersive membranes are often applied in post-combustion capture systems [14]. This type of membrane configuration has a higher degree of operational flexibility because of the independent control of the gas and liquid flow rates, as well as an interfacial area which can be controlled and makes it easier to predict the performance of the membrane contactor. In addition, the modularity of the membrane contactors allows linear scale-up, and the system is compact and energy efficient. Issues regarding the flooding, channelling and entrainment are also avoided since the two phases flow on opposite sides. Furthermore, the mass transfer of CO<sub>2</sub> from the gas to liquid phase does not have a large impact on the gas flow due to the low concentration of CO<sub>2</sub> in the gas phase [14]. Other membrane separation systems such as distillation, extraction and electrophoresis can also be utilised. Stripping is another common separation process where single or multiple components are

absorbed from a liquid stream by a vapour stream for the separation of dilute volatile organic compounds from an aqueous solution [15,16].



**Fig. 1** Schematic of systems for CO<sub>2</sub> removal: (a) non-dispersive contact via a microporous membrane; (b) gas permeation; (c) supported liquid membranes [12]. Reprinted with permission of Elsevier from [12].

In this work, a systematic literature review was conducted to inform the reader as to the current knowledge about all the published studies performed in the area of CO<sub>2</sub> removal using various membrane-type contactors. Figure 2 depicts how the use of membrane contactors for CO<sub>2</sub> removal has increased throughout the years and will continue to do so. A detailed description of the methodology is provided to deliver an insight into how the review was performed, based on the guidelines for conducting systematic literature reviews [17]. Subsequently, the methodology section is followed by the results and discussion to assess and analyse the findings of the study. To our knowledge this is the first systematic review in the topic of CO<sub>2</sub> removal using membrane-type contactors.



**Fig. 2** Popularity of membrane contactors for CO<sub>2</sub> removal throughout the years

## 2 Methods

A systematic enquiry was applied by using a defined search strategy and run on the 8th January 2018 across three databases: web of science (WoS), google scholar and PubMed. This was done to gather peer-review articles, conference proceedings, editorial letters, books and grey literature with no language, time or geographical restrictions in our search. We imported all references to an EndNote library, removed duplicates and screened for relevance based on title and abstract.

### 2.1 Search strategy

A search strategy was devised using only “CO<sub>2</sub>” and “membrane” in the title. Although a third keyword could have been added to refer to the process of isolating the CO<sub>2</sub>, this was intentionally left out. This was done as many studies use for example a specific absorbent membrane type of membrane and hence, do not use words like capture, removal, separation or other synonyms as such to describe this. Similarly, instead of trying to gather a list of potential solvents used to absorb CO<sub>2</sub>, we kept a few basic search terms and the search strategy simple. We then used the information from the collected studies to fill the knowledge gap around the type of membranes used so far by researchers in the area of CO<sub>2</sub> removal. Compared to systematic reviews performed in the field of healthcare where the titles can be longer and more descriptive, most engineering articles contain information in the title about the equipment and material(s) used.

### 2.2 Inclusion/exclusion criteria

Only studies that concerned the absorption of CO<sub>2</sub> in membrane contactor systems were included. For example, studies focusing on membrane systems for medical and nature applications were excluded.

We adopted the widely recognised preferred reporting items for systematic reviews and meta-analyses (PRISMA) flowchart to demonstrate the steps in the undertaken methodology and results. Figure 3 details the PRISMA flowchart for this work.

### 2.3 Outcome analysis

Prior to conducting the review, we have considered that the following items would be important variables in synthesising the research in this area: membrane material, contactor type, flow configuration, absorbent (molarity), wetting, average flux ( $\text{mol}\cdot\text{min}^{-1}\cdot\text{cm}^{-2}$ ), gas flowrate ( $\text{mol}\cdot\text{min}^{-1}$ ), liquid flowrate ( $\text{mL}\cdot\text{min}^{-1}$ ),  $\text{CO}_2$  in inlet feed (%),  $\text{CO}_2$  removal (%). The above information was extracted for each paper. Unless the average flux was provided by the authors, we have manually calculated it using the formula below:

$$\bar{f} = \frac{\text{inlet molecular flow } \left(\frac{\text{mol}}{\text{time}}\right) * \text{conversion } (\%)}{\text{membrane surface area}}. \quad (1)$$

where the conversion (%) refers to the amount of  $\text{CO}_2$  removed.

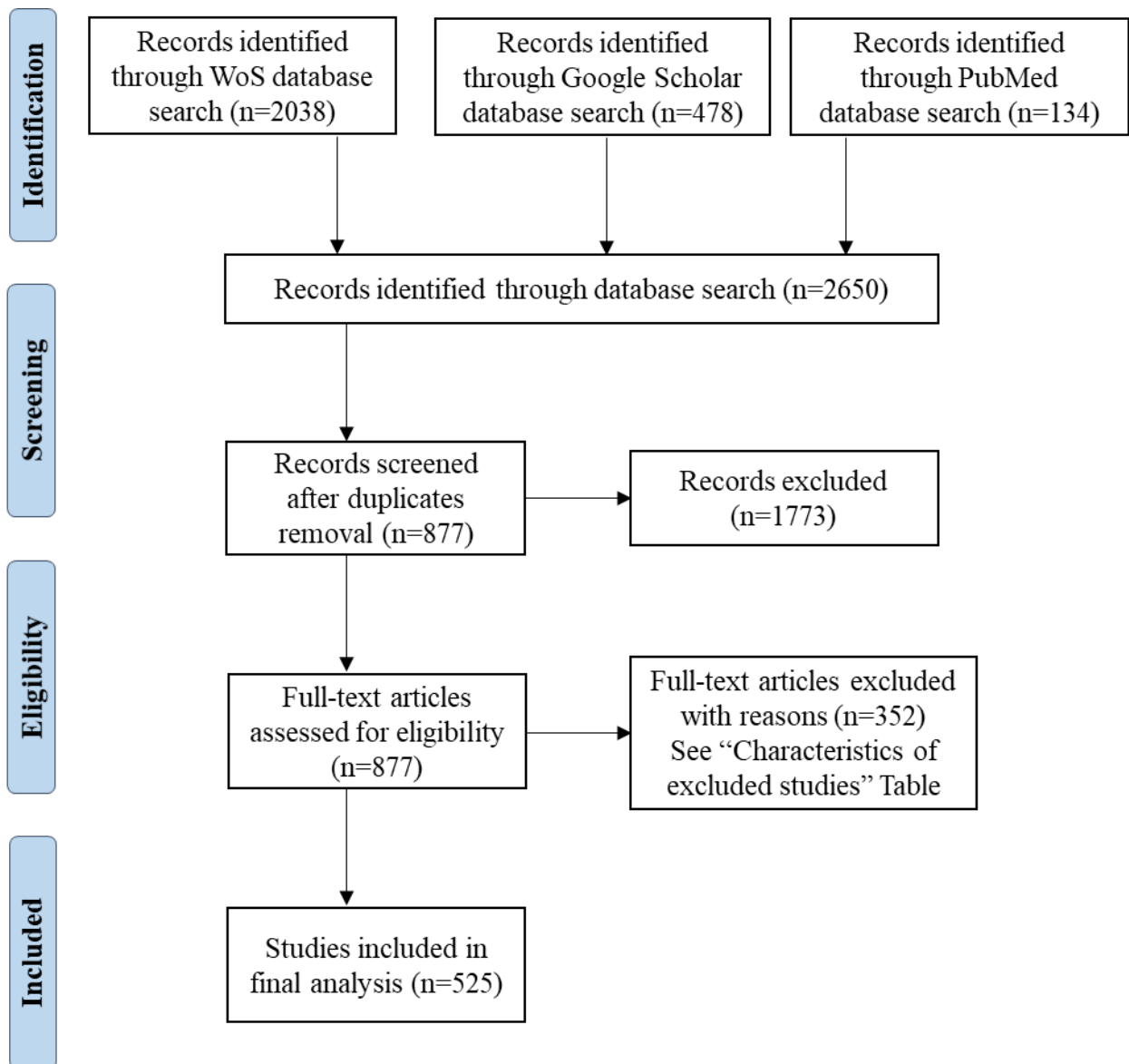


Fig. 3 The PRISMA flowchart of the methodology

### 3 Results

We have identified 2,650 studies through electronic searches of WoS (n = 2,038), Google Scholar (n = 478) and PubMed (n = 134). There were 341 removal of duplicated records. We have excluded 1,773 through scanning titles and reading abstracts and retrieved a total of 877 full-text articles for further assessment. The 352 upon full-text did not meet the study criteria, and hence were subsequently removed. We included 525 studies in the final review.

#### 3.1 Excluded studies

The excluded studies encompassed those that did not meet the criteria of using a membrane structured reactor. Of the total 877 studies, 122 of the papers had no text available or could not obtain access, leaving 755 studies. To keep the focus of this paper on published and established CO<sub>2</sub> membrane applications, non-peer-reviewed sources such as masters or doctoral degree theses were later excluded (17). Also, since patents do not provide scientific results, were also excluded (16). Lastly, as the aim of this paper was to compile data on CO<sub>2</sub> membranes, biological membranes, such as plant or animal-based membranes for CO<sub>2</sub> transfer were neglected as they demonstrated applications in biological systems (17). CO<sub>2</sub> capture and separation has been of interest to many research possibilities, especially with novel membrane technologies. A further 181 studies discussed the potential application of new membrane into CO<sub>2</sub> capture by either theoretical modelling or conducting preliminary experiments to study the permeances of the membrane. However, they did not provide enough parameters to be included in the review as these new innovations need further research before being presented for industrial capture applications.

