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Ionic liquids combined with membrane separation processes: A review

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1 Contents

| | | |
|----|--|----|
| 2 | Ionic Liquids Combine with Membrane Separation Processes: A Review | 1 |
| 3 | 1. Introduction | 3 |
| 4 | 2. Physicochemical properties of ionic liquids..... | 10 |
| 5 | 3. Membrane processes containing ILs | 12 |
| 6 | 3.1 Supported ionic liquids membranes (SILMs)..... | 13 |
| 7 | 3.1.1 Separation of VOCs by SILMs..... | 16 |
| 8 | 3.1.2 Separation of CO ₂ by SILMs..... | 17 |
| 9 | 3.2 IL-Polymer membranes (ILPMs) | 21 |
| 10 | 3.2.1 Separation of VOCs by ILPMs..... | 23 |
| 11 | 3.2.2 Separation of CO ₂ by ILPMs..... | 25 |
| 12 | 3.3 Ionic liquids composited mixed matrix membranes (ILMMMs) | 27 |
| 13 | 3.3.1 Separation of VOCs by ILMMMs..... | 28 |
| 14 | 3.3.2 Separation of CO ₂ by ILMMMs..... | 28 |
| 15 | 3.4 Poly(ionic liquid)s membranes (PILMs) | 30 |
| 16 | 3.4.1 Separation of VOCs by PILMs..... | 32 |
| 17 | 3.4.2 Separation of CO ₂ by PILMs..... | 32 |
| 18 | 3.5 Ionic liquid-gel membranes (ILGMs)..... | 35 |
| 19 | 3.5.1 Separation of VOCs by ILGMs | 36 |
| 20 | 3.5.2 Separation of CO ₂ by ILGMs | 36 |
| 21 | 3.6 Ionic liquid membrane contactors (ILMCs) | 37 |
| 22 | 3.6.1 Separation of VOCs by ILMCs | 38 |
| 23 | 3.6.2 Separation of CO ₂ by ILMCs | 40 |
| 24 | 4. Conclusion..... | 43 |
| 25 | Acknowledgments | 46 |
| 26 | References | 47 |
| 27 | | |
| 28 | | |

1. Introduction

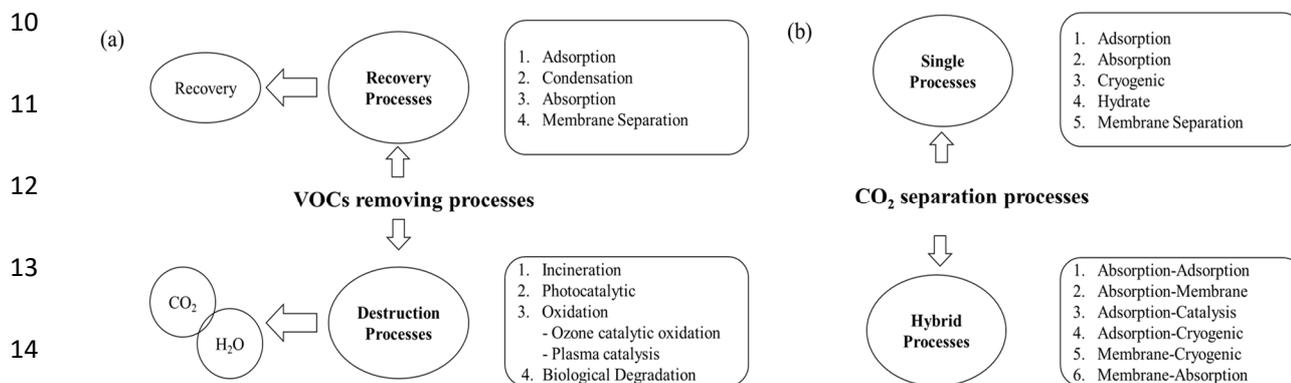
Environment pollution and climate change caused by volatile organic compounds (VOCs) and carbon dioxide (CO₂) have attracted increasing concerns. VOCs with the boiling point ranging from 323 to 523 K are among the main air pollutants [1, 2]. They include a multitude of substances and are easily evaporate at room temperature [2 - 4]. The volatility of VOCs allows them to spread around the sources of pollution. They often cause serious environmental problems, such as greenhouse effect [5 - 7], photochemical smog [8, 9], stratospheric ozone depletion [10 - 13] and so on. These potential risks have resulted in stricter environmental legislations in most countries, forcing industry companies to treat their air emissions before discharging them into the atmosphere [14]. Table 1 shows the common VOCs discharged into the atmosphere and the negative effects on human health. Therefore, developing effective VOCs elimination techniques and optimizing VOCs removing processes become societal issues. The representations of VOCs were selected based on (i) by sources of emissions and dangerous to human health and (ii) the concentrations of VOCs make immediately dangerous to health (National Institute of Occupational Safety and Health).

Table 1. Health effects of common VOCs [15 - 23].

| Classification | Representation | Health Effects |
|--------------------|--|---|
| Aldehydes | Acetaldehyde Formaldehyde | -Nasal tumors -Predecessor of ozone -Irritation of eyes, skin, and throat |
| Ketones | Ethyl butyl ketone | -Central nervous system depression -Headache and nausea |
| Aromatic compounds | Benzene Toluene O-xylene and P-xylene Ethylbenzene | -Carcinogen -Damage the ozone layer -Irritation the central nervous system -Produce photochemical smog, and pose mutagenic hazards |
| Halogenated VOCs | Carbon tetrachloride Trichloroethylene Tetrachloroethylene | -Strong bioaccumulation potential -Cause greenhouse effects |

| | | |
|----------------------------------|-------------------------------|---|
| Alcohols | Isopropyl alcohol Methanol | -Eyes irritation -Central nervous system depression -Throat irritation and shortness of breath |
| Alkenes | Propylene Ethylene | -Photochemical ozone creativity -Potentially carcinogenic and adversely affects the odor and taste of drinking water |
| Polycyclic aromatic hydrocarbons | Phenanthrene | -Carcinogen |

1
2 The VOCs separation processes in industrial fields are mainly divided into two parts
3 according to the destruction or recovery of the pollutants, as shown in Figure 1(a): the processes
4 based on destruction including incineration [24 - 28], photocatalytic [29 - 35], catalysts [36 -
5 39], oxidation [40 - 43] and biological degradation [44 - 47], and those based on recovery
6 including adsorption [7, 48 - 52], condensation [53, 54], absorption [55 - 60] and membrane
7 processes [61 - 66]. Compared to the destruction processes, the recovery processes are more
8 economic, because it is possible to regenerate both important compounds and value pollutants.
9 Table 2 resumes the advantages and disadvantages of VOCs separation processes.



16 **Figure 1.** Main processes of VOCs (a) and CO₂ (b) separation.

17 Adsorption is economical and environment-friendly, in which a suitable adsorbent is
18 critical. Numerous adsorbents have been studied, such as active carbon [7, 67 - 71], zeolite [72
19 - 77], polymeric adsorbents [78 - 83], to selectively adsorb VOCs. However, the high cost and
20 the regeneration of adsorbents are the major limitations of adsorption processes. For high
concentrations of VOCs, adsorption with a suitable solvent is a commonly method. It is an

1 undeniable that the spent solvent as the absorber and the disposal of VOCs are the key problems.
 2 Condensation processes can be used to remove VOCs but one of major restriction is the disposal
 3 of the spent coolants. Because of the energy intensive nature of the process, condensation is
 4 limited to only evaporative solvents.

5 **Table 2.** Characteristics of VOCs removal processes.
 6

| VOCs processes | Methods | Waste generation | VOCs | Efficiency | VOCs concentration | Advantages (+) Disadvantages (-) | Ref. |
|------------------|---------------------|------------------|-------------------------------------|--------------------|--------------------|--|------|
| Recovery process | Adsorption | Spent adsorbent | Benzene Toluene | >90% | 700-1000 ppm | (+) Fast removal (+) Recovery of pollutants (-) Desorption phenomena during adsorption process (-) Competitive adsorption of other contaminants | [67] |
| | Condensation | - | Methanol Methyl-iso-butyl ketone | - | >5000 ppm | (+) Recovery of pollutants (+) Treat high concentration of contaminants (-) High energy consumption (-) Low efficacy | [84] |
| | Absorption | - | Toluene Dimethyl disulfide | - | - | (+) Fast removal (-) Gas-liquid absorber depend on equilibrium solubility | [55] |
| | Membrane separation | Clogged membrane | Dichlorobenzene | Benzene Toluene | <25% | (+) High efficacy (+) Treat a wide range of VOCs (-) Cleaning requirements (-) Low concentration of VOCs is limited (-) Increasing the pressure drop results in a greater mechanical resistance during operation | [66] |

| | | | | | | | |
|---------------------|---------------------------|---|--|--------------------|-----------|--|----------|
| Destruction process | Incineration | CO ₂ , CO, NO _x | - | >99% (40 min) | 20% - 25% | (+) Efficient destruction within short time (-) High construction cost (-) Potential formation of high toxic byproducts | [25, 26] |
| | Photocatalytic oxidation | Strong oxidant OH-radicals | Toluene Decane Trichloroethylene | 100 % (5 min) | - | (+) Fast and high efficacy (-) High energy consumption (-) Byproducts formation (-) Low recovery of catalysts | [86] |
| | Ozone-catalytic oxidation | Organic aerosols | Dichlorobenzene Trichlorobenzene | 100% (2 h) | - | (+) Effective and fast removal of contaminants (-) Low solubility of ozone in water (-) Ozone scavengers commonly found in environment (-) Incomplete oxidation | [86, 87] |
| | Plasma catalysis | Carboxylic acids NO _x O ₃ | Phenol | 74%-81% | - | (+) Efficient (-) Poor energy efficiency for the treatment of low VOCs (-) Formation of undesirable byproducts | [88] |
| | Biological degradation | Acetaldehyde | Formaldehyde Benzene Toluene Xylene | 100% (7 months) | <5000 ppm | (+) Low cost (+) In site remediation (-) Slow kinetics (-) Inhibitive nature for local microorganisms | [89] |

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In addition, it should not be ignored that the climate change is caused by increasing concentration of CO₂ emission. Therefore, it is necessary to develop effective processes for CO₂ separation. Recently, in order to against global warming, the CO₂ separation processes has attracted higher concern. They are mainly divided into two parts according to the numbers of stage processes required to meet the separation targets, as shown in [Figure 1\(b\)](#).

7

The major challenge of CO₂ separation processes is high energy consumption. Also, there

1 are some disadvantages of each process which restrict their application and development, such
 2 as the secondary pollution [90], degradation of solvent [91, 92], high equipment cost [93, 94],
 3 and limited selectivity [95, 96]. To overcome the challenges of single separation process, hybrid
 4 processes should be a promising alternative. Combination of different processes can enhance
 5 the separation performance and avoid their disadvantages. However, the hybrid process is
 6 subject by standalone process. Table 3 summarized the advantages and disadvantages of CO₂
 7 separation processes.

8 **Table 3.** The advantages and disadvantages of CO₂ separation processes [97 - 100].
 9

| CO ₂ separation processes | | Advantages (+) | Disadvantages (-) |
|--------------------------------------|------------|--|--|
| Single processes | Adsorption | (+) High capacity of CO ₂ adsorption (+) Withstands high-pressure operation (+) High selectivity over other gases | (-) High energy consumption (-) Difficulty of regeneration (-) Not suitable for cyclic operation |
| | Absorption | (+) Suitable for industrial scales (+) Operating for low CO ₂ feed concentration | (-) High energy consumption (-) Corrosion caused by amines degradation (-) Emissions of harmful by-products (-) Expensive for regeneration |
| | Cryogenic | (+) High product purity | (-) High energy requirement (-) Only suitable for high CO ₂ feed concentration (> 50%) (-) Need pre-processing step to remove humidity from mixed feed stream |
| | Hydrate | (+) High product purity | (-) High pressure and low temperature (-) Cause secondary pollution (-) High capital cost |
| Hybrid processes | Membrane | (+) Installation easily (+) Operating simply (+) Low energy consumption | (-) High surface area of membrane (-) Humidity of feed gases affect the permeability, especially polymeric membrane (-) Performance is affected by operating conditions |
| | | | |

| | | |
|---------------------------|---|--|
| Absorption -Adsorption | (+) Higher CO ₂ carrying capacity (+) Low heat capacity | (-) High sorbent regeneration energy (-) Emission of harmful by-products (-) High-pressure drop across the absorber of small particles |
| Absorption -Membrane | (+) Low regeneration energy (+) Low capital cost (+) Membrane process can reduce amine emission | (-) Only suitable for relative low CO ₂ feed concentration (13%-23%) (-) Corrosion caused by amines degradation |
| Adsorption -Catalysis | (+) Relative high separation rate (+) Increasing the conversion of CO ₂ (+) Capital cost is reduced (+) Some absorbents allow two processes occur in on single simultaneous | (-) Need pre-processing step to remove H ₂ from mixed feed stream (-) Need to find a suitable catalyst |
| Adsorption -Cryogenic | (+) Low energy consumption | (-) Not mature enough to apply in industrial scales |
| Membrane -Cryogenic | (+) The size of cryogenic equipment is reduced (+) Process efficiency improved (+) Energy requirement reduced | (-) Need to select a condensing agent (-) Need future reduce energy consumption |
| Membrane -Absorption | (+) High purity (+) High recovery | (-) Need future reduce energy consumption (-) Only suitable for relative low CO ₂ feed concentration (15%-36%) |

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Several CO₂ separation processes have been developed in the last decades, for example absorption, adsorption, membrane, cryogenic, and hydrate etc. In the CO₂ capture approaches, cryogenic and hydrate are low-temperature processes. Cryogenic process is gases convert into their liquid phase at very low temperatures. Hydrate is a process to form a solid hydrate. Hydrate units are introduced in this process to enhance the hydrate formation and reduce the equilibrium conditions. Low temperature and high pressure are necessary conditions for cryogenic and hydrate [101, 102]. Absorption, and adsorption have been widely applied to many industrial fields and they are relative matured.

