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Comprehensive review on the efficiency of ionic liquid materials for membrane separation and environmental applications

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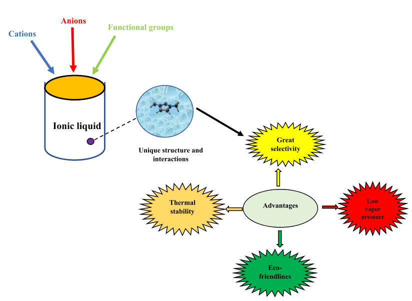
HIGHLIGHTS

- State-of-the-art review was performed on the efficiency of IL-based absorbents to separate CO_2.
- A techno-economic analysis was carried out to compare the cost-effectiveness of ILs with alkanolamine absorbents.
- Studying major environmental impacts of the ILs applications in industries was conducted.
- Discussion of future perspectives towards solving the operational challenges was implemented.

GRAPHICAL ABSTRACT



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ABSTRACT

In the current twenty years, industrial applications of ionic liquids (ILs) have been of paramount attention due to their indisputable positive characteristics like negligible volatility and chemical/thermal stability. These brilliant advantages open new horizons towards environmentally friendly application of ILs in several industrial activities like membrane-based CO_2 separation, electrolyte, bioprocessing, targeted drug delivery and solar panels. The principal intention of this article is to prepare a comprehensive review on the potential efficiency of IL-based absorbents to separate CO_2 acidic contaminant from industrial gaseous streams compared to alkanolamine absorbents as the benchmark. For this purpose, a techno-economic evaluation is presented to compare the cost-

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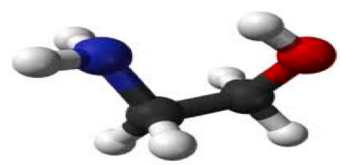
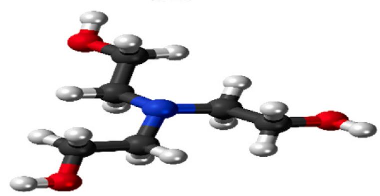
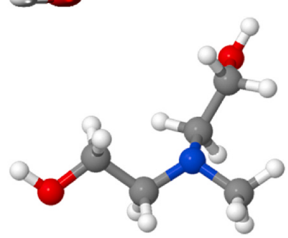
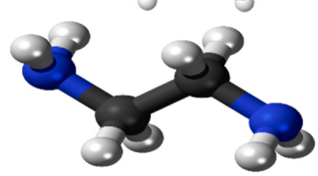
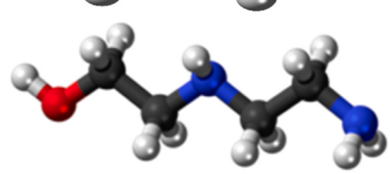
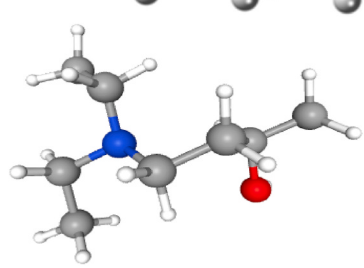
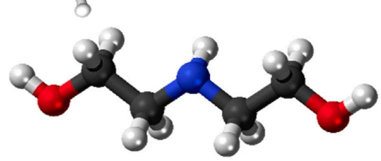
Ionic liquids
Mass transfer

effectiveness of ILs compared to alkanolamine absorbents. Finally, major environmental impacts of the ILs applications in industries are discussed and future perspectives towards solving the operational challenges are presented in detail.

