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Review

Polymeric and Crystalline Materials for Effective and Sustainable CO_2 Capture

David Gendron * and **Maria Zakharova**

Kemitek, Cégep de Thetford, 835 Rue Mooney, Thetford Mines, QC G6G 0A5, Canada; mzakharova@kemitek.org
* Correspondence: dgendron@kemitek.org

Abstract: Carbon dioxide (CO_2) is recognized as the primary cause of global warming due to its greenhouse potential. It plays a significant role in contributing to the emissions arising from a variety of anthropogenic activities, such as energy production, transportation, the construction industry, and other industrial processes. Capturing and utilizing CO_2 to mitigate its impact on the environment is, therefore, of significant importance. To do so, strategies such as net-zero strategies, deploying capture and storage technologies, and converting CO_2 into useful products have been proposed. In this review, we focused our attention on the preparation and performance of polymeric and crystalline materials for efficient CO_2 capture. More precisely, we examined MOFs, petroleum-based polymers (amine-based, polymeric ionic liquid, ionic polymer, conjugated macro/micro-cyclic polymer, and porous organic polymer) as well as bio-based polymers for CO_2 capture. In brief, the present work aims to guide the reader on the available crafted polymeric and crystalline materials offering a promising avenue towards innovative carbon dioxide capture strategy.

Keywords: polymer; carbon capture; carbon dioxide; biobased; MOFs; hybrids; composites; CO_2

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1. Introduction

Carbon dioxide (CO_2) is the ultimate byproduct arising from the combustion of carbonaceous matter and is characterized by exceptional stability [1]. In 2021, the annual CO_2 emission rate was approximately 34.9 Gt CO_2, primarily originating from the power industry (combustion of fossil fuel) as well as the steel and cement industries, which account for the majority of the global emissions [2]. These activities stand as the primary cause of the greenhouse effect, consequently contributing to global warming and sparking environmental concerns regarding mitigation efforts. Recently, the scientific community has increased its focus on capturing, utilizing, and studying CO_2 chemistry [3].

Foreseeing that carbon-based fossil fuels will maintain their significance as a key energy source for the next few decades, ongoing efforts are necessary to counter the escalating CO_2 emissions and their resulting impact on the greenhouse effect. Strategies like directly reducing emissions through the efficient use of energy, using carbon-free duels, deploying CO_2 capture and storage, and converting CO_2 into components for platform chemicals and fuels are among a few of the important solutions proposed [4–9].

Herein, our attention will be focused mainly on polymeric materials as an effective means to capture CO_2. We have explored the latest studies (2014–2024) with an important emphasis on the preparation (synthesis) of these polymers and their CO_2 uptake capacity as one figure of merit. It is important to point out that several reviews have been published on the topic of CO_2 absorption/capture/uptake based on polymeric materials. Therefore, the reader is referred to the works of Yuan, Gao, Han, Sattari, Song, Wang and others in which they reported an extensive list of polymer materials together with their corresponding performances [10–16].

The present review explores the fundamentals of CO_2 capture and summarizes the leading materials used in the process, both inorganic and organic. It examines, particularly, how they are made, contrasting petroleum-based with bio-based approaches. A range of petroleum-derived polymers, such as amine-based, polymeric ionic liquids, ionic polymers, conjugated macro/micro-cyclic polymers, and porous organic polymers are examined and compared with their bio-based counterparts. A section also examines the latest crystalline materials (i.e., MOFs) used for CO_2 capture. The review assesses the performance and sustainability of these materials to provide a comprehensive overview of the current landscape to consider when developing innovative and performing organic-based materials for CO_2 capture [17]. In brief, this review aims to inform the reader about which polymer structures to consider when developing innovative and well-performing materials for CO_2 capture.

2. Background

2.1. General Principals of Carbon Dioxide (CO_2) Capture and Sequestration

To mitigate CO_2 accumulation in the atmosphere, its capture and sequestration are among the suggested techniques. They can be divided into three main processes: (1) capture, (2) transport, and (3) sequestration. The capture step is the most costly process and thus poses a serious challenge to the reduction of its operational costs. Indeed, several studies have suggested that CO_2 capture alone can contribute up to 75% of the overall capture and sequestration cost [18]. In particular, this step involves capturing CO_2 from industrial process emissions before they are released into the atmosphere. Various technologies are employed for carbon capture, including post-combustion capture, pre-combustion capture, and oxyfuel combustion [19,20]. Among them, the most widely used is post-combustion capture due to its flexibility and ease of retrofitting to existing industrial processes, such as power plants and cement factories. It involves removing CO_2 from flue gases emitted during combustion processes using sorbents or solvents.

Pre-combustion capture involves capturing CO_2 from the gasified feedstock or fuel before combustion occurs. Oxy-fuel combustion involves burning fuel in a mixture of oxygen and recycled flue gas, resulting in a concentrated stream of CO_2 that is easier to capture. Another novel alternative, namely chemical looping combustion, recently attracted interest because the use of a solid oxygen carrier (typically a transition metal oxide) provides the oxygen needed for combustion. This technique helps to avoid diluting the combustion effluent stream with the N_2 released from the air and, therefore, minimizes the requirement of CO_2 separation, a major cost of CO_2 capture [21]. While post-combustion capture is currently the most used method, ongoing research and development efforts are focused on improving all four capture technologies to make them more cost-effective, energy-efficient, and applicable to a wider range of industrial processes.

As for sequestration, viable storage sites for technology encompass depleted gas fields, oceans, and saline aquifers [22]. However, it is important to point out that these locations require a geological stratum with a high impermeability to avert CO_2 leakage from the storage cluster [23]. Despite the significant progress in capture and sequestration, fundamental constraints endure. For instance, struggles with high energy consumption, suboptimal capture efficiency, and slow adsorption kinetics, the stability and longevity of storage sites, as well as safety concerns (i.e., leakage from storage sites) are the main barriers to overcome [4,24].

Once captured, the CO_2 can be converted into valuable products through chemical, biological, or electrochemical processes. Carbon utilization pathways include the production of chemicals, fuels, building materials. Examples of carbon utilization technologies include the conversion of CO_2 into methane (methanation); the synthesis of chemicals such as methanol, ethylene oxide, or formic acid; the production of polymers and plastics; and the mineralization of CO_2 into inorganic carbonates for use in construction materials.

CCU (carbon capture and utilization) offers several potential benefits, including carbon emission reduction, resource conservation, an important economic opportunity by generating revenue from the sale of carbon-derived products, diminution of the costs associated with carbon capture and storage (CCS), and waste reduction. In this regard, various large-scale multinational projects have been developed recently, including the CO2EXIDE project, which was granted in 2018 by the European Commission in the frame of the Horizon 2020 program. Its main goal is the establishment of an electrochemical, energy-efficient, and near-to-CO_2-neutral process to produce bulk chemical ethylene from CO_2, water, and renewable energy [25].

To tackle the capture of CO_2, several approaches have been proposed. Firstly, technologies such as zero emission are poised to significantly simplify the separation process, which is often the most expensive step in CO_2 recovery. Also, membrane separation processes offer several advantages over traditional techniques such as a high selectivity for CO_2 over oxygen or nitrogen while utilizing polymeric and/or inorganic materials [26–28]. Porous membranes, when combined with supporting amine solutions, have demonstrated effectiveness in CO_2 separation [13,29,30]. Specifically designed materials, capable of separating molecules based on molecular weight or size, show promise for CO_2 adsorption.

2.2. CO_2 Capture Using Adsorbents and Membranes

Adsorption is the process by which molecules adhere to the surface of a solid material (adsorbent) [31–33]. In CO_2 capture using adsorbents, porous materials with high surface areas and/or carbon dioxide-specific functional groups are utilized to selectively adsorb CO_2 molecules from gas streams. CO_2 capture via adsorption is a widely used method for separating and concentrating carbon dioxide from gas mixtures, such as flue gas or ambient air [34]. The gas stream, with most of the CO_2 removed, is then emitted to the atmosphere or utilized for a different purpose. The solid is then purified in stages using differences in pressure, temperature, or other stimuli to remove the CO_2 and further compress it for storage. The advantages of this process are numerous, including the high selectivity, flexibility of the process, scalability, and regenerability. Nevertheless, certain limitations can accompany the process, including the high energy cost, low adsorbent stability, and the spaciousness of the equipment.

