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Latest Development of Ionic Liquid Membranes and Their Applications

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ABSTRACT

Ionic Liquids (ILs) are semi-solid electrolytes with higher thermal stability, they also have the property of low flammability and slight volatility. Due to its unique properties of this electrolyte, they have wider applications in the field of science. Ionic Liquid Membranes (ILMs) are membranes embedded with ILs. ILMs technology was initiated to meet the low-cost investment on an industrial scale. However, the stability of the membranes depends on the ILs. This paper summarizes the recent fabrication of different types of ILMs along with their applications to various fields and also termed with the limitations to help further researchers in the field of membrane technology.

Keywords: Ionic liquids, gas separation, supported ionic liquid membranes

INTRODUCTION

The fast-growing field of membrane technology helps to solve the issues of high power consumption and waste generation. Ionic liquids (ILs) are used because of their unique properties but due to some limitations of ILs, they are impregnated inside the membrane pores to form ionic liquid membranes (ILMs) [1]. In general, there are different types of ILMs such as bulk liquid membranes, emulsion liquid membranes and supported liquid membranes. Supported ionic liquid membranes (SILMs) have strong stability towards organic solvents, alcohols, hydrocarbons, etc. [2–6]. But due to the limitation of SILMs, there are still doubts about them for the industrial application [2]. ILMs are also more stable when compared to traditional separation methods such as centrifugation, evaporation, and adsorption [2, 3]. SILMs have been widely used for various gas separations such as carbon-di-oxide (CO_2), methane (CH_4), oxygen (O_2), and nitrogen (N_2) [7–10]. Emulsion liquid membranes (ELMs) have higher efficiency in treating effluents by impregnating ionic liquid inside them [11]. The application of ILMs further moves with liquid-liquid or solid-liquid extraction too. This paper summarizes the fabrication of various ILMs along with their application in various fields.

The general methods of fabrication of SILMs are direct immersion, vacuum and pressure method. The most generally utilized two methodologies for using ILs as membrane materials are shown in Figure 1. While quasi solidified ionic liquid membranes (QSILMs) can be prepared by making a solution of IL with a special gel to form quasi solidification film this is the most efficient method when comparing SILMs [1]. The hydrophilic polyacrylonitrile (PAN) flat sheet

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membrane was used as a supporting membrane for the filtration of the dimethyl carbonate/methanol mixture. The hydrophobic membrane was not able to hold the IL inside the pores even after applying high vacuum pressure. But, the hydrophobic microporous membrane was used to protect the gel layer in gas permeation experiment [1, 2, 10]. The high porosity hydrophobic polytetrafluoroethylene (PTFE) and polyvinylidene fluoride (PVDF) polymers were selected for the support of ILs [11].

