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## Key factors influencing direct CO<sub>2</sub> capture from air

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Excessive CO<sub>2</sub> emission into the atmosphere has caused serious climate change that draws global concerns. Direct air capture (DAC) plays a crucial role in reducing atmospheric CO<sub>2</sub> concentration and mitigating climate change. In this review, we summarize the latest advances and emerging opportunities in DAC through three key aspects. First, we introduce two different types of air contactors (adsorption- and absorption-based) and their functions, unveiling their critical roles in DAC. Second, we discuss a number of effective capture agents for DAC, including solid adsorbent materials and liquid absorbent solutions, with emphasis on the capture mechanism and efficiency. Third, we present three typical methods for CO<sub>2</sub> release and capture agent regeneration that match DAC effectively, including temperature swing, precipitation-phase separation, and electrolysis, focusing on the CO<sub>2</sub> release process and energy consumption. In the end, we provide insights into the existing challenges, potential solutions, and future directions for advancing DAC technologies.

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### 1 Introduction

The CO<sub>2</sub> concentration in the atmosphere has risen to 420 ppm, bringing a significant crisis to the Earth's climate system, such as global warming.<sup>1</sup> In response to this

challenge, the historic Paris Agreement was adopted at the 2015 Conference of the Parties to the United Nations Framework Convention on Climate Change, which emphasizes the significance of reducing greenhouse gas emissions and calls for global cooperation to limit temperature rise to <2 °C, with efforts to strive for a limit of 1.5 °C.<sup>2</sup> The agreement has spurred increasing efforts to reduce CO<sub>2</sub> emissions in many countries and regions by setting carbon neutrality targets, which is defined as equilibrating CO<sub>2</sub> emissions with removal and represents a state of net-zero emissions essential for climate stabilization.<sup>3–7</sup>

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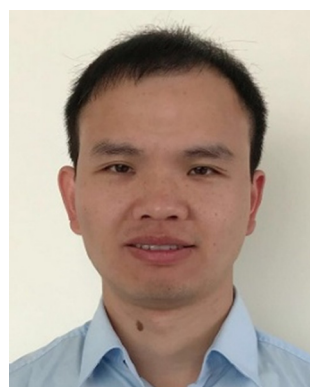
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Carbon capture, utilization, and storage (CCUS) is a crucial technology for achieving carbon neutrality by reducing CO<sub>2</sub> emissions and enhancing carbon sinks across various industrial sections, offering both environmental benefits and economic opportunities.<sup>8,9</sup> CO<sub>2</sub> capture is the first step in CCUS, laying the foundation for subsequent processes of CO<sub>2</sub> storage and utilization. At present, research on CO<sub>2</sub> capture is mainly focused on point sources (*e.g.*, power, petrochemical, and steel plants) due to their enormous and manageable CO<sub>2</sub> emissions. However, about 1000 gigatons (Gt) of CO<sub>2</sub> must be removed from the atmosphere by 2100 to meet the target set by the Paris Agreement, which sounds like an impossible task if only relying on conventional CCUS technologies.<sup>10</sup> In addition to point sources, emissions from decentralized sources such as transportation, residence, and agriculture account for 36% of total annual CO<sub>2</sub> emissions.<sup>11</sup> This portion of CO<sub>2</sub> is almost entirely emitted into the atmosphere, constituting another important emission source. Uncontrolled CO<sub>2</sub> emissions could lead to a global temperature increase exceeding 1.5 °C within 10 years.<sup>12</sup> Gür proposed a vivid picture for CO<sub>2</sub> removal: offsetting just 1 Gt atmospheric CO<sub>2</sub> would require reforesting an area of 900 000 km<sup>2</sup> (nearly the total land area of Germany and France).<sup>13</sup> Therefore, it is imperative to explore new technologies for capturing CO<sub>2</sub> from air.

Direct air capture (DAC) emerges as a new technology aimed at reducing atmospheric CO<sub>2</sub> concentrations. Unlike carbon capture from flue gas which targets specific emission sources, DAC removes CO<sub>2</sub> directly from ambient air. This capacity makes it particularly suitable for sectors where controlling CO<sub>2</sub> emissions is challenging, such as transportation, residence, agriculture, and construction. Moreover, DAC technology can be integrated with other emerging technologies to create a more comprehensive strategy for addressing climate change.<sup>14</sup> For example, powering DAC systems with renewable energy such as solar and wind power can significantly reduce their carbon footprint during operation, and converting the captured CO<sub>2</sub> into valuable fuels and chemicals enables carbon recycling.<sup>15–19</sup>



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Thus, the implementation of DAC not only offers a technological means for mitigating climate change but also holds the potential to promote the development of emerging industries and green economy.

Despite of the great prospect, DAC is currently in its early stages of development. Fig. 1a summarizes the reported projects for DAC and point source CO<sub>2</sub> capture all over the world, according to the data from the International Energy Agency, April 2025. It shows that the number of DAC projects is very limited, and only a few of them (5) are in operation. Most DAC initiatives are still under construction (5) or in the planning (45) stages. In contrast, there are already 53 operational point-source CO<sub>2</sub> capture projects, 36 under construction, and 337 planned, which all outnumbers DAC dramatically. The total capacity of operational CO<sub>2</sub> capture plants suggests that the 53 point source plants can capture approximately 50.9 million tons CO<sub>2</sub> per year, while the DAC plants only capture 0.017 million tons CO<sub>2</sub> annually (Fig. 1b).

The slow development of DAC is primarily due to the large energy consumption and low efficiency of extracting low concentration of CO<sub>2</sub> from air. Consequently, developing efficient DAC systems is crucial to advance this emerging technology and making it viable for practical applications. A complete DAC system can be divided into three major components: air contactor, capture agent, and CO<sub>2</sub> release/capture agent regeneration unit. Each component critically influences the overall efficiency of the DAC process. Previous research has generated substantial knowledge and progress into these key aspects, paving the way for practical applications of the DAC technologies.<sup>21,22</sup> Recently, a handful of reviews have summarized the important progress of DAC technologies, primarily focusing on either the development of efficient sorbent materials,<sup>23,24</sup> the system's water management,<sup>25</sup> or mobile application scenarios.<sup>26</sup> However, a review that explicitly links the design and optimization of air contactors, capture agents, and regeneration methods remains absent in the literature. Addressing this gap through a systematic examination of the entire DAC process would significantly benefit the DAC community.

This review is structured around the three key factors influencing the DAC efficiency, including air contactors, capture agents, and CO<sub>2</sub> release/capture agent regeneration approaches (Fig. 2). We start by introducing the types and functions of the core setup for DAC, *i.e.*, the air contactors. Following this, we discuss two types of capture agents for DAC, *i.e.*, solid adsorbent materials and liquid absorbent solutions, focusing on their mechanism and efficiency for CO<sub>2</sub> capture. Finally, we present three primary approaches for CO<sub>2</sub> release and capture agent regeneration that match DAC. Additionally, we outline the challenges faced by current DAC technologies and propose potential solutions in conjunction with our perspectives on the future directions of DAC.

## 2 Air contactors

The air contactor is the core unit of the DAC system, responsible for maintaining substantial airflow and facilitating the



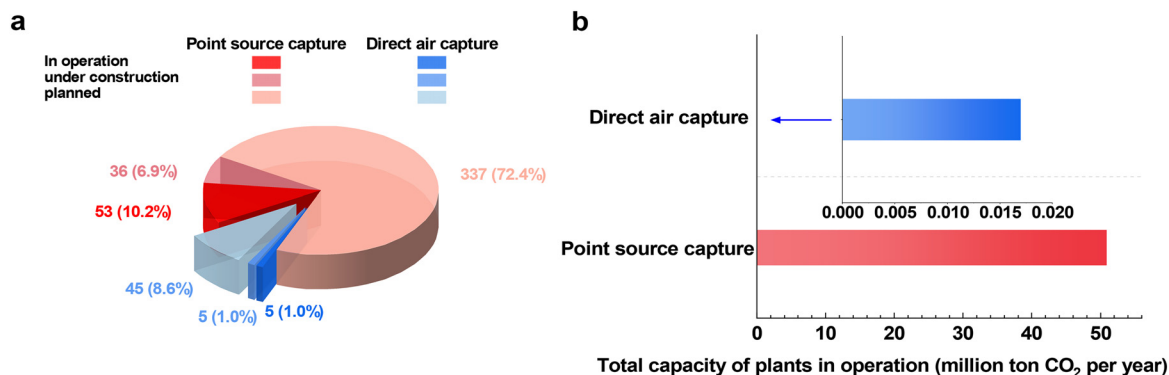


Fig. 1 (a) Reported plants for CO<sub>2</sub> capture which are either in operation, under construction, or planned; (b) total annual CO<sub>2</sub> capture capacity of operational carbon capture plants. Data from International Energy Agency.<sup>20</sup>

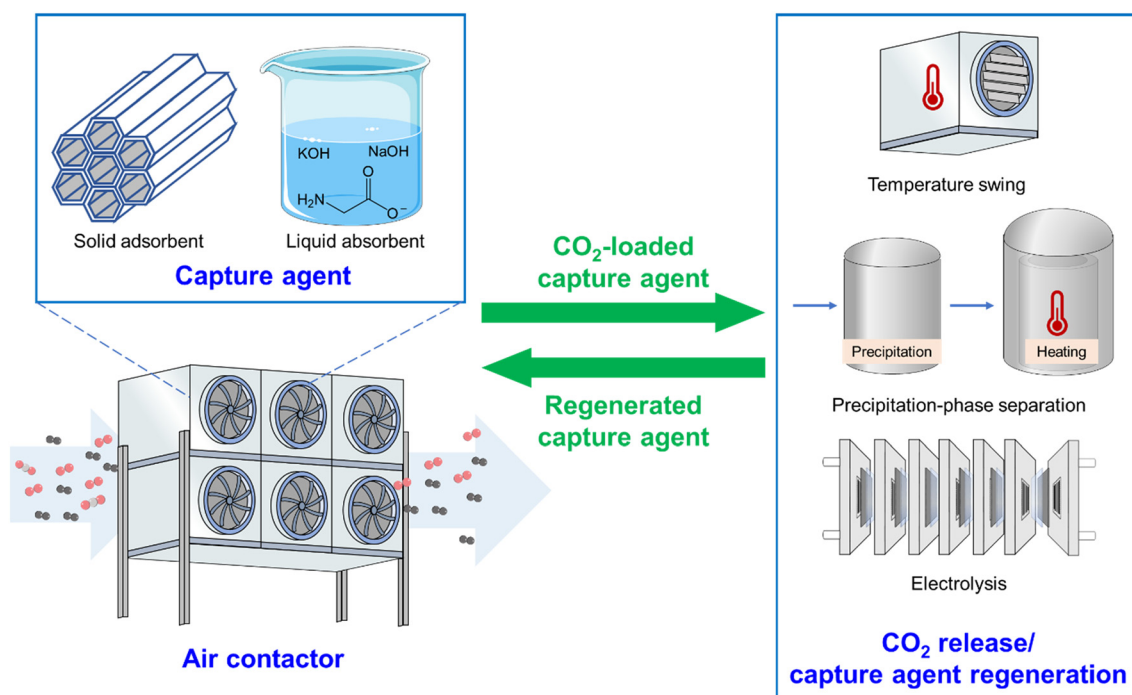


Fig. 2 Three key factors influencing the DAC efficiency.

interaction between ad/absorbents and CO<sub>2</sub> molecules. It typically comprises axial fans and packing layers. The fan system draws in air, while the packing layer, made from lightweight and corrosion-resistant materials such as polyvinyl chloride (PVC), is designed with numerous pores and channels to maximize the surface area.

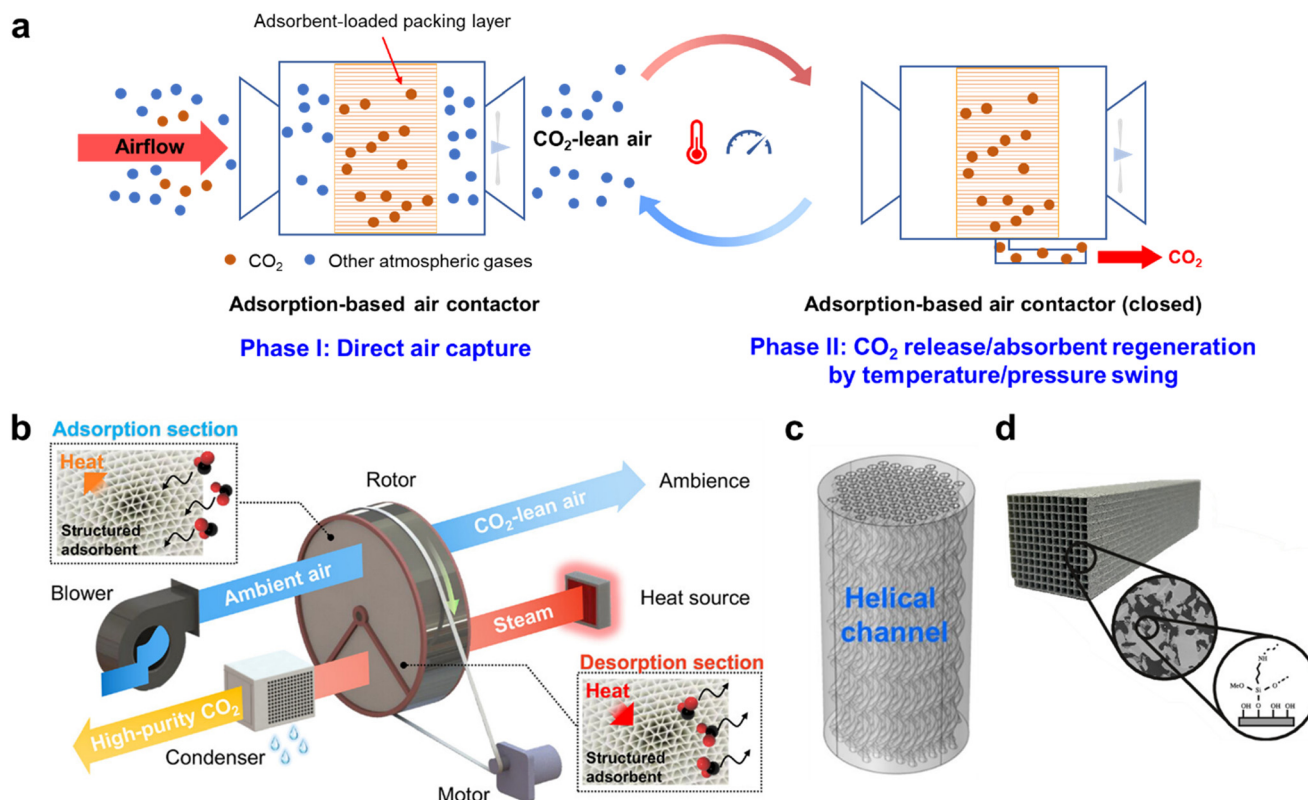
Air contactors can be categorized into two types based on CO<sub>2</sub> capture modes: adsorption and absorption. In general, adsorption-based air contactors employ solid adsorbents for CO<sub>2</sub> capture, whereas absorption-based air contactors utilize liquid solutions. However, it is important to note that some CO<sub>2</sub> capture systems may involve hybrid mechanisms. For example, Liu *et al.* reported a slurry-based sorbent consisting of the metal-organic framework (MOF), ion liquid, amine, and membrane, which achieved efficient CO<sub>2</sub> capture

through a hybrid adsorption-absorption mechanism that leverages the advantages of both solid and liquid sorbents.<sup>27</sup> To provide conceptual clarity, we classify DAC systems as either adsorption- or absorption-based types according to the sorbent phase and the dominant CO<sub>2</sub> capture mechanism.

