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Unravelling the nanoarchitectonics of –C–N– linkages in covalent organic frameworks for CO₂ capture—a mini-review

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Covalent Organic Frameworks (COFs) featuring carbon–nitrogen (C–N) linkages represent a versatile class of crystalline, porous materials constructed through the dynamic covalent bonding of organic monomers. Schiff-base, C–N imine-linked COFs have garnered considerable interest for carbon dioxide (CO₂) capture owing to their highly tunable structures, excellent thermal and chemical stability, and enduring porosity, which stem from their robust architectures and nitrogen-rich composition. The inherent polarity and Lewis basicity of C–N bonds promote strong CO₂ adsorption *via* dipole–quadrupole and hydrogen bonding interactions, enhancing uptake and selectivity, particularly under low-pressure or humid conditions, making them ideal for real-world applications in CO₂ capture. Key structural attributes, such as nanoarchitectures based on small pore sizes, electron-rich substituents, and three-dimensional multi-layered frameworks, further enhance both stability and CO₂ affinity by reinforcing π -conjugation and improving weak supramolecular interactions. This mini-review delves into the mechanistic role of C–N connectivity in influencing CO₂ gas affinity and framework stability, highlighting a promising pathway for the rational design of next-generation COFs for carbon dioxide capture and storage technologies.

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1. Introduction

Greenhouse gases (GHGs), which trap heat at the Earth's surface, play a crucial role in maintaining the planet's energy balance and supporting life. In the absence of these gases, the global average surface temperature would be approximately 32.5 °C lower than the current average of 14.4 °C, resulting in an estimated temperature of –18 °C.¹ However, elevated con-

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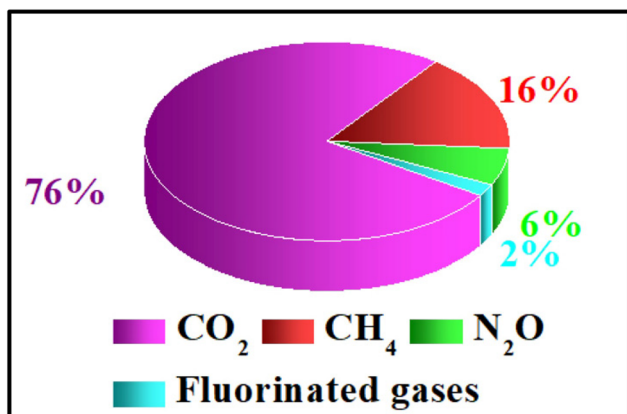


Fig. 1 Source: forcing greenhouse gases – Global emissions (2012), International Energy Agency.

centrations of GHGs – such as carbon dioxide (CO₂), nitrous oxide (N₂O), methane (CH₄), water vapour (H₂O), and fluorinated gases – pose significant challenges to sustainable development.^{2,3} According to the Intergovernmental Panel on Climate Change (IPCC) Sixth Assessment Report, radiative forcing – defined as the change in net radiative flux (W m⁻²) due to drivers such as CO₂ emissions, volcanic aerosols, and variations in solar output – is a key metric in assessing climate impact.^{4,5} Since the 1980s, CO₂-induced radiative forcing has increased by approximately 57%, and CO₂ currently accounts for about 80% of the total radiative forcing attributed to GHG emissions.^{2,6} As illustrated in Fig. 1, rising atmospheric CO₂ concentrations are the primary driver of contemporary climate change. Recent studies by the European Chemical Industry Council propose that CO₂ utilized in fuel production and other short-lived applications can be classified as “negative emissions”, regardless of its source or sequestration pathway.^{7,8}

According to the 2023 annual report from the National Oceanic and Atmospheric Administration (NOAA) Global Monitoring Laboratory, the global average atmospheric CO₂ concentration reached 419.3 ppm.⁹ This rise in atmospheric CO₂ is of critical concern for two main reasons. First, CO₂ is the most impactful anthropogenic greenhouse gas, with a strong capacity to absorb and re-radiate infrared radiation, unlike diatomic gases such as oxygen and nitrogen. The continuous increase in CO₂ concentration, primarily driven by human activities, is directly linked to the rise in global temperatures.¹⁰ Second, CO₂ readily dissolves in ocean water, forming carbonic acid and contributing to ocean acidification – a process that alters marine chemistry and threatens aquatic ecosystems.¹¹

Given these environmental consequences, the development of effective CO₂ capture and conversion technologies is imperative to mitigate atmospheric CO₂ levels and reduce associated ecological risks. Historically, anthropogenic CO₂ emissions began to escalate significantly during the Industrial Revolution, initiated around 1750 with the advent of steam power. The large-scale combustion of fossil fuels – initially coal, followed by oil and natural gas – has since accelerated the release of CO₂ into the atmosphere. To limit global warming and meet the objectives outlined in the Paris Agreement, it is essential to implement sustainable strategies aimed at reducing global CO₂ emissions by 45% by 2030 and achieving net-zero emissions by 2050.¹² Fig. 2 presents normalized annual CO₂ emissions from fossil fuel combustion and industrial activities. Additionally, the figure highlights the regional contribution from Asia (Fig. 2b), illustrating the exponential increase in CO₂ emissions over recent decades.

Technologies enabling the efficient utilization of CO₂ are critical to establishing a sustainable, circular carbon economy, where CO₂ is treated as a valuable resource rather than a waste



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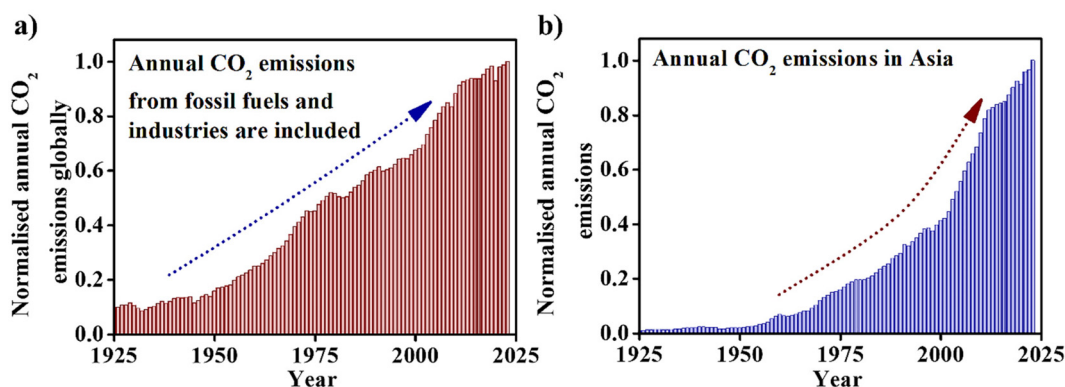


Fig. 2 (a) Annual CO₂ emission data from fossil fuels and industries globally (divided the values with maximum value obtained 37.79 billion tonnes). (b) Annual CO₂ emissions in whole region of Asia (divided the values with maximum value obtained 19.75 billion tonnes). Source: Global Carbon Budget (2024).

product. As such, carbon dioxide utilization is considered a viable short- to mid-term strategy for mitigating atmospheric CO₂ levels due to its relatively straightforward implementation. The three principal CO₂ mitigation approaches include pre-combustion,¹³ post-combustion,¹⁴ and oxy-fuel combustion methods.¹⁵ A comparison of the key advantages and disadvantages of different approaches is discussed in Table 1. In conjunction, five major CO₂ separation techniques have been developed: chemical looping,¹⁶ membrane separation, adsorption, absorption, and cryogenic distillation.^{17,18} Table 2. provides a comparison of the main merits and demerits of various approaches. Among these, adsorption-based systems have gained significant attention due to their high CO₂ uptake capacities, low energy requirements, and reduced operational and maintenance costs.¹⁹ The development of efficient, porous adsorbent materials that demonstrate high selectivity for CO₂

in the presence of other gases has thus become a research priority.

Adsorbents are generally classified into low-temperature and high-temperature categories. High-temperature materials, such as reactive metal oxides (*e.g.*, hydrotalcites and calcium oxide), are not considered within the scope of this article.³⁰ Low-temperature adsorbents can be further categorized as chemisorbents or physisorbents. Chemisorbents typically include amine-functionalized materials supported on oxides, polymers, or metal-organic frameworks (MOFs), while physisorbents comprise zeolites, MOFs, activated carbons, and related porous compounds.³¹ Porous materials are characterized by a network of voids, with pore sizes ranging from micropores (<2 nm), mesopores (2–50 nm), to macropores (>50 nm), facilitating gas diffusion and molecular separation.³² MOFs, in particular, have emerged as promising materials due to their

Table 1 Merits and demerits of different CO₂ mitigation approaches

CO ₂ capture methods	Merits	Demerits
Post-combustion	(a) It can capture CO ₂ from flue gases with low concentrations of CO ₂ (typically 10–15%). ²⁰ (b) Use of activated carbon as one of the adsorbents makes the process environmentally friendly. ²² (c) Captured CO ₂ can be utilized for a variety of purposes, such as enhanced oil recovery (EOR) or as a feedstock for chemical production, making the process more economically viable. ¹³	(a) CO ₂ is captured from a low-pressure (1 bar) gas stream with low CO ₂ content (3%–20%), often at high temperatures (120–180 °C), and containing impurities such as SO _x and NO _x . ²¹
Pre-combustion	(a) Useful in CO ₂ capture at high pressure and concentrated gas streams, leading to carbon-free fuel. ²³ (b) Key adsorbents include zeolites, metal-organic frameworks (MOFs), carbon molecular sieves, and various polymer-based materials. ²⁰ (c) Syngas, the main product of pre-combustion capture, can be used in combined cycle power generation or as a feedstock for producing various chemicals. ²⁴	(a) High costs and increased risks limit the process. ²³
Oxy-fuel combustion	(a) Oxy-fuel combustion results in a significant reduction in NO _x emissions. ²⁵ (b) Compared to other CO ₂ removal technologies, purifying the stream is easier once trace contaminants have been removed. ²⁰	(a) A major drawback of oxy-fuel combustion is the energy-intensive air separation process required. ²⁵

Table 2 Merits and demerits of different CO₂ separation techniques

CO ₂ separation techniques	Merits	Demerits
Chemical looping	(a) It has lower energy requirements because it does not need solvent regeneration or a separate air separation unit. ²⁶ (b) It uses a reusable oxygen carrier that cycles through reduction and oxidation states, which is more energy-efficient and stable. ²⁶	(a) The oxygen carrier used in chemical looping may undergo deactivation over time due to carbon deposition, sintering, or oxidative wear. ²⁶
Membrane separation	(a) High separation efficiency and high selectivity over other gases. ²³	(a) Limited CO ₂ purity. ²⁷ (b) Lower efficiency at higher CO ₂ concentrations. ²⁷
Adsorption	(a) Suitable for temperature or pressure swing adsorption operations – low energy consumption and reduced operational and maintenance costs. ²⁰ (b) Adsorbents can often be regenerated by pressure or temperature changes, making them reusable for multiple cycles. ²⁸	(a) Scale-up challenges and the cost of adsorbent materials can hinder large-scale deployment. ²⁸ (b) Limited CO ₂ storage capacity. ²⁹
Absorption	(a) Absorption processes, especially amine scrubbing, exhibit high selectivity for CO ₂ , effectively separating it from other gases, even in diluted streams. ²³	(a) The removal of CO ₂ for the regeneration of the absorbent is energy-demanding process compared to that of adsorption techniques. ²³ (b) Solvent degradation over time leads to corrosion, which increases the maintenance and costs. ²³
Cryogenic distillation	(a) Separation of CO ₂ from streams having high CO ₂ concentrations. (b) No chemical reagents. ²³	(a) Cryogenic distillation requires large and bulky equipment. ²³

structural tunability and dual-mode interaction with CO₂. Depending on their functionalization and framework design, MOFs can exhibit both chemisorptive and physisorptive behavior, making them highly versatile candidates for CO₂ capture from flue gases and other emission sources.^{33,34}

Metal–organic frameworks are a class of crystalline materials composed of metal nodes coordinated with organic ligands, forming highly porous, extended network structures.³⁵ The concept of linking molecular building blocks *via* strong covalent bonds to yield either amorphous or crystalline materials was pioneered by Omar M. Yaghi, who later extended this approach to create MOFs and Covalent Organic Frameworks (COFs).³⁶ In 1995, O. M. Yaghi *et al.* reported the first MOFs by integrating the microporosity and crystallinity of zeolites with metal–organic linkages, marking a significant advancement in porous materials chemistry.³⁷ Since their discovery, MOFs have gained widespread attention due to their high surface area, tunable pore sizes, and structural versatility, positioning them at the forefront of applications such as gas separation,³⁸ catalysis,³⁹ sensing,⁴⁰ and energy storage.⁴¹ Despite these advantages, certain limitations – such as moisture sensitivity, complex synthesis, and limited mechanical stability – can restrict their practical deployment. In contrast, porous organic polymers (POPs) offer superior thermal and chemical stability, more facile synthesis, and greater mechanical flexibility.⁴² However, POPs are generally amorphous, in contrast to the crystallinity of MOFs, which can be a disadvantage in applications requiring ordered frameworks. Over the past two decades, several subclasses of POPs have emerged, including hypercrosslinked polymers (HCPs, developed in 1980),⁴³ polymers of intrinsic microporosity (PIMs, 2004),⁴⁴ covalent organic frameworks (COFs, 2005),⁴⁵ conju-

gated microporous polymers (CMPs, 2007),⁴⁶ and porous aromatic frameworks (PAFs, 2009).⁴⁷ The increase in research activity in this area is reflected in the growing number of publications related to porous materials for gas adsorption, as shown in Fig. 3(a–c).

