

Review: Novel physical and chemical CO₂ capture methods

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ABSTRACT

Methods for CO₂ capture that are essential for maintaining a clean atmosphere rely on amine group technology because of their unique physical and chemical characteristics and diverse absorption behavior. This study examined the latest advancements in a novel class of materials called liquid-like nanoparticle organic hybrid materials. These materials exhibit unique properties, and this research encompasses experimental findings and molecular simulations. CO₂ is absorbed by physical and chemical ionic liquids using metal-organic frameworks for CO₂ adsorption and membrane vacuum regeneration technology. The findings presented here illustrate the benefits and drawbacks of these approaches.

Introduction

The greenhouse effect resulting from elevated CO₂ concentrations in the atmosphere prompted the investigation of CO₂ gas collection methods.^{1, 2} The Industrial Revolution ushered in an era of unprecedented CO₂ emissions, permanently altering the Earth's atmosphere. Prior to industrialization, atmospheric CO₂ concentration remained stable at approximately 280 parts per million (ppm). However, this delicate balance has been disrupted, with CO₂ levels now reaching around 390 ppm.

This significant increase highlights the profound impact human activities have had on our planet's climate and underscores the urgency of addressing climate change.³ Global CO₂ emissions in 2008 were estimated to be around 29 gigatons (Gt), an increase of roughly 40% over emissions of 20.9 Gt in 1990.⁴ Currently, the transport sector accounts for 23% of all energy-related CO₂ emissions, followed by the industry sector (20%), the buildings sector (10%), and other sectors (41%).^{4,5} The significant increase in projected global CO₂ emissions, which are expected to reach 40.2 Gt by 2030, has drawn attention to fossil fuels on a global scale.⁶ Consequently, interest in creating technology for the

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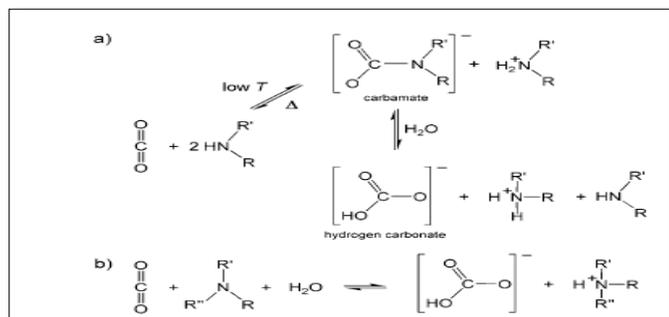
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effective collection and sequestration of vast amounts of CO₂ is beginning to increase.⁷ The existing literature presents a diverse landscape of innovative CO₂ capture technologies, encompassing membrane separation, molecular sieves, carbamate, amine physical absorption, amine dry scrubbing, and mineral carbonation as key methodologies. Although these strategies offer promising solutions for CO₂ capture and separation, chemical absorption using an aqueous solution of monoethanolamine (MEA) has established itself as the dominant technology employed on an industrial scale.^{8,9} This technique depends on the functional groups, including amines, for CO₂ capture. Several novel methods used the functional amine group for CO₂ capture, particularly highlighting nanoparticle organic hybrid materials (NOHMs). NOHMs exhibit high selectivity for CO₂. A new class of metal–organic frameworks (MOFs) presents alternative methods for CO₂ removal via metal-containing nodes bonded to organic bridging ligands based on amine group absorption and adsorption.¹⁰ A less costly and energy-intensive method of CO₂ separation than chemical absorption is CO₂ capture by physical absorption. Chemical absorption is essential, and a recent development in this area is the use of ionic liquids (ILs). These liquids have demonstrated considerable potential in absorbing CO₂ and are also environmentally friendly. CO₂ is reversibly sequestered as a carbamate salt by the IL that contains a cation with an attached amine group. First, IL consists of a primary amine in an imidazolium ion [NH₂p-bim][BF₄], indicating the proposed reaction mechanism between IL and CO₂^{11–13}.

Extensive research has focused on the zwitterion mechanism by which carbon dioxide reacts with the amine in the main alkanol¹⁴ amine MEA to produce carbamates (Scheme 1a).



