



Green solvents for CO₂ capture

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The development of specific 'green' solvents with unique combination of properties and associated techniques for target applications, capable of minimizing the environmental impact from their use in chemical production or by developing sustainable and renewable energy and resources, has been gathering increasing attention over the last years. Emerging alternatives or undervalued self-claimed greener solvents, such as ionic liquids, amino acid-functionalized ionic liquids, ionic liquid-mixed solvents, and eutectic solvents, have been proposed as promising materials with unique properties not achievable by means of any other material. Nonetheless, in spite of all these greener compounds' outstanding properties and potential for acid gas separation, important limitations have hampered the development of separation units and processes capable of fulfilling industrial demands. This study intends to carry a critical analysis upon the solvents proposed for CO₂ capture and the pursued technologies to make carbon capture, utilization, and storage a reality.

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Introduction

Concerns with increasing average global temperature due to CO₂ concentration in the atmosphere, with 36.8 gigatons released in 2017 alone, have spurred a worldwide debate on CO₂ emissions, ultimately leading to challenging climate frameworks. Even though gas separation has long been used in natural gas industry, because of the increased value of the posttreated gas, postcombustion is still seen as a nonefficient, costly, and, ultimately, unfeasible process [1–3]. Nonetheless, postcombustion technology is particularly appealing to tackle the climate framework targets because one can retrofit existing power plants, one of the main sources of anthropogenic CO₂ emissions. Several physical and chemical processes, based on absorption, adsorption, membranes, and cryogenic separation, are commercially available and widely used by the natural gas processing

industry [3]. However, when envisioning post-combustion, the current technologies, requiring large separation units and high CO₂ partial pressures, stand unfeasible for postcombustion processes. Thus, innovative postcombustion technological development for removing acid gases, envisioned using green solvents, is indispensable aiming at a clean energy production, anthropogenic CO₂ capture, transportation, and/or reconstitution into value-added products.

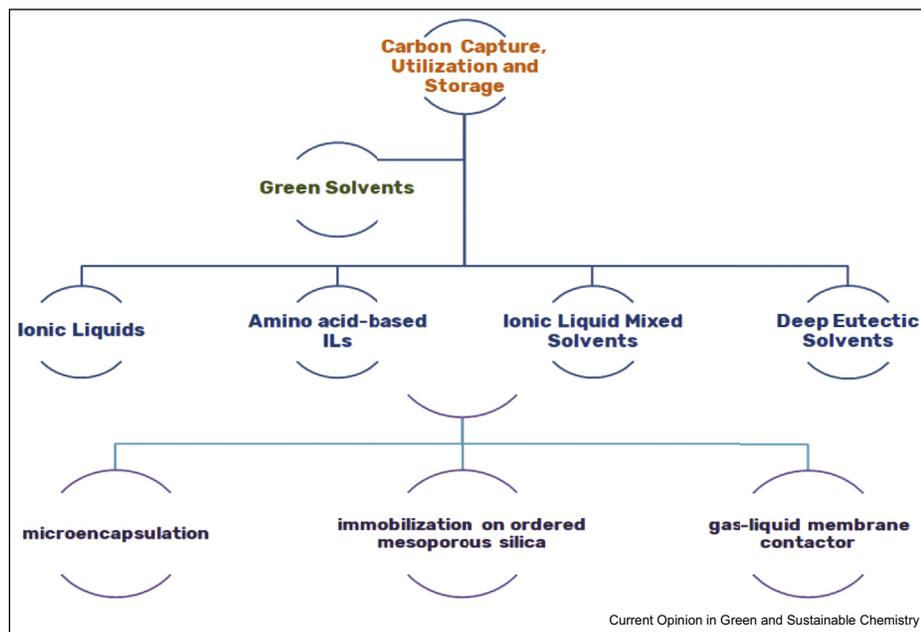
Although the idea of an 'ideal green solvent' suitable for all sorts of chemical transformations sounds unrealistic or even pretentious, the development of specific 'green' solvents with unique combination of properties and associated techniques for target applications, capable of minimizing the environmental impact from their use in chemical production or by developing sustainable and renewable energy and resources, has been gathering increasing attention over the last couple of years. Emerging alternatives or undervalued self-claimed greener solvents, such as task-specific ionic liquids (ILs), amino acid-functionalized ILs, IL-mixed solvents, and eutectic solvents, are promising materials with unique properties not achievable by means of any other material.