1 Among all the separation processes, membrane-based separations are deemed as one of
2 the most widely studied and fastest growing separation processes for efficient pollutants
3 treatment [103 - 105]. In order to overcome some disadvantages of membrane-based separation
4 processes and further improvement of their separation performance, researchers are committed
5 to not only optimizing operating parameters but also developing new membrane materials and
6 membrane processes.

7 Ionic liquids (ILs) are organic molten salts with many excellent properties such as no vapor
8 pressure above the liquid surface, thermal stability, and low volatility [106 - 108]. Within this
9 regard, they have been found uses in a widely range of research fields such as electrolytes [109
10 - 113], chemical solvents [114 - 117], and catalysts [118, 119]. In addition, they have been
11 successfully applied to several separation processes, such as removing heavy metal ions [120,
12 121], greenhouse gases treatment [122 - 125], and VOCs removal [1, 126 - 128]. Besides, the
13 most attractive property of ILs is tailor-made, which means IL's properties can be modified via
14 alteration of cations and anions to produce specific compounds for given applications [129,
15 130]. Therefore, ILs are regarded as effective green solvents alternative to traditional organic
16 solvents. However, several studies have underlined that there are some negative effects of ILs
17 on environment resulting in a limitation of their applications in industry [131 - 133]. It is
18 necessary to find a method to break this limit. Although, it is possible to change the ILs chemical
19 compounds to reduce their environmental risk [134, 135]. It will increase difficulty to meet the
20 given processes requirements with reduction of their hazard potential. In this case, stability of
21 the ILs into a supported media provides a promising way to realize the use of ILs with
22 environmental damages reducing.

23 ILs combined with membrane processes have becoming a promising approach in
24 separation fields in recent years. It can promote solvent properties of ILs and enhance
25 membrane separation performances. In the past few years, many different types membrane
26 separation processes containing ILs have been studied, including supported ionic liquid
27 membranes (SILMs), ILs composite polymer membranes (ILPMs), ionic liquids composite
28 mixed matrix membranes (ILMMMs), poly(ionic liquid)s membranes (PILMs), ionic liquid gel
29 membranes (ILGMs), , and ionic liquid membrane contactors (ILMCs). They are mainly focus

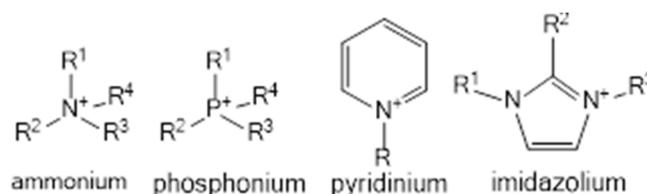
1 on VOCs or CO₂ removing. This review firstly provides comprehensive overview on the use of
 2 membranes processes with ILs for VOCs or CO₂ removal. Moreover, the problems and
 3 challenges in ionic liquid membrane separation processes are identified and discussed.

4 2. Physicochemical properties of ionic liquids

5 Ionic liquids (ILs) have been got lots of attentions because of their physicochemical
 6 properties and potential roles in sustainable development. Specifically, room temperature ionic
 7 liquids (RTILs) are salts in liquid at room temperature [136]. The most significant application
 8 is RTILs as solvents to replace volatile solvents due to their excellent physicochemical
 9 properties. Table 4 exhibits the main difference of features between RTILs and other traditional
 10 solvents. This table 4 is given for comparison and the majority of ILs display these properties.

11 **Table 4.** Comparison of RTILs and traditional organic solvents [136, 140]

| Features | RTILs | Traditional solvents |
|------------------------------|---------------------|-----------------------------|
| Applicability | Multifunctional | Single functional |
| Chirality | Tunable | Rare |
| Vapor pressure | Negligible | Follow C-Clapeyron equation |
| Flammability | Nonflammable | Flammable |
| Solvation | Strongly solvating | Weakly solvating |
| Polarity | Polarity depends | Conventional polarity |
| Tuneability | Designer solvents | Limited range |
| Recyclability | Economic imperative | Green imperative |
| Viscosity (cP) | 22-40 000 | 0.2-100 |
| Density (g cm ³) | 0.8-3.3 | 1.3-1.6 |



12 **Figure 2.** Common organic cations of ILs.

13 Furthermore, RTILs exist as liquid at ambient temperatures due to their chemical
 14 structure. The anion and cation are chosen precisely to destabilize the solid-phase crystal.
 15 Therefore, even though there are no set rules to making an IL, this can be achieved within ion
 16 structures by balancing ion-ion interactions. Structure and nanostructure control ionic liquid

1 properties [137]. Two special types of mesoscale structures exist of in the ILs system. i)
 2 hydrogen bond, ii) ionic cluster. The viscosity of IL was effect by hydrogen bond. For example,
 3 strength of hydrogen bonds is determined items to improve viscosity of pyridinium ionic liquids
 4 [138]. Ionic clustering will influence the IL properties and behaviors including viscosity,
 5 dissolution, and acidity or alkalinity [139]. The multiple combinations cation/anion can lead to
 6 more than 10^6 different ILs, each of them with specific physicochemical properties. In this case,
 7 ILs are also regarded as designer solvents. The cations are generally compositing organic
 8 compounds as ammonium, phosponium, pyridinium or imidazolium (Figure 2) with alkyl
 9 chains (R^n) [142]. The anions of the ILs could be organics or minerals (Table 5). Usually, the
 10 physical properties were controlled by cations, and the anions reflect chemical properties [141].
 11 Based on steric hindrance, the cation/anion asymmetry and the alkyl chains length (Table 6)
 12 have dramatic effects on the melting point which decrease with that parameters increasing [143,
 13 144]. The low melting point and the thermic stability ($>350^\circ\text{C}$) allow the use of ILs in a large
 14 thermic operating range (300 - 400°C). The density of ILs with exception of the tetraalkylborates
 15 is generally higher than water. Moreover, the density decreases with the cation alkyl length
 16 increasing [145]. In addition, the density is also controlled by the anion. The viscosity of ILs is
 17 higher than organic solvents. When the cation alkyl length increase, the viscosity is higher.
 18 [146]. ILs are considered as polar solvents and their polarity close to the alcohols with short
 19 alkyl length [147]. Generally, the polarity can affect the solubility of ILs. The aromatic
 20 compounds are more than ten times the alkyl compounds (Table 7).

21

Table 5. Common anions of ILs.

| Organic anions | Mineral anions |
|---|---|
| CH_3CO_2^- $\text{OTs} (= \text{C}_6\text{H}_5\text{SO}_3^-)$ $\text{NTf}_2 (= \text{N}(\text{SO}_2\text{CF}_3)_2^-)$ $\text{OTf} (= \text{SO}_3\text{CF}_3^-)$ $\text{CTf}_3 (= \text{C}(\text{SO}_3\text{CF}_3)_3^-)$ | $\text{X} (= \text{F}, \text{Cl}, \text{Br}, \text{I})$ BF_4^- , PF_6^- Lewis Acid + X^- |

22

23

Table 6. Effects of ILs characterizes on their properties.

| Effects of the cation/anion asymmetry on the melting point | | |
|--|---------|-------------------|
| X^- | r (Å) | Melting point (K) |
| | | |

| | | | | |
|--|--------------------------|--------------------------------|----------------------------------|----------------|
| | | Na ⁺ X ⁻ | Emim ⁺ X ⁻ | |
| Cl ⁻ | 1.7 | 1074 | 360 | |
| PF ₆ ⁻ | 2.4 | > 473 | 333 | |
| AlCl ₄ ⁻ | 2.8 | 458 | 279 | |
| Alkyl length effects on melting point of the bromide salts | | | | |
| Cations | | Melting point (K) | | |
| [NMe ₄] ⁺ | | > 573 | | |
| [Net ₄] ⁺ | | 557 | | |
| [NBu ₄] ⁺ | | 399 | | |
| [NHex ₄] ⁺ | | 373 | | |
| [NOct ₄] ⁺ | | 368 | | |
| Physicochemical values of some ILs | | | | |
| ILs | M (g mol ⁻¹) | T (K) | Density (g cm ³) | Viscosity (cP) |
| [Bmim][BF ₄] | 226 | 193 | 1.12 | 233 |
| [Bmim][AlCl ₄] | 321 | - | 1.33 | 19 |
| [Bmim][NTf ₂] | 419 | 267 | 1.44 | 69 |
| [Bmim][PF ₆] | 284 | 265 | 1.37 | 257 |
| [Bmim][OP(O) (OBu) ₂] | 348 | - | 1.04 | 1896 |
| [Mmim][OP(OMe) ₂] | 222 | - | 1.26 | 363 |

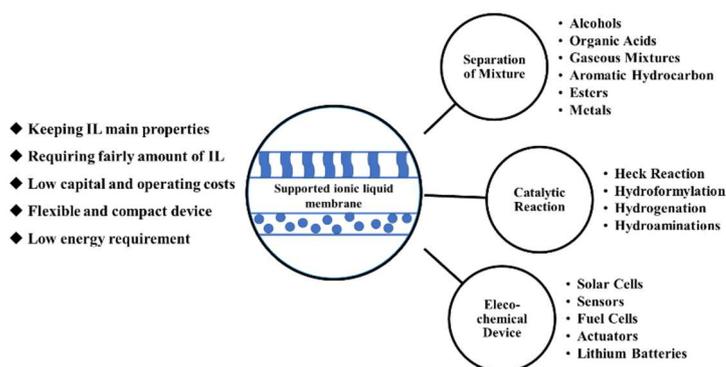
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3. Membrane processes containing ILs

ILs are made by associating large organic cations with a wide variety of anions. This results in a large variety of ILs can be adapted to a given process. Combination of these features can bring new opportunities in the use of ILs-membranes and processes in VOCs and CO₂ separation applications [148 - 150], which are energy-saving and environmentally-friendly (Figure 3). In recent years, many studies focus on the use of ILs in membranes and membrane processes. The disadvantages of ILs, such as high viscosity, high production costs, unclear toxicities, and potential environment effects [151 - 155], limit their industrial applications. However, consisting of immobilizing ILs into support materials in separation processes, which allows the amount of active phase needed for a given process to be minimized and greatly facilitates the recovery and reusability of ILs. The use of matrix to support ILs enables it to obtain a membrane that serves as a separator between a feeding and a receiving phase. Different types of membrane and membrane processes containing ILs are listed, discussed and compared

1 in this review, including supported ionic liquid membranes (SILMs), ILs composite polymer
 2 membranes (ILPMs), ionic liquids composite mixed matrix membranes (ILMMMs), poly(ionic
 3 liquid)s membranes (PILMs), ionic liquid gel membranes (ILGMs), and ionic liquid membrane
 4 contactors (ILMCs). For each configuration, advantages and disadvantages are presented and
 5 more especially for the treatment of VOCs and CO₂ which are the most significant compounds
 6 treated.



12 **Figure 3.** Properties and applications of membrane processes containing ILs for VOCs separations.

13 **3.1 Supported ionic liquids membranes (SILMs)**

14 Supported ionic liquid membranes (SILMs) are porous supports which pores are
 15 impregnated with ILs [156 - 158]. They have been showing several potentials in different
 16 separation applications, such as organic compounds removing [159, 160], separation of mixed
 17 gases [161 - 163], and vapor permeation [164 - 167]. Generally, the driving force is due to the
 18 concentration or pressure gradient between the feed and receiving phase which results in energy
 19 conservation [168, 169]. In addition, high selectivity and low solvent holding of SILMs are also
 20 attractive advantages. Whereas, the primary disadvantage is insufficient membrane stability for
 21 large scale and long-time operations, which has been proven by several studies [164, 168, 170,
 22 171]. The selectivity of SILMs will decreased strongly because ILs was pushed out of porous
 23 support under over-time operation.

24 For industrial application, the most important property is the process stability. In recently,
 25 most researchers focus on improving the stability of SILMs for long-time operation. According
 26 to the previous studies, the strategies to improve SILMs stability are mainly from following

1 three points: properties of support membrane, viscosity of ILs and preparation ways.

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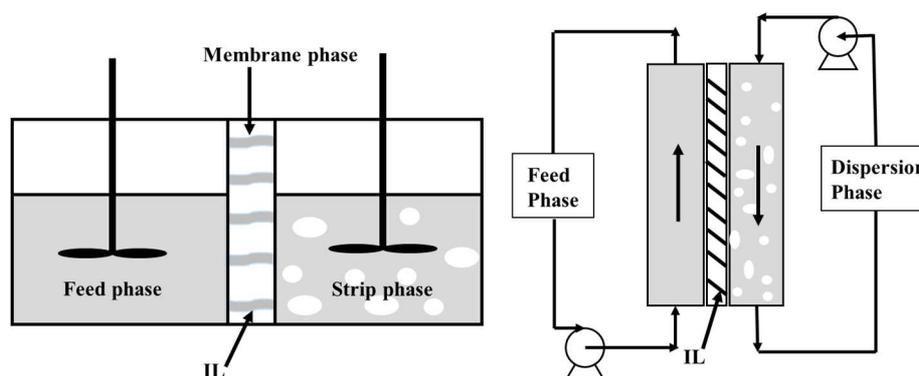


Figure 4. Schematic of SILMs [216].