Scheme 1. General reaction schemes for the chemical absorption of CO₂ by primary or secondary and tertiary amine-containing solvents.¹⁵

Scheme 1 illustrates the general chemical reaction mechanisms for CO₂ absorption by primary/secondary and tertiary amines, which are two classes of widely studied solvents for CO₂ capture. By examining these pathways, we gain valuable insights into the key steps involved in this process, ultimately facilitating the development and optimization of amine-based CO₂ capture technologies.¹⁶

The regeneration of the chemical solvent requires elevated temperatures (100 °C–140 °C) but relatively low pressures, typically near atmospheric pressure. This energy-intensive process requires the exploration of alternative materials and methods for efficient CO₂ capture¹⁷.

Innovative organic and inorganic materials, including amine-functionalized mesoporous sorbents and novel liquid solvents, such as ILs and aminoalkyl silanes, are being developed in this field and signify promising advances¹⁸. This research highlights the potential of emerging technologies like NOHMs, MOFs for gas adsorption, and CO₂ capture systems using membrane vacuum regeneration (MVR) in ILs. Further research exploring these methodologies and their synergistic combinations is crucial for developing next-generation solutions for efficient and sustainable CO₂ capture¹⁹. Exciting progress in this area involves the creation of cutting-edge organic and inorganic materials, such as amine-functionalized mesoporous sorbents, as well as new liquid solvents such as ILs and aminoalkyl silanes.

1.1 NOHMs

NOHMs are particularly intriguing because of their ability to absorb gases selectively for favorable CO₂ capture capacity. Consequently, one of the promising solutions for CO₂ collection has been suggested: NOHMs. Studies revealed that the presence of amine groups significantly amplifies the effectiveness of CO₂ capture compared with the presence of ether groups. To investigate this phenomenon, we synthesized two NOHMs: NOHM-I-PEI (incorporating polyethyleneimine) and NOHM-I-tPE (synthesized using polyether amines containing tertiary amine

groups). Both samples were evaluated for their CO₂ capture capabilities at 25 °C and a CO₂ partial pressure of 3.2 atm (Fig. 1). These conditions purposefully improved the NOHMs' ability to collect CO₂.²⁰ To measure the increase in CO₂ absorption resulting from chemical bonding, scientists focused on studying the impacts of two distinct functional groups: ether and amine groups.²⁰

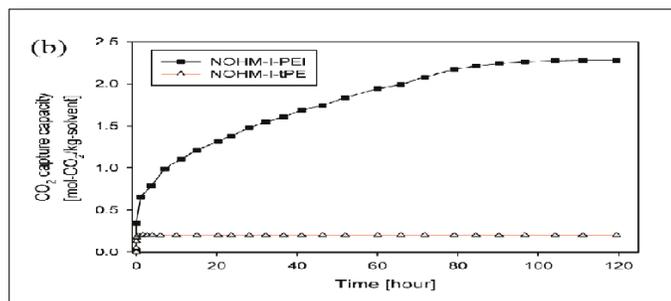


Fig. 1. Variety of amine groups under a pressure of 3.4 atm and a temperature of 30 °C

NOHM samples were synthesized and analyzed to elucidate the interaction between primary amines and the surface functional groups of nanoparticle cores (ionic bonding with sulfonate groups in NOHM-I and covalent bonding with glycidyl ether groups in NOHM-C). At 1530 (symmetric NH³⁺) and 1630 cm⁻¹ (asymmetric NH³⁺), characteristic peaks associated with protonated amines were observed in the infrared spectra of NOHM-I. These findings confirmed the formation of an ionic bond between polyether amine and the sulfonate groups on the NOHM-I core. The synthesis of NOHMs revealed a unique phenomenon: complete utilization of the functional groups present in polyether amines during the grafting process. By utilizing the bonds established between the nanoparticle core and the grafted polymer chains as primary anchors, rather than relying on functional groups, the resulting NOHMs exhibited exceptional stability. Analysis of the NOHM samples confirmed the absence of any unreacted free polymers, indicating a highly efficient synthesis process. The synthesis of polyether-silane in NOHM-C-HPE was verified by the hydroxyl groups present at 3400–3600 cm⁻¹ and the absence of a band at 3056 cm⁻¹ (ν_{C-H} epoxy) (Fig. 2(b)). Comparable FT-IR spectra patterns were discovered for the polyether-silane that corresponded with NOHM-C-MPE.²¹

