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EDITED BY

Eric C. D. Tan,
National Renewable Energy Laboratory (DOE),
United States

REVIEWED BY

Sean McCoy,
University of Calgary, Canada
Yuan Jiang,
Pacific Northwest National Laboratory (DOE),
United States

*CORRESPONDENCE

Burcu Gurkan
✉ beg23@case.edu

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Direct air capture of CO₂: from insights into the current and emerging approaches to future opportunities

Muhammad Zeeshan¹, Michelle K. Kidder², Emily Pentzer^{3,4},
Rachel B. Getman⁵ and Burcu Gurkan^{1*}

¹Department of Chemical and Biomolecular Engineering, Case Western Reserve University, Cleveland, OH, United States, ²Energy Science and Technology Directorate, Oak Ridge National Laboratory, Oak Ridge, TN, United States, ³Department of Chemistry, Texas A&M University, College Station, TX, United States, ⁴Department of Materials Science and Engineering, Texas A&M University, College Station, TX, United States, ⁵Department of Chemical and Biomolecular Engineering, Clemson University, Clemson, SC, United States

The rapid development of direct air capture (DAC) technologies has become critical in order to remove CO₂ from the atmosphere and limit global warming to a maximum of 1.5°C. In this perspective, we provide a mini review of the current research on the emerging liquid- and solid-based sorbent materials to capture CO₂, summarize the existing challenges of DAC technologies, and suggest future research directions to accelerate the development of DAC systems. In particular, the desired properties for a breakthrough sorbent that efficiently captures CO₂ from the air and releases it for sequestration are described.

KEYWORDS

eutectic solvents, ionic liquids, carbon capture, zeolites, metal organic frameworks, capsules, climate change, negative emissions

1. Introduction

The growing global reliance on fossil fuels for energy and material production has resulted in record-high carbon dioxide (CO₂) emissions in the atmosphere (Kumar et al., 2015; Sanz-Pérez et al., 2016). To mitigate the potential impact on climate change, ~10 gigatons of CO₂/year needs to be removed to limit global warming to a maximum of 1.5°C by 2050 according to multiple reports (World Resources Institute, 2002; Philander, 2012; Ozkan, 2021; IEA, 2022). Direct air capture (DAC), a process that captures CO₂ directly from air is critical to negative emissions technologies (NETs), and plays a significant role in achieving net zero emissions by the 2050 Scenario, among other CO₂ removal (CDR) approaches (National Academies of Sciences, 2019; IEA, 2021). According to the International Energy Agency (IEA) projection for achieving net-zero emissions by 2050 Scenario, the CDR removal target by DAC is ~980 Mt CO₂ per year (House et al., 2011). However, capturing CO₂, particularly from the air is a challenging and energy-intensive process, because CO₂ in the atmosphere is highly diluted, ca. 400 ppm (0.04%), when compared to the CO₂ concentration in a flue gas stream emitted from a power station, cement or natural gas plant (~4–15%). Further, a significant amount of CO₂ must be removed from the atmosphere and sequestered to have a quantifiable positive impact on the environment, and thus the technological challenge is formidable.

In this perspective, we provide an overview of the current approaches of DAC systems and emerging sorbent materials associated with CO₂ capture from dilute streams. Sorbents typically capture CO₂ at ambient temperatures and are regenerated by releasing the captured CO₂ at elevated temperatures (e.g., by applying convective heat) or using alternative heat sources (e.g., microwave) (Gomez-Rueda et al., 2022), as well as non-thermal techniques such as electrochemical (Stern and Hatton, 2014; Voskian and Hatton, 2018; Rahimi et al., 2020), ultrasound (Ying et al., 2014) and pressure-swing (Wiheeb et al., 2016). Lackner and co-workers introduced a moisture-swing process for DAC where the sorbent absorbs CO₂ when dry and releases it when wet (Lackner, 2009; Wang T. et al., 2011; Shi et al., 2020). Sorbents can be liquid, solid, or a hybrid material that has both liquid-like and solid-like components. The development of sorbents requires the consideration of numerous performance parameters, such as high CO₂ capacity, selectivity, fast sorption/desorption kinetics, and chemical stability under variable temperature and humidity. Furthermore, industry-related challenges such as the operational cost and energy demand can only be addressed when an efficient sorbent material is integrated into a DAC system. Thus, a molecular-level understanding of sorption kinetics, thermodynamics, and structure-property relations are imperative for the discovery of a breakthrough sorbent. Further, research focusing on alternative regeneration techniques is essential to reduce the overall energy consumption and cost of CO₂ removed, i.e., joule per ton of extracted CO₂ in a DAC system.