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It is important for the stability of SILMs to choose a suited support membrane. Generally, the support membranes are including inorganic and polymeric membranes. For the inorganic membrane as a support, ceramic membranes are the most widely used due to high mechanical strength and thermal stability. In this case, the SILMs with ceramic membranes as supports can be used at relatively high temperature and pressure of operations. For example, Karousos et al. [172] developed a ceramic membrane with 1-methyl-3-octylimidazolium tricyanomethanide ([Omim][TCM]), which was subjected to a cyclic heating process up to 453 K. Additionally, a thin ionic liquid membrane with a ceramic support that can resist up to 55 bar transmembrane pressure, which was presented by Kreiter et al. [173]. In addition, carbon materials also are a good choice for the inorganic support in SILMs. Chai et al. [174] used a carbon-graphitic carbon nanocomposite membrane to support imidazolium-based RTILs (1-alkyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([C_nmim][TNf₂], n = 2, 4, and 6). This supported membrane greatly enhanced the stability of SILMs. The SILMs can operate under transmembrane pressures much higher to 1000 kPa without degrading their separation performances. For the polymeric membrane as a support, Nylon, Polypropylene (PP), Polyvinylidene fluoride (PVDF), polytetrafluoroethylene (PTFE), and Polysulfone (PS) are widely used to prepare SILMs. According to research of Riso et al [175], they studied the Nylon membrane as a support based on three ionic liquids such as 1-butyl-3-methylimidazolium hexafluorophosphate ([Bmim][PF₆]), 1-butyl-3-methylimidazolium tetrafluoroborate ([Bmim][BF₄]) and 1-butyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([Bmim][NTf₂]). As shown in Figure 4, the feed and strip phases were separated by an organic membrane phase. They found that the

1 mass of ILs retained in membrane pores was constant during operation. Cichowska-
2 Kopczynska et al. [176] investigated the imidazolium ionic liquids supported on PP. They found
3 that maximum pressure possible to apply without membrane damage was 59 kPa. They also
4 revealed that thickness of PP support was higher after swelling ILs, which could change the
5 structure of polymer network and enhance its mechanical stability [177]. The mechanical
6 property of PVDF support on 1-ethyl-3-methylimidazolium ethyl sulfate ([Emim][EtSO₄]) and
7 1-ethyl-3-methylimidazolium acetate ([Emim][Ac]) were studied by Gomez-Coma et al. [178],
8 and these membranes showed better mechanical properties than others. Moreover, to improve
9 stability of SILMs, PVDF was also used as a support in Kim's study [179]. This SILMs based
10 on 1-n-octyl-3-methylimidazolium hexafluorophosphate ([C₈mim][PF₆]) and hydrophobic
11 PVDF, in which the ideal selectivity was improved even after 8 cycles under 298 K and 1.2 bar
12 [180]. Hanioka et al. [181] examined the long-term stability of a PTFE membrane supported n-
13 aminopropyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([C₃NH₂mim][NTf₂]).
14 They found that membrane kept its permeability and selectivity under atmospheric pressure
15 even after 260 days. Therefore, PS has been used to synthesize support membranes due to
16 excellent mechanical strength, high thermal and chemical stability [182, 183]. Ilconich et al.
17 [184] studied the stability of a SILM with 1-n-hexyl-3-methylimidazolium
18 bis(trifluoromethanesulfonyl)imide ([Hmim][NTf₂]), and a PS membrane used as a support.
19 They found that membrane was stable up to 398 K. Alkhouzaam et al. [185] synthesized dense
20 SILMs using PS as a support membrane. They investigated the applicability of these SILMs in
21 industrial gas separation processes under high pressure. Respectively, the membrane showed
22 only 30% and 20% IL loss with 5 wt% tributylmethylphosphonium formate ([P₄₄₄][formate])
23 and tributylmethylammonium formate ([N₄₄₄][formate]) at 10 bar after 12 h. Moreover, there
24 were even not loss of ILs for di-iso-propyl 1-alkyl-3-methylimidazolium bistriflamide ([DIP-
25 C₄mim][NTf₂]) and 1-alkyl-3-methylimidazolium bistrifamide ([C₄mim][NTf₂]) at the same
26 operating conditions.

27 In addition, the SILMs can tolerate higher transmembrane pressure by choosing a support
28 with suitable pore sizes, because ILs extrusion from the large pores of membrane could be
29 mainly contribute to IL loss of SILMs [186]. Some studies are suggested that supported
30 membranes with pore size range from 100 to 200 nm are suitable for composed SILMs [168,

1 186]. Fatyeyeva et al. [187] observed that the presence of ILs influences strongly the
2 permeability of the membrane by reducing the pore sizes. Besides, it should be noted that the
3 high porosity of supports provides more active surface area, which results in a stronger diffusion
4 during the support membrane. However, the mechanical strength of membrane will be
5 decreased with increasing porosity.

6 Also, different preparation methods of SILMs affect stability. Preparation methods of
7 SILMs fall into three main categories: immersion, vacuum, and pressure. For the immersion, a
8 porous support or substrate is soaked in an IL bath, and then the IL is filled into the pores of the
9 support by capillary forces [168]. This method is the easiest and available way to prepare a
10 SILM. However, the SILMs prepared by this way have less stable. It means the IL within the
11 pores of the supported membrane will be pushed out because of the transmembrane pressure.
12 Fortunato et al. [188] found that immersion method was not suitable for high viscosity of ILs
13 because capillary forces decrease with increasing viscosity of ILs. For the vacuum, IL was
14 spread out the membrane surface by vacuum. Similarly, when the viscosity of ILs increases, the
15 loss of ILs was increased for SILMs prepared by vacuum [189]. This phenomenon can be
16 explained by the fact that ILs are on the external surface of the supported membrane by vacuum
17 method. During operation, it is easy for ILs to break away from the support. For the pressure,
18 IL was forced by certain pressure to get into the pores of supports. This method is suitable for
19 high viscosity of ILs due to independence of ILs. Furthermore, SILMs by pressure method was
20 verified by Hernandez-Fernandez et al. [189]. Recently, the new method cold plasma was used
21 to treat SILMs, they found that the loss of IL by plasma treated was lower than untreated
22 membrane due to the higher affinity of the support and IL [190].

23 3.1.1 Separation of VOCs by SILMs

24 This process doesn't need high energy consumption due to moderate operation conditions
25 requiring. One the most important application of the SILMs is separate organic compounds
26 selectively. From the study of Matsumoto et al. [1, 192], the selective separation of benzene,
27 toluene, and p-xylene from n-heptane was analyzed using SILMs based on [Bmim][PF₆], 1-
28 hexyl-3-methylimidazolium hexafluorophosphate ([Hmim][PF₆]), and 1-methyl-3-

1 octylimidazolium hexafluorophosphate ([Omim][PF₆]) supported in a PVDF membrane. It was
2 found that aromatic hydrocarbons were successfully transported through the membrane based
3 on these ionic liquids, the maximum selectivity to n-heptane being reached using benzene in
4 the aromatic permeation and [Bmim][PF₆] in the liquid membrane phase. The feasibility of the
5 prepared SILMs for the separation of water mixed with ethanol or cyclohexane were
6 investigated by Abdellatif Dahi et al. [191]. SILMs were prepared by immobilizing the room
7 temperature ionic liquids (RTILs), 1-butyl-3-methylimidazolium tetrafluoroborate
8 ([C₄c₁im][BF₄]) and 1-butyl-3-methylimidazolium hexafluorophosphate (n = 4) and 1-hexyl-3-
9 methylimidazolium hexafluorophosphate (n = 6) ([C_nc₁im][PF₆] (n = 4 or 6)), in a porous
10 matrimid membrane. For the first study of separating water and VOC vapor, only [C₄c₁im][PF₆]
11 showed the good sorption. In addition, the SILMs showed satisfactory as well as durability.
12 Tadashi Uragami et al. [193] focused on the addition of hydrophobic ionic liquid 1-allyl-3-
13 butylimidazolium bis(trifluoromethylsulfonyl)imide ([Abim][NTf₂]) to poly(methyl
14 methacrylate)-graft-poly(dimethylsioxane) (PMMA-graft-PDMS) membranes for the removal
15 of dilute benzene from an aqueous solution. They found the membrane showed high benzene
16 perm-selectivity and permeability, when the aqueous solution of 0.05 wt% benzene was
17 permeated. Cichowska-Kopczyrska et al. [176] investigated SILMs for toluene separation from
18 gas phase. The selectivity of toluene/N₂ can reach 306, which is significantly high. They also
19 performed that permeation of toluene increase with increasing alkyl chain length of
20 bis(trifluoromethylsulfonyl)imide ([TNf₂]). From Cichowska-Kopczyrska study [195], the
21 selectivity of SILMs was increased with increasing the alkyl chain length. Furthermore,
22 selectivity of SILMs with a given anion of ILs increased with decreasing the alkyl chain length
23 on the imidazolium ring [196, 197]. The separation performance of SILMs can be predicted by
24 the relationship between permeability and partition coefficients [198]. Selectivity of SILMs is
25 dependent on the alkyl chain length of ILs.

26 **3.1.2 Separation of CO₂ by SILMs**

27 CO₂ separation processes have become urgent focuses due to global warming and climate
28 change. Recently, the most publications specifically researched on separating CO₂ from gas

1 pairs such as CO₂/N₂, CO₂/CH₄, CO₂/H₂ [199, 200], CO₂/SO₂ [201, 202] and CO₂/He [184].
2 Because the selectivity and permeability are very important, Table 8 and Table 9 show the
3 different SILMs performance on separation of CO₂/N₂ and CO₂/CH₄.

4 For the separation of CO₂ from CO₂/N₂ gas pair. Lee et al. [203] added CrO₃ particles into
5 a SILMs composited by [Bmim][BF₄] and polyethylene oxide (PEO) support. They found that
6 the selectivity and the permeance of CO₂ from CO₂/N₂ were enhanced. The maximum
7 permeance and selectivity of composite membrane were 144 GPU and 30. Schott et al. [204]
8 researched the CO₂ solubility in several larger cyclic skeleton ILs. They suggested that amino
9 functionality of basic ILs compounds are highly affected on CO₂ solubility. The maximum
10 selectivity CO₂ can reach 53 by [Emim][B(CN)₄] and highest solubility is 0.13 mol L⁻¹ atm⁻¹ at
11 298 K by [EtDBN][TNf₂]. The larger CO₂ solubility was attributed to electrostatic interactions
12 or binding energy decreasing between the cation and anion in ILs. Grunaure et al. [205]
13 compared the selectivity and permeability of CO₂ from CO₂/N₂ on SILMs with three different
14 ILs including 1-ethyl-3-methylimidazolium dicyanoamide ([Emim][DCA]), 1-ethyl-3-
15 methylimidazolium bis(trifluoromethylsulfonyl)imide ([Emim][NTf₂]), and [Bmim][PF₆]. The
16 results showed that [Emim][DCA] performed best in selectivity of CO₂ reach 65, and the
17 highest permeability was from [Emim][NTf₂]. For the high selectivity, CO₂ showed high
18 affinity to [Emim][DCA] due to highly delocalized anions of IL [206], and low N₂ bonding
19 [207]. For the high permeability, it is mainly because of the flexible structure and CF₃ groups
20 of TNf₂, which permit fast throughput [208]. Yahia et al. [209] described a new method for
21 SILMs to enhance the selective CO₂ at high temperature. They presented good permeability
22 (733 barrer) at high temperature (373 K), and transport selectivity of CO₂ was 35.6 from CO₂/N₂.
23 Fan et al. [210] design a SILMs with (2-hydroxyethyl)-trimethyl-ammonium(S)-2-
24 pyrrolidinecarboxylic ([Choline][Pro]) and polyethylene glycol 200 (PEG 200). When the
25 weight ration of [Choline][Pro] to PEG 200 was 1:2, the permeability of CO₂ from CO₂/N₂
26 reached to 1798.6 barrer at 343.15 K, while the selectivity of CO₂ was decreased to 7.9 under
27 same operating conditions.