### 2.1 Adsorption-based air contactors

An adsorption-based air contactor is usually equipped with a solid adsorbent on the packing layer and connected with an *in situ* system for CO<sub>2</sub> release and adsorbent regeneration (Fig. 3a). The operational process is straightforward: (i) air is drawn into the contactor *via* the fan system and brought into contact with the solid adsorbent, where CO<sub>2</sub> is adsorbed; (ii) after the capture process, the contactor is sealed, and a





**Fig. 3** Different configurations of adsorption-based air contactors: (a) the common fixed-bed configuration with *in situ* temperature/pressure swing for  $\text{CO}_2$  release; (b) the rotating-bed configuration with steam-assisted  $\text{CO}_2$  release. Reprinted with permission from ref. 35. Copyright 2026 Wiley; different configurations of packing layers: (c) helical packing structure. Reprinted with permission from ref. 36. Copyright 2023 Elsevier; (d) wash-coated honeycomb monolithic configuration. Reprinted with permission from ref. 37. Copyright 2023 American Chemical Society.

temperature/pressure swing program is initiated to release  $\text{CO}_2$  and regenerate the adsorbent. Most reported DAC systems rely on adsorption-based air contactors, including those from Climeworks, Heirloom, and CarbonCapture Inc.<sup>28–30</sup> These adsorption-based air contactors integrate all functions for  $\text{CO}_2$  capture, release, and adsorbent regeneration, showing significant advantages such as small space occupation and flexible modularity for placement in any location. Notably, the size of the air contactors determines the scale of  $\text{CO}_2$  capture. In 2021, Climeworks launched its Orca facility in Iceland, which was the world's largest adsorption-based DAC plant at the time, achieving an annual  $\text{CO}_2$  capture capacity of 4000 tons.<sup>31</sup> Building on this milestone, the company launched its Mammoth facility in 2024, with a capacity of 36 000 tons  $\text{CO}_2$  per year, nearly ten times that of Orca. This scale-up marks a significant step toward megaton-scale DAC deployment.<sup>32</sup>

The common fixed-bed reactors can serve as air contactors for DAC. However, they impose stringent requirements on the size of adsorbent particles, which limits adsorbent selection and makes them susceptible to attrition.<sup>33</sup> Moreover, multiple fixed beds must operate in parallel to maintain continuous  $\text{CO}_2$  capture, significantly increasing system complexity.<sup>34</sup> To address these challenges, Wu *et al.* recently developed a rotating-bed contactor (Fig. 3b).<sup>35</sup> In this design, powdered

capture agents are fabricated into structured sorption modules and loaded into a rotatable reaction bed, which is divided into two functional zones for adsorption and desorption. In the adsorption zone, air flows through the module, where  $\text{CO}_2$  is captured by the adsorbent. In the desorption zone,  $\text{CO}_2$  is desorbed with hot steam. A motor drives the bed to rotate slowly and continuously, enabling each sorption module to cycle periodically between adsorption and desorption. This configuration allows for continuous  $\text{CO}_2$  capture from air.

The packings inside the adsorption-based air contactor can be of various cross-sectional shapes, with the adsorbent material loaded on the walls of the channels. As air flows through the contactor,  $\text{CO}_2$  diffuses through the channel walls, where it is captured by the adsorbent. Enhancing the mass transport of  $\text{CO}_2$  to the adsorbent is crucial for improving  $\text{CO}_2$  capture efficiency and reducing operational cost. Therefore, optimizing the structure of the packing layer is very important. Tegeler *et al.* presented a high-surface-area packing with helical channels instead of straight ones, significantly enhancing the transport of  $\text{CO}_2$  to the adsorbent (Fig. 3c).<sup>36</sup> The air contactor with such a packing configuration reduced the overall DAC cost by about 30%. The research by Grossmann *et al.* indicated that the structure of the adsorbent loaded onto the packing layer also affected  $\text{CO}_2$  capture efficiency (Fig. 3d).<sup>37</sup> They evaluated two adsorbents: one with a pellet structure and the other with a



wash-coated honeycomb monolith structure, and found that the latter enhanced mass transport kinetics and reduced pressure drop, making it a more promising option for DAC contactors.

It is important to note that temperature and vacuum are key driving forces for the regeneration of adsorbents in adsorption-based air contactors, necessitating the integration of control valves, vacuum pumps, and heat sources.<sup>38</sup> This makes the internal piping and control processes of such air contactors relatively intricate, as a single contactor incorporates too many subsystems. Moreover, given the need to release high-purity CO<sub>2</sub>, the sealing integrity of the air contactor deserves special attention.

## 2.2 Absorption-based air contactors

An absorption-based air contactor shares a similar structure with the adsorption-based counterpart but has completely different CO<sub>2</sub> capture and release processes. In this type of air contactor, the liquid absorbent flows uniformly through the packing layer, ensuring sufficient contact with CO<sub>2</sub> in the air stream to form a CO<sub>2</sub>-loaded solution, which is then transported to a separate unit for CO<sub>2</sub> desorption (Fig. 4a). In this system, CO<sub>2</sub> release is decoupled from the capture process, enabling on-demand adjustment of the release strategy to accommodate actual needs and varying operating conditions.

To our knowledge, Carbon Engineering is the only company in the world using absorption-based air contactors, which employ a calcium loop system involving precipitation, phase separation, and calcination for CO<sub>2</sub> release and KOH absorbent solution regeneration (Fig. 4a).<sup>39</sup> The first commercial facility using Carbon Engineering's technology is under construction in the USA and is designed to capture 500 000 ton CO<sub>2</sub> per year.<sup>40</sup> Beyond conventional absorption-based air contactors, the hollow fiber membrane contactor (HFMC) offers a compelling alternative by uniquely integrating CO<sub>2</sub> absorption in solutions with membrane-based gas separation (Fig. 4b). While still under development, the modular design and stable phase interface of HFMC render it a promising platform for DAC applications.<sup>41</sup>

The packing is also the core component of the absorption-based air contactor, directly determining the system's mass transport efficiency and CO<sub>2</sub> capture performance. The packing used in traditional absorption towers for flue gas capture is well established, and recent research has been directed to optimizing the packing layer structure with advanced technologies such as 3D printing.<sup>42</sup> However, due to the significant difference in CO<sub>2</sub> concentration between air and flue gas, using the same packing from absorption towers is not effective for DAC, necessitating the development of special packing structures tailored to DAC applications. For example, Kasturi *et al.* developed a hybrid packing structure

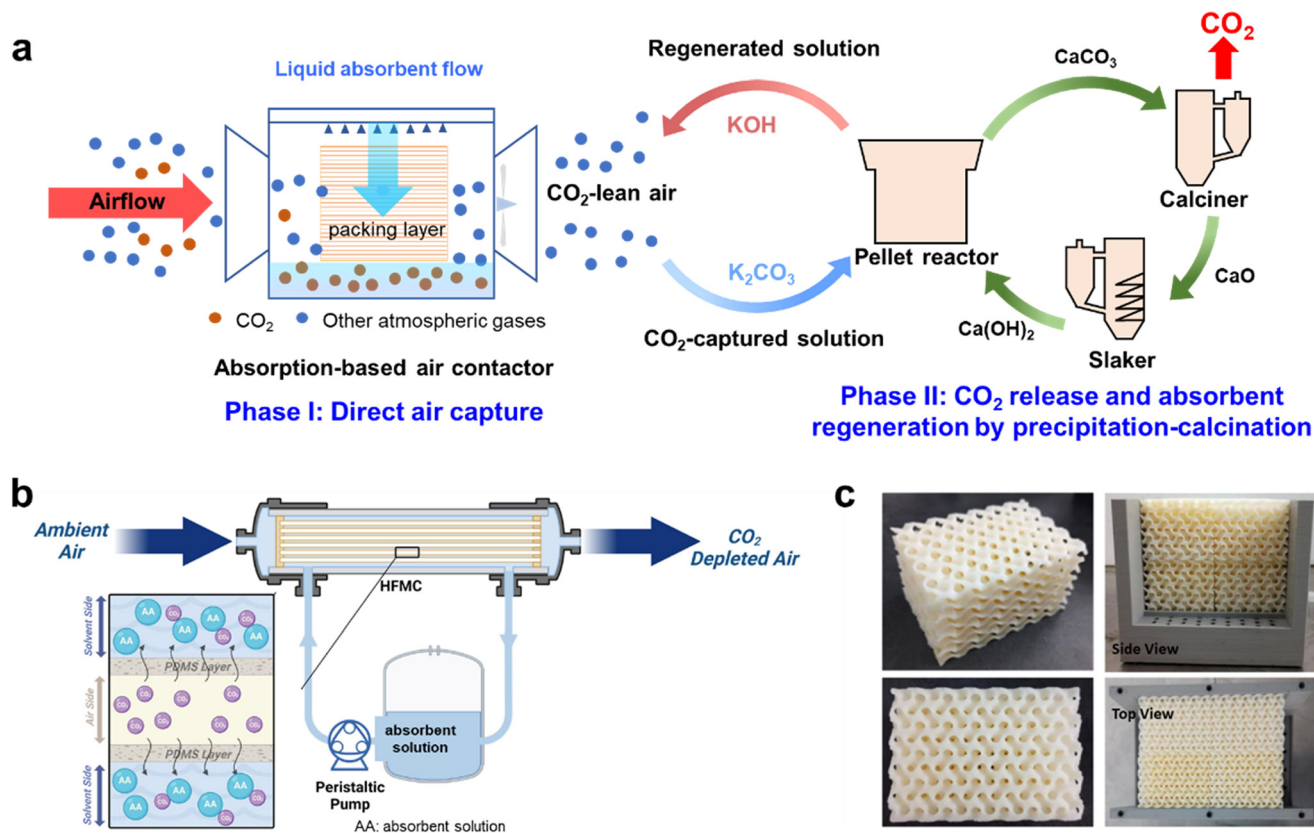


Fig. 4 (a) The common absorption-based air contactor with separate precipitation, phase separation, and calcination steps for CO<sub>2</sub> release and capture agent regeneration; (b) the HFMC-type contactor. Reprinted with permission from ref. 41. Copyright 2024 Elsevier; (c) packings with 3D printed triply periodic minimal surfaces. Reprinted with permission from ref. 43. Copyright 2024 Wiley.



utilizing two specialized materials, structured packing made of PVC and random packing made of stainless steel, to optimize mass transport and enhance carbon capture performance.<sup>21</sup> Such a packing layer exhibits a relatively high specific surface area ( $885 \text{ m}^2 \text{ m}^{-3}$ ), achieving a  $\text{CO}_2$  absorption efficiency of 75% and a  $\text{CO}_2$  capture rate of 550 g per day, highlighting the critical role of packing materials in high-throughput contactors. An *et al.* proposed a packing geometry known as triply periodic minimal surfaces (TPMS), which consisted of two interpenetrating fluid domains separated by thin walls and exhibited periodicity in three dimensions (Fig. 4c).<sup>43</sup> Such TPMS geometry enhanced mass transport efficiency by 49–61% and increased the effective gas–liquid interfacial area by 91–140% compared to the conventional packing layer.

Since absorption-based air contactors are inevitably exposed to strong alkaline absorbents, the corrosion resistance of the contactor material (*e.g.*, steel) is particularly pivotal.<sup>44,45</sup> Furthermore, prolonged gas–liquid contact generally leads to significant water loss *via* evaporation. For instance, An *et al.* reported that the water evaporation for per ton of  $\text{CO}_2$  captured was 6.6 tons under the conditions of 21 °C and 69% RH (relative humidity), while the water evaporation reported by Keith was 2–8 ton  $\text{H}_2\text{O}$  for each ton  $\text{CO}_2$  (20 °C, 35–75% RH).<sup>39,46</sup> Therefore, it is essential to develop efficient water management strategies tailored to the local climate conditions for the absorption-based air contactor.

No matter which type of air contactor is used, the low concentration of atmospheric  $\text{CO}_2$  necessitates a vast number of large-size contactors to maintain adequate air intake for large-scale DAC, which increases construction and operational costs and demands substantial land use, thereby eroding its practical benefits.<sup>47</sup> Previous estimation indicates that the theoretical energy consumption required for DAC is about twice as high as or even more than that of flue gas capture.<sup>48</sup> Therefore, it is essential to develop efficient and cost-effective air contactors or explore alternative solutions.

### 2.3 Air contactors retrofitted from existing facilities

Integrating DAC with existing buildings or facilities can accomplish efficient  $\text{CO}_2$  capture and meanwhile effectively reduce construction and operational costs for air contactors.<sup>49</sup> For example, panels made from  $\text{Ca}(\text{OH})_2$  can serve as replaceable components of buildings for DAC, with estimated  $\text{CO}_2$  capture cost of as low as  $\$102 \text{ per ton}_{\text{CO}_2}^{-1}$ , comparable to  $\text{CO}_2$  capture from flue gases.<sup>50</sup> These replaceable panels can be easily affixed to building surfaces, addressing the large land use issue of similarly sized DAC systems.

Recently, Bachman *et al.* proposed a concept of combining DAC with railcars (Fig. 5a).<sup>51</sup> This specialized train system is equipped with a  $\text{CO}_2$  capture system, a battery array, compression equipment, and auxiliary devices, eliminating the need for traditional fan array-based air contactors. They estimated the scaling potential of the DAC-railcar system, indicating that each railcar could capture approximately 3000 tons of  $\text{CO}_2$  annually, and potentially 0.45 gigatons of  $\text{CO}_2$  per year by 2030. Alternatively, Zheng *et al.* demonstrated the proof-of-concept of integrating DAC with industrial cooling towers that are operated worldwide, which could be retrofitted to capture  $\text{CO}_2$  from air by adding a suitable absorbent into the circulating water (Fig. 5b).<sup>52</sup> Based on the experimental data, they projected that the existing cooling towers in the North American region alone could achieve an annual  $\text{CO}_2$  capture capacity of  $\sim 4$  gigatons if they could all be retrofitted with DAC function. These examples reveal the great potential of retrofitting existing facilities for DAC.

The air contactor is a key determinant of DAC's practical feasibility, governing both capital expenditure and  $\text{CO}_2$  capture efficiency. As summarized in Table 1, current contactor designs fall into two broad categories, adsorption-based and absorption-based systems, each with distinct trade-offs. Among adsorption-based configurations, traditional fixed bed contactors with structured packings offer high modularity and scalability, whereas the emerging rotating-bed contactors enhance mass transfer within a compact footprint. In comparison,

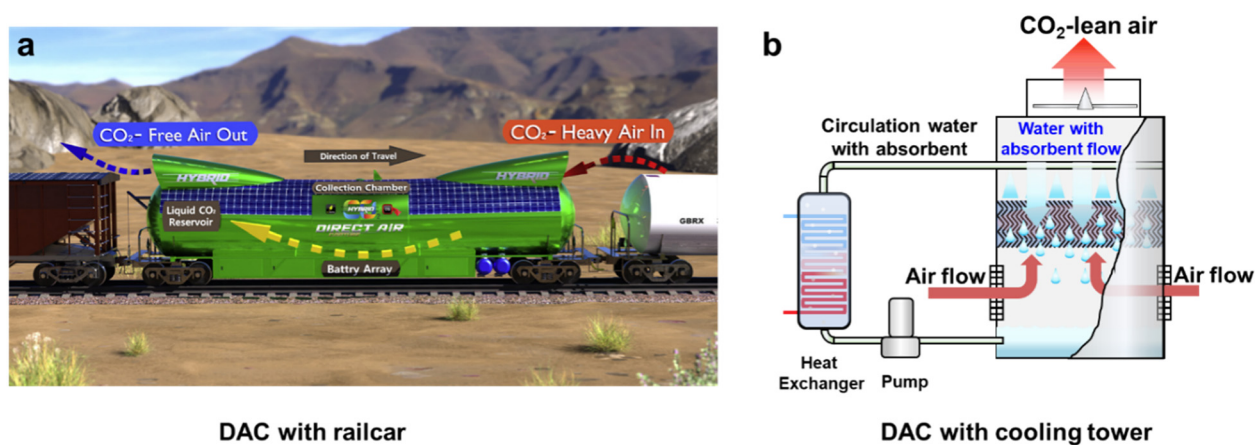


Fig. 5 Integrating DAC with existing facilities: (a) railcar. Reprinted with permission from ref. 51. Copyright 2022 Elsevier; (b) cooling tower. Reprinted with permission from ref. 52. Copyright 2025 Wiley.