2. Current status and challenges

In general, CO₂ capture or removal from a mixture stream process includes adsorption, absorption, and/or membrane-based separation. Adsorption and absorption-based processes are typically considered more promising for directly capturing of CO₂ at ultradilute conditions (McQueen et al., 2021). In an absorption-based process, CO₂ dissolves in a liquid solvent such as an aqueous amine, aqueous sodium hydroxide, ionic liquid (IL), or a mixture of solvents via physisorption or chemisorption (Mahmoudkhani and Keith, 2009; Kumar et al., 2020). Alternatively, in an adsorption-based process CO₂ is chemisorbed or physisorbed onto a solid surface that has chemical functionality for CO₂-philicity, such as in the case of zeolites, metal organic frameworks, or covalent organic frameworks (Choi et al., 2011). CO₂ binding processes are exothermic in nature and requires active cooling to maintain CO₂ capacity. This challenge is mitigated to some extent due to distributed heat of the solid surface material and the density of sites in adsorption. Thermodynamically, the affinity between the sorbent and the target gas molecule is quantified through the heat of absorption or the isosteric heat of adsorption (Builes et al., 2013). Recent studies comparing the energetics of temperature swing vs. pressure swing CO₂ separation processes for a generic adsorbent (with a heat of adsorption of -65 kJ/mol) found the temperature swing adsorption to be more efficient under dilute CO₂ concentrations (i.e., 50% removal from feed and 95% product purity) (Lackner, 2013; Lively and Realf, 2016). Conversely, pressure swing adsorption is more efficient for bulk gas separations,

as pressurizing the inlet feed with low CO₂ concentration makes the process energetically costly.

Even with temperature swing processes, both adsorption and absorption processes consume large amounts of thermal energy during the sorbent regeneration, in addition to the energy required to move air through the sorbents. In particular, the trade-off between the strength of CO₂ molecular interactions with the sorbent and the required regeneration energy remains a challenge. An aqueous solution of calcium hydroxide is an effective sorbent for capturing CO₂ from the air due to its high affinity to CO₂. For example, pilot plant of Carbon Engineering has the capacity to remove 1 Mt of CO₂/year (requires 8.81 GJ per ton of CO₂ removed) by using an aqueous-sorbent (Keith et al., 2018). First, air is passed through a series of filters to remove any particles and pollutants. Next, the purified air flows into a contactor, a container filled with a solution of potassium hydroxide (KOH). As the air passes through the contactor, the CO₂ in the air reacts with KOH to form potassium carbonate (K₂CO₃) and water. The aqueous solution is then regenerated for reuse by heating at 900°C to release the captured CO₂. Table 1 summarizes the key comparisons for DAC plants around the world, including Carbon Engineering (Ozkan et al., 2022).

In contrast to absorption and adsorption processes, membrane-based CO₂ separation works on the principle of preferential permeation, where the target gas molecules diffuse across the membrane film under isothermal conditions. The gas molecule can permeate through the membrane via different mechanisms including size sieving, solution diffusion, surface diffusion, ion and facilitated transport. For CO₂ separations from air using a membrane, combination of these diffusion mechanisms is needed since there is a lack of high-pressure driving force to transport CO₂ across the membrane by solution diffusion alone. Few recent studies explored facilitated transport membranes for DAC (Lee and Gurkan, 2021; Matsuoka et al., 2021; Nabity et al., 2021; Lee et al., 2022). Lee et al. (2022) prepared a thin film of poly(ionic liquid) (PIL)-IL impregnated graphene oxide membrane supported on a poly(ethersulfone)/poly(ethylene terephthalate) substrate, and reported excellent CO₂ permeance of 3,923 GPU and CO₂/N₂ selectivity of 1,200 under 410 ppm CO₂ with a 1 bar feed gas (CO₂/N₂/H₂O mixture) at 40% RH and 22°C. In this example, helium was used as the sweep gas on the effluent side which limits the purity of the separated CO₂ and the post-process application. For post-sequestering purposes, higher purity CO₂ is needed and the study showed the separation performance of the membrane decreases under vacuum conditions on the effluent side due to the need to reinforce the membrane further. However, the purity requirement does not have to meet prior targets set (i.e., > 95%) for CO₂ removal from post-combustion flue gas. While the recent reports are encouraging to further develop facilitated transport membranes for DAC, they also suggest the need of an integrated approach based on multiple membrane modules or integration with other separation units. Another emerging approach alternative to temperature swing processes is the Faradaic electro-swing process developed by Hatton and co-workers where the CO₂ binding is regulated by electroactive species [e.g., amine sorbents (Wang M. et al., 2020) and quinone (Gurkan et al., 2015) carriers] (Voskian and Hatton, 2019). Recently, they demonstrated a bench scale,

TABLE 1 Comparison of CDR metrics of current and future planned (*) DAC plants around the world.

Company (plant type)	Location and country	Sorbent type	Regeneration temperature (°C)	CO ₂ removal capacity (tCO ₂ /year)	Thermal energy (GJ/tCO ₂)	Estimated cost (GJ/t CO ₂)
Carbon Engineering (2023) (Pilot)	British Columbia, Canada	Liquid	900	350	~8	500–600
Carbon Engineering* (Commercial)	Texas, USA	Liquid	900	1,000,000	N/A	94–232 (Keith et al., 2018)
Climeworks (2023) (Pilot)	Zurich, Switzerland	Solid	80–100	900	~7	600
Climeworks (Commercial)	Hellisheidi, Iceland	Solid	80–100	4,000	N/A	600 (Giving Green Earth, 2023)
Climeworks (Pilot/Commercial)	Europe	Solid	80–100	2,000	N/A	600
Global Thermostat (2023)* (Commercial)	Oklahoma, USA	Liquid	100–120	4,000	N/A	50
Infinitree LLC (2023) (Pilot)	New York, USA	Solid (Ion exchange)	Humidity Swing	100	N/A	N/A

The * symbol indicates future planned. The N/A stands for not available.

solid-state Faradaic electro-swing reactive adsorption unit that achieved > 90% Faradaic efficiency with a CO₂ feed < 0.1%, demonstrating a work of 40–90 kJ per mole of CO₂ captured, representing favorable thermodynamics comparison to most other thermally regenerated adsorbents (Voskian and Hatton, 2018).