The membranous process can serve as an attractive alternative, offering a complementary approach, where the selective permeation of CO_2 through a membrane material takes place, while other gases are retained [35]. The resulting separation is based on differences in molecular size, solubility, and/or diffusivity. Membranes for such systems consist of thin, porous layers through which the gas mixture is passed. The captured CO_2 can then be collected on the permeate side of the membrane for further processing or storage. Such a gas separation technique is currently widely used for air separation, hydrogen production, natural gas purification, and post- and pre-combustion carbon dioxide capture. An advantage of this process over adsorbent-based separations is that it is continuous, without any alterations in temperature or pressure. Upstream pressure is the only energy input required to induce the separation. This way, the challenges associated with energy efficiency, the spaciousness of the design, and modularity can be easily solved, while the selectivity of the separation and membranous degradation problems must be addressed. While the aforementioned technologies offer the same results via two complementary approaches, the appropriate method must be carefully selected depending on the process requirements, scale, and specifications. More precisely, membrane separation offers alternative energy- and carbon-efficient approaches to the current gas separation technologies, such as pressure swing adsorption (PSA), temperature swing adsorption (TSA), and cryogenic distillation [36]. Their advantages are numerous: membranes do not require thermal or pressure-driven phase changes, offering economic competitiveness, with the market steadily growing since 1977. Furthermore, the emergence of MOFs and their successful formation into crystalline films, followed by the success in mixed matrix membrane synthesis, has enabled the development of new materials with unique and outstanding properties [37]. To be considered for post-combustion flue gas treatment and to be economically competitive with amine absorption technology, membranes should demonstrate a high CO_2 permeance of 1000–5000 GPU and a CO_2 selectivity of 30–50 [37–39]. A flue gas produced by a typical 550 MW power plant contains 12–14 % by volume of CO_2, necessitating an enormous CO_2 permeance relative to traditional polymer membrane systems. For the pre-combustion capture process, the requirements are slightly easier, with a CO_2 permeance greater than 200 GPU and a CO_2/H_2 selectivity greater than 20 desired to be competitive with typical PSA processes [37,40]. Permeance is commonly expressed in units like gas permeance (e.g., GPU—gas permeation units, with 1 GPU corresponding to 10^{-6} $cm^3(STP)/(cm^2{\cdot}s{\cdot}cmHg)$) for gas separations, and it can vary significantly depending on the type of polymer, the substance being permeated, and the conditions like temperature and pressure [41].This concept is crucial in the design of membranes for gas separation, filtration, and various industrial applications.

Several polymeric membranes showed interesting capabilities for the selective permeation of nitrogen and carbon dioxide [42]. For instance, oxygen-rich polymers, polymeric ionic liquids, perfluoro polymers, and iptycene-based polymers are a few examples of performing membranes for CO_2/gas separation [42–45]. More precisely, it is well known that a high content of ether oxygen enhances the CO_2 affinity via the creation of a quadrupole–quadrupole interaction [46]. Liu and coworkers recently reported that a highly branched amorphous polymer incorporating a 1,3-dioxolane unit exhibited excellent CO_2/N_2 separation properties [47]. Likewise, crosslinked polyethylene glycol (PEG) derivatives prepared by Harrigan et al. have been shown to hinder the diffusion of CH_4, thus improving the CO_2/CH_4 selectivity [48].

As mentioned, polymeric ionic liquids showed potential for CO_2/N_2 and CO_2/CH_4 separations. They can be defined in several categories such as ion-gel membranes, poly(ionene)s, and poly(ionomer)s [49–51]. Deng and coworkers recently reported a PEG-diacrylate incorporating a 2-aminoethyl motif [52]. Several ionic liquids were blended with the polymer network, demonstrating targeted selectivity towards CO_2 in the presence of N_2. Bara et al. reported the preparation of a novel rigid polyamide–ionene material (Im-TB-PA ionene) using Menshutkin reactions between *N*,*N*′-(1,4-phenylene)bis(4-(chloromethyl)benzamide) and diimidazole-functionalized Tröger's base monomers [53]. The resulting membrane displayed excellent selectivity for CO_2. Schwartz et al. studied the catalytic activity and CO_2 kinetic separation performances of anionic ionomer-based membranes utilizing an N_2, O_2, and CO_2 gas mixture [54].

Perfluoropolymers are fluorinated polymers with C-H bonds replaced by C-F bonds. In terms of CO_2 separation, several derivatives have been studied [55,56]. For example, the gas separation of perfluoro(butenyl vinyl ether)co-perfluoro(2,2-dimethyl-1,3-dioxole) polymer was reported [57]. The resulting polymeric membrane showed increasing H_2/CO_2 selectivity with temperature. Another example is the iptycene polymers, a class of 3D materials incorporating a [2,2,2]bicyclooctatriene moiety. In terms of CO_2 separation, these materials have shown promising results as reported by Luo et al. [58,59]. In particular, the authors studied a triptycene rearranged–polybenzoxazole (TR-PBO) polymer membrane for highly selective and permeable gas separation membranes with potential for H_2 purification and CO_2 removal from natural gas [60].

2.3. MOFs, ZIFs, and Other Crystalline Materials for CO_2 Capture

For efficient CO_2 capture and utilization, various classes of porous solid materials were brought to the table as a potential effective solution: metal–organic frameworks (MOFs) [61], carbon-based materials [62], zeolites [63], zeolitic imidazolate frameworks (ZIFs) [64], nanoporous silica materials [65], porous aromatic frameworks (PAFs) [66], covalent organic frameworks (COFs) [67], hybrid ultramicroporous materials (HUMs) [68], and others. Moreover, when selecting adsorbent materials for effective CO_2 capture applications, it is essential to consider critical parameters such as adsorption capacity, selectivity, kinetics, regenerability, stability, mechanical robustness, cost, and energy consumption [69].

MOFs belong to a class of crystalline materials that consist of coordination bonds between metal or metal oxide connected by organic linkers (referred to as secondary building units [SBUs]) via a self-assembly process forming a three-dimensional structure [70]. In this regard, metal–organic frameworks (MOFs) are very promising candidates for CO_2

adsorption [71]. They are perhaps the most thoroughly studied class of porous adsorbents that offer tremendous potential due to (1) their high versatility (tunable composition and structure), offering an important CO_2 adsorption capacity, (2) high CO_2 affinity, (3) large surface areas (greater than 7000 m^2g^{-1}), and (4) high pore volumes [72]. They can be readily self-assembled from metal ions with an organic linker through metal–organic bonds. The following are the most reported groups of MOF materials. An elegant classification was proposed by the researchers from the Scientific Research National Center of Paris, based on the nature of an organic linker and a metal centre [73].

1. polycarboxylates of trivalent cations (Fe^{3+}, Cr^{3+} and Al^{3+}) associated with inorganic units in the form of oxo-trimers (MIL-88, MIL-100, MIL-101, MIL-127) or the chains of octahedrons MO6 (MIL-53, MIL-160, NOTT-300 (NOTT states for Nottingham University)), CAU-23 (CAU states for Christian Albrechts University). Among them, the MOFs carrying Cr^{3+} ions are generally the most chemically stable due to the strong bond between the cation and a ligand.

2. carboxylates of tetravalent cations (Zr^{4+} and Ti^{4+}) have experienced considerable growth since the discovery of MOF UiO-66. They are generally constructed based on oxo-clusters Zr6, such as NU-1000 (NU stands for Northwestern University), MIP-200 (MIP stands for Materiaux Institut Paris), but also Zr12 (MIP-206) and sometimes ZrO8 polyhedral chains (MIL-140).

3. azolates of divalent cations, such as zeolitic imidazolate frameworks or those based on Ni12 oxo-clusters (fcu-Ni(DP), where FCU stands for face-centred cubic) and PCN-601 (PCN stands for porous coordination network).

The properties of both metal ions and organic linkers, as well as synthetic conditions (the solvent system, pH, metal–ligand ratio, and temperature), determine the physical, structural, and morphological features of MOF networks (e.g., porosity, pore size, and pore surface) [74]. Figure 1 illustrates common MOFs structures. Some of the most well-studied MOF structures include MOF-5 (also known as IRMOF-1) [75], one of the earliest and most extensively studied MOF. It consists of zinc ions coordinated with benzene-1,4-dicarboxylate (BDC) linkers; HKUST-1 (also known as Cu-BTC) [76] is composed of copper ions coordinated with benzene-1,3,5-tricarboxylate (BTC) linkers; MIL-101 is a family of MOFs with various metal ions (e.g., aluminum, chromium, iron) coordinated with terephthalic acid linkers, characterized by an exceptional porosity [77]; ZIF-8 is composed of zinc ions coordinated with imidazolate linkers, characterized by an excellent chemical stability and intensely studied in cancer-treatment therapy [78,79]; MOF-74 consists of metal ions (e.g., magnesium, zinc) coordinated with 2,5-dihydroxyterephthalic acid linkers, characterized by an exceptional CO_2 adsorption capacity [80,81]; UiO-66 is composed of zirconium ions coordinated with terephthalic acid linkers [82]; MOF-177 is constructed from octahedral $Zn_4O(\text{-COO})_6$ and triangular 1,3,5-benzenetribenzoate (BTB), which possesses an exceptionally large pore size for microporous MOFs up to 1.8 nm [83].