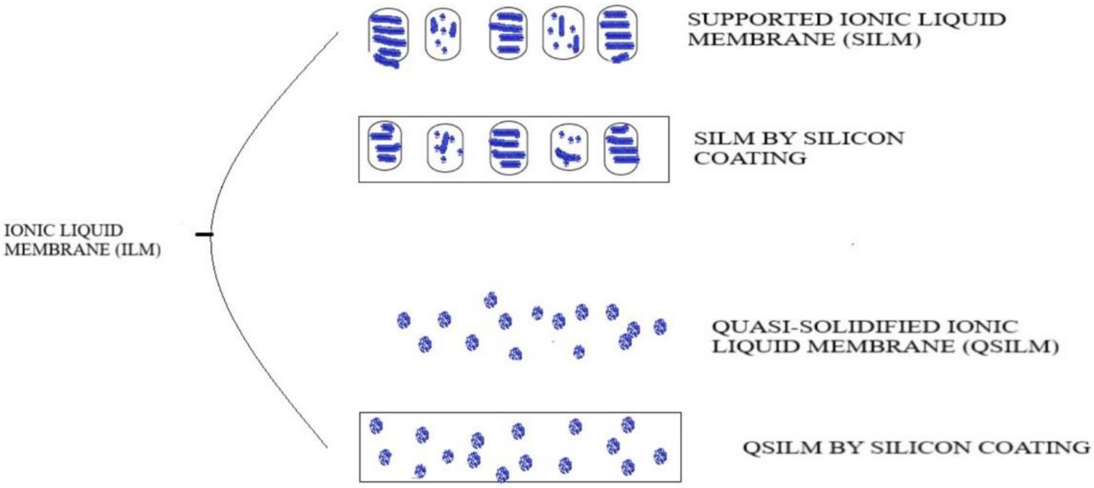


Figure 1 Two main ways of using ILs as membrane materials

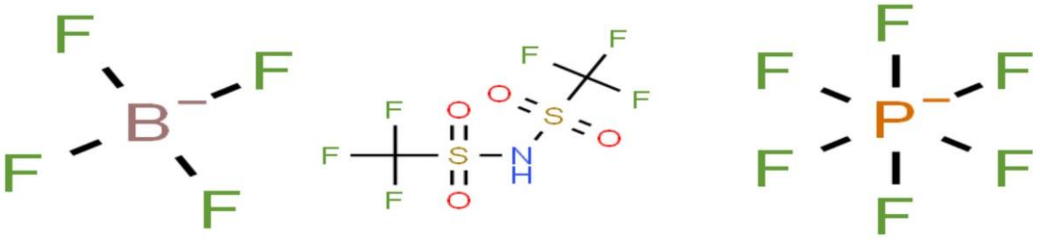
GAS SEPARATION USING IONIC LIQUID MEMBRANES

ILMs have been considered attributable to the crucial properties of ILs like insignificant instability, low and high combustibility and thermal stability creating them as a whole liquid phase for SILMs [12, 13]. The gas transport in SILMs was depended on a solution-diffusion mechanism, and their main thrust for ideal selectivities is the variance in the solubility of different gases in the IL [14].

IMPORTANCE OF MATERIALS DEVELOPMENT FOR ILS

ILs are molten state salts that entirely consist of cations and anions. The chemical structures of generally used anions and cations of ILs are presented in Figure 2 & 3. The choice of ionic constituents determines the physiochemical properties of ILs. The reaction rates of alcoholysis, amminolysis, perhydrolysis, and enantioselective acylation processes were better by using Candida Antarctica lipase as an enzyme catalyst in BuMeIm-based ILs. The room temperature ionic liquid (Vinylbenzyl) trimethylammonium acetate [Vbtma][Ac] was prepared by passing the solution of (Vinylbenzyl) trimethylammonium chloride [Vbtma][Cl] with methanol along with OH resin through the column. Vinylbenzyl trimethylammonium hydroxide [Vbtma][OH] was the solution obtained from the column. A mixture of acetic acid with methanol is added to the obtained solution and mixed well for 30 minutes. The solvent extraction is done by applying a vacuum [15]. The reaction of methylimidazole with chlorobutane yielded 1-n-butyl-3-methylimidazolium chloride [C4mim][Cl], C4mim][Cl] on further reacting with lithium bis (trifluoromethylsulfonyl) imide (LiTf2N) produced 1-butyl-3-methylimidazolium bis (trifluoromethyl sulfonyl) imide [C4mim][Tf2N] IL. Similarly, the reaction of methylimidazole with 2-bromopropylamine hydrobromide produced 1-propylamine-3-methylimidazolium bromide [C3NH2mim][Br] and on the ion exchange with LiTf2N, N-aminopropyl-3-methylimidazolium bis (trifluoromethylsulfonyl) imide [C3NH2mim][Tf2N] IL was fabricated [16]. 1-butyl-3-methylimidazolium tetraisothiocyanatocobaltate (II) (Bmim2Co(NCS)4) IL was prepared by mixing cobalt (II) chloride (CoCl2), potassium thiocynanate (KNCS), and 1-butyl -3-methylimidazoilum chloride [Bmim]Cl with acetone. The sample was then stirred for a certain time. To remove the solvent, the mixture was subjected to evaporation and filtration. Then the sample was washed with dichloromethane and dried under vacuum [17]. A reactive methoxysilane-functionalized ionic liquid (Si-IL) was designed by reacting (3-chloropropyl)triethoxysilane (CPS) and N,N-dimethyl propyl amine (NDPA) to form methoxysilane-functionalized quaternary ammonium component in the first step. The second step involves replacing chloride ion (Cl−) with tetrafluoroborate ion (BF4 −) [18].