A number of interesting reviews have been published during the last few years addressing various aspects of CO₂ capture processes [2,4–6]. This study intends to carry a critical analysis upon the solvents proposed for CO₂ sorption and the pursued technologies to make carbon capture, utilization, and storage (CCUS) a reality (Figure 1).

Ionic liquids

ILs have been, at least for a certain time, considered as not only designer solvents but also green solvents mainly because their negligible vapor pressure, not contributing thus to the volatile organic compounds' problematic. But, often their synthesis is not green; many are badly biodegradable and show a significant toxicity [7]. Nonetheless, ILs are so 'species-rich', comprising a large set of chemically very different solvent and liquid classes with remarkably different characteristics—having in common the fact that all are salts and liquid at low to moderate temperatures—that similarly to the term 'green solvents', the term IL becomes almost meaningless. Nonetheless, ILs still retain the label of promising novel solvents for a wide range of potential applications; with their application as solvents for CO₂ capture processes still attracting most of the spotlight. Although most ILs, even those highly CO₂-philic, were

Figure 1



Schematic representation of the carbon capture, utilization, and storage. IL, ionic liquid.

shown not to be feasible solvents for CO₂ capture [6,8,9], either because of their similar sorption capacity to common organic solvents or to their high viscosity, leading to the design of unfeasible separation units, many authors have redirected their focus from a simple IL functionalization, such as fluorination of either the IL anion or cation [10,11], oxygenation of the anion [11], or the use of basic anions or cations, to explore mechanisms of enhancing the CO₂ chemisorption through the optimization of acetate-based ILs or the use of greener precursors such as fatty acids, carboxylic acids, and amino acids [12–15].