#### 3.2 Included studies

We have included 525 studies in this systematic review. There were 77 studies on computational modelling, where different programmes such as Aspen Plus, COMSOL, Aspen HYSYS and MATLAB were utilised to stimulate a preliminary application of CO<sub>2</sub> membrane capture. A total of 21 review papers that discussed existing membrane capture technologies, showcased a range of membrane from zeolites to polymeric to ionic liquid membranes (ILMs). Other studies varied between demonstrating an application of CO<sub>2</sub> membrane capture to small scale lab experiments to determine the potential of the proposed method.

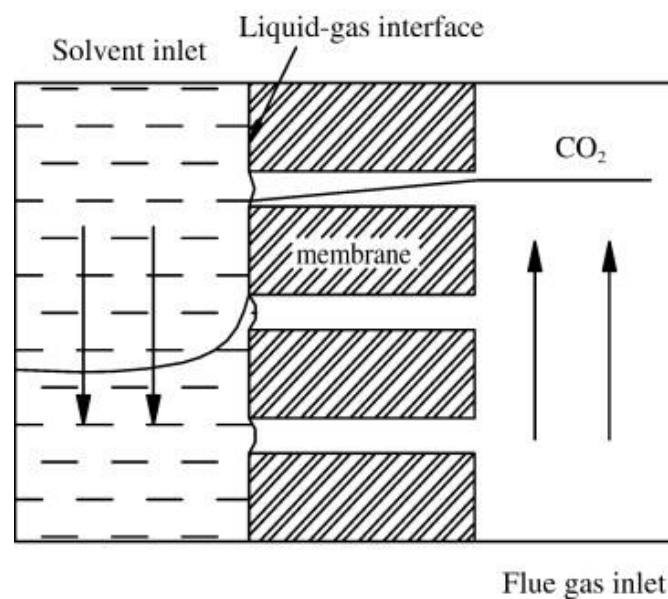
#### 3.2 Summary of main results

##### 3.2.1 Membrane material

The porosity and pore size of the membrane are the most significant factors to take into consideration since the contact between the gas and liquid phases occur solely on the pores of the membrane. It is imperative to have a good chemical suitability between the membrane contactor and the solvent as the absorption liquid determines the selectivity of the separation [12]. Figure 4 demonstrates the principle



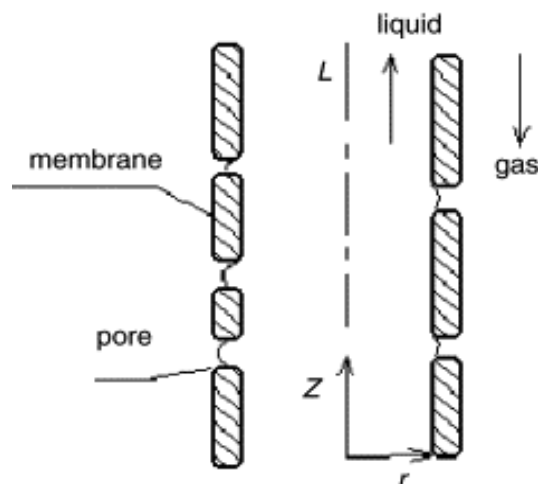
of CO<sub>2</sub> separation using a membrane. Here, the membrane determines the permeability and selectivity of the process and so the use of liquids is not required [18]. Gas permeation membrane technology is predominantly used in pre-combustion systems. However, such membranes are being developed for post-combustion systems as well. The use of supported liquid membranes for CO<sub>2</sub> removal have gained increasing attention due to ionic liquids being used as solvents. In this membrane configuration, the pores of the membrane are saturated with a liquid, or the liquid is supported on the surface of the membrane. Ionic liquids are mostly attractive in a membrane separation device due to their very low volatility which minimises solvent losses from the membrane [19].



**Fig. 4** Principle of CO<sub>2</sub> absorption using membranes [20]. Reprinted with permission of Elsevier from [20].

The most popular type of membrane (28%) was found to be the hollow fibre ( $n = 149$ ) [20–168]. Figure 5 shows a schematic of how absorption occurs in a HFM. The second most common membrane (15%) was observed to be mixed matrix membranes (MMMs) ( $n = 78$ ) [169–246]. The average flux for these ranges from  $3.95 \times 10^{-3}$  to  $1.8 \times 10^{-12} \text{ mol}\cdot\text{cm}^{-2}\cdot\text{min}^{-1}$ . HFM work to imitate the function of pulmonary capillary bed packing function where the oxygenation can be optimised by manipulating the gas delivered to the oxygenator. HFM combines chemical absorption with membrane separating technology, allowing for higher selectivity and smaller dimensions (compared to typical separation columns) to be achieved. The mass transfer mechanism resembles that of the shell-tube heat exchanger, thus causing the concentration gradient to be the driving force. Since HMFs are modular systems, interfacial area is significant increased and scale-up operations can be relatively simple when compared to conventional separation systems [247]. Due to better performance, HFMs were one of the first membrane systems to be investigated for gas separation systems, which is supported by the high number of studies testing HFM with relatively higher flux values. However, the flux values sit within

a huge range, with the majority being in the lower end. This could be the result of membrane wetting, as membranes can become partially wet by the absorption liquid and significantly reduce performance.



**Fig. 5** Schematic showing how absorption occurs in a HFM [25]. Reprinted with permission of Elsevier from [25].

MMMs combine the inorganic fillers with polymeric properties thus giving rise to a huge potential for gas separation industry. These have exhibited versatile performance, with different kind of solvents, ranging from water, ethanol, hydrochloric acid to amine solvents. Since this utilises polymeric properties, the majority of MMMs (3%) are made from different types of polymer materials but face commercialisation issues on a large scale. Metal-organic frameworks (MOFs,  $n = 14$ , 3%) [211,248–260] are one of the recent inventive solutions for CO<sub>2</sub> separation, with 14 studies exploring the potential. The physical characteristics such as high porosity and surface area along with adjustable pore size and versatile structural arrangement makes it an ideal membrane. The studies above have shown better results than conventional zeolites and polymeric membranes, in conjunction with different kinds of solvents (from alcohol to amine solvents). Poor mechanical properties along with thermal and chemical stability are amongst one of the major limitations of MOFs. Intercrystalline voids or any internal damage within the layers significantly reduces the membrane selectivity. As MOFs are a relatively new membrane advancement than zeolites or polymers and coupled with their limitation makes them a less popular choice amongst other types.

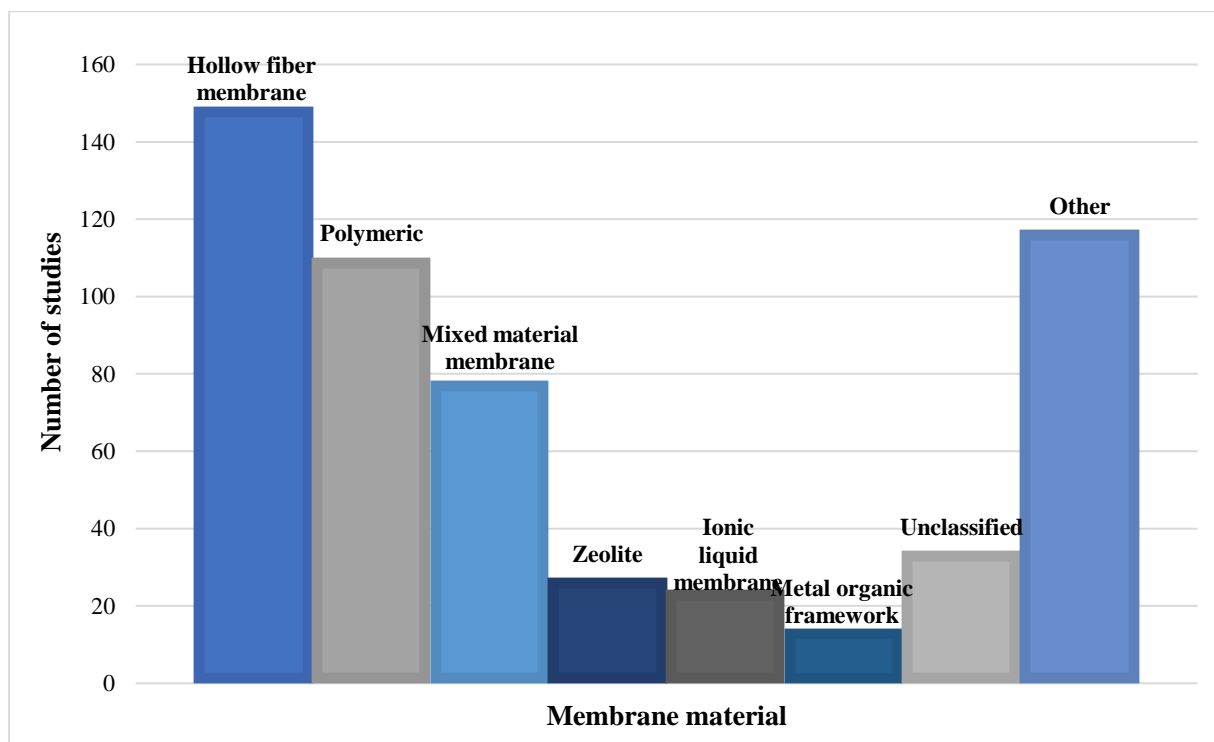
A total of 110 studies utilised polymeric membranes (21%) [46,48,52,62,77,102,117,134,152,227,229,250,261–358] or zeolites ( $n = 27$ , 5%) [359–385] and have been a major part of CO<sub>2</sub> separation, through membrane technology. These membranes have been applied and used throughout industry for the last three decades and have shown great potential. The studies have shown enhanced performance of these membranes in hybrid and composite forms. The average flux ranged from  $1.3 \times 10^{-1}$  to  $3.33 \times 10^{-9} \text{ mol}\cdot\text{cm}^{-2}\cdot\text{min}^{-1}$ , thus displaying the great potential. Furthermore, these can be applied with organic and inorganic solvents, making them more versatile,