1. Introduction

Significant requisition of different fossil fuels for energy production is the main cause of air pollution (Mac Dowell et al., 2017; Cao et al., 2021/04). An estimation has implied the annual release of approximately thirty-three thousand million metric tons of CO_2 to the atmosphere (Hospital-Benito et al., 2021). Intergovernmental Panel on Climate Change (IPCC) provided a comprehensive report about the global warming and mentioned the 1.5 °C increment of global temperature compared to pre-industrial era in 2030 (Lian et al., 2021). Continuous increment of global temperature will effectively eventuate in climate deterioration and leads to the emergence of a series of socioecological challenges (Li et al., 2019; Sun et al., 2018). Therefore, efficient mitigation of anthropogenic CO_2 emission into the atmosphere is of great importance to decrease perilous impacts of global warming like acid rain, climate deterioration, fog, and haze.

Table 1
Comprehensive information about common alkanolamine absorbents for CO_2 separation (Nakhjiri et al., 2018a; Nakhjiri and Heydarinasab, 2019/10; Akinola et al., 2019; Razavi et al., 2016/01; Zhang, 2016; Rahim et al., 2014; Shirazian et al., 2020).

Amine type	Chemical formulation	Molecular structure	Molecular weight (g/mol)
MEA	[C_2H_7NO]		61.08
TEA	[$C_6H_{15}NO_3$]		101.19
MDEA	$CH_3N\ [C_2H_4OH]_2$		119.164
EDA	[$C_2H_8N_2$]		60.1
EEA	[$CH_3CH_2NHCH_2CH_2OH$]		89.14
DEAB	[$C_8H_{19}NO$]		145.24
DEA	[$C_4H_{11}NO_2$]		105.14

Gas separation technologies (focusing on CO_2) including cryogenic, adsorption, absorption, membrane-based separation, and bio-fixation have been extensively applied to mitigate CO_2 release in industrial plants (Tan et al., 2016; Cao et al., 2021a; Babanezhad et al., 2020a; Marjani et al., 2021/03; Hart and Gnanendran, 2009; Yu et al., 2012). Alkanolamine absorbents are known as industrial benchmark for efficient separation of CO_2 from gaseous mixtures considering the formation of carbamate and carbonate (Hospital-Benito et al., 2021; Cabral et al., 2019; Vega et al., 2020). Table 1 presents various applied alkanolamine absorbents for CO_2 separation.

Despite the great performance of alkanolamine absorbents for CO_2 separation such as high efficiency and low cost, the emergence of important drawbacks like significant consumption of energy during the regeneration process, high volatility, and equipment corrosion have confined their applications in separation (Mota-Martinez et al., 2017; García-Gutiérrez et al., 2016; Taghvaie Nakhjiri et al., 2020/10; Cao et al., 2022a). In recent decades, various novel chemical absorbents such as amino acid salt solutions (i.e., potassium glycinate (PG), potassium argininate (PA), potassium lysinate (PL) and potassium threonate (PT)), alkali solution (NaOH), ionic liquids (ILs) ([Bmim][BF_4]) and nanofluids (i.e., MDEA-based nanofluids) have been under investigations to modify the performance of the CO_2 separation process from gaseous mixtures (Nakhjiri et al., 2018a, 2018b).

ILs are known as the molten salts that their physical state is interpreted as a liquid at temperatures less than the value of 100 °C (Wilkes et al., 1981; Wilkes, 2007; Taghvaie Nakhjiri et al., 2022/04; Taleghani et al., 2021/04). These promising absorbents include cations and anions when no molecular solvent is presented. The first utilization of ILs was for military battery applications. Then after, ILs have shown their brilliant potential of application in disparate processes such as catalysts, lubricants, and gas separation using porous membranes (Torimoto et al., 2010; Welton, 2018). Walden was the pioneer of IL introduction over 100 years ago (Walden, 1914). Despite the discovery of this category of chemical absorbents wasn't of great attraction at the time, ILs have been of paramount attention in current decades owing to their superior characteristics. It is worth pointing out that the chemical characteristics of ILs may be modified by changing the functional groups, which eventuates in developing "task-specific" ILs for efficient separation of CO_2 molecules. One of the most important features of ILs is the fact that although they are liquid at room temperature, their vapor pressure and density is much lower than water (Lian et al., 2021; Giernoth, 2010; DavisJames, 2004). Fig. 1 schematically demonstrates ball-and-stick models of commonly-applied ILs in the field of CO_2 separation.