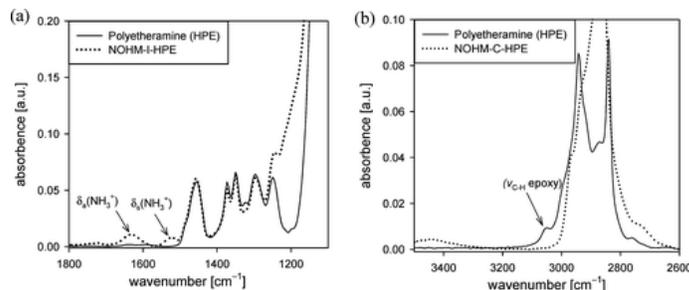


Fig 2. Synthesized NOHMs' ATR FT-IR spectra: (a) NOHM-I-HPE and (b) NOHM-C-HPE.²¹

1.2 Gas adsorption in MOFs for carbon dioxide capture

The discovery of MOFs, a new class of crystalline porous materials, has generated considerable interest in chemistry and materials science. MOFs possess framework structures that are sufficiently well-defined geometrically and crystallographically to permit the removal of guest species. Many MOFs have undergone experimental evaluation for CO₂ adsorption and separation. Pore size, pore volume, and surface influence CO₂ absorption. Numerous MOFs have large pore volumes and surface areas.²² The crystallinity of MOFs enables detailed structural characterization using diffraction methods, supporting the derivation of structure–function connections and rational design of these materials.

MOFs can be conceptually synthesized and constructed based on how building components combine to form a net (as illustrated in Fig. 3).²³ The remarkable flexibility of MOFs stems from their ability to precisely control their structures and properties via careful selection of building blocks. This is achieved through pre-design during synthesis and post-synthetic modifications that have proven effective in tailoring the pore characteristics of MOFs.

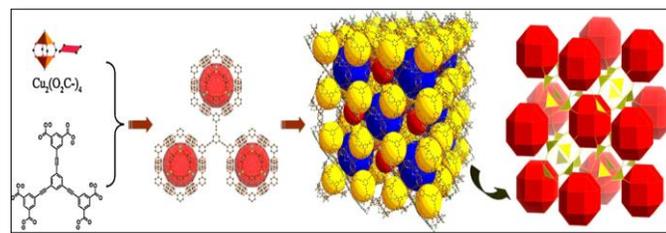


Fig. 3 Metal-containing node and bridging organic ligand are the first steps in the construction of a proposed MOF. Next, a supramolecular building block is added. Lastly, a 3D framework with pores is created.

The concluding figure illustrates the geometric structure of the framework, with ligands acting as connecting nodes, represented in yellow and red for 3 and 24 nodes, respectively. Notably, some ultrahigh pore MOFs, such as $Zn_4O(bdc)_3$ (MOF-5 or IRMOF-1), $Zn_4O(btb)_2$ (MOF-177), and $Zn_4O(bte)_{14/9}(bpdc)_6/9$ (MOF-210), exhibit sigmoidal isotherms for CO_2 adsorption at near-room temperature and high pressure, indicating potentially cooperative adsorption behavior.²⁴

MOFs are the optimal solution for gas storage and separation, including CO_2 capture, because of their unmatched adjustable pore surface properties, vast surface areas, and highly tunable pore diameters. Small pores, particularly those facilitated by large cations in the MOF structure, offer excellent CO_2 condensation capabilities at realistic temperatures. Furthermore, cation exchange in flexible MOFs, such as $Ni(bpe)_2(N(CN)_2)_2$: cations (where bpe = 1,2-bis(4-pyridyl)ethane), allows for the precise control of CO_2 adsorption. Low-pressure CO_2 adsorption within an amine-functionalized MOF, $Zn_2(Atz)(ox)$ (Atz = aminotriazolato, ox = oxalate), was investigated using X-ray crystallography, revealing the unique CO_2 interactions within the framework (Fig. 4).²⁵

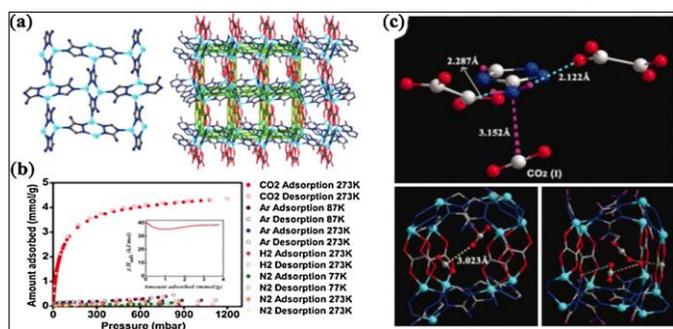


Fig. 4 (a) Structure of $Zn_2(Atz)(ox)$ and its gas adsorption properties; (b) binding of CO_2 inside the pores of $Zn_2(Atz)(ox)$, which was ascertained immediately by X-ray structural refinement at 173 K. Adapted with permission from Reference.²⁵

