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Ionic liquid–formulated hybrid solvents for CO_2 capture

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Ionic liquids are believed to be a class of green solvents with potential applications in the post-combustion capture of CO_2. However, some inherent defects of ionic liquids, such as high cost and viscosity, significantly limit their industrial applications. Integrating ionic liquids with other materials represents a promising strategy to adjust the physical properties of ionic liquids and promote their real applications in industry. In this mini review, the progress to date in the fabrication of hybrid solvents based on ionic liquids for CO_2 capture was described. The advantages and disadvantages associated with these hybrid solvents were discussed. Future directions and prospects for CO_2 capture with ionic liquids were also outlined.

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Introduction

There is growing political and economic pressure to reduce emissions of CO_2 as a solution to the environmental and climatic issues caused by global warming [1]. Most CO_2 is emitted as a byproduct of burning fossil fuels, and fossil fuels will still be the major source of energy for industrial activity in the coming decades. This face imposes a critical emphasis on the post-combustion capture of CO_2 from industrial streams, for which aqueous organic amines are widely used [2]. Unfortunately, organic amines are highly volatile and corrosive, and the regeneration process is highly energy-intensive. These defects make it urgent to develop new materials that are green and sustainable to replace organic amines for CO_2 capture. Within this regard, ionic liquids (ILs) have attracted extensive attention owing to their *state-of-the-art* properties, such as wide liquid range, high thermal stability, extremely low volatility, and structural designability [3,4]. To date, great progress has been achieved in designing ILs for CO_2 capture [5–7]. However, the high cost and viscosity of ILs significantly limit their real applications in industry. Integrating ILs with other materials provides a platform for adjusting the physical properties of ILs and makes them more feasible for industrial applications. In this mini review, we summarized the most recent advances in CO_2 capture with ILs, mainly focusing on the fabrication of IL-based hybrid solvents for CO_2 absorption (see Fig. 1).