In the following sections, we focus on the conventional and emerging sorbents based on absorption and adsorption-based processes that have demonstrated promising CO₂ capture capabilities at ultradilute (400 ppm) conditions.

2.1. Conventional sorbents for CO₂ capture

To date, most of the research on DAC has been focused on hydroxide- or amine-based aqueous sorbents due to their efficacy in capturing CO₂ under ultradilute conditions (Sanz-Pérez et al., 2016; Sodiq et al., 2023). The hydroxide based solvents need 900°C for regeneration with energy requirement of 6–9 GJ/tCO₂—equivalent to the energy utilized by an average household in the US for 2–3 months (Baciocchi et al., 2006; National Academies of Sciences, 2019; McQueen et al., 2021; Lebling et al., 2022). Besides this substantial energy requirement, sorbent loss incurred during the sorption/desorption process via evaporation and degradation is of significant concern. Further, water loss in the air contactor of solvent-based carbon capture systems can also significantly limit their deployment and technomic effectiveness, particularly in dry climates (Rosa et al., 2021; An et al., 2022). On the other hand, porous solid-supported amines present higher stability under moisture in ambient air, lower heat capacities (≈1–1.5 J/g K compared to the CO₂-loaded monoethanolamine ≈4 J/g K) (Weiland et al., 1997; McQueen et al., 2021), and mild regeneration temperatures (≈50–120°C) (Wang et al., 2015). However, when compared to liquid sorbents, amine supported solid sorbents are limited by their slow sorption kinetics, limited CO₂ capacities at low partial pressures, and poor cyclability (absorption-desorption cycles with maintained capacity) (Wang Q. et al., 2011; Lai et al.,

2021). The following sections discuss the improvements made over conventional sorbents in consideration of these challenges.

2.2. Emerging liquid sorbents

Aqueous amino acids have been investigated for carbon capture, which are relatively non-volatile, environmentally friendly, and regenerable using mild heating (≈100°C). For instance, Custelcean and co-workers reported amino acid-based sorbent with a capacity of 0.7 mol of CO₂ per mol of aqueous solution at ambient air conditions (Custelcean et al., 2019). The CO₂-saturated bicarbonate species in the solution were then crystallized using a 2,6-pyridine-bis(iminoguanidine) (PyBIG) to form a solid hydrated carbonate in an aqueous solution. Heating the solid carbonate produced at a comparatively mild temperature (120°C) removes most of the bound CO₂ and water via the reaction mechanisms shown in Figure 1A. In general, amino acids with a highly basic functional group are desirable for CO₂ capture (Ramezani et al., 2022), but the reported amino acids present lower cyclic capacities typically in the range of 0.12–0.4 mol CO₂/mol sorbent (Recker et al., 2022).

Researchers have also explored the use of amino acid derived ILs as an alternative liquid sorbent for CO₂ capture. ILs are unequivocally versatile materials for CO₂ capture with negligible volatility and high chemical stability (Zhang et al., 2012). For instance, Gurkan et al. synthesized an amino acid-based IL, trihexyl(tetradecyl)phosphonium proline, ([P₆₆₆₁₄][Pro]), which exhibited a CO₂ capacity of ~0.9 mol CO₂/mol IL in low partial pressure region at 22°C (Gurkan et al., 2010a). A significant disadvantage of amine-functionalized ILs for CO₂ capture application is their high viscosity leading to slow sorption kinetics. In this regard, Schneider, Brennecke, and co-workers developed aprotic heterocyclic anion (AHA) ILs for CO₂ capture and showed that the viscosity of ILs remained unchanged before and after the CO₂ absorption while maintaining a 1:1 CO₂ sorption