MOF's pore surfaces can be readily fine-tuned to provide a highly selective bonding of CO_2 [84]. However, some MOFs were found to be unstable at high temperatures, under moisture conditions, or unable to meet industrial standards. A discussion on the subject was reported by Zhang et al., in which they reviewed the performances of a variety of microporous MOFs for CO_2 capture and separation while emphasizing the most important role in the CO_2 capture: capacity, selectivity, and regeneration [84]. Some of the novel trends for the chemistry of MOFs include the post-synthetic modifications, such as metal ions [85], ligand exchange [86], the introduction of novel functional groups [87], and surface functionalization with polymers [88] or nanoparticles to enhance the adsorption capacity of CO_2. Kim and colleagues obtained MOFs with exceptional porosities and gas (hydrogen, methane, and carbon dioxide) uptake capacities using the concept of crystalline structure extension via the addition of secondary organic links [89]. These impressive materials demonstrated Brunauer–Emmett–Teller and Langmuir surface areas of up to 10,400 square meters per gram, the highest reported for crystalline materials, and a total carbon dioxide storage capacity of 2870 milligrams per gram (65 mmol CO_2 per gram of sorbent) at 50–60 bars. For the capture of CO_2 at atmospheric pressure, the best performance belongs to another MOF, namely TMU-42 $\{[Zn_2(fum)_2(4\text{-bpdb})]\cdot 2H_2O\}n$ (fum = fumarate; 4-bpdb = 1,4-bis(4-pyridyl)-3,4-diaza-1,3-butadiene), which demonstrated an uptake of 320 milligrams per gram (7.29 mmol CO_2 per gram of sorbent) [90].

With the advancement in the synthesis of water and thermally stable MOF structures, some industrial-scale projects have recently emerged. The project titled MOF4AIR is a European Commission's four-year initiative in the frame of the Horizon 2020 program, gathering 14 partners from 8 countries to develop and demonstrate the performances of MOF-based CO_2 capture technologies in power plants and energy-intensive industries, with a total budget of over EUR 11.1 M [91]. MOF4AIR stands for metal–organic frameworks for carbon dioxide adsorption processes in power production and energy-intensive industries.

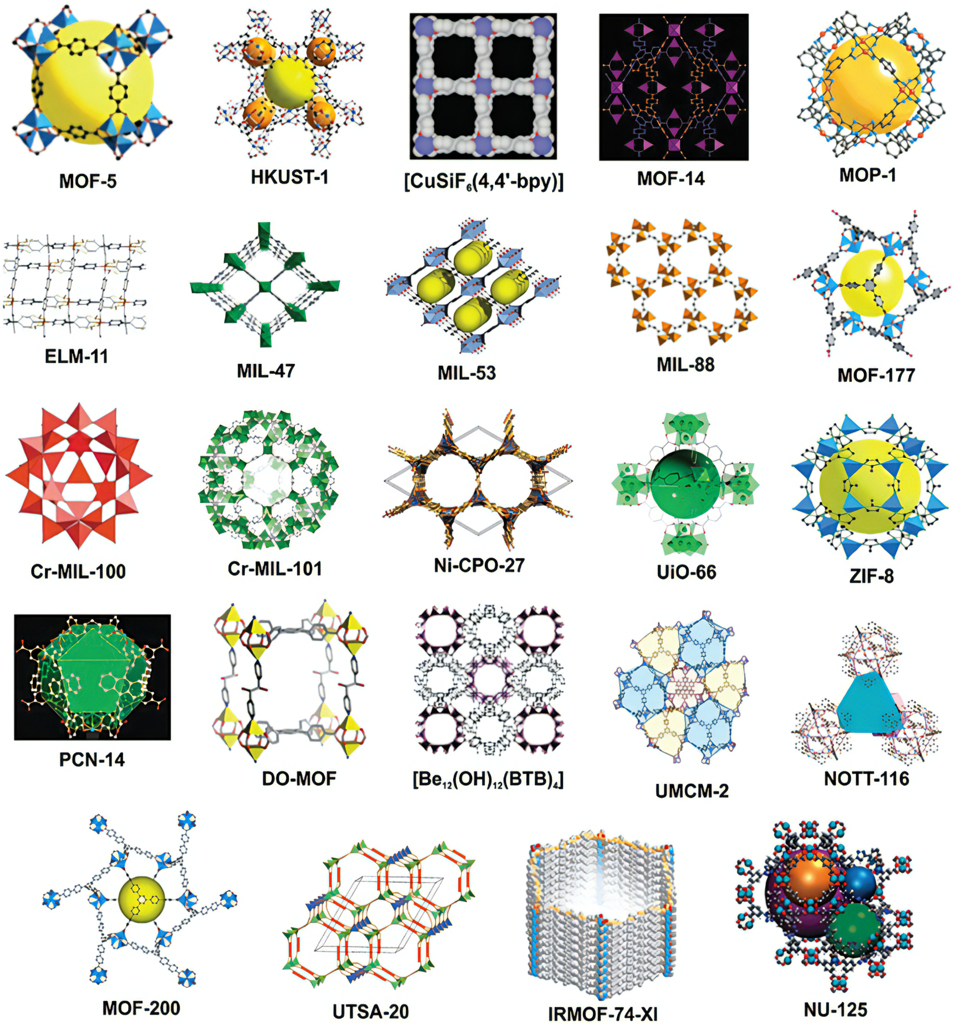


Figure 1. Representation of different MOF structures. Reproduced with permission from the Royal Society of Chemistry from [92].

Zeolites and zeolitic imidazolate frameworks (ZIFs) represent another important class of CO_2 adsorbents [64,93,94]. These two classes of porous materials share several common characteristics, such as a porous structure, high surface area, tunable pore size, high crystallinity, and the possibility to be functionalized via the surface and in-bulk modification [95]. While zeolites are crystalline aluminosilicate minerals that can be synthesized through hydrothermal or solvothermal methods [96], ZIFs belong to the class of MOFs composed of metal ions or clusters coordinated with imidazolate ligands and can be synthesized through hydrothermal, mechanochemical, or solvothermal methods [97]. The choice of synthesis parameters influences the resulting zeolite structure, composition, and properties [98]. Natural zeolites occur in geological formations, while synthetic ones should be synthesized in the laboratory. Synthetic zeolites offer advantages such as uniform pore sizes, high purity, and tailored properties, making them preferred for many industrial applications [99]. A large segment of the current global economy (350 Billion United States dollars (USD)) is based on the use of crystalline microporous zeolites in various essential processes, such as petrochemical cracking [100], ion exchange for water softening and purification [101], and in the separation of gases [99]. Even though the zeolites generally exhibit higher thermal and chemical stability compared to ZIFs due to the strong covalent bonds between tetrahedral Si(Al)O_4 units, some ZIFs have been reported to exhibit exceptional stability under harsh conditions, including high temperatures, humidity, and chemical environments [102]. Such enhanced stability is attributed to the strong coordination bonds between metal ions and imidazolate ligands, as well as the rigid nature of the framework structure.

As for their CO_2 adsorption capacity, ZIFs may offer advantages over zeolites in terms of enhanced surface areas and stronger chemical interactions with CO_2. Calero and others performed a computational screening study to examine the families comprised of the experimentally known zeolites and their respective ZIF counterparts in the context of several environmental and industrial separations involving carbon dioxide, nitrogen, methane, oxygen, and argon [103]. In regard to pore size, ZIFs have demonstrated pore sizes approximately twice as large as those of their zeolite counterparts due to the larger size of the imidazole linkers (the average first neighbors Si–Si distance is 3.05 Å in zeolites, compared with the average Zn–Zn distance of 6.00 Å in ZIFs). Thus, while some zeolites can act as molecular sieves based on the relative sizes of the adsorbates under study, the larger apertures of the ZIFs make them lose the molecular size-sieving ability. Thus, zeolites are more likely to be used in the size exclusion separation of CO_2 from N_2, as in the case of CO_2 from CH_4 separation. In terms of the accessible surface area and accessible volume of the structures, the larger pore sizes of ZIFs in relationship to zeolites imply larger spaces to accommodate the guest molecules. This fact makes them especially useful for gas storage applications. Finally, based on interatomic potential-based calculations, ZIFs appear to be better candidates for gas separation processes involving mixtures of non-polar and quadrupolar molecules, such as the removal of carbon dioxide for natural gas purification. According to Calero and coworkers, ZIFs also seem to surpass the performance of zeolites for the separation of carbon dioxide from flue gas. Finally, the work of Yaghi et al. seems to prove the predictions of Calero. Synthesized series of very stable ZIFs, termed ZIF-68, ZIF-69, and ZIF-70 exhibit unusual selectivity for CO_2 capture from CO_2/CO mixtures and extraordinary capacity for storing CO_2: 1 liter of ZIF-69 can hold approximately 83 liters of CO_2 at 273 K under ambient pressure [104].