with water solvent displaying the best results. However, polymeric membranes are subject to mechanical and structural change over time, as well as low surface area per unit of volume, and results in low selectivity and permeability values on a large scale. This is supported by the lower range of flux values, where the majority lie under  $1 \text{ mol}\cdot\text{cm}^{-2}\cdot\text{min}^{-1}$ . The main current challenge is the plasticisation of suppression of polymer membranes along with the economic implication involved in increasing the membrane area to obtain higher flux values. To overcome this, many studies have experimented on hybrid polymer membranes by integrating them into other membrane structures. Peters et al. [302] studied acid gas sweetening using amine absorption and a two-polymer membrane structure and achieved a flux of  $2.3 \times 10^{-3} \text{ mol}\cdot\text{cm}^{-2}\cdot\text{min}^{-1}$ . The value of flux refers to the performance of membrane and the effect it has on the molar flux of the membrane, considering the ratio of its permeability against the thickness. The membrane technology was reported to achieve a content of 2%  $\text{CO}_2$  in the product gas as a final target, with a two-stage configuration for a purity of 90%  $\text{CO}_2$  within the permeate stream of the second membrane stage. The flux of the membrane was also considered in the simulation environment exercises conducted. Though a good membrane performance was exhibited, the group also reported further work to evaluate the capital costs of the separation system and thus indicating the persistent challenge between price and performance. Zeolites have been extensively used as catalyst throughout the industry and have shown potential in membrane technology in the last twenty years but have not been as successful as novel MMMs or HFMs. These have exhibited average flux in the range of  $1 \times 10^{-2}$  to  $3.02 \times 10^{-6} \text{ mol}\cdot\text{cm}^{-2}\cdot\text{min}^{-1}$  and have displayed poor mass transfer within the membrane. Zeolites are desired for numerous reasons, the main one being the durability and economic cost, as well as their ability to work with different kinds of solvent [386]. However, further research is required to study and establish better reaction conditions to achieve better mass transfer within the system.

ILMs ( $n = 24, 4\%$ ) [112,360,387–408] are one of the recent advancements in membrane technology. They have a liquid component in the system which allows for the system to have a higher diffusivity, thus resulting better permeability as well allowing the system to be modified by adding on complexes to enhance the  $\text{CO}_2$  solubility. Nevertheless, ILMs still need to be further developed to withstand high temperatures and how their hydrodynamics work. This can explain why there only 23 (4%) studies on ILMs, with an average flux in the range of  $1 \times 10^{-3}$ – $5.03 \times 10^{-9} \text{ mol}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ . Due to the great potential of these ILMs, some studies have experimented on different ways forming ILMs. Karousos et al. [390] developed ILMs through physical inhibition of ionic liquid in ceramic tube, consisting of mesoporous separation layer. The group tested different types of ionic liquids,  $8.1 \times 10^{-7} \text{ mol}\cdot\text{cm}^{-2}\cdot\text{s}^{-1}$  was only exhibited by one type ionic liquid as well as being limited to high temperatures and  $\text{CO}_2$  mole fractions. ILMs are yet to be successfully applied in industry for long-term systems, most studies are focused on investigating support membranes, viscosity of ionic liquids and preparation methods. The

upcoming and major challenges for ILMs can be summarised in the following points, thus reflecting the piqued interest but low popularity: (1) Adsorption capacity of ionic liquids in membrane separations; (2) ionic liquids, though show great performance promise, are toxic; (3) overcoming selectivity, stability and recycling issues; (4) finding an economically feasible method of ILM development and membrane set-up.

The flux range and the number of studies in this paper are higher than zeolite and MOFs, showing great promise and potential for CO<sub>2</sub> capture. There were 22% (n = 117) of studies [89,409–524] which tested different materials that do not fit the mentioned categories due to the vast variation in materials, such as ceramics, caesium incorporated, bio-catalytic membrane materials, and capillary membrane as well hybrid membrane forms of polymer and MMMs for example. The remaining 34 studies (6%) [525–546] have not conveyed any information relating to the membrane material. Figure 6 displays the percentage of studies utilising the various mentioned membranes.



**Fig. 6** Percentile of types of membranes utilised for CO<sub>2</sub> capture determined from this work

### 3.2.3 Contactor type

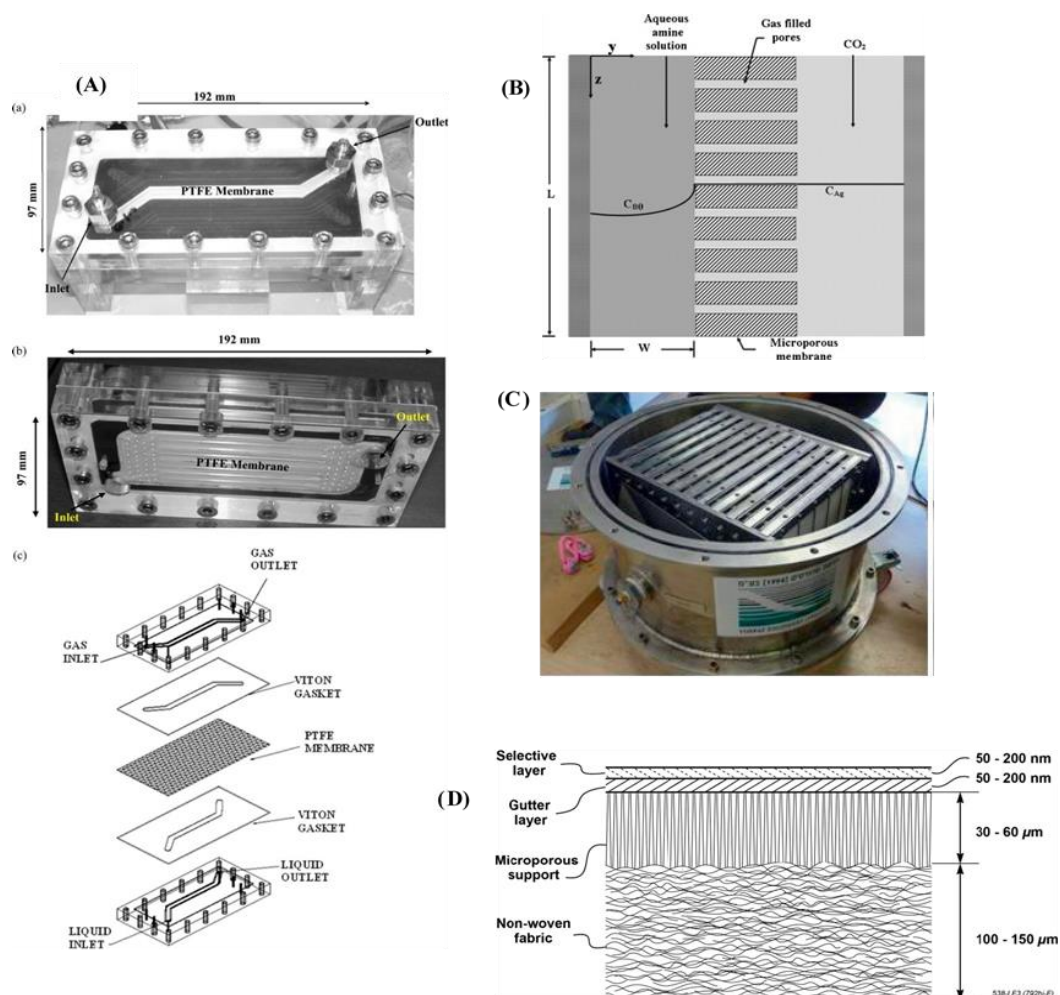
There were three types of membrane contactors observed across the 525 studies: flat sheet (FS) (n = 79, 15%), [90,93,98,117,127,137,180,181,187,193,199,212,218,222,230,238–240,243–245,248,249,256,267,268,278,282,284,287,289,302,305,312–315,317,321,323,324,326,329,333,336,338,343–345,347,352,356,358,360,391,398,399,404,408,422,426,438,449,450,453,465,467,476,481,500,503,508,515,521,522,524,531,538,544], facilitated transport (FT) (n = 36, 6%)

[21,32,50,86,135,136,188,272,276,283,288,325,335,387–390,392–397,399–403,421,423,431,468,482,510,517,518], and HFM (n = 176, 32%) [20,22–49,51–63,65–84,87–89,91,94–97,99–111,113–116,118–126,128–134,138–148,150–168,174,183–185,192,195–197,204,261,262,276,284,288,291–293,307,311,322,331,364,387,390,392,412,426,429,430,434,435,439,489,492,507,516,519,527,528,530,535,540,541]. There were 175 (33%) studies that had other membrane contactor types (for example, water gas shift/membrane hybrid, and poly(ethylene oxide) based block copolymers contactors), which do not fit into the above mentioned categories [64,85,170–173,175,176,178,179,182,189,191,194,200–203,206–211,231–233,241,242,246,247,253,260,263–266,271,273,274,277,279,281,283,285,286,290,295–301,303,304,306,308–310,316,318–320,327,328,332,334,337,339,340,342,346,348,350,351,353–355,357,359,361,363,366,367,370–372,374–378,380,381,385,386,405–407,410,411,413–418,420,424–428,432,433,436,440,442–448,451,454–458,460–462,464,466,469,470,472–475,477–480,483–488,490,491,493,494,497–499,501,502,505,509,512–514,520,523,532,533,536,537,539,542,545]. The remainder 72 studies (14%) do not contain any information on the membrane contactor type [92,112,149,169,177,186,190,198,205,213–217,219–221,223–229,234–237,250–252,254,255,257–259,269,270,275,280,294,330,341,349,362,365,368,369,373,379,382–384,409,419,437,441,452,459,463,471,495,496,504,506,511,525,526,529,534,543,546].