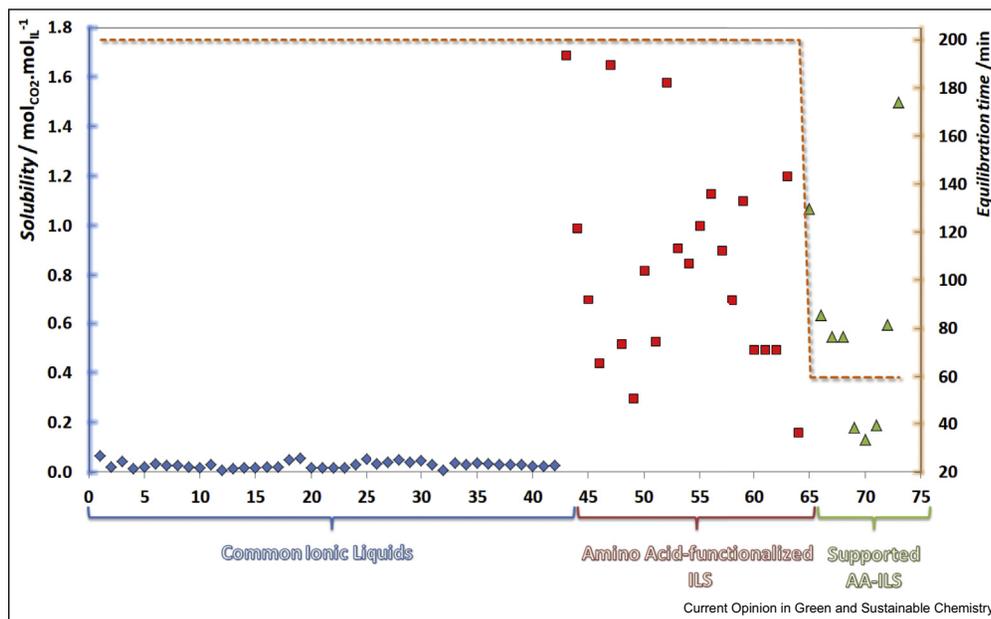
Amino acid-based ILs

Amino acid-based ILs (AA-ILs) are particularly promising because of their low cost, abundant availability, and nontoxic biodegradability [16]. However, although AA-ILs present high chemical absorption capacity [12,17] (Figure 2), their CO₂ absorption presents important kinetic limitations related to the high viscosity and low diffusion coefficients that result from the CO₂-complex and the salt bridge hydrogen-bonded network formed upon the reaction with CO₂ [18]. Aiming at circumventing these limitations different approaches have been proposed, such as the use of a cosolvent able to lower the mixture viscosity and improve mass transfer limitations, the use of a solid phase to immobilize the IL, and the use of the IL in membrane gas absorption processes [13,16,17,19–21]. Confining ILs in nanoporous matrices, with one or more of their spatial dimensions subjected to a geometrical restriction, results

in nanoconfined ILs with improved mechanical integrity and ionic conductivity that overcomes the major drawbacks of bulk ILs, such as high viscosity, slow gas diffusivity, and ultimately, requiring reduced quantities of IL to achieve the same separation [22]. Recently, the Palomar's group [17,23] developed a microencapsulation technique to encapsulate ILs in a hollow carbon submicrospheres, with a diameters between 400 and 700 nm, able to retain a large amount of IL (up to 80% in weight) allowing discretizing the IL fluid in microdrops, drastically increasing the gas–liquid contact surface and ultimately the sorption kinetics [17,23]. The authors reported a dramatic increase in the CO₂ absorption rates, maintaining the high absorption capacity reported for the neat IL [17]. Hiremath et al. [20] proposed a highly reversible CO₂ capture using AA-ILs synthesized from four different amino acid sources immobilized on ordered mesoporous silica. The authors reported fast kinetics (with equilibrium times lower than 30 min), moderate chemisorption, and an easy and low energy demanding desorption processes finely controlled by the suitable amino acid effective loading, temperature, and CO₂ concentration [20].

Membrane gas absorption provides a high specific surface area, independent controllable gas and liquid flow rates, a compact and energy efficient separation unit, and a linear scale-up design. On the other hand, on a gas–liquid membrane contactor, the absorbents allow high selectivity and a high mass transfer driving force. Lu et al. [21] proposed the use of a nondispersive

Figure 2



Carbon dioxide solubility and equilibration time of common ILs, amino acid-functionalized ILs, and amino acid-functionalized ILs supported in ordered mesoporous silica and hollow carbon microspheres [17,20,24]. IL, ionic liquid; AA-IL, amino acid-based ionic liquid.

sustainable absorption by a hydrophobic polypropylene microporous hollow fiber membrane contactor using the monoethanolamine glycinate amino acid-functionalized IL. The authors reported a high membrane flux and low gas outlet CO₂ concentration, highlighting the potential of the duality 'environmentally friendly and efficient CO₂ absorbent' and gas-liquid membrane contactor separation unit [21].

IL-mixed solvents

IL-mixed solvents stand as another approach to overcome the known limitations of ILs. Mixtures of ILs with water or organic solvents have been proposed as new absorbents for CO₂ separation [9]. At present, the research on ILs mixed solvents for CO₂ capture focus mainly on IL-water solutions, IL-alkanamine blends, IL-organic solvents, and IL-IL mixtures [13,14,25-27].

Although not a trivial task, owing to the strong dependence on the nature of the constituents, ions/compounds, several authors evaluated mixtures of ILs as a mechanism to increase the sorption capacity by manipulating specific interactions, excess volume, and viscosity. As reported by Moya et al. [28], mixtures of ILs with unfavorable intermolecular interactions, presenting, thus, positive deviations to ideality, show increased CO₂ solubility and favorable transport properties, with negative viscosity deviations, whereas mixtures with near-ideal behavior present averaged CO₂ absorption capacity of the neat ILs. Although the results denote new opportunities to design IL-based absorbents with better