**Table 1** Comparative analysis of reported air contactors

Type	Configuration	Key advantages	Challenges/limitations	Ref.
Adsorption-based	Static structured packings	Highly modular Scalable	Batch/cyclic operation	36, 37
	Rotating bed	Intensified mass transfer Compact footprint Continuous capture/release	Mechanical complexity Extra energy consumption for rotation	35
	Building	Using existing urban surfaces Significant capital expenditure saving	Low capture flux Uncontrolled capture rate	50
Absorption-based	Railcar	Using “free” high-speed airflow	Influence on railcar	51
	Static structured packings/spray	Mature gas–liquid contact technology Decoupled capture and regeneration units Cost-effective absorbent solutions	Water loss by evaporation Corrosion	39, 42, 43
	Membrane	Stable gas–liquid interface High surface-to-volume ratio	High membrane cost Low capture flux	41
	Cooling tower	Using existing industrial facilities Significant capital expenditure saving	Balancing DAC operation with cooling function	52

conventional absorption-based contactors employing structured packings and sprays leverage mature gas–liquid contacting technologies and decouple CO<sub>2</sub> capture from absorbent regeneration, offering operational flexibility for industrial scaling. However, they remain susceptible to water loss and corrosion. Membrane contactors present a promising alternative by providing a stable, high-surface-area interface, albeit at higher material costs.

Retrofitting existing facilities into adsorption- or absorption-based air contactors represents a promising approach to reduce DAC costs. However, its practical implementation faces several engineering challenges. For instance, although the laboratory study has demonstrated the feasibility of integrating DAC into industrial cooling towers, large-scale deployment may compromise the primary heat dissipation function of such towers, necessitating a comprehensive assessment. These challenges highlight that the success of retrofitting strategies hinges not only on the capture technology itself but also on optimized integration with the host infrastructure.

### 3 Capture agents

The choice of capture agent is of great importance in DAC, as it directly governs the efficiency of the entire process, from initial CO<sub>2</sub> capture to subsequent CO<sub>2</sub> release and its own regeneration. This section examines the characteristics of two typical types of capture agents, solid adsorbent materials and liquid absorbent solutions, discussing their advantages and limitations for DAC applications.

#### 3.1 Solid adsorbents

Adsorbents can be classified into physical and chemical types based on their properties and mechanisms for CO<sub>2</sub> capture. Physical adsorption is driven by intermolecular attraction with weaker binding forces, resulting in lower heat enthalpy and relatively higher rates for both CO<sub>2</sub> adsorption and desorption. Common physical adsorbents include zeolites, activated carbons, alumina, MOFs, and covalent organic frameworks (COFs), which are porous, possess large surface

areas, and can be modified and functionalized.<sup>53</sup> At present, physical adsorbents are primarily utilized for post-combustion CO<sub>2</sub> capture from flue gases, with limited applications in DAC.<sup>54</sup> This limitation is largely due to their low selectivity toward CO<sub>2</sub> whose concentration is low in air. Additionally, air humidity can lead to competitive adsorption between water and CO<sub>2</sub> molecules on physical adsorbents, significantly impacting CO<sub>2</sub> adsorption performance.

Accordingly, a major focus of recent research on physical adsorbents is to understand and alleviate the detrimental effects of water co-adsorption. For example, Hu *et al.* systematically compared the CO<sub>2</sub> and water adsorption behaviour of two typical zeolites, FAU and MFI, and evaluated their CO<sub>2</sub> capture performance under high relative humidity (75%).<sup>55</sup> They show that increasing the Si/Al ratio of the zeolite slightly reduces its CO<sub>2</sub> uptake but markedly enhances hydrophobicity, enabling the material to retain relatively high CO<sub>2</sub> capacity under humid conditions. More importantly, they developed an advanced zeolite adsorbent that achieves high CO<sub>2</sub> capacity, fast adsorption kinetics, and excellent humidity resistance by repairing structural defects and eliminating hydrophilic silanol groups of the MFI zeolite.

In comparison, chemical adsorbents exhibit greater tolerance to moisture and show a stronger affinity toward CO<sub>2</sub>. Thus, DAC studies mainly utilized chemical adsorbents. Metal oxides, such as CaO, capture CO<sub>2</sub> *via* carbonation reactions to form the corresponding carbonates (*i.e.*, CaCO<sub>3</sub>). The adsorbent can then be regenerated with CO<sub>2</sub> released through high-temperature calcination. However, the inherently low specific surface area of metal oxides limits the availability of their active sites for CO<sub>2</sub> adsorption. A widely adopted strategy to overcome this limitation is to disperse metal oxide onto a highly porous support. For instance, Huang *et al.* developed a composite adsorbent for DAC by loading CaO onto acid-modified attapulgite (HcATP).<sup>56</sup> They revealed that the CO<sub>2</sub> uptake of the composite arises from a synergistic effect of physical and chemical adsorption. Specifically, the HcATP support alone shows a CO<sub>2</sub> uptake of 212.4 μmol g<sup>-1</sup> through physisorption, while the loaded CaO



provides additional adsorption *via* carbonation, significantly increasing the total CO<sub>2</sub> uptake to 426.7 μmol g<sup>-1</sup>, underscoring the important contribution of chemisorption. However, it should be noted that the CO<sub>2</sub> adsorption capacity of this inorganic adsorbent is relatively low, while the regeneration temperatures for adsorbents are usually high (carbonate calcination occurs at >800 °C).

To date, amine-functionalized porous materials are the most extensively studied chemical adsorbents for DAC. These amine-based adsorbents utilize traditional physical adsorbents as supports whose porous structure facilitates the dispersion of amines, while the amines serve as the adsorption sites for CO<sub>2</sub> capture (Fig. 6). Thus, selecting the appropriate amine, support material, and method to link them together is crucial for optimizing the performance of the resulting adsorbent.

Amine-based adsorbents are categorized into two classes according to their preparation methods: physical impregnation and covalent binding (Fig. 6a). Amine-impregnated adsorbents involve the physical impregnation of amine oligomers or polymers onto porous support materials by stirring the support materials with the desired amines in an organic solvent. The advantages of easy preparation and good performance make amine-impregnated adsorbents attract intensive investigation. A variety of amine oligomers or polymers can be used to prepare amine-impregnated adsorbents, including polyethyleneimine (PEI), poly(propyleneimine) (PPI), tetraethylenepentamine (TEPA) (Fig. 6b); and the support can be any type of porous material, such as silica, alumina, and MOFs, and COFs (Fig. 6c). Table 2 summarizes a variety of amine-based adsorbents and their performance for DAC applications.

PEI has been widely utilized to prepare amine-impregnated adsorbents for its low degree of polymerization and large abundance of primary amine groups (Table 2). PEI has two typical types of structures, including linear chains with amine groups mostly secondary and dendritic structure with tertiary amines serving as branching points (Fig. 6b). The desorption enthalpy of linear PEI (74–75 kJ mol<sup>-1</sup>) is slightly lower than that of branched PEI (77–80 kJ mol<sup>-1</sup>), giving linear PEI a higher CO<sub>2</sub> desorption rate.<sup>78</sup> A significant limitation of PEI is its high susceptibility to oxidative degradation in high-temperature, oxygen-rich environments. Consequently, the regeneration of PEI requires a vacuum or inert atmosphere, which raises process complexity and energy consumption.<sup>79</sup>

PPI demonstrates better oxidation resistance compared to PEI. Yang *et al.* compared the performance of amine-impregnated adsorbents with 20 wt% PEI and 20 wt% PPI supported on a MOF (UiO-67(Zr)) under ambient air conditions (~400 ppm CO<sub>2</sub>).<sup>80</sup> The PPI-based adsorbent exhibited a higher CO<sub>2</sub> capture capacity (0.42 mmol g<sup>-1</sup>) than the PEI counterpart (0.38 mmol g<sup>-1</sup>). After exposing both adsorbents to ambient air for 7 days, PPI remained fresh, while PEI experienced a 22% loss in amine efficiency. Rosu *et al.* conducted the prolonged aging of PPI (about 2 years) and studied its performance for DAC.<sup>81</sup> Adsorbents made from aged PPI showed a slight decline in CO<sub>2</sub> capture performance (approximately 20%) compared to the fresh

polymer, but the overall CO<sub>2</sub> cycling performance remained excellent (the number of cycles is 20, the average CO<sub>2</sub> capacity is 0.96 mmol g<sup>-1</sup>). However, the synthesis of the PPI monomer is relatively complex, involving the use of various acid initiators (*i.e.*, HBr, HClO<sub>4</sub>, HCl, CH<sub>3</sub>SO<sub>3</sub>H).<sup>82</sup>

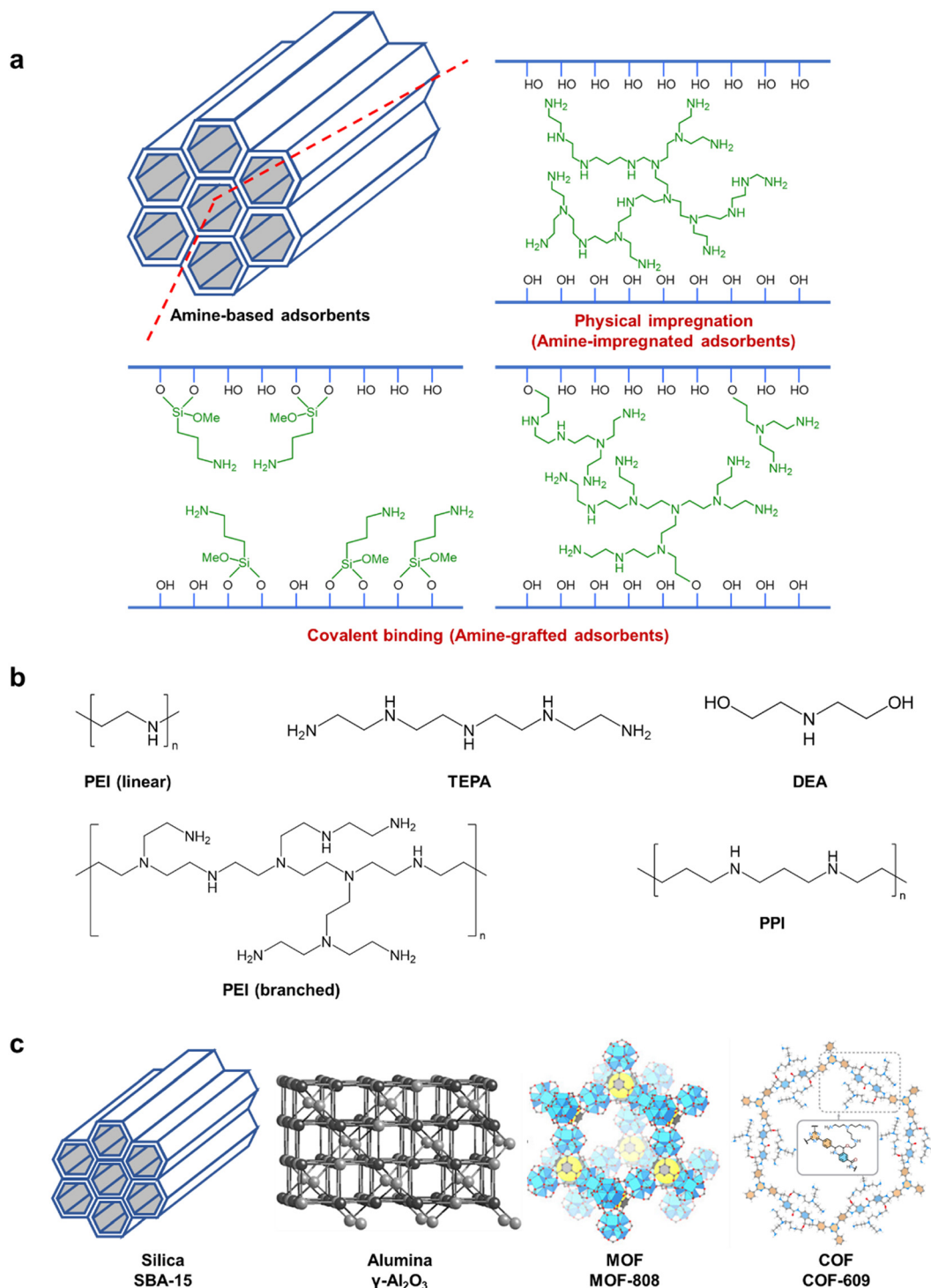
The molecular weight of the amine significantly influences the performance of amine-functionalized adsorbents. While high molecular weight PEI can increase the content of amine groups, it may impede CO<sub>2</sub> diffusion within the support structure, resulting in reduced CO<sub>2</sub> uptake. Smaller amines such as TEPA are reported to improve DAC owing to their high nitrogen content and reduced stacking tendency, which enhances CO<sub>2</sub> mass transport within porous supports.<sup>83</sup>

The performance of amine-impregnated adsorbents can be influenced by the structure of support materials, because the porous structure of the support governs amine dispersion and CO<sub>2</sub> accessibility. Rim *et al.* demonstrated this by comparing TEPA impregnated on two types of support materials, MIL-101(Cr) and γ-Al<sub>2</sub>O<sub>3</sub> (Fig. 7a and b).<sup>84</sup> *In situ* IR spectroscopy revealed that weak chemisorption (forming carbamic acid) was the dominant pathway for CO<sub>2</sub> capture over MIL-101(Cr)-supported TEPA, whereas strong chemisorption (forming carbamate) prevailed on γ-Al<sub>2</sub>O<sub>3</sub>-supported TEPA. This mechanistic divergence was attributed to the stronger amine-support interaction in MIL-101(Cr), which possessed a significantly higher surface area to pore volume ratio than γ-Al<sub>2</sub>O<sub>3</sub>. This strong interaction likely stabilized the carbamic acid intermediate *via* surface hydroxyls or water. In contrast, the weaker interaction of amine with γ-Al<sub>2</sub>O<sub>3</sub> allowed for greater amine-amine interaction, facilitating the formation of the ammonium carbamate pair. Therefore, the CO<sub>2</sub> capture pathway of impregnated amines can be engineered by modulating the amine-support interaction, highlighting the important effect of the support materials.