ILs as environmentally-friendly absorbents have recently been of paramount attention in the field of gas separation owing to possessing numerous functional/operational positive points including suitable chemical/thermal resistance against undesirable conditions, negligible saturated vapor pressure, great selectivity, tunable structure, and the need of lower energy in the regeneration step compared to benchmark alkanolamine absorbents (Gao et al., 2015/09; Fukaya et al., 2007; Nguyen et al., 2020). Fig. 2 schematically illustrates the CO_2 molecular separation through the CO_2-MEA-[Bmim]BF_4 system.

Numerous investigations have evaluated the feasibility of ILs application in CO_2 separation processes. Imidazole-/pyridine-based ILs have been the first investigated ILs for CO_2 separation (Yunus et al., 2012/05; Schuur et al., 2019; Dai et al., 2016; Babanezhad et al., 2020b). Bates et al. made an effort to develop "a task-specific" imidazolium ion-based IL to separate CO_2. According to their study, functionalized imidazolium ion-based IL could separate 0.5 mol CO_2/mol IL when the exposure of synthesized IL and CO_2 took place for 180 min, which was entirely the same as the MEA separation efficiency (Bates et al., 2002). Some studies in current decades have shown that the amino acid-based ILs comprising of several amine sites possess greater capability to separate CO_2 molecules compared to those ILs that have just one primary amine group owing to the mechanism of CO_2 separation by interaction with amine groups (Lian et al., 2021; Lv et al., 2016/03; Sistla and Khanna, 2015/08). As an example, the CO_2 solubility of 1.23 mol CO_2/mol ILs was obtained by Lv et al. via the simultaneous introduction of amine groups to the anion and cation of ILs (Lv et al., 2016/04).

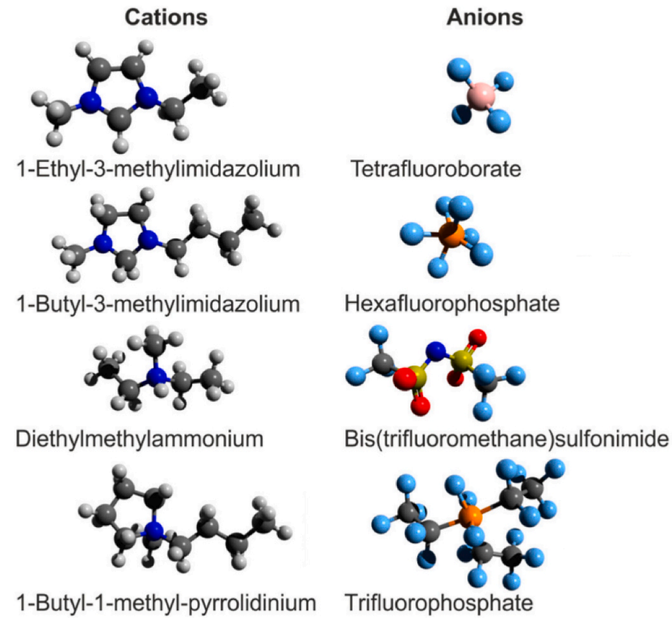


Fig. 1. Ball-and-stick models of commonly-applied ILs for CO_2 separation. Reprinted from (Rodenbücher et al., 2019).

Apart from the significant impact of functional groups, the important role of alkyl chain length and the anions on the CO_2 separation efficiency of ILs can't be ignored. Aki et al. conducted an experiment and corroborated that the increment of the alkyl chain length from butyl to octyl may possess encouraging influence on the solubility of CO_2 (Aki et al., 2004). Sharma et al. evaluated the effect of anions in the separation of CO_2 molecules in amino-functionalized ILs. Based on their investigation, the range of CO_2 separation using ILs including disparate anions was $BF_4^- < DCA^- < PF6^- < TfO^- < Tf_2N^-$ (Sharma et al., 2012/02; Sharma et al., 2012/06).