Among these, covalent organic frameworks have gained considerable interest as promising candidates for CO₂ capture and conversion due to their modularity, thermal and chemical robustness, and well-defined porosity. COFs are crystalline porous materials constructed from light elements such as carbon, hydrogen, nitrogen, oxygen, and boron, linked *via* covalent bonds to form extended, periodic frameworks.^{48–52} Their well-ordered structures, functional tunability, and high surface areas render them suitable for a wide range of applications.^{53–59} The first COF was reported by A. P. Cote *et al.* in 2005, using borate linkages formed through the self-condensation of 1,4-phenylenediboronic acid.⁴⁵ In conventional COF synthesis, reversible interactions facilitate the self-correction of structural misalignments, thereby promoting higher crystallinity. However, in systems dependent on irreversible bonds, the inability to self-correct significantly complicates the formation of well-ordered and highly crystalline structures.⁶⁰ Nevertheless, B. Zhang *et al.* synthesised dioxin-linked irreversible COF from linear tetrafluorophthalonitrile (TFPN) and 2,3,5,6-tetrafluoro-4-pyridinecarbonitrile (TFPC), with triangular 2,3,6,7,10,11-hexahydroxytriphenylene (HHTP), which has excellent chemical stability.⁶¹ Later, Y. Su *et al.* developed stable cyano-substituted benzofuran-linked COFs through irreversible cascade reactions.⁶² Despite its chemical stability in irreversible COFs, the majority of COFs are produced through reversible covalent bonding. Consequently, the advancement of irre-

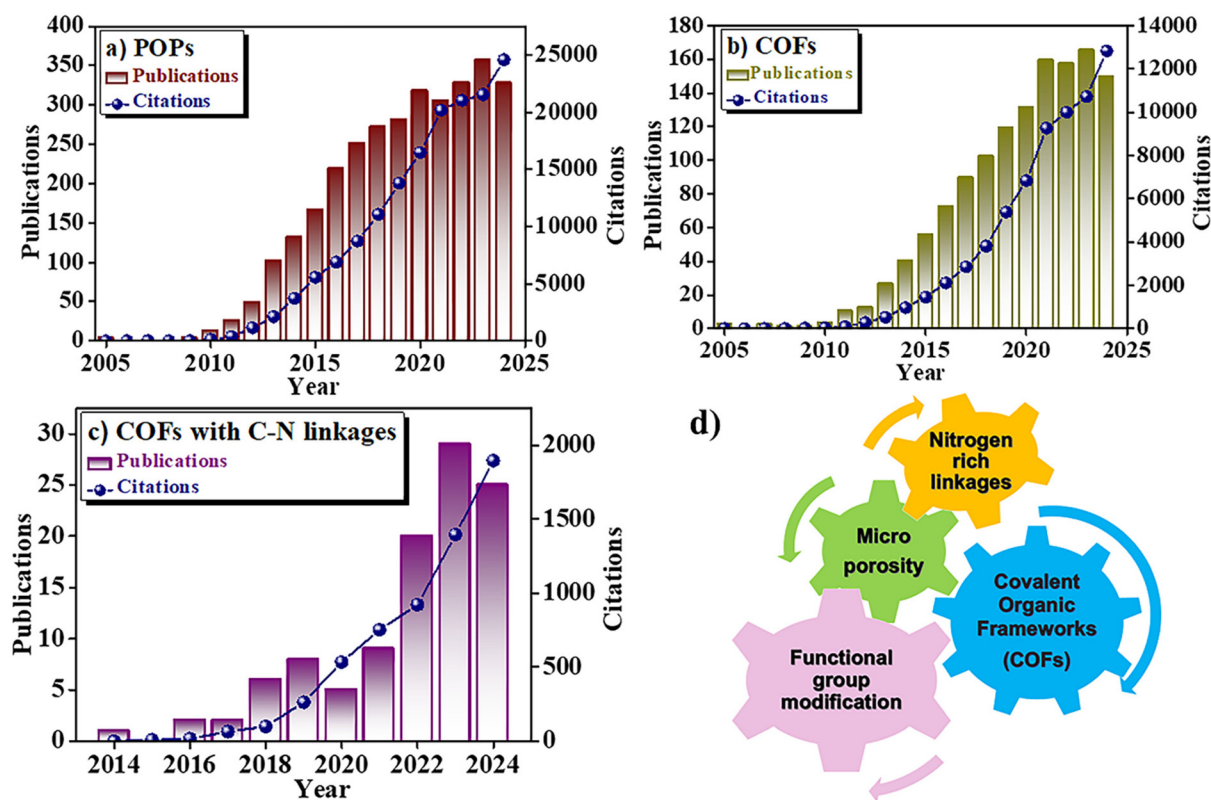


Fig. 3 (a) Annual publications and citations of Porous Organic Polymers (POPs). (b) Covalent Organic Frameworks (COFs). (c) Covalent Organic Frameworks (COFs) with C–N linkages for CO₂ capture over the past 20/10 years (data collected from Web of Science). (d) Designed properties of COFs for CO₂ capture.

versible bonding chemistry in COFs is promising, but it remains a time- and energy-demanding process.⁶⁰ This necessitates fine control over reaction kinetics and conditions, such as mild temperatures and pressures, making COF synthesis inherently more challenging than other POPs.^{63,64} COFs have been explored for numerous applications, including gas storage and separation,^{65,66} catalysis,^{67,68} optoelectronics,⁶⁹ chemical sensing,^{70–73} water purification,⁷⁴ energy storage and conversion,^{75,76} and biomedical uses such as drug delivery.^{77–79} In the context of environmental sustainability, COFs offer significant potential for CO₂ capture, particularly through functionalization and porosity strategies that enhance CO₂-philic behaviour (Fig. 3d).

The COVID-19 pandemic, while temporarily reducing global CO₂ emissions, also highlighted the need for long-term strategies to address climate change. It presented an opportunity to reassess global energy systems, urban infrastructure, and material technologies. In this regard, COFs have emerged as a sustainable solution, particularly due to their adaptability and efficiency in CO₂ capture and conversion. In celebration of the 20th anniversary of COFs, this mini-review highlights recent advancements in COFs featuring enriched nitrogen content through C–N linkages. We explore their design strategies, mechanisms of interaction with CO₂, and

their potential as high-performance adsorbents for carbon capture.

2. Historical background

The pioneering work by Furukawa and Yaghi in 2009 represented a significant milestone in the application of covalent organic frameworks for CO₂ capture. Their systematic investigation demonstrated the strong correlation between CO₂ adsorption capacity and structural features (structures are illustrated in Fig. 4), particularly surface area and microporosity. The study concluded that micropore volume is a critical determinant of CO₂ uptake, highlighting the role of pore architecture in enhancing gas storage performance.⁶⁵ Subsequent advancements have focused on functional modifications to improve selectivity and adsorption efficiency. In 2015, Q. Gao *et al.* synthesized a nitrogen-rich imine-linked COF using 1,3,5-triformylbenzene and 2,4,6-tris(4-aminophenyl)-1,3,5-triazine under solvothermal conditions. The high nitrogen content in the framework enhanced CO₂ selectivity over CH₄ and N₂ due to favorable interactions with CO₂ molecules.⁸⁰ In 2016, L. Wang *et al.* incorporated thiadiazole units into an imine-linked COF through Schiff-base condensation. Thiadiazole, a dipolar heterocycle, offers strong dipole–quad-

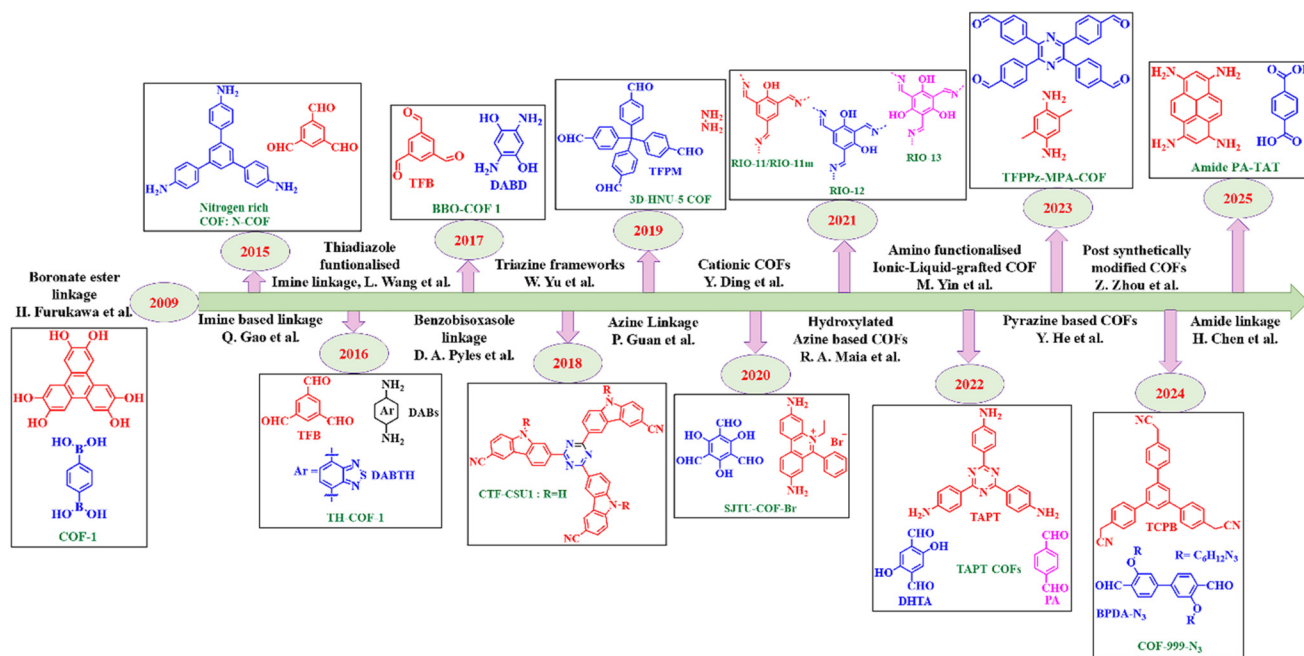


Fig. 4 Progression of Covalent Organic Frameworks (COFs) with various monomers for CO₂ capture over the past decade. This chart illustrates the evolution and advancements in COF design, highlighting the diversity of materials used in CO₂ sequestration.