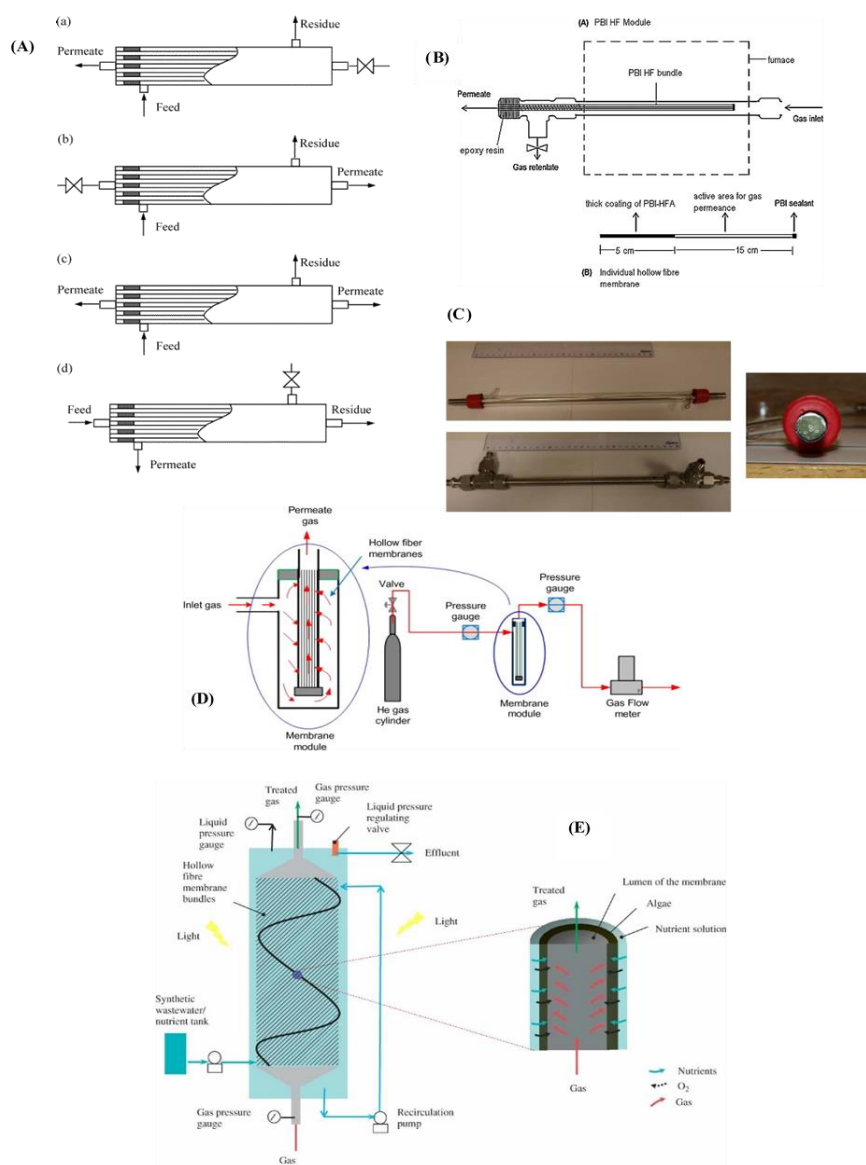
FT is also known in the application of supported liquid membranes, referring to ILMs in this review. The FT mechanism refers to a form of passive transport where external species are used to aid the transport. The molecules move across the membrane with the help of membrane proteins and possesses the ability to transport larger molecules. This is usually affected by the temperature, which is supported by the fact that ILMs have better stability, and thus can withstand higher temperatures, resulting in higher flux values. Concentration is another influential factor that affects the transport mechanism [547].

FS membrane configurations (Fig. 7) are most known for their application in bioreactors [548]. Hollow fibre reactor configurations (Fig. 8) provide higher fluxes, this is supported by review data presented in the supplementary material. HFM provide better gas permeability across the membrane, evidently supported by the number of studies utilising HFM. They are also easy to maintain with minimal pre-screening and requiring mild cleaning to maintain the fibre exterior. FS membrane configurations do not allow for the membrane to back pulse, and so the risk of membrane fouling increases because the impurities cannot be frequently removed [548]. However, FS is a common choice from a maintenance perspective because of the application of gravity flow, saving the systems from using effluent pumps thus saving cost and energy in operation [549]. They have a longer lifetime but are not commonly manufactured across industry, making the initial investment costly. The arrangement of FT membranes (Fig. 9) enables high selectivity and high flux as well as better stability.

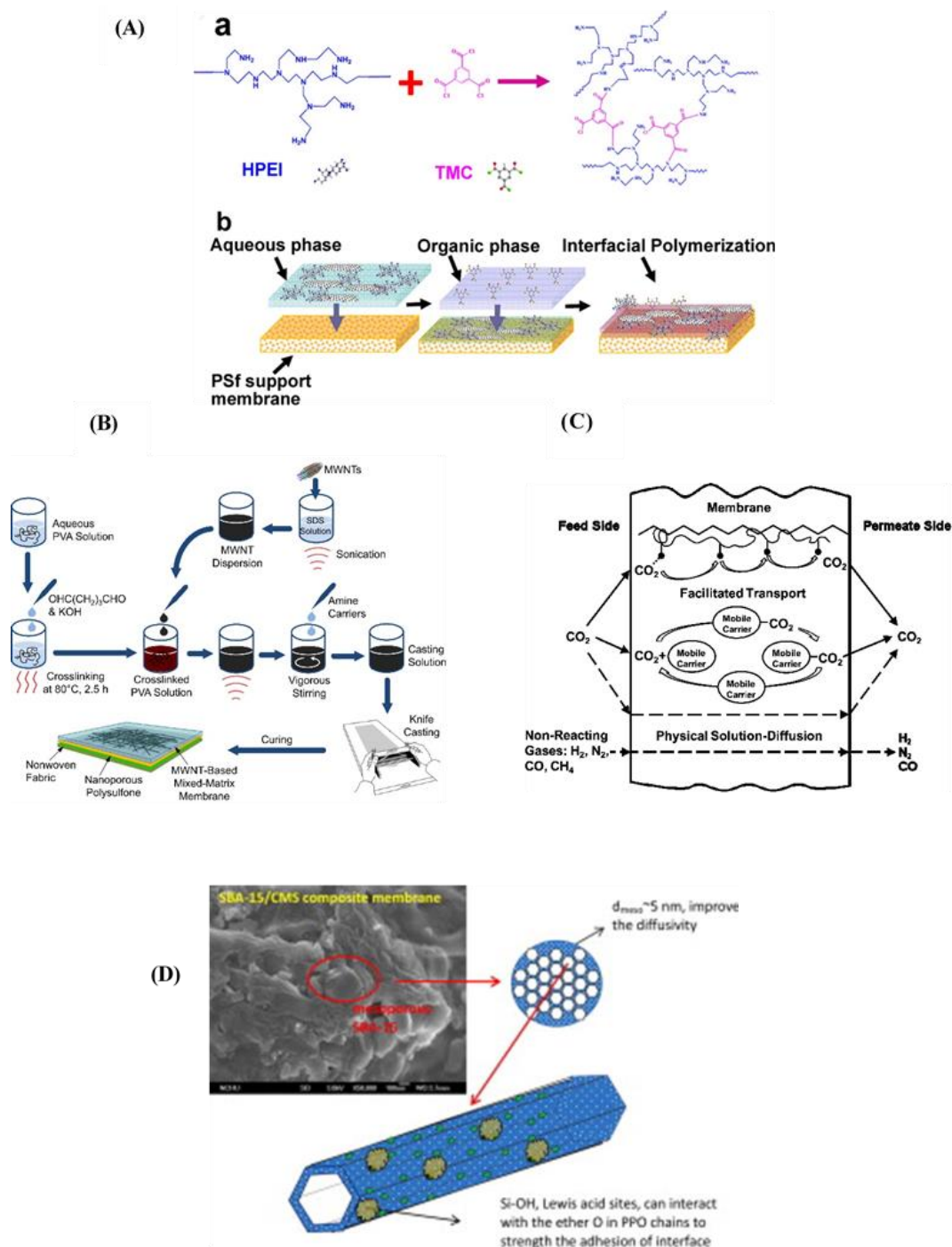
Fixed carrier membranes where the ionic liquids were adsorbed on the support exhibited better stability in terms of higher reaction pressures and temperatures, when compared to the flat liquid sheet membrane configuration. Hence, they have higher potential for recyclability. The reason being simply that adsorbed ionic liquids are stronger anchor on the support than the freely standing ionic liquids. Table S1 (cf. Electronic Supplementary Material) shows that higher flux values are exhibited by HFM, followed by FT and then FS.



**Fig. 7** (A) Flat membrane microstructured contactors: (a) picture of assembled device of the Polytetrafluoroethylene (PTFE) single channel contactor; (b) picture of the assembled device of the 8-channel PTFE contactor; (c) exploded schematic view of the single channel contactor [321]. Reprinted with permission of ACS Publications from [321]; (B) schematic representation of absorption in hydrophobic FS membrane [422]. Reprinted with permission of Elsevier from [422]; (C) FS pilot scale membrane module [282]. Reprinted with permission of Elsevier from [282]; (D) schematic illustration of thin-film composite polaris membranes [323]. Reprinted with permission of Elsevier from [323].