1.3 CO_2 capture system via MVR in IL

MVR technology utilizes amine-based solutions for CO_2 desorption primarily due to their advantageous properties: low viscosity, low cost, and high CO_2 absorption capacity, even at low CO_2 partial pressures. Two approaches can be employed to further develop MVR technology for CO_2 capture using ILs:

(i) Modeling and design: This involves simulations and modeling studies to investigate the influence of various operating factors on low-temperature CO_2 desorption using different ILs. (ii) Investigating IL-based membrane contactors: This focuses on optimizing the compatibility between ILs and membranes in membrane contactors for efficient CO_2 separation.²⁶

Optimizing the solvent regeneration performance in IL-based MVR systems necessitates a thorough investigation of the IL's nature (physical or chemical absorption), thermodynamic properties, and kinetic characteristics.

1-Ethyl-3-methylimidazolium methyl sulfate ([emim][MS]) and 1-ethyl-3-methylimidazolium acetate ([emim][Ac]) are viable options for chemical and physical ILs, respectively. This study examined the influence of temperature and vacuum grade on the CO_2 desorption flux and overall desorption performance.²⁷ The IL [emim][MS], a physical CO_2 absorbent with favorable solvent properties due to its low volatility and very low viscosity, emerged as a promising candidate for applications at high CO_2 partial pressures. However, considering isotherm data, the IL [emim][Ac] displayed superior overall CO_2 absorption capacity, particularly at low CO_2 partial pressures. Further investigation is needed to elucidate the interplay between the specific type of interaction between CO_2 and the IL (chemical or physical) and contactor properties, including membrane geometry and fluid dynamics, to optimize CO_2 capture efficiency.²⁸ The effectiveness of IL characteristics on CO_2 desorption performance should be determined to examine the regeneration conditions.

1.4 Physical processes of CO_2 capture by ILs

The previous part of the study examined the effectiveness of the CO_2 release process and the flow of released CO_2 by utilizing the ILs [emim][Ac] as a chemical absorbent and [emim][MS] as a physical absorbent within a continuous, stable absorption–desorption system for CO_2 capture under different conditions. Equation (1) was employed to assess the impact of the MVR-based CO_2 desorption stage on the overall CO_2 capture system performance. Fig. 5 displays the membrane contactor specifications and operating

conditions of the steady-state absorption–desorption system in two different scenarios. This allows for the assessment of how solvent temperature (289 K and 313 K) and CO₂ desorption vacuum pressure (0.2 and 0.04 bar) collectively influence the overall CO₂ capture efficiency for each IL investigated in this study.²⁹

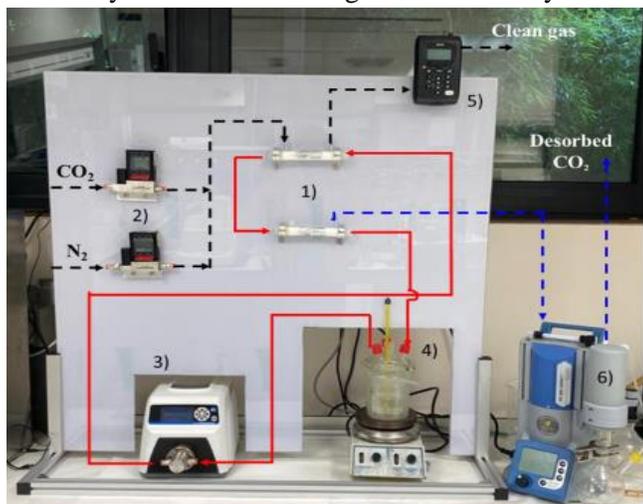


Fig. 5 Absorption–desorption process using MVR technology²⁹

$$Gv = \frac{Fv}{vmA} \dots (1)$$

The terms can be identified as follows:

- **Gv**: Volumetric gas flow rate (often referred to as the volumetric gas velocity or gas flux), which describes the volume of gas passing through a unit area per unit time.
- **Fv**: Molar flow rate of gas, representing the amount of gas passing per unit time (typically in moles per second or minute).
- **vm**: Molar volume of gas, which is the volume occupied by one mole of gas under specific conditions of temperature and pressure (commonly in cubic meters per mole, m³/mol).
- **A**: Cross-sectional area through which gas flows (usually measured in square meters, m²), representing the area of the surface where gas desorption occurs.