At last, covalent organic frameworks (COFs) are covalently linked to two- and three-dimensional (2D and 3D) organic crystalline structures obtained by employing reticular synthesis. These synthetic routes typically involve the self-assembly of organic building blocks under mild reaction conditions, leading to the formation of crystalline frameworks with predictable structures and properties [105]. COFs were first reported in 2005 and quickly became the subject of intensive research in the domain of catalysis, gas separation, and optoelectronics [106]. COFs can be synthesized through various methods, including condensation reactions, Schiff base reactions, and dynamic covalent chemistry. The main difference between COF and MOF materials, as reported by Yaghi, is that COFs, despite being reticulated solids, are more versatile in molecular synthesis. This is primarily because COFs use purely organic linkers, typically aromatic and polyaromatic compounds like boroxine and boronic esters [107–109], imines, hydrazones, azines [108,110], *b*-ketoenamines [111], imides, or cyanovinylene linkages. In terms of their adsorption capacity, the highest uptake of CO_2 belongs to the group of 3D medium-sized pores structures (COF-102 and COF-103), which reached 1180 milligrams per gram of sorbent (26.8 mmol CO_2) at 35 bars, as reported by Yaghi [112]. COFs will be discussed in more detail in the upcoming section.

3. Petroleum-Based Polymers

The following section provides a brief overview of the main types of petroleum-based polymeric materials used for CO_2 capture: amine-based polymers, polymeric ionic liquids, conjugated macrocyclic polymers, porous organic polymers, and ionic polymers. For information purposes, petroleum-based polymers can be defined as artificial organic polymers (e.g., polyethylene, polypropylene, nylon, polyester, polytetrafluoroethylene, epoxy), obtained from natural gas or oil hydrocarbons [113–115]. These synthetic petroleum-based polymers possess highly desirable properties, such as strength, flexibility, resistivity, and chemical inertness, to name a few [115].

3.1. Amine-Based Polymers

Amine-functionalized adsorbents typically consist of porous support to which amines are introduced, either by weak physical adsorption (impregnation) or through strong chemical bonding (grafting/covalent attachment). Impregnated systems involve the direct deposition of amines onto the support, whereas grafted/covalently bonded systems refer to the chemical incorporation of amines, either during the material's synthesis (in situ) or via post-synthesis modification [116].

Although impregnated CO_2 sorbent systems often exhibit high capture capacity, they encounter stability issues such as amine leaching and degradation, as well as limited CO_2 transport to active sites due to diffusion hindrance. To overcome these challenges and enhance long-term performance, a grafted/covalently bonded CO_2 sorbent systems approach offers improved stability over multiple regeneration cycles. More specifically, polymeric amines, a group of porous materials containing multiple amino groups, are gaining significant attention for improving CO_2 capture capacity of supported amine-based sorbent systems due to their higher amine content [116–118]. It is worth noting that polymeric amines with primary and secondary amine groups are preferred for such applications due to their effectiveness in capturing CO_2 in various environmental conditions [119]. For instance, primary amine can efficiently capture CO_2 by the formation of stable carbamate species. Secondary amines are considered the ideal functional group (in a polymer porous network) as they give a balance between the adsorption and the regeneration [120]. At last, the tertiary amine can regenerate more easily under mild conditions while being detrimental to the adsorption capacity and selectivity.

Polyethylenimine (PEI) is among one of the most utilized and researched polyamines for CO_2 capture. Its widespread adoption is due to factors such as its easy availability, high CO_2 capture efficiency, elevated amine density, presence of primary amine chain ends, and stability, which enable it to retain CO_2 adsorption capability even under elevated temperatures [121]. Figure 2 depicts the main and most reported polymeric amines for CO_2 capture. In the same vein, Varghese and coworkers recently reported a complete summary encompassing the CO_2 uptake performance of several PEI derivatives [117].

In this regard, Chen and coworkers have shown the preparation of a polyvinyl amine (PVAm)/piperazine glycinate membrane for efficient CO_2 capture from flue gas [122,123]. The PVAm polymer was prepared by polymerizing *N*-vinyl formamide using AIBA. The obtained polymer was then subjected to acid hydrolysis followed by an ion exchange to generate the PVAm (Figure 3). Optimizing the amine layer thickness, the molecular weight and the piperazine glycinate ratio of the PVAm led to a CO_2 permeance up to 1100 GPU. Polymer permeance refers to the ability of a polymeric material to allow substances, typically gases or liquids, to pass through. It is a measure of how easily a specific substance can permeate or diffuse through a polymer film or membrane.

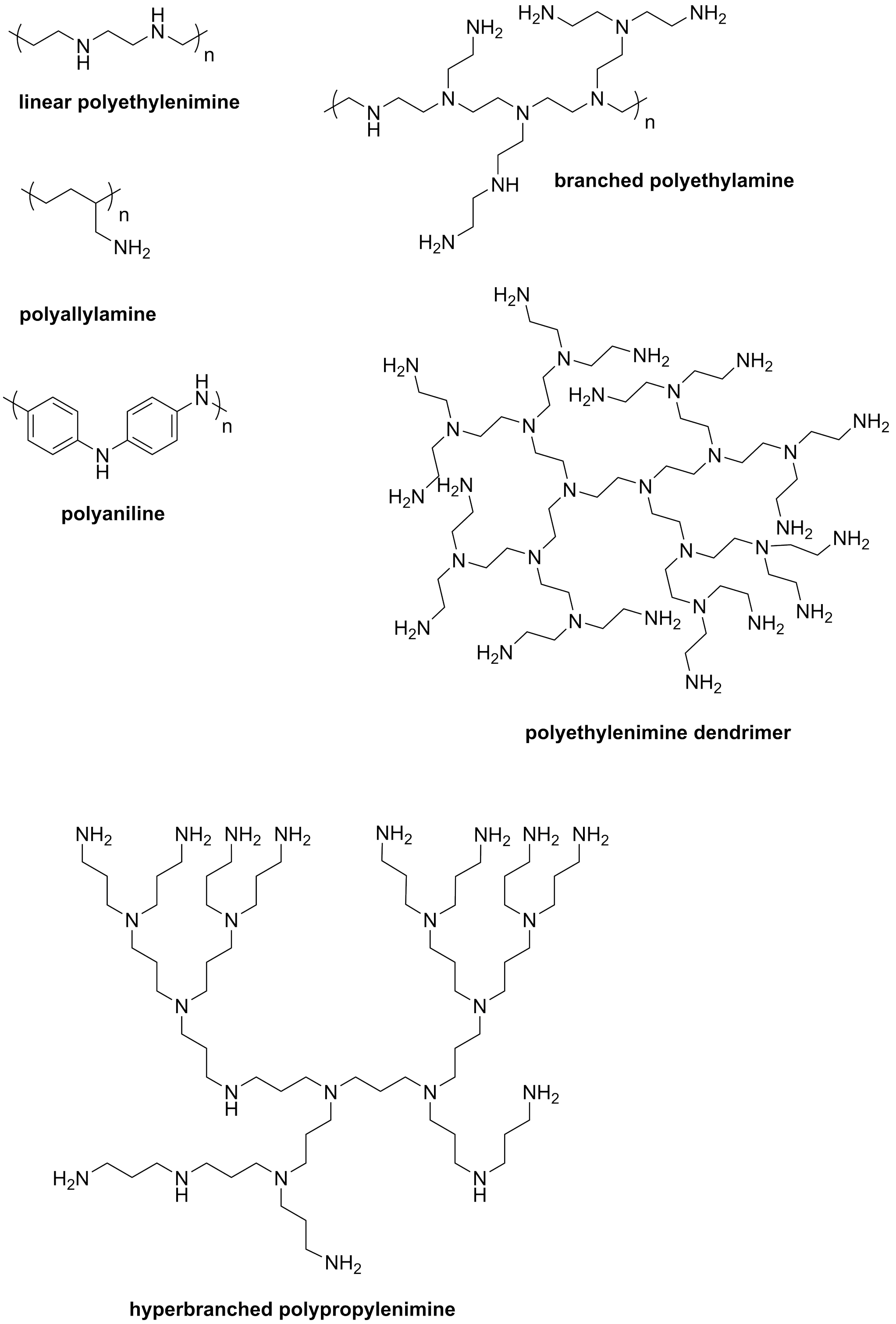


Figure 2. Most studied polymeric amines for CO_2 capture.