**Fig. 8** (A) Configurations of membrane modules: (a) shell side feed, counter-current flow; (b) shell side feed, co-current flow; (c) shell side feed, counter-/co-current flow (permeate withdrawal from both ends of the fibre bores); (d) bore side feed, counter-current flow [30]. Reprinted with permission of ACS Publications from [30]; (B) schematic representation of (a) polybenzimidazole (PBI) HFM module used for permeation at high temperature, (b) Individual HFM partially coated with polybenzimidazole-4,4'-(hexafluoroisopropylidene)bis(benzoic acid) (PBI-HFA) and lumen plugged with PBI sealant [63]. Reprinted with permission of Elsevier from [63]; (C) hollow fibre modules for gas permeation experiments [56]. Reprinted with permission of Elsevier from [56]; (D) schematic diagram of hollow fibre gas permeation test apparatus [61]. Reprinted with permission of Elsevier from [61]; (E) schematic of the bench-scale HFM photo-bioreactor (HFMPB) system [43]. Reprinted with permission of Wiley from [43]



**Fig. 9** (A) Schematic diagram of (a) the reaction between hyperbranched polyethylenimine (HPEI) and trimesoyl chloride (TMC), and (b) the fabrication process for the HPEI/graphene oxide-TMC composite membrane [481]. Reprinted with permission of ACS Publications from [481]; (B) schematic of the preparation procedure for the MMM [449]. Reprinted with permission of Elsevier from [449]; (C) schematic of gas permeation through a FT [98]. Reprinted with permission of ACS Publications from [98]; (D) schematic of mesoporous silica sieve SBA-15/carbon molecular sieve composite membrane [283]. Reprinted with permission of Elsevier from [283].



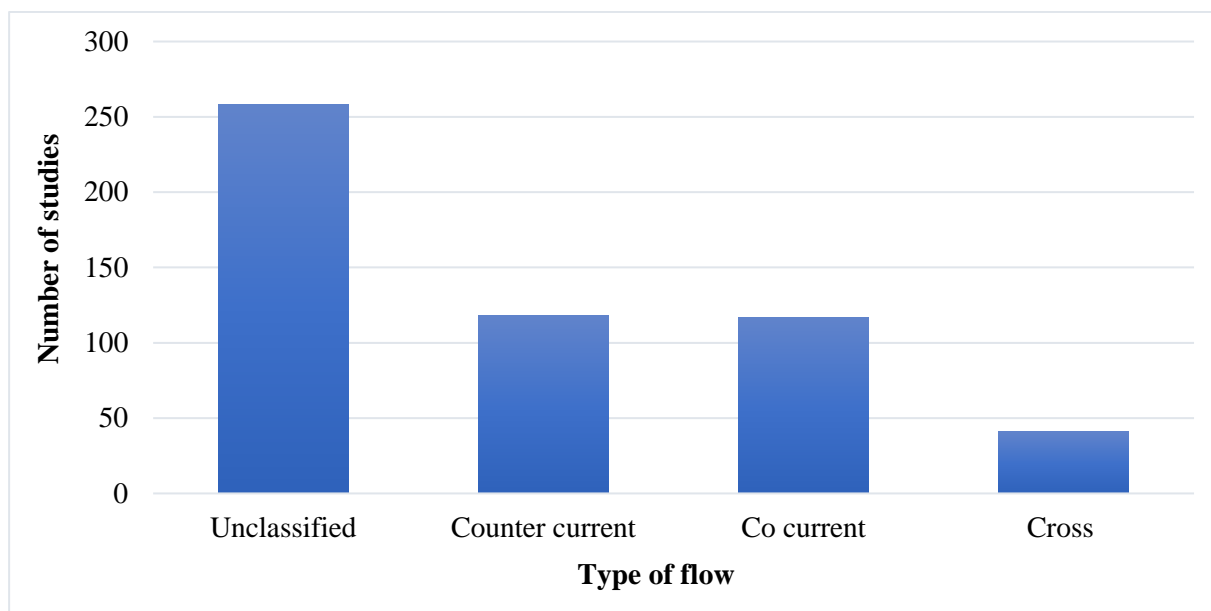
### 3.2.4 Flow configuration

Figure 9 shows different types of flow configuration that were used across the studies: co-current (n = 117, 22%) [21–24,26,32,34,36–38,41,46,52,53,58,61,63–65,67,73,74,76,77,81,83,86,97,109–111,113,123,132,165–167,171,177–179,182,195,217,226,236,238,241,242,244,245,253,261,262,275,282,284,288,290,292,300,311,312,314,319–322,327,336,338,344,346,347,356,357,359,365–367,376,387,388,390,392,394,395,397,400,403,407,408,410,412,419,422,424,434,446,447,462,468,472,487,488,490,507,510,515,522,528,531,533,535,536,538,546], counter current (n = 118, 22%) [20–22,24,25,27,30,33,40,42,44,45,47–49,54,56–58,60,65,66,69,71,72,74,75,80,82,85,87–89,91,98–102,104,105,108,113,115,117,119,120,124,125,128,134,138,140–144,147,148,151,153,154,156,157,160,161,163,168,186,187,196,197,249,263,278,279,300,302,326,339,344,345,350,358,382,383,396,397,411,413,418,421,428–430,433,439,453,458,461,465–467,470,477,489,505,509,514,516,520,521,526–528,530,534,544], and cross flow (n = 41, 8%) [51,78,92,94–96,103,114,130,131,139,146,150,152,155,194,196,225,287,293,308,322,323,333,344,351,353,406,413,417,432,451,454,456,486,491,494,495,502,523,526]. The rest 258 studies (49%) contained no information regarding the flow configuration used [28,31,35,43,50,55,59,62,84,90,93,106,107,112,116,118,121,122,126,127,129,133,135–137,145,149,158,159,162,164,169,170,172–176,180,181,183–185,188–193,198–216,218–224,227–235,237,239,240,243,246,248,250–252,254–260,264–274,276,277,280,281,283,285,286,289,291,294–298,299,301,303–307,309,310,313,315–318,324,325,328–332,334,335,337,340–343,348,349,352,354,355,360–364,368–375,377–381,384,385,389,391,393,398,399,401,402,404,409,414,415,420,423,425–427,431,435–438,440–445,448–450,452,455,457,459,460,463,464,469,471,473–476,478–485,492,493,496–501,503,504,506,508,511–513,517–519,524,525,529,532,537,539–543,545].

In many cases both co- and counter- current flow were studied to see the effect on mass transfer and membrane performance. Co-current and counter flow configurations are most utilised across various disciplines, due to the developed understanding of mass transfer phenomena. However, it is not possible to say whether one is better than the other. It can be summarised that these flow configurations provide better performance (with their respective membrane application) when compared to other types of flows. Studies that solely used co-current configuration displayed an average flux in the range of  $1 \times 10^{-1}$ – $1.72 \times 10^{-17}$  mol·cm<sup>-2</sup>·min<sup>-1</sup> with the higher values corresponding to ILMs suggesting that co-current flow is better suited for ILMs.

The study of counter current flow was mostly exhibited in HFMs and displayed larger mass transfer

range,  $5.5 \times 10^{-1}$ – $8.7 \times 10^{-11}$  mol·cm<sup>-2</sup>·min<sup>-1</sup>. This indicated the better performance to be due to better concentration gradients being established at the gas liquid interfaces. However, the lower flux value suggest that this is flow is subjective to situation and experimental conditions. A flux of  $8.7 \times 10^{-11}$  mol·cm<sup>-2</sup>·min<sup>-1</sup> was obtained at a lower gas flow rate and 0% N-Methyl-2-Pyrrolidone solvent. Counter current configuration seems to display better mass transfer rate with amine and salt solvent, with high and low inlet feed conditions. Some studies experiment with cross-flow, where the feed travels tangentially across the membrane. Theoretically, this provides better contact as there is more random contact between the membrane and the gas, but the results do not provide promising mass transfer. The flux range was between  $2.94 \times 10^{-4}$  and  $1.2 \times 10^{-12}$  mol·cm<sup>-2</sup>·min<sup>-1</sup>. However, these were tested with butanol and amine solvents, further testing with different types of solvents could potentially provide a different result.



**Fig. 10** Types of flows (by count) determined from the included studies

### 3.2.5 Solvent (with molarity)

The solvent choice is an important factor in membrane separation, as it directly impacts the economic aspect of the process as well as aid to finding the right low energy solution for CO<sub>2</sub> processing. Three distinctive types of solvents were identified from the studies; amine solvents (n = 104, 20%) [20,21,25,32–34,36,37,39,40,42,49,51–53,57,59,60,67,68,70,72,79,85,87,91–96,98,100,103,105,107,109,114,115,119,120,122,124,125,129–131,138,140,141,143,144,146,151–155,157,166,168,171,206,213,217,233,241,263,271,276,287,288,296,300,302,309,310,317,324,327,331,339,357,396,402,404,408,411,412,422,425,434,439,453,458,469,476,491,498,503,505,521,529,538]. CO<sub>2</sub> capture using amine solvents has been practiced since the 1950s and therefore is a well understood and developed process. This is supported by the number of studies testing CO<sub>2</sub> separation