In practical DAC applications, environmental conditions vary with seasons. Understanding the effect of air temperature and humidity is important to accomplish optimal performance of amine-impregnated adsorbents for DAC. Priyadarshini *et al.* studied a series of TEPA-impregnated γ-Al<sub>2</sub>O<sub>3</sub> with varying amine loadings (10, 20, and 40 wt%) for CO<sub>2</sub> capture under ambient (25 °C) and sub-ambient (-20 °C) conditions.<sup>63</sup> At 25 °C, the CO<sub>2</sub> capacity of these adsorbents increased with amine loading, with 40 wt% TEPA-impregnated γ-Al<sub>2</sub>O<sub>3</sub> showing the highest CO<sub>2</sub> uptake capacity (1.8 mmol g<sup>-1</sup>). At -20 °C, however, the CO<sub>2</sub> uptake increased for the adsorbent with 10 wt% TEPA but slightly decreased for the adsorbents with 20 and 40 wt% TEPA. This was because high pore filling and restricted mobility of amine chains arising from high amine loadings caused pore blocking of the adsorbents at sub-zero temperatures. This hindered CO<sub>2</sub> diffusion, leading to reduced capacity for CO<sub>2</sub> capture. A similar phenomenon was observed in TEPA and PEI-impregnated MOFs.<sup>64</sup>

Humidity is another critical factor influencing CO<sub>2</sub> adsorption, typically leading to higher CO<sub>2</sub> capture capacity under high humidity conditions. Under dry conditions, CO<sub>2</sub> reacts with two amine molecules to form ammonium carbamate





**Fig. 6** Preparation methods and key components of amine-functionalized adsorbents. (a) Two typical methods for immobilizing amines onto support materials; common (b) amines and (c) support materials for preparing amine-functionalized adsorbents. The alumina in (c) is reprinted with permission from ref. 57. Copyright 2020 Elsevier; the MOF in (c) is reprinted with permission from ref. 58. Copyright 2024 American Chemical Society; the COF in (c) is reprinted with permission from ref. 59. Copyright 2022 American Chemical Society.

ion pairs (*i.e.*,  $\text{RNH}_3^+\text{COO}^-$  and  $\text{R}_1\text{R}_2\text{NH}_2^+\text{COO}^-$  for primary and secondary amines, respectively). This reaction limits the capacity to 0.5 mol  $\text{CO}_2$  per mole of amine and contributes to cross-linking within or between amine chains, which significantly impedes  $\text{CO}_2$  diffusion. In contrast, under moist

conditions, water-stabilized ions (*e.g.*, ammonium bicarbonate or hydronium carbamate) are thought to bind through a single amino group.<sup>85</sup> This monodentate binding can promote  $\text{CO}_2$  diffusion and enhance  $\text{CO}_2$  capture capacity. The humidity also affects the durability of adsorbents. Carneiro *et al.* studied the



**Table 2** Various amine-based adsorbents and their performance for DAC applications

Type	Amine	Support	Capture conditions <sup>a</sup>	RH <sup>b</sup>	CO <sub>2</sub> uptake (mmol g <sup>-1</sup> )	Ref.
Physical impregnation	40 wt% TEPA & 10 wt% DEA <sup>c</sup>	SBA-15	400 ppm, 25 °C	Dry	1.93	60
	50 wt% PEI <sup>d</sup>	Commercial resin particles	400 ppm, 25 °C	25%	2.95	61
	70 wt% PEI	SBA-15	400 ppm, 35 °C	78%	3.48	62
	40 wt% PEI, 40 wt% TEPA	Mesoporous $\gamma$ -Al <sub>2</sub> O <sub>3</sub>	400 ppm, 25 °C	Dry	0.9, 1.8	63
	30–50 wt% TEPA	MOF MIL-101(Cr)	400 ppm, -20 °C	Dry	0.72–1.11	64
	45 wt% PEI	Mg–Al–CO <sub>3</sub> layered double hydroxide	400 ppm, 25 °C	Dry	1.82	65
	50 wt% PEI	Mesoporous cellular foam	400 ppm, 25 °C	Dry-80%	0.41–0.71	66
	40 wt% PEI	Commercial resins	400 ppm, 28 °C	50%	1.47–1.60	67
	25 wt% DEA & 25 wt% PEI	SBA-15	400 ppm, -5 to 25 °C	20%	1.29	68
	30 wt% PEI	Mg <sub>x</sub> Al–CO <sub>3</sub> layered double hydroxides & Mg <sub>x</sub> Al–O mixed metal oxides	400 ppm, -20 to 25 °C	70%	2.0–3.2	69
	70 wt% TEPA	Hierarchical silica	420 ppm, 30 °C	50 ± 3%	5.2	70
	71 wt% TEPA	Meso-macroporous melamine formaldehyde	420 ppm, 30 °C	Dry	2.65	71
	70 wt% PEI	Zeolite 13X & MOF MIL-101(Cr)	400 ppm, -20 °C	Dry-70%	1.05–1.43	72
Covalent binding	AEEA <sup>e</sup>	MOF MIL-100 (Cr)	400 ppm, -25 to 0 °C	1%	1.91–2.42	73
	N <sup>1</sup> -(3-Trimethoxysilylpropyl)-diethylenetriamine	Hierarchical bimodal mesoporous silica	415 ppm, 25 °C	Dry	1.04	74
	47 wt% PEI	Mesoporous silica foam	400 ppm, 25 °C	65%	1.50	75
	SH-bPEI	COF (synthesized by TFTDA and TAPPy) <sup>g</sup>	400 ppm, 25 °C	Dry-75%	0.48–1.24	76
	Aziridine	COF (synthesized by TCPB and BPDA-N <sub>3</sub> ) <sup>f</sup>	400 ppm, 25 °C	Dry-50%	0.96–2.05	77

<sup>a</sup> Simulated air is used for all the studies listed. <sup>b</sup> Relative humidity. <sup>c</sup> TEPA is tetraethylenepentamine, DEA is diethanolamine. <sup>d</sup> PEI is polyethyleneimine. <sup>e</sup> AEEA is N-(2-aminoethyl)ethanolamine. <sup>f</sup> TCPB is 1,3,5-tris(4-cyanomethylphenyl)benzene, BPDA-N<sub>3</sub> is 3,3'-bis[(6-azidoheptyl)oxy]4,4'-biphenyl-dicarbaldehyde. <sup>g</sup> TFTDA is 2',3',5',6'-tetrafluoro-[1,1':4',1''terphenyl]-4,4''-dicarbaldehyde, TAPPy is 1,3,6,8-tetrakis(4-aminophenyl)pyrene.

role of water in the degradation of amine-based adsorbents using elemental analysis, solid-state NMR, and *in situ* infrared spectroscopy.<sup>86</sup> They established the cleavage of terminal C–N bonds as the dominant deactivation mechanism in PEI-based amine adsorbents, and the presence of highly reactive oxygen species (*e.g.*, OH<sup>•</sup>) in water exacerbates such cleavage. In consequence, drier air minimizes the impact of adsorbed water on amines, while low temperature leading to reduced absolute humidity enhances adsorbent durability.

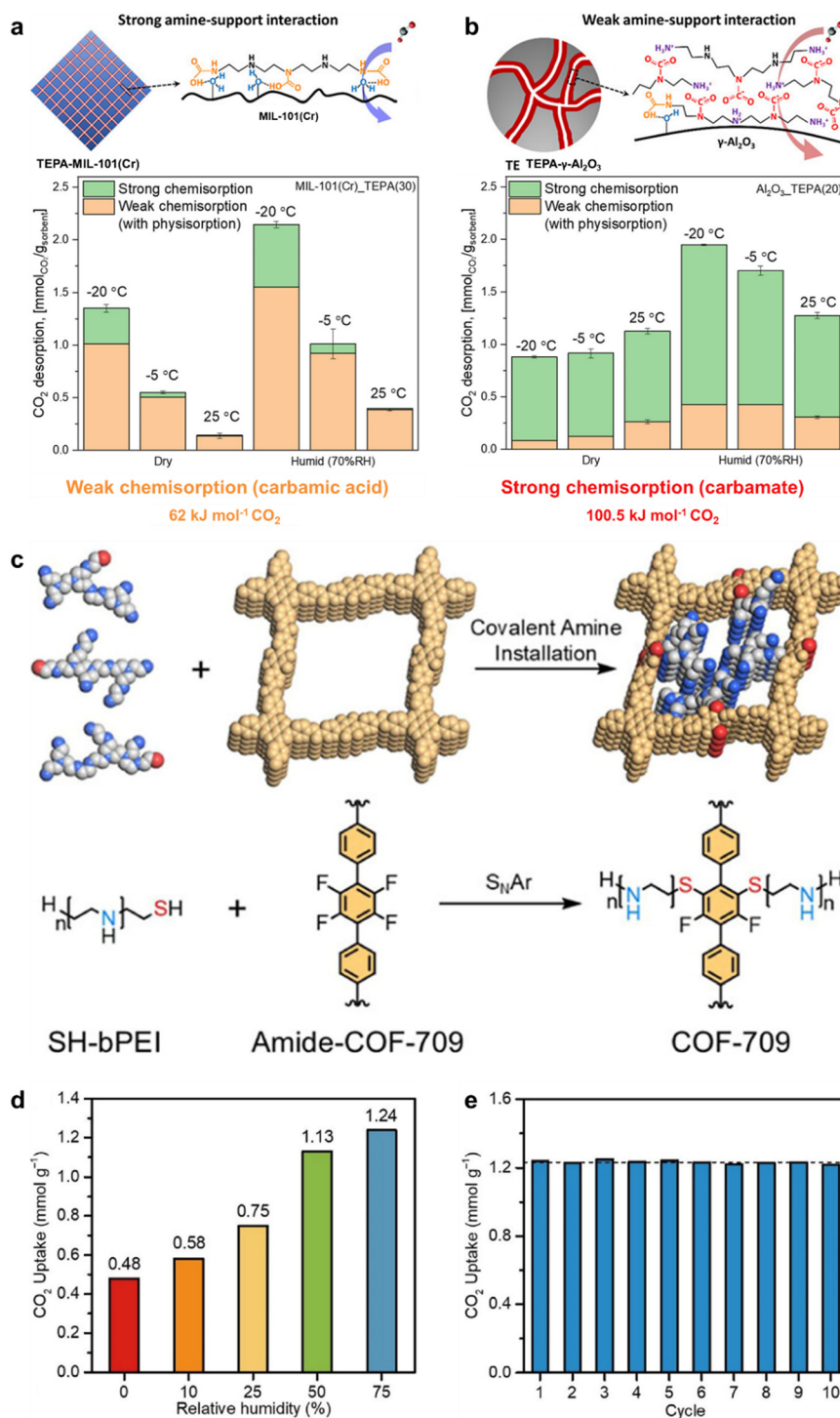
A significant issue of amine-impregnated adsorbents is that the physical impregnation of amines leads to disordered amine stacking, which has adverse effects on CO<sub>2</sub> diffusion. Additionally, the weak interaction between the amine and support makes these adsorbents more susceptible to detach. In contrast, amine-grafted adsorbents through covalent binding are more stable since their amines are chemically bonded to the support materials.

Amine-grafted adsorbents are typically prepared *via* two primary methods: one is through the silanization reaction between a silica support and aminosilanes; the other is through C–O/C–S bond formation (Fig. 6a). Anyanwu *et al.* grafted N<sup>1</sup>-(3-trimethoxysilylpropyl)diethylenetriamine onto mesoporous silica under anhydrous (dry-grafting) and hydrous (wet-grafting) conditions. A significantly higher amine loading was accomplished in the wet-grafting adsorbents (49%) than in the dry-grafting ones (28%).<sup>74</sup> This was because the presence of water enhanced the hydrolysis

of unreacted alkoxy groups and introduced additional hydroxyl groups, thereby increasing amine loading in wet-grafting. The higher amine loading allowed the amine-grafted silica to achieve a CO<sub>2</sub> uptake of 1.04 mmol g<sup>-1</sup> at 415 ppm of CO<sub>2</sub>.

Beyond silica, a variety of other support materials can also be functionalized with amines to fabricate efficient DAC adsorbents. A notable example is the PEI-grafted COF (COF-709) developed by Li *et al.* *via* an aromatic nucleophilic substitution reaction, forming C–S bonds between PEI and COF (Fig. 7c).<sup>76</sup> When tested with 400 ppm CO<sub>2</sub> at 25 °C, COF-709 exhibited a remarkable CO<sub>2</sub> uptake that was highly dependent on RH, increasing significantly from 0.48 mmol g<sup>-1</sup> under dry conditions to 1.24 mmol g<sup>-1</sup> at 75% RH (Fig. 7d), similar to amine-impregnated adsorbents.<sup>85,86</sup> <sup>13</sup>C NMR spectroscopy revealed that this enhancement was due to a shift of the adsorption products: while carbamate and carbamic acid formed under dry conditions, the presence of moisture promotes the formation of bicarbonate along with carbamate, increasing CO<sub>2</sub> capture capacity. Furthermore, the C–S bonds used to anchor PEI within the COF pores provided exceptional chemical stability, preventing amine loss (Fig. 7e). As a result, COF-709 exhibited excellent cycling stability, maintaining its CO<sub>2</sub> uptake without degradation over 10 consecutive adsorption–desorption cycles under simulated DAC conditions. In another work, the same group developed a polyamine-functionalized COF through a ring-





**Fig. 7** The influence of different support materials on CO<sub>2</sub> capture. (a) MIL-101(Cr) and (b)  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>. Reprinted with permission from ref. 84. Copyright 2023 American Chemical Society; (c) synthesis of PEI-grafted COF-709, (d) CO<sub>2</sub> uptake performance under varying RH conditions, and (e) stability over multiple CO<sub>2</sub> adsorption–desorption cycles using PEI-grafted COF-709 for DAC. Reprinted with permission from ref. 76. Copyright 2024 American Chemical Society.

opening polymerization reaction with aziridine, which exhibited similarly good performance for DAC.<sup>77</sup> These studies confirm the effectiveness and durability of amine-grafted COFs for efficient CO<sub>2</sub> capture in an open air environment.

In addition to amine-based adsorbents, Li *et al.* recently reported a new class of adsorbents known as “charged sorbents”. Such adsorbents (also named as PCS-OH) were synthesized by electrochemically inserting reactive hydroxide ions into a porous carbon (Fig. 8a).<sup>87</sup> The hydroxide ions



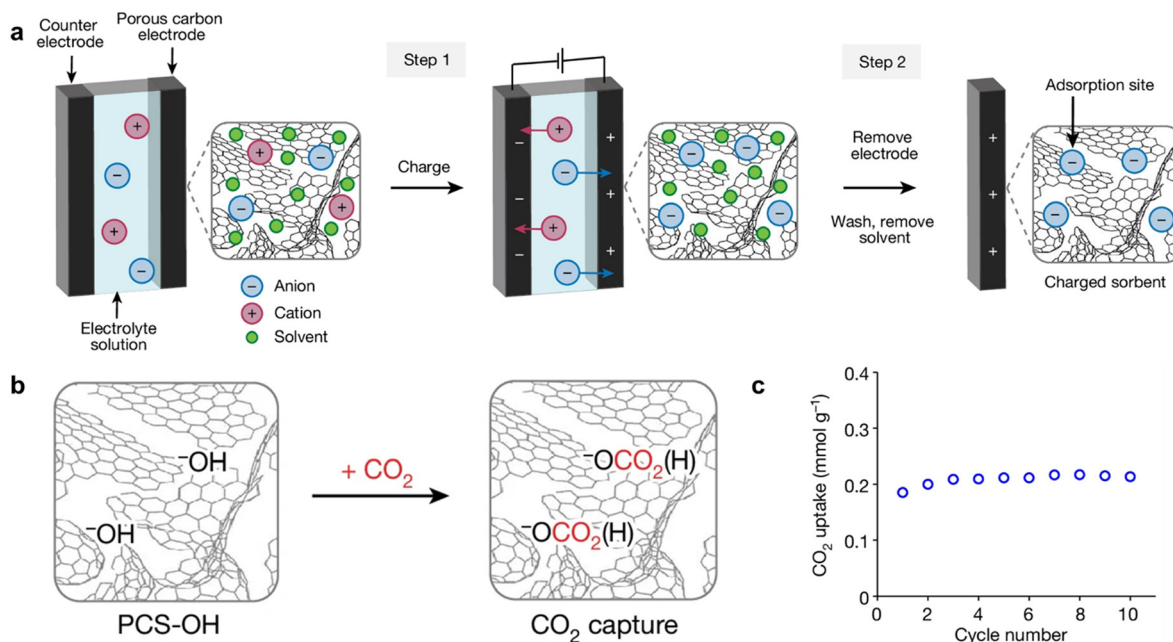


Fig. 8 Charged sorbents for DAC. (a) The preparation steps, (b) proposed mechanism for CO<sub>2</sub> capture, and (c) cycling capacities for ten CO<sub>2</sub> adsorption-desorption cycles over the charged sorbents. Reprinted with permission from ref. 87. Copyright 2024 Springer Nature.