Many researchers have been done, where SILMs were prepared for future applications in gas separations. Synthesis of SLIMs is the most commonly used soaking porous support membrane in the preferred IL. As ILs have comparatively trivial vapour pressure and high viscosity, the use of vacuum is normally used to eliminate gas bubbles in the resultant membranes.



Bistriflimide **Bistriflimide** **Hexafluorophosphate**

Triflate ion **Tetrafluoroborate**

Figure 2 Chemical structures of generally used anions of ILs for gas separation

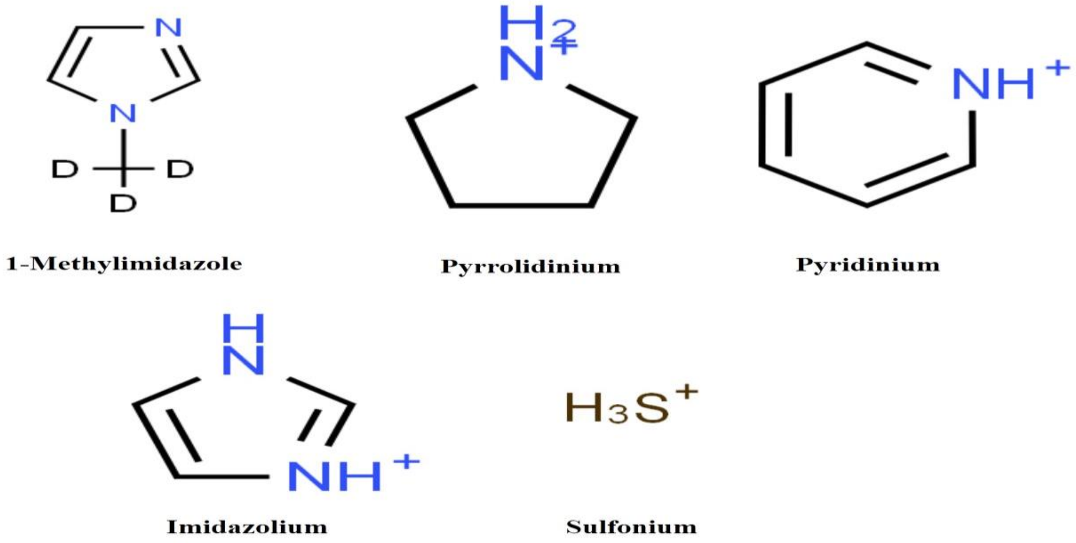


Figure 3 Chemical structures of generally used cations of ILs for gas separation

STABILITY OF IONIC LIQUID MEMBRANES

SILMs have greater stability than supported liquid membranes. But then there are several factors involved in influencing the stability of SILMs; they are membrane pore size, properties of ILs, transmembrane pressure, the interfacial tension between the aqueous phase and membrane support. SILMs with membrane pore size 100–200 nm offer good support. Inorganic nanofiltration membranes are the suitable choice for defined pore structure and stability. The hydrophobic support membranes are stable than hydrophilic support. The loss of ILs in high transmembrane pressure can be controlled by nanofiltration membranes; the selection of proper pore size of the membrane can increase the transmembrane pressure. Figure 4 shows the graphical representation of mixed gas permeation test cell.