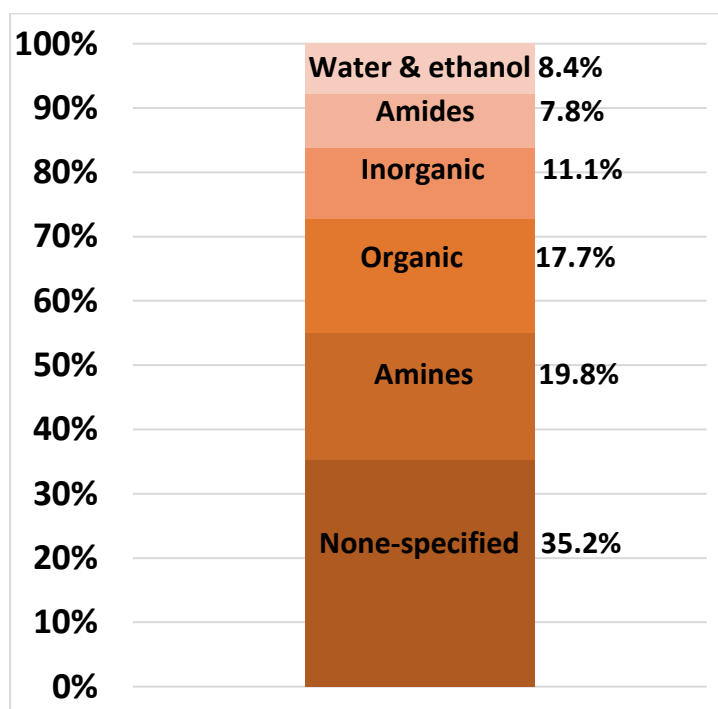
using amine solvents. Typical membrane separation operates at 60 °C, which makes it extremely desirable to be an energy efficient option. The flux for amine solvents range from  $1.1 \times 10^{-1}$ – $1.75 \times 10^{-16} \text{ mol}\cdot\text{cm}^{-2}\cdot\text{min}^{-1}$ . For  $1.1 \times 10^{-1} \text{ mol}\cdot\text{cm}^{-2}\cdot\text{min}^{-1}$  an MEA solvent was used in a polymer matrix. For  $1.75 \times 10^{-16} \text{ mol}\cdot\text{cm}^{-2}\cdot\text{min}^{-1}$  the group used same MEA solvent in FT membrane contactor with a feed inlet of 41%. Large variation in flux ranges support the low absorption capacity of amine solvents as well as high reactivity, stability and thermal degradation issues.

There were 58 studies (11%) which utilised various kinds of inorganic solvents such as metal nitrates and silane [22,47,61,71,73,89,90,106,123,149,150,159,162,170,172,173,192,195,200,207,208,229,237, 239,240,245,256,269,311,315,320,321,325,367,370,372,378,416,419,420,424,427,444,448,450,452,456,460,473,478,492,501,502,506,511,512,515,523], and amides (n = 44, 8%) [24,26,63,82,99,101,133,139,176,184,187,197,198,209,216,227,231,252,262,270,272,274,279,301,326,334,336,346,347,349,351,352,379,385,395,400,401,407,465,467,474,475,490,514]. Amide solvents are typically known for their use in pharmaceuticals and manufacturing materials such as Kevlar [550]. Their widespread application led to new and upcoming ideas for organic amide solvents for membrane operations. Particularly due to their relatively easy synthesis process as well introducing a huge variety of amide solvents possibilities that can be utilised. The flux values range from  $9.66 \times 10^{-5}$ – $9.55 \times 10^{-14} \text{ mol}\cdot\text{cm}^{-2}\cdot\text{min}^{-1}$ . Though amides are known to have comparatively better permeability and selectivity further research is required to find the optimum operating conditions to achieve stable values of flux.

41 studies (8%) used water and ethanol solvents [38,55,108,112,135,164,175,178,181,201,204,214,215,222,223,238,242,246,260,278,285,298,304,307,318,328,329,335,350,361,369,375,382,388,403,451,480,481,486,500,518]. It was found that 93 studies (18%) showcased the use of general organic solvents such as alcohols, acids and salts [23,27,30,31,44,46,48,62,65,66,75–78,80,84,88,102,116,118,132,161,163,169,180,182,186,191,193,194,196,203,210–212,218,221,225,228,235,236,243,248,249,257,261,264,266,268,273,280,286,289,295,297,316,319,332,340,342,348,352,355,360,365,377,387,389,391,397,399,405,406,410,414,415,423,428,433,435,438,454,459,464,479,482,487,494,508,517,519,521,522]. The use of organic solvents is preferred due to economic opportunities that arise as well as being more environmentally friendly, hence a considerable interest in organic solvents. The flux range between  $3 \times 10^{-3}$ – $1.17 \times 10^{-6} \text{ mol}\cdot\text{cm}^{-2}\cdot\text{min}^{-1}$ . As organic solvents do not provide high flux values, this might be linked to the membrane roughness and some structural changes a membrane can undergo when in contact with organic solvents [551]. The absorption efficiency of organic solvents is theoretically better than amine solvents. This is supported by a smaller flux range for studies that used organic solvents, indicating

consistent behaviour. More recently, membrane contactors which utilise immobilised enzymes, such as carbonic anhydrase (CA), for effective CO<sub>2</sub> removal have been studied. For applications at low concentration CO<sub>2</sub> (< 1%, v/v) and near atmospheric reaction conditions, CA is the most efficient catalyst for CO<sub>2</sub> hydration and dehydration, with a turnover number of 10<sup>6</sup> mol CO<sub>2</sub> mol<sup>-1</sup> CA s<sup>-1</sup>. The reaction rate catalysed by CA is much faster than the rate at which CO<sub>2</sub> complexes with other solvents such as MEA [552,553].

The remaining 185 studies (35%) had no information about solvent types [28,29,35,41,43,45,50,54,56,58,64,69,81,83,86,97,104,110,111,113,117,121,126–128,134,136,137,142,145,147,148,156,158,160,165,167,174,177,179,183,185,188–190,199,202,205,219,220,224,226,230,232,234,244,250,251,253–255,258,259,265,267,275,277,281–284,290–294,299,303,305,306,308,312–314,322,323,330,333,337,338,341,343–345,353,354,356,358,359,362–364,366,368,371,373,374,376,380,381,384,390,392–394,398,409,413,417,418,421,426,429–432,436,437,440–443,445–447,449,455,457,461–463,466,468,470–472,477,483–485,488,489,493,495–497,499,504,507,509,510,513,516,520,524–528,530–537,539–546]. Figure 11 shows a visual representation of the different types of solvents and the number of studies that utilised them. Inorganic membranes provide better flux values because organic solvents have can cause denaturing of membranes at high temperature operations.



**Fig. 11** Types of solvent used in the included studies

### 3.2.6 Wetting

Wetting refers to the angle the solvent makes with the membrane, hence determining the solvent

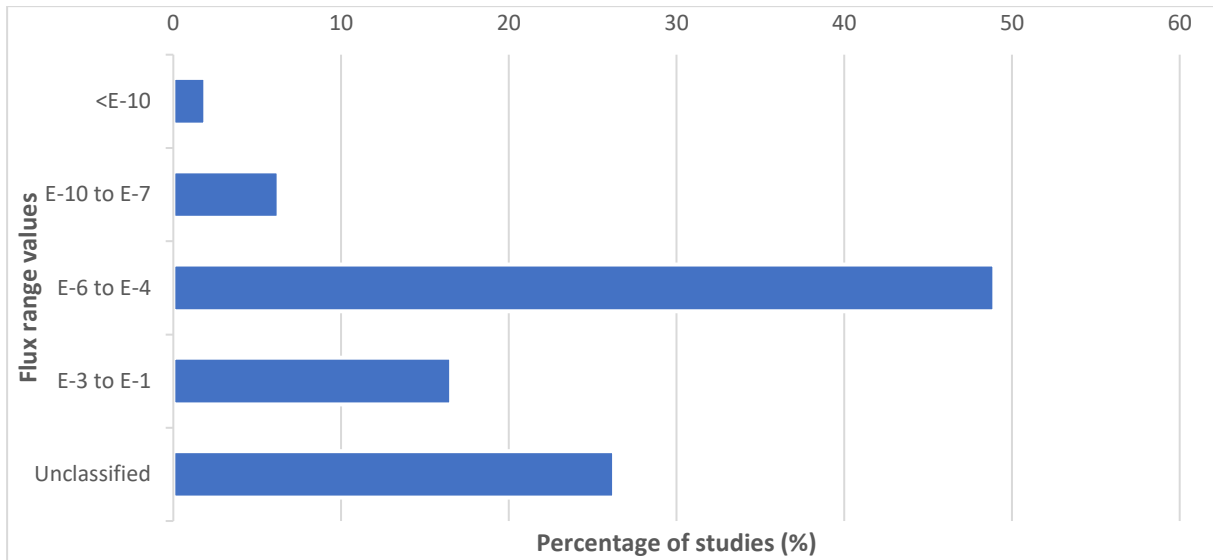
dispersion on the membrane surface. It was found that 8 studies (1.3%) used hydrophilic membranes [54,225,337,433,491,492,515,522]. The majority of the studies (n = 341, 65%) exhibit a phobic behaviour between the solvent and the membrane [20,22–24,26,27,30,31,34–40,42,44,46–53,55,56,58–69,71–78,80,82–86,88–91,94–109,111,114–118,121–123,125,127,129,130–132,133,134,135,137,139,141,143–146,151–157,159,161,163,164,166–171,174,176–181,183–185,188–194,198–211,213,214,216–218,222–224,226–228,230,233–36,238–240,242,244,246,250,252,253,258,261,262,264–270,272–274,276–283,285,286,288–305,307–336,338–356,358,359,360,362,364,365,368,376,377,380,383,384,393,394,400,405,408,410,411,415,419,426–428,431,434,435,437,438,443,445,447–451,453,454,456,458,459,463–467,472,473,475,478–480,482,483,484–489,496,497,500–502,505,506,508,513,514,516,517,519,521,523,524,528,533,538,543]. One of reasons for the low popularity of hydrophilic membranes is the low thermal and chemical stability of membranes, which in turn has an effect on the flux, demonstrated in Table S1, with the values ranging from  $1.1 \times 10^{-3}$ – $1.2 \times 10^{-10} \text{ mol}\cdot\text{cm}^{-2}\cdot\text{s}^{-1}$ . Hydrophobic membranes have better thermal stability and along with lower transport resistance making them more appropriate for gas separation applications, and hence making it a popular option for gas separation studies [554]. The remainder 176 studies (34%) had no information about wetting [21,25,28,29,32,33,41,43,45,57,70,79,81,87,92,93,110,112,113,119,120,124,126,128,136,138,140,142,147–150,158,160,162,165,172,173,175,182,186,187,195–197,212,215,219–221,229,231,232,237,241,243,245,248,249,251,254–257,259,260,263,271,275,284,287,306,357,361,363,366,367,369–375,378,379,381,382,385,387–392,395–399,401–404,406,407,409,412–414,416–418,420–425,429,430,432,436,439–442,444,446,452,455,457,460–462,468–471,474,476,477,481,490,493–495,498,499,503,504,507,509–512,518,520,525–527,529–532,534–537,539–542,544–546].