28

29

1

Table 8. Gas permeability and selectivity for CO₂ and N₂ through SILMs.

| ILs | Supports | Temperature K | Pressure bar | Permeability (barrers) | | Selectivity | Ref. |
|---|-------------------|------------------|-----------------|------------------------|----------------|-------------|-------|
| | | | | CO ₂ | N ₂ | | |
| [MeDBU][B(CN) ₄] | Polyether sulfone | 298 | 1 | 834 | 17 | 49 | [204] |
| [EtDBU][B(CN) ₄] | | | | 672 | 14 | 48 | |
| [EtDBU][NTf ₂] | | | | 432 | 16 | 26 | |
| [Emim][DCA] | PS-b-P4VP | 303 | - | 480 | 7.4 | 65 | [205] |
| [Emim][NTf ₂] | | | | 120 | 10 | 12 | |
| [Bmim][Ac] | | | | 100 | 6.7 | 15 | |
| [C ₄ mim][NTf ₂] | PVDF | 373 | 0.7 | 733.7 | 20.6 | 35.6 | [209] |
| [AEmim][NTf ₂] | Ceramic | 303 | 0.78 | - | - | 13 | [211] |
| [AEmim][BF ₄] | | | | - | - | 2 | |
| [Toma][An] | | | | - | - | 70 | |
| [Emim][Ac] | Ceramic | 303 | 2 | - | - | 34.7 | [212] |
| [Bmim][NTf ₂] | Ceramic | 303 | 2 | - | - | 127 | [213] |
| [C ₈ F ₁₃ mim][NTf ₂] | | | | - | - | 72 | |
| [Emim][NTf ₂] | Ceramic | 303 | 2 | - | - | 21 | [186] |
| [BzPy][NTf ₂] | PTEF | 298 | 0.35 | 518 | 18.4 | 27.9 | [214] |
| [Bz ₂ Py][NTf ₂] | | | | 358 | 10.8 | 33.1 | |
| [Bz ₃ Py][NTf ₂] | | | | 446 | 20.3 | 22 | |
| [Bz ₄ Py][NTf ₂] | | | | 496 | 21.9 | 22.6 | |
| [Choline][Pro] 50 wt% | PEG200 | 308 | 1.8 | 343.3 | 9.9 | 34.8 | [210] |
| [DMAPH][EoAc] | PES | 313 | - | 3028 | 20 | 151.4 | [215] |
| [DMAPH][TFA] | | | | 3352 | 26 | 129 | |

2

3 For the separation of CO₂ from CO₂/CH₄ gas pair. Bara et al. [194] synthesized three
4 molten salts with increasing lengths of fluoroalkyl substituents for SILMs and tested their gas
5 separation properties. The results exhibited that ideal selectivity for CO₂/CH₄ separating was
6 higher than CO₂/N₂. Zhang et al. [216] studied 1-butyl-3-methylimidazolium acetate
7 ([Bmim][Ac]) supported by PVDF selected separation of CO₂. The results showed high
8 permeability CO₂ (2148 barrer) and perm-selectivity of CO₂/CH₄ (40) under 1 bar and 313 K.
9 In addition, Zhang's group increased the CO₂ selective separation furtherly [215]. In this
10 membrane, they used PES supported [DMAPH][EoAc] to separate CO₂ from CO₂/CH₄. The
11 highest permeability was 3028 barrer and perm-selective reached to 72 from CO₂/CH₄ and 151
12 from CO₂/N₂. Liu et al. [217] used RTIL [Tespim][BF₄] modified a membrane tested the CO₂

1 permeances and selectivity of original and modified. They found that the selectivity of CO₂ of
 2 the modified membrane was enhanced. Moreover, ideal CO₂ selectivity of original membrane
 3 was 17, and increased to 76 for modified membrane with the same operating conditions. While,
 4 the CO₂ permeance of modified membrane decreased by 44%. One important phenomenon
 5 observed in several researches was a trade-off process. It means the selectivity decreases, when
 6 having high permeability [218].

7 **Table 9.** Gas permeability and selectivity for CO₂ and CH₄ through SILMs.

| ILs | Supports | T K | P bar | Permeability (barrer) | | Selectivity | Ref. |
|--|----------|--------|----------|-----------------------|-----------------|-------------|-------|
| | | | | CO ₂ | CH ₄ | | |
| [Bmim][PF ₆] | PVDF | 313 | 0.1 | 512 | 63.9 | 8 | [216] |
| [Bmim][BF ₄] | | | | 1104 | 111 | 9.9 | |
| [Bmim][NTf ₂] | | | | 1233 | 131 | 9.4 | |
| [Bmim][OTf] | | | | 1078 | 84.8 | 12.7 | |
| [Bmim][Ac] | | | 1 | 2148 | 53.6 | 40 | |
| CPTES | zeolite | 298 | 2 | - | - | 26 | [217] |
| [Tespimim][BF ₄] | | | | - | - | 87 | |
| [APTMS][Ac] | P-84 | 298 | 10 | - | - | 38.1 | [218] |
| [DMAPH][TFA] | PES | 303 | | 3352 | 50 | 67 | [215] |
| [DMAPH][EoAc] | | | | 3028 | 42 | 72 | |
| [P ₆₆₆₁₄][Cl] | PES | 298 | 0.2 | 350 | 89 | 3.9 | [219] |
| [C ₂ mim][NTf ₂] _{0.75} [Ac] _{0.25} | PVDF | 293 | 1 | 503 | 16 | 31.4 | [219] |
| [N ₂₂₂₄] ₂ [maleate] | PES | 313 | 0.1 | 2840 | 13 | 218.5 | [201] |
| [C ₃ NH ₂ mim][OTf] | PTFE | 298 | 0.025 | 2600 | 21 | 123.8 | [181] |
| [P ₄₄₄₁][formate] _{5wt%} | PSF | 295 | 10 | 11.5 | 0.4 | 32 | [185] |
| [P ₄₄₄₁][formate] _{25wt%} | | | | 17.3 | 0.48 | 31 | |
| [DIPc ₄ mim][NTf ₂] _{0.5wt%} | | | | 12.2 | 0.19 | 61 | |
| [DIPc ₄ mim][NTf ₂] _{2.5wt%} | | | | 13.8 | 0.22 | 63 | |
| [C ₄ mim][NTf ₂] _{0.5wt%} | | | | 10.9 | 0.19 | 57 | |
| [C ₄ mim][NTf ₂] _{2.5wt%} | | | | 11.5 | 0.16 | 70 | |
| [N ₄₄₄₁][formate] _{0.5wt%} | | | | 12.5 | 0.26 | 47 | |
| [N ₄₄₄₁][formate] _{2.5wt%} | | | | 10.2 | 0.22 | 46 | |
| Pr[mim] ₂ [NTf ₂] | alumina | 300 | 0.3 | 190 | 7 | 27.1 | [219] |
| h[mim] ₂ [NTf ₂] | | | | 230 | 14 | 16.4 | |
| [Hmim][NTf ₂] | | | | 1101 | 31.5 | 31.5 | |

8
 9 Generally, the selectivity and permeability of CO₂ through SILMs are mainly influenced by
 10 properties and structures of ionic liquids, gas pair, supports, and operating conditions. The
 11 effects from structures and properties of ILs are studied. Usually, modified or synthesized ILs

1 can increase the CO₂ solubility based on synergistic effect between CO₂ and the IL [217]. For
2 example, adding inorganic particles into ILs [203, 212] or using different types of precursors to
3 synthesize ILs [219]. For example, CO₂ have high affinity to delocalized anions [220] and -CN
4 group [205]. ILs containing acetate [205] and amine [213, 221] functional groups can increase
5 the CO₂ solubility or promote reactions to capture CO₂. This method is also limited by the gas
6 pair. The viscosity and molar volume of ILs are important parameters for selectivity and
7 permeability of CO₂. The higher viscosity leads to a lower diffusivity of CO₂ and permeability
8 [209]. Camper et al. [208] and Shannon et al. [214] showed the solubility and selectivity of gas
9 affected by molar volume, and they also gave a method to estimate gas permeability and
10 selectivity for ideal CO₂/N₂ and CO₂/CH₄. Ilyas [218] suggested that their SILMs which showed
11 high selectivity for CO₂/CH₄ can be attributed to the high solubility of CO₂ in the selected IL
12 as well as its molar volume. The selectivity and permeability of CO₂ not only depend on the
13 selected IL but also on the porous support including effective pore size and surface porosity.
14 The ideal selectivity was found to be independent of supported membrane pore size [186].
15 However, it is opposite for permeance. Close et al. [186] found the permeance value of
16 supported membrane with larger pore size was higher than small pore size. It is value to note
17 for polymeric supported membrane, the effective pore size diameter was reduced due to in the
18 presence of water [221], resulting a change of selectivity and permeability. It can be stated that
19 the higher porosity results directly in higher permeability of CO₂ by comparing the membranes
20 with different porous structures. Finally, the operating conditions such as temperature and
21 pressure are also significant. The selectivity was decreased with increasing temperature [217,
22 218]. In addition, when the pressure increase, selectivity was decreased [185].

23 **3.2 IL-Polymer membranes (ILPMs)**

24 ILs composite polymer membranes provide a successful method to solve the instability of
25 SILMs [225]. The IL was stabilized into a polymeric membrane by entrapped it in the space of
26 polymer chains or clusters [226, 227], which provides a mechanically stable membrane as
27 shown in Figure 4.

28

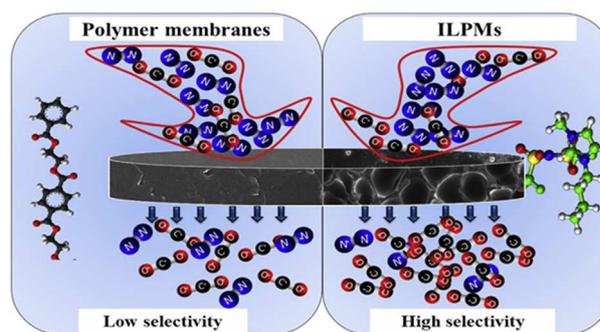


Figure 5. Schematic of polymer membranes and ILPMs [259].

Although, polymeric membranes exhibited excellent mechanical stability, which can overcome the main drawback of SILMs, it should be noted that PILMs already show a certain trade-off between permeability and selectivity for gas separation [218]. Therefore, several researches were donated to find a solution to reduce the trade-off and to develop a desirable membrane.

According to previous studies, the IL loading content and polymeric membranes affect the stability of ILPMs [228, 229]. Mass transfer in ILs is much faster than in a solid polymeric matrix, resulting higher gas flux and faster separation for ILPMs with higher IL loading [227]. For example, CO₂ permeability increased from 400 to 1800 barrer when the 1-ethyl-3-methylimidazolium tetracyanoborate ([C₂mim][TCB]) loading from 0 to 75 vol% to a PVDF matrix [230]. In addition, they observed that polymer matrix be more amorphous with the increase of ILs content which supports the enhance the permeability of gas species through polymers. Furthermore, the stability of polymeric matrix directly determines the operating pressure and temperature of the ILPMs. For example, Mannan et al. [231] synthesized the [Emim][NTf₂] (50 %) and PES composite membrane. This ILPM displayed great stability at a trans-membrane pressure up to 25 bar. It not observed plasticization phenomena on their membrane even under such high operating pressure. Kim et al. [179] found that the ILPMs membrane composited by 40 % of 1-ethyl-3-methylimidazolium tetrafluoroborate ([C₂mim][BF₄]) and PVDF-HFP showed high temperature resistance up to 673 K. In conclusion, the polymer host matrix was selected based on high mechanical strength, high thermal and chemical stability, and the IL was selected in ILPM mainly based on its high affinity of CO₂ and less viscosity [232].

3.2.1 Separation of VOCs by ILPMs

PILMs have caused wide public concern over the recent years for VOCs separating. Tadashi Uragami et al. [233] studied the removal of chloroform, benzene and toluene from aqueous solutions of dilute VOCs by poly(styrene)-b-poly(dimethylsiloxane) (PSt-b-PDMS) membranes containing [Abim][NTf₂] (loading content from 0 to 30 wt%). The results showed that strong VOC/water selectivity when aqueous solutions of 0.05 wt% VOCs. [Abim][NTf₂] localized in the PSt-b-PDMS membrane acts as an absorbent to selectivity partition VOCs from the aqueous solution. Both the permeability and the VOCs perm-selectivity of ILPMs were promoted due to the affinity of IL and VOCs. Furthermore, they used the same IL with another polymeric matrix composited a new ILPM. The poly(methyl metacrelate)-g-poly(dimethylsiloxane) (PSMMA-g-PDMS) membranes containing [Abim][NTf₂] (loading content including 10 wt%, 15 wt%, and 20 wt%) showed high benzene selectivity from an aqueous solution. The permeability and the benzene selectivity were enhanced with increasing the IL content [193]. When the IL content increased, the solubility of VOCs in membrane was increased because of the preferential absorption of VOCs in membrane and the high diffusivity of VOCs in the polymer matrix with IL. The imidazolium-based ILs (1-n-althyl-3-methylimidazolium tetrafluorobrate, n=4, 6 - 8, 12) and PVDF membranes were used to characterize ethanol and cyclohexane vapor sorption performance [187]. It was revealed increasing the sorption capacity with increasing alkyl chain length because of the enhancement of ability of interaction via van der waals interactions [234 - 236]. Jebur et al. [237] composited imidazolium based ionic liquids and hydrophobic PTFE based membrane to separate aromatic compounds from non-aqueous medium. They found the order of separation efficiency was divinylbenzene > styrene > toluene due to affinities with the IL. Moreover, the stability of this membrane was evaluated up to 60 days. More other applications of PILMs to remove VOCs are shown in Table 10. Briefly, the performance of ILPMs in the separation processes of VOCs is mainly dependents on two factors [238, 239]: i) polymeric membrane parameters, including thickness, pore size, and hydrophilicity; ii) properties and content of ILs.