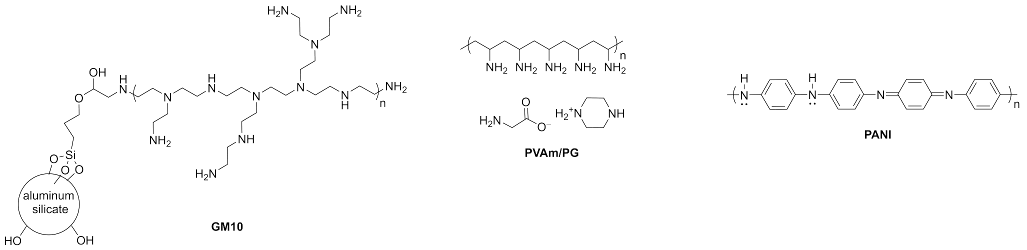


Figure 3. Recent examples of polymeric amines for CO_2 capture [122,124,125].

Liu et al. reported the preparation of an impregnated poly(ethyleneimine) (PEI) on a poly(divinylbenzene (PDVB) scaffold [126]. The PDVB support was first prepared by a solvothermal and template-free method using AIBN and DVB in a mixture of THF/H_2O. Heating the solution mixture to 120 °C afforded a PDVB solid scaffold, which was then physically impregnated with PEI at different weight loadings. In terms of CO_2 capture, the PEI–PDVB scaffold was able to reach a CO_2 capacity of 3.70 mmol g^{-1} at 298 K. The authors attributed its performance to its abundant meso-macropores and large volume pores. Sujan and coworkers have reported the direct capture of CO_2 from air using a PEI-loaded silica sorbent [127]. They showed that by implementing a vacuum- and temperature-assisted desorption step, high-purity CO_2 recovery from ambient air was successfully achieved regardless of moisture presence.

Recently, Mirkovic et al. reported the preparation of a PEI–amine-modified metakaolin brushite hybrid [124]. First, 3-glycidoxypropyltrimethoxysilane was reacted with a sodium silicate solution using the sol-gel method. Then, the epoxy-grafted intermediate was functionalized with PEI at 60 °C for 24 h. At last, the N1-(3-trimethoxysilylpropyl) diethylenetriamine (TMPTA) was used to finalize the formation of the targeted composite, allowing polymer GM10 to exhibit a CO_2 uptake of 0.685 mmol g^{-1} at 348 K (Figure 3).

Porous carbon derived from polyaniline has recently been reported as a potential CO_2 capture material [125]. Aniline was polymerized by chemical oxidation in the presence of sulfuric acid and ammonium peroxydisulfate. The corresponding porous carbon derivative was obtained by the carbonization of PANI (polyaniline) with $ZnCl_2$ at elevated temperatures of 500 °C and 650 °C for 1 h. A maximum CO_2 capacity of 3.54 mmol g^{-1} at 298 K and 1 bar was observed for the PANI derivative Zn-1500 (Figure 3).

3.2. Polymeric Ionic Liquids

Polymeric ionic liquids (PILs) are defined as polyelectrolytes containing ionic liquid species (cations or anions) and thus, forming a large molecular structure [128]. Ionic liquids (ILs) are liquids at room temperature due to weak intermolecular forces, whereas PILs possess strong intermolecular forces between polymer chains and are, therefore, solid. In addition, PILs, synthesized from ionic liquid monomers (ILMs), offer improved processability, stability, and control over structure. For PILs to be successful candidates for CO_2 capture, the effect of structural variation in PILs (i.e., choice of cation, anion, backbone, alkyl chain length, porosity, and cross-linking) needs to be evaluated. For a complete overview of the CO_2 capture capacity of PILs up to 2015, the reader is referred to the work of Zulfiqar and coworkers [129]. For the reader's benefit, typical structures of PILs for CO_2 sorption are illustrated in Figure 4. Depending on the structure and the choice of the ion, CO_2 capture capacity ranging from 0.03 to 0.3 mmol g^{-1} at 298 K and 1 bar could be obtained [129].

Recently, Sang and coworkers reported the preparation of benzimidazole-based hypercross-linked poly(ionic liquid)s (HPILs) [130]. The HPILs were obtained by Friedel–Crafts alkylation reaction between the ionic liquid monomer (i.e., benzimidazole) and benzyl halide moieties. A CO_2 uptake of 1.47 mmol g^{-1} at 298 K and 1 bar were obtained. Interestingly, the polymeric substrate also showed catalytic activity for CO_2 cycloaddition (i.e., epoxides to cyclic carbonates).

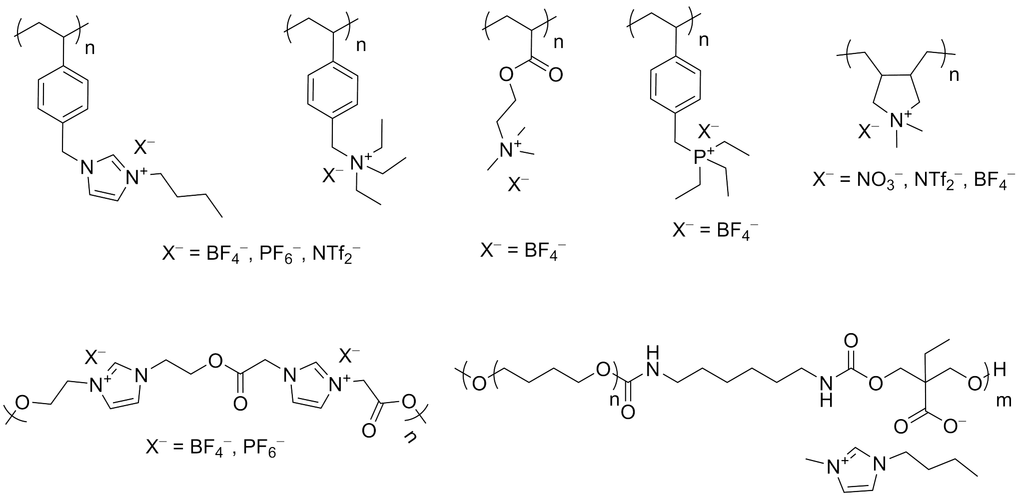


Figure 4. Typical structures of polymeric ionic liquids used for CO_2 sorption [129].

3.3. Conjugated Macrocyclic Polymers

Conjugated macro/microcyclic polymer combined the properties and structures of the porous organic network and macrocycles [131,132]. For instance, Dai et al. reported the preparation of an arene-based conjugated macrocycle (BpP6-OTf-CMP-2) using a Sonogashira–Hagihara palladium-catalyzed cross-coupling reaction [133]. They design the ring to be selective towards CO_2 by varying the internal diameter. They observed a CO_2-adsorption capacity of 1.83 mmol g^{-1} at 273 K at 1 atm.

Ren and coworkers reported the preparation and characterization of 1,3-diyne-linked conjugated microporous polymer for CO_2 capture [134]. Using a Sonogashira cross-coupling reaction, 1,3,6,8-tetraethynylpyrene was reacted in the presence of $Pd(PPh_3)_4$, CuI, and *N,N′*-diisopropylethylamine in DMF to afford microporous-conjugated polymer LKK-CMP-1 (Figure 5). A CO_2 adsorption uptake of 2.27 mmol g^{-1} at 273 K and 1 bar was measured.