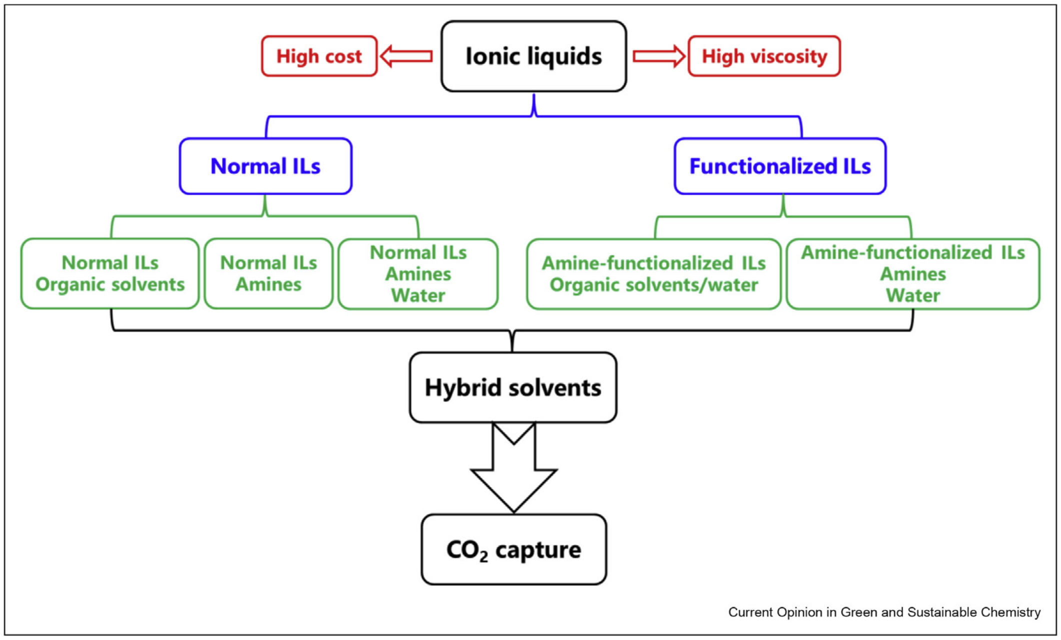
Normal IL-based hybrid solvents

Normal IL–organic mixed solvents

Initially, normal ILs were investigated as physical absorbents for CO_2 capture because of their relatively weak interaction with CO_2 [8,9]. In this case, ILs are more favorable for use in the low-temperature capture of CO_2 to offer high CO_2 capacity. However, the viscosity of ILs increases exponentially as the temperature decreases, which makes it difficult to employ ILs directly for the low-temperature capture of CO_2. Taking advantage of the low-viscosity of organic solvents and non-volatility of ILs, Lei et al. [10] proposed a binary mixture of methanol and 1-octyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([omim][Tf_2N]) for the capture of CO_2 at temperatures down to 228.2 K. This is a modification to the well-known Rectisol process, which uses methanol as the capture medium. Based on thermodynamic modeling with UNIFAC (UNIversal QUAsiChemical Functional-group Activity Coefficients) and process simulation with ASPEN Plus, a conceptual process was designed for the capture of CO_2 from syngas (see Fig. 2). It was found that the overall energy consumption, operating cost, and heat duty are reduced compared with the traditional Rectisol process.

Generally, physical absorption is more energy-efficient than chemical absorption because of the ease of desorption. Therefore, it is important to extend the work of Lei et al. [10] to the mixtures of many other ILs and organic solvents in the future to supplement the

Figure 1



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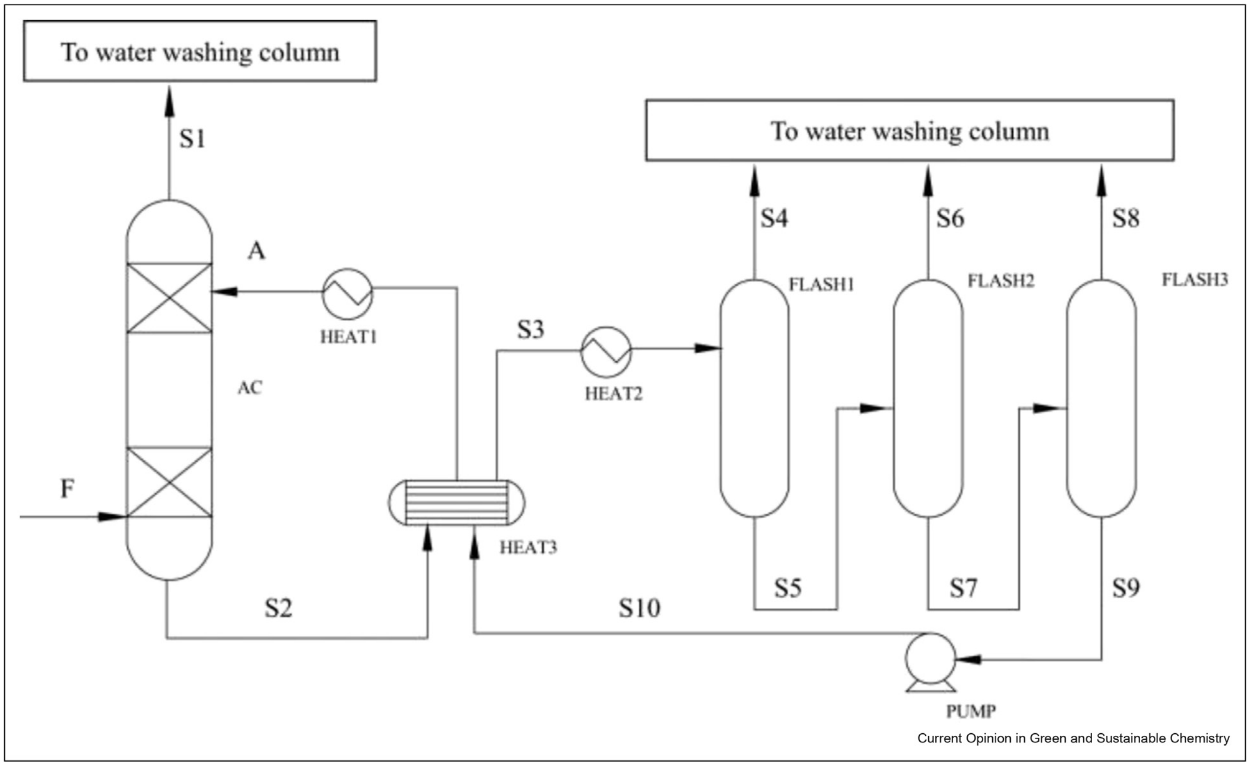
Ionic liquid–formulated hybrid solvents for CO_2 capture.

