

Carbon Dioxide Capture: Current Status and Future Prospects

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Abstract: The surge in greenhouse gas emissions, predominantly in the form of carbon dioxide (CO₂) spurred by the Industrial Revolution, has surpassed the critical threshold of 400 ppm, fueling global warming, ocean acidification, and climate change. To mitigate the adverse effects of these emissions and limit the global temperature rise to below 2 °C, the ambitious target of achieving net zero emissions by 2050 was established in the Paris Agreement. Current state-of-the-art technologies, such as amine scrubbing, remain problematic owing to their high energy requirements, susceptibility to corrosion, and other operational challenges. Owing to the lack of suitable technologies coupled with escalating energy demand, there is still a significant amount of carbon dioxide being released into the atmosphere. Accordingly, there is an urgent need for the development of alternative technologies that offer high efficiency, low energy consumption, cost-effective installation, and operation. In this review, we delve into the emerging technologies poised to address these challenges, evaluating their maturity levels in comparison to existing commercially available solutions. Furthermore, we provide a brief overview of ongoing efforts aimed at commercializing these innovative technologies.

Keywords: Absorbents · Adsorbents · Carbon capture and storage · CO₂ emissions · Global warming · Membranes



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1. Introduction

Greenhouse emissions, primarily stemming from the burning of fossil fuels, represent a critical component of anthropogenic climate change. These emissions, predominantly in the form of carbon dioxide (CO₂), contribute to the greenhouse effect, trapping heat within the Earth's atmosphere and leading to global warming.^[1–6] This surge in carbon emissions has led to a rise in global temperatures, causing adverse impacts on ecosystems, weather patterns, and sea levels.^[7] In response to the escalating climate crisis, the international community came together to address the issue through the Paris Agreement. Adopted in 2015 at the 21st UN Climate Change Conference (COP21), the accord aims to limit global warming to well below 2 °C above pre-industrial levels, with efforts to pursue a more ambitious target of limiting the increase to 1.5 °C (Fig. 1).^[8] A pivotal aspect of the Paris Agreement is the commitment of participating nations to achieve net-zero carbon emissions by 2050. In 2015, the United Nations (UN) formally adopted the Sustainable Development Goals (SDGs) as a part of the 2030 agenda, encompassing **Goal 13: Climate Action**. This milestone is imperative to combat climate change and its cascading effects through rigorous measures, including mitigating greenhouse gas emissions, the adoption of renewable energy sources, optimizing energy efficiency, and other related initiatives.^[9]

The global emissions primarily stem from the absence of reliable, cost-effective technologies capable of capturing CO at point sources, particularly from flue gases. Currently, the forefront of carbon capture technology lies in amine scrubbing, a process centered on the chemical reaction between CO₂ and aqueous amine solutions followed by their regeneration at elevated temperatures. Despite its advantages, including high CO₂ selectivity (~95%) and relatively low cost, amine scrubbing faces significant drawbacks such as the requirement for high regeneration temperatures, limited uptake capacity (typically 10–15 wt.%), adsorbent degradation, loss, and equipment corrosion, rendering it less appealing to industries. For example, a coal power plant employing amine

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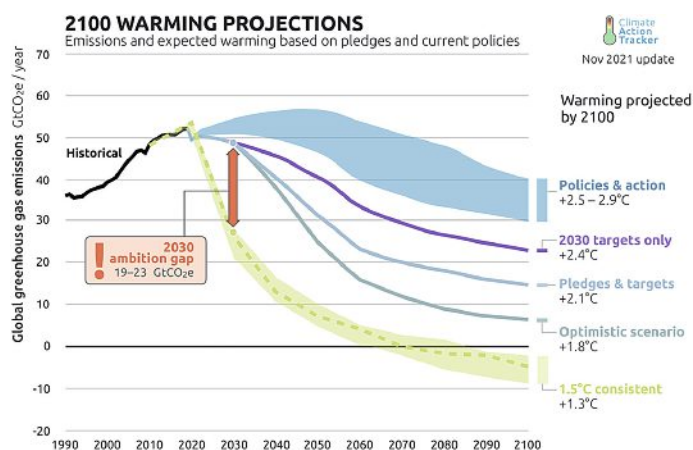


Fig. 1. Projections for global warming until 2100 indicate a substantial disparity between the targeted limit of 1.5 °C, as outlined in the Paris Agreement, and the existing policies and actions. Figure Copyright © 2021 by *Climate Analysis and New Climate Institute*. All rights reserved.^[4]

scrubbing consumes approximately 30% of its energy for carbon capture.^[10] Given the rising energy prices, the economic feasibility of releasing CO₂ rather than capturing it becomes more apparent. In light of escalating energy demands and prevailing policies, global temperatures are projected to increase by 2 to 3 °C by 2100, surpassing the target outlined in the Paris Agreement (Fig. 1). Limiting the temperature rise to 1.5 °C necessitates substantial reductions in greenhouse gas emissions alongside intensified efforts in advancing carbon capture and storage (CCS) technologies. Hence, in this review, we analyze emerging CCS solutions, encompassing both point source and direct air capture (DAC), from both scientific and industrial perspectives.