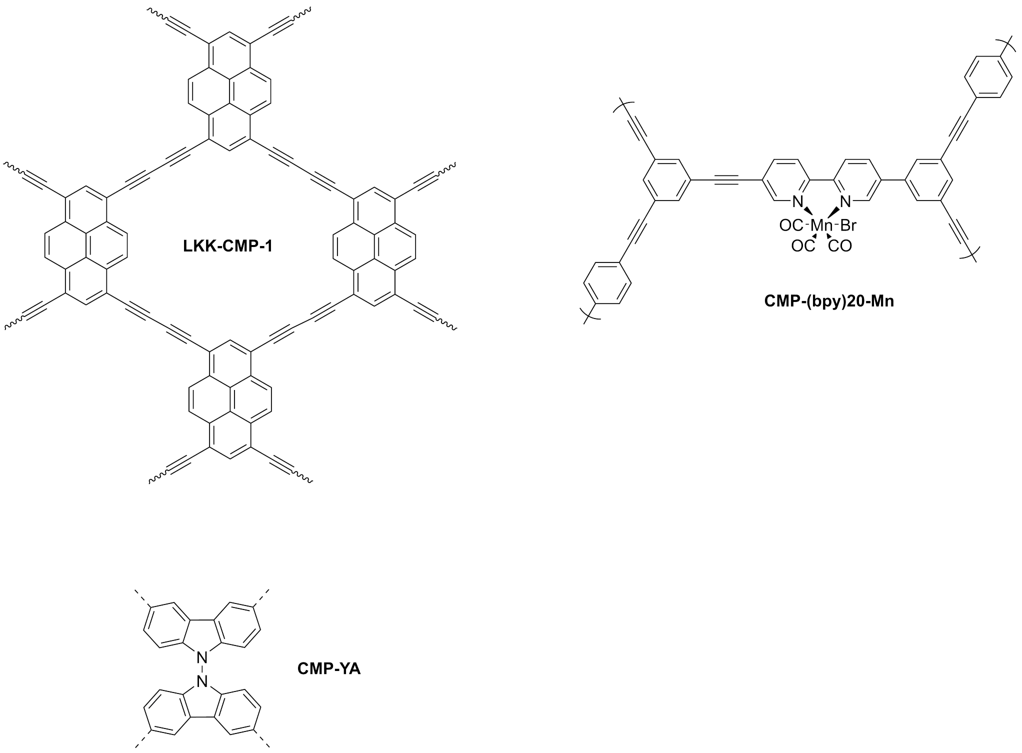


Figure 5. Structures of conjugated macrocyclic polymers for CO_2 capture [134–136].

An interesting report from Smith et al. showed the preparation of a metal–organic conjugated microporous polymer incorporating a bipyridine moeity [135]. In short, the CMP-(bpy)20 was prepared by a Sonogashira cross-coupling reaction of 1,3,5- triethynylbenzene with 5,5′-dibromo-2,2′-bipyridine and 1,4-dibromobenzene (Figure 5). The obtained polymer was purified and then refluxed in diethyl ether for 24 h with [$Mn(CO)_5Br$] to afford the targeted CMP-(bpy)20-Mn. A CO_2 uptake of 1.06 mmol g^{-1} at 298 K and 1 bar was observed.

Another example of a conjugated polymer network was reported by Yuan et al. [136]. They prepared a *N*-*N*′-bicarbazole homopolymer by Yamamoto cross-coupling reaction using 3,3′,6,6′-tetrabromo-*N*,*N*′-bicarbazole as the starting material and ([$Ni(cod)_2$] as the catalyst. The obtained polymer CMP-YA showed a CO_2 uptake of 1.27 mmol g^{-1} at 298 K and 1 bar (Figure 5).

3.4. Porous Organic Polymers

POPs, or hyper-crosslinked polymeric materials constructed solely from organic covalent bonds, have emerged as versatile substances with a wide array of applications, including CO_2 capture. These materials are known by different names, including porous polymer networks (PPNs), porous organic frameworks (POFs), conjugated microporous polymers (CMPs), microporous organic polymers (MOPs), polymers of intrinsic microporosity (PIMs), hypercrosslinked polymers (HCPs), covalent triazine-based frameworks (CTFs), porous aromatic frameworks (PAFs), and crystalline covalent organic frameworks (COFs) [137]. For convenience, the term POPs broadly encompasses these materials.

POPs offer several advantages for carbon capture applications. Firstly, their construction from relatively rigid monomers allows for the formation of pores with rigid walls, leading to permanent porosity comparable to the most porous materials. Notably, PPN-4 has demonstrated the highest surface area among POP materials published thus far [10]. Secondly, the synthesis of POPs draws from a wide range of modern bond-forming methodologies (i.e., Yammoto coupling [10], boronic acid condensation [138], metal-catalyzed coupling [139], Schiff-base [140,141], and Friedel–Craft alkylation [142]), resulting in numerous structural topologies and diversified porosities. Additionally, POPs, composed of lightweight elements, tend to have higher gravimetric carbon capture capacities. Moreover, their covalent bond nature confers high stability and exceptional chemical and water stability, enabling reusability. These properties, combined with large surface area, tunable pore size, high stability, and easy functionalization, make POPs ideal candidates for carbon capture [10,143–145].

As stated previously, covalent triazine frameworks (CTFs) have gained significant attention for their easy and scalable synthesis methods, as well as their tunable structures. A prevalent method for CTF preparation involves ionothermal trimerization of aromatic nitriles in the presence of $ZnCl_2$ at high temperatures, forming triazine linkages [146]. Alternative synthetic routes include acid-catalyzed and elemental sulfur-mediated approaches [147,148]. With high surface area and thermal stability, CTFs' high nitrogen content renders them attractive for CO_2 capture and separation. Additionally, their chemical and physical stability make CTFs suitable as catalytic supports for various reactions, including CO_2 reduction [149]. However, challenges such as low conversion yield and selectivity have been observed despite promising catalytic activity. Recently, Buyukcakir and coworkers have proposed the preparation of a charged covalent triazine framework based on cyanophenyl-substituted viologen dication [150]. The latter was then trimerized by an ionothermal reaction involving $ZnCl_2$ with better material porosity when the reaction was carried out at 500 °C. A CO_2 adsorption of 3 mmol g^{-1} at 273 K and 1 bar was observed. Interestingly, the presence of charged functionalities in the CTFs also allowed the conversion of epoxide functional groups into cyclic carbonates. For instance, propylene oxide and epichlorohydrin were converted to their corresponding cyclic carbonates with a yield higher than 95%.

Shao and co-workers also studied the preparation of a triazine-based POP [151]. In a typical procedure, they reacted the chosen organic amine (i.e., ethylene diamine, triethylenediamine, or *p*-phenylenediamine) with 2,4,6-trichloro-1,3,5-triazine under basic conditions, affording the triazine-based POP (cCFTs) (Figure 6). These were then precarbonized at 200 °C, followed by a carbonization step at 500 °C to furnish the desired N-enriched porous carbons. In terms of performance, a CO_2 uptake of 0.93 mmol g^{-1} at 273 K and 1 bar was noted.

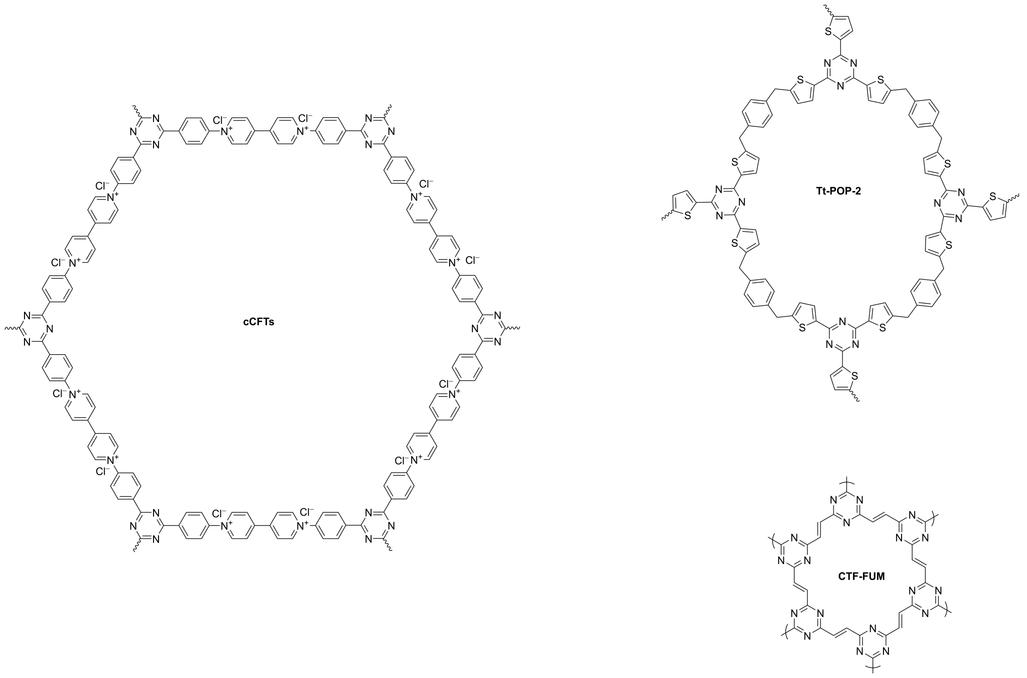


Figure 6. Examples of triazine-based polymers [151–153].