Figure 2



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A conceptual process for the low-temperature capture of CO_2 from syngas by the binary mixture of [omim][Tf_2N] and methanol. Reprinted with permission from Ref. [10]. Copyright 2015 Elsevier Ltd.

exploration of IL-based media for CO_2 capture. However, it is time-consuming to investigate all the possible mixtures by experiments. It is suggested that thermodynamic models and process programs be used first to screen promising candidates, and then experimentally validate the performance of optimized ones. The challenge is lack in the knowledge of the thermodynamic properties of ILs, which is necessary for the implementation of thermodynamic models and process programs.

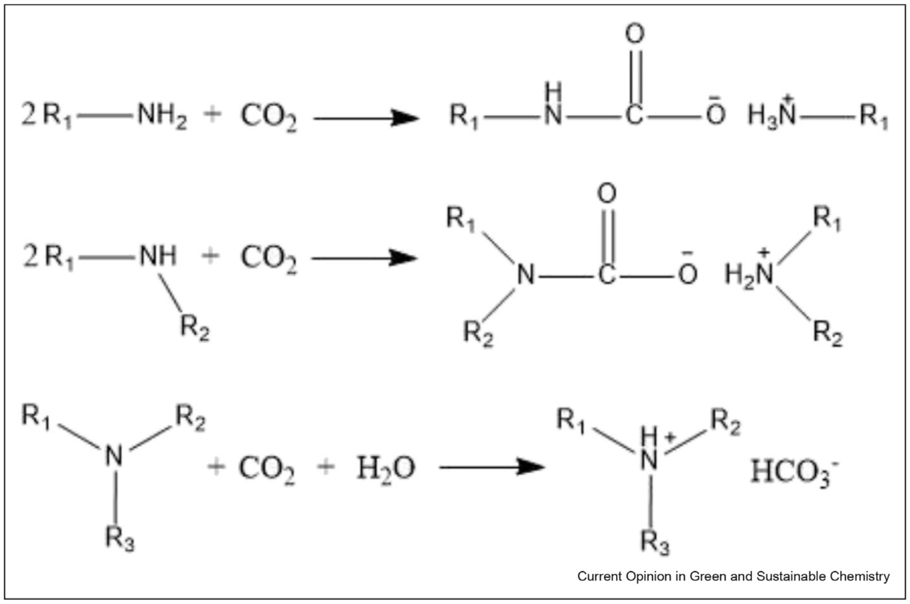
Normal IL-amine solutions

To tailor the properties of normal ILs and make them suitable for capture of CO_2 from dilute sources (e.g., flue gas), Noble et al. [11] dissolved commercially available MEA in hydrophobic 1-hexyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([hmim][Tf_2N]). Resultant mixtures are capable of the rapid and reversible absorb 1 mol of CO_2 per 2 mol of MEA to give an insoluble carbamate precipitate. The mechanism for reaction of amine compounds with CO_2 is shown in Fig. 3. The [hmim][Tf_2N]-MEA solution shows some significant advantages over traditional aqueous solutions of MEA for CO_2 capture: (1) the low heat capacity of ILs saves a considerable amount of energy during heat-induced desorption; (2) the evaporation of water solvents is efficiently avoided by using the extremely low-volatility ILs; and (3) the precipitation-assisted capture process drives the reaction of CO_2 with amine to offer both high CO_2 capacity and fast absorption rate and provides the possibility of separating the carbamate from the bulk ILs for energy-efficient desorption.