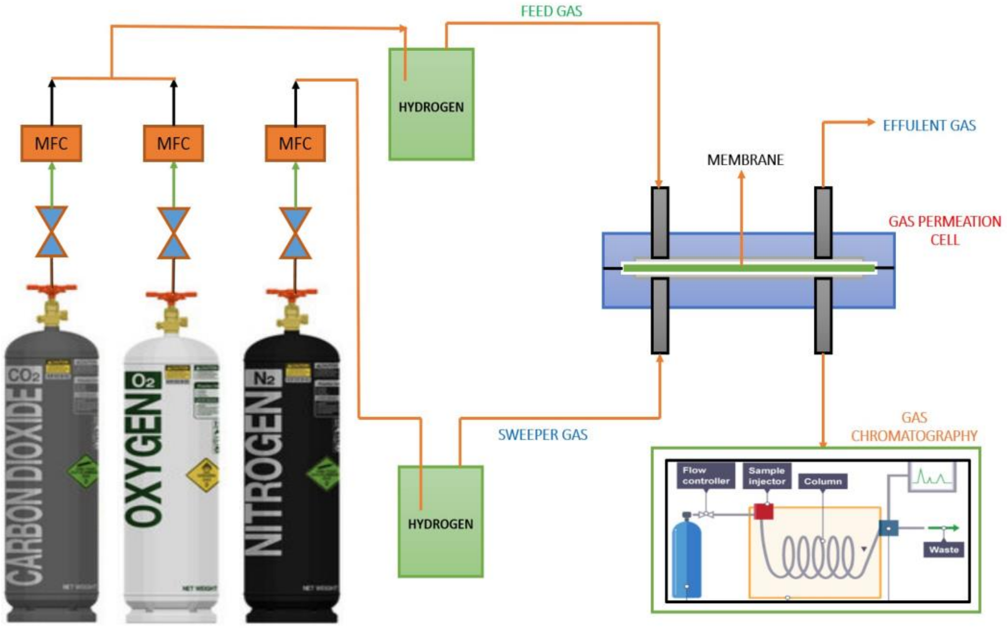


Figure 4 Graphical representation of gas permeation setup

Silver-based ILMs is a novel innovative membrane. The liquid was made by mixing silver tetrafluoroborate ($AgBF_4$) with the proportional other liquids to prepare the membrane solvent [19]. The fabrication of SILMs using supercritical fluids was tested with supercritical fluid deposition [4]. The composition of 80% IL with 20% acetone was selected as a supporting phase for the absorption of CO_2/O_2 by aqueous monoethanolamine (MEA) [20]. After degassing the polymer supports ILs, the polymeric support was saturated with ILs, which provides a suitable supported imidazolium ionic liquid membrane for toluene separation [2,21]. The immobilization of the membrane solvent inside the pores requires pressure through the inert gas nitrogen (N_2), then the procedure was repeated to ensure that all membrane pores were filled with liquid [22,23].

A polyionic liquid matrix can withstand heavy ionic liquid in the self-standing membrane, the higher amount of liquid may make the membrane too soft and this can be sorted out by adding alumina particles [24]. The vacuum was applied for 1 hour to remove the air inside the PVDF porous membrane by placing it in a desiccator [7]. For the easier introduction of IL, the support membranes were immersed in ethanol [10]. For preparing the emulsion liquid, the internal membrane and external phases were individually prepared at room temperature [25]. Composite membranes were fabricated by adding various ILs to the polymer solution to prepare polymer-IL membranes [26]. The hollow fiber-supported ionic liquid membranes (HFSILMs) were prepared after the synthesis of hollow fiber substrate [27].

Silver or aprotic ILs (Ag/AIL) were homogeneously distributed inside the membrane pores, only the biggest macropores were partially filled [28]. Leaching is the major problem related to the immobilization of an IL so membranes with a double polyether-block-amide (PEBA) layer are preferred, the self-produced membranes were compared and tested with commercial membranes but those results haven't proposed still [29]. The porous polyionic liquid membranes were immersed in distilled water to achieve phase inversion [30]. The polyethylene membranes were immersed in homogeneous mixtures containing ethanol to ILs in the ratio of 15:5, 12:8, and 10:10, respectively. The ethanol was purposely used to enhance ILs into the membranes [31]. Applications of various gas separations using ILMs were presented in Table 1.

APPLICATION OF IONIC LIQUID MEMBRANES (ILMS)

SILMs were used in the separation of methanol/dimethyl carbonates (DMC). However, an increased amount of methanol decreases the separation process due to the coupling effects of DMC and methanol, which also shows that membrane performance depends slightly on the concentration (1). High solubility capacity for various gases was observed in ILs. The membranes containing IL of weight 20%, 30%, and 40% offer good selectivity and permeability. The transportation rate and selectivity of mixed gases were affected by the nature of support membranes. The increased concentration of IL resulted in notable gas permeability, but the mechanical stability of SILMs declined with an enhance in the hydrocarbon chain of IL cations. While in the separation of CO_2 from other gases, supported membranes with fluoroalkyl-functionalized imidazolium IL was found to be lower and higher selectivity of CO_2/N_2 and CO_2/CH_4.