### 3.2.7 Average flux

Molar flux is known as the amount of substance passing across the membrane, per unit of area and is one of the key parameters to evaluate the performance of a membrane. A higher indication of flux represents effective utilisation of the membrane surface. The flux values ranged from  $10^{-17}$  to  $10^{-13} \text{ mol}\cdot\text{cm}^{-2}\cdot\text{min}^{-1}$ . In 10 studies (2%) the flux values were lower than  $10^{-10} \text{ mol}\cdot\text{cm}^{-2}\cdot\text{min}^{-1}$  [21,65,96,143,194,292,343,379,411,524], and 33 studies (6%) were in the range  $10^{-10} \leq \text{flux} \leq 10^{-7}$  [51,63,68,79,100–102,112,149,155,163,215,224,235,267,295,314,326,380,385,390,401,408,423,431,438,448,458,467,483,514,520,522]. There were 257 studies (48%) which determined the average flux to be within  $10^{-6} \leq \text{flux} \leq 10^{-4}$ , [20,22–27,30,31,33,34,36,40–42,44,46–49,55,60–62,66,67,69,72–78,80–82,84,87–

89,93,97,99,103–106,108,109,111,115,116,118,123,127,129,131–134,137,139,145–148,150,152,153,156,159,161,164,165,167,169,170,171–174,177–179,182–187,191–193,195,197,199–201,204,205,209–214,216–219,221–223,225,226,229–233,236,238–243,246,248,256,261,262,265,266,270,271,273–275,278,279,281,283,284,289–291,294,296–298,300–305,313,316–318,323–325,327,328,334,335,338–340,342,345–347,349–355,357,359,361,365,367,370–372,375–378,381,382,384,387–389,391,392,397,398,399,402,404–406,410,414,416,417,419,420,422,425,427,428,432,433,439,440,452,456,459,460,463,464,474–476,478–482,490–492,496–503,510–513,518,519,526,534] and 87 studies (17%) had the flux range of  $10^{-3} \leq \text{flux} \leq 10^{-1}$  [32,38,50,53,56,57,70,83,86,90,94,98,110,119,135,140–142,175,176,180,181,188,198,206–208,220,237,252,255,257,264,268,277,288,299,307,312,315,319,321,329,332,336,337,341,348,356,358,362–364,369,373,374,393,395,396,407,415,421,426,435,441,447,449–451,453,454,457,465,466,469,484,486,488,505,506,508,515–517,521,523]. The remainder 138 studies (26%) contained no information on the flux [28,29,35,37,39,43,45,52,54,58,59,64,71,85,91,92,95,107,113,114,117,120–122,124–126,128,130,136,138,144,151,154,157,158,160,162,166,168,189,190,196,202,203,227,228,234,244,245,249–251,253,254,258,259,263,269,272,276,280,282,285–287,293,306,308–311,320,322,330,331,333,344,360,366,368,383,394,400,403,409,412,413,418,424,429,430,434,436,437,442–446,455,461,462,468,470–473,477,485,487,489,493–495,504,507,509,525,527–533,535–546].

Figure 12 shows a visual representation of the flux ranges. Table S1 shows that the highest flux was exhibited by polymeric membranes ( $7.6 \times 10^{-1} \text{ mol}\cdot\text{cm}^{-2}\cdot\text{min}^{-1}$ ). However, some ILM studies exhibited a relatively higher flux values when compared to conventional membranes such as polymeric membrane material. These are made up of microporous supports containing cation and anions. The arrangement and structure of these membrane allow for the vapour pressure to be neglected within the system and provide greater viscosity and reducing the solubility thus resulting in effective utilisation of the membrane. These recent studies on ILMs open a new research opportunity for gas separation processes. Some HFMs also displayed high flux in combination with amine solvents. The tubular and small capillary arrangement of these membranes allows the membrane to utilise maximum surface area for CO<sub>2</sub> separation. However due to fouling and breaking issues, the best result is not always achieved.



**Fig. 12** Flux range percentages

### 3.2.8 Gas and liquid flow rate

It was found that 17 studies (3%) had the gas flow rate under  $10 \text{ mL}\cdot\text{min}^{-1}$  [21,44,54,63,88,118,127,157,186,205,225,278,356,407,421,508,529], and 51 studies (10%) set the flow rate between  $10$  and  $100 \text{ mL}\cdot\text{min}^{-1}$  [32,51,59,73,77–79,81,90,98,101,104,115,156,187,188,200,209,217,239,246,272,279,283,294,298,314,325,336,350,357,358,365,372,382,414,423,427,433,449,459,469,472,474,475,492,517,518,520,524,529]. A further 43 studies (8%) set the gas flow rate in the range of  $100 < \text{flow} < 1000$  [25,27,37,38,40,47,49,60,70,72,73,76,85,104–106,115,122,128,131,134,150,152–154,161,162,167,195,285,321,327,364,397,402,417,423,439,469,479,515,521,522]. 13 studies (2%) set the flow rate to  $\leq 1000 \text{ mL}\cdot\text{min}^{-1}$  [87,94,96,139,143,146,151,166,168,266,302,428,467]. The remaining 406 studies (78%) contained no information on the flow rate [20,22–24,26,28–31,33–36,39,41–43,45,46,48,50,52,53,55–58,61,62,64–69,71,74,75,80,82–84,86,89,91–93,95,97,99,100,102,103,107–114,116,117,119,120,121,123–126,129,130,132,133,135–138,140–142,144,145,147–149,155,158–160,163–165,169–185,189–194,196–199,201–204,206–208,210–216,218–224,226–238,240–245,248–265,267–271,273–277,280–282,284,286–293,295–297,299–301,303–313,315–320,322–324,326,328–335,337–349,351–355,359–363,366–371,373–381,383–385,387–395,398–401,403–406,408–413,415,416,418–420,422,424–426,429–432,434–438,440–448,450–458,460–466,468,470,471,473,476–478,480–491,493–507,509–514,516,519,523,525–528,530–546].

It was found that 25 studies (5%) had the liquid flow rate of under  $10 \text{ mL}\cdot\text{min}^{-1}$  [21,29,31,41,47,54,59,82,88,98,157,164,268,278,321,324–326,331,342,348,356,440,467,515]. Forty-two studies (8%) had the flow rate between  $10$ – $100 \text{ mL}\cdot\text{min}^{-1}$

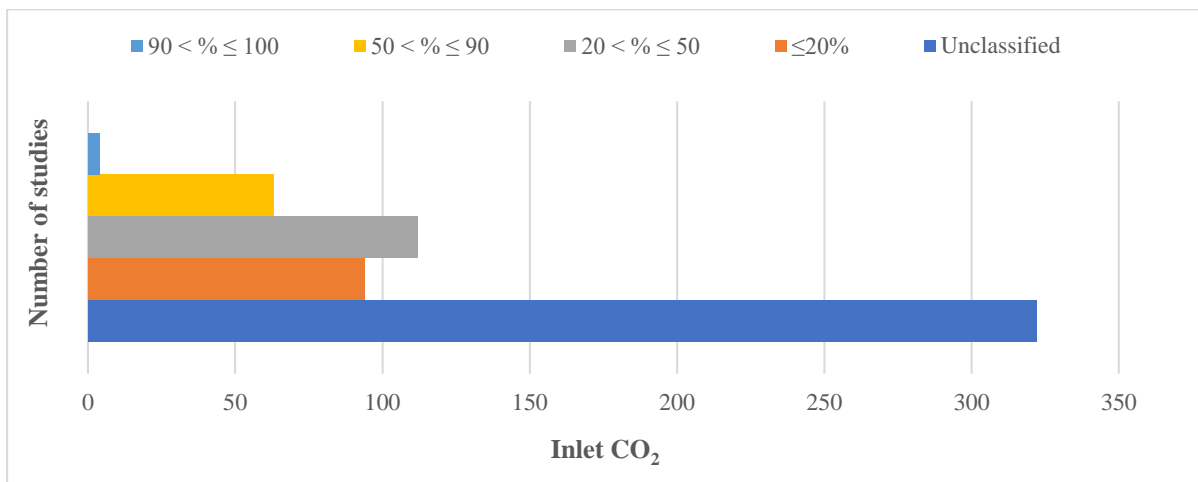
[25,29,32,44,49,65,74,76,77,79,85,87,96,101,106,110,118,128,131,146,151,153,154,156,161,162,166,195,217,246,272,279,298,357,397,432,433,440,447,467,474,520]. Thirty-eight studies (7%) had flow rate in the range of  $100 < \text{flow} \leq 1000$  [32,37,38,40,44,46,48,51,57,70,75–78,84,85,94,101,102,104,106,122,128,131,134,143,152,188,195,337,364,365,396,408,440,472,521,522], and 5 studies (1%) had the flow rate greater than  $1000 \text{ mL}\cdot\text{min}^{-1}$  [145,147,168,496,516]. However, 429 studies (82%) did not provide enough data [20,22–24,26–28,30,33–36,39,42,43,45,50,52,53,55,56,58,60–64,66–69,71–73,80,81,83,86,89–93,95,97,99,100,103,105,107–109,111–117,119,120,121,123–127,129,130,132,133,135–142,144,148–150,155,158–160,163,165,167,169–187,189–194,196–216,218–245,248–267,269,270,271,273–277,280–297,299–320,322,323,327–330,332–336,338–341,343–347,349–355,358,359–363,366–385,387–395,398–407,409,410–431,434–439,441–446,448–466,468–471,473,475–495,497–514,517–519,523–546]. The average gas and liquid flow rates ranged between  $100\text{--}800 \text{ mL}\cdot\text{min}^{-1}$ . A general correlation between flow rates and flux can be deduced. Lower flow rates result in lower flux. This was the expected result, higher flowrate results in more contact with the membrane resulting in higher flux, at any given concentration. However, these relationships do not necessarily hold on smaller preliminary lab scale experiments.