Siaj et al. [12,13] then systematically investigated the decomposition behavior of carbamate resulting from the reaction of CO_2 with diethanolamine (DEA) and 2-amino-2-methyl-1-propanol (AMP) in [hmim][Tf_2N] solutions. They found that the separation of carbamate as a solid phase offers a dual advantage: it requires less volume to regenerate and it narrows the temperature gap between CO_2 capture and amine regeneration. The same group further found that the inclusion of ILs helps suppress the corrosion of amines [14]. In light of the high cost of lithium bis(trifluoromethylsulfonyl)imide ($LiTf_2N$), a raw material for the synthesis of [Tf_2N]-based ILs, Yu et al. [15] studied the absorption capability of CO_2 in solutions of MEA in 1-ethyl-3-methylimidazolium tetrafluoroborate ([emim][BF_4]), 1-butyl-3-methylimidazolium tetrafluoroborate ([bmim][BF_4]), 1-ethyl-3-methylimidazolium hexafluorophosphate ([emim][PF_6]), and 1-butyl-3-methylimidazolium hexafluorophosphate ([bmim][PF_6]). They found that the CO_2 capacity in [emim][BF_4]-MEA and [bmim][BF_4]-MEA solutions is higher than that in [emim][PF_6]-MEA and [bmim][PF_6]-MEA solutions because the resulting carbamate is soluble in [PF_6]-based ILs.

In spite of the impressive features of normal IL-amine solutions, it remains a challenge to design a capture system in which gas–liquid absorption units and liquid–solid separation units can be effectively integrated. However, precipitated carbamate is an intermediate that can be catalytically converted to urea derivates [16], which are a class of important fine chemicals in industry. Transforming carbamate to urea

Figure 3



Reactions between amine compounds and CO_2.

derivates can eliminate the energy-intensive desorption process, as well as the transportation and storage of enriched CO_2 gas. Therefore, such a process intensified with both the capture and the conversion of CO_2 would be of great interest from an industrial perspective.

Aqueous normal IL + amine solutions

Aqueous solutions of MEA and activated aqueous MDEA solutions are the most widely used absorbents for CO_2 capture in industry. They can be modified by using normal ILs as additives to reduce the use of water, which is highly volatile and has a high heat capacity. Moreover, the excellent physical affinity of ILs with CO_2 is beneficial for the enhancement of CO_2 capacity. Li et al. [17] proposed to use aqueous [bmim][BF_4]+MEA solutions for CO_2 capture and found that the mixed absorbent shows better stability and antioxidant activity than do aqueous solutions of MEA. A kinetic study demonstrated that [bmim][BF_4] has an active effect on the hydration of CO_2 [18]. Zhang et al. [19] investigated the ability of different ILs – [bmim][BF_4], 1-butyl-3-imidazolium nitrate ([bmim][NO_3]), and 1-butyl-3-imidazolium chloride ([bmim][Cl]) – to tailor the performance of piperazine (PZ)–activated aqueous MDEA solutions for CO_2 capture. They found that aqueous [bmim][BF_4]+PZ + MDEA solutions show the largest CO_2 cyclic capacity. Calorimetric measurements and theoretical modeling revealed that the addition of [bmim][BF_4] can reduce the enthalpy of CO_2 absorption and reduce the sensible heat at 313 K by 30.1% and 20.3%, respectively.

Functionalized IL-based hybrid solvents

Organic/aqueous solutions of amine-functionalized ILs

Inspired by amine chemistry (see Fig. 3) [20], an array of amine-functionalized ILs has been specifically designed for CO_2 capture [21–23]. The highest CO_2 capacity has reached 1.69 mol/mol (13 wt%) in di(tributylethylphosphonium) iminodiacetate ([P_{4442}]$_2$[IDA]), a aminopolycarboxylate-based IL recently reported by our group [23]. However, amine-functionalized ILs suffer from a dramatic increase in viscosity after they are complexed with CO_2 [24]. A simple solution is to dilute amine-functionalized ILs with other solvents to form mixed absorbents. For example, Han et al. [25], Wu et al. [26], and Deng et al. [27] dissolved choline prolinate ([Ch][Pro]), diethylenetriamine hydrochloride ([DETA][*n*HCl]), and tetrabutylphosphonium glycinate ([P_{4444}][Gly]), respectively, in polyethylene glycol (PEG) for highly efficient and reversible capture of CO_2. Wu et al. [28], Zhou et al. [29], and Zhang et al. [30] investigated the absorption of CO_2 in a wide range of aqueous solutions of amine-functionalized ILs, including amino acid ILs and dual amine-functionalized ILs. These efforts demonstrated that the addition of dilute solvents helps enhance the adsorption/desorption rate of CO_2 and increases its loading capacity.