This paper aims to review the recent investigations on the potential use of IL-based absorbents to separate CO_2 from various types of industrial-based gaseous streams. To do this, a techno-economic study is aimed to be presented to compare the cost-effectiveness and efficiency of ILs and alkanolamine absorbents as the benchmark. Eventually, prominent environmental effects of the ILs applications in industries are discussed and future perspectives towards solving the operational challenges are presented in detail.

2. Techno-economic feasibility

From economic point of view, numerous investigations have predicted the average value of the CO_2 separation applying MEA alkanolamine solution in the range of 51–147 \$/tCO_2 for post-combustion process, 173 \$/tCO_2 for biogas upgrading and 27 \$/tCO_2 for natural gas sweetening (Akinola et al., 2019; Mota-Martinez et al., 2017; Shiflett et al., 2010; Mac Dowell and Hallett; Huang et al., 2014).

Despite superior capability of ILs to separate CO_2 molecules compared to other conventional absorbents, their high price has substantially confined their applications in industrial-based activities (Mota-Martinez et al., 2017; García-Gutiérrez et al., 2016; Mac Dowell and Hallett; de Riva et al., 2017; Palomar et al., 2019). Many researchers have recently investigated the techno-economic feasibility of CO_2 separation applying various types of ILs. For instance, de Riva et al. predicted an operation cost of 83 \$/tCO_2 for post-combustion CO_2 separation applying [Emim][NTf_2] IL (de Riva et al., 2017). Mota-Martinez et al. corroborated that the cheapest IL for efficient

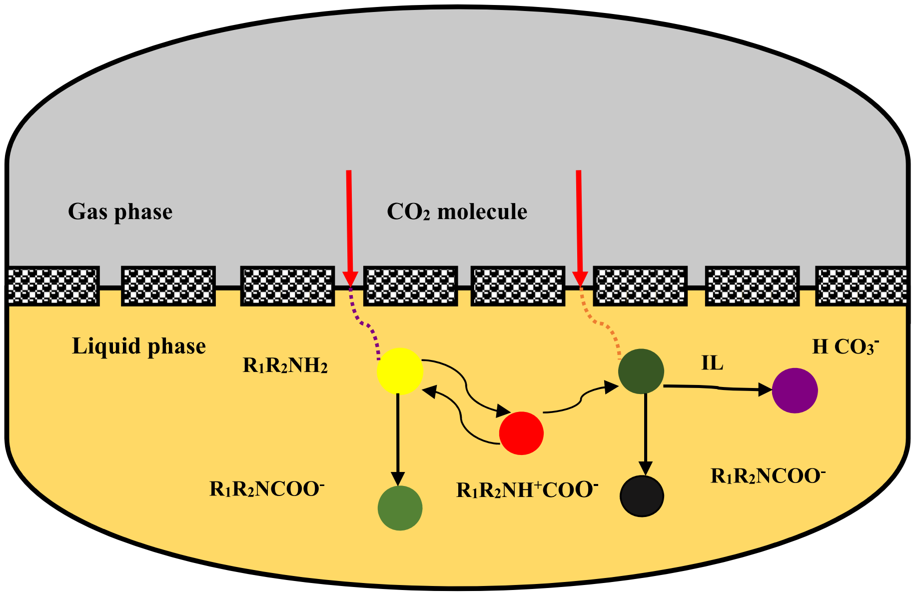


Fig. 2. Schematic demonstration of CO_2 molecular separation through the CO_2-MEA-[Bmim]BF_4 system. Reprinted from (Pishnamazi et al., 2020/09a) with permission from Elsevier.

separation of CO_2 would be [Emim][DCN] with total cost of 90 \$/tCO_2 (Mac Dowell and Hallett).