2. Carbon Capture and Storage

The initial strategy for mitigating global greenhouse gas emissions involves prevention through the capture of CO₂ from point sources. Roughly 72% of CO₂ emissions are attributed to energy production, where fossil fuels such as coal, natural gas, and petroleum products are combusted to generate heat. The resulting exhaust, commonly known as flue gas, primarily comprises nitrogen (75–85%), water vapor (5–15%), and CO₂ (7–15%).^[11] In many applications, including geological storage, it is necessary to enrich CO₂ to purities exceeding 90% to minimize energy inefficiency.^[12] The primary challenge lies in efficiently separating CO₂ from these mixtures to enrich it to the required level. In this context, we will examine prominent and competing technologies: absorbents, adsorbents, and membranes.

2.1 Amine Scrubbing

Absorbents play a crucial role in the separation of CO₂ from flue gas, leveraging the distinct chemical and physical properties that differentiate it from nitrogen gas. A prime example is the amine scrubbing process, utilizing solutions of amines such as monoethanolamine (MEA), methyl diethanolamine (MDEA), piperazine (PZ), and ethyl diethanolamine (EDEA) as absorbents (Fig. 2A).^[10,13] Amines exploit the reactivity of CO₂ in contrast to the inertness of nitrogen for effective separation. Despite their high selectivity in a single step due to a reversible reaction with CO₂, amines face limited popularity owing to the energy penalty associated with regeneration (Fig. 2A).

Another promising category of chemical absorbents for CO₂ capture involves metal and metal oxides, such as MgO and CaO.^[14] These oxides react with CO₂ to form respective bicarbonates and carbonates, offering high theoretical uptake capacities of approximately 0.78g CO₂/g CaO and 0.79g CO₂/g MgO.^[15] Additionally, these absorbents boast high selectivities for CO₂ in a single step. Despite these advantages, CO₂ capture using CaO/MgO encounters challenges including high regeneration temperatures (600–900 °C), significant capacity loss upon cycling, and slug-

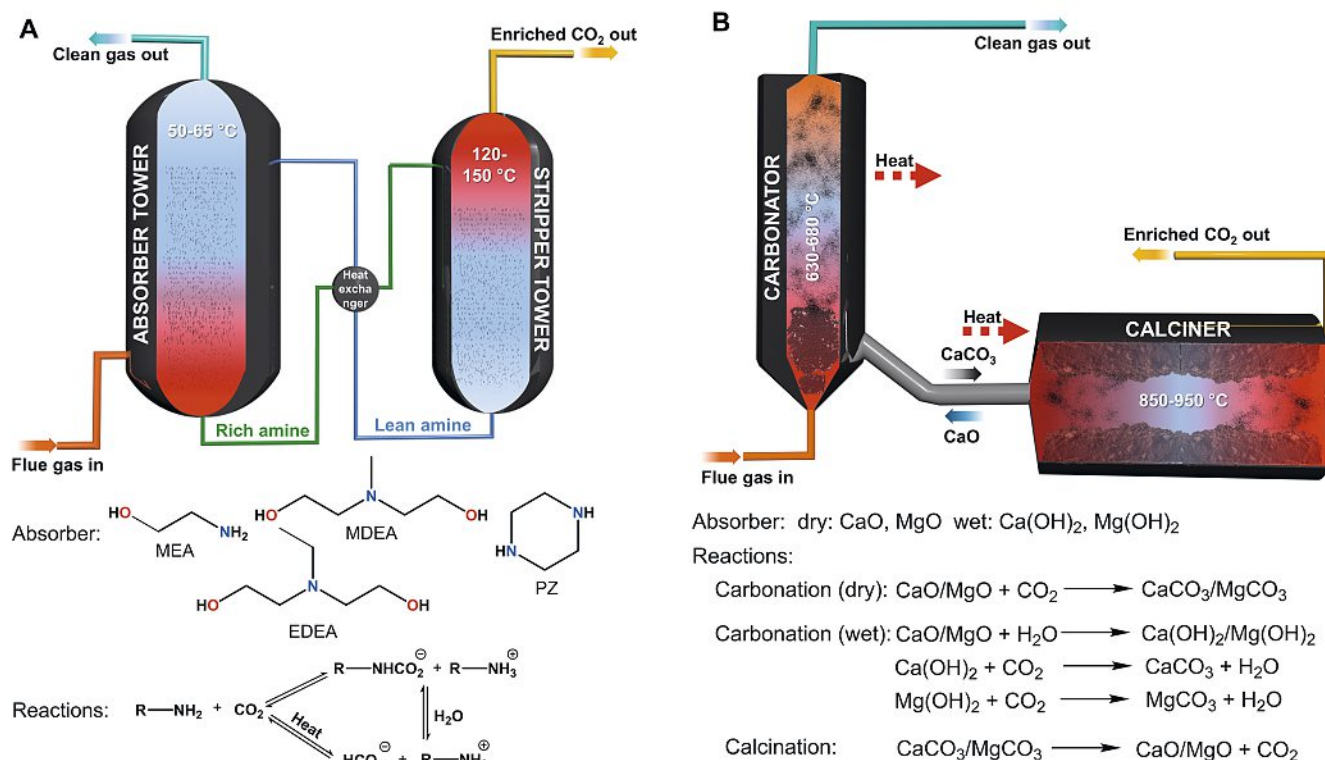


Fig. 2. Simplified schematics of A) the amine scrubbing process adapted from ref. [5], and B) carbonate looping (lime scrubbing) adapted from ref. [6] including commonly used absorbents and process reactions.