Recently, the group of Das and coworkers proposed an interesting example of a fully conjugated triazine–thiophene porous organic scaffold (Tt-POP-2) [153] (Figure 6). First, the Tt monomer was prepared by a cyclic trimerization of 2-thiophenecarbonitrile. The polymer was then obtained by a Friedel–Craft allylation in the presence of 1,4-bis(bromoethyl)benzene and $FeCl_3$ as Lewis acid in 1,2-dichloroethane. Different organic linkers were used. A CO_2 uptake capacity of 1.79 mmol g^{-1} at 273 K and 1 bar was observed for Tt-POP-2.

Similarly, Wang et al. proposed the preparation of covalent triazine frameworks based on fumaronitrile [152]. Starting from fumaronitrile, $ZnCl_2$ is added, and the mixture is sealed in a quartz ampule. The latter was heated to a temperatures between 350 and 500 °C for 40 h to complete the preparation of the covalent triazine framework (CTF-FUM) (Figure 6). A maximal CO_2 uptake of 2.55 mmol g^{-1} at 298 K was observed.

The introduction of an N/O/S-containing building block is an advised approach to improve adsorption capacity and selectivity. Functional groups and aromatic rings such as imines [154], imidazole [155], triazine [156], as well as azo linkages [157], and Troger bases [158] are usually used on the surface of the POP to increase its CO_2 adsorption.

Sang et al. reported the preparation of a POP Schiff base based on a benzene-1,3,5-triyltris((9H-carbazol-9-yl) methanone) [159]. To do so, the authors use a synthetic approach involving a Friedel–Crafts reaction with melanin to generate the corresponding TPOP-3 (Figure 7). An adsorption capacity of 4.09 mmol g^{-1} at 273 K and 1 bar was reached.

Sun and co-workers have reported the preparation of a benzene–ethylamine polymer network (PPN-81) [154] (Figure 7). The latter was prepared in one step from the reaction of 2,4,6-tris(chloromethyl)mesitylene and ethylene diamine. The presence of secondary amines in the structure of the network afforded a CO_2 uptake of 1.92 mmol g^{-1} at 295 K at 1 bar. The same group also reported the preparation of polymers based on a methylbenzene precursor [160]. More specifically, 2,4,6-tris(chloromethyl)-mesitylene (TCM) was reacted with ethylene diamine by a nucleophilic substitution reaction affording polymer NUT-1

(Figure 7). The latter reached a CO_2 uptake of 1.87 mmol g^{-1} at 273 K and 1 bar with enhanced selectivity towards CO_2 (compared to CH_4 and N_2).

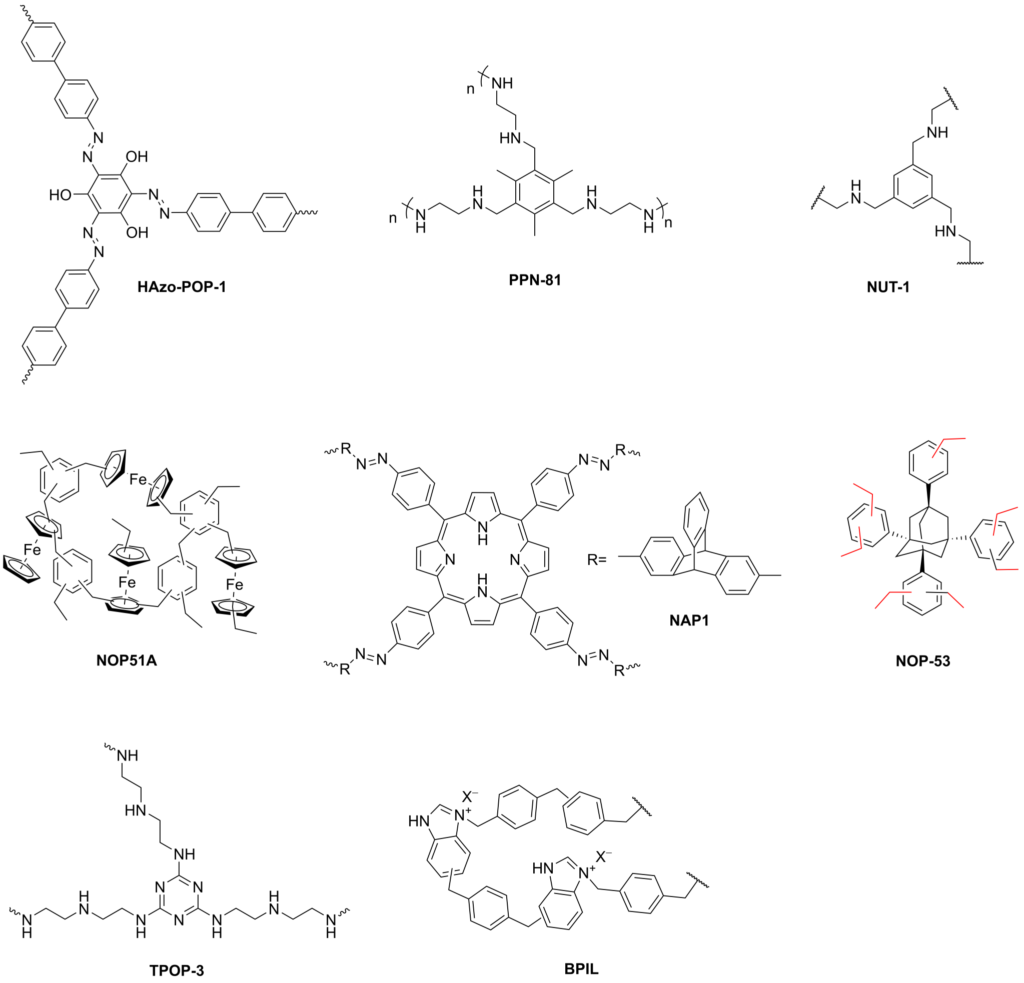


Figure 7. Examples of porous, nanoporous, and microporous organic polymers [154,159–165].

Adamantane moiety has also been used in the preparation of nanoporous organic polymers (NOPs) [164]. Using a Friedel–Crafts approach, 1,3,5,7-tetraphenyladamantane was reacted with formaldehyde dimethyl acetal in the presence of $FeCl_3$ to afford NOP-53 (Figure 7). The latter shows a CO_2 uptake of 2.27 mmol g^{-1} at 1 bar and 273 K while also demonstrating a high selectivity toward CO_2 over N_2 and CH_4. By following the same approach, Bera and coworkers reported nanoporous azo polymers based on triptycene for selective CO_2 uptake [163]. Derivative NAP-1 is obtained by direct coupling of the 2,6-diaminotriptycene with 5,10,15,20-tetrakis (4-nitrophenyl)-21H,23H-porphine, through the formation of azo-linkage without any metal catalyst (Figure 7). The authors reported a CO_2 uptake of 2.25 mmol g^{-1} at 298 K at 1 bar with a good selectivity over N_2. Similarly, Chen et al. proposed the preparation of tunable nanoporous triptycene, carbazole, and ferrocene-based polymers [165]. The polymers (NOP-50x) were prepared by Friedel–Craft alkylation using, for example, carbazole in the presence of dichloroxylene and $FeCl_3$ as catalyst (Figure 7). A CO_2 uptake up to 4.22 mmol g^{-1} was observed at 273 K and 1 bar for carbazole-based NOP.

Ji et al. reported a mesoporous *o*-hydroxyazobenzene polymer for CO_2 capture and conversion [162]. The polymers were prepared via a diazo coupling reaction of aryl tri/diamine with tri/diphenol in an aqueous solution at 0–5 °C leading to HAzo-POP compounds showing a CO_2 adsorption capacity of 1.68 mmol g^{-1} at 273 K and 1 bar (Figure 7).

Sekizkardes and coworkers have reported the preparation of a benzimidazole-based material (BILB-101) as an efficient ultra-microporous organic polymer [161] (Figure 7). In terms of synthesis, the latter was prepared by a simple and efficient polycondensation reaction between the 1,2,4,5-benzenetetramine and 1,3,5-triformylbenzene. A CO_2 uptake of around 1 mmol g^{-1} (4 wt %) at 298 K and 0.15 bars was reported for this material as well as an exceptional CO_2/N_2 selectivity.