Table 1 shows generally used support membranes and their relating ILs for different gas separation applications

Ionic liquids	Support membrane	Characterization	Application	Weight % of ionic liquids	Gas permeability barrier					Ref
					CO_2	N_2	O_2	CH_4	H_2	
[BMIM] [Tf2N]	PI	NMR, FTIR, SEM, XRD, TGA, DSC	Separation of CO_2	35	33.4	1.3	4.8	1.4	25	[32]
[emim][B(CN)4]	PVDF	PLM, DSC	Separation of CO_2 from hydrogen and flue gas	50	1778	43.2	-	-	13.9	[33]
[emim][Tf2N]	Hydrophilic PES	-	Separation of CO_2, N_2 and CH_4	-	1050	51	-	94	-	[34]
[emim] [BF4]	PVDF-HFP	FTIR, DSC, SEM	Separation of CO_2 from N_2	50	400	6.5	-	-	-	[35]
[C4MPyrr] [B(CN)4]	PES	-	Separation of CO_2 and N_2	-	1633	44	-	-	-	[36]
[C2mim][DCA]	Hydrophilic PTFE	-	Separation of CO_2	-	476	7.0	-	20.7	-	[37]

A double network (DN) ion gel-based membrane act as a venting device for spaceship by capturing low CO_2 concentration [8]. For the competitive thermostable carbon capture, SILMs with immobilized carbon anhydrases resulted in positive news and also for selective separation of hydrogen sulphide (H_2S) from natural gas, SILMs based on ILs (1-butyl-3-methylimidazolium acetate) [bmim] [Ac] were used [10]. The cellulose-based CO_2 capture membrane is a cheap and renewable method; this also offers advantages to work under unstable conditions. The supported hollow fiber membranes were used in green engineering for carbon capture. SILMs also help to achieve better recovery and reuse rate by offering excellent permeability of sulphur dioxide (SO_2). In the separation of chromium (III) from aqueous solution, using pseudo-emulsion-based hollow fiber strip dispersion (PEHFSD) technology increased the transport of chromium with carrier concentration and the notable extraction rate was 95% under operating conditions.

Supported liquid membrane systems can be used for the separation of lithium from sodium chloride, but the dissolution of IL may reduce the lifetime of the process. For the separation of organic compounds, ILMs technology was preferred for its low energy consumption, from the separation of trans-esterification reaction products found that nylon and iso-pore compounds were suitable than others [1, 5, 27]. Using SILMs for 1,3-propanediol the separation factor was significantly increased from 0.4 to 177. Water-based SILMs resulted in a low permeation rate due to their high viscosities of the ILs, hence SILMs greatly enhanced the selectivity of aromatic hydrocarbons. The extremely high selectivity transport of secondary amine over tertiary amine was achieved with SILMs with 1-butyl-3-methylimidazolium hexafluorophosphate [BMIM] [PF6] and PVDF as a support membrane [5, 24, 31].

The novel ILs offers high CO_2 solubility values but further physical properties are required for the transport of CO_2 through membranes [6–10, 21, 26, 27, 38]. Support liquid membranes based on silver was a novel method of IL, but for the practical application, the selectivity of the membrane could not last after 24 hours of operation [1]. IL@beta-CD-Gel membrane for tea polyphenols evaluated that this provides better selectivity filtration from natural compounds [7]. The imidazolium-based SILMs exhibit higher performance concerning mass transfer coefficients for the separation of toluene. However, while considering other compounds molecular volume or viscosity should be considered [2].

Ion gel membrane offered better ionic conductivity for solid electrolytes [24]. The role of anion, pH of wastewater plays a significant role in SILMs extraction. Then the use of ILs exhibits advantages of non-volatility, greater efficiency, stability, and durability [21]. In biobutanol concentration by pervaporation, the selection of materials needs to be analyzed because the stability of the membrane may depend on any property of the material [20]. The need for toxic inorganic solvents was reduced by replacing them with edible oil in preparing ELM for metal solution treatment [23, 25]. For recyclable industrial applications, poly (amino acid ionic liquid) (PAAILMs) provides excellent antibacterial and antifouling properties [5, 22].