García-Gutierrez et al. techno-economically evaluated the efficiency of novel IL-based physical absorbing agents including [Emim][NTf_2], ([Hmim][NTf_2]) and ([P66614] [NTf_2]) for biogas upgrading (García-Gutiérrez et al., 2016). On the basis of their study, [Emim][NTf_2] was introduced as the most economical IL-based physical absorbent for CO_2 separation with a total cost of 271 \$/tCO_2. The combination of [Bpy] [BF_4], MEA and H_2O (30/30/40 wt%) was applied by Akinola et al. for CO_2 separation from natural gas. They corroborated the optimum cost of 25 \$/tCO_2 to obtain a cost-efficient separation process (Akinola et al., 2019). In addition, Huang et al. presented a study to assess the techno-economic feasibility of [Bpy][BF_4]-MEA system for separating CO_2 from flue gas. They have illustrated that the application of the above-mentioned absorption system significantly decreased the operation cost of the traditional MEA-based system from 70 to 60–62.5 \$/tCO_2 (Huang et al., 2014).

As explained above, application of IL-based absorbents as an alternative for conventional solutions in the CO_2 separation process possesses numerous positive points. Negative aspects of ILs such as solvent loss can be appropriately eradicated owing to the nature of the solvent. Additionally, the cost management during the separation of CO_2 from flue gas is optimistic (Zhang et al., 2016/01). Albeit some operational/functional problems of ILs must be solved before large-scale commercialization of CO_2 separation. Amino-functionalized ILs often possess greater viscosity, which eventuates in longer separation process (Bhattacharyya and Shah, 2016). Despite solving high viscosity problems of ILs via enhancing the water content, it can result in increasing the volume accordingly (Navarro et al., 2019/05; Ziobrowski and Rotkegel, 2017/05).

Recently, various analyses have been conducted applying various solutions such as alkanolamine absorbents, alkanolamine-IL mixtures or ILs in order to investigate the efficiency of CO_2 separation (Hospital-Benito et al., 2021; Ovejero-Pérez et al., 2021; Xie et al., 2018). Based on the knowledge of the authors, very few papers systematically investigate the application of disparate types of ILs for CO_2 chemical separation. Aspen Plus and Aspen Process Economic Analyzer (APEA) can be considered as two promising software to numerically simulate CO_2 separation efficiency and predicting their costs, respectively (Hospital-Benito et al., 2021; García-Gutiérrez et al., 2016; Ashkanani, 2021; Sultan et al., 2021; Babanezhad et al., 2020c). To study the techno-economic feasibility of using ILs in industrial plants, three CO_2 separation capacities including 1, 10 and 100 kmol/h were evaluated in a pilot plant, a small biogas upgrading plant and an industrial scale, respectively (de Riva et al., 2017; Ma et al., 2018; Ortloff et al., 2018). In this case, three ILs including [P2228][CNPyr], [P66614][CNPyr] and [Bmim][acetate] were investigated owing to their excellent efficiency for separating CO_2 compared to other ILs-based absorbents (Hospital-Benito et al., 2020).

APEA in integration with COSMO-based/Aspen Plus has recently been a prosperous methodology to predict the post-combustion, biogas and pre-combustion expenditure of CO_2 separation applying ILs absorbents (Hospital-Benito et al., 2021; Wang et al., 2021/06; Moya et al., 2022; Babanezhad et al., 2020d). It is perceived that the expenditure of the CO_2 separation process can significantly benefit from economy of scale. Application of [P2228][CNPyr] IL has significantly reduced the total annualized expense per metric ton of CO_2 in each process (post-combustion: 104.05 \$/tCO_2, biogas: 94.79 \$/tCO_2, pre-combustion: 93.86 \$/tCO_2 (Hospital-Benito et al., 2021). Management of cost can be considered as a vital key point to apply ILs as potential alternatives instead of conventional organic alcohol ammonia solutions for CO_2 separation. To investigate the techno-economic feasibility of ILs application as a chemical absorbent in CO_2 separation, Ma et al. compared the separation performance of CO_2 between ILs-based and an MEA-based processes. They resulted from their simulation that the ILs-based process was techno-economically more efficient for the CO_2 separation and sequestration and saved 30.01% of energy consumption and 29.99% in initial cost (Ma et al., 2018).