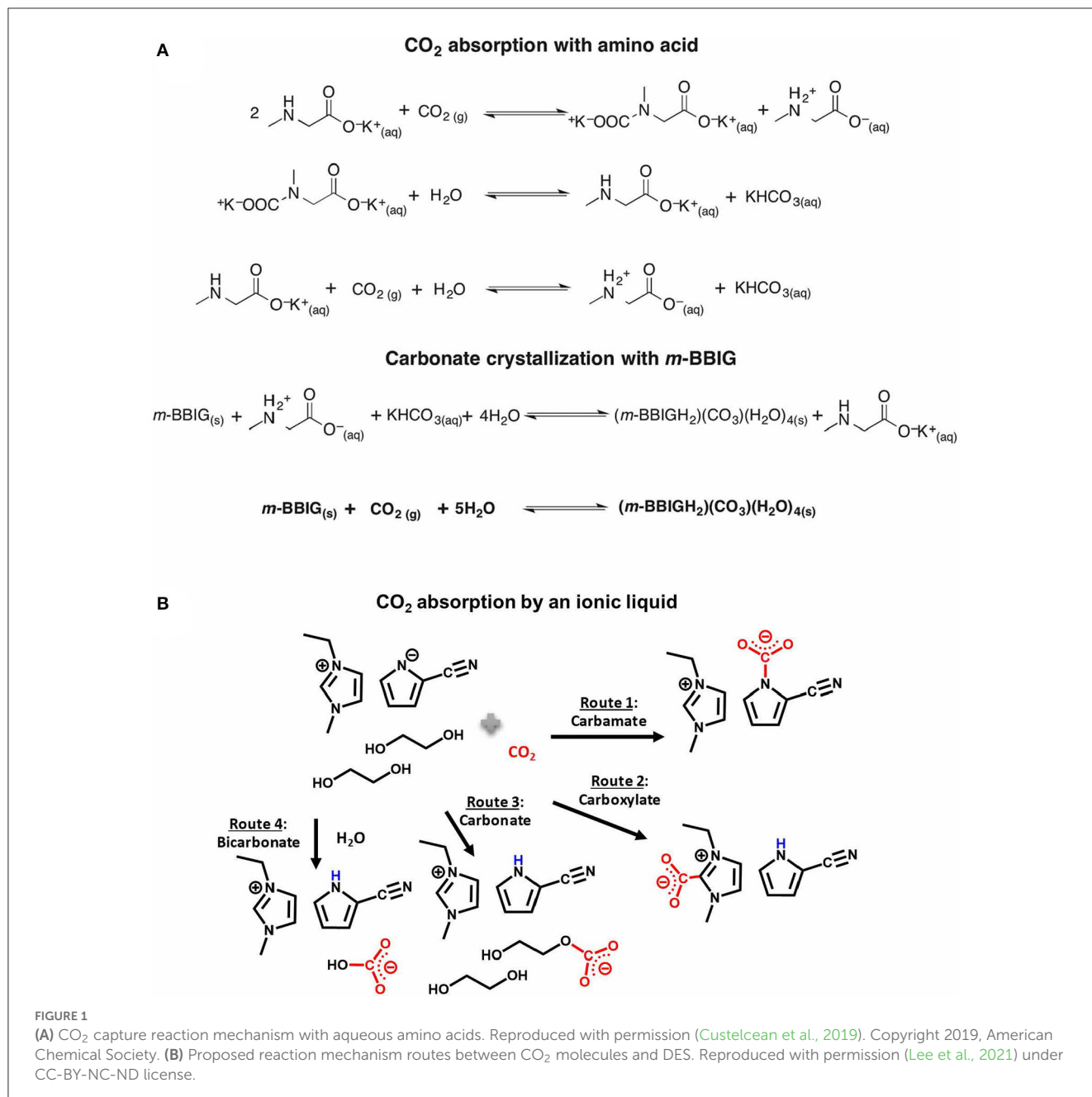


FIGURE 1

(A) CO₂ capture reaction mechanism with aqueous amino acids. Reproduced with permission (Custelcean et al., 2019). Copyright 2019, American Chemical Society. (B) Proposed reaction mechanism routes between CO₂ molecules and DES. Reproduced with permission (Lee et al., 2021) under CC-BY-NC-ND license.

stoichiometry (Gurkan et al., 2010b; Seo et al., 2014). In addition, the reaction enthalpy (~ 50 kJ/mol) was lower than conventional amines such that regeneration was possible at temperatures < 100°C, giving a reasonable working capacity (capacity difference between absorption and desorption). However, the demonstrated AHA ILs had low gravimetric CO₂ capacities (~9%) due to their large molar mass. Although there has been extensive research on the use of ILs for capture and utilization of pure CO₂ (Aghaie et al., 2018; Shukla et al., 2019), studies on their application in DAC have been limited (Yang and Dai, 2021).

Similar to ILs, deep eutectic solvents (DESs) are known to have significant physisorption capacity for CO₂ (García et al., 2015; Trivedi et al., 2016). DESs are mixtures of two or more components that form stable, low-melting-eutectics and are typically composed

of a hydrogen bond acceptor, such as a halide salt, and a hydrogen bond donor (Smith et al., 2014; Hansen et al., 2021). Recent studies demonstrated functionalized DESs and eutectics that demonstrate CO₂ chemisorption capacity, similar to amine functionalized ILs (Zhang et al., 2018; Yan et al., 2020; Lee et al., 2021; Klemm et al., 2023). Lee et al. reported a functionalized DES for CO₂ capture that overcomes the limitations of traditional ILs, such as high viscosity and poor gravimetric CO₂ capacity at low partial pressures (Lee et al., 2021). In that study, a reactive IL, 1-ethyl-3-methylimidazolium 2-cyanopyrrolide, was used as a hydrogen bond acceptor component and ethylene glycol (EG) was the hydrogen bond donor. The 1:2 molar mixture of IL:EG showed a capacity of 0.85 mol CO₂/mol solvent at 1 bar of CO₂ and 0.31 mol CO₂/mol solvent at 410 ppm of CO₂ at 25°C. Here, it