gish uptake kinetics (Fig. 2B).^[16] Currently, chemical absorption processes such as amine and lime scrubbing remain unattractive for capturing CO₂ from the emissions sources.

In the ongoing efforts to combat climate change, a significant avenue is the capture of CO₂ directly from the atmosphere, known as direct air capture (DAC). Despite their inherent disadvantages, chemical adsorbents remain unparalleled in this context, primarily due to the low concentrations of CO₂ in the atmosphere that render alternative techniques inefficient.^[17] Companies like Climeworks AG (www.climeworks.com), a spin-off from ETH Zurich, exemplify this approach by utilizing amine-functionalized sorbents for DAC.^[18] Their technology relies on amine-functionalized cellulose membranes to capture CO₂ directly from the atmosphere for subsequent geological storage.^[19] Other notable companies such as Global Thermostat (www.globalthermostat.com), Antecy B. V., and Oy Hydrocell Ltd. (www.hydrocell.fi) are also among the companies that use amine functionalized sorbents for DAC.^[20]

Furthermore, companies like Carbon Engineering ULC (www.carbonengineering.com), based in Canada, utilize the calcium looping process, which employs CaO as an absorbent for capturing atmospheric CO₂. Similarly, Heirloom Carbon Technologies (www.heirloomcarbon.com) employs calcium oxide (CaO) as a promising avenue for sustainable carbon capture. Their method involves wet calcium looping, wherein CaO is hydrated to produce calcium hydroxide (Ca(OH)₂), which subsequently reacts with atmospheric CO₂ to yield limestone (CaCO₃).^[21] This limestone is then decomposed back into CaO and CO₂. Finally, the captured CO₂ is injected underground for permanent geological storage. These diverse approaches underscore the evolving landscape of carbon capture technologies and their potential contributions to mitigating climate change.

The reaction of carbon dioxide with lime (CaO) to form carbonate presents an opportunity for permanent carbon storage, particularly in the context of reducing emissions associated with cement production.^[22] This phenomenon has acquired attention for its potential role in mitigating greenhouse gas emissions by sequestering CO₂ within concrete structures. Another avenue of research explores the utilization of cement, primarily composed of CaO, as a permanent means of CO₂ storage.^[23] This innovative approach is exemplified by CarbonCure (www.carboncure.com), an American company that injects CO₂ into cement during production. Similarly, Swiss companies such as Neustark AG (www.neustark.com) and Zirkulit Beton AG (www.beton.zirkulit.ch) have embraced this concept, actively capturing CO₂ and incorporating it into cement for long-term storage purposes. These initiatives showcase the potential of leveraging cementitious materials as a possible strategy for carbon capture and storage, contributing to sustainable development and environmental impact.

2.2 Adsorbents via PSA

2.2.1 Activated Carbon and Composites

Porous adsorbents, exemplified by activated carbon and zeolites, exploit disparities in physical properties between nitrogen (N₂) and CO₂ to achieve separation. These materials predominantly consist of micro (<2 nm) and mesopores (2-50 nm), with CO₂'s smaller kinetic diameter of 3.30 Å allowing faster diffusion into these pores compared to N₂ (kinetic diameter of 3.64 Å).^[24] The adsorption process, being purely physical, facilitates the regeneration of activated carbon at lower temperatures in contrast to amine scrubbing, often utilizing pressure swing adsorption (PSA) at room temperature with the aid of vacuum assistance (Fig. 3A).

Despite its lower energy penalty and cost-effectiveness, activated carbon's reliance solely on pore size results in limited selectivity, necessitating multiple steps to attain CO₂ purity exceeding 90%.^[25] Additionally, these adsorbents suffer from slow kinetics, requiring prolonged equilibration times.^[23] Another significant consideration regarding adsorbents is the impact of temperature; the uptake capacity exhibits an inverse relationship with temperature. This necessitates flue gas cooling to near-room temperatures to optimize performance, further diminishing the competitiveness of activated carbon. To enhance its competitiveness with scrubbing processes, research efforts have been directed towards augmenting activated carbon's CO₂ selectivity, achievable through the formation of composite materials or the enhancement of pore structures.^[26]