Subsequently, aqueous solutions of amino acid ILs were thoroughly studied as promising CO_2 absorbents because of their low viscosity and high capacity [31,32]. To provide a reliable basis for the process design, kinetic data for CO_2 absorption are necessary. To this end, Luo et al. [33] and Wu et al. [34] quantitatively measured the absorption rate of CO_2 in the aqueous solutions of some amino acid ILs, such as 1-ethyl-3-methylimidazolium glycinate ([emim][Gly]) and 1-butyl-3-methylimidazolium glycinate ([bmim][Gly]). The results showed that the absorption rate of CO_2 in aqueous solutions of amino acid ILs is slightly lower than that in aqueous solutions of monoethanolamine (MEA) because of the higher viscosity of amino acid ILs.

With the aid of quantum mechanical calculations, Yamada [35] compared the solvation effects on CO_2 absorption in aqueous solutions of MEA and [emim][Gly]. It was found that the dielectric constant of aqueous solutions of MEA decreases with an increase in CO_2 loading, which possibly contributes to the decrease in the CO_2 absorption rate. In contrast, the energy diagram of the reaction between CO_2 and [emim][Gly] barely depends on the solvation effect. As a result, the trade-off relationship between the absorption rate and absorption heat in the capture of CO_2 with aqueous solutions of organic amines [36] does not necessarily apply to aqueous solutions of amino acid ILs; this finding explains the superiority of aqueous solutions of amino acid ILs for CO_2 capture.

Besides PEG and water, other low-volatility and low-viscosity organic solvents such as sulfolane, propylene carbonate, *N*-methylpyrrolidone, and *N*,*N*-dimethylformamide can be used as dilute solvents for amine-functionalized ILs. Although they are well-known physical absorbents for CO_2 capture, their applications in the facilitated absorption of CO_2 in amine-functionalized ILs remain to be explored. Another promising alternative is to dilute amine-functionalized ILs with low-viscosity normal ILs. Since the heat capacity of ILs is much lower than that of water [37], using ILs in place of water as the bulk solvents for amine-functionalized ILs [38] can help reduce the energy penalty for desorption. Some preliminary work also investigated the absorption of CO_2 in IL solutions of 1-ethyl-3-methylimidazolium acetate ([emim][Ac]) [39,40], another type of functionalized IL that exhibits chemical reactivity with CO_2. Therefore, it is suggested to extensively investigate other organic solvents and normal ILs as dilute solvents for amine-functionalized ILs in the future.

Aqueous amine-functionalized IL + amine solutions

The reaction of CO_2 with primary or secondary amines is subject to 1:2 stoichiometry *via* the carbamate route; however, the reaction of CO_2 with tertiary amines in the presence of water is subject to 1:1 stoichiometry *via* the bicarbonate route (see Fig. 3) [20]. The reaction rate of CO_2 with tertiary amines is much slower than its reaction rate with primary or secondary amines. Therefore, primary or secondary amines are commonly used as activators for aqueous solutions of tertiary amines in the traditional amine-scrubbing process to offer both high CO_2 capacity and a fast absorption rate.

Inspired by this fact, Wu et al. [41] proposed to use amino acid ILs as the activators of aqueous solutions of methyldiethanolamine (MDEA) for CO_2 capture. The amine group of amino acid can react quickly with CO_2 to form zwitterions, which will transfer protons to MDEA, and the absorption rate of CO_2 is therefore increased greatly in comparison with aqueous solutions of MDEA (see Fig. 4). They suggested that tetramethylammonium glycinate ($[N_{1111}]$[Gly]) is highly attractive as the activator because of its low cost. Subsequently, the effects of $[N_{1111}]$[Gly] and MDEA concentrations on CO_2 capacity, regeneration performance, and absorption rate were investigated systematically [42,43]. Compared with traditional MEA-activated aqueous MDEA solutions, amino acid IL–activated aqueous MDEA solutions show higher stability during absorption-desorption cycles owing to the negligible volatility of the ILs, yet still exhibit comparable performance for CO_2 absorption and desorption.