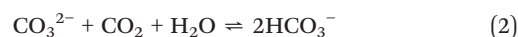
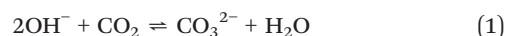
present in PCS-OH serve as reactive sites for selective CO<sub>2</sub> capture from air, forming (bi)carbonate species (Fig. 8b). PCS-OH was evaluated for DAC under simulated dry air with 400 ppm CO<sub>2</sub> at 30 °C, showing a stable CO<sub>2</sub> capacity of ~0.2 mmol g<sup>-1</sup> over 10 repeated adsorption and desorption cycles (Fig. 8c). It should be noted that the CO<sub>2</sub> capacity of PCS-OH is generally lower than amine-based adsorbents due to its limited hydroxide content, but this can be improved by optimizing the synthesis procedure to increase the hydroxide content in the adsorbents. This study lays the foundation for developing a new family of effective and low-cost adsorbents for DAC.

### 3.2 Liquid absorbents

In CO<sub>2</sub> capture by absorbent solutions, CO<sub>2</sub> first comes into contact and reacts with the absorbent, getting separated from air. Then, the CO<sub>2</sub>-rich absorbent solution is transported to the regeneration unit to release CO<sub>2</sub> and regenerate the absorbent, where the absorbent effluent can be reused for the next cycle of CO<sub>2</sub> capture. The absorption of CO<sub>2</sub> depends on an interfacial equilibrium between air and absorbent solutions. Thus, the capture capacity of CO<sub>2</sub> in the absorbent solution is influenced by the absorbent property as well as the air temperature and pressure. Common absorbents used in DAC are alkaline solutions, including inorganic and organic bases dissolved in water (Fig. 9a and b).

Hydroxide aqueous solutions (*e.g.*, NaOH, KOH) have been widely utilized to eliminate CO<sub>2</sub> from air in closed spaces like submarines and spacecraft. They can easily react with CO<sub>2</sub> to form carbonate, enabling selective CO<sub>2</sub> capture from air (eqn (1) and (2)). These alkaline solutions are extremely attractive as CO<sub>2</sub>

absorbents because of their excellent stability and affordable price. However, there are some drawbacks to be addressed when using them for large-scale applications. For instance, when Ca(OH)<sub>2</sub> comes into contact with CO<sub>2</sub>, it generates CaCO<sub>3</sub>, which may plug the packing and piping components of the air contactor, posing challenges for long-term operation. Furthermore, the limited solubility of Ca(OH)<sub>2</sub> in water results in a low CO<sub>2</sub> capture capacity. As for other hydroxide solutions (*e.g.*, NaOH and KOH), the regeneration process requires extremely high temperatures (*e.g.*, >340 °C for Na<sub>2</sub>CO<sub>3</sub> solution), leading to the complete boiling of the aqueous solution.<sup>89</sup> Thus, the NaOH and KOH absorbent solutions after CO<sub>2</sub> capture are typically subjected to reaction with Ca(OH)<sub>2</sub> to regenerate the absorbent and form CaCO<sub>3</sub>, which is then calcined to release CO<sub>2</sub>. It should be emphasized that this calcination process occurs at even higher temperatures (≥700 °C), resulting in large energy penalty.<sup>90</sup> The mixture of carbonate and hydroxide solutions (*e.g.*, K<sub>2</sub>CO<sub>3</sub>/KOH, Na<sub>2</sub>CO<sub>3</sub>/NaOH) has also been used for DAC, because mixing carbonate and hydroxide solutions creates a synergistic system, in which hydroxides ensure fast CO<sub>2</sub> capture kinetics, while carbonates offer low corrosivity.<sup>91</sup> However, the mixture also encounters challenges similar to those associated with pure hydroxide solutions.<sup>92,93</sup>



Amine solutions are well-established absorbents for CO<sub>2</sub> capture mostly from flue gas. Compared to hydroxide solutions, the amine regeneration process is milder, usually at lower



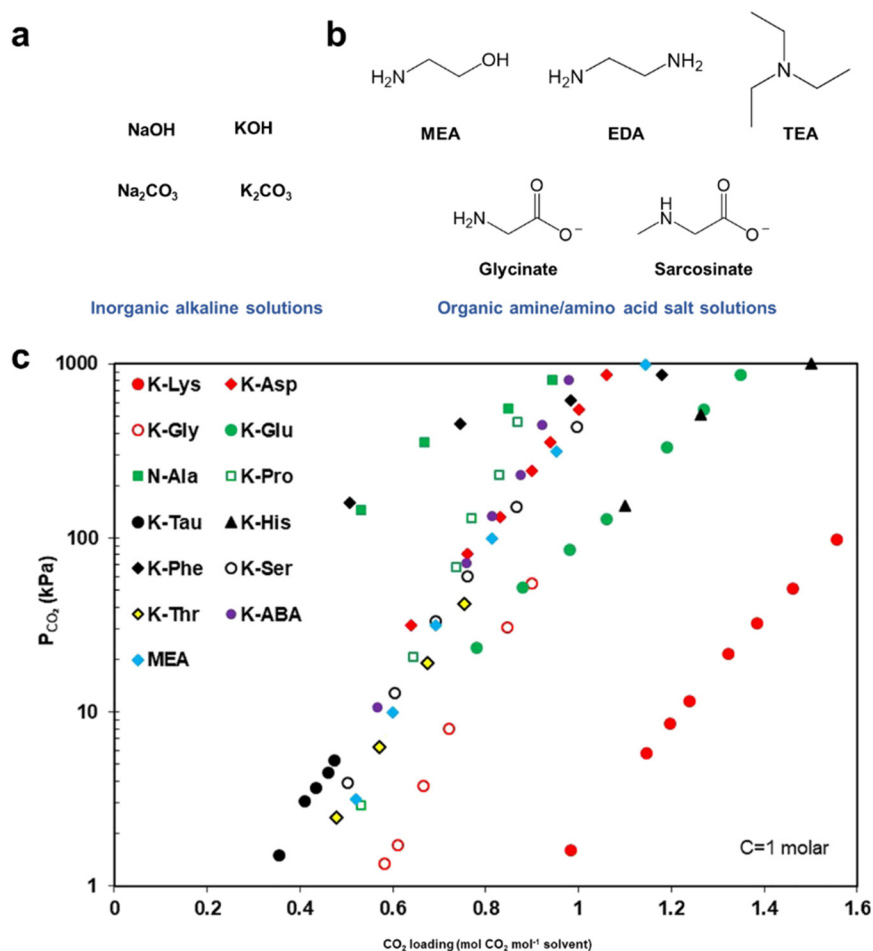
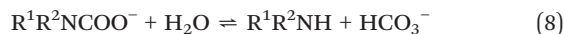
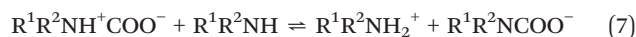
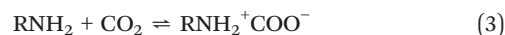


Fig. 9 Two common types of liquid absorbents: (a) inorganic alkaline solutions and (b) organic amine and amino acid salt solutions; (c) the CO<sub>2</sub> loading capacity of amino acid salt and MEA solutions at 313 K. Reprinted with permission from ref. 88. Copyright 2022 De Gruyter.

temperatures (*i.e.*, 80–120 °C), thereby featuring lower energy consumption. Commonly used amines include monoethanolamine (MEA), ethylenediamine, triethylamine, *etc.*<sup>94</sup> CO<sub>2</sub> absorption by amine occurs *via* the zwitterion mechanism, where the amine reacts with CO<sub>2</sub> to form protonated amine and carbamate/bicarbonate. Both primary and secondary amines undergo similar reactions to form carbamate (eqn (3), (4), (6) and (7)), with a maximum theoretical CO<sub>2</sub> uptake of 0.5 mol CO<sub>2</sub> per mol amine. However, a certain carbamate can undergo hydrolysis in the presence of water to form bicarbonate (eqn (5) and (8)), which can slightly increase the CO<sub>2</sub> uptake. In contrast, tertiary amines have a much higher theoretical CO<sub>2</sub> loading (1.0 mol CO<sub>2</sub> per mol amine) because they cannot directly capture CO<sub>2</sub> but instead act as catalysts to promote the hydrolysis reaction of CO<sub>2</sub> (eqn (9)). Thus, the performance of amines for CO<sub>2</sub> capture is highly dependent on their chemical structures. Barzagli *et al.* carried out a screening study on the performance of different aqueous amine solutions for DAC.<sup>95</sup> They found that unhindered primary amines, such as MEA and isopropanolamine, are the most effective absorbents for DAC, as efficient as aqueous hydroxides. This high efficiency was attributed to the formation of a high-

concentration carbamate. In contrast, tertiary amines show poorer performance as they cannot form carbamate, but instead rely on the slower kinetics of bicarbonate formation.<sup>96</sup> Interestingly, aqueous 2-amino-2-methyl-1-propanol (AMP), an efficient flue gas absorbent, was found unsuitable for capturing extremely diluted CO<sub>2</sub>, highlighting the differences between DAC and conventional CO<sub>2</sub> capture. Therefore, the optimal absorbents for point-source capture are not necessarily the best choice for DAC.





Indeed, many amine-based absorbents are unsuitable for DAC due to their oxidative degradation in the capture of low concentration CO<sub>2</sub>. On the one hand, amine degradation compromises carbon capture stability, particularly in the presence of high concentration of oxygen.<sup>86</sup> On the other hand, amine degradation products are directly released into the atmosphere.<sup>97</sup> In DAC systems with large air flow, these emissions may lead to severe environmental impacts. Consequently, most amine-based absorption solvents are primarily utilized in flue gas capture rather than DAC.

Amino acid salts (AAS) are promising alternative absorbents to traditional amines, showing multiple advantages such as low toxicity, low volatility, antioxidant properties, resistance to thermal degradation, and fast reaction kinetics with CO<sub>2</sub>.<sup>98,99</sup> Currently, the most extensively studied AAS for DAC applications include glycine (Gly), lysine, sarcosine, leucine, valine, phenylalanine, and arginine.<sup>100–103</sup> AAS absorbent solutions are typically prepared by mixing amino acids with a hydroxide solution like NaOH or KOH.<sup>104</sup> Similar reactions occur when using AAS for DAC (eqn (10) and (11)), but AAS solutions are slightly more alkaline than common amine solutions due to the presence of both amino and carboxylate groups. Fig. 9c shows that many AAS absorbent solutions, such as lysinate, glycinate, histidinate, and glutamate, exhibit superior CO<sub>2</sub> capture capacity compared to the conventional MEA solution.<sup>88</sup> In a following study, McQuillan *et al.* found that K-Gly exhibited comparable CO<sub>2</sub> uptake to MEA in DAC but showed lower energy consumption for regeneration.<sup>105</sup> More recently, Xiao *et al.* compared the stability of AAS and amine absorbents in air.<sup>106</sup> After exposure in air for 7 days, 3 M K-Gly showed a degradation rate of 16.7 mmol per mole K-Gly, while the degradation rate of common amines (3 M MEA and AMP) approximately doubled (*e.g.*, 29.2 mmol per mol MEA and 31.6 mmol per mol AMP). For these advantages, increasing efforts are directed to exploring AAS for DAC.



## 4 CO<sub>2</sub> release and capture agent regeneration

CO<sub>2</sub> release accompanied by adsorbent/absorbent regeneration is a critical step in continuous DAC processes. In general, the methods used for CO<sub>2</sub> release and adsorbent/absorbent regeneration in DAC show some similarity to those in flue gas carbon capture. However, differences also exist due to the significantly lower CO<sub>2</sub> uptake in DAC-based systems as a result of low atmospheric CO<sub>2</sub> concentration. Currently, the main approaches to release CO<sub>2</sub> from DAC systems are temperature swing and precipitation-phase separation, which are deployed by two representative DAC companies, Climeworks and Carbon

Engineering, respectively. With the growing emphasis on sustainable energy, electrochemical methods are emerging as a compelling option for CO<sub>2</sub> release due to their advantages of using renewable electricity, controllable reaction rates, and mild operation conditions. In this section, we will scrutinize these regeneration methods.

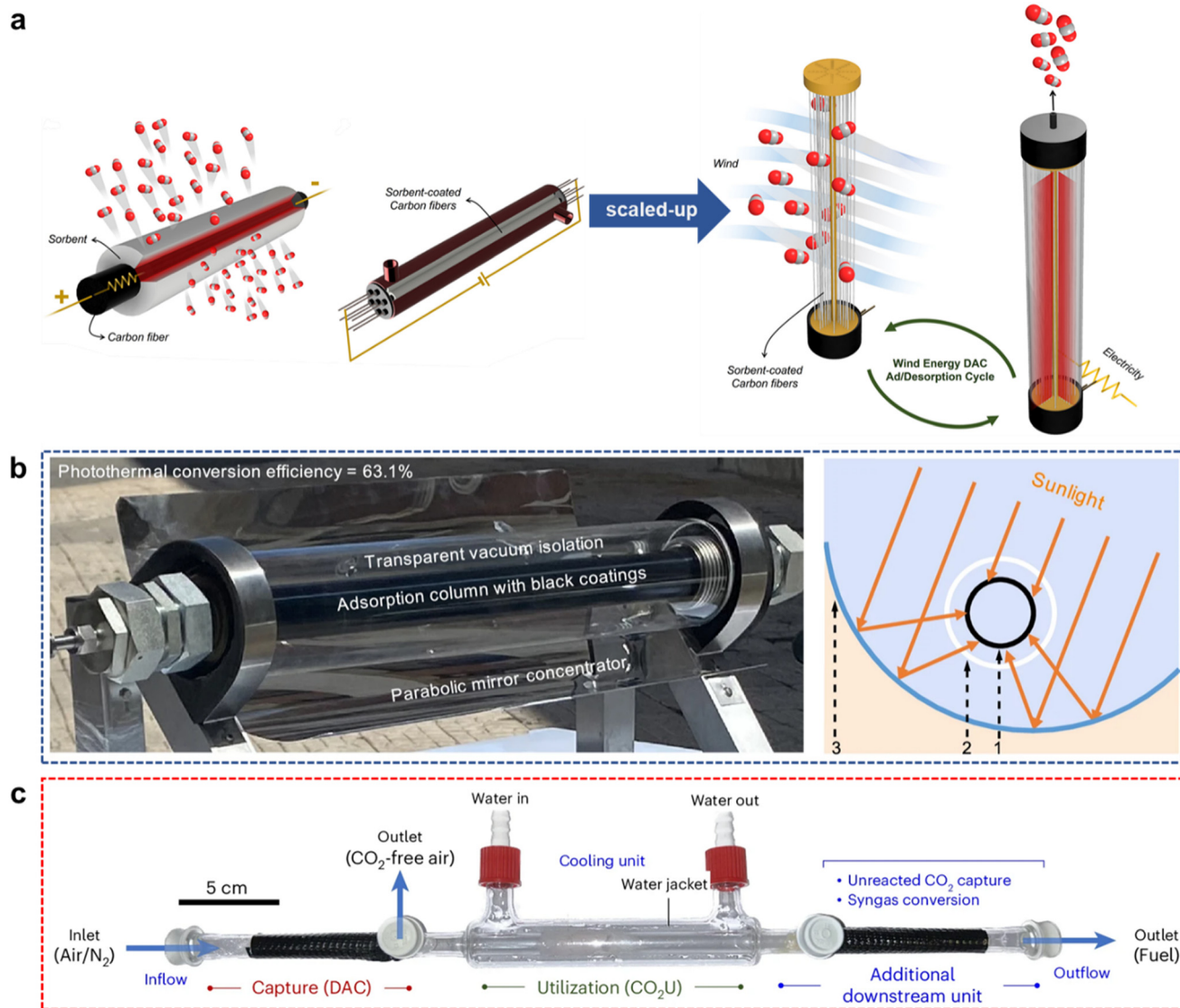
### 4.1 Temperature swing

Thermal desorption based on temperature swing is a common method for CO<sub>2</sub> release and regeneration of solid adsorbents. At elevated temperatures, the bond energy between CO<sub>2</sub> and the adsorbent is weakened, facilitating the release of CO<sub>2</sub> gas. In temperature swing adsorption (TSA) systems, CO<sub>2</sub> capture occurs under ambient conditions, while CO<sub>2</sub> release operates at elevated temperatures (100–120 °C).<sup>107</sup> The desorption temperature is the most critical parameter in TSA, directly influencing the energy consumption of the systems. Generally, CO<sub>2</sub> release at higher temperature leads to higher energy consumption ( $\geq 6.0$  GJ ton<sub>CO<sub>2</sub></sub><sup>-1</sup>).<sup>38</sup>

To lower the operation temperature, TSA can be integrated with vacuum systems, which is known as temperature vacuum swing adsorption (TVSA).<sup>108</sup> As a result, TVSA shows reduced energy consumption and improved stability for cyclic CO<sub>2</sub> capture. A good application of the TVSA process in DAC is demonstrated by Climeworks. Their system operates at temperatures below 100 °C with an energy consumption of 5.40–7.20 GJ ton<sub>CO<sub>2</sub></sub><sup>-1</sup>.<sup>109</sup> Similar energy consumption (4.20–8.60 GJ ton<sub>CO<sub>2</sub></sub><sup>-1</sup>) for TVSA was also reported in the literature.<sup>110</sup> Recently, the company has introduced a new generation of DAC technology, which incorporates novel filter materials and is reported to reduce energy consumption by roughly 50% compared to earlier systems.<sup>111</sup> This upgraded system is planned for deployment in the United States, aiming at reducing the DAC cost to \$250–300 per ton<sub>CO<sub>2</sub></sub><sup>-1</sup> by 2030.