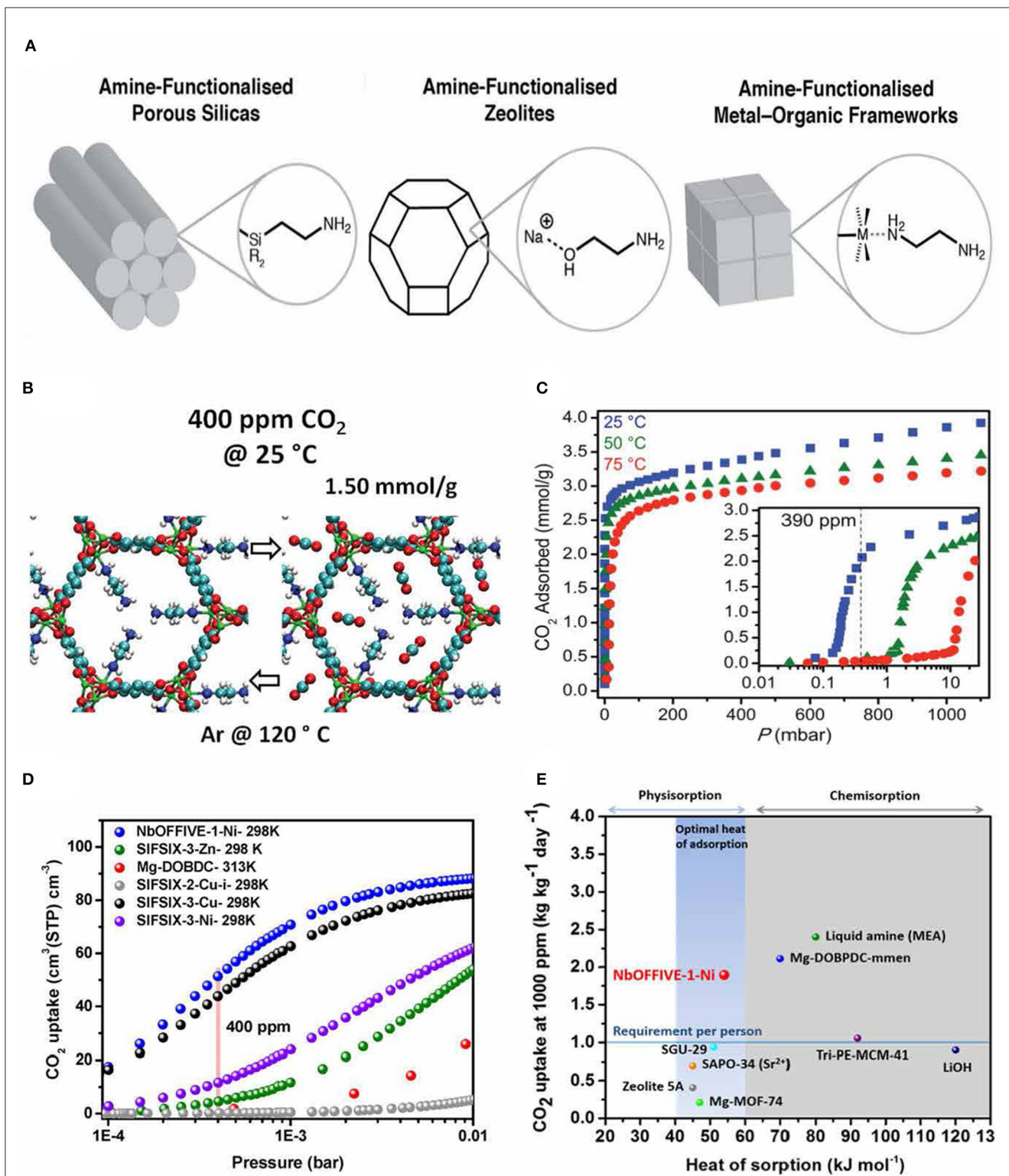


FIGURE 2
 (A) Schematics of amine-functionalized solid adsorbents for CO₂ capture. Reproduced with permission (Forse and Milner, 2021). Copyright 2021, Royal Society of Chemistry. (B) CO₂ uptake of MOF (Mg/DOBDC) under ambient air conditions. Reproduced with permission (Choi et al., 2012). Copyright 2012, American Chemical Society. (C) Adsorption of CO₂ in MOF [Mg₂(dobpdc)] in the pressure range of 1–1,000 mbar. Reproduced with permission (McDonald et al., 2012). Copyright 2012, American Chemical Society. (D) CO₂ uptake of NbOFFIVE-1-Ni MOF at low pressure and 25°C and (E) CO₂ heat of adsorption in comparison with other benchmark materials. Reproduced with permission (Bhatt et al., 2016). Copyright 2016, American Chemical Society.

is worth noting that the gravimetric CO₂ capacity of the DES (2.7 mol CO₂/kg sorbent) was higher than that of the neat IL (2.1 mol CO₂/kg sorbent) at 410 ppm CO₂. This was attributed to carbonate formation with the deprotonated EG, in the presence of the pyrrolide anion, which was stabilized by the hydrogen bonding network (Figure 1B). This study demonstrates that while the H-bonding network in DESs may increase viscosity, it can also modulate the CO₂ interactions with the sorbent, leading to an alternative reaction route as compared to neat IL. To tune functionalized DESs for DAC beyond the capture capacity, thermal and oxidative stability upon cycling must be established, since some of the parent compounds are volatile (unlike ILs).