This equation was used to analyze the volumetric behavior of the gas (CO₂) in the desorption stage of the CO₂ capture system, particularly in relation to MVR.

The total efficiency of CO₂ capture for the chemical IL [emim][Ac] increased from 29% to 61% due to the elevated temperatures of the solvent and low vacuum pressures for CO₂ desorption. The chemisorption effect increased the CO₂ absorption amplitude, whereas the viscosity of [emim][Ac]

decreased as the temperature increased. Thus, temperature contributed more to CO₂ capture than the CO₂ desorption vacuum pressure (P_v). The use of a high vacuum during the MVR process increased the overall CO₂ capture efficiency from 12% to 21%. The IL [emim][MS] is shown in Fig. 6.³⁰

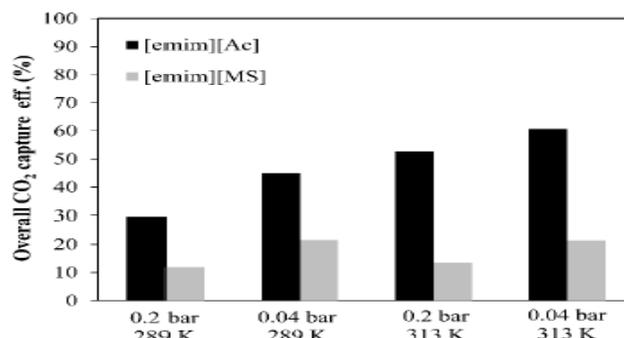
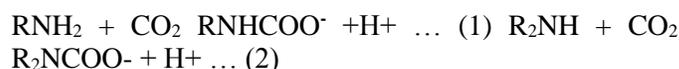


Fig. 6 Overall CO₂ capture performance by using two ILs at different vacuum pressures and temperature³⁰

1.5 Chemical processes of CO₂ capture by ILs

The study aimed to investigate the innovation of new properties of protic ionic liquids (PILs). This work studied the potential of a cost-effective PIL with multiple amine functional groups for CO₂ capture. PILs are formed through proton transfer between a Brønsted acid and a Brønsted base. The specific PIL employed in this study contained two primary and two secondary amine groups. The elevated basicity of secondary amines resulted in the preferential proton transfer from the acid to their nitrogen atoms. Converting existing amine and alkanolamine solvents into their ionic forms through protonation is an effective strategy for minimizing solvent evaporation at high temperatures.

In an amine-based CO₂ capture plant, the primary and secondary amines efficiently capture CO₂ through reaction pathways and then undergo conversion into carbamate salt.



The results of the reaction were CO₂ with tertiary amines in the formation of a bicarbonate salt.



In the reaction mixture, nine main species were present: precursor amines (RNH₂/ R₂NH/R₃N), protonated amines (am H⁺), carbamates (RNHCOO⁻, R₂NCOO⁻), carbonate (CO₃²⁻), bicarbonate (R₃NHCO₃)

⁻), hydronium ion (H₃O⁺), hydroxyl ion (OH⁻), CO₂, and water.³¹

The zwitterion mechanism involves the formation of a zwitterion as an intermediate during the interaction between carbon dioxide and an amine.



When a base B, such as amine, water or an OH group, deprotonates this zwitterion, producing carbamate:



Carbamate has poor stability, so it can easily withstand hydrolysis to create bicarbonates and release free amine molecules. CO₂ and the regenerated amine reconstitute. Initially, the carbamate species are dominant, and 13C NMR can subsequently verify comparable amounts of bicarbonate ions. The total rate of reaction between CO₂ and an amine aqueous solution can be represented at a steady state.³²

$$\frac{k_2(\text{CO}_2)(\text{R}_2\text{NH})}{1 + \frac{k_{-1}}{k_B(\text{B})}} \dots\dots\dots (6)$$

Here,

k₂ = second-order reaction rate constant, L·mol⁻¹·s⁻¹

k₋₁ = backward rate constant, s⁻¹ and

k_B represents the kinetic constant that quantifies the rate at which the zwitterion loses a proton while reacting with any base, measured in s⁻¹.

An example is shown in Fig. 7.