1
2

Table 10. Summary of the applications of ILPMs on VOCs removal.

| ILs | Polymers | VOCs | Conditions | Major inclusions | Ref |
|---|--|----------------------------------|--|--|-------|
| [Bmim][PF ₆] [Hmim][PF ₆] [Omim][PF ₆] [Et ₂ MeMeON][NTf ₂] | polyvinylidene e (PVDF) | benzene toluene p-xylene | - effective area: 20 cm ² - pore size of PVDF film: 45 um - feed solutions: mixed of heptane and aromatic hydrocarbon - receiving solution: hexadecane - room temperature | The permeation rate of hydrocarbon -based on ILs: heptane>toluene>p-xylene>benzene -based on water: Toluene ≤ heptane < benzene < p-xylene | [240] |
| [C ₄ mim][PF ₆] [C ₆ mim][PF ₆] [C ₈ mim][PF ₆] | elcosene copolymer (VP) | benzene cyclohexane | -effective area: 38.4 cm ² -feed solutions: benzene cyclohexane -sweep stream: 50 mL min ⁻¹ -temperature: 303 K | -benzene selectively permeated the membrane -permeability was mainly controlled in the membrane by diffusivity -the separation factor increased as the hydrophilicity of ILs increased | [241] |
| [Abim][TFSI] | poly(methyl methacrylate) -graft- poly(dimethyl siloxane) (PMMA-g-PDMS) | benzene | -feed solution: 0.05 wt% and water -temperature: 313 K | -PMMA-g-PDMS increased benzene-permselectivity with increasing DMS content -introducing molecular recognition into membrane matrix can improve permeability for removing benzene | [193] |
| [Abim][TFSI] | poly(styrene)- b- poly(dimethyl siloxane) | benzene chloroform toluene | -feed solution: 0.05 wt% VOCs mix water -pressure: 1.33 Pa -Temperature: 313 K -effective area: 13.8 cm ² | -the permeability of VOC and perm-selectivity were enhanced on increasing the ILs content -introduction of ILs to membrane increased the solubility of VOCs into the membrane | [233] |

| | | | | | |
|---|---|-----------------------|---|---|-------|
| [Abim][TFSI] | poly(vinyl chloride) (PVC) | benzene | -feed solution: 0.05 wt% mix aqueous solution | -the addition of IL into PVC membrane enhanced both the permeability and benzene/water selectivity -the sorption selectivity increased by adding IL into PVC membrane | [242] |
| [C ₂ mim][Tf ₂ N] | poly(vinylidene fluoride-co-hexafluoropropylene) P(PDF-HFP) | n-hexane isooctane | -sweep gas: N ₂ -pressure: 100 kPa -sweep gas flow: 10 ml min ⁻¹ -temperature: higher than that saturators | -the vapour sorption and transport properties were enhanced when membrane containing 80 mass% of IL - with the temperature range of 298-318 K, the permeability of hexane and of isooctane were found to be independent of both temperature and activity of the permeant | [243] |

1

2 3.2.2 Separation of CO₂ by ILPMs

3 Blending a task-specific IL with proper polymer membrane could provide remarkable
4 selectivity for CO₂. In this case, choosing a proper polymer membrane is very important. For
5 gas separation, membranes prepared from glassy polymers have been widely studied, including
6 PIM-1 [258], cellulose acetate (AC) [244, 245], polysulfone (PSF) [183, 246], Nafion [253]
7 and polyimides (PI) [247, 248]. However, these polymer membranes show low gas permeability
8 and high gas selectivity even at high operating pressure due to low free volume [249]. In
9 addition, membranes prepared from rubbery polymers, such as poly(ethylene oxide) (PEO)
10 [250], poly(amide-6-b-ethylene oxide) (Pebax) [251], poly(dimethyl siloxane) (PDMS) [252],
11 and Nylon [254], show excellent permeability because of the strong chemical affinity between
12 polymer chains and gas molecules [249]. Moreover, the task-specific ILs combined to polymer
13 membranes provide a remarkable selectivity and permeability for CO₂ separation, which
14 contain some specific anion or cation with promoted CO₂ affinity [255, 256]. In the ILPMs, the
15 polymer membranes usually provide the mechanical strength, whereas ILs play a significant
16 role in CO₂ separation.

1 For CO₂ separation, ILs of ILPMs show more important for permeability and selectivity.
 2 First, the low viscosity and high CO₂ permeability of IL are vital. They are attributed to CO₂
 3 solubility, and play significant roles in CO₂ separation [230]. Second, higher IL contents results
 4 in a higher gas fluxes and faster separation for ILPMs. That because the mass transfer in IL is
 5 faster than in polymer matrixes. For example, Mannan et al. [231] suggested that incorporation
 6 of higher IL up to 50 % into polymer matrix not only enhanced the permeability of the ILPMs
 7 but also improved the separation factor of the synthesized membranes. Chen et al. [230] found
 8 that permeability of CO₂ increased rapidly with increasing IL content, which is attributed to the
 9 increments of CO₂ diffusivity and solubility. They prepared [Emim][B(CN)₄]/PVDF blend
 10 membranes showed a high CO₂ permeability of 1778 barrer with CO₂/H₂ and CO₂/N₂ selectivity
 11 of 12.9 and 41.1. However, the mechanical properties of composited membrane including
 12 Young's modulus, tensile strength, and extension at break, were decreased with increasing IL
 13 content. Third, it is necessary to check the operating pressure for ILPMs due to the plasticization
 14 phenomena [249, 257]. Plasticization is a pressure-dependent phenomenon, which is showed
 15 by rise in permeability and fall selectivity suddenly after a threshold pressure [249]. Mannan
 16 group found for glassy polymer (PES) membrane that the permeability decreased with
 17 increasing pressure. When the IL content was 50 wt%, the permeability of CO₂ was 355.76
 18 barrer at 5 bar and 298.84 barrer at 25 bar. There was no plasticization occurred in
 19 [Emim][NTf₂]/PES membranes [249]. More other applications of PILMs to remove CO₂ are
 20 shown in Table 11.

21 **Table 11.** Summary of removing CO₂ by ILPMs.

| Polymers | ILs | Permeability (barrer) | | | Selectivity | | Conditions | Ref. |
|----------|---|-----------------------|----------------|-----------------|---------------------------------|----------------------------------|----------------|-------|
| | | CO ₂ | N ₂ | CH ₄ | CO ₂ /N ₂ | CO ₂ /NH ₄ | | |
| PIM-1 | [C ₂ mim][NTf ₂] 5 wt% | 6650 | 332 | 593 | 20 | 11 | 303 K - | [258] |
| | [C ₄ mim][NTf ₂] 5 wt% | 4590 | 212 | 349 | 22 | 13 | | |
| | [C ₆ mim][NTf ₂] 5 wt% | 2240 | 90 | 165 | 25 | 14 | | |
| PSF | [Bmim][TFSI] 50 wt% | 3000 | 107 | - | 28 | - | 308 K 7 bar | [183] |
| | [Bdim][TFSI] 50 wt% | 3600 | 129.9 | - | 27.7 | - | | |
| | [Dems][TFSI] 50 wt% | 5800 | 193 | - | 30 | - | | |
| PSF | [P ₄₄₄₁][formate] 12.5 wt% | 17.3 | - | 0.48 | - | 31 | 295 K | [259] |
| | [DIP-C ₄ mim][NTf ₂] 2.5 wt% | 13.8 | - | 0.22 | - | 63 | 10 bar | |

| | | | | | | | | |
|------|---|------|------|------|------|------|-------------------|-------|
| | [C ₄ mim][NTf ₂] 2.5 wt% | 11.5 | - | 0.16 | - | 70 | | |
| | [N ₄₄₄₁][formate] 0.5 wt% | 12.5 | - | 0.26 | - | 47 | | |
| PVDF | [Bmim][BF ₄] | 93.9 | 5.04 | 20.3 | 52.3 | 8.18 | 303 K 3 bar | [260] |
| | AMMOEN™100 | 93.9 | 1.79 | 5.76 | 52.6 | 7.93 | | |
| | ECOENG™1111P | 127 | 11.6 | 15.6 | 10.9 | 6.38 | | |
| | Cyphos 102 | 637 | 15.3 | 76.5 | 41.5 | 6.87 | | |
| | Cyphos 103 | 487 | 11.3 | 65.1 | 43.1 | 5.62 | | |
| | Cyphos 104 | 642 | 20.3 | 11.3 | 31.6 | 5.17 | | |
| | [Emim][OTf] | 486 | 14.3 | 21.1 | 34.0 | 13.1 | | |
| | [Set ₃][NTF ₂] | 747 | 18.4 | 81.2 | 26.2 | 6.67 | | |
| PVDF | [C ₄ mim][PF ₆] | - | - | - | 20 | 200 | 303 K 1.7 bar | [261] |
| | [C ₄ mim][BF ₄] | - | - | - | 32 | 102 | | |
| | [C ₄ mim][NTf ₂] | - | - | - | 30 | 161 | | |
| | [C ₈ mim][PF ₆] | - | - | - | 21 | 98 | | |
| PES | [MpFHim][NTf ₂] | 320 | 12 | 17 | 27 | 19 | 296 K 1.85 bar | [262] |
| | [MnFHim][NTf ₂] | 280 | 14 | 17 | 21 | 17 | | |
| | [MtdFHim][NTf ₂] | 210 | 13 | 16 | 16 | 13 | | |
| PI | [Bmim][NTf ₂] | 34.4 | 1.34 | 1.43 | 25.7 | 24.1 | 308 K - | [248] |

1

3.3 Ionic liquids composited mixed matrix membranes (ILMMMs)

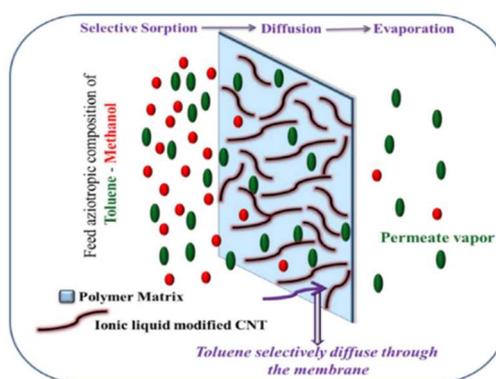
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Figure 6. Schematic of ILMMMs selective separation of toluene [269].

9

10 Mixed matrix membranes (MMMs) are comprised of inorganic fillers and polymers. The separation performance of MMMs depends on the selective sorption/diffusion of the fillers, and

1 the mechanical strength relies on polymer matrices [263]. According previous studies,
2 polymeric membranes are subjected to the trade-off effect between permeability and selectivity
3 [218], and poor processability [264]. MMMs combine outstanding gas separation performance
4 of inorganic materials and mechanical strength of polymer membranes. However, there are still
5 some challenges of MMMs should be solved. For example, selecting an appropriate filler and
6 polymer [263], the filler dispersion [265], and the compatibility of filler and polymer [266].
7 Combining the ILs with inorganic fillers in MMMs (Figure 6) will enhance the membrane
8 separation performance due to the synergistic effect [267]. In ILMMMs, ILs are used to increase
9 the absorption capacity, and combine with polymer matrixes to further improve membranes
10 separation performance [268].

11 **3.3.1 Separation of VOCs by ILMMMs**

12 Numerous MMMs combine with ILs have been synthesized using different fillers and
13 polymers to enhance the compatibility between filler and polymer. But for their applications, a
14 few studies are focus on VOCs separation. Abraham et al. [269] presented an IL modified
15 multiwalled carbon nanotubes (MWCNT) based styrene butadiene rubber (SBR) composite
16 membrane, which showed impressive separation performance for toluene selective separation
17 from methanol/toluene mixture. The highest separation factor reached to 128.4, which is 1.6
18 times of SBR membrane. This excellent performance was owing to the closeness of solubility
19 parameter between toluene and SBR and interaction between benzyl groups of IL. Furthermore,
20 they also discussed the influence of loading MWCNT and the effect of IL loading. The
21 separation factor was increased first and then decreased with the MWCNT loading increasing.
22 The effect of IL loading was similar to MWCNT. When the 5 phr f-MWCNT and mixed with
23 IL in ration 1:5 loaded membrane, the separation factor reached to the maximum 128.4.
24 Moreover, the glass temperature was reduced with increasing IL content due to the plasticizing
25 effect.

26 **3.3.2 Separation of CO₂ by ILMMMs**

27 Recently, various types coupling agents have been investigated to improve the compatibility

1 and CO₂ separation performance of ILMMMs. For example, Li et al. [270] suggested a new
 2 method to introduce [Bmim][NTf₂] into zeolite imidazolate framework-8 (ZIF-8) and combine
 3 with poly(ether-block-amide) (Pebax) polymer membrane. The results showed that mechanical
 4 and gas separation properties of ILMMMs were improved simultaneously. On the other hand,
 5 polymeric ILs (PILs) have polymer properties and ILs characteristics [271]. When RTILs were
 6 loaded into PILs matrix, those composite membranes show excellent stability. For example, A
 7 high performance PIL/IL/zeolite MMMs was prepared by Cowan et al. [272]. This membrane
 8 showed ideal CO₂ permeability was 261 barrer and CO₂ selectivity from CO₂/CH₄ was up to
 9 93. This outstanding performance can be explained by the presence of coulombic attraction
 10 between non-polymerizable IL and PIL matrix. Huang et al. [265] facilitated a new MMMs by
 11 incorporating IL functionalized graphene oxide (GO-IL) into Pebax 1657. This membrane
 12 showed high CO₂ permeance up to 9000 GPU and the CO₂/N₂ and CO₂/H₂ selectivities of about
 13 45 and 5.8 due to enhanced CO₂ solubility by IL. More information of separating CO₂ by
 14 ILMMMs are shown in Table 12.