Chemical absorbents like metals and metal oxides (*e.g.* MgO, CaO) that facilitate enhanced CO₂-selectivity *via* chemisorption can be integrated into activated carbon structures.^[27] Previous endeavors involved combining activated carbon with various metals and metal oxides to promote chemisorption, thereby enhancing both CO₂ selectivity and capture capacity.^[26] Alternatively, porous ordered materials such as zeolites, silicates, and metal-organic frameworks (MOFs) have been utilized as fillers to augment pore structure and increase CO₂ selectivity.^[26,28]

While these strategies enhance the efficacy of activated carbon in terms of CO₂ selectivity, composite materials frequently encounter obstacles such as elevated regeneration temperatures, steep production expenses, diminished porosity, and uneven filler distribution.^[26,28a] Furthermore, a drawback arises from the fact that CaO is ultimately generated through the thermal decomposition of limestone, a process that releases a stoichiometric amount of CO₂.

Another vital component in the flue gas mixture is water, which presents its own set of challenges. Amines are employed as aqueous solutions, and the presence of water expedites the scrubbing process. Moreover, research indicates that the presence of water enhances the uptake properties of lime-based oxides for CO₂ capture.^[29] However, water also competes with CO₂ for the same binding sites within these composite materials.^[30]

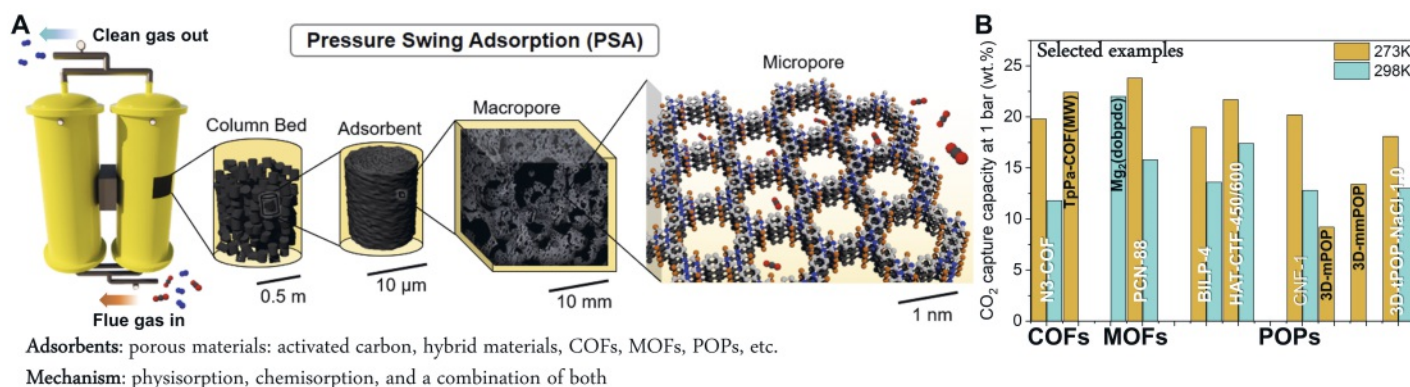


Fig. 3. A) Simple schematics of PSA setup and commonly used adsorbents with respective mechanisms. Reproduced from ref. [1] with permission. B) Selected examples of COFs, MOFs, and POPs for CO₂ capture at 273 K and 298 K. Data is reproduced from original reports.

2.2.2 COFs and MOFs

A more promising strategy involves designing porous materials with tailored structures for efficient CO₂ capture. Hence, ongoing research focuses on developing novel porous materials that leverage differences in chemical properties to enhance both selectivity and kinetics. Recently, design principles for porous organic polymers, POPs, aimed at enhancing CO₂-selectivity and uptake capacity have been summarized by our group, emphasizing the importance of ultramicropores (<0.7 nm) and heteroatoms such as nitrogen to enhance selectivity *via* dipole-quadrupole interactions.^[25,31]

In the realm of CO₂ capture, covalent organic frameworks (COFs) and metal-organic frameworks (MOFs) have garnered considerable attention owing to their remarkable properties, including high porosity, tunable chemical structures, low structural density, adjustable pore sizes, and robust thermal and chemical stabilities.^[32] A pivotal aspect of COF and MOF design lies in the flexibility afforded by linker selection, enabling tailored frameworks capable of CO₂ capture *via* physisorption and/or chemisorption.^[28c,33] By integrating various functional groups such as imines, boronates, and triazines, it becomes feasible to fine-tune the CO₂ uptake capacities of COFs.^[34] For instance, the triazine-linked N₃-COF exhibited notable CO₂ uptake performances of 19.8 and 11.8 wt.% at 1 bar and temperatures of 273 and 298 K, respectively (Fig. 3B).^[35] Similarly, Wang *et al.* synthesized the imine-linked TpPa-COF (MW), demonstrating a remarkable CO₂ uptake performance of 22.4 wt.% and a CO₂/N₂ selectivity of 32 at 273 K and 1 bar.^[36] Despite the promising performance of COFs as CO₂ adsorbents, challenges related to scalability, reproducibility, and crystallinity hinder their commercial utilization for CO₂ separation from flue gas.^[32,37]