2.3. Emerging solid sorbents

Traditional amine functionalized sorbents such as zeolites (Fu et al., 2022), mesoporous silicas (Shi et al., 2020) (Figure 2A), and moisture-swing resins/polymers (Wang et al., 2017; Wang T. et al., 2020) have been investigated for CO₂ capture, yet they have considerable shortcomings for DAC. This includes low CO₂ capacities, slow kinetics, diffusion-limited sorption, small surface area, and poor multi-cycle capacity, all of which significantly impact the sorbent material's overall performance. In this context, MOFs are considered superior to conventional solid sorbents because of their highly accessible surface area and adjustable chemical functionality (Sadiq et al., 2020). Choi et al. (2012) developed ethylene diamine (ED) modified ED-Mg/DOBDC (Figure 2B), which showed a capacity of 1.5 mmol/g under ambient air conditions together with excellent thermal stability and regenerability. The amine groups grafted on the open metal sites introduced additional sites for chemisorption, which improved the CO₂ capacity compared to the parent MOF (1.35 mmol/g). Similarly, McDonald et al. (2012) demonstrated that alkylamine-loaded Mg₂(dobpdc) have exceptional CO₂ uptake capacity (2 mmol/g) under 390 ppm at 25°C (Figure 2C). This improvement in the CO₂ uptake at very low partial pressure was ascribed to the interaction of electrophilic carbon of CO₂ with the nitrogen electron pair in diamine.

Eddaoudi and coworkers synthesized a MOF, NbOFFIVE-1-Ni, which exhibited excellent CO₂ sorption capacity (1.3 mmol/g) under 400 ppm CO₂ at 25°C due to the favorable interactions of CO₂ with the fluorine centers of the MOF, with a regeneration energy of 54 kJ/mol (Figures 2D, E) (Bhatt et al., 2016). Alternatively, Darunte et al. illustrated that the CO₂ capacity of a simple MOF [MIL-101(Cr)] could be improved by incorporating amine molecules into the MOF pores (Darunte et al., 2016). Accordingly, tris(2-amino ethyl) (TREN)-loaded MIL-101(Cr) exhibited eight-times higher CO₂ capacity (2.8 mmol/g) than the corresponding pristine MOF (0.35 mmol/g) at 0.4 mbar (400 ppm CO₂ in He) and 25°C. The composite sample, however, showed a significant loss in cyclic capacity due to the excessive amount of TREN loading (more than the available unsaturated coordination sites). Therefore, future research on MOFs for DAC should focus on stability under various temperature and humidity conditions, cycling capacity, high CO₂ selectivity, and cost-efficient scalability.

3. The way forward: key matrix for a breakthrough sorbent

Fundamental studies aiming to develop structure-property-performance relations are still needed to further develop sorbents for widespread application in DAC. In addition, computational studies, for example utilizing machine learning, are needed to accelerate sorbent discovery (Mohan et al., 2022). For example, simulations can provide insight into material properties that maximize CO₂ sorption (Guan et al., 2022) as well as CO₂ binding sites and their corresponding thermodynamics and kinetics (Qazvini et al., 2021; Ahmad et al., 2022). In the following sections, we discuss the significance of individual properties of the sorbents that make up the key matrix for a breakthrough sorbent which would significantly expedite the development of DAC technologies to help meet the global goal of decreasing atmospheric CO₂ levels.

3.1. Sorption, kinetics, and diffusion

Sorbents with high basicity can increase affinity to CO₂ which is an acidic gas. However, strong binding of CO₂, such as with primary and secondary amines relative to tertiary amines for example, also increases the overall energy required to regenerate the sorbent. Therefore, both the enthalpy of the reaction (recommended ≈ -49 to 65 kJ mol⁻¹) (Lively and Realff, 2016; Yang and Dai, 2021) and the CO₂ capacity must be considered when designing a sorbent for DAC. Similarly, there is a trade-off between capacity and adsorption rate for solid sorbents. Grafting of CO₂-philic moieties such as amine, hydroxy, sulphonate, imidazole, triazine, and imine groups onto solid sorbent pore surfaces to improve CO₂ capacity (Petrovic et al., 2021) can result in blocking of the pore openings and prevents CO₂ diffusion. In a solid adsorbent, gas diffusion can be tuned by modification of pore openings and creating a high surface-to-volume ratio to facilitate the accessibility to the active surface area.

For liquid sorbents, gas diffusion is impacted by the viscosity. Mota-Martinez et al. examined the impact of physiochemical and transport properties of CO₂ capture solvents in terms of the cost of the overall separation process for the emission mitigation purposes. They report that viscosity is a critical parameter determining the absorber size and the associated operational and capital costs (Mota-Martinez et al., 2017). For example, the aqueous amine benchmark solvent with a viscosity of 2.5 cP requires a 50 m tall absorber column whereas a solvent with a viscosity of 16 cP requires 133 m in height for the absorber. For post-combustion CO₂ capture, to maximize the efficiency of CO₂ capture process, it is generally recommended to keep the viscosity of a CO₂-capturing solvent under 5 cP when operating in an absorber column (Song et al., 2017). However, similar analysis are not yet available for emerging DAC solvents. In complement to designing novel liquid sorbents with low viscosity, packaging sorbents, e.g., via encapsulation, is an attractive strategy for enhancing performance. Provided a capsule shell that is permeable to CO₂ is used, encapsulation can increase the effective surface area of the liquid sorbent and lead to more rapid gas uptake. Capsules of IL can be prepared by impregnation of a hollow carbon

shell using a co-surfactant (Moya et al., 2018), extrusion (Zhang and Cai, 2012), or interfacial polymerization (Weiss and Abu-Reziq, 2017). For example, Pentzer, Gurkan, and coworkers used interfacial polymerization in IL-in-water or IL-in-oil emulsions to prepare capsules a core of IL and polyurea-based shell (Huang et al., 2019; Luo et al., 2019; Gaur et al., 2021). The same group encapsulated a task specific IL for the chemisorption of CO₂ and performed breakthrough and regeneration experiments; the capsules were stable under humid conditions, outperformed zeolites at low pressure, and were stable through the multiple absorption-desorption cycles (Lee et al., 2020). Thus, in addition to enhancing the physical properties of liquid sorbents, e.g., by decreasing viscosity, such composites can give access to new structures with various knobs to tune performance related to capacity, selectivity, and regeneration.