15 **Table 12.** Summary on ILMMMs for CO₂ separation.

| ILMMMs | Permeability (barrer) | | | Selectivity | | Conditions | Ref. |
|--|-----------------------|----------------|-----------------|---------------------------------|----------------------------------|-----------------------------|-------|
| | CO ₂ | N ₂ | CH ₄ | CO ₂ /N ₂ | CO ₂ /CH ₄ | | |
| PSF/[bmim][NTf ₂]/@ZIF-8 | 350.7 | 2.18 | 8.77 | 161.1 | 40 | 303 K, 6 bar | [277] |
| PEBAX1657/[bmim][NTf ₂]/@ZIF-8 | 104.9 | 1.25 | 3.01 | 83.9 | 34.8 | 298 K, 1 bar | [270] |
| [emim][dca]/P([vbim][dca]) | 272.9 | 5.1 | - | 53.5 | - | 308 K, 1-2 bar | [278] |
| [emim][B(CN) ₄]/P([vbim][dca]) | 340.1 | 8.10 | - | 42.0 | - | | |
| [bmim][CF ₃ SO ₃]/Pebax 1657 | 320 | 8 | 11.85 | 40 | 27 | 298 K, 1 bar | [251] |
| [bmim][NTf ₂]/PI | 510 | 18.48 | 35.17 | 27.6 | 14.5 | 308 K, 75 cmHg | [247] |
| [emim][NTf ₂]/SAPO-34/P(vinyl-IL) | 527.2 | 20.28 | 21.16 | 26 | 24.9 | 296 K, 1-1.5 bar | [279] |
| [emim][NTf ₂]/SAPO-34/P([smim][NTf ₂]) | 260 | - | 2.89 | - | 90 | 298 K, 2 bar | [280] |
| [emim][B(CN) ₄]/ZIF-8/P([vbim][NTf ₂]) | 906.4 | 43.17 | - | 21 | - | 308 K, 3.5 bar | [271] |
| | 928.7 | - | 80.06 | - | 11.6 | | |
| [bmim][NTf ₂]/@ZIF-8/PSF | 279 | 2.15 | - | 130 | - | 303 K, 6 bar | [277] |
| | 253 | - | 5.54 | - | 45.7 | | |
| (3-aminopropyl)trimethoxysilano acetate/@GO/Pebax 1657 | 118.6 | 1.67 | - | 71 | - | 298 K, 4 bar | [264] |
| [emim][NTf ₂]/SAPO-34/PES | - | - | - | - | 37.23 | -, 4 bar | [267] |
| [emim][NTf ₂]/SAPO-34/PES | - | - | - | - | 62 | room temperature, 30 bar | [273] |

1 Membrane separation performance is not only affected by filler and polymer, but also by
2 operating conditions. Mohshim et al. [273] observed that CO₂/CH₄ selectivity of PES-SAPO-
3 34 modified [Emim][NTf₂] membrane increased with increasing pressure. The similar results
4 were observed by Huang et al. [265]. In addition, it was noticed that the CO₂/CH₄ ideal
5 selectivity increased as the IL concentration increased. In conclusion, it is very important for
6 development of ILMMMs to select an appropriate material for both sieve phase and matrix [97,
7 274]. Although lots of researchers have been devoted to carry out preparation and modification
8 fillers and polymers in ILMMMs [275, 276], it is necessary to broaden the applications of
9 ILMMMs, because few groups focus on removing VOCs by ILMMMs.

10 **3.4 Poly(ionic liquid)s membranes (PILMs)**

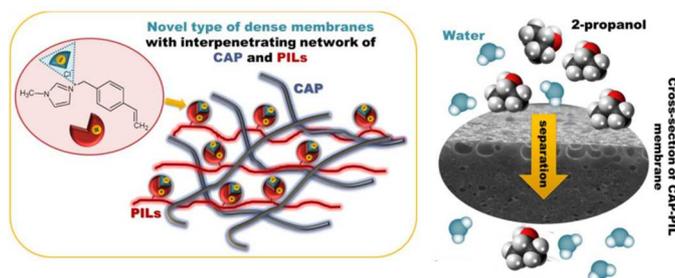
11 PILs were synthesized by polymerizable ILs. Usually, to obtain highly gas permeable and
12 selective, ILs can be directly imbibed into porous membranes. In this case, ILs can be
13 polymerized to form dense polymerized ionic liquid (PIL) membranes to resolve the mechanical
14 stability issue. Poly(ionic liquid)s (PILs) possess many of the unique combination properties of
15 ILs together with intrinsic polymer properties [281 - 284]. When PILs are used for greenhouse
16 gas separation [285, 286], they exhibited excellent sorption capacities and desorption rates than
17 the corresponding ionic monomers. For example, Cardiano et al [285] presented a series of PILs,
18 also the properties of sorption for CO₂ have been researched. The results showed that PIL
19 exhibited significant fast and reversible CO₂ sorption properties at room temperature. It is
20 confirmed that ILs assembling on polymer network represents a useful method to get more
21 processable ILs-materials for greenhouse gas separation. Additionally, Yuan et al. [286]
22 synthesized amine-functionalized PIL brushes which exhibited enhanced CO₂ adsorption
23 capacity. When the polymer brush with polymer loading of 46 wt%, the carbon dioxide
24 adsorption capacity was 2.43 mmol g⁻¹ at 298 K under 0.2 bar (CO₂ partial pressure), much
25 higher than that value of the free polymer (1.28 mmol g⁻¹) under the same conditions.

26 Due to their polymer macrostructure, PILMs not only have enhanced processability and
27 durability, but also improved mechanical stability. To overcome low gas permeability and
28 diffusivity, various strategies have occurred to improve the transport properties of PILMs

1 include loading certain amounts of free room temperature ionic liquids (PIL-IL) [287 - 291],
2 controlling chain packing by synthesizing PIL copolymer membranes [291, 292], and
3 incorporating inorganic material [294, 295]. The synthesis of composite membranes by
4 blending PILs with free non-polymerizable ILs seems to be the most promising strategy to
5 obtain membranes that preserve the mechanical and thermal stability of common polymers
6 which providing high permeation rates and separation factors, which are attractive for industrial
7 purposes. Researchers [290] prepared composite membranes by polymerization of
8 imidazolium-IL monomers in the presence 20 wt% of free IL, which results showed that the
9 CO₂ permeability increased by approximately 40 %. Then, PIL-IL composite membranes draw
10 lots of attentions of researchers, and several PIL and IL combinations are studied [287 - 291].
11 Recently, there is reported that supported thin-film (nano layers) of PIL-IL composites exhibit
12 excellent CO₂ permeation rates together with acceptable CO₂/N₂ selectivities [289], thus
13 indicating that there is still a lot of spaces for improvement in the development of this type of
14 membranes. Phosphonium-based PILs have primarily been synthesized by radical
15 polymerization of IL monomers containing a quaternary phosphonium cation with three
16 attached alkyl chains and a vinylbenzyl polymerizable group, along with an associated free
17 cation. For example, controlled reversible addition-fragmentation chain-transfer
18 polymerizations of [P_nVB][Cl] (where n = the number of carbons in the alkyl chains and VB =
19 the vinylbenzyl polymerizable group on the phosphonium cation) monomers have been used to
20 produce linear PILs and block copolymers containing the phosphonium the phosphonium
21 moiety [292]. Matthew G. Cowan et al. [293] studied the effect of cation alkyl chain length on
22 phosphonium-based PILMs. They found that the gas permeability increased approximately
23 linearly with increasing alkyl chain length on the phosphonium group. In addition, gas
24 selectivity was observed to decrease with an increase in phosphonium alkyl chain length from
25 n = 4 to n = 6, then remain approximately constant between n = 6 and n = 8. The prior works
26 showed that the physical properties of phosphonium-based PILs were influenced on the length
27 of alkyl chain length. Combining PILs with inorganic materials to improve the gas separation
28 and permeability is a promising approach. For example, Yeny C. Hudiono et al. [279] showed
29 that the non-volatile RTIL component enhanced the interaction between the zeolites particles
30 and the poly(RTIL) matrix. Zeolites, RTILs and poly(RTIL)s have been found to be very

1 promising materials for the separation of CO₂ from other gases when formed into membranes.

2 **3.4.1 Separation of VOCs by PILMs**



6 **Figure 7.** Schematic of poly(ionic liquid)s membrane [296].

7 Recently, the application of PILs as membrane materials has become one of the fast-
8 growing fields. Rynkowska et al. [296] prepared a new type of dense membrane (Figure 7)
9 containing interpenetrating network of cellulose acetate propionate (CAP) and PIL. They also
10 found that CAP-PIL membranes are efficient in separation of water and propan-2-ol.
11 Furthermore, the physicochemical and transport properties of CAP-PIL membranes were
12 investigated [297]. With increasing the PIL content, both the thermal stability and mechanical
13 strength were increased. The immobilization of RIL in CAP membranes enhanced the
14 hydrophilic properties of CAP-membrane surface. In addition, it was observed that PIL
15 modified CAP membrane increase the swelling ability, which resulted the swelling of CAP-
16 based membranes in contact with organic such as ethanol and propan-2-ol decreased.

17 PIL-based membranes have been investigated for many applications, such as gas
18 separation [291], and wastewater treatment [298]. The properties and performance of PILMs
19 can be tuned for the solvent properties by the PIL monomer and polymerization conditions
20 [227]. The selectivity of membranes can be changed by swelling properties and surface charge,
21 which depended on polymer monomer, polymerization methods and conditions [228, 299]. Due
22 to the wide variety of PIL monomers, the PIL-based membranes with different properties can
23 be developed to meet different industrial separation requirements.

24 **3.4.2 Separation of CO₂ by PILMs**

25 According to the approaches of improving gas separation, PILMs can be divided into three

1 types such as homo-PIL membranes, PIL-IL membranes, and PIL-inorganic particle
2 membranes. For homo-PIL membranes, Zhang et al. [300] found that the CO₂ permeability
3 gradually increased with increase of PIL content, and the best permeability and perm-selectivity
4 of the membrane reach 36.1 barrer and 59.6. The semi-interpenetrating polymer network
5 membrane was synthesized by incorporating polyvinyl acetate into a cross-linked PIL network,
6 which showed an improved mechanical strength and a depressed glass transition temperature.
7 Cong et al. [301] reported a PILs composited poly (2, 6-dimethyl-1, 4-phenylene oxide)
8 membrane. However, this membrane enhanced CO₂/N₂ selectivity but sacrificed the
9 permeability. Sum up, homo-PILMs usually show small CO₂ permeability [288]. For PIL-IL
10 membranes, ILs are regarded as a plasticizer and enhanced the CO₂ permeability [302]. More
11 importantly, when increase the IL loading, permeability improved without changing the
12 selectivity [255]. Tome et al. [303] compared the gas permeation properties of PIL-IL
13 membranes with different cation pendants, such as imidazolium, pyridinium, pyrrolidinium,
14 ammonium, and cholinium, combined with the same counter-anion ([NTf₂]). When the free
15 IL loading was 10%, CO₂ perm-selectivities of different anions PIL-IL membranes were
16 ranging from 28.3 to 43.0 for CO₂/CH₄ and from 25.8 to 34.7 for CO₂/N₂. The best perm-
17 selectivity were obtained by cholinium and pyrrolidinium better than other anions. In addition,
18 they also tested the PIL-IL membranes with different anions including [N(CN)₂], [C(CN)₃],
19 [B(CN)₄], and [NTf₂]. They found that the CO₂ and N₂ permeabilities were controlled by
20 diffusivity mechanism, while the perm-selectivities of CO₂ and N₂ was a solubility-controlled
21 process. Moreover, The CO₂/N₂ separation performance of PIL C(CN)₃-40 IL N(CN)₂, PIL
22 N(CN)₂-60 IL C(CN)₃, PIL C(CN)₃-60 IL B(CN)₄, and PIL B(CN)₄-60 IL C(CN)₃ were on top
23 of Robeson 2008 upper bound [304]. Some researchers indicated that increasing the content of
24 free IL enhanced the permeabilities of gases. For PIL-inorganic particle membranes, composite
25 PIL-IL membranes containing CuCl have been obtained by Zarca et al [289]. Results showed
26 that enhancement of both gas permeability and ideal selectivity were observed for CO₂/N₂ and
27 H₂/N₂. However, presence of Cu⁺ did not promote facilitated transport of CO₂. And decreased
28 gas diffusion. More information of separating CO₂ by PILMs are shown in Table 13.