Conversely, MOFs leverage inorganic polynuclear clusters known as secondary building units alongside pore structure and linker modulation to enhance CO₂ capture capabilities.^[38] Incorporating metals such as Mg, Zn, and Cu, in conjunction with nitrogen moieties *via* linkers, can augment the CO₂ affinity of MOFs.^[39] For instance, Long and colleagues synthesized Mg₂(dobpdc), which exhibited a substantial CO₂ capture capability of 22.0 wt.% at 298 K and 1 bar (Fig. 3B).^[40] Further modifications of Mg₂(dobpdc) with alkylamines yielded a CO₂ capture efficiency of 8.1 wt.% under atmospheric conditions (298 K and 0.39 mbar). Similarly, Li *et al.* designed the Cu MOF, PCN-88, capable of capturing 23.8 wt.% of CO₂ at 273 K and 15.8 wt.% at 296 K and 1 bar.^[41] MOFs hold promise as potential adsorbents for both point source CO₂ capture and atmospheric extraction. In line with this potential, NovoMOF AG (www.novomof.com), a Swiss startup, endeavors to commercialize MOFs for CO₂ capture applications. Moreover, water constitutes a critical component of flue gas, necessitating consideration in CO₂ capture applications from both flue gas and the atmosphere. It is known that certain COFs and MOFs exhibit poor water stability and inferior uptake performance under wet conditions.^[34,37,42]

2.2.3 Porous Organic Polymers

Porous organic polymers (POPs), serving as the amorphous counterparts of COFs and MOFs, boast superior chemical and thermal stabilities, tunability, and high surface areas and pore volumes.^[25,43] Notably, the diverse synthetic approaches available for POPs enable large-scale synthesis and offer greater flexibility in linker selection, thereby expanding their application scope.^[25,43,44] Consequently, POPs have been extensively explored for various applications, including CO₂ capture and separation.^[45]

For instance, El-Kaderi and colleagues demonstrated the efficacy of benzimidazole-linked polymer (BILP-4) with a CO₂ uptake capacity of 19.0 wt.% at 273 K, decreasing to 13.6 wt.% at 298 K and 1 bar, alongside CO₂/N₂ selectivities of 79 and 32, respectively.^[46] Similarly, Zhu *et al.* synthesized modified covalent triazine frameworks (CTF), a subclass of POPs, for efficient CO₂

capture. The HAT-CTF series exhibited promising results, with HAT-CTF-450/600 showing a CO₂ uptake capacity of 21.7 wt.% and 17.4 wt.% at 273 and 297 K and 1 bar, respectively, along with a CO₂/N₂ selectivity of 110 at 297 K.^[47]

Our research group has been active in developing novel porous organic polymers for CO₂ capture, separation, and conversion.^[48] For instance, we synthesized carbon nitride frameworks (CNF) *via* a chemical blowing approach, with CNF-1 demonstrating CO₂ uptake capacities of 20.2 wt.% and 12.8 wt.% at 273 K and 298 K and 1 bar, respectively.^[48c] Moreover, by incorporating highly CO₂-selective cyclotetrazinone macrocycles into POP structures, we obtained 3D-mPOP, exhibiting a CO₂ uptake of 9.2 wt.% at 1 bar and a CO₂/N₂ selectivity of 137 at 273 K.^[31,49] Subsequent modifications of linker length led to an increase in CO₂ capture capacity to 13.4 wt.%.^[50] Additionally, we prepared POPs by incorporating tetraphenylene moieties to enhance CO₂ selectivity and used NaCl as a template to tailor pore size and porosity. The resulting 3D-tPOP-NaCl-1.0 demonstrated remarkable CO₂ uptake capacities of 18.1 wt.% at 273 K and 13.0 wt.% at 298 K and 1 bar, attributed to the presence of ultramicropores and nitrile groups.^[51] Furthermore, we recently reported porous polyphenylenes (pPPs) *via* Diels-Alder cycloaddition for CO₂ capture. We synthesized 2D and 3D-pPPs exhibiting CO₂ uptake performances of 60.7 wt.% and 65.2 wt.% at 296 K and 27 bar, respectively.^[52] These findings underscore the immense potential of POPs for CO₂ capture and separation, further enhanced by the wide range of available organic linkers and post-modification strategies, facilitating modification of functional groups to target specific CO₂ capture conditions.

One prevailing challenge associated with these porous adsorbents for CO₂ capture is their comparatively higher production costs relative to activated carbon, compounded by the lack of comprehensive long-term stability studies. Furthermore, performance loss upon scaling up poses a significant obstacle to the active commercialization of such materials for industrial applications, both for point source and atmospheric carbon capture. Thus, further research efforts are needed to enhance the accessibility and applicability of these materials for industrial-scale carbon capture endeavors.