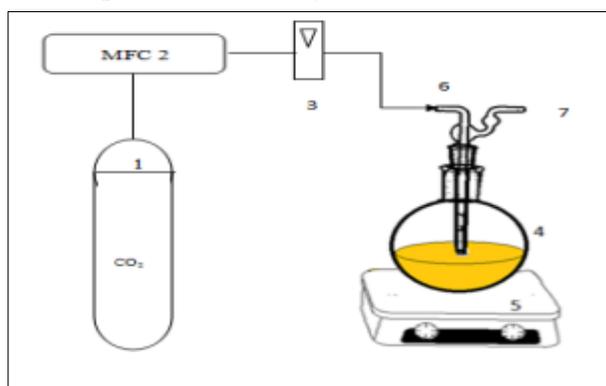


Fig. 7. Setup for CO₂ absorption. 1. CO₂ cylinder, 2. Digital mass flow controller, 3. CO₂ rotameter 4. Reaction vessel, 5. Magnetic stirrer, 6. Gas inlet port, 7. Gas outlet port.

1.6 Functionalized ILs for CO₂ capture

1.6.1 Cation-functionalized ILs for CO₂ capture

1-Propylamide-3-butyl imidazolium tetrafluoroborate ([apbim][BF₄]) was absorbed in IL by

a carbamate mechanism (1 CO₂: 2 amino). When amino-grafted IL cations were compared with a standard aqueous solution of alkanol amine (30 weight percent MEA) for CO₂ capture, the amino grafted on ILs demonstrated superior capture capacity and high thermostability. Certain amino-grafted cation-functionalized ILs may successfully extract CO₂. The mechanisms underlying amino-CO₂ reactions in ILs closely resemble those observed in aqueous alkanolamine solutions. However, under anhydrous conditions, tertiary amines are generally considered non-reactive with CO₂.³³

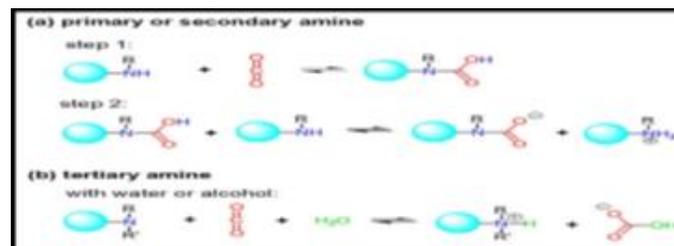


Fig. 8. General mechanisms of amino-CO₂ reactions for (a) primary or secondary amine and (b) tertiary amine.³³

1.6.2 Anion-functionalized ILs for CO₂ capture

An effective method for CO₂ collection involves the grafting of different kinds of functional groups onto anions. Anion-functionalized ILs are classified into two groups based on their CO₂ interaction: mechanisms at one site and mechanisms at several sites. Functionalized ILs contain single-site anions, including amino, carboxylate, alkoxide, phenolate, and azolate. Fig. 9a shows the typical mechanisms involved in the non-amino anion-CO₂ reaction.³⁴

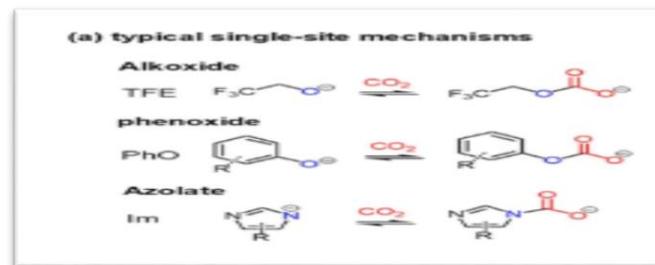


Fig. 9. Typical (a) mechanisms of non-amino anion-CO₂ reactions.³⁴

Advantages:

1-By NOHMs

This work showed that NOHMs possess robust better thermal stabilities, which allows them to operate

at a wide range of temperatures for CO₂ capture and solvent regeneration. Moreover, NOHMs exhibit potential for recycling and demonstrate high selectivity for CO₂ compared with N₂, O₂, and N₂O. Given these unique characteristics, NOHMs have demonstrated considerable potential for CO₂ capture.²¹

2-By MOFs

A new class of crystalline porous materials called MOFs offers several benefits, such as high porosity, easily adjustable pore properties, and ease of design and synthesis. Therefore, MOFs are remarkable candidates for materials used in CO₂ capture and separation.³⁵