3.2. CO₂ selectivity

Selective capture of CO₂ from atmospheric air is crucial since molecules such as N₂, H₂O, and O₂ can also be physisorbed. Further, volatile organic components can permanently bind to active sites that would otherwise be available to bind CO₂, leading to a decrease in the overall CO₂ capacity of a sorbent. It is important to note that CO₂ capacity of liquid sorbents generally improves under moist conditions, since the interaction between CO₂ and water can also lead to the formation of carbonic acid and increase the CO₂ capacity (Avelar Bonilla et al., 2019). However, the presence of co-adsorbed moisture consumes additional thermal energy during the solvent regeneration. This is because water has a high heat capacity (4.2 J/g K), and the presence of moisture in the sorbent will consume more energy for regeneration (Quang et al., 2015). Additionally, in the context of adsorptive processes, water has higher selectivity compared to CO₂ toward sorption sites, thus hindering CO₂ sorption at the amine sites (Shaik et al., 2022). Recently, Young et al. (2021) proposed a mechanistic isotherm model for amine-containing sorbents that successfully predicted CO₂ co-adsorption performance in humid conditions on Lewatit[®] VP OC 1065 (a commercially available benchmark sorbent comparable to first-generation sorbent used by Climeworks for DAC process). However, the applicability of such accurate co-adsorption models to other types of amine-functionalized sorbents, especially under high relative humidity representing real-world condition remains unclear and requires further development to uncover potential opportunities for enhancing the overall efficiency of a DAC process. Finally, high-purity CO₂ must be obtained so that it can be further sequestered or utilized as a feedstock to produce renewable fuels or value added chemicals. Therefore, to produce highly pure CO₂ from a mixture stream, pore openings of solid sorbent materials should be tuned according to the kinetic diameter of CO₂ (3.3 Å) together with high chemical affinity toward CO₂.

3.3. Regeneration energy

The regeneration of sorbents in DAC currently requires energy in the range of 6–10 GJ/tCO₂ and 4–6 GJ/tCO₂ for liquid and solid sorbents, respectively (Ozkan et al., 2022). As mentioned earlier,

the regeneration energy is related to the heat capacity and the reaction enthalpy of CO₂ binding. To make DAC cost-effective, developing sorbents that can be regenerated using energy similar to the flue gas CO₂ capture process (≈ 2 GJ/tCO₂) is crucial (Zhang et al., 2016). Furthermore, theoretical calculations estimate that the energy requirement to produce a highly concentrated stream of CO₂ (>90% purity) is 20 kJ/mol CO₂ under ≈ 400 ppm of CO₂ concentration (House et al., 2011). However, DAC technologies are currently not as efficient, with typical energy requirements of ~ 400 kJ/mol CO₂, assuming an efficiency of 5% for DAC system (House et al., 2011). Thus, increasing the CO₂ capture efficiency, ideally above 20%, is a critical target for making DAC more economically sustainable. Additionally, the high regeneration temperature requirement (e.g., 900°C for hydroxide-based solvents) (Keith et al., 2018; National Academies of Sciences, 2019) poses a challenge in terms of thermal energy and heating rate efficacy; necessitating the need for alternative energy sources and the targeted delivery of energy (i.e., overcome reliance on bulk convective heating). In this context, electrical energy can be utilized to regenerate sorbents through techniques such as microwave (MW) and induction-based heating, which offer rapid dielectric heating rates compared to conventional thermal heating (Wilcox, 2020; Mohd Pauzi et al., 2022). Ozkan et al. (2022) demonstrated that the use of electricity for both liquid and solid sorbent regeneration yields a lower thermal energy equivalent for DAC. In addition, MW based-heating has several potential benefits compared to conductive heating methods, including energy efficiency, rapid heating rates, and the ability to provide instantaneous dielectric heating without heat transfer limitations (Gomez-Rueda et al., 2022). Lee et al. (2023) recently demonstrated the susceptibility of IL sorbents to dielectric heating and rapid CO₂ desorption. These benefits may help to reduce the overall energy consumption, regeneration rate, and cost of DAC operations. However, further research is needed to fully understand the potential of MW assisted regeneration method and its impacts on sorbent stability and absorption-desorption cyclability for DAC.