rupole interactions with CO₂, whose quadrupolar nature results in enhanced adsorption. Additionally, the functional group induced ultra-microporosity (<0.7 nm), facilitating efficient pore-filling at low pressures.⁸¹ In 2017, D. A. Pyles *et al.* reported the synthesis of benzobisoxazole-linked COFs (BBO-COFs) using C₂-symmetric *o*-aminophenol and C₃-symmetric formyl building blocks, catalyzed by cyanide ions. These COFs demonstrated not only substantial CO₂ uptake but also excellent water stability, marking the first successful synthesis of ordered BBO-based COFs.⁸² The following year, W. Yu *et al.* developed mesoporous COFs (CTF-CSUs) *via* ionothermal polymerization of triazine- and carbazole-based monomers using ZnCl₂ as a catalyst. The presence of nitrogen-rich triazine and carbazole units significantly enhanced the CO₂ capture capacity of the frameworks.⁸³

In recent years, the development of three-dimensional (3D) Covalent Organic Frameworks (COFs) has gained significant attention due to their unique structural properties and promising applications in areas such as CO₂ capture and gas storage.⁸⁴ Unlike traditional two-dimensional (2D) COFs, 3D COFs exhibit interconnected networks that enhance their stability, adsorption capacity, and exceptional surface area.⁸⁴ Recently, H. M. El-Kaderi *et al.* studies have introduced new 3D COFs, such as COF-102, COF-103, COF-105, and COF-108, which are synthesized through the self-condensation of boronic acids to form boroxine rings and co-condensation with catechol to produce boronate ester rings.⁸⁵ These frameworks represent a significant advancement in the design of highly functional materials for industrial-scale applications. In 2019, P. Guan *et al.* synthesized a novel 3D azine-linked COF

(3D-HNU5) with a two-fold interpenetrated diamond topology *via* room-temperature condensation of tetrakis(4-formylphenyl)methane and hydrazine. Recyclability studies confirmed the material's stability and consistent CO₂ adsorption capacity over multiple cycles, supporting its potential for practical deployment.⁸⁶ Y. Ding *et al.* explored the effect of counter anions on CO₂ adsorption in cationic COFs for the first time in 2020. They synthesized a series of anion-exchanged COFs (SJTU-COF-Br, -Cl, -AcO, -CF₃SO₃) *via* microwave-assisted solvothermal methods. Among these, SJTU-COF-AcO exhibited the highest CO₂ uptake, attributed to the interaction between CO₂ and the acetate anion. This interaction, characterized as mild charge transfer between the carbon atom of CO₂ (a Lewis acid) and the oxygen atom of the carboxylate group (a Lewis base), demonstrates the potential of anion exchange as a strategy to tailor COF adsorption properties.⁸⁷ Next year, R. A. Maia *et al.* synthesized hydroxylated azine-based COFs (RIO-11, RIO-11m, RIO-12, RIO-13) *via* the condensation of hydrazine hydrate with 1,3,5-triformylbenzene. Their study focused on understanding the structure–property relationships governing CO₂ adsorption by correlating surface chemistry and textural features with gas uptake performance.⁸⁸

Targeted functionalization continues to drive advances in COF design. In 2022, M. Yin *et al.* fabricated imine-linked triazine COFs functionalized with amino-terminated ionic liquids. These modified materials demonstrated enhanced CO₂ capture due to strong electrostatic, acid–base, and van der Waals interactions between CO₂ and the functional groups. Additionally, the nucleophilic halide groups in the ionic liquids facilitated chemical conversion of captured CO₂, indi-

cating dual functionality.⁸⁹ In 2023, Y. He *et al.* developed pyrazine-based COFs (TFPPz-MPA-COF and TFPPz-BD(OMe)₂-COF) through the condensation of tetra(4-formylphenyl)pyrazine with respective diamines. These frameworks exhibited significant potential for both CO₂ and iodine adsorption, providing a versatile platform for multifunctional separation applications.⁹⁰ Olefin-linked COFs have recently emerged as highly promising materials for direct air capture due to their exceptional chemical stability, high CO₂ capacity, and adaptability for functionalization. Recently, Z. Zhou *et al.* reported the synthesis of COF-999-N₃, an olefin-linked precursor COF that was further modified through reduction and aziridine functionalization to yield COF-999. Outdoor testing in Berkeley, California over 100 adsorption–desorption cycles demonstrated complete retention of performance, underscoring the robustness of these frameworks.⁹¹

Additionally, linkage engineering presents a novel route to enhance COF performance. H. Chen *et al.* developed a method to convert imine-linked COFs into amide-linked frameworks. The imine-PA-TAT COF, synthesized from 4,4',4''-(1,3,5-triazine-2,4,6-triyl)trianiline and 1,4-phthalaldehyde, was oxidized to form amide-PA-TAT. Mechanistic studies revealed that the carbonyl groups in amide linkages create strong electrostatic fields, resulting in a fourfold increase in CO₂ adsorption relative to imine linkages due to stronger interaction with the CO₂

molecule.⁹² Collectively, these studies demonstrate the rapid evolution of imine-linked and functionalized COFs as viable materials for CO₂ capture. Through structural innovation, functional group incorporation, and linkage transformation, COFs continue to expand their role as high-performance adsorbents in gas separation technologies.

3. Architectural design

Adsorption is a key process for CO₂ capture and plays a significant role in many emerging technologies aimed at reducing carbon emissions. This process involves the adsorption of carbon dioxide molecules onto adsorbent surfaces through weak, non-covalent interactions, including van der Waals forces and electrostatic interactions between the quadrupole moment of CO₂ and the dipole moments within the porous frameworks.^{93,94} There are various adsorption-based methods for CO₂ capture, with material selection and techniques being influenced by factors such as cost, effectiveness, and specific application requirements.⁹⁵ Adsorption can either be physical (physisorption) or chemical (chemisorption), depending on the nature of the interactions between CO₂ and the adsorbent, illustrated in Fig. 5. Carbon dioxide adsorption using physical adsorbents is driven by weak van der Waals and electrostatic

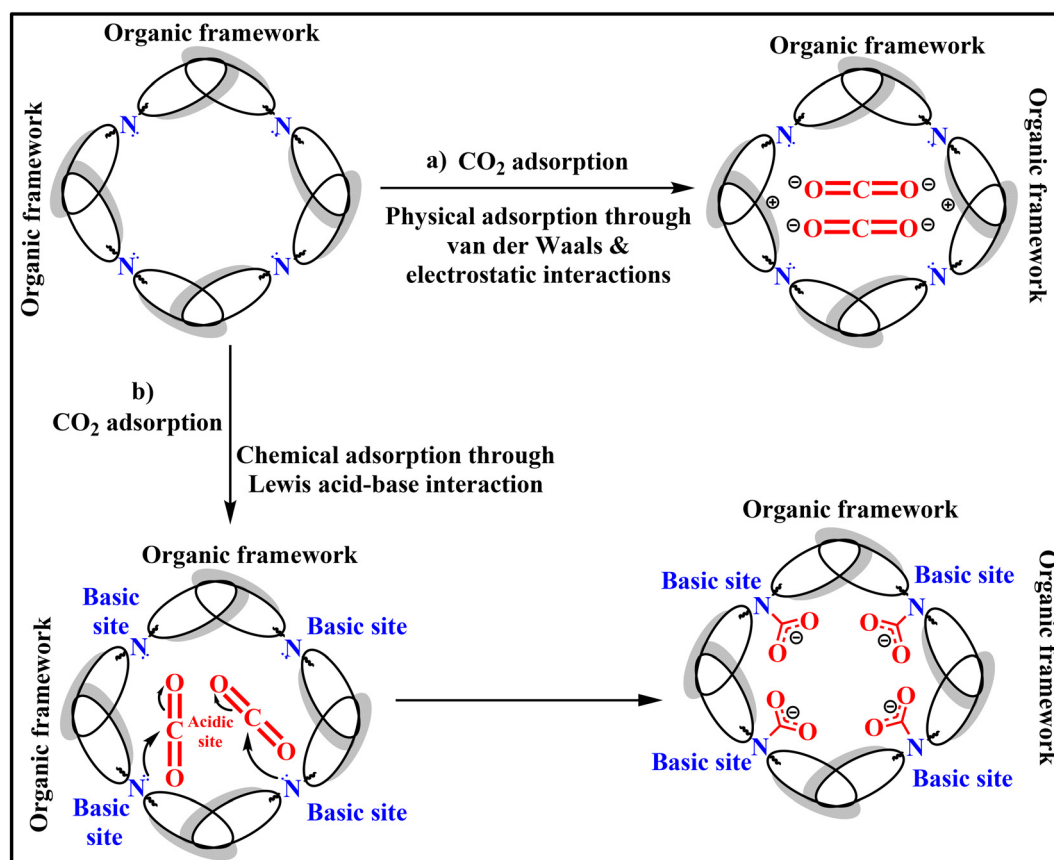


Fig. 5 Schematic representation of the mechanisms involved (a) physical adsorption and (b) chemical adsorption in CO₂ capture.

interactions, which require less energy compared to chemical adsorbents.⁹⁶ This is because no new bonds are formed between CO₂ and the adsorbent surface, leading to a lower energy penalty during CO₂ regeneration. The reaction mechanism in chemical adsorption is driven by the Lewis acid–base interaction between CO₂ and nitrogen-rich groups, where the nitrogen-rich group acts as a base and CO₂ as an acidic gas, creating a strong attraction between the two.⁹⁷ On the other hand, chemical adsorption involves the formation of stronger bonds between CO₂ and the adsorbent, resulting in a higher CO₂ uptake capacity compared to physical adsorption. This makes chemical adsorption especially useful in post-combustion capture, pre-combustion capture, and direct air capture (DAC) technologies.⁹⁸ While physical adsorption of CO₂ on COFs is considered more energy-efficient due to the ease of desorption, chemisorption offers significant benefits in enhancing adsorption capacity, particularly at low pressures.⁹⁸

Both mechanisms are governed by the diffusion of CO₂ into porous structures with pores tailored to CO₂ size, followed by interaction and, ultimately, reversible CO₂ release, contingent on the external conditions.⁹⁹ To effectively capture and separate CO₂ from air or flue gas streams using nanoporous adsorbents, it is crucial to account for the impact of humidity. H. Veldhuizen *et al.* explored the effectiveness of a polyimide COF (TAPB-NDA-COF) for CO₂ separation from humid CO₂/N₂ streams, given that water vapour, common in flue gas, can significantly reduce the adsorbent's efficiency by competing with CO₂ for adsorption sites.¹⁰⁰ The TAPB-NDA-COF, which has substantial supermicropore and mesopore volumes, appears advantageous for enhancing water-assisted CO₂ adsorption. Additionally, the abundance of aromatic planes in the COF may facilitate the formation of aromatic motifs that preferentially adsorb CO₂ over water.¹⁰¹ Moreover, the kinetic diameter of CO₂ is approximately 3.30 Å (0.33 nm), closely comparable to that of N₂, which is 3.64 Å (0.36 nm).¹⁰² This similarity in molecular size highlights the importance of selective sorbent design for effective CO₂ separation, which is influenced by both textural and geometric properties. Key textural properties include thermal conductivity and heat capacity, while geometric considerations involve pore architecture and material morphology.¹⁰³ An essential thermodynamic parameter for evaluating adsorbents is the isosteric heat of adsorption (Q_{st}). For optimal CO₂ capture performance, Q_{st} should ideally fall within the range of –20 to –50 kJ mol^{–1}, balancing adsorption strength and energy efficiency. Values more negative than –50 kJ mol^{–1} suggest chemisorption, while less negative values typically indicate physisorption-dominated interactions.¹⁰⁴ In a notable study, N. Huang *et al.* synthesized positively charged imine-linked COFs incorporating 6-bis(4-formylbenzyl)-1,3-dimethyl-benzimidazolium bromide (BFBIm) as a cationic building unit and 4,4',4'',4'''-(pyrene-1,3,6,8-tetrayl)tetraaniline (PyTTA) as a neutral node. The resulting PyTTA-BFBIm-iCOF exhibited an excellent CO₂ uptake capacity of 177 mg g^{–1} at 273 K and 1 bar, with a Q_{st} of –30.2 kJ mol^{–1}, indicating efficient physisorptive interaction.¹⁰⁵ C. Ji *et al.* reported the synthesis of 3D cage-based COFs utilizing a linear

ditopic ligand as a 2-connected linker and an amino-functionalized D_{3h} -symmetric organic cage as a 6-connected node. Structural distortion and the presence of polar functional groups led to variations in aperture sizes; however, all resulting 3D-OC-COFs demonstrated high thermal and chemical stability along with permanent porosity. Among these, 3D-OC-COF-OH displayed a CO₂ uptake of 174.83 mg g^{–1} and a Q_{st} of 22.4 kJ mol^{–1} under standard conditions (273 K, 1 bar).¹⁰⁶ Thermal conductivity in porous materials is inversely related to pore size, as increased porosity introduces more voids filled with air or vacuum, reducing the material's ability to conduct heat.¹⁰⁷ Furthermore, the development of polymeric frameworks with ultra-micropores (<0.7 nm) has been shown to enhance CO₂ capture, with microporosity contributing linearly to total pore volume and, consequently, gas uptake capacity.¹⁰⁸