balanced properties for CO₂ capture, its applicability is restricted to physical absorption [14,28,29]. Aiming at overcoming these limitations, IL-amine hybrid solvents have been proposed as energy-saving systems for CO₂ capture, compared with the conventional amine scrubbing process [14]. Ma et al. [30] reported IL-amine hybrid solvents composed of MEA (Monoethanolamine) with 1-butyl-3-methylimidazolium tetrafluoroborate and 1-butyl-3-methylimidazolium hexafluorophosphate with 24%–26% lower energy consumption. However, the amine and CO₂ reaction products impose such a high viscosity to the solvent that hinders pumping or precipitate them from the solvent. Furthermore, the formed carbamate promotes severe corrosion during the absorbent regeneration [12]. Focusing on overcoming the problems resulting from the amine and CO₂ reaction in mixed IL-amine solution and eliminating the energy requirements, compared with conventional amine scrubbing process, several authors, following the idea proposed by Bara et al. [31,32], evaluated the use of water as a cosolvent [14,26,27,33,34]. As stated by the authors, the IL-amine hybrid solvents have shown to be, through the manipulation of the IL concentration, an energy-saving system for CO₂ capture with reduced heat of absorption and manageable diffusivities and kinetics [14].

Aiming at enhancing the CO₂ chemisorption, through the optimization of acetate-based ILs, Gómez-Coma et al. [13] proposed the use of aqueous solutions of 1-ethyl-3-methylimidazolium acetate in nondispersive

absorption in a polyvinylidene fluoride hollow fiber membrane contactor. As reported by the authors, a mixture of 70% in volume of IL allows an overall mass transfer coefficient five times higher than that obtained for the neat IL, concluding that the use of aqueous solutions allows not only to overcome the known mass transfer limitations but mostly position the solvent as a competitive solvent when compared with traditional alkanolamine solvents [13].

Deep eutectic solvents

The concept of deep eutectic solvents (DESs), a mixture of a hydrogen bond acceptor (HBA) and a hydrogen bond donor (HBD), has surfaced recently as a new class of solvents with high potential for acid gas separation. Although the definition of the term deep, proposed by Abbott et al. [35], on most of the reported DES is questionable, the advantage of these eutectic solvents on many applications stands indisputable [36]. DESs have been proposed for CO₂ capture by Li et al. [37] back in 2008 using eutectic mixtures composed of choline chloride and urea. Since then, many evaluated other HBDs and HBAs, such as glycerol, ethylene glycol, ammonium based, phosphonium based and amine based, aiming at enhancing the CO₂ solubility [38–41]. Sze et al. [42] and Bhawna et al. [43] evaluated the DES basicity on the CO₂ sorption by preparing the DES with a superbase, aiming at promoting the formation of active alkoxide anions through the deprotonation of the HBD and HBA OH groups, reporting increased CO₂ capture. With the increasing number of DES evaluated, the concept of natural DES—DES composed of ‘natural’ or of ‘biological origin’ components—has been proposed as a greener class of a DES [44]. However, this term is somewhat misleading, because although most of the components of natural DESs occur in living things in many cases, they are chemically synthesized for the formulation.

Even though DESs have shown high potential for CO₂ capture, the number of experimental studies on gas solubility is remarkably limited to CO₂ and minor studies on SO₂ with studies on carbon capture from complex gas mixtures or selectivity nonexistent. Furthermore, on those available DESs, they are often mixed with a significant amount of water to, similar to the approach followed for the ILs, minimize energy requirements, heat of absorption, and mass transfer limitation [4,45,46]. Ma et al [46] and Sarmad et al [45] evaluated 35 DESs in terms of the influence of salt and HBD type and structure, as well their molar ratio on the CO₂ solubility and viscosity. As reported by the authors, stronger HBA–HBD intermolecular hydrogen bonds lead to lower CO₂ solubility, whereas increasing the alkyl chain length or the number of carbon atoms in the HBA or HBD leads to higher CO₂ solubility because of increased free volume within the DES. Among the DES evaluated, the authors highlighted 15 DES on the basis of glycerol-containing DES and fluorinated ILs,