2.3 Membranes

Gas separation membranes represent a prevalent method for separating gases, including applications such as flue gas treatment.^[53] A membrane functions as a semipermeable barrier with a nanoporous structure, allowing one component of the gas mixture to selectively permeate through. The performance of a membrane is characterized by two critical factors: permeance (or permeability) and selectivity. Permeance measures the rate at which gas passes through the membrane, affecting pressure drop across the membrane, while selectivity gauges the purity of the permeated component gas. Permeance, in a nutshell, represents the flow rate across the membrane, standardized by transmembrane pressure and membrane area, commonly denoted in gas permeation units (GPU) where 1 GPU equals to 3.35×10^{-10} mol·s⁻¹·m⁻²·Pa⁻¹.^[54] Ideal membranes would exhibit high values for both parameters; however, in practice, there exists a trade-off between them (Fig. 4). Membranes with high permeance typically demonstrate low selectivity and vice versa.^[55]

One notable advantage of membranes is their ability to facilitate continuous processing without the need for regeneration. Nevertheless, the current state-of-the-art membranes predominantly consist of polymers, which exhibit high selectivity but low permeance, necessitating substantial pressurization of up to 7–13 bars.^[56] Moreover, due to the inherently low concentration of CO₂ in flue gas, membranes typically require multiple stages to achieve CO₂ purity comparable to that of amine scrubbing, thereby increasing operational costs and diminishing their attractiveness for widespread applications.^[57] Consequently,

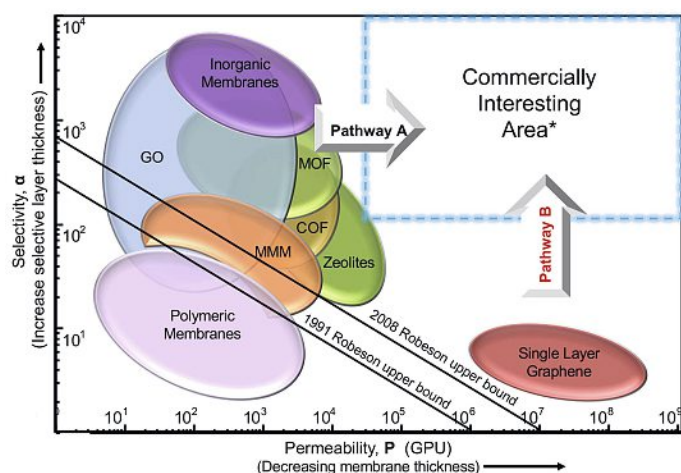


Fig. 4. Schematic representation of the trade-off between permeability and selectivity including 1991 and 2008 Robeson's upper bonds. The position of commercially interesting area is relative to the upper bounds and can vary depending on the application. Adapted from ref. [3]

significant efforts have been directed towards developing membranes that exhibit both high selectivity and permeance. Breakthroughs in this domain include the discovery of mixed-matrix membranes (MMMs)^[3] and two-dimensional (2D) membranes based on porous graphene, which enables unprecedented permeance values, that are a million times higher than those of polymer membranes.^[58] In light of these advancements, the exploration of MMMs and graphene-based membranes holds considerable promise.

2.3.1 Mixed-matrix Membranes

Mixed-matrix membranes represent a class of composite membranes comprising a filler material integrated within a polymer matrix. While polymeric membranes demonstrate high selectivity, they often exhibit limited permeance. Conversely, inorganic membranes such as silicates and aluminates exhibit high permeance but lower selectivity. MMMs capitalize on the strengths of both materials, combining the high selectivity of the polymer matrix with the elevated permeance of inorganic membranes, thereby overcoming the selectivity-permeance trade-off.^[3,59] The incorporation of fillers not only enhances the separation performance of the polymeric membrane but also improves its mechanical and chemical stability and robustness. Additionally, fillers can retard polymer membrane aging by providing structural rigidity. These advantages of MMMs over conventional polymeric membranes render them appealing for advanced gas separation applications, including flue gas treatment and CO₂ separation.^[60]

The performance of MMMs hinges on the choice of filler materials and their interaction with the polymer matrix. A primary challenge lies in achieving uniform filler distribution within the polymer matrix and ensuring effective interaction between filler materials and the polymer matrix. Common strategies for preparing MMMs involve incorporating filler materials during polymer casting. Furthermore, it is feasible to modify filler materials in MMMs to enhance their performance and properties. A wide array of materials, including zeolites, MOFs, COFs, POPs, and shape-persistent molecules, have been investigated as filler materials.^[61]