3. Applications of ILs in CO_2 separation and other industrial-based activities

The most attractive use of IL is as a chemical absorbent for separation of CO_2 molecules from disparate gases inside the hollow-fiber membrane contactors. HFMC is a relatively novel and promising membrane-based process, which can use the ILs as an efficient liquid absorbent (Nakhjiri and Heydarinasab, 2020a; Qazi et al., 2020/02). ILs demonstrate greater solubility rates to some gases such as CO_2, as a function of P and T (Yan et al., 2019; Zeng et al., 2017; Sasikumar et al., 2018; Zhang et al., 2017; Wu et al., 2020; Polat et al., 2020; Pishnamazi et al., 2020/09b). Table 2 aims to enlist prevalent use of ILs in disparate applications. ILs have been recently of great application for various sorts of

Table 2
Prevalent use of ILs in disparate industries.

Industrial-based application	Target	Positive points	Drawbacks	Ref.
Biomass processing	Absorbent	High efficiency Ability of application under the processing conditions	Difficulty of Operation in membranes	(Piemonte et al., 2016; Nguyen et al., 2022; Bakonyi et al., 2013)
Gas chromatography	Adsorbent	Separation of difficult-to-separate mixtures	Challenges of finding appropriate ILs	(Wlazło et al., 2016; Poole and Poole, 2011)
Gas separation	Absorbent	Selective separation of gaseous molecules	Large-scale industrial application is still ambiguous	(Zeng et al., 2017; Rashid, 2021; Liu et al., 2016; Hu et al., 2018; Friess et al., 2021; Cao et al., 2021b; Li et al., 2017; Yang et al., 2021)
Extraction	Chemical extractant	Excellent affinity for disparate natural/bioactive components	High cost High corrosion	(Berthod et al., 2018; Vidal et al., 2012; Nakhjiri and Roudsari, 2016; Cao et al., 2021c, 2022b)
Targeted drug delivery	Delivery agent	Safe/stable formulation	Hypersensitivity impact	(Zhuang et al., 1181; Shu et al., 2017; Monti et al., 2017)
Batteries and fuel cell	Electrolyte	High conductivity Non-volatile nature Very little corrosiveness	low ionic conductivity at room temperature	Mishra et al. (2020)
Solar panels	Electrolyte	Increment of efficacious charge transfer Preparation of low corrosive environment	The requirement of good sealing to prevent leakage	(Ghosh and Singh, 2019; Yu et al., 2017)

vital chemical reactions such as hydrogenations, biomass processing, hydroformylations, membrane-based separation and alkylation (Singhal et al., 2019; Yao et al., 2021; Swati et al., 2021; Babanezhad et al., 2020e). Nowadays, ILs have been of great attention to be extensively applied for the separation of different gases from industrial gaseous flows. The majority of industrial gases (i.e., CO_2, SO_2, NO_2 and H_2S) have acidic characteristics. Therefore, great motivations have been made towards studying the selectivity of ILs to separate acidic gases. Currently, Carvalho et al. experimentally studied the selectivity amounts of CO_2/CH_4 and H_2S/CH_4 gaseous mixtures using various ILs. They perceived from their study that the acidic gases/methane selectivity is in direct relationship with the polarity of ILs (Carvalho and Coutinho, 2011).