29

1
2
3

Table 13. Summary on major results of PILMs for CO₂ separation.

| PILMs | Gas pairs | Conditions | Main conclusions | Ref. |
|---|---|--------------------------|--|-------|
| Styrene-based PILMs Acrylate-based PILMs | CO ₂ /N ₂ CO ₂ /CH ₄ | 293 K | <ul style="list-style-type: none"> - CO₂ permeability was increased in a nonlinear fashion as the n-alkyl substituent was lengthened - CO₂/N₂ separation performance was relatively unaffected as CO₂ permeability increase - CO₂/CH₄ ideal solubility selectivity was highest when alkyl substituent was a methyl group - PILMs can dissolve about twice as much volume of CO₂ per cubic centimeter of material than their liquid analogues at similar temperature | [291] |
| Poly(1-[(4-ethenyl phenyl)methyl]-3-alkylimidazolium) bis-(trifluoromethane)sulfonamide) with PES membranes | CO ₂ /CH ₄ | 283 – 313 K 10-50 bar | <ul style="list-style-type: none"> - PILs at single and mixed gas pressures up to 40 bar and over a temperature range from 283 – 313 K - The permeability of CO₂ increase by more than 60% over a pressure range of 40 bar - Plasticizing effect of the absorbed CO₂ resulting in a strong increase in CH₄ permeability with increasing feed pressure - The permeability of CO₂ is not suppressed by the presence of CH₄ | [302] |
| Poly(diphenylacetylene)s containing imidazolium salts membranes | CO ₂ /N ₂ | 298 K - | <ul style="list-style-type: none"> - High free volume polymers with ionic groups enhanced CO₂ separation selectivity - CO₂ permeability reached 250 barrer, and selectivity of CO₂/CH₄ was ranging from 31 to 44 - Introduction of 1-methylimidazole into the membranes remarkable increased the CO₂/N₂ selectivity, but decreased the CO₂ permeability | [305] |

| | | | | |
|---|---|--------------------|---|--------------|
| <p>DVB-cross-linked poly($[P_{nnnVB}][NTf_2]$) ion gel membrane containing the free ILs $[P_{4448}][NTf_2]$, $[P_{8888}][NTf_2]$, and $[emim][NTf_2]$</p> | <p>CO_2/N_2 CO_2/CH_4 CO_2/H_2</p> | <p>298 K -</p> | <p>- Shorter alkyl chain lengths on the phosphonium cation of the IL resulted in increased thermal stability of the ionic conductivity for the resulting PIL/IL membrane materials with negligible changes in ionic conductivity performance at temperatures up to 383 K - Increasing of free IL loading resulted in an increase in overall membrane mechanical stability and gas permeabilities - Significantly higher gas permeabilities were observed for ion gel membranes containing the $[P_{8888}]$ cation (162 barrer)</p> | <p>[306]</p> |
| <p>Styrene-based IL monomers containing branched- and cycloalkyl groups formed PILMs</p> | <p>CO_2/N_2 CO_2/CH_4</p> | <p>-</p> | <p>- PILs with branched and cyclic functionalities exhibited around 20% larger CO_2/N_2 and CO_2/CH_4 selectivities, but gas permeability decreased more than 50% - PILMs with n-alkyl groups displayed increasing CO_2 permeability with increasing number of carbons in the pendant group</p> | <p>[307]</p> |

1

2 3.5 Ionic liquid-gel membranes (ILGMs)

3 In contrast to chemical polymerization, IL can be gelled physically using low organic
4 gelators or polymers as a simpler fabrication alternative. Typically, an IL-gel membrane is
5 prepared by mixing small amount of a low-molecular weight organic gelator (LMOG) with the
6 chosen IL at elevated temperatures [308, 309]. When the samples were cooled down, a physical
7 gel forms due to hydrogen-bonding or /and van der Waals forces from the gelator and IL to
8 generate a dilute noncovalent network throughout the sample [310]. The IL-gel membranes
9 suffer from a “flux-mechanical stability trade-off”, which means increasing the weight fraction
10 of the gelator that will improve the membrane mechanical stability, however, sacrifice the
11 permeability. LOMG-gelled membranes are difficult to be processed into some shapes like thin
12 film, which inhibits the applications of the membranes. On the contrary, polymers as gelling

1 agents are more attractive as the gelled membranes are capable of being packed into large
2 modules for easy retrofitting. IL-gel materials are booming because they maintain liquid-like
3 gas transport properties in a solid state, which is much easier to handle and to prepare membrane
4 modules for industrial applications.

5 **3.5.1 Separation of VOCs by ILGMs**

6 Although some of the separation performances of ILGMs are promising, the number of
7 gelators known to gel room temperature ILs is limited. Moreover, the thermal stability of the
8 IL-gel membranes is another limitation by the temperature at which the gel reversibly becomes
9 a fluid [251, 311]. Therefore, new gelators could gel IL with higher thermal stability and have
10 the ability gel functionalization room temperature ILs with enhanced VOCs affinity. This kind
11 of gelators are required the further development of the IL-gel membrane platform. Plaza et al.
12 [312] researched the separation of acetone-butanol-ethanol mixtures from aqueous solution by
13 membrane which synthesized by the gelation of an ionic liquid ([Bmim][PF₆]) and
14 polytetrafluoroethylene hollow fibers. Improved butanol/ethanol selectivity was verified and
15 they also found the membrane have potential for separation of butanol from these mixtures.
16 Furthermore, the selectivity of membrane was mainly depended on the partition coefficient
17 between the IL gel and aqueous solution. A gel membrane based on 20 wt% of poly(vinylidene
18 fluoride-co-hexafluoropylene) with 80 wt% of [C₂mim][NTf₂] for ethanol removing from
19 polluted air was reported by O. Vopicka et al. [308]. In this system, ethanol permeability reached
20 around 25000 barrer in the membrane at the highest ethanol vapor activity. Moreover, the
21 ethanol vapor sorption in membrane increased with temperature.

22 **3.5.2 Separation of CO₂ by ILGMs**

23 The main separation performances of ILGMs focus on biogases separating. Fam et al. [229]
24 reported a defect-free Pebax1675/[Emim][BF₄] (1-ethyl-3-methylimidazolium tetrafluoroborate)
25 gel membranes with high mechanical strength and improved CO₂ separation performance. The
26 stability of high IL-loaded gel membrane with up to 8 bar feed pressure also has possibility to
27 meet industrial gas separation required. When the IL loading 80 wt%, the CO₂ permeability was

1 increased 300% and ideal CO₂/CH₄ and CO₂/N₂ selectivities were 36 and 15. Friess et al. [314]
2 synthesized epoxy-amine-based ion-gel membranes showing remarkable CO₂/CH₄ separation
3 performance even in presence of humidity. Introduction of humidity in the mixed-gas feeds
4 leads to higher CO₂/CH₄ separation factor (up to 35) compared with dry mixed-gas feeds. In
5 addition, an increase of free IL [Emim][NTf₂] content from 50 to 75 wt% leads to higher CO₂
6 permeability from 195 to 525 barrer with slight loss of CO₂/CH₄ ideal selectivity from 20 to 18.
7 Moghadam et al. [315] also studied ILGMs for CO₂ separation under humid conditions. At
8 room temperature, the membrane exhibited good stability at high relative humidity (RH). More
9 importantly, at 303 K, RH 70%, and a CO₂ partial pressure of 0.1 kPa, the membrane showed
10 high CO₂ permeability (around 50000 barrer) and CO₂/N₂ selectivity (up to 8100). Mahdavi et
11 al. [310] introduced inorganic particles into ILGMs to promote gas permeabilities and ideal
12 selectivities. The results showed that incorporation of 8 wt% SiO₂ increased CO₂ and CH₄
13 permeabilities values from 104.3 to 153.6 barrer and from 5.6 to 8.1 barrer. The ideal
14 selectivities of CO₂/CH₄ increased from 18.5 to 19.1 at 2 bar 298 K. However, Couto et al. [313]
15 developed a new gel membrane, which showed much lower permeability and ideal selectivity
16 for CO₂, comparing to other gel membranes.

17 Above all, although some of, ILGMs show high mechanical strength and gas permeability,
18 the gelators to gel ILs are limited. In addition, the thermal stability of ILGMs is hindered its
19 applications. On the other word, the gelled ILs will change the morphology with temperature
20 increasing. Therefore, development of new gelators with high thermal stability is necessary.
21 And it is also significant to find new method to gel functionalized ILs with higher affinity of
22 target gas, and exploitation of new applications of ILGMs.

23 **3.6 Ionic liquid membrane contactors (ILMCs)**

24 Membrane absorption [54, 323 - 325] is a hybrid process that integrates the advantages of
25 membrane separation and absorption, where liquid absorbents provide the selectivity and the
26 porous/unselective membrane acts as the contacting interface between liquid and gas phases.
27 There are lots of advantages of membrane contactor [326 - 329], such as determined interfacial
28 area, independent control of gas and liquid flow rates, and avoidance to drop dragging. For most

1 of the cases, membrane contactor is applied for gas capture at relatively low pressure and
2 temperature [330 - 332]. Various aqueous absorbents have been studied in membrane contactors,
3 including aqueous amine solutions [335, 343], amino salt solutions [335 - 337], enzyme
4 solutions [338, 339], and ammonia solutions [340 - 342]. Based on liquid flow direction, there
5 are co-current, cross flow, and counter-current configurations of membrane contactors [343].
6 Based on the structures of membrane, there are flat and hollow fiber membranes. Moreover,
7 there are polymeric and ceramic membranes according to the materials of membrane used. For
8 the type and geometry, hollow fiber membrane contactors are widely applied due to large mass
9 transfer area [329, 344 - 346]. Hollow fiber membrane contactors (HFMCs) have become
10 attractive for gas separation due to high volume, higher active surface areas, and toleration of
11 long-term operation. [316]. The gas separation performance of HFMCs is mainly affected by
12 pore size and its distribution, selective skin layer thickness, membrane materials and fabrication
13 conditions [316, 318]. A lot of researchers are attention to improve gas separation performance
14 of HFMCs [319 - 322]. The use of ILs in HFMC processes can bring new opportunities in the
15 use of HFMCs in gas separation. For the type of materials, hydrophobic polymeric membranes
16 are popular due to high porosity and wide availability [347, 349].

17 Currently, ILs-based membrane contactor is a new concept that needs to be demonstrated.
18 The excellent thermal stability of the IL component will enable higher temperature applications,
19 and the negligible vapor pressure of the IL will sharply reduce solvent loss and regeneration
20 energy, compared to traditional absorbents.

21 **3.6.1 Separation of VOCs by ILMCs**

22 ILs in membrane contactors used as absorbents based on affinities for VOCs. FHILMCs are
23 preferred by industrial applications over flat membranes because the separation processes take
24 the advantages of the hollow fiber morphology by trapping IL within the micro-porous of the
25 fiber wall [317, 318]. Hollow fibers ionic liquid membrane contactors (HFILMCs), have been
26 developed to separate VOCs with long-term operation of membrane processes. Kim et al. [320]
27 fabricated a polyacrylonitrile HFMC by less toxic IL solvent instead of DMF. From this method,
28 they obtained a uniform pattern and high porosity of the inner surface of HFM. Zhang et al.

1 [321] obtained the hollow fiber renewal ionic liquid membranes that are interested because they
2 allow the solvent phase to be continuously renewed. Because of low melting point, viscosity
3 and relatively low cost, n-butylpyridinium tetrafluoroborate ([Bpy][BF₄]) and [Bmim][BF₄]
4 were used to separate toluene from cyclohexane solution (Figure 8). The selectivity of HFILMC
5 processes was larger than 10 at the temperature 323 K. Moreover, this membrane showed a
6 good long-term stability due to the high viscosity of IL. Furthermore, hollow fiber
7 configurations have been also studied with gelled ionic liquids to promote the mechanical
8 strength. The processes of gelation enable the ILs into the polymeric supported, which
9 preventing ILs release to the surrounding phase. Plaza et al. [312] synthesized a membrane by
10 the gelation of an ionic liquid [Bmim][PF₆] into porosity of polytetrafluoroethylene (PTFE)
11 hollow fibers. During the whole experiments, losses of IL of gelled ILHFMs were not observed,
12 which were more stable than the membrane without 12-hydroxystearic acid gelator. They
13 suggested that the gelled ionic liquid HFMC favored the separation of butanol, while the HFMC
14 without IL preferred acetone. Snochowska et al. [343] reported polypropylene HFILMC with
15 1-butyl-3-methylimidazolium methyl sulfate ([Bmim][MeSO₄]) separated ethanol from
16 aqueous solutions. The experiments were carried out with feed concentrations range from 1 to
17 5 wt.% and feed flow rates range from 1 to 8 dm³ h⁻¹. The results showed the efficiency of
18 ethanol used this contactor was around 20 %, which was considered satisfactory of industrial
19 requirement. Ortiz et al. [349] compared separation performance of two different contactors
20 including a membrane contactor (MC) and a semi-tank reactor with same IL Ag⁺ modified
21 [Bmim][BF₄] as reactive media. These contactors were used to separate propylene from
22 propylene/propane gas mixtures. They found that the contactor showed lower mass transfer
23 coefficient, however, the overall resistance of mass transport was decreased, and absorption rate
24 and capacity of contactor were increased. Finally, the separation performance of contactor was
25 more attractive than semi-tank reactors under same operating conditions.

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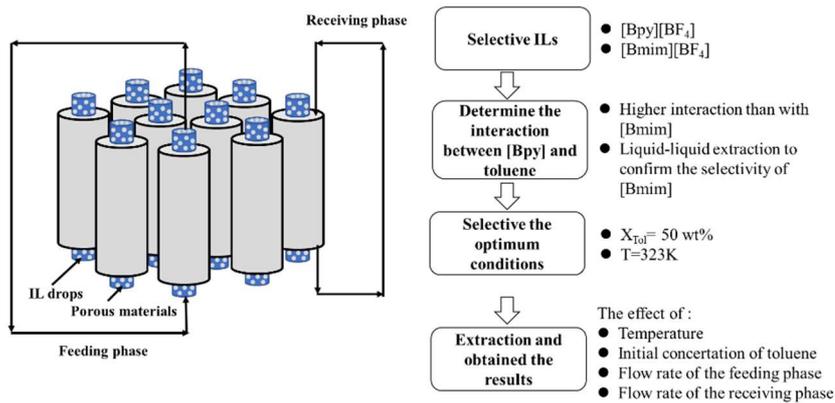


Figure 8. Hollow fiber supported ILMs by Zhang et al [321]

3.6.2 Separation of CO₂ by ILMCs

ILMCs processes with ionic liquids also widely used to CO₂ separation, especially HFILMCs. Until now, there are lots of reports focus on removing gas, such as CO₂, CO [350], and SO₂ [351], using ILs in membrane contactors due to their extremely high interfacial area from membranes and high selectivity from ILs. The main drawback of membrane contactors is wetting of membrane pores, which decreased the mass transport [355, 356] and pressure difference between the liquid and gas side of membrane [352 - 354, 357]. The methods used to reduce wetting effects are divided into following two parts: i) using composite hollow fiber membrane contactors; ii) using a dense membrane contactor with high permeable to CO₂.