It has to be noted that the TSA/TVSA system requires closing the inlet and outlet during CO<sub>2</sub> release and heating the entire interior space of the air contactor, leading to unnecessary heat loss. To further optimize the system, researchers have developed a novel electrically driven temperature swing adsorption (e-TSA) method, which can minimize the heat loss as it employs joule heating (also known as resistive heating) to generate thermal energy only on the conductor or the material itself to release CO<sub>2</sub>.<sup>112,113</sup> This method allows the heat to be transferred directly from the inside to the outside of the adsorbent, thereby enhancing the energy efficiency of the temperature swing. For example, Lee *et al.* developed a DAC module made of a PEI-impregnated adsorbent-coated carbon fiber, which reached the CO<sub>2</sub> release temperature within one minute when a 7 V voltage was applied (Fig. 10a).<sup>114</sup> This DAC module released CO<sub>2</sub> and regenerated the adsorbent six times faster than the traditional TSA thermal desorption, with an energy consumption of 7.20 GJ ton<sub>CO<sub>2</sub></sub><sup>-1</sup> and only 7% of the total heating lost to the surrounding environment. Later, Li *et al.* employed joule heating at a voltage of 7–8 V to release CO<sub>2</sub> from “charged sorbents” at low temperatures of 90–100 °C, showing an energy





**Fig. 10** (a) An electrically driven temperature swing adsorption module for DAC. Reprinted with permission from ref. 114. Copyright 2023 Elsevier; (b) solar-driven CO<sub>2</sub> release coupled with thermal adsorbent regeneration. Reprinted with permission from ref. 116. Copyright 2024 Springer Nature; (c) solar-assisted DAC system enabling *in situ* CO<sub>2</sub> conversion. Reprinted with permission from ref. 117. Copyright 2025 Springer Nature.

consumption of 6.50 GJ ton<sub>CO<sub>2</sub></sub><sup>-1</sup> under 11% RH.<sup>87</sup> More recently, Lee *et al.* established an electrothermal-vacuum swing adsorption (e-TVSA) process by integrating joule heating with a vacuum system to release CO<sub>2</sub> from an ethylenediamine-grafted Y zeolite (EDA-Y) adsorbent.<sup>115</sup> In this case, a significantly reduced voltage of 3 V was sufficient to release CO<sub>2</sub> and regenerate the adsorbent, resulting a lower energy consumption of 6.1 GJ ton<sub>CO<sub>2</sub></sub><sup>-1</sup>.

Solar concentrators offer a way of direct use of solar energy for thermal regeneration of adsorbents. A solar-driven DAC system developed by Wang *et al.* accomplished near-complete CO<sub>2</sub> release (~100%) in 50 minutes under 500–700 W m<sup>-2</sup> solar irradiance with 63.1% photothermal conversion efficiency (Fig. 10b).<sup>116</sup> Kar *et al.* reported a similar CO<sub>2</sub> release method, compatible with most commercial adsorbents regenerable at 80–100 °C.<sup>117</sup> Additionally, they explored *in situ* CO<sub>2</sub> conversion

with the DAC system, showing a maximum yield of ~24 μmol g<sub>TiO<sub>2</sub></sub><sup>-1</sup> for syngas production *via* solar photocatalysis (Fig. 10c). These two studies demonstrate the feasibility of integrating renewable energy into the process of CO<sub>2</sub> release and adsorbent regeneration. Solar-driven CO<sub>2</sub> release/adsorbent regeneration systems enable decentralized deployment in remote locations without extensive thermal infrastructure, positioning them as a promising DAC technology.

For the regeneration of liquid absorbents, direct heating of the CO<sub>2</sub>-loaded absorbent solutions is commonly applied to organic amines but is unsuitable for inorganic hydroxide solutions due to the high temperatures required.<sup>118,119</sup> Although organic amine solutions have been widely deployed in flue gas CO<sub>2</sub> capture, they are prone to oxidative degradation in air, which explains the scarce examples of their applications in DAC. AAS solutions are more stable and employed as promising



absorbents for CO<sub>2</sub> capture from air. After CO<sub>2</sub> capture, AAS solutions can be regenerated by heating them to a certain temperature (70–90 °C) at a lowered pressure (21 kPa).<sup>41</sup> However, the direct heating of absorbent solutions faces challenges such as solvent evaporation, oxidative degradation, and the need to heat large volumes of liquid.<sup>120,121</sup> Therefore, new technologies have been developed to regenerate liquid absorbents in DAC (*vide infra*).

#### 4.2 Precipitation-phase separation

DAC systems based on aqueous sodium/potassium hydroxide solutions usually rely on an additional calcium cycle to release the captured CO<sub>2</sub> and regenerate the sodium/potassium hydroxide solutions. This is a typical precipitation-phase separation method that precipitates the CO<sub>2</sub> captured, significantly reducing the volume or mass of substance for follow-up thermal treatment.

Fig. 11a shows a potassium–calcium loop for CO<sub>2</sub> release in the DAC process developed by Carbon Engineering.<sup>39</sup> In this process, a K<sub>2</sub>CO<sub>3</sub> solution formed after CO<sub>2</sub> capture by KOH solutions reacts with Ca(OH)<sub>2</sub> to regenerate KOH and yield the CaCO<sub>3</sub> precipitate, which is separated and calcined at 900 °C to release gaseous CO<sub>2</sub> and produce solid CaO. Ca(OH)<sub>2</sub> is then regenerated through hydration of CaO. The major challenge of the potassium–calcium loop is its high energy intensity and complex operation steps. The regeneration energy of this process is estimated to be 5.25–8.81 GJ ton<sub>CO<sub>2</sub></sub><sup>-1</sup>, which represents 80% of the total energy consumption in the DAC system.

In order to reduce the energy consumption, Brethome *et al.* developed a new precipitation-phase separation system relying on the reaction between CO<sub>2</sub>-loaded AAS aqueous solution (glycinate and sarcosinate) and a guanidine compound (2,6-pyridinebis(iminoguanidine), PyBIG), which forms insoluble

carbonate crystals and regenerates the AAS absorbent (Fig. 11b).<sup>122</sup> The carbonate crystals can decompose at mild temperatures (80–120 °C) to release CO<sub>2</sub> and regenerate PyBIG at a percentage of 92%. Liu and co-workers uncovered the structural evolution and decomposition mechanism of the GBIG-bicarbonate (GBIGH<sub>2</sub>(CO<sub>3</sub>)<sub>2</sub>(H<sub>2</sub>O)<sub>2</sub>) crystal formed by the reaction of CO<sub>2</sub>-loaded AAS with glyoxal-bis(iminoguanidine) (GBIG) during the CO<sub>2</sub> release process using *in situ* wide-angle X-ray scattering, attenuated total reflectance-Fourier transform infrared spectroscopy, and other measurements.<sup>123</sup> Their findings indicate that CO<sub>2</sub> and water molecules are released simultaneously from the GBIG-bicarbonate crystal during the thermal regeneration process at 97–134 °C.

The guanidine compound can also be utilized directly to capture CO<sub>2</sub> without the use of AAS. For instance, Cai *et al.* reported a trichelating iminoguanidine (BTIG) for DAC.<sup>124</sup> Unlike previous works, this guanidine can directly and strongly bind with CO<sub>2</sub> to form insoluble carbonate crystals without the aid of AAS. In the reversible process, BTIG-CO<sub>2</sub> crystals release CO<sub>2</sub> and regenerate the guanidine at mild temperatures (60–150 °C), showing much lower energy consumption (2.27 GJ ton<sub>CO<sub>2</sub></sub><sup>-1</sup>) than CaCO<sub>3</sub> calcination.

#### 4.3 Electrolysis

In recent years, electrolysis has shown significant potential in CO<sub>2</sub> release, as it avoids external heating or vacuum conditions. Electrolysis exhibits excellent compatibility with absorbents that are sensitive to heating and can be applied in various scenarios for CO<sub>2</sub> removal, particularly in DAC. There are two reasons for this. Firstly, the modular design of electrolyzers allows them to be deployed in diverse environments, including urban, industrial, and remote areas, which aligns with the geographically flexible nature of the DAC technologies. Secondly, the electrochemical equipment

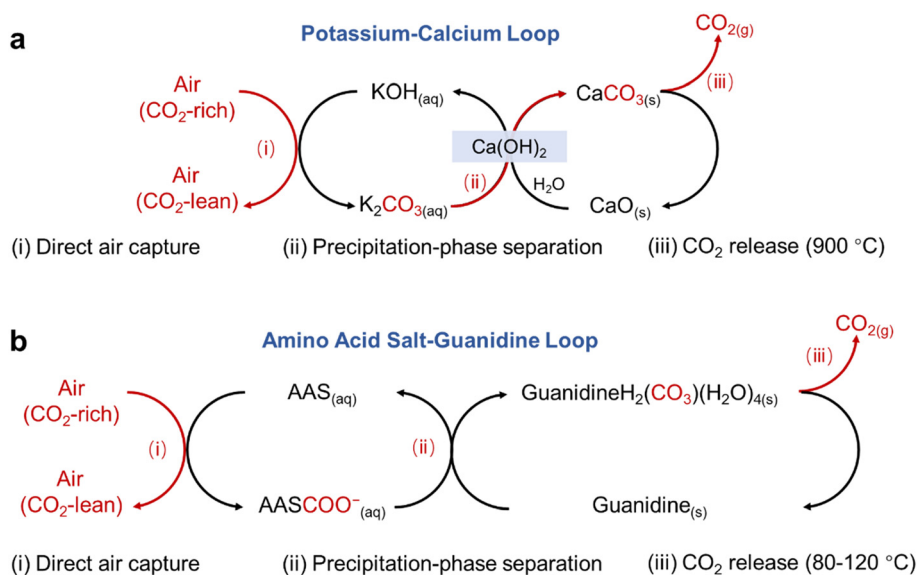
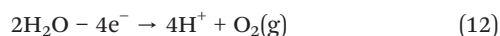


Fig. 11 CO<sub>2</sub> release and absorbent regeneration by precipitation-phase separation. (a) Potassium–calcium loop; (b) AAS–guanidine loop.



is easy to maintain and can be connected to renewable energy sources, making the process of CO<sub>2</sub> release and absorbent regeneration more sustainable and cost-effective. The methods for electrochemical CO<sub>2</sub> release mainly include pH swing and redox mediation.

**4.3.1 pH swing.** The pH determines the carbon species in various absorbent solutions, with acidification leading to CO<sub>2</sub> release and basification resulting in CO<sub>2</sub> capture. Thus, pH swing can mediate CO<sub>2</sub> capture/release based on the redox reaction of water (eqn (12) and (13)). At the anode, H<sub>2</sub>O is oxidized and loses electrons to produce protons and oxygen, which lowers the solution pH and facilitates CO<sub>2</sub> release. Concurrently, H<sub>2</sub>O is reduced to yield OH<sup>-</sup> and H<sub>2</sub> at the cathode, causing an increase in pH of the solution, which can then be used for CO<sub>2</sub> capture. For instance, Kim *et al.* utilized a classic pH-swing electrochemical system for recovery of LiOH from spent CO<sub>2</sub> adsorbents.<sup>125</sup> In this setup, electrocatalytic water-splitting electrodes are separated by a cation exchange membrane. This configuration enables lithium ions to selectively migrate from the oxygen-evolving anode chamber to the hydrogen-evolving cathode chamber, thereby directly producing a LiOH solution in a single step. Ellis *et al.* employed the pH-swing process to decompose calcium carbonate coming from CO<sub>2</sub> mineralization for CO<sub>2</sub> release, thereby avoiding the high temperature calcination.<sup>126</sup> However, the pH swing based on water splitting also produces a substantial amount of O<sub>2</sub> at the anode, forming a mixture with released CO<sub>2</sub>. Thus, an additional step for O<sub>2</sub> separation is required to obtain pure CO<sub>2</sub>.



The pH swing performance can be significantly improved using bipolar membrane electrodialysis (BPMED). The core of this system is the bipolar membrane (BPM), which is constituted by a cation- and an anion-exchange layer. When applying a potential, the BPM dissociates water into H<sup>+</sup> and OH<sup>-</sup>, generating a sharp pH gradient across the membrane (Fig. 12a). A typical BPMED stack comprises multiple BPMs alternating with anion exchange membranes (AEMs), with acidification and basification chambers in between. During operation, carbonate/bicarbonate ions migrate through the AEM into the acidification chamber, where H<sup>+</sup> triggers CO<sub>2</sub> release. Simultaneously, OH<sup>-</sup> in the basification chamber regenerates the absorbent solution.

In an early work, Eisaman *et al.* reported a BPMED stack for releasing CO<sub>2</sub> from potassium carbonate and bicarbonate solutions, which allows for CO<sub>2</sub> release and absorbent regeneration at the energy of 100 to 200 kJ mol<sub>CO<sub>2</sub></sub><sup>-1</sup> (*i.e.*, 2.27 to 4.54 GJ ton<sub>CO<sub>2</sub></sub><sup>-1</sup>) at the low current density (5 mA cm<sup>-2</sup>).<sup>127</sup> This work utilized an electrolyzer stack composed of seven unit cells, with the AEM reaching 0.14 m<sup>2</sup>, demonstrating the scalability potential of this technology. Recently, Castaño *et al.* systematically investigated the influence of parameters such as

current density on the performance of a BPMED.<sup>128</sup> Among the tested current densities, 100 mA cm<sup>-2</sup> showed the highest CO<sub>2</sub> desorption efficiency but also the highest energy consumption, whereas 25 mA cm<sup>-2</sup> exhibited the lowest energy consumption (8.8 GJ ton<sub>CO<sub>2</sub></sub><sup>-1</sup>) but lower CO<sub>2</sub> desorption. These results reveal that the current density is a key factor governing the overall performance of BPMED, affecting both CO<sub>2</sub> release efficiency and energy consumption.