In the context of COF design, the selection of topologies, linkages, and monomer geometries plays a fundamental role in defining the material's structural and functional properties. Nanoarchitectonics, the design and manipulation of materials at the nanoscale to achieve specific structural and functional outcomes, plays a pivotal role in optimizing these parameters.¹⁰⁹ In COFs, nanoarchitectonics refers to the assembly of nanoscale components such as organic linkers, functional groups, *etc.*, into ordered porous crystalline structures with customized properties suited for applications like gas storage, catalysis and sensing.¹¹⁰ Monomer symmetries – such as C_1 , C_2 , C_3 , C_4 , C_6 , and T_d – are predictive of the resulting COF topology, which directly impacts CO₂ capture efficiency. For example, T. Banerjee *et al.* synthesized a series of imine-linked [4 + 3] 2D COFs using tetra- and tritopic linkers, achieving a **bex** net topology and a CO₂ uptake of 127 mg g^{–1} at 273 K and 1 bar.¹¹¹ Y. Meng *et al.* developed porphyrin-based COFs from [$C_4 + C_2$] symmetric linkers for 2D structures, and from C_4 and T_d symmetric monomers for 3D frameworks with **pts** topology. The resulting 3D **pts** COFs displayed significantly higher CO₂ uptake (205 mg g^{–1} at 273 K and 1 bar), approximately 3.1 times greater than their 2D counterparts, owing to their narrower pore size distribution, higher free volume, and reduced density.¹¹² Other notable examples of 3D COFs include an azine-linked framework with a **dia** topology that showed a CO₂ uptake of 123.1 mg g^{–1} under the same conditions.⁸⁶ Additional topologies reported for imine-linked COFs with promising CO₂ capture performance include **ljh**,¹¹³ **lon**¹¹⁴ (featuring 4-connected nodes); **tbo**,¹¹⁵ **fjh**,¹¹⁶ **stp**,¹¹⁷ **hea**,¹¹⁸ and **she**¹¹⁹ (based on 6-connected nodes); and **scu**,¹²⁰ **flu**¹²¹ (with 8-connected nodes). These studies collectively demonstrate that the precise engineering of COF topology, monomer symmetry, and pore architecture can significantly enhance their applicability for selective and efficient CO₂ capture.

4. Structural linkages in COFs

The term “linkage” in covalent organic frameworks refers to the specific covalent bonds that connect organic building

blocks (monomers) within their two- or three-dimensional architectures. These linkages are central to the exceptional versatility of COFs, as they allow for the precise modulation of properties such as porosity, thermal and chemical stability, electrical conductivity, and catalytic functionality.⁸⁵ The selection of linkage chemistry in covalent organic frameworks is primarily guided by the target application, which may include gas storage and separation, catalysis, or energy storage. A diverse range of covalent linkages has been explored in COF synthesis, including C–C, C–N, B–N, B–O, C–S, C=N (e.g., imine), and P–O/P–N linkages. The chemical nature of these linkages significantly influences the framework's stability, porosity, and interaction with guest molecules. H. Furukawa *et al.* investigated the CO₂ adsorption capacities of COF-1 and COF-102, both featuring a boroxine linkage, at low (1 bar) and high pressures (35 and 55 bar), finding that at 273 K and 1 bar, COF-1 and COF-102 exhibited CO₂ uptakes of approximately 98 mg g⁻¹ and 68.6 mg g⁻¹, respectively.⁶⁵ Z. Kahveci *et al.* synthesized a 2D mesoporous COF called TDCOF-5, formed from 1,4-benzenediboronic acid and hexahydroxytripitycene, which demonstrated a CO₂ uptake of 92.12 mg g⁻¹ at 273 K and 1 bar.¹²² However, C. Jia *et al.* reported the CO₂ uptake capacities of COF-1^{45,123} and tetraethylenepentaamine (TEPA)-modified COFs, TEPA(*n*)COF-1 having boroxine linkage, with *n* = 5 and *n* = 10, which were 78.79, 97.61, and 105.84 mg g⁻¹, respectively. The modification of COF-1 with oligoamine (TEPA) introduces additional interaction sites for CO₂, owing to the abundance of amine groups, resulting in a significant increase in CO₂ adsorption.¹¹⁸ Nitrogen-rich groups provide active sites that enhance CO₂ adsorption by promoting interactions through electrostatic forces and hydrogen bonding,

distinguishing them from other linkages, such as boron-containing groups, in COF materials designed for CO₂ capture.

B. P. Biswal *et al.* developed a β-ketoenamine-linked COF (TpBD) *via* a [3 + 2] reaction between 1,3,5-triformylphloroglucinol (Tp) and benzidine (BD), which demonstrated a CO₂ adsorption capacity of 85 mg g⁻¹ at 273 K.¹²⁴ Building on this, S. Chandra *et al.* synthesized modified β-ketoenamine-linked COFs incorporating various electron-donating and electron-withdrawing groups to improve CO₂ capture. Among these, nitro-substituted COFs [TpBD-NO₂ and TpBD-(NO₂)₂] showed enhanced CO₂ uptake capacities of 124 and 103 mg g⁻¹ at 273 K, respectively. In contrast, the fluoro-substituted COF (TpBD-F₄) showed a lower uptake of 69 mg g⁻¹. COFs modified with methyl and methoxy groups [TpBD-Me₂ and TpBD-(OMe)₂] had CO₂ adsorption capacities of 73 and 53 mg g⁻¹, respectively, at the same temperature.¹²⁵ A. F. El-Mahdy *et al.* synthesized β-ketoenamine-linked COFs (TFP-TPA, TFP-Car, TFP-TPP) *via* a one-pot [3 + 3] polycondensation reaction between 1,3,5-triformylphloroglucinol and tris(aminophenyl) derivatives, and they achieved higher CO₂ uptakes of 183, 190, and 200 mg g⁻¹, respectively, at 273 K and 1 bar.¹²⁶ D. Kaleeswaran *et al.* synthesized imine and β-ketoenamine tethered COFs by the condensation between 1,3,5-tris(40-aminophenyl)benzene (TAPB) and 1,3,5-tris(40-amino-30,50-isopropylphenyl)benzene (iPrTAPB) with 1,3,5-tris(40-formylphenyl)benzene (TFPB) and 1,3,5-triformylphloroglucinol (TFP) resulting crystalline 2D frameworks, TAPB-TFPB, iPrTAPB-TFPB, TAPB-TFP and iPrTAPB-TFP having CO₂ uptake of 40.1, 31.2, 180, 105.2 mg g⁻¹ respectively.¹²⁷ β-Ketoenamine linkages provide more effective CO₂ capture than imine linkages due to their stronger CO₂ interactions. However, their

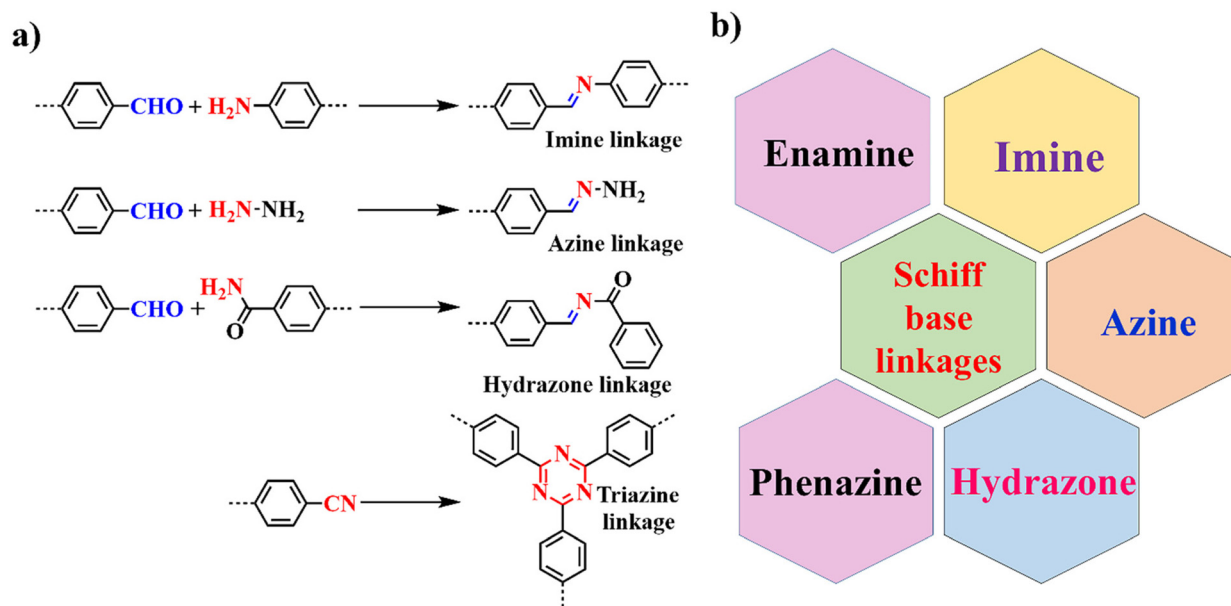


Fig. 6 (a) Schematic representation of -C=N- bonding interactions in COFs discussed in this class of materials. (b) The diagram illustrates the different bonding interactions constructed *via* Schiff base chemistry that contribute to the structure of COFs.

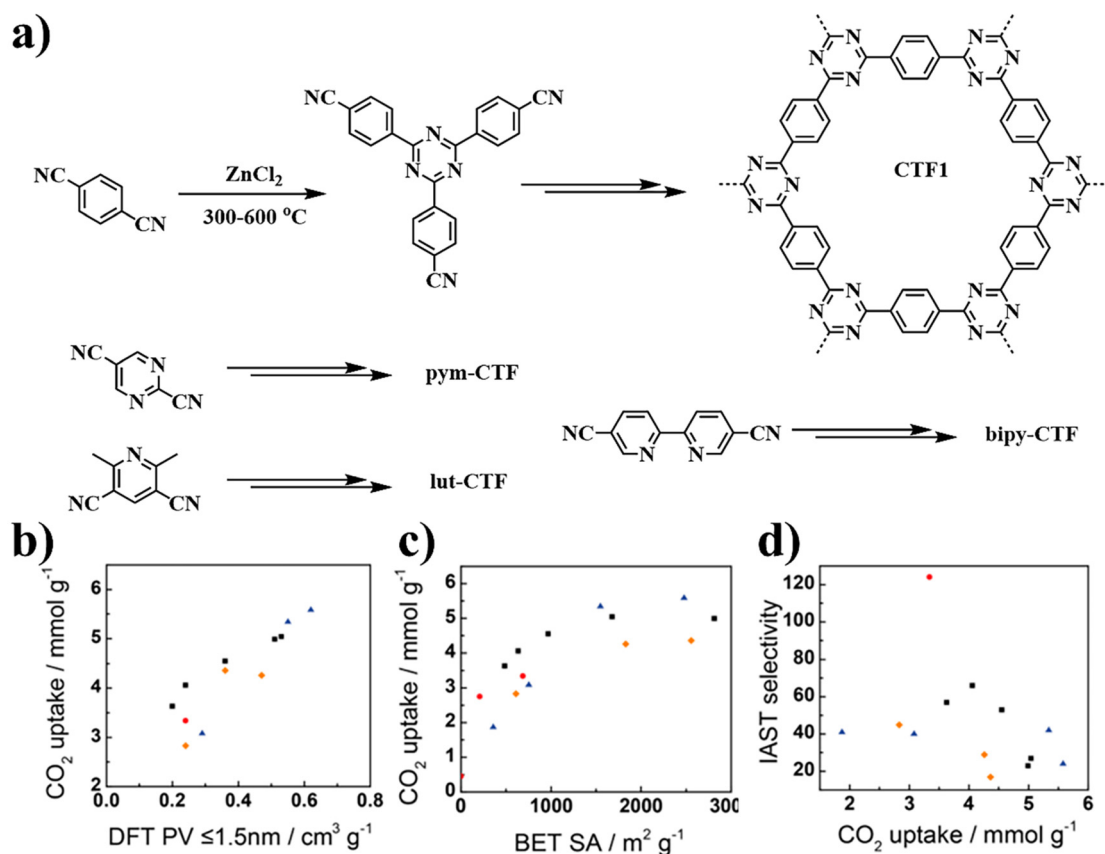


Fig. 7 (a) Synthetic scheme of the CTFs: CTF1, pym-CTF, lut-CTF, and bipy-CTF. (b) CO_2 uptake on the micropore volume (≤ 1.5 nm). (c) BET SA and (d) IAST selectivity on the CO_2 uptake. lut-CTFs (black squares), pym-CTFs (red circles), bipy-CTFs (blue triangles), and CTF1s (orange diamonds). Reproduced from ref. 129 with permission from the American Chemical Society, Copyright 2015.