Because of the wetting-effect, porous polymeric hollow fiber membrane contactors are more popular. Rostami et al. [332] researched the effects of [Bmim][BF₄] on the absorption CO₂ using polypropylene hollow fiber membrane contactors. They also investigated the effects of different operating parameters including concentration of IL in water, liquid and gas flow rates, and flow direction. The results showed that adding IL into the water, the CO₂ solubility and viscosity of the solution were increased and diffusion coefficient of CO₂ in the solution was decreased. The resistance of mass transfer in liquid phase decrease by increasing the flow rate of liquid. Moreover, when the gas flow rate increase, higher amount of gas enters the membrane contactor resulting in the increase of mass transfer flux. For the direction of gas and liquid flows, the counter-current flow has a higher efficiency than the co-current flow because the counter-current flow keeps high mass transfer through the length of membrane. Besides, the effects of

1 temperature on CO₂ capture were studied by Gomez-Coma et al. [178]. They demonstrated that
2 the CO₂ removal efficiency was dependent on temperature. For instance, when the temperature
3 increased from 291 to 348 K, the CO₂ removal efficiency was increased from 30 to 45%.
4 Furthermore, this research group studied the dry and wet PVDF hollow fiber membrane
5 contactors with 1-ethyl-3-methylpyridinium ethylsulfate ([Emim][EtSO₄]) to absorb CO₂
6 [358]. They indicated that the fibers in dry form showed higher CO₂ permeability than in the
7 wet form, and the permeability increased by 43 % due to the presence of immobilized IL.
8 Wickramanayake et al. [321] designed a four morphology of polymeric HFILMCs, which were
9 fabricated using Matrimid and [Hmim][NTf₂]. The primary of this study was to optimize the
10 porosity of the support affecting gas separation performance. The porosity of membranes was
11 subjected to the polymer content. The porosity of membrane was increased by decreasing the
12 polymer content. It was found that the CO₂ permeance and the CO₂/H₂ selectivity of membrane
13 were increased with increasing the porosity in micro-porous region because of higher IL content
14 in selective layer of fibers. However, it should be noted that a high porosity is beneficial to
15 the gas separation, while there are some negative effects on mechanical strength of membrane.
16 Furthermore, they also studied the structural stability by same types of HFILMCs with different
17 polymeric matrixes [322]. In this system they fabricated polymeric HFILMs containing 1-
18 hexyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([C₆mim][NTf₂]) with Matrimid
19 and Torlon matrixes. By using sufficiently high fiber porosity and thus maximizing the IL
20 content of the membrane, it was found that Torlon fibers have improved mechanical strength
21 compared with Matrimid. The study of Kim et al. [179] focused on improving the CO₂
22 permeability and stability of the HFILMs composited by [Emim][NTf₂] and PVDF. They
23 indicated that the gas permeability and stability were affected by support structure. Two
24 structures of hollow support obtained such as sponge-like and finger-like, and sponge-like
25 hollow support showed higher stability than finger-like under 1.5 bar. And for gas separation,
26 they observed CO₂ permeability over 2600 barrer and selectivity of CO₂/N₂ was from 30-33.

27 The gas separation performance of HFILMCs is depended on the following factors [317,
28 318, 321]: i) porosity and pore size, which control the selectivity of membrane; ii) selective
29 skin layer thickness, which decides the membrane flux; iii) materials and fabrication of

1 membrane, which govern the gas permeability and perm-selectivity. However, the HFILMCs
2 also show a few problems, for example poor stability and the fabrication condition of membrane,
3 which limit the applications of HFILMCs. For the industrial applications, the HFILMCs should
4 meet the requirements of both good transport and mechanical properties, as well as find suitable
5 approaches to address these competing interests.

6 Additionally, the nonporous composite membrane with thin dense top layer also provides
7 an opportunity to eliminate the effect of pore wetting. For example, Bazhenov et al. [357]
8 designed a gas-liquid membrane contactor to capture CO₂. This dense flat-sheet membrane
9 contactor was composed by [1-(trimethylsilyl)-1-propyne] and nine different RTILs. According
10 to the solvent-polymer membrane compatibility, [Emim][BF₄] were selected to apply in
11 contactor. They revealed that the solvent-membrane interaction is mainly determined by the
12 liquid surface tension rather than viscosity and molecular size of RTILs. In addition, membrane
13 contactors using both porous and nonporous polymeric HFMCs with 1-butyl-3-
14 methylimidazolium tricyanomethanide ([Bmim][TCM]) were developed and compared as CO₂
15 capture by Dai et al. [359]. According to the compatibility and stability of different polymeric
16 membranes, the porous PTFE and nonporous Teflon-PP membranes were selected to separate
17 CO₂. The effects of operating pressure and gas flow rate were investigated both porous and
18 nonporous membranes. It was found that the operating pressure has opposite effect on CO₂ flux
19 of two membranes, with a more significant effect on the porous membrane. Additionally, higher
20 gas flow rate results in higher CO₂ flux in both membranes, while the porous membrane
21 contactor presents a higher CO₂ flux and nonporous membrane contactor exhibits better long-
22 term stability.

23 Besides, many challenges for IL-based membrane contactors still need to be overcome.
24 Firstly, low surface tension of ILs, which leads to easy membrane pore-wetting [1, 168, 355,
25 356]. Another challenge is that the ILs are generally good solvents also for polymers [228, 229],
26 which means ILs might dissolve some polymeric membranes or polymer-based sealing
27 materials gradually. In addition, the toxicity and potential environmental impacts of many ILs
28 [154, 360 - 362] are still not clear, although commonly ILs are accepted as green solvents.
29 Moreover, the high price of ILs is also a drawback.

1 4. Conclusion

2 Different kinds of membrane processes relating to use of ILs for VOCs or CO₂ removal
 3 are discussed, which showed better performance than conventional processes. Table 14 shows
 4 a comprehensive summary of advantages and disadvantages on ILs membrane processes for
 5 VOCs or CO₂ separation.

6 **Table 14.** Advantages and disadvantages of membrane processes containing ILs.
 7

| Membranes | Advantages | Disadvantages |
|-----------|---|--|
| SILMs | <ul style="list-style-type: none"> - High interfacial area per unit volume for mass transfer - Low solvent holding - More efficient in application over other liquid membrane processes | <ul style="list-style-type: none"> - In over-time operation, the liquid phase evaporates or is pushed out of the membrane pores, resulting in a non-selective transport - Thick membranes will exhibit improved stability and reasonable lifetimes but lower gas flux - Regeneration is complicated and difficult |
| ILPMs | <ul style="list-style-type: none"> - Unique combination properties of ILs and intrinsic polymer properties - Prevent the membrane from excessive swelling and maintain the gas separation performances even at increased temperature and pressure conditions | <ul style="list-style-type: none"> - Gas permeability and diffusivity are hindered through the solid polymer matrix - Plasticization phenomena occurred - Limited by Robeson upper bound line - Weak thermal and mechanical stability |
| ILMMMs | <ul style="list-style-type: none"> - Membrane performances including permeability and selectivity were enhanced due to the synergetic effect between ILs and fillers - Added inorganic fillers will increase the sorption/diffusion - ILs help to improve the interfacial voids in the MMMs - Relative higher thermal and mechanical properties than others | <ul style="list-style-type: none"> - ILs could cause pore blockage in porous matrix - Surface voids and filler agglomeration affect the gas separation and structures of membranes - Poor polymers and inorganic fillers compatibility |

| | | |
|-------|---|--|
| PILMs | <ul style="list-style-type: none"> - PILs modified membranes show tunable charge, wettability - Improved mechanical properties - Promoted both gas permeability and ideal selectivity - RTILs with polymerizable groups can convert into solid, which composite dense membranes resulting in a friendly process | <ul style="list-style-type: none"> - Separation performances of PILMs are mainly depended on polymer monomer, polymeric methods - The stability of membrane needs improved by adding plasticizers |
| ILGMs | <ul style="list-style-type: none"> - Liquid-like gas transport properties in a solid state, which is easier to handle and to prepare membrane modules for industrial applications - Gas permeability is increased | <ul style="list-style-type: none"> - Thermal and mechanical stability are limited - Gas permeability and ideal selectivity are low - Gelators are limited - High packing density is required - Suffering the trade-off effect between signal gas permeability and selectivity of gas pair |
| ILMCs | <ul style="list-style-type: none"> - Long-time operation - Higher active surface area to volume ratio - Allow the solvent phase to be continuously renewed -low resistance of gas flow - Self-supporting structure - Ability to perform at high pressure - Determined interfacial area - Independent control of gas and liquid flow rates - Avoidance the drop dragging - Reduce the solvent loss and regeneration energy | <ul style="list-style-type: none"> - Poor stability and processability - Selection of materials is limited - Some ILs can dissolve the polymeric or polymer-based sealing membrane - Low surface tension of ILs |

1 Regardless of the limitation of some ILs, the use of them as absorbents for VOCs or CO₂
2 separation processes have become increasingly important. Although great progress has been
3 done on removal of VOCs or CO₂ on ILs and membrane separation processes, there are still
4 knowledge gaps that need to be filled. Additional studies are still needing to: i) improve VOCs
5 or CO₂ adsorption capacity of ILs and membrane separation processes; ii) reduce toxicity of
6 ILs and overcome the limitations related to the stability, which can broaden the range of
7 applications of ILs; iii) increase the selectivity and stability of ILs and membrane; iv) simplify
8 recycling processes of ILs and adsorbed VOCs or CO₂; v) decrease the cost of adsorbent and
9 membrane set-up equipment; vi) solve the difficulty of high boiling VOCs desorption.

10 All these improvements will make possible to replace the traditional methods for VOCs or

- 1 CO₂ removal by green and economic processes based on ILs and membrane separation in the
- 2 not distant future.
- 3

Names and abbreviations of ILs

| Abbreviations | Names |
|--|--|
| [Abim][NTf ₂] | 1-allyl-3-butylimidazolium bis(trifluoromethanesulfonyl)imide |
| [Bmim][Ac] | 1-butyl-3-methylimidazolium acetate |
| [Bmim][PF ₆] | 1-butyl-3-methylimidazolium hexafluorophosphate |
| [Bmim][BF ₄] | 1-butyl-3-methylimidazolium tetrafluoroborate |
| [Bmim][NTf ₂] | 1-butyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide |
| [Bpy][BF ₄] | n-butylpyridinium tetrafluoroborate |
| [Bmim][TCM] | 1-butyl-3-methylimidazolium tricyanomethanide |
| [Bmim][MeSO ₄] | 1-butyl-3-methylimidazolium methylsulfate |
| [C _n mim][NTf ₂] (n=2, 4, 6) | (1-alkyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide |
| [C ₂ mim][BF ₄] | 1-ethyl-3-methylimidazolium tetrafluoroborate |
| [C _n mim][BF ₄] (n=4, 6-8, 12) | 1-n-alkyl-3-methylimidazolium tetrafluoroborate |
| [C ₈ mim][PF ₆] | 1-n-octyl-3-methylimidazolium hexafluorophosphate |
| [C ₃ NH ₂ mim][NTf ₂] | n-aminopropyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide |
| [C ₄ C ₁ im][BF ₄] | 1-butyl-3-methylimidazolium tetrafluoroborate |
| [C _n C ₁ im][PF ₆] (n = 4 or 6) | 1-butyl-3-methylimidazolium hexafluorophosphate (n=4) 1-hexyl-3-methylimidazolium hexafluorophosphate (n=6) |
| [Choline][Pro] | (2-hydroxyethyl)-trimethyl-ammonium(S)-2-pyrrolidinecarboxylic |
| [C ₂ mim][TCB] | 1-ethyl-3-methylimidazolium tetracyanoborate |
| [Emim][BF ₄] | 1-ethyl-3-methylimidazolium tetrafluoroborate |
| [Emim][DCA] | 1-ethyl-3-methylimidazolium dicyanoamide |
| [Emim][EtSO ₄] | 1-ethyl-3-methylimidazolium ethylsulfate |
| [Emim][NTf ₂] | 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide |
| [Emim][Ac] | 1-ethyl-3-methylimidazolium acetate |
| [Empy][EtSO ₄] | 1-ethyl-3-methylpyridinium ethylsulfate |
| [Hmim][PF ₆] | 1-hexyl-3-methylimidazolium hexafluorophosphate |
| [Hmim][NTf ₂] | 1-hexyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide |
| [N ₄₄₄][formate] | tributylmethylammonium formate |
| [Omim][TCM] | 1-methyl-3-octylimidazolium tricyanomethanide |
| [Omim][PF ₆] | 1-methyl-3-octylimidazolium hexafluorophosphate |
| [P ₄₄₄][formate] | tributylmethylphosphonium formate |

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