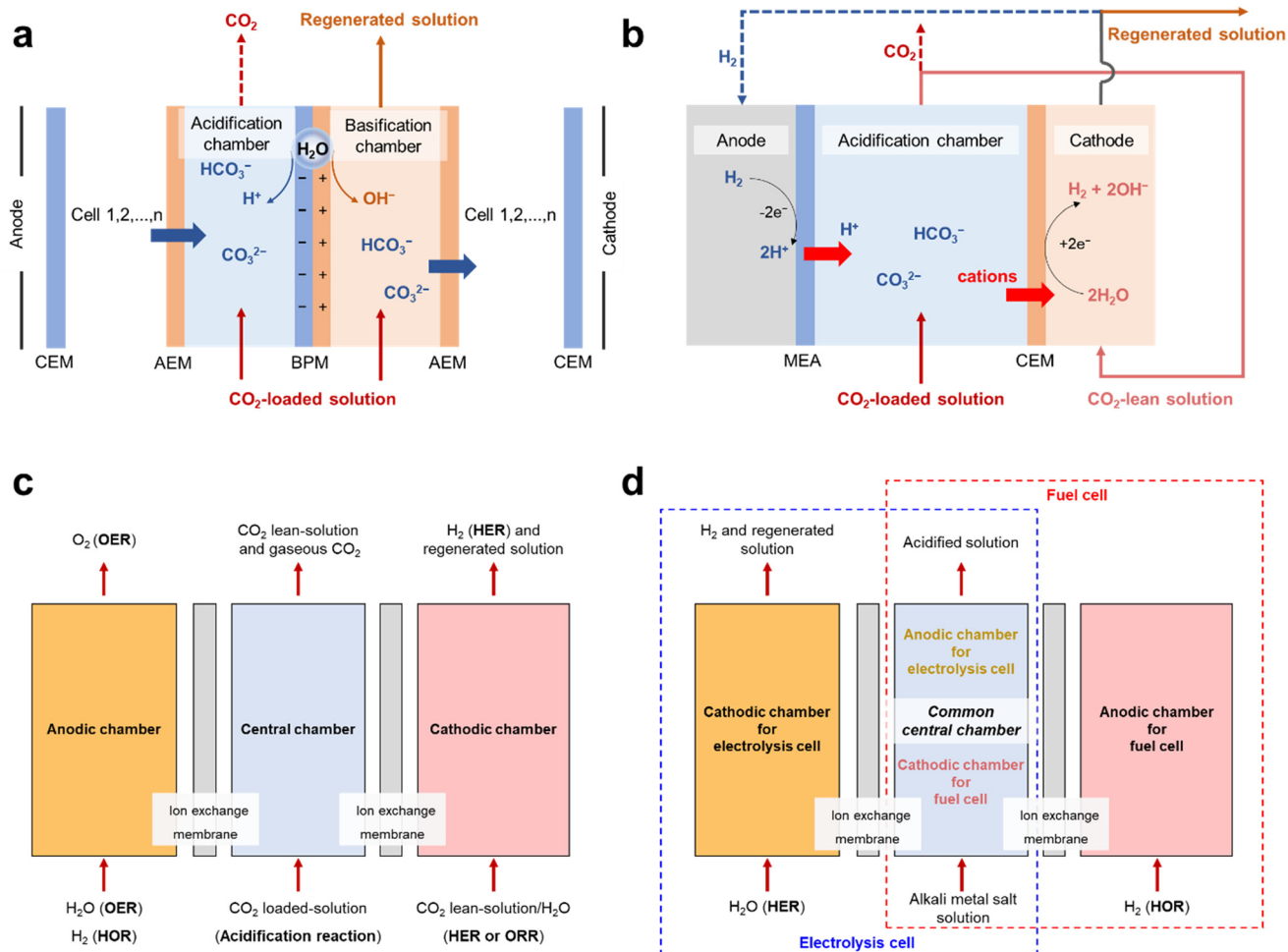
The main challenge of BPMED technology is its high energy consumption and high expense of ion exchange membranes. Sabatino *et al.* conducted a techno-economic analysis of BPMED and proposed that future development should be aimed at enhancing the electrical conductivity and overall stability of ion exchange membranes.<sup>129</sup> With more durable and cost-effective membranes, the total cost of BPMED could potentially fall below \$250 per ton<sub>CO<sub>2</sub></sub><sup>-1</sup>.

Fig. 12b shows another efficient system for electrochemical CO<sub>2</sub> release and absorbent regeneration, called three-chamber electrolysis system comprising cathode, acidification, and anode chambers.<sup>130</sup> In this system, the anode oxidizes H<sub>2</sub> to produce H<sup>+</sup>, which is transferred through the ion exchange membrane to the acidification chamber, while the cathode produces OH<sup>-</sup> by water reduction to regenerate the absorbent solution and H<sub>2</sub>. Notably, this system avoids the generation of O<sub>2</sub> from water reduction and instead utilizes the hydrogen generated at the cathode for proton supply. In the chemical industry, the storage and transportation of H<sub>2</sub> often incur more costs and safety risks. This system addresses these challenges by effectively utilizing H<sub>2</sub>, making it particularly promising for application.

For example, Shu *et al.* employed amine-functionalized ion-exchange resins for DAC and utilized a three-chamber electrolytic cell for CO<sub>2</sub> release.<sup>131</sup> This study reported a very high energy consumption of 537 ± 33 kJ mol<sub>CO<sub>2</sub></sub><sup>-1</sup> (*i.e.*, 12.20 ± 0.75 GJ ton<sub>CO<sub>2</sub></sub><sup>-1</sup>) at a current density of 20 mA cm<sup>-2</sup>, which was attributed to the large electrode overpotential of the cell. Zheng *et al.* also employed a similar three-chamber electrolytic cell to release CO<sub>2</sub> with a lower energy consumption of 5.20–8.08 GJ ton<sub>CO<sub>2</sub></sub><sup>-1</sup> and a high CO<sub>2</sub> purity of ≥98% from a CO<sub>2</sub>-loaded AAS solution.<sup>52</sup> It was found that reducing the thickness of the acidification chamber from 5 mm to 1.5 mm led to an approximately 70% reduction in energy consumption. Recently, Zhang *et al.* proposed a different three-chamber electrolysis system for CO<sub>2</sub> release from 0.5 M NaHCO<sub>3</sub> solution.<sup>132</sup> This system showed a very low energy consumption of 118 kJ mol<sub>CO<sub>2</sub></sub><sup>-1</sup> (*i.e.*, 2.68 GJ ton<sub>CO<sub>2</sub></sub><sup>-1</sup>) at 100 mA cm<sup>-2</sup> due to the use of a thinner solid electrolyte, instead of the acidification chamber. The solid electrolyte allowed the cell voltage to remain stable below 2 V when the current density was doubled from 100 to 200 mA cm<sup>-2</sup>. Without this solid interlayer, the voltage surged beyond 8 V even at current densities below 100 mA cm<sup>-2</sup>.

Xu *et al.* reported an alternative three-chamber electrolytic system that switches between the electrolytic cell and the fuel cell in the same middle chamber by applying voltage alternately.<sup>133</sup> This system released CO<sub>2</sub> from Li<sub>2</sub>CO<sub>3</sub> solution, with an energy consumption of 6.40 GJ ton<sub>CO<sub>2</sub></sub><sup>-1</sup> at a current density of 100 mA cm<sup>-2</sup>. To optimize the system, Liu *et al.*





**Fig. 12** Divergent pH swing methods for CO<sub>2</sub> release and absorbent regeneration: (a) bipolar membrane electrodiolysis and (b) three-compartment electrolytic cell; two types of three-chamber electrolytic cells: (c) synchronous reaction type, in which all three chambers operate simultaneously and (d) alternating reaction type, in which two electrolytic cells share one central chamber.

combined two electrolytic cells into a three-chamber structure, utilizing cyclic viologen electrocatalysis to facilitate CO<sub>2</sub> release.<sup>134</sup> This system could operate stably for 200 hours with a reduced energy consumption of 3.80 GJ ton<sub>CO<sub>2</sub></sub><sup>-1</sup>.

It is important to note the distinct structural and functional differences between the two and three-chamber electrolytic cells. In the first configuration, the anode and cathode are physically separated by two ion-exchange membranes with a central chamber between them, allowing the chambers to operate independently (Fig. 12c).<sup>52,132</sup> This configuration is well-suited for continuous-flow systems, offering benefits in operational stability and simplicity. In contrast, the second design employs current switching to control all chambers, enabling operational transitions between electrolysis and fuel cell modes (Fig. 12d).<sup>133,134</sup> This approach necessitates a more complex voltage control system, and its practical stability remains unclear and requires further validation.

**4.3.2 Redox mediation.** Redox-mediated electrochemical CO<sub>2</sub> capture and release rely on absorbents/adsorbents with redox-active couples, such as quinone and phenazine. In this process, the redox state of the mediator (redox couple) is

reversibly altered by applying voltages, triggering the release or binding of CO<sub>2</sub>. This method offers a significant advantage over traditional pH-swing by substantially lowering energy consumption.

Quinones are the most widely studied compounds for electrochemical CO<sub>2</sub> capture and release. The mechanism of quinone in CO<sub>2</sub> capture and release is shown in Fig. 13a. Generally, quinone undergoes a proton coupled electron transfer reaction in a neutral aprotic electrolyte. At reduction potentials, quinone is reduced to semiquinone and radical anions. As the reduction potential becomes more negative, the radical anions are reduced further to form hydroquinone and divalent radical anions, which are capable of binding CO<sub>2</sub>. In contrast, during the oxidation, CO<sub>2</sub>-loaded quinones release CO<sub>2</sub> to regenerate hydroquinone and divalent radical anions. The capture and release of CO<sub>2</sub> occur upon reduction and oxidation, respectively, thereby constituting a complete electrochemical carbon capture cycle.

In a protic medium (*e.g.*, water), the formation of hydroquinone becomes more favorable than CO<sub>2</sub> binding, making CO<sub>2</sub> capture inefficient. To address this issue, Liu *et al.*



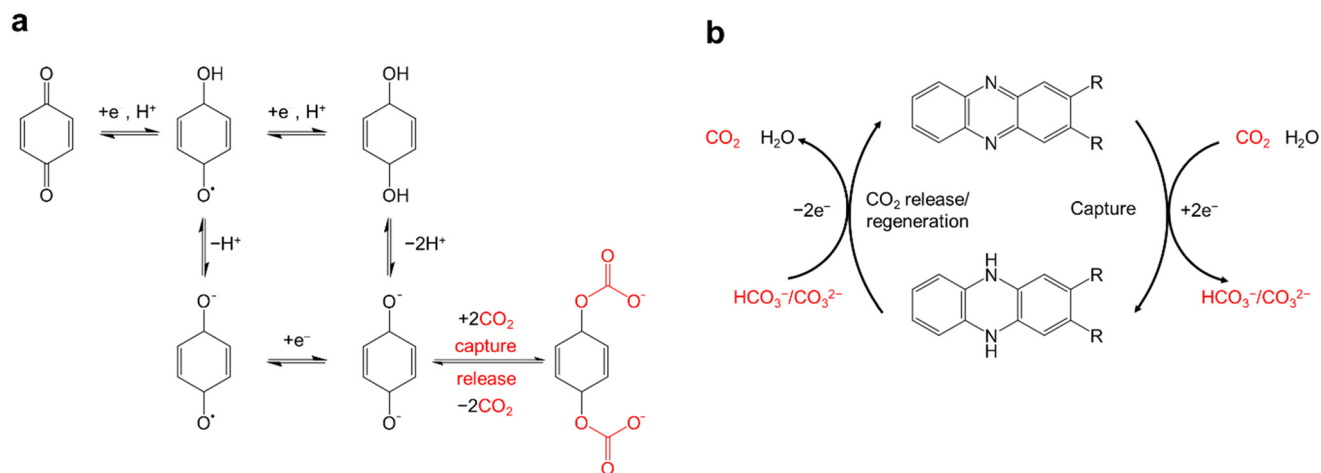


Fig. 13 Redox-mediated CO<sub>2</sub> capture and release using (a) quinone and (b) phenazine compounds.

proposed to use salt-concentrated aqueous media.<sup>135</sup> In this electrolyte, the mass and volume of dissolved salts exceed those of water, resulting in the absence of free water, which effectively suppresses water molecule activity and extends the electrochemical stability window. Compared to conventional diluted quinones, this approach demonstrates enhanced thermodynamic reactivity toward CO<sub>2</sub> in salt-concentrated electrolytes, achieving a high CO<sub>2</sub> capture uptake (8.9 mmol g<sup>-1</sup>) and low regeneration energy consumption of 56 kJ mol<sub>CO<sub>2</sub></sub><sup>-1</sup> (i.e., 1.27 GJ ton<sub>CO<sub>2</sub></sub><sup>-1</sup>). Voskian and co-workers demonstrated that quinones can also be made into electrode materials for DAC.<sup>136</sup> The adsorbent consisting of polyanthraquinone and carbon nanotubes captures CO<sub>2</sub> during charging while releasing it during discharging. A preliminary economic analysis revealed the high potential of this system, with costs ranging from \$50–100 per ton<sub>CO<sub>2</sub></sub><sup>-1</sup>.

Phenazine is an alternative redox mediator for CO<sub>2</sub> capture and release. During reduction (Fig. 13b), phenazine is hydrogenated by protons from water, producing OH<sup>-</sup> that captures CO<sub>2</sub>, forming HCO<sub>3</sub><sup>-</sup> and CO<sub>3</sub><sup>2-</sup>. In the oxidation process, the hydrogenated phenazine is oxidized to produce H<sup>+</sup>, which then reacts with HCO<sub>3</sub><sup>-</sup> and CO<sub>3</sub><sup>2-</sup> to release gaseous CO<sub>2</sub> and regenerate phenazine. Xie *et al.* developed a CO<sub>2</sub> capture/release cell utilizing a sulfonated phenazine derivative, which changed pH in aqueous NaHCO<sub>3</sub>/Na<sub>2</sub>CO<sub>3</sub> electrolyte to facilitate the release of CO<sub>2</sub> and the regeneration of the absorbent solution.<sup>137</sup> The system showed a faradaic efficiency of 95.8% at 10 mA cm<sup>-2</sup>, with an electrolytic energy consumption as low as 21.6 kJ mol<sub>CO<sub>2</sub></sub><sup>-1</sup> (i.e., 0.49 GJ ton<sub>CO<sub>2</sub></sub><sup>-1</sup>). Pang *et al.* reported an electrochemical cell based on 2,2'-(phenazine-1,8-diyl)bis(ethane-1-sulfonate) for CO<sub>2</sub> capture and release, achieving an energy consumption of 36–55 kJ mol<sub>CO<sub>2</sub></sub><sup>-1</sup> (i.e., 0.82–1.25 GJ ton<sub>CO<sub>2</sub></sub><sup>-1</sup>).<sup>138</sup> Notably, this system exhibits excellent charge and discharge capabilities, with a remarkably low degradation rate (less than 0.01% per day), enabling flexible adjustment of the capture and release time in response to fluctuations in the electricity market. In principle, the

phenazine mediated electrolysis for CO<sub>2</sub> capture/release is similar to the pH swing, but its energy consumption is significantly reduced as a result of the phenazine mediation.

We make a table to have a better comparison of the presented methods for CO<sub>2</sub> release and capture agent regeneration (Table 3). As a mature technology validated in large-scale DAC applications, TSA/TVSA can operate under mild conditions (~100 °C), but it still features moderate to high energy consumption, which is primarily attributed to the thermal energy required for desorption. As an emerging alternative, e-TSA/e-TVSA shows great promise for adsorbent regeneration due to its high efficiency and rapid response, yet it currently exhibits relatively high overall energy consumption (6.10–11.40 GJ ton<sub>CO<sub>2</sub></sub><sup>-1</sup>, see the entries for temperature swing in Table 3). Another alternative is solar-driven TSA, which can directly utilize renewable solar energy. However, its operational stability is highly dependent on weather conditions, posing a major challenge for maintaining continuous and stable performance at scale.