3.4. Thermal stability and oxidative degradation

Cyclic capacity is dependent on the thermal and oxidative stability of a sorbent. Oxidative degradation can be particularly problematic in amine-based CO₂ capture systems where the material is exposed to oxygen; amines can undergo chemical reactions in the presence of O₂ resulting in the formation of undesired byproducts, such as ammonia and amine-derived carbonates (Spietz et al., 2018; Vevelstad et al., 2022). Indeed, oxidative and thermal degradation in amines leads to significant CO₂ capacity loss (Vevelstad et al., 2022). In addition, degradation of solvent can increase corrosion, foaming, and fouling in a CO₂ capture unit, thus decreasing the lifetime of a system and increasing the cost of the CO₂ capture process (Saeed et al., 2018). Using a thermally stable solvent can help to minimize the need for frequent solvent replacement and reduce the overall cost of the process, as well as minimize environmental impacts. Recently, we reported on the oxidative and thermal degradation mechanism of a functionalized IL where its superior stability,

compared to conventional CO₂ capture solvents, was shown to maintain its high capacity (Lee et al., 2023). Hence, developing sorbent materials with minimal thermal degradation and excellent oxidative stability is essential for the development of practical DAC technologies. For most of the emerging sorbents, detailed stability and cyclability studies are scarce and thus their practicality cannot be fully assessed.

3.5. Design and cost

DAC technology has an estimated cost of \$264–1,000 per tCO₂ assuming 75% air capture efficiency and 95% CO₂ purity (National Academies of Sciences, 2019), which is still significantly higher than the cost of CO₂ capture from flue gas (\$50–100/tCO₂) (Lebling et al., 2022). It is important to note that the current capture efficiency of DAC technology is <10% (Zeman, 2007; Long-Innes and Struchtrup, 2022), which significantly impacts the cost, making the debate to achieve the highly ambitious target of \$100–300/tCO₂ viability skeptical (Küng et al., 2023). Since, DAC is an emerging technology, it is imperative to comprehensively explore the general energy requirements and economic feasibility, as well as the environmental viability of a breakthrough sorbent. An optimized design of an air-sorbent contactor column is critical to efficiently remove CO₂ from the air, especially in the case of powder sorbents. However, driving large volumes of atmospheric air and the associated pressure drop in a sorbent unit incur a substantial operational cost (Zolfaghari et al., 2022). Thus, developing an effective contactor to process the large volume of air will also help lower the overall cost of capturing CO₂. Recent studies have indicated that fossil fuels remain a cost-effective energy source, but their use may also render the CO₂ capture process ineffective as it releases significant amounts of CO₂ (Terlouw et al., 2021). Therefore, for net zero emissions, in a DAC unit that utilizes fossil fuel-derived thermal energy, the downstream emissions should be captured while controlling the upstream emissions (Ozkan et al., 2022). On the other hand, integrating renewable energy resources such as wind and solar to a DAC unit would enable a cost-effective and environmentally friendly solution for reducing the current atmospheric CO₂ concentration (Zolfaghari et al., 2022).

3.6. Environmental impacts of sorbents

Environmental assessment of sorbents in DAC technologies is often overlooked. To fully evaluate DAC feasibility, it is crucial to consider the environmental impacts of sorbent production, disposal, and lifecycle costs (Leonzio et al., 2022). For example, sodium hydroxide used in DAC process is corrosive and generates toxic chlorine gas during production (Realmonte et al., 2019). ILs, on the hand, are relatively benign, in particular due to their negligible volatility, but require proper disposal (de Jesus and Maciel Filho, 2022). A recent study linked the presence of high levels of imidazolium-based cations in soil (i.e., landfill site sampled in United Kingdom) to human health issues such as primary biliary cholangitis which is a chronic autoimmune liver disease (Abdelghany et al., 2020).

Likewise, MOF synthesis often involves solvothermal strategies that use toxic solvents (Kumar et al., 2019; He et al., 2022). To address these issues, further research should focus on developing environmental friendly synthesis routes for emerging CDR sorbents that avoid the use of hazardous solvents and explore innovative techniques like microwave-assisted synthesis (Thomas-Hillman et al., 2018), electrosynthesis (Al-Kutubi et al., 2015), or 3D printing (Lieu et al., 2022). For MOF synthesis, these approaches are also shown to reduce synthesis time, promote rapid growth, and enable homogeneous packing assemblies as additional benefits.

4. Conclusions

In this perspective, we have highlighted the emerging sorbents to capture CO₂ directly from the air. Most current research focuses on improving CO₂ working capacity sorption kinetics and does not typically address cyclability or the design of scalable DAC infrastructure. In the development of new sorbents, computational studies identifying descriptor for molecular design and approaches that accelerate sorbent discovery and prediction of properties are required. These will likely require multiscale modeling aimed at interrogating both bulk sorbent properties as well as detailed CO₂ binding energetics (Alizadeh et al., 2021; Heydari Dokoochaki and Zolghadr, 2021; Malik et al., 2021). The field is still in search of a breakthrough sorbent that possess high selectivity, high working capacity under varied humidity and temperature conditions, improved thermal and oxidative stability and cyclability. Even with a breakthrough sorbent, the critical factor in ensuring the successful net and sustainable removal of CO₂ is integrating the DAC system with low-carbon energy sources and establishing the lifetime of the sorbent, including appropriate operating conditions and stability to CO₂ uptake-release cycles. Therefore, there is also the need to develop technology platforms that transfer energy efficiently. Taken together, intensive research focus, government policy and support, and private industry funding are needed to overcome the technological challenges of developing viable DAC systems to operate at a net-zero cost.

Data availability statement

The original contributions presented in the study are included in the article, further inquiries can be directed to the corresponding author.

Author contributions

MZ and BG created the first draft of the manuscript. All authors contributed to the revisions.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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