For example, Ahmad and Hagg prepared an MMM using zeolite 4A as filler and polyvinyl acetate as the polymer, achieving a CO₂/N₂ selectivity of 100.54 with a CO₂ permeance of 0.03 GPU at 35 wt% zeolite loading.^[62] Attempts have also been made to synthesize MMMs using unconventional materials as fillers. For instance, Kawakami and colleagues modified nonporous silica particles into pearl-necklace-shaped nanoparticles (P-NP) and

embedded them into the PIM-1 matrix. At 50 wt.% loading, they obtained a CO₂ permeance of 153.5 GPU and CO₂/N₂ selectivity of 16.6.^[63] However, the CO₂ permeance obtained was not competitive enough for CO₂ separation compared to conventional scrubbing processes. Better CO₂ permeation results were achieved by Zhu *et al.* who utilized UiO-66-CN MOF as a filler and PIM-1 as a matrix, obtaining UiO-66-CN@sPIM-1 membranes with a CO₂/N₂ selectivity of 53.5 and CO₂ permeance of ~464 GPU at 20 wt.% MOF loading.^[64] Even more promising results were reported by Wang *et al.* who incorporated MKP nanoparticles into a poly(vinylamine) matrix. The prepared MKP-PVAm/mPSf membrane, with ~44 wt% loading, demonstrated a CO₂ permeance of 823 GPU and CO₂/N₂ selectivity of 242 at 5 bar transmembrane pressure.^[65]

Given these impressive performance metrics, MMMs have garnered significant attention for commercialization. Their foundation on polymeric membranes, a well-established technology, makes the commercialization of MMMs relatively straightforward. For instance, Zurich-based startup UniSieve (www.unisieve.com) is actively developing MMMs using polymers such as poly(ether sulfone) as matrix and ZIF-8 as filler for gas separation applications, including CO₂ separation from flue gas.^[66]

2.3.2 Porous Graphene Membranes

Ever since its discovery, graphene has found widespread applications across various fields, spanning from electronics and photovoltaics to medical sciences, sensors, desalination, and gas separations.^[67] Notably, graphene's atomic thickness endows it with unparalleled permeability, resulting in minimal pressure drops across membranes.^[68] Therefore, graphene is considered an ideal membrane material. However, for separation applications, artificial pores must be introduced, as pristine graphene lacks inherent porosity. Particularly for achieving high gas selectivities, the creation of numerous small pores (<3 nm) is imperative to enable molecular sieving, thereby ensuring high permeance and selectivity concurrently.

Early endeavors in graphene membranes explored gas permeation through intrinsic defects in chemical vapor deposition graphene.^[69] Subsequent efforts focused on etching graphene using ultraviolet (UV)/ozone plasma to observe gas permeation, demonstrating the potential of precisely fabricated pores to confer high gas selectivities.^[70] However, UV/ozone etching often yields low porosity, thereby limiting permeability.^[70,71]

To address this limitation, ion beam irradiation in conjunction with oxygen, ozone, and UV plasma has been employed to enhance the porosity of graphene-based membranes, albeit predominantly leading to pore enlargement rather than pore creation.^[72] A significant breakthrough in graphene membrane technology was achieved through focused-ion beam (FIB) irradiation, which generated a multitude of pores, resulting in a porous graphene membrane exhibiting remarkable CO₂ permeance (~1.6 × 10⁷ GPU).^[58] However, the inherent challenge associated with FIB irradiation lies in the large pore size generated (>5 nm), which adversely affects gas selectivities and poses significant scalability constraints, thereby limiting their practical applicability to fundamental scientific investigations. In contrast, UV/O₂/ozone plasma etching techniques offer scalability potential, making them conducive for large-scale commercial applications.^[71]

Recent advancements have introduced novel etching methodologies, such as millisecond ozone gasification at elevated temperatures, as an effective means to fabricate porous graphene membranes, yielding competitive CO₂ permeance values suitable for carbon capture from flue gas streams (CO₂ permeance of ~9600 GPU with CO₂/N₂ selectivity of 24.4).^[73] Encouraged by these developments, concerted efforts are underway to advance commercialization endeavors, exemplified by initiatives

such as Divea (www.divea.ch), spearheaded by École Polytechnique Fédérale de Lausanne (EPFL), aimed at commercializing graphene-based membranes for carbon capture and separation applications.

Our research efforts are similarly focused on the frontier of 2D membrane technologies, with a primary emphasis on graphene-based membranes.^[74] For example, we employed FIB irradiation to create pores on graphene and deposited metal layers in a checkerboard pattern to enable gas separation based on their affinities, thereby introducing adsorptive separation.^[4] This novel approach entails one of the gases in the mixture adhering to the adsorbent on the membrane and being retained while non-interacting components pass through, theoretically enabling the separation of any gas mixture. As a proof of concept, we successfully demonstrated the complete separation of helium and hydrogen, a notoriously challenging task using conventional methods. Additionally, we achieved a helium permeation rate of 2.6×10^7 GPU (Fig. 5A). Furthermore, we explored the potential of separating CO₂-containing mixtures by introducing more cost-effective nickel micro islands, achieving an H₂/CO₂ separation factor of 26 with an H₂ permeance of 2.0×10^5 GPU (Fig. 5A).