The precipitation-phase separation method allows the transformation of captured CO<sub>2</sub> in the solution into a solid form, thereby avoiding the treatment of large volumes of absorbent solutions. This makes the method particularly suitable for large-scale DAC systems. The separated CO<sub>2</sub> capture and release units can also provide greater flexibility and controllability, enabling the accumulation of sufficient quantities of carbonate solid from multiple capture cycles before heating treatment. The precipitation-phase separation method through the calcium loop has a relatively high energy consumption due to high-temperature calcination (see the entries for precipitation-phase separation in Table 3). In comparison, the CO<sub>2</sub> release through the guanidine loop opens up new avenues for reducing the heating temperature required (60–150 °C), significantly reducing the energy consumption (2.27 GJ ton<sub>CO<sub>2</sub></sub><sup>-1</sup>) and also making solar heating a viable option. However, while specific guanidine compounds have shown good performance in previous studies, their potential for scaled-up synthesis remains uncertain.



**Table 3** Comparison of three typical methods for CO<sub>2</sub> release/capture agent regeneration

Methods	Specific types	Capture agent	Working temperature (°C)	Energy consumption (GJ ton <sub>CO<sub>2</sub></sub> <sup>-1</sup> )	Ref.	
Temperature swing	TSA/TVSA	Amine loaded adsorbents	80–100	5.40–7.20	109	
		PEI loaded silica	80–120	7.20	114	
	e-TSA/TVSA	Hydroxide-functionalized porous carbon	90–100	6.50–11.40	87	
		EDA-Y zeolite/cellulose acetate fiber sorbent	110	6.10	115	
Solar-driven TSA		Lewatit VP OC 1065 (Lanxess) <sup>a</sup>	110	No data available	116	
		PEI-loaded SBA-15	80–100	No data available	117	
		KOH/K <sub>2</sub> CO <sub>3</sub>	900	5.25–8.81	39	
Precipitation-phase separation	Potassium-calcium loop	K-Gly/K-Sar	80–120	No data available	122	
	Amino acid salt-guanidine loop	BTIG	60–150	2.27	124	
Electrolysis	pH swing with BPMED	K <sub>2</sub> CO <sub>3</sub> /KHCO <sub>3</sub> <sup>b</sup>	RT <sup>c</sup>	2.27–4.54 <sup>d</sup>	127	
		K <sub>2</sub> CO <sub>3</sub> /KHCO <sub>3</sub> <sup>b</sup>	RT	8.80 <sup>d</sup>	128	
	pH swing with three-chamber reactors	Na <sub>2</sub> CO <sub>3</sub> /NaHCO <sub>3</sub> <sup>b</sup>	RT	8.49	130	
		NaOH	RT	12.19 ± 0.75 <sup>e</sup>	131	
		NaOH	RT	2.68 <sup>e</sup>	132	
		Na-Gly/Gly/NaCl	RT	5.20–8.08	52	
		LiOH	RT	6.40 <sup>f</sup>	133	
		LiOH	RT	3.80 <sup>f</sup>	134	
	Redox mediation by quinones	1,4-Napthoquinone and 1-ethyl-3-methylimidazolium tricyanomethanide	Hydroquinone/quinone	RT	1.27	135
			Anthraquinone	RT	2.57	139
			Poly(vinylanthraquinone)	RT	0.48	140
			2,3,5,6-Tetrachloro-p-benzoquinone	RT	1.09–3.18	141
		Redox mediation by phenazines	Na <sub>2</sub> CO <sub>3</sub> /7,8-dihydroxyphenazine-2-sulfonic acid	RT	0.49	137
			1,8-ESP	RT	0.82–1.25	138
Cyclic poly(phenazine sulfide)			RT	2.36	142	

<sup>a</sup> A commercial amine-functionalized weakly basic anion exchange resin. <sup>b</sup> Using a simulated CO<sub>2</sub>-captured solution (*e.g.*, NaHCO<sub>3</sub>/Na<sub>2</sub>CO<sub>3</sub>). <sup>c</sup> Room temperature. <sup>d</sup> The differences in electrolytic cell configuration, <sup>e</sup> The middle acidification chamber thickness, and <sup>f</sup> The reaction type lead to the variations in energy consumption.

The traditional electrochemical pH swing usually results in the generation of byproducts such as O<sub>2</sub>, which necessitates an additional separation step to obtain pure CO<sub>2</sub>. BPMED and the three-chamber electrolytic system offer alternative approaches for releasing CO<sub>2</sub> with high purity and moderate to high energy consumption (2.68–12.19 GJ ton<sub>CO<sub>2</sub></sub><sup>-1</sup>, see the entries for electrolysis in Table 3). The thickness of the central acidification chamber dictates the electrode spacing, while the electrolyte ion concentration governs the system's conductivity. These two parameters are key factors leading to the variations in energy consumption reported across different studies. Replacing the central acidification chamber with the solid electrolyte layer can increase the current density significantly without increasing the cell voltage, representing a promising direction for electrolytic CO<sub>2</sub> release.

Using redox mediators for electrolysis exhibits the lowest energy consumption across all systems, making it a highly promising approach (0.48–3.18 GJ ton<sub>CO<sub>2</sub></sub><sup>-1</sup>, see the entries for electrolysis–redox mediation in Table 3). The energy consumption of the redox-mediated approach has reached close to the theoretical minimum for CO<sub>2</sub> release (0.43 GJ ton<sub>CO<sub>2</sub></sub><sup>-1</sup>).<sup>143</sup> However, the complex synthesis process for the

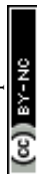
organic mediators remains the primary limitation for its scale-up.

## 5 Summary and outlook

### 5.1 Summary

DAC has shown great potential in promoting carbon recycling and achieving carbon neutrality, garnering increasing attention in recent years. In this work, we discuss three key factors influencing its efficiency and development, which are summarized in Table 4.

(1) The air contactor is an indispensable component of the DAC system which guarantees air flow and CO<sub>2</sub> supply. The adsorption-based air contactor allows for CO<sub>2</sub> capture and release in a compact design. However, this integrated functionality requires a more complex piping and process design for the DAC system. Thus, the system can achieve large CO<sub>2</sub> capacities primarily through modular scale-out (*i.e.*, running many modular units in parallel). In contrast, the absorption-based air contactor operates independently from the CO<sub>2</sub> release unit and shows high compatibility with a wide range of CO<sub>2</sub> release technologies. Accordingly, it



**Table 4** Summary of three key factors influencing DAC efficiency and development

Key factors	Types	Key benefits	Key challenges
Air contactor	Adsorption-based	Compact and small-size devices CO <sub>2</sub> capture and release on one site Modular scale-out	Operational complexity from cyclic switching
	Absorption-based	Modular scale-out & centralized scale-up Compatible with various CO <sub>2</sub> release technologies	Water evaporation Corrosion risk of facility
Capture agent	Solid adsorbents	Capable of modularization Wide adaptability to environmental temperature	Competing with H <sub>2</sub> O adsorption Poor stability
	Liquid absorbents	Relatively low cost and easy to scale up High CO <sub>2</sub> selectivity	Corrosion to the air contactor/pipeline Water evaporation
CO <sub>2</sub> release/capture agent regeneration	Temperature swing	Waste heat can be utilized	Low thermal energy efficiency Degradation of capture agent
	Precipitation-phase separation	Treating a small amount of carbonate solid	Involving multiple steps High operating temperature
	Electrolysis	Compact equipment Controllable reaction rate Coupling with renewable energy	Relying on precious electrocatalysts, mediators, and ion-exchange membranes

offers greater flexibility for scaling, *via* both modular scale-out and centralized scale-up (*i.e.*, expanding a single plant). For example, Carbon Engineering is leading the front-end engineering and design for a DAC facility with a CO<sub>2</sub> capture capacity of 500 000 tons per year.<sup>144</sup> However, it is important to note that the absorption-based air contactor usually utilizes a strongly alkaline absorbent solution, which requires careful consideration of water evaporation and corrosion risk to the contactor and piping framework, as well as the potential for precipitate formation.

(2) The capture agent determines the CO<sub>2</sub> capture capacity. There are two typical types of capture agents, including solid adsorbents and liquid absorbents, which are used in adsorption- and absorption-based air contactors, respectively. Amine-functionalized porous materials are the most important solid adsorbents, and their performance for DAC is influenced by a number of factors such as the type of amine, amine loading, support material, and environmental temperature and humidity. However, the high production cost and limited lifespan of adsorbent materials hinder their widespread use in DAC. Alkaline hydroxide and amine solutions are both efficient and cost-effective absorbents for CO<sub>2</sub> capture. Alkaline hydroxides have found practical applications in the absorption-based DAC facility. However, the use of amine compounds for DAC is limited, because of the volatility and degradation issues. Thus, the stability of organic amines under atmospheric conditions requires further improvement, for example, by developing new amine compounds with low volatility and resistant to oxidative degradation. AAS, as easily accessible and environmentally friendly absorbents with rapid reaction kinetics for CO<sub>2</sub> capture, hold significant potential for DAC.

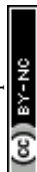
(3) The process of CO<sub>2</sub> release and capture agent regeneration contributes to most energy consumption in DAC. There are several types of effective methods to release the captured CO<sub>2</sub>. Temperature swing, which usually operates at elevated temperatures of 80–120 °C and under vacuum conditions, is primarily used to release CO<sub>2</sub> from adsorption-based DAC systems. This technology enables the utilization of waste heat,

offering potential sustainability benefits.<sup>47</sup> However, the degradation of capture agents at high regeneration temperature leads to decreased CO<sub>2</sub> capture performance. The precipitation-phase separation method separates CO<sub>2</sub> capture and release steps. This method converts captured CO<sub>2</sub> in liquid solution into a solid precipitate, significantly reducing the volume/mass of substance for processing. However, the high calcination temperature required for CO<sub>2</sub> release from CaCO<sub>3</sub> remains the primary limitation of this method. Additionally, the multiple processing steps lead to operational complexity. The electrochemical approach for CO<sub>2</sub> release provides distinct advantages including (i) mild reaction conditions, (ii) electrochemically tunable reaction rates, (iii) compatibility with renewable electricity, and (iv) low energy consumption when mediated by redox couples. To date, these developments have been primarily limited to lab-scale prototypes, lacking industrial-scale implementation cases. Scaling it up remains challenging, as it depends on stable and costly electrocatalysts and ion exchange membranes to achieve high efficiency and durability.

Overall, existing research has largely focused on the development and optimization of individual DAC components, such as the air contactor, capture agent, or regeneration unit, under idealized conditions. In practice, however, these three components form a tightly connected system, where the choice of air contactors determines the choice of capture agents, which in turn dictates the feasible regeneration approaches. The lack of a system-level study on the entire DAC process poses a major obstacle for achieving efficient and large-scale DAC implementation.

## 5.2 Outlook

Despite the challenges and issues mentioned above, the DAC technology is highly promising for its great prospect of removing CO<sub>2</sub> from the atmosphere directly. To overcome the issues and promote the development and practical applications of DAC technology, research efforts may be directed to the following aspects.



**(1) Integrating DAC with existing facilities.** Due to the relatively low concentration of CO<sub>2</sub> in air, the massive investment in construction and operation of large-scale dedicated air contactors is a significant challenge. Therefore, integrating DAC with existing industrial or urban facilities is a very promising solution, which avoids the large capital and operational investment. Previous work has showcased that buildings, trains, and industrial cooling towers can all be retrofitted into DAC facilities.<sup>50,51</sup> In principle, any facility with air flow can be retrofitted for DAC. In this way, DAC is expected to become as routine as wastewater treatment in everyday life with reduced cost. However, effective integration must move beyond simple retrofitting. Its success depends on the careful optimization of the fluid dynamics between the airflow of the original facilities and the retrofitted DAC unit.

**(2) Exploring new capture agents for DAC.** In adsorption-based DAC, the primary challenge is the competitive adsorption of atmospheric moisture, which reduces CO<sub>2</sub> capture capacity. However, this issue can be reframed as an opportunity by treating the adsorbed water as a valuable co-product. Notably, porous solid adsorbents have shown great promise not only for DAC but also for atmospheric water harvesting (AWH), which addresses global freshwater scarcity.<sup>145–147</sup> Given that AWH and DAC can be operated under similar conditions, future adsorbent design should leverage co-adsorption of CO<sub>2</sub> and water molecules to enable simultaneous CO<sub>2</sub> capture and water collection.<sup>148</sup> For absorption-based DAC, common organic amines are not ideal for use as DAC capture agents owing to their susceptibility to oxidative degradation. Inorganic alkaline solutions (*e.g.*, KOH) are stable and effective CO<sub>2</sub> absorbents. However, their capture stoichiometry requires two OH<sup>-</sup> ions for one CO<sub>2</sub> molecule, leading to carbonate formation, which shows poor kinetics for capturing CO<sub>2</sub> further from the atmosphere.<sup>149</sup> AAS stands out as a superior option, exhibiting both high stability and efficient CO<sub>2</sub> capture performance. While two AAS molecules also capture at most one CO<sub>2</sub> molecule, utilizing AAS as a promoter for hydroxide absorbents in DAC systems could enhance CO<sub>2</sub> capture capacity.<sup>150,151</sup>

Notably, machine learning, leveraging datasets of atomic and molecular properties, is becoming a powerful tool for CO<sub>2</sub> capture studies.<sup>152,153</sup> While numerous efficient adsorbents have already been identified, future efforts can focus on constructing comprehensive databases and employing text mining to extract insights from previous reports. Building on this, developing robust machine learning algorithms will be crucial for guiding the rational design of new capture agents. The ultimate goal is to discover new capture agents that deliver both high CO<sub>2</sub> uptake and long-term stability across a range of contactor configurations and operating environments.

**(3) Developing DAC technologies driven by renewable electricity.** The adoption of renewable energy (*e.g.*, wind or solar power) to drive DAC systems represents a viable strategy for improving their societal and economic feasibility.<sup>14</sup> Several reports have demonstrated that both solid adsorbents

and liquid absorbents can be coupled with renewable energy sources to achieve sustainable CO<sub>2</sub> release and capture agent regeneration.<sup>116,117,122,154</sup> With the continuing decline in the cost of green electricity, the integrated “renewable energy + DAC” approach is expected to become a key negative emission technology.<sup>155,156</sup> Due to the intermittency of wind and solar energy, future electrically driven DAC systems must tolerate frequent start-stop cycles without compromising the stability or kinetics of the capture medium. Integrating electrochemical regeneration with energy storage buffers is essential to maintain consistent performance under fluctuating in renewable power supply.

**(4) On-site conversion of captured CO<sub>2</sub>.** Currently, the captured CO<sub>2</sub> is usually released somehow and then converted or stored, which involves multiple steps and high energy consumption. A more promising way is to directly convert the captured CO<sub>2</sub> without release, which can avoid the energy-intensive steps such as CO<sub>2</sub> release, purification, and compression.<sup>157,158</sup> The production of high-value chemicals such as CO, formic acid, and ethylene from captured CO<sub>2</sub> *via* direct thermal,<sup>159,160</sup> photo,<sup>161,162</sup> or electrocatalytic conversion<sup>163–165</sup> can compensate DAC operating costs, enhancing its commercial feasibility. This integrated strategy effectively closes the carbon cycle, transforming the DAC facility into a versatile platform for sustainable synthesis of chemicals.

Looking forward, future research should shift from studying individual components toward a system-level paradigm. The air contactor, capture agent, and regeneration method are not independent but tightly connected components, whose collective performance determines overall DAC efficiency. For instance, a highly efficient sorbent cannot realize its potential unless paired with an air contactor and a regeneration process enabling effective mass and energy transfer. Research efforts on linking the three components as a single system are expected to unlock synergistic gains in enhancing DAC efficiency for practical applications.

## Author contributions

All authors contributed to the manuscript's writing, reviewing, and revising. X.-M. Hu coordinated and supervised the overall project.

## Conflicts of interest

The authors declare no conflict of interest.

## Data availability

No primary research results, software or code have been included and no new data were generated or analysed as part of this review.

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