3-By ILs

ILs are simple and inexpensive to synthesize. This study demonstrated that adjusting the fundamental properties of ILs through precise control of acid and base concentrations during synthesis can significantly enhance their CO₂ absorption capacity. In this study, the chemical IL [emim][Ac] achieved a maximum CO₂ desorption efficiency of 92%, whereas the physical IL [emim][MS] reached 83%. Notably, these optimal achievements were conducted under identical operating conditions: a liquid flow rate of 60 mL·min⁻¹, a temperature of 313 K, and a vacuum pressure of 0.04 bar. Within the continuous absorption–desorption CO₂ capture system investigated, the chemical IL exhibited superior process performance compared with the physical IL. However, physical ILs may serve as energy-efficient CO₂ capture absorbents if more advanced physical ILs with enhanced CO₂ absorption capacities are developed.³⁰

Disadvantage:

1-By NOHMs

NOHMs containing functional groups with limited CO₂ targeting capabilities demonstrated lower measured CO₂ capture capabilities than 15–30 weight percent MEA solvents. To maximize CO₂ capture efficiency, researchers are currently synthesizing the next generation of NOHMs, and kinetic mechanistic investigations are underway. In addition to providing critical elements for the use of NOHMs in CO₂ collection, research into this area will shed light on the

chemical and physical properties of these innovative nano-scale materials.²¹

2-By MOFs

Given that MOFs are promising candidates for CO₂ capture, numerous challenges must be investigated. Failure to resolve these issues may hinder the development of practical applications. Only a fraction of the numerous MOFs synthesized and structurally characterized in the past decade have undergone testing for CO₂ adsorption, storage, and associated gas separation.

Measuring all of these new materials is impractical. Therefore, scanning techniques should be developed as an alternative. Besides identifying valuable materials, they can also be used to assess structure–function correlations, thereby guiding the development and improvement of novel MOF materials. Molecular simulation plays a major role in searching MOFs for CO₂ capture.³⁵

3-By ILs

To enhance the CO₂ capture process, we examined IL-based methods. Further research on continuous absorption–desorption systems employing vacuum regeneration and ILs is required to enhance the practical application and economic viability of non-dispersive absorption technology.³⁰

Conclusions

Although amine-based technology continues to dominate CO₂ capture because of its unique chemical and physical properties, concerns about its limitations persist. This study explored the promising potential of liquid-like NOHMs as a novel class of capture materials with remarkable properties. Employing experimental and simulation data, researchers investigated several capture methods involving physical and chemical ILs, MOFs for adsorption, and MVR technology for regeneration. By meticulously analyzing the benefits and drawbacks of each approach, this study elucidated the trade-offs inherent in various CO₂ capture strategies. This work suggests that amine-based capture remains significant, but a future focused on innovative materials such as NOHMs and a diverse array of capture

techniques may be essential for achieving a clean environment.

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مراجعة: أساليب جديدة لالتقاط ثاني أكسيد الكربون في المجال الفيزيائي والكيميائي

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الخلاصة:

الطرق الضرورية لالتقاط ثاني أكسيد الكربون التي تلعب دوراً مهماً في الحفاظ على الغلاف الجوي نظيفاً تعتمد على تقنية مجموعات الأمين بسبب خصائصها الفيزيائية والكيميائية الفريدة وسلوك الامتصاص المتنوع لها. تتناول هذه الدراسة أحدث التطورات في فئة جديدة من المواد تُعرف باسم المواد الهجينة العضوية ذات الجسيمات السائلة المشابهة للنانو (NOHMs) تتمتع هذه المواد بخصائص فريدة، وتشمل البحث الذي تم تقديمه هنا النتائج التجريبية والمحاكاة الجزيئية على حد سواء. يتم امتصاص ثاني أكسيد الكربون عن طريق السوائل الأيونية الفيزيائية والكيميائية باستخدام الهياكل المعدنية العضوية (MOFs) لامتصاص ثاني أكسيد الكربون وتقنية التجديد بالفراغ للأغشية (MVR) توضح النتائج المقدمة هنا مزايا وعيوب هذه النهج **الكلمات المفتاحية:** امتصاص ثاني أكسيد الكربون، تقنية المجموعات الأيونية، مواد الهجين العضوية ذات الجسيمات السائلة المشابهة للنانو (NOHMs) الهياكل المعدنية العضوية (MOFs) تقنية تجديد الغشاء بالفراغ (MVR).