In another approach, we employed stepwise gold deposition to precisely adjust the size of pores on graphene, thereby facilitating the transition of gas transport from Knudsen diffusion to surface diffusion, and ultimately to molecular sieving when the pore size decreased below 3 nm.^[75] This method not only yielded a record-high H₂/CO₂ separation factor of 31.3 with an H₂ permeance of 2.23×10^5 GPU but also established an upper bound for porous graphene membranes (Fig. 5B). While our initial studies utilizing FIB milling showcased the potential of graphene as a membrane material, we recognized the limitations of this method for large-scale production. Consequently, one of our objectives was to identify a scalable pore generation method on graphene for commercialization. As a result, we recently developed a large-scale method based on inorganic silica deposition, enabling the creation of a large number of pores in various sizes.^[76]

To further advance the development and commercialization of graphene-based membranes for carbon capture and hydrogen recovery, we are currently in the process of establishing our startup, SEPARATIC (www.separatic.com). Membranes based on porous graphenes are gradually being integrated into carbon capture and separation applications. With the roll-to-roll production of graphene already established for large-scale production, the key factor for the commercialization of porous graphene membranes

lies in achieving competitive production costs that can rival more mature technologies.

3. Conclusions and Perspectives

In summary, we have provided an overview of carbon capture and separation technologies, along with ongoing research efforts aimed at enhancing the performance of existing materials. We have also briefly examined the commercial implementation of emerging technologies in this field. Mature technologies including amine scrubbing and polymeric membranes are constrained by various limitations, ultimately resulting in increased energy demand and the inadvertent release of CO₂ rather than its capture – a valuable chemical commodity. Additionally, the high installation costs associated with amine scrubbing and membrane technologies further incentivize the economically favorable option of CO₂ release. To achieve net zero emissions by 2050, there has been a shift in focus towards exploring new emerging technologies. In this direction, some of the start-up companies have already progressed to ton-scale demonstration plants and are poised for further expansion. It appears that a significant portion of the carbon capture market will be dominated by these innovative companies, offering more cost-effective alternatives for both operation and installation, especially considering the anticipated rise in carbon prices due to stricter regulations in the years to come. In conclusion, it is essential to recognize the importance of supporting these early-stage companies, either through government initiatives or market incentives, despite the inherent risks associated with their developmental stages. This support is crucial in facilitating the achievement of the net zero emissions goal. The emerging technologies outlined here primarily focus on carbon capture and separation. However, an important consideration arises once these technologies mature: what should be done with the captured CO₂? While permanent underground storage in basalt salts shows promise, it remains experimental and geographically limited at this stage. Therefore, as the costs of carbon capture and separation decrease, it is imperative to advance technologies that convert CO₂ into value-added C1 and C_n products, thereby completing the circular economy loop. The ultimate objective is to establish a fully functional carbon circular economy, aligning with the net zero emissions goal outlined in the Paris Agreement.

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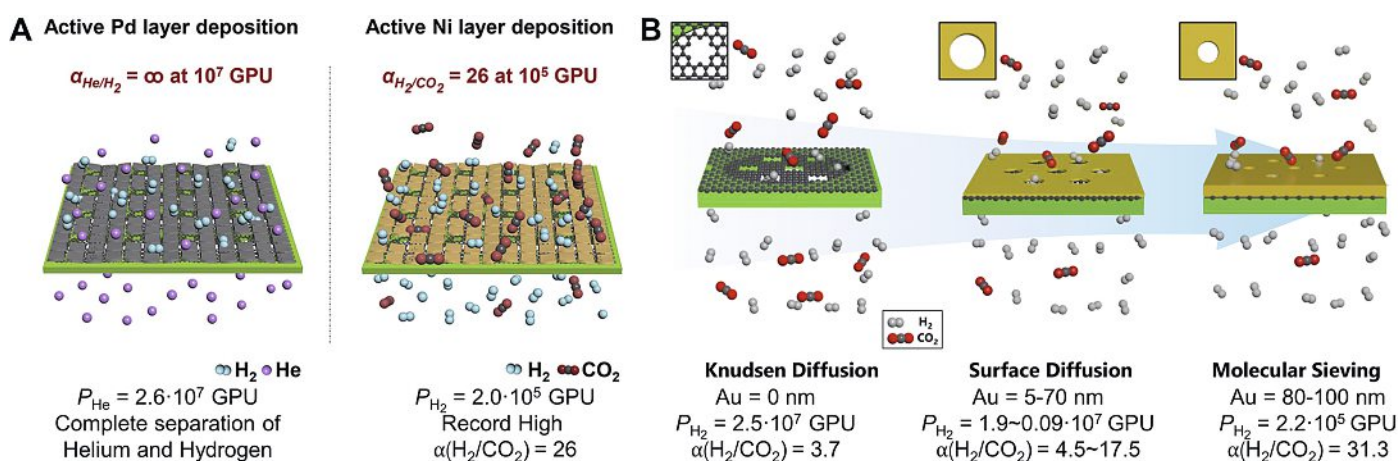


Fig. 5. A) Schematic view of adsorptive separation that led to complete separation of helium/hydrogen, record high H₂/CO₂ selectivity at H₂ permeance of 2.0×10^5 GPU. Reproduced from ref. [4] B) Transition of the gas transport mechanism from Knudsen diffusion to surface diffusion and molecular sieving with increasing gold layer thickness. Data is reproduced from original reports.

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