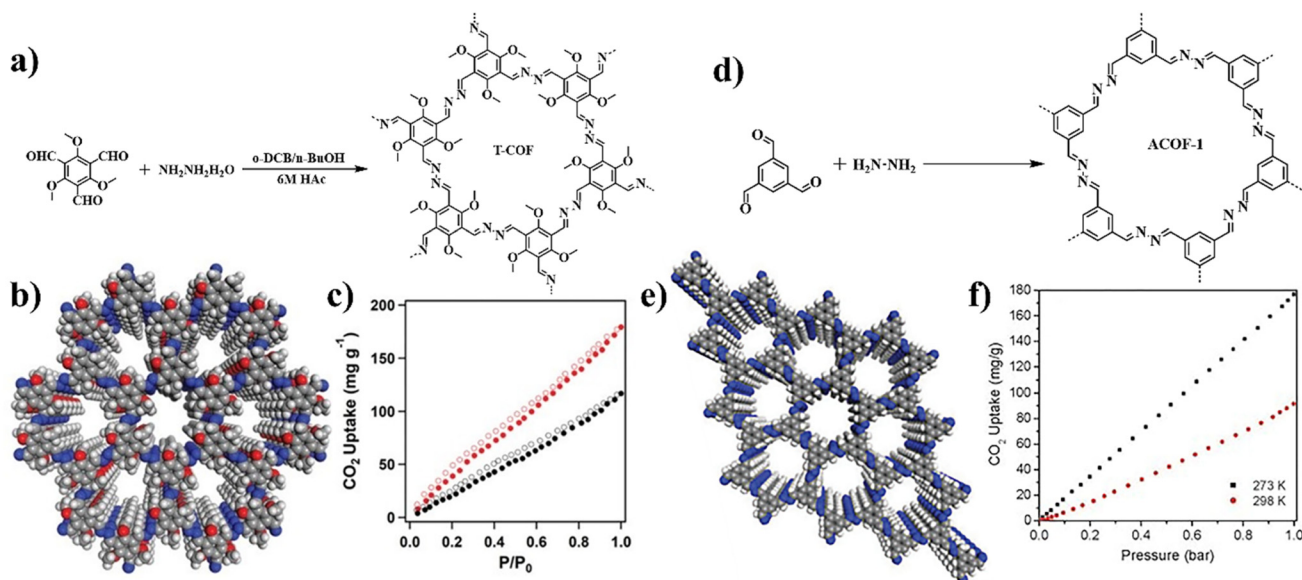


Fig. 8 (a) Schiff base reaction scheme of T-COF. (b) Crystal structure of T-COF with AA stacking model. (c) CO_2 isotherms at 273 K and 298 K of T-COF. Reproduced from ref. 131 with permission from the Royal Society of Chemistry, Copyright 2022. (d) Synthetic scheme of ACOF-1. (e) Top view of AA stacking structure of ACOF-1. (f) CO_2 isotherms of ACOF-1 at 273 K and 298 K. Reproduced from ref. 132 with permission from the Royal Society of Chemistry, Copyright 2014.

synthesis often requires specific monomers and conditions, such as tautomerization, making them less commonly used compared to other C=N linkages.¹²⁷

For CO₂ capture, functionalization of the COF backbone with electron-rich or basic groups such as triazine, imide, tetrazole, and amine moieties has proven particularly effective. These functional groups, due to their high polarizability and strong quadrupole interactions with CO₂, enhance the CO₂-philic nature of the materials. For that, Schiff base chemistry has been widely employed for constructing COFs, especially those involving C=N and C-N linkages. Common examples include imine, azine, and hydrazone linkages,¹²⁸ as well as triazine-based frameworks,¹²⁴ as illustrated in Fig. 6. Among these, this review places emphasis on COFs containing C-N linkages, owing to the favorable chemical stability and tunable interaction dynamics afforded by the carbon-nitrogen bond network. These features make C=N linked COFs particularly promising candidates for CO₂ capture and related gas separation applications.

S. Hug *et al.* have developed a series of efficient physisorbent materials for gas adsorption and storage using triazine-based frameworks. Covalent Triazine Frameworks (CTFs) were synthesized *via* an isothermal polymerization approach incorporating various functional units such as lutidine, bipyridine,

pyrimidine, and phenyl groups shown in Fig. 7. Their findings indicate that while nitrogen content in the CTFs contributes marginally to CO₂ adsorption through weak interactions, the predominant factor responsible for high uptake is the presence of ultramicropores. These pores not only enhance gas sorption by providing high surface area but also increase the accessibility of nitrogen sites, facilitating interactions through Lewis acid-base mechanisms. The CO₂ interaction energy is thus dependent on both the quantity and nature of nitrogen functionalities present in the framework.¹²⁹ In a complementary study, J. Du *et al.* reported the synthesis of benzodiazole-functionalized CTFs from a dicyano-substituted benzodiazole monomer. This material exhibited a notable CO₂ uptake of 89.2 cm³ g⁻¹ (174.83 mg g⁻¹) at 273 K and 1 bar, with isosteric heats of adsorption reaching up to 52 kJ mol⁻¹, highlighting the combined contributions of microporosity and nitrogen enrichment in enhancing adsorption capacity.¹³⁰

Beyond triazine-based frameworks, Schiff base linkages, particularly azine bonds, have shown considerable promise for CO₂ capture. Y. Zhang *et al.* synthesized an azine-linked covalent organic framework (T-COF) through the condensation of 2,4,6-trimethoxybenzene-1,3,5-tricarbaldehyde and hydrazine demonstrated in Fig. 8a. The T-COF exhibited a high CO₂ uptake of 178 mg g⁻¹ at 273 K, attributed to its nitrogen- and

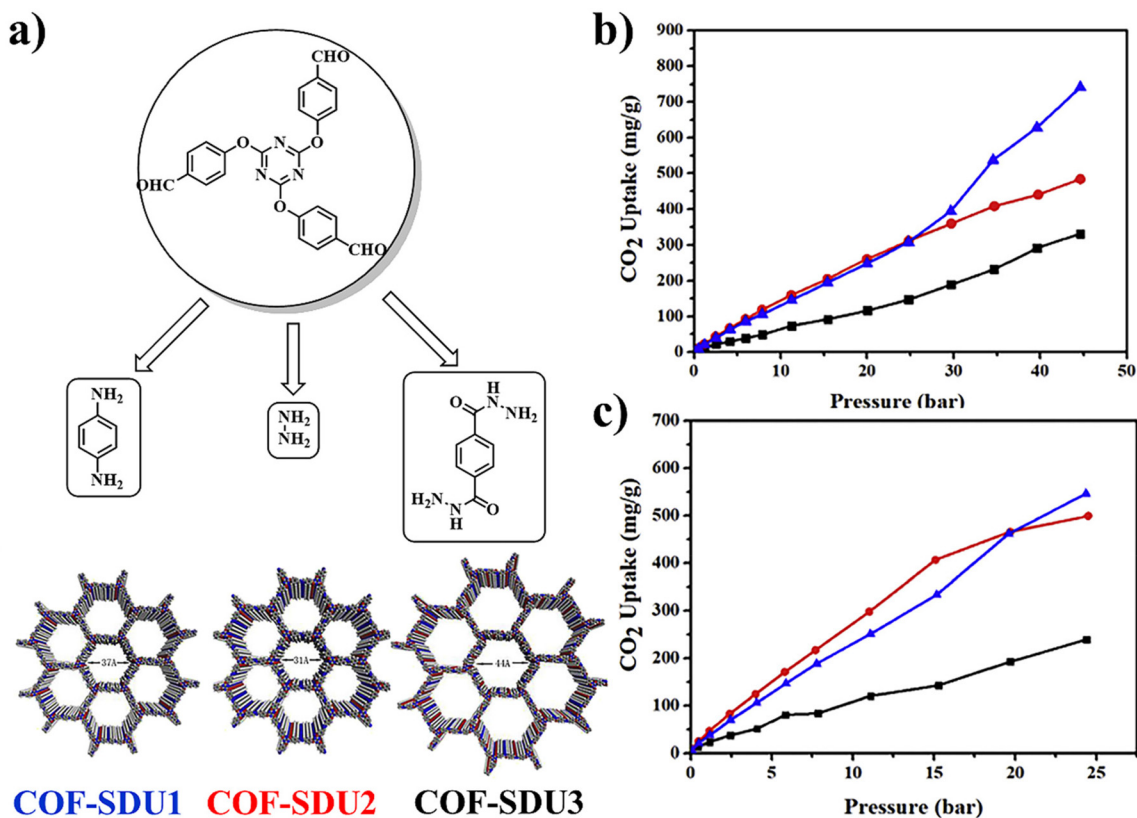


Fig. 9 (a) Illustration of different bonding motifs in COFs with imine, azine, and hydrazone linkages. (b) CO₂ uptake of COFs, COF-SDU1 (blue triangle), COF-SDU2 (red cycle), COF-SDU3 (black square) at 298 K. (c) CO₂ uptake at 273 K. Reproduced from ref. 134 with permission from the Elsevier, Copyright 2019.

oxygen-rich skeleton and uniform pore size of approximately 0.82 nm, as determined by non-local density functional theory (NLDFT). These embedded polar functional units and microporous architecture synergistically promote high CO₂ affinity.¹³¹ Z. Li *et al.* further demonstrated the potential of azine-linked COFs by synthesizing ACOF-1 *via* solvothermal condensation of 1,3,5-triformylbenzene with hydrazine hydrate. ACOF-1 exhibited a high surface area (1318 m² g⁻¹) and a CO₂ uptake of 17.7 wt% at 273 K and 1 bar shows in Fig. 8f. The isosteric heat of adsorption (Q_{st}) was measured at 27.6 kJ mol⁻¹, surpassing that of many previously reported imine-linked COFs, organic cages, and diimide polymers. The enhanced gas uptake is attributed to the interplay between high surface area, optimal pore size, and the availability of nitrogen sites.¹³²

Hydrazone-linked COFs also present attractive characteristics for CO₂ capture, owing to their high nitrogen content, structural tunability, synthetic versatility, and robust thermal and chemical stability. N. Bagherian *et al.* synthesized a hydra-

zone-linked COF (TPT/OH COF) *via* solvothermal condensation of oxalyl dihydrazide and 2,4,6-tris(*p*-formylphenoxy)-1,3,5-triazine (TPT) illustrated in Fig. 9. This framework demonstrated a CO₂ uptake of 0.9 mmol g⁻¹, attributed to its extended π -conjugation, nitrogen-rich pore walls, high surface area, narrow pore size, and stable hydrazone linkages.¹³³ Additionally, M. Zhang *et al.* constructed three COFs using tri-(4-formylphenoxy)-1,3,5-triazine (TRIF) and various linkers, including *p*-phenylenediamine (imine linkage), hydrazine (azine linkage), and terephthalic dihydrazide (hydrazone linkage). Their study revealed that larger pore sizes favoured CO₂ uptake under high-pressure conditions (>25 bar), whereas BET surface area was the dominant factor under low-pressure conditions (<25 bar). Among the three, COF-SDU1 exhibited the highest CO₂ uptake (compared in Table 4), which was attributed to its high surface area, large pore volume, and the presence of both triazine rings and imine functionalities.¹³⁴ Imine-linked COFs offer the benefits of high surface area and easy modifiability, making them ideal