Mechanism of gas separation applying ILs may be physical or chemical, which profoundly relies on the connection between the structures of employed ILs and the separation performance (Dai et al., 2016; Babanezhad et al., 2020d; Budzianowski, 2015; Nakhjiri and Heydarinasab, 2020b). For conventional IL-based absorbents, the amount of CO_2 solubility is in direct contact to the cations/anions structures accompanying with CO_2-ILs interactions (Blanchard and Brennecke, 2001; Ramdin et al., 2015; Babanezhad et al., 2020f). The dissolution of CO_2 molecules in ILs can be controlled by interaction with anions of the ILs. On the basis of different reports, functionalization of conventional ILs considerably increase the separation efficiency of CO_2 molecules owing to improving physical interaction between ILs and gaseous molecules compared to non-functionalized ILs (Zeng et al., 2015). With the aim of obtaining better separation efficiency, the functionalization process of ILs is carried out via introduction of disparate reactive groups onto cation or anion. Better separation efficiency can be justified due to the chemical reactions between ILs and gaseous molecules (Ding et al., 2014; Rashid, 2021). Bates et al. offered amino-functionalized IL ([NH_2p-bim][BF_4]) for the first time, which possessed great efficiency to separate CO_2 (Bates et al., 2002). The mechanism of CO_2 chemical separation applying IL-base absorbents is attributed to the reaction of ILs with CO_2 and the formation of carbamate. Although initial endeavors were on the basis of the amino functionalization on the cation, investigators all over the world are attempting to functionalize anionic compartment of ILs to obtain equimolar separation of CO_2 (Gurkan et al., 2010).

An interesting use of ILs is the chemical synthesis of various materials. Application of ILs as an efficient chemical absorbent in disparate industrial-based activities has restricted the use of traditional organic solvents owing to having remarkable superiorities such as better reaction rates, negligible volatility, and excellent selectivity (Rashid, 2021; Ghandi, 2014; Lei et al., 2017). ILs possess noteworthy potential of utilization for various sorts of important chemical reactions such as hydrogenation, hydroformylation, isomerization and Diels-Alder reactions. Phase transfer catalysis and metal extraction can be identified as other applications of ILs-based chemical absorbents. ILs have significant capability to control the chemistry of reaction in both homogeneous/heterogeneous catalysis systems via stabilizing the ionic transition states (Babanezhad et al., 2020e; Marsh et al., 2004; Piemonte et al., 2016). Additionally, ILs can significantly increase the reaction rate in numerous catalytic reactions, especially microwave-/ultrasound-assisted processes. An attractive scientific area for ILs is electrolytes. The existence of brilliant advantages such as extensive electrochemical window, the feasibility of application in inorganic membranes, suitable conductivity, negligible dielectric constant, insignificant explosiveness and non-volatility has made ILs promising for use in batteries and fuel cells (Brennecke and Maginn, 2001). Application of ILs for targeted drug delivery can be an interesting field of study due to their brilliant characterizations to solve (at least decrease) the operational challenges related to the application of traditional organic solvents (i.e., poor drug solubility, toxicity and weak stability) (Zhuang et al., 1181; Adawiyah et al., 2016; Pedro et al., 2021; Marjani et al., 2020; Ghadiri et al., 2020/11). An attractive use of ILs is in the field of water treatment. The emergence of disparate noteworthy advantages including inherent non-volatility and low vapor pressure has increased the interest of scientists to use ILs as promising solvents and extractants in industrial water treatment processes (Khraisheh et al., 2021/09; Cao et al., 2022/07). Incorporation of ILs in solvent-based or membrane-based processes of water treatment caused a significant enhancement in the selective recovery of various organic contaminants like dyes, amines, herbicide, and phenolic compounds (Zhu et al., 2019/07; Nguyen et al., 2022). Fig. 3 schematically illustrates the application of ILs in supported liquid ionic membranes.