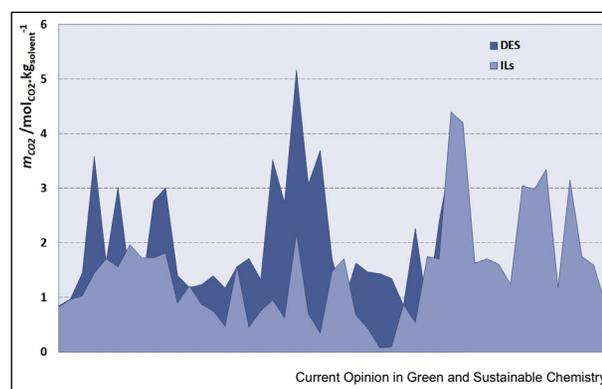
with viscosity lower than 200 mPa s, as those with the highest CO₂ absorption capacity. Although the CO₂ absorption reported by the authors stands within that reported for conventional ILs, as depicted in Figure 3, the reduction on the mixture viscosity allowed to achieve better kinetics of absorption. Furthermore, by adding water to the glycerol-based DESs, the authors were able to further improve the kinetics of absorption and enhance the mixture CO₂ absorption capacity [46].

Summary and future perspectives

Being an energy-intensive process, with almost 80% of the total costs for the whole CO₂ mitigation effort related to the capture process, the development of novel absorbent materials for high-performance post-combustion CO₂ capture stands vital on the pursuit for a sustainable use of fossil fuels in a low-carbon economy with decreasing greenhouse gas emissions. Emerging alternatives or undervalued self-claimed greener solvents, such as ILs, amino acid–functionalized ILs, IL-mixed solvents, and eutectic solvents, have been proposed as promising materials with unique properties not achievable by means of any other material.

The outstanding thermophysical properties of ILs have gathered a remarkable interest from both industry and academia. The possibility of fine-tuning the IL properties through suitable anion–cation combinations and from the possibility of developing task-specific ILs for target applications allowed to envision ILs as feasible solvents for CO₂ capture. Exploring mechanisms of enhancing the CO₂ chemisorption, the community proposed approaches ranging from the optimization of acetate-based ILs or the use of greener precursors such as fatty acids, carboxylic acids, and amino acids to IL-mixed solvents of IL–water solutions, IL–alkanolamine blends, IL–organic solvents, and IL–IL mixtures. DESs, a mixture of a HBA and a HBD, have surfaced recently as a new class of solvents with high potential for

Figure 3



Carbon dioxide molality in DESs (dark blue) and in conventional ILs (light blue). Data taken from Refs. [4,45,46]. IL, ionic liquid; DES, deep eutectic solvent.

acid gas separation. Although, DESs have shown high potential for CO₂ capture, the number of experimental studies on gas solubility is remarkably limited.

Nevertheless, in spite of all these greener compounds' outstanding properties and potential for acid gas separation, important limitations have hampered the development of separation units and processes capable of fulfilling industry demands. Academia tends to focus on enhancing equilibrium capacity, neglecting other properties, such as transport properties, that impose important penalties on process performance, and therefore on cost, that ultimately prove the process unfeasible. If one aims at developing a technical and economically viable process for carbon capture, the pursuit on improving CO₂ solubility, viscosity, and heat capacity must be addressed from a process engineering perspective. Leclaire and Heldebrant [2], in a recent 'call to arms' perspective review, call the attention to critical research needs, with emphasis on the principles of green chemistry and green engineering, to make CCUS a reality.

Aiming at enhancing absorption and desorption processes, several authors have evaluated the use of a solid phase to immobilize the solvents and the use of gas-liquid membrane contactors for gas absorption processes. Although confining solvents in nanoporous matrices results in improved mechanical integrity capable of overcoming the major drawbacks of the bulk solvents, such as high viscosity and slow gas diffusivity, the technology is still in an initial stage of development with major challenges to be addressed before their scale-up and ultimately industrial application. On the other hand, membrane gas absorption poses as a mature technology in which high specific surface area, independent controllable gas and liquid flow rates, compact and energy efficient separation units, and a linear scale-up design allow one to envision its use for carbon capture. Aiming at taking advantage of the membrane technology characteristics, several authors proposed the use of gas-liquid membrane contactors for the intended separation. The use of absorbents on a membrane contactor improves the separation selectivity and the mass transfer driving force, allowing high membrane fluxes and low gas outlet CO₂ concentration, highlighting the potential of the technology to take advantage of the green solvents discussed here, and making the CCUS a reality.

Conflict of interest statement

Nothing declared.

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