Impregnating polyethylene membrane with 1-butyl-3-methylimidazolium dicyanamide, IL has its application in heavy metal wastewater treatment [31]. Three SIL-based imidazolium used for the pie electron separation of toluene, styrene, and divinylbenzene in a hexane medium: Congo red, Erichrome black T, and Ramazol brilliant blue R in an aqueous solution. The membrane showed a significant reduction in the colony of bacteria, which was investigated using pseudomonas aeruginous and staphylococcus aureus, thus suggesting better antibacterial property for the membranes [2, 19, 31].

CONCLUSIONS

ILMs are a great alternative to traditional methods of operations. The main advantages of ILMs are cost efficiency, longer stability, low energy consumption, and even some are eco-friendly. But the stability of membranes also depends on properties of the materials and some novel method of ILMs that utilizes silver may cost a bit higher. ILMs provide great efficiency towards the separation of gas mixtures, which paves the way for monitoring air pollution and to meet the solution of clean water treatment by removing organic and inorganic compounds from wastewater. However, every separate method of fabrication of ILMs uses different ILs to succeed in their end goal. Concentrating on the limitations of ILMs, we can update ILMs technology for a better world.

NOMENCLATURE

AIL	Aprotic Ionic Liquid
Ag	Silver
AgBF4	Silver tetrafluoroborate
CA	Cellulose Acetate
CH4	Methane
CO2	Carbon -di-oxide
CoCl2	Cobalt (II) chloride
CPS	(3-chloropropyl)triethoxysilane
DMC	Dimethyl Carbonates
DN	Double Network
DSC	Differential Scanning Calorimetry
FID	Flame Ionization Detector
FTIR	Fourier - Transform Infrared Spectroscopy
GC	Gas Chromatography
H_2S	Hydrogen Sulphide
ILs	Ionic Liquids
ILMs	Ionic Liquid Membranes
KNCS	Potassium thiocynanate
LiTf2N	Lithium bis(trifluoromethylsulfonyl)imide
MEA	Monoethanolamine
N2	Nitrogen
NDPA	N,N-dimethylpropyl amine
NMR	Nuclear Magnetic Resonance
OH	Hydroxide ion
PAN	Polyacrylonitrile
PAAILMs	Poly Amino Acid Ionic Liquid Membranes
PEBA	Polyether-Block-Amide
PVDF	Polyvinylidene fluoride
QSILM	Quasi Solidified Ionic Liquid Membrane
SCFD	Supercritical fluid deposition
SILMs	Supported Ionic Liquid Membranes
Si - IL	Methoxysilane- functionalized ionic liquid
SEM	Scanning Electron Microscopy
SSL	Split/Splitless Injection
TCD	Thermal Conductivity Detector
TGA	Thermo-Gravimetric Analyses
XRD	X-ray Diffraction
[BF4]	Tetrafluoroborate ion
[BMIM] [Ac] [10]	1-Butyl-3-Methylimidazolium Acetate
[BMIM]Cl	1-butyl -3-methylimidazoilum chloride
[Bmim2Co(NCS)4]	1-butyl-3-methylimidazolium tetraisothiocyanatocobaltate(II)
[BMIM] [PF6]	1-Butyl-3-Methylimidazolium Hexafluorophosphate
[BMIM][Tf2N]	1-butyl-3-methylimidazolium bis(tri fluoromethylsulfonyl)imide
[C2mim][DCA]	1-ethyl-3-methylimidazoilum dicyanamide
[C3NH2mim][Br]	1-propylamine -3-methylimidazolium bromide
[C3NH2mim][Tf2N]	N-aminopropyl-3-methylimidazolium bis (trifluoromethylsulfonyl) imide
[C4mim][Cl]	1-n- butyl-3-methylimidazolium chloride
[Cl−]	Chlorine ion
[emim][B(CN)4]	1-ethyl-3-methylimidazoilum tetracyanoborate
[emim] [BF4]	1-ethyl-3-methylimidazoilum tetrafluoroborate
[emim][Tf2N]	1-ethyl-3-methylimidazolium bis (trifluoromethanesulfonyl) amide
[Vbtma][Ac]	(Vinylbenzyl) trimethylammonium acetate

[Vbtma][Cl]	(Vinylbenzyl) trimethylammonium chloride
[Vbtma][OH]	(Vinylbenzyl) trimethylammonium hydroxide

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