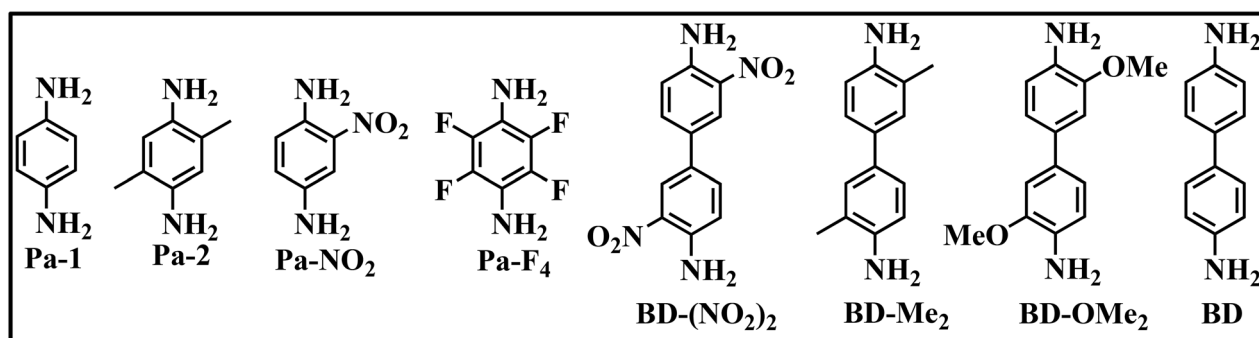


Fig. 10 Different functionalized amino monomers utilized by Chandra *et al.* for the condensation with 1,3,5-triformylphloroglucinol (Tp).¹²⁵

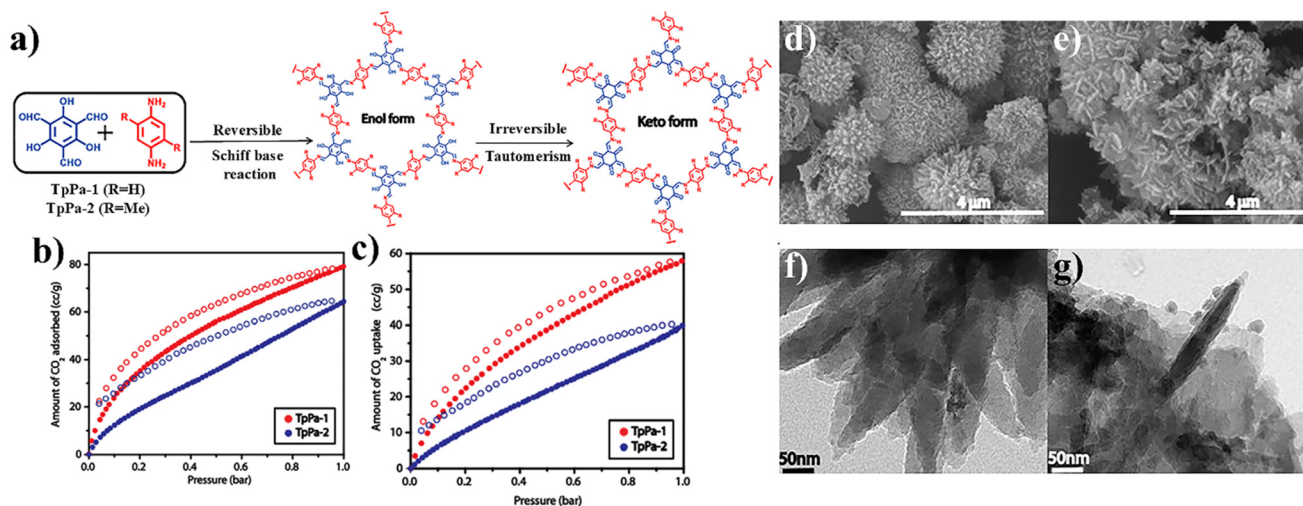


Fig. 11 (a) Synthetic scheme of the COFs, TpPa-1 & TpPa-2. (b) CO₂ uptake of TpPa-1 & TpPa-2 at 273 K. (c) CO₂ uptake of TpPa-1 & TpPa-2 at 298 K. (d and e) TEM images of TpPa-1. (f and g) TEM images of TpPa-2. Reproduced from ref. 141 with permission from the American Chemical Society, Copyright 2012.

for optimizing CO₂ adsorption by post-synthetic functionalization.

5. Schiff base imine linkage

Among the various linkages developed to date, imine linkages have emerged as particularly significant for applications in carbon dioxide (CO₂) capture and sequestration. This prominence is due to their synthetic accessibility, reversible formation *via* Schiff base condensation, tunable reactivity, and ability to support microporosity while accommodating diverse topologies. Generally, they demonstrate significant stability in a wide range of organic solvents and exhibit minimal sensitivity to aqueous conditions, as well as both acidic and basic environments.¹³⁵ Schiff base imine-linked COFs often rely on amines and aldehydes, which are relatively inexpensive but require high purity for high-performance frameworks.¹³⁶ An initial advancement in this area was reported by F. J. Uribe-Romo *et al.* in 2009, with the synthesis of COF-300, a three-dimensional COF featuring a five-fold interpenetrated diamondoid topology. This framework was constructed *via* the reversible condensation of aldehydes and amines to form imine bonds, demonstrating the combined application of reticular chemistry and dynamic covalent chemistry to produce structurally ordered and functionally robust materials.¹³⁷ Also, the ability to scale up and the cost-efficiency of Schiff base imine-linked COFs are the crucial factors that will determine their feasibility for large-scale industrial applications in CO₂ capture.¹³⁶ The synthesis of Schiff base imine-linked COFs often relies on solvent-based condensation (solvothermal method) reactions between amines and aldehydes, which can be challenging to scale efficiently. The combination of mesitylene,

dioxane, and acetic acid (AcOH) is commonly used as the most effective solvent mixture.¹³⁸ Other solvent systems, such as dioxane-aqueous acetic acid and *n*-butanol (*n*-BuOH)/*o*-dichlorobenzene (*o*-DCB)/3 M AcOH, have also been widely employed. AcOH, typically serving as the catalyst, plays a critical role in the formation of the imine linkage.¹³⁹ Alternatively, microwave synthesis and mechanochemical (MC) synthesis are widely explored in the synthesis of imine-based COFs.¹³⁶ B. P. Biswal *et al.* were the pioneers in employing mechanochemical techniques to efficiently synthesize two chemically stable COFs (TpPa-1 and TpPa-2) using a mortar and pestle system at room temperature. These COFs, synthesized *via* solvent-free mechanochemical methods, demonstrated moderate crystallinity, impressive stability, and high scalability.¹²⁴ Subsequently, H. Wei *et al.* employed a microwave-assisted solvothermal method to synthesize the 2D imine-linked COF (TpPa-COF) *via* a Schiff base reaction between 1,3,5-triformylphloroglucinol and *p*-phenylene diamine, achieving a higher yield in less time and TpPa-COF (MW) showing CO₂ uptake of 224.4 mg g⁻¹ at 273 K and 1 bar.¹⁴⁰ These methods eliminate the need for toxic solvents and facilitate large-scale production, potentially making them suitable for industrial applications. The scalability of imine-based COFs is also tied to the cost of synthesis. The economic viability of scaling up production will rely on factors such as the availability of affordable monomers, the chosen synthesis methods, and the energy demands associated with large-scale manufacturing.

5.1. Tunable Schiff base imine linkages in COFs: functionalization

The initial study of carbon dioxide (CO₂) adsorption by imine-linked covalent organic frameworks was reported in 2012 by

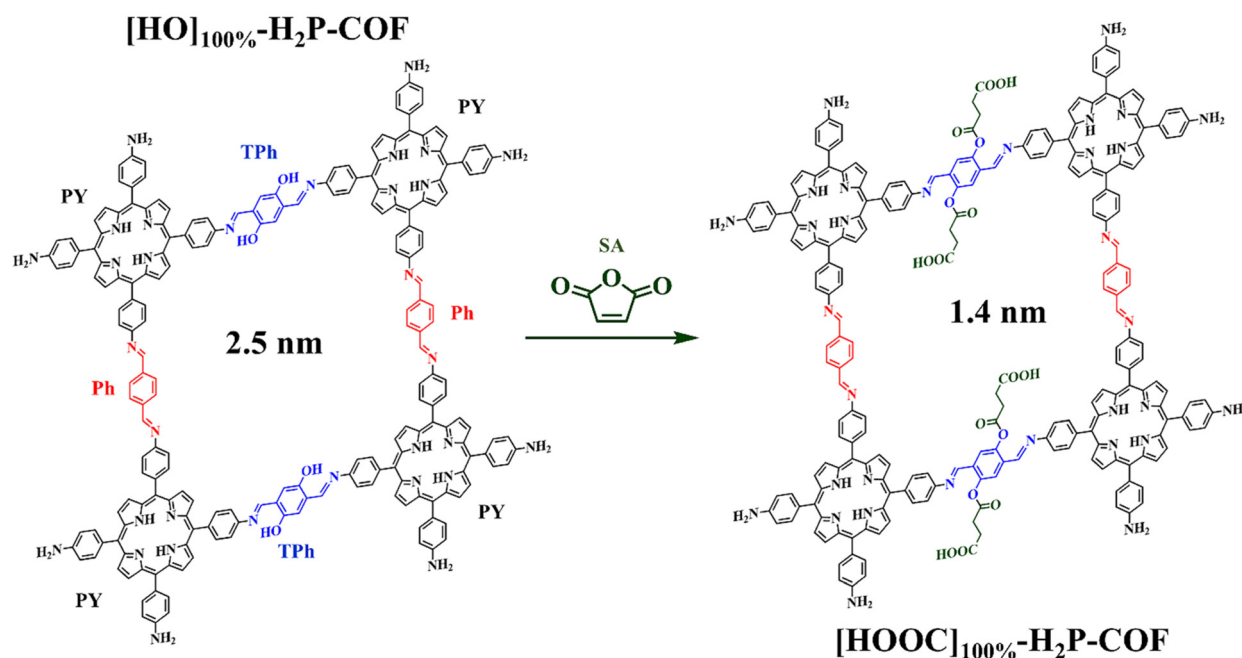


Fig. 12 Structural representation of [HO]_{100%}-H₂P-COF and [HOOC]_{100%}-H₂P-COF.¹⁴²

S. Kandambeth *et al.*, who synthesized two chemically robust two-dimensional COFs, TpPa-1 and TpPa-2, *via* reversible and irreversible Schiff base solvothermal reactions. These frameworks were obtained through the condensation of 1,3,5-triformylphloroglucinol (Tp) with *p*-phenylenediamine (Pa-1) and 2,5-dimethyl-*p*-phenylenediamine (Pa-2), respectively shows in Fig. 11a. The resulting materials exhibited CO₂ uptake capacities of 80 cm³ g⁻¹ and 63 cm³ g⁻¹ at 273 K and 1 bar, respectively. Both frameworks underwent irreversible tautomerism from the enol-imine to keto-enamine form, conferring excellent acid (9 N HCl) and thermal stability due to the absence of hydrolyzable imine linkages. Notably, both COFs demonstrated a characteristic flower-like morphology upon crystallization.¹⁴¹ Subsequently, S. Chandra *et al.* expanded this platform by synthesizing a series of functionalized imine-linked COFs, including TpPa-NO₂, TpPa-F₄, TpBD-(NO₂)₂, TpBD-Me₂, TpBD(OMe)₂, and TpBD, using various substituted diamine and dialdehyde monomers (Fig. 10). These COFs exhibited pore sizes ranging from 1.5 to 2.4 nm.¹²⁵