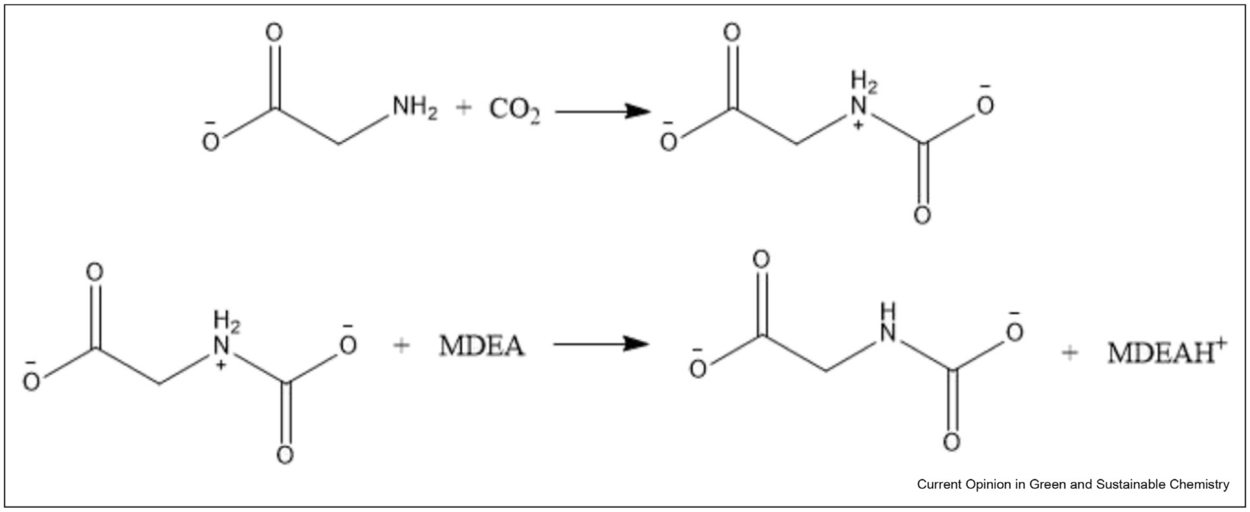
Based on the work of Wu et al. [41–43], Fu et al. [44] further evaluated the performance of [bmim][Gly]–activated aqueous MDEA solutions for CO_2 capture. They concluded that the optimized mass fraction of [bmim][Gly] is 0.10–0.15 and the total mass fraction of [bmim][Gly] + MDEA is ~0.5 when the absorption capacities, absorption rates, and viscosities of absorbents are comprehensively taken into account. Similarly, Jing et al. [45,46] found that the addition of $[N_{1111}]$[Gly] or 1-hexyl-3-methylimidazolium glycinate ([hmim][Gly]) into aqueous solutions of AMP enhances the reactivity of AMP with CO_2, and the kinetics region is considered to be a fast pseudo-first order.

Other functionalized ILs with chemical reactivity with CO_2, such as 1-hydroxyethyl-3-methyl imidazolium glycinate ([HOemim][Gly]) [47,48] and [emim][Ac] [49] were also investigated as additives to aqueous solutions of MEA for CO_2 capture. Those studies proved that the inclusion of functionalized ILs is a viable strategy to tailor the properties of aqueous solutions of amines and their performance for CO_2 capture.

Conclusions

In summary, the most recent progress in the fabrication of IL-formulated hybrid solvents for CO_2 capture was reviewed. Five classes of hybrid solvents were discussed: normal IL-organic mixed solvents, normal IL-amine solutions, aqueous normal IL + amine solutions, organic/aqueous solutions of amine-functionalized ILs, and aqueous amine-functionalized IL + amine solutions. Integrating ILs with other materials was demonstrated to be an effective method of overcoming the high viscosity of ILs and making them more applicable in industrial use. Obviously, IL-based hybrid solvents exhibit significant advantages over aqueous solutions of amines in many aspects, especially in reducing the volatile loss of solvents and reducing energy consumption.

Figure 4



Mechanism for the activation of MDEA by amino acid ILs.

Therefore, employing IL-based hybrid solvents for CO_2 capture is believed to be a green and sustainable approach. Although the high cost of ILs is still a challenge, the operation cost of CO_2 capture with IL-based hybrid solvents is expected to be much lower than the cost of the traditional amine-scrubbing process.

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