4. Environmenental impacts of ILs application

Existence of strong belief on the ecofriendly nature of ILs following with negligible toxicity, excellent biodegradability, and acceptable recyclability has motivated investigators to use them at laboratory and industry (Nguyen et al., 2020; Khraisheh et al., 2021/09; Kianfar and Mafi, 2021; Llaver et al., 2021). Recently, researchers have made disparate endeavors to evaluate the ILs greenness. Basic investigations have been conducted with the aim of studying the toxicity, biodegradability and bioaccumulation risk of ILs in the environment (Costa et al., 2017; Magina et al., 2021). Considering low vapor pressure, negligible

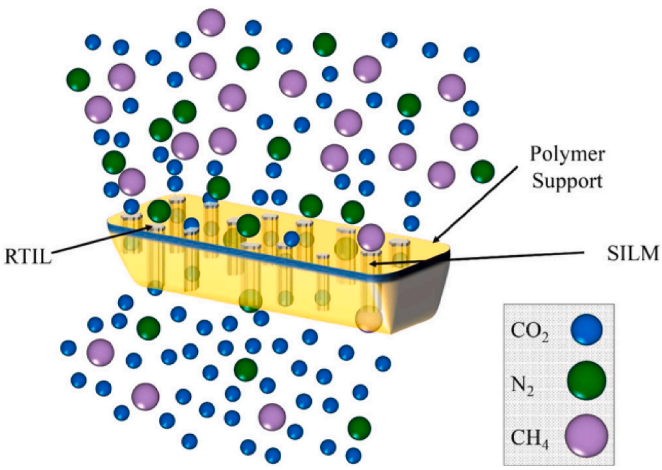


Fig. 3. Schematic demonstration of ILs application in supported liquid ionic membranes. Reprinted from (Shamair et al., 2020) with permission from Elsevier.

volatility and inflammability, ILs have been regarded as a safe and environmentally-friendly absorbent in comparison with traditional organic solvents and amine absorbents (Magina et al., 2021). Therefore, despite the existence of low concern about air pollution due to the gaseous release, this challenge must not be entirely ignored because of the feasibility of protic ILs distillation. Owing to the considerable solubility of ILs in water, these absorbents demonstrate great thermal/-chemical stability, which may result in environmental pollution during their release in wastewater sources. Hence, the ILs release through the soil or water is a serious environmental challenge that can be attributed to the dispersion of persistent contaminants to environment (Earle et al., 2006). Most ILs have demonstrated low biodegradability and therefore present an extensive range of toxicities. Thus, selection of more-environmentally friendly ILs for increasing the separation efficiency of GHGs is of great importance (Wu et al., 2018). For instance, great amounts of 1-octyl-3-methylimidazolium as a common type of an imidazolium-based IL has been found in the soil of Newcastle (a city in England), which has increased the risk of an auto-immune liver disorder (Leitch et al., 2020).

5. Future perspectives

As the future outlook, more fundamental investigations must be done to evaluate any feasibilities of photodegradation and biodegradation accompanying thermal degradation of ILs. Moreover, conduction of an R&D multi-scale approach is of great necessity to the advancement of low-cost technologies. Additionally, despite robustness of the recently-employed economic assessment models, development of faster and cost-effective economic assessment methodologies is of great importance to provide better opportunities to exactly estimate the techno-economic possibility of using ILs for separation of CO_2.

6. Conclusions

ILs are novel liquid absorbents that have been widely employed in CO_2 separation process. Summarization of numerous scientific investigations about IL-based absorbents concludes that these environmentally-friendly materials recommend a noteworthy platform for advancement of efficient membrane-based CO_2 separation with several operational, chemical and functional advantages. Although IL-based absorbents have illustrated excellent efficiency for CO_2 separation, alkanolamine absorbents still allocate more than 90% of the global market of CO_2 separation technology notwithstanding various operational/functional disadvantages like solvent loss and corrosion. An important obstacle towards the industrial use of ILs is trouble in the development of economical approaches for the separation of acidic contaminants particularly in the post-combustion flue gases. This review paper concludes that ILs can be introduced as a safe and environmentally-friendly alternative to toxic alkanolamine absorbents in GHGs separation industry. Selection of cost-effective IL can significantly increase the separation efficiency and decrease the environmental hazard compared to amine absorbents.

Credit author statement

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Declaration of competing interest

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

Data availability

No data was used for the research described in the article.

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