However, the densely packed, π -stacked architecture of 2D COFs can restrict their gas adsorption performance due to

limited accessible pore volume and surface area. To address this limitation, N. Huang *et al.* (2015) introduced wall-functionalized imine-linked COFs constructed from porphyrin units and phenol-containing aldehydes, facilitating post-synthetic modification of the pore walls with carboxylic acid groups *via* a ring-opening reaction with succinic anhydride (Fig. 12).¹⁴² By modulating the proportion of phenol groups through a three-component condensation strategy, the CO₂-philic functionality was precisely tuned. A 100% carboxylate-functionalized COF demonstrated an enhanced CO₂ uptake of 174 mg g⁻¹ at 273 K and 1 bar, despite a concurrent reduction in porosity.^{142,143}

The incorporation of triazine moieties into polymer frameworks has been widely reported to enhance their suitability for CO₂ adsorption and sensing applications.¹⁴⁴ In the same year, R. Gomes *et al.* reported TRITER-1, an imine-linked COF prepared *via* a one-pot Schiff base condensation of terephthalaldehyde with 1,3,5-tris(4-aminophenyl)triazine. This work also marked the first example of a superacid-catalyzed trimerization of 4-aminobenzonitrile to form the triazine core.¹⁴⁵ These nitrogen-rich frameworks, characterized by their mesoporous structure (~1.7 nm), large surface area (716 m² g⁻¹), and

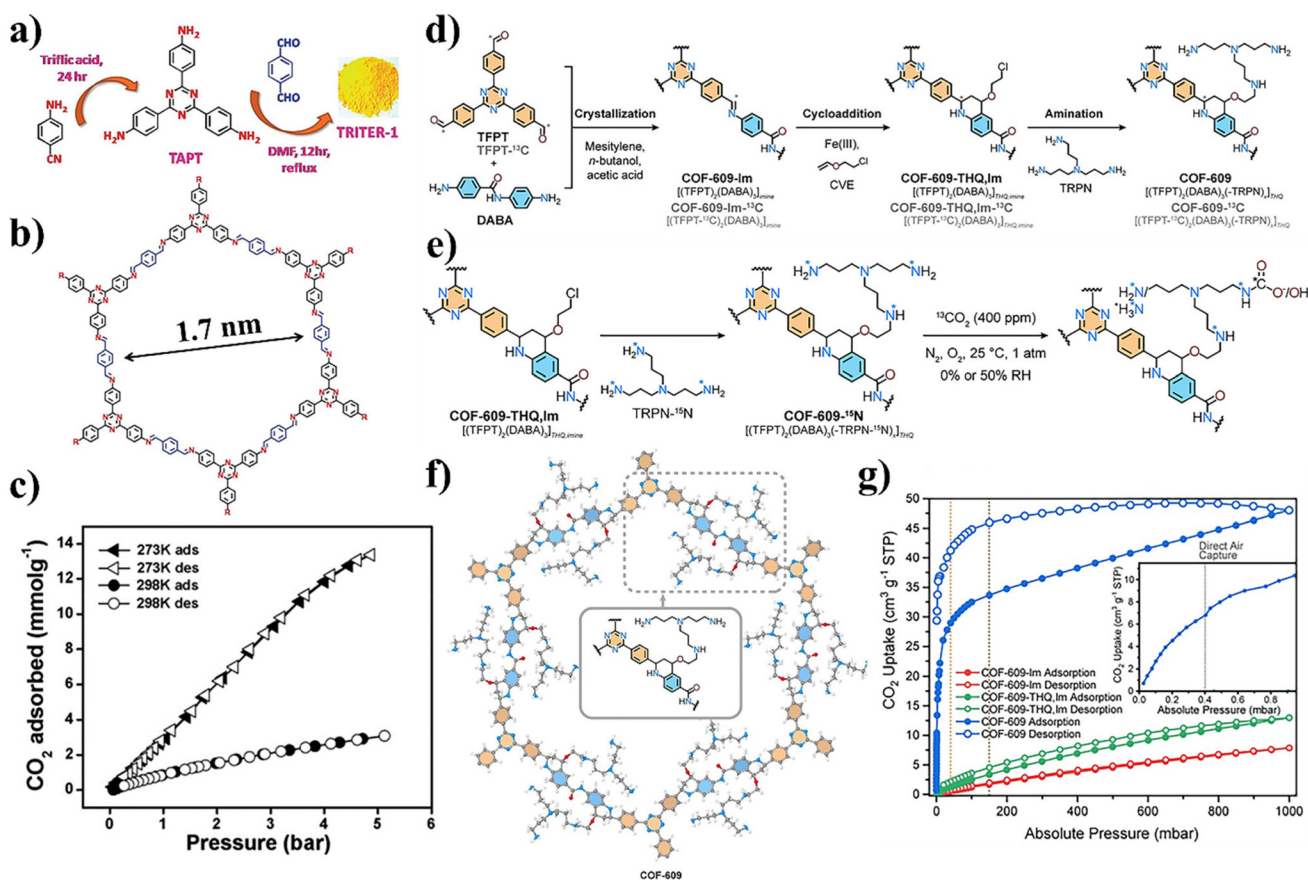


Fig. 13 (a) Schematic outline for the synthesis of COF, TRITER-1. (b) Designed structure of TRITER-1. (c) CO₂ isotherms of TRITER-1 at 273 K and 298 K. Reproduced from ref. 145 with permission from the American Chemical Society, Copyright 2015. (d) Synthetic pathway of COF-609 with labelled C13. (e) COF-609-¹⁵N synthetic scheme and its chemisorption with ¹³CO₂. (f) Proposed structure of COF-609. (g) CO₂ adsorption and desorption isotherm of COF-609-Im, COF-609-THQ,Im & COF-609. Reproduced from ref. 147 with permission from the American Chemical Society, Copyright 2022.

thermal stability, exhibited exceptional CO₂ adsorption capacities, up to 58.9 wt% at 273 K and 13.7 wt% at 298 K under 5 bar pressure illustrated in Fig. 13(a–c).¹⁴⁵ Similarly, S. Wu *et al.* synthesized imide-functionalized triazine COFs (TPIs@IC) through a two-step process, achieving a CO₂ uptake capacity of 3.2 mmol g⁻¹ at 273 K and 1 bar.¹⁴⁶

In a notable advancement, H. Lyu *et al.* developed aliphatic amine-linked COFs derived from imine precursors *via* a post-synthetic tetrahydroquinoline (THQ) linkage transformation. The resulting framework, COF-609, displayed strong chemisorption characteristics and significantly enhanced CO₂ uptake (6.8 cm³ g⁻¹ or 0.304 mmol g⁻¹ at 0.4 mbar), in contrast to its precursor (COF-609 THQ-Im), which exhibited only 0.005 cm³ g⁻¹ (0.00022 mmol g⁻¹) under identical conditions shown in Fig. 13(d–g). This 1360-fold improvement provides compelling evidence that incorporating aliphatic amines into COF structures enables efficient CO₂ capture under ambient conditions, supporting their potential for direct air capture and post-combustion applications.¹⁴⁷

5.2. Three-dimensional (3D) imine linkages: expanding the dimensionality of COFs

While the majority of covalent organic frameworks reported to date are two-dimensional (2D) layered materials featuring one-dimensional (1D) pore channels, the development of three-dimensional (3D) COFs has attracted growing interest due to their enhanced porosity and potential for gas adsorption.⁸⁴ These 2D structures typically rely on non-covalent interactions for layer stacking, whereas 3D COFs offer interconnected networks that may improve guest molecule accessibility and framework stability. Z. Li *et al.* successfully synthesized a 3D COF, termed 3D-ceq-COF, *via* solvothermal condensation between 1,3,5-tris(4-aminophenyl)triazine (TAPT) and 2,3,6,7,14,15-hexakis(4-formylphenyl)triptycene (HFPTP).¹⁴⁸ The incorporation of the triptycene scaffold facilitated the formation of a **ceq** topology and enhanced the exposure of aromatic units, promoting stronger interactions with CO₂ molecules. This framework demonstrated a CO₂ uptake of 179 mg

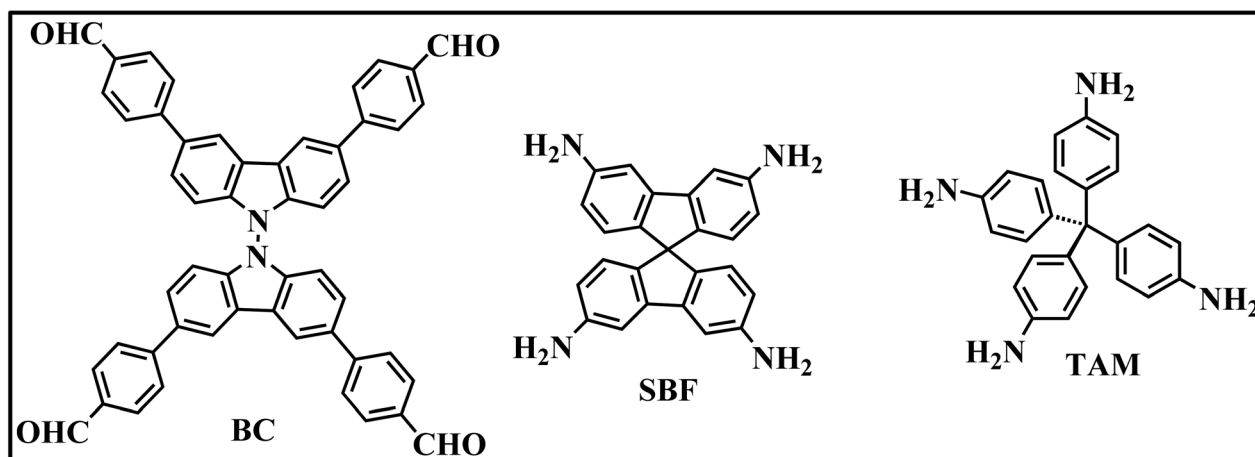


Fig. 14 Representative monomeric scaffolds utilized for the 3D polymeric architectures.

Table 3 Comparison of 2D and 3D COFs: advantages and limitations in CO₂ capture performance, stability, and synthesis complexity

Properties	3D-COFs	2D-COFs
Surface area and porosity	Higher surface area and porosity.	2D COFs have a smaller surface area than 3D COFs.
Stability	It offers superior stability, particularly in terms of thermal and chemical resistance, due to its interconnected framework.	2D COFs have less stability compared to 3D COFs due to their layered structure, which can delaminate under harsh conditions.
CO ₂ adsorption capacity	Well-defined and stable pores for selective CO ₂ capture.	Lower CO ₂ adsorption due to a less rigid structure.
Scalability	Difficult to scale up due to the limited availability of topology design strategies and building blocks.	Constructing layered structures in 2D COFs is challenging, but both diverse building blocks and topology designs provide potential solutions.
Cost	More expensive due to the need for precise monomer design and complex synthesis, which often involves advanced techniques like self-assembly or template-directed methods, adding to the overall cost.	Typically less expensive due to simpler synthesis and diverse building blocks, but costs can rise with scaling and maintaining uniformity.