### 3.2.9 Feed $\text{CO}_2$ concentration

About a fifth of the studies 18%, ( $n = 94$  studies) had the inlet feed at less than and including 20% [22,37–39,49,50,56,61,64,68–70,73,87,90,94,112,115,120,121,124,126,127,137,139,140,142,143,145,147,148,151,158,160–163,165,167,168,173,181,187,199,209,217,240,245,251,260,267,270,275,277,290,303,311,312,315,316,319,320,322,323,330,336,338,367,381,390,409,412,421,423,428–430,436,437,453,455,480,488,489,493,497,507,516,520–522,526,529,533]. There were 112 studies (21%) that set the inlet feed between 20%–50% [21,28,34,36,40,42,48,55,57,60,67,71,72,88,90,95–97,99,103,110,117,126,129,131,137,139,155–157,164,172,183,184,187,188,200,205,209,218,221,237,240,248,251,261,265,277,279,281,292,298–300,313,320,321,323,325,327,329,341,344,352,356–359,362,365,370,371,373,375,379,381,382,385,388–390,392,393,398,407,410,414,416,419,425,427,435,439,440,444,446,455,457,470,491,507,510,513–515,519,523,529,531,532,534,536]. For 63 studies (12%) the inlet feed composition was set between 50%–90% [27,30,40,55,71,79,88,90,92,95,97,99,126,132,139,146,172,183,184,200,213,218,221,237,248,251,281,291,298,314,320,341,359,362,371,373,375,379,381,382,388–390,392,393,398,407,410,414,416,425,427,435,440,457,507,510,513,514,523,524,528,529]. Four studies (1%) had inlet compositions up and including 100% and this was done to study the permeability and solubility of the membrane [54,79,92,266]. Figure 13 shows a visual representation



of inlet CO<sub>2</sub> ranges and the number of studies. The inlet feed CO<sub>2</sub> ranged from 1.8% to 100%, with many of the studies (142) keeping the inlet feed at the standard feed composition of industrial feed (between 14%–50%). As a lot of studies were lab scale experiments along with simulated models, 14%-50% provides a better representation of CO<sub>2</sub> capture feed. The remaining 322 studies (62%) had no information on the CO<sub>2</sub> feed [20,23–26,29,31–33,35,41,43–47,51–53,58,59,62,63,65,66,74–78,80–86,89,91,93,98,100–102,104–109,111,113,114,116,118,119,122,123,125,128,130,133–136,138,141,144,149,150,152–154,159,166,169–171,174–180,182,185,186,189–198,201–204,206–208,210–212,214–216,219,220,222–236,238,239,241–244,246,249,250,252,253–259,262–264,268,269,271–274,276,278,280,282–289,293–297,301,302,304–310,317,318,324,326,328,331–335,337,339,340,342,343,345–351,353–355,360,361,363,364,366,368,369,372,374,376–378,380,383,384,387,391,394–397,399–405,406,408,411,413,415,417,418,420,422,424,426,431–434,438,441–443,445,447–452,454,456,458–462,463–469,471–479,481–487,490,492,494–496,498–506,508,509,511,512,517,518,525,527,530,535,537–539,540,541,542,543,544,545, 546].



**Fig. 13** The inlet feed of CO<sub>2</sub> concentration for the included studies

#### 4 Quality of the evidence and bias assessment

Although we did not perform a quality assessment on the included studies, we could not identify a validated tool that can be used in the engineering field, in a similar way that various ones are being widely used for the appraisal of healthcare interventions. For the purposes of our study, we considered adequate that the conducted studies have undergone peer-review to publication. Potentially, it would be useful to perform future studies on the construction and validation of such quality assessment tools

specifically for experimental and theoretical studies in membrane contactor systems. We did not assess conflicts of interests, such as industrial collaboration or funding, on any of these studies, but we consider these to be important aspects in checking for biases in the reported methods and results. Our review process was systematic in that we defined a search strategy, run it across three key databases where engineering work is published, with no language, time or geographical restrictions. At every stage at least two authors were independently screening and extracting data, reducing the potential for error. However, this impacted the duration of the overall process from the initial design, search, data extraction and reporting of the primary studies which took almost three years.

## 5 Implications for research and practice

The review highlights for the first time, the research evidence on the capture of CO<sub>2</sub> using various membrane systems. Although patents, books and conference proceedings were excluded from this review, the included peer-reviewed studies have indicated that HFMs are the most common practice of gas separation methods in industry, along with the use of inorganic solvents in these separation methods providing best results. From the 525 published research studies on the CO<sub>2</sub> capture using different membranes there are three types of membrane contactors identified: FS (15%), FT (6%), and hollow fibre (33%). The flow configuration was co-current in 22% of the studies and counter-current in 22%. Although three main solvent types were used: amines, amides, and water and ethanol solvents, there were inorganics such as metal nitrates and silane, general organic solvents such as alcohols, acids and salts (18%) also explored. The majority of studies (65%) favour a phobic behaviour between the solvent and the membrane and future studies should avoid hydrophilic membranes. The inclusion of more information around the membrane material, membrane contactor type, flow configuration and other identified parameters can lead to the design of better studies on optimally capturing higher concentrations. Future studies should try to address the issue of efficient CO<sub>2</sub> capture by using membranes tested under ILMs and facilitated membrane transport. ILMs and FT have advantages from a chemical and economical perspective, however, further research should focus on how to overcome the issue of thermal stability and lack of reliability on hydrodynamic application in industry.

## 6 Conclusions

This study started from 2,650 papers down to 525 final included studies (shown in Fig. 2). This displays that membrane technology for CO<sub>2</sub> capture has attracted a lot of research attention from research in the past three decades. An efficient method for CO<sub>2</sub> post and pre-processing is yet to be established. Membrane carbon capture and storage, if established, can be operated in a continuous system as opposed to current adsorption and absorption of CO<sub>2</sub> in batch systems. Different kinds of membranes have been investigated to study how membrane systems can be applied and optimised on an industrial scale.

Polymeric membranes have low operating costs and zeolite membranes have high durability and

recyclability making them both an attractive common starting place for investigations. Zeolites were initially preferred due to their durability of high temperatures and sorption-diffusion mechanism in separating CO<sub>2</sub>. However, they cannot be widely applied due to high manufacturing costs, which may explain why they have only been tested in 5% of the studies. Some have proposed the solution of modifying the zeolite structures by integrating polymers and MMMs but that is yet to be researched further. Polymeric membranes were found to be very popular due to the range of structural possibilities they hold, as well as being economically feasible. ILMs were one of the least popular choice amongst the studies. Although recent advancements established them with better performance at low concentrations when compared to other membranes, ILMs are not widely applied because the membranes cannot withstand high temperatures, and the hydrodynamics of the membranes is yet to be properly understood.

The application of polymer membranes has transitioned into the use of MMMs where organic polymers are imbedded into inorganic casings. This structural arrangement provides higher flux and better separation performances than simple conventional polymer membranes. In 15% of the studies experiments were conducted with MMMs and found great potential. However, issues of incorrect solvent application and inconsistency in the flux values require further investigation. HFMs were found to be the most popular choice due to their versatility and wide range of applications. These are known for gas separation applications which may justify their use in 28% of the studies which experimented with different kinds of HFMs. The HFMs can have various configuration possibilities with different combinations of polymers and have gained a lot of interest as they display good performances. However, further research is required to overcome fouling issues and developing a more economical manufacturing and operating processes. Different kinds of amine solvents were found to be the most popular choice for the membrane studies (20%). Amine solvents have a high CO<sub>2</sub> capacity, low solvent degradation during the absorption and regeneration process, as well as exhibiting better tolerance for regeneration at high pressures. Counter-current flow was the most popular choice of flow configurations over concurrent flow as it provides a better thermodynamic environment as well along with larger concentration gradients promotes gas separation. The main limitation of CO<sub>2</sub> membrane capture can be evaluated by a compromise between flux, membrane stability and economic implications. The systematic review of all of the studies in the CO<sub>2</sub> removal and capture is an important milestone in the recognition of the most up to date research work. It also provides additional benefits by serving as a rich databank for further research and benchmarking and in identifying areas of further research priority.

This study did not focus on papers that involved biological membrane for CO<sub>2</sub> transfer. This was done to keep the focus on CO<sub>2</sub> separation in the energy sector. Studies that modelled membrane systems using different computational programmes, the effect of programmes was not discussed, rather the flux and other parameters were included in the table ([supplementary material](#)). Future

research should be focused around CO<sub>2</sub> capture using ILMs and facilitated membranes tested under organic and inorganic solvents to form a well-rounded evaluation of these membrane applications in industry, from a chemical and economical prospective. Stability issues of HFM should be investigated to better understand their potential to widely commercialised. Some research could be focused around optimising polymeric and zeolite CO<sub>2</sub> membrane separations systems or upgrading the existing systems into MMMs systems.

Electronic Supplementary Material    Supplementary material is available in the online version of this article at <http://XXXX> and is accessible for authorized users.

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