Table 4 Reported characteristics and CO₂ capture performance of reviewed COF materials

COF materials [Ref.]	Linkage	Surface area (m ² g ⁻¹)	Pore size (nm)	Thermal stability	Chemical stability	CO ₂ uptake at 273 K & 1 bar (mg g ⁻¹)	CO ₂ uptake at 298 K & 1 bar (mg g ⁻¹)
PyTTA-BFBIm-iCOF ¹⁰⁵	Imine	1532	2.3	450 °C	Stable in water, DMF, HCl (3 M) and NaOH (3 M) for 1 day.	177	93
3D-OC-COF-OH ¹⁰⁶	Imine	923	0.65	430 °C	Stable in organic solvents and water.	174.83	100.94
PY ₂ B-COFs ¹⁰⁴	Imine	1984	1.83, 2.37	—	Unstable in HCl (1 M) and NaOH (1 M).	127	—
3D-PdPor-COF ¹⁰⁴	Imine	1406	0.58	500 °C	—	205.8	93.49
COF-1 ⁶⁵	Boroxine	750	0.9	500 °C	—	98	230 (55 bar)
COF-102 ⁶⁵	Boroxine	3620	1.2	400 °C	—	68.6	1200 (55 bar)
TD-COF-5 ¹²²	Boroxine	2497	2.6	420 °C	—	92.12	—
TpPa-1 ¹⁴¹	Imine	535	1.25	350 °C	Stable in water and HCl (9 M) for 7 days.	152.88	—
TpPa-2 ¹⁴¹	Imine	339	1.35	350 °C	Stable in water, HCl (9 M) and NaOH (9 M) for 7 days.	125.44	—
CTF1 ¹²⁹	Triazine	2557	—	—	—	191.84	97.24
pym-CTF ¹²⁹	Triazine	689	—	—	—	146.96	94.6
lut-CTF ¹²⁹	Triazine	2815	—	—	—	219.56	110.88
bipy-CTF ¹²⁹	Triazine	2479	—	—	—	245.52	129.8
T-COF ¹³¹	Azine	587	0.82	350 °C	Stable in water, DMF, HCl (1 M) and NaOH (1 M) for 12 hours.	178	117
ACOF-1 ¹³²	Azine	1318	0.94	300 °C	—	177	~92
TPT/OH COF ¹³³	Hydrazone	424	3.35	~300 °C	Stable in organic solvents and HCl (9 M) for 1 day.	—	39.6
COF-SDU1 ¹³⁴	Imine	1008	3.70	400 °C	—	547 ^a	741 ^a
COF-SDU2 ¹³⁴	Azine	1070	2.96	350 °C	—	484 ^a	500 ^a
COF-SDU3 ¹³⁴	Hydrazone	419	4.30	350 °C	—	331 ^a	239 ^a
[HO ₂ C] _{100%} -H ₂ P-COFs ¹⁴²	Imine	364	1.4	~300 °C	—	174	76
TRITER-1 ¹⁴⁵	Imine	716	1.7	400 °C	—	588.72 ^b	136.84 ^b
TPis@IC ¹⁴⁶	Imide	1053	0.54, 1.48	450 °C	Stable in organic solvents and dil. HCl (10 wt%) for 2 days.	141.68	92.84
3D-ceq-COF ¹⁴⁸	Imine	1148.6	—	550 °C	Stable in organic solvents and water for 1 day.	179	—
3D CageCOF-1 ¹⁴⁹	Imine	1040	—	400 °C	Stable in organic solvents and water for 1 day.	204	107
SP-CA-COF-IM ¹⁵¹	Imine	1551	0.95	~400 °C	—	110	48.4
SP-CA-COF-AM ¹⁵¹	Amine	1557	0.95	~400 °C	—	162.8	80.52
COF-300-SO ₃ H ¹⁵³	Imine	428.42	—	—	—	—	274.12

^a CO₂ uptake of COF-SDU1, COF-SDU2, and COF-SDU3 was measured at 273 K and 298 K under 45 bar and 25 bar pressures, respectively. ^b CO₂ uptake of TRITER-1 was measured at 273 K and 298 K under 5 bar pressure.

g⁻¹ at 273 K and 1 bar, highlighting the role of π - π interactions and molecular recognition sites in enhancing CO₂ adsorption. Despite this progress, expanding the library of 3D COFs remains challenging due to the scarcity of suitable 3D organic building blocks and the limited reversibility of the covalent bonds used in framework construction. Addressing this, Cooper *et al.* developed a 3D organic cage-based COF (3D-CageCOF-1) using a hydrothermal reaction between 4,6,16,18,31,33-hexamino-2,8,14,20,29,35-hexaoxabicyclo[3.3.3]octane-based calixarene (Cage-6-NH₂) and 2,5-dihydroxyterephthalaldehyde (DHTPA).¹⁴⁹ The resulting framework features a microporous environment enriched with oxygen and nitrogen functionalities, forming a polar and hydrophilic cavity ideal for CO₂ adsorption. 3D-CageCOF-1 exhibited an exceptional CO₂ uptake capacity of 204 mg g⁻¹ under the same conditions.

In a separate study, C. Gao *et al.* synthesized a series of isostructural 3D COFs with **pts** topology *via* [4 + 4] imine condensation using linkers bearing -H, -CH₃, or -F substituents.¹⁵⁰ Among these, the F-substituted COF displayed the highest CO₂ uptake, attributed to stronger binding interactions as indicated by a higher isosteric heat of adsorption ($Q_{st} = 28.4$ kJ mol⁻¹), despite comparable porosities and pore architectures. L. Zhang *et al.* introduced a rigid, imine-linked 3D COF (SP-CA-COF-IM), synthesized by the condensation of bicarbazole (BC) and spirobifluorene (SBF)-based orthogonal building blocks (Fig. 14).¹⁵¹ This framework, possessing a dia topology, restricted molecular rotation to produce a structurally brittle lattice. Post-synthetic modification using NaBH(OAc)₃ converted the imine linkages into secondary amines, yielding SP-CA-COF-AM. The incorporation of amine functionalities introduced chemisorptive sites, significantly enhancing

the selectivity and capacity for CO₂ capture compared to its imine-linked precursor.

F. Yuan *et al.* employed grand canonical Monte Carlo (GCMC) simulations and density functional theory (DFT) calculations to examine the influence of polar functional groups (-CH₂NH₂, -COOH, -SO₃H) on CO₂ uptake in 3D COFs.¹⁵² Functionalization led to elevated binding energies (19.4–35.1 kJ mol⁻¹) compared to the unfunctionalized frameworks (19.2 kJ mol⁻¹), thereby promoting improved adsorption performance. Similarly, S. Wei *et al.* synthesized a series of functionalized derivatives of COF-300-THF, derived from tetrakis(4-aminophenyl)methane (TAM) and terephthalaldehyde (BDA), modified with -OH, -NO₂, -SO₃H, and -NH₂ groups.¹⁵³ The CO₂ adsorption capacity at 298 K and 1 bar progressively increased from 57.2 mg g⁻¹ for COF-300-THF to 82.28 mg g⁻¹ (COF-300-NH₂), 91.08 mg g⁻¹ (COF-300-OH), 187 mg g⁻¹ (COF-300-NO₂), and peaked at 274.12 mg g⁻¹ for COF-300-SO₃H. These enhancements are attributed to stronger guest–host interactions facilitated by the introduced polar groups and the accompanying reduction in pore size. 3D COFs offer superior stability and higher adsorption capacity, making them ideal for demanding industrial applications.¹⁵⁴ However, they are more complex to synthesize and come at a higher cost. In contrast, 2D COFs are easier to synthesize and more cost-effective, but their lower stability and fragility can limit their long-term effectiveness in CO₂ capture and under harsh conditions, as illustrated in Table 3.¹³⁹ Table 4 summarizes the carbon dioxide uptake capacities of various COFs featuring -C-N- linkages, as discussed in this review.

6. Future outlook and conclusions

The Paris Agreement, a landmark global accord ratified by over 190 nations, aims to mitigate climate change by limiting the rise in global average temperatures to well below 2 °C above pre-industrial levels, with efforts directed toward a 1.5 °C threshold. Despite this widespread commitment, current global emissions trajectories remain misaligned with these temperature targets, underscoring the urgent need for advanced carbon capture technologies. In this context, covalent organic frameworks (COFs) have emerged as promising candidates due to their structural tunability, permanent porosity, and functional modularity. To meet both performance and scalability demands, it is imperative to develop adsorbent materials with superior CO₂ capture capabilities. The design of imine-linked 3D COFs represents a significant advancement in the field, effectively combining the chemical robustness and design flexibility of extended frameworks with the synthetic modularity of organic chemistry. However, several critical challenges remain that must be addressed to realize the full potential of these materials.

(1) The targeted construction of ultra-microporous 3D COFs – defined by pore sizes less than 0.7 nm – should be emphasized for their ability to enable high-density CO₂ packing, especially under elevated pressure conditions (>25 bar), aligning with industrial operational demands.¹⁵⁵

(2) There is a pressing need to explore Schiff base-derived 3D topologies that incorporate CO₂-philic moieties such as nitrogen-rich triazine and polar functional groups like -NH₂, COOH, OH, SO₃H. Modifying the electron density or steric hindrance at these linkage sites can tune the material's ability to

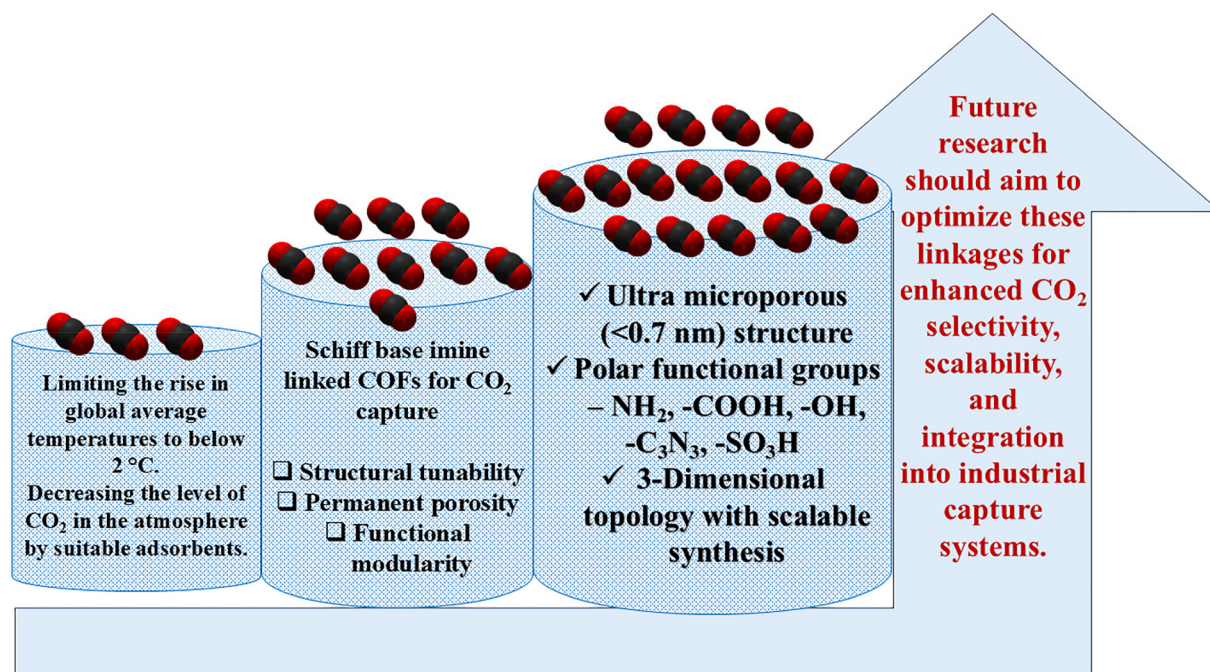


Fig. 15 Schematic diagram summarizing the challenges and solutions for effective CO₂ capture with emphasis on Schiff base imine-linked COFs.

selectively adsorb CO₂. These functionalities could enhance host-guest interactions through Lewis acid-base interactions and significantly improve CO₂ capture ability.

(3) Introducing non-covalent interactions strategies – such as dipole-dipole interactions, van der Waals forces (hydrogen bonding), and electrostatic attractions – offers a promising route to modulate the physicochemical characteristics of Schiff base imine-linked frameworks while preserving their crystallinity and robustness.

In conclusion, while COFs have already demonstrated utility across a range of sectors, further research is essential to address existing limitations. By integrating these design principles with linkage engineering and scalable synthesis techniques, we can enable the rational development of next-generation COFs optimized for efficient, selective, and regenerable CO₂ capture in industrial environments illustrated in Fig. 15. Furthermore, interdisciplinary collaboration, combining computational modeling with experimental synthesis, will be crucial for translating these laboratory findings into practical, real-world industrial applications. These approaches will be crucial for developing next-generation COF materials with enhanced CO₂ capture performance and long-term environmental sustainability. This mini-review underscores the potential of Schiff base imine-linked 3D COFs as a platform for the rational design of chemically and thermally stable, functional porous materials for greenhouse gas adsorption and beyond.

Conflicts of interest

There are no conflicts to declare.

Data availability

No new data were generated or analyzed as part of this review; all data are available in the appropriate references.

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