Benzoxazine-based porous organic polymers were reported recently by Ejaz et al. [166]. They prepared the benzoxazine (BZ) unit through a sequence of steps by first reacting triphenylamine (TPA) and dihydroxy terephthalaldehyde (DHPT) units, followed by a Sonogashira–Hagihara coupling reaction between BZ monomers functionalized with TPA and DHTP was carried out to give the resulting polymer poly(DHTP-Ea BZ) (Figure 8). The BZ monomers were prepared using a multistep process that involved Schiff base formation, reduction, and Mannich reactions. A CO_2 capture capacity of 3.29 mmol g^{-1} at 298 K and 1 bar was recorded.

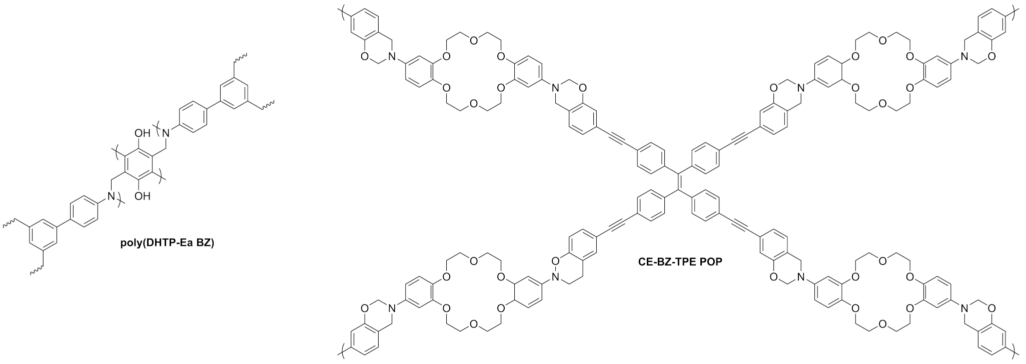
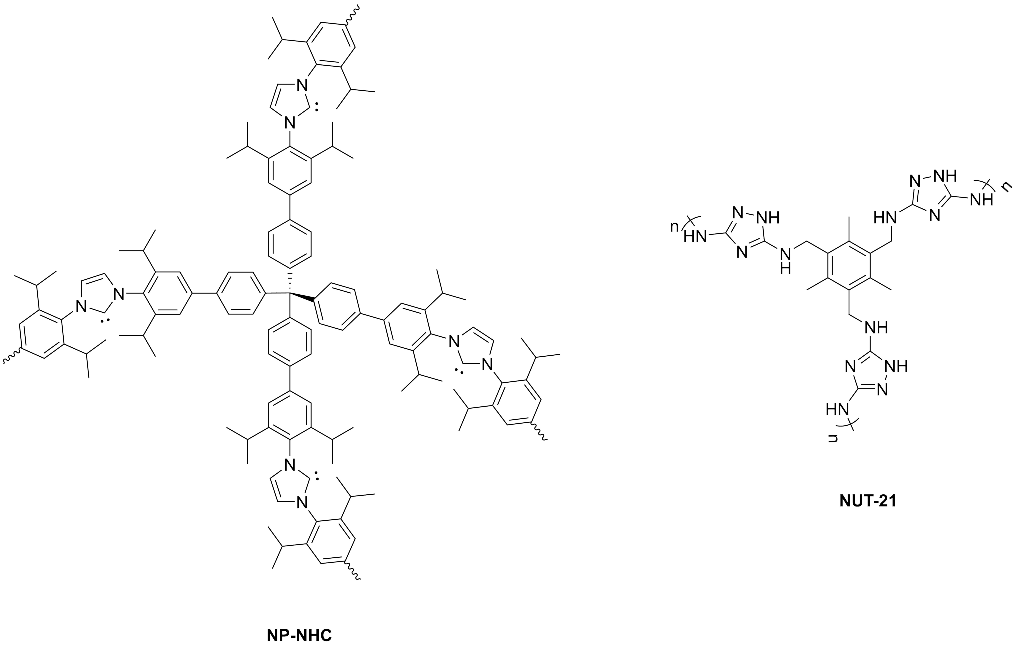


Figure 8. Benzoxazine-based polymers [166,167].

Mohamed et al. have reported a crown ether benzozazine-linked porous polymer [167]. In particular, they synthesized the CE-BZ monomer by a Schiff-base formation in the presence of 4-bromosalicylaldehyde, followed by a reduction in $NaBH_4$ and a Mannich condensation with formaldehyde in dioxane/water to furnish the targeted monomer. The polymer (CE-BZ-TPE-POP) was obtained by a Sonogashira–Hagihara Pd cross-coupling reaction in the presence of a copper catalyst and tetraphenylethylene (Figure 8). In terms of CO_2 capture capacity, an uptake of 4.39 mmol g^{-1} (298 K, 1 bar) was obtained after thermal polymerization at high temperature (200–300 °C).

N-heterocyclic carbenes (NHC) have been reported as CO_2 capture materials [168]. The *N*-heterocyclic carbenes were prepared in a three-step synthesis. First, a reaction of tetraphenylmethane incorporating 2,6-diisopropyl aniline with glyoxal gave the bis(2,6-diisopropyl phenyl) diazabutadiene. Second, the latter was reacted with paraformaldehyde in the presence of HCl to yield the corresponding imidazolium chloride derivative. Third, the NHC intermediate is obtained upon the neutralization of the imidazolium chloride with potassium *tert*-butoxide furnishing the NP-NHC (Figure 9). It is important to note that the isopropyl moieties were effective for steric confinement, leading to reduced stability of the NP-NHC-CO_2 adduct. This facilitates the efficient release of captured CO_2 at elevated temperatures and concurrently enhances its catalytic activity in the CO_2 conversion process. CO_2 capture capacity was found to be 0.78 mmol g^{-1} at 298 K and 1 bar. Finally, the NP-NHC substrate also demonstrated a cycloaddition of CO_2 to epoxide capability for selected substrates only (i.e., 2-methyloxirane, 2-(chloromethyl)oxirane, and 2-butyloxirane).

Interestingly, Peng et al. reported an *N*-doped porous carbon from a polymer precursor based on triazole moeity [169]. Concerning the synthetic route, the 2,4,6-tris(bromomethyl) mesitylene and 3,5-diamino-1,2,4-triazole were reacted in tetrahydrofuran, at 62 °C without the use of any catalysts under a N_2 atmosphere according to a nucleophilic substitution reaction affording polymer NUT-21. Then, a carbonization step (KOH, then 500 °C, 600 °C, and 700 °C for 3 h) furnished the NPC-T material. The CO_2 capacity was evaluated at 8.3 mmol g^{-1} at 273 K and 1 bar.


NP-NHC

NUT-21

Figure 9. Examples of *N*-heterocyclic carbene and *N*-doped polymers [168,169].

3.5. Ionic Polymers

Imidazolium salt-modified porous hyper-crosslinked polymers were introduced as a suitable CO_2 adsorbent by Wang et al. [170]. More specifically, the benzyl halide derivatives were directly self-polymerized by the Friedel–Craft reaction. Then, the unreacted benzyl halide groups were further reacted with *N*-methylimidazole to furnish the POM-IM structure (see Figure 10). The evaluation of the performances of the POM-IM material showed a CO_2 uptake of 3.68 mmol g^{-1} at 273 K and 1 bar. In addition, POM-IM can act as a catalyst for the conversion of epoxide to cyclic carbonate in yields ranging from 68 to 93% depending on the substrate. The same group also reported the preparation of phosphonium salt incorporated into a hyper-crosslink porous polymer [171]. In this case, a phosphonium salt was reacted with benzene and formaldehyde dimethyl acetal to give a series of polymers. A CO_2 capture capacity of up to 1.68 mmol g^{-1} was obtained at 298 K and 1 bar. These phosphonium-based polymers were also able to convert epoxide functions into cyclic carbonates.

Another example of imidazolium-based hypercrosslinked ionic polymer was reported by Li et al. [172]. In their study, they synthesized a 2-phenylimidazolinium HIP by a Friedel–Craft reaction between 2-phenylimidazoline and dichloro-*p*-xylene in the presence of $FeCl_3$ at 80 °C (Figure 10). The CO_2 uptake for this material was found to reach 2.1 mmol g^{-1} at 298 K and 1 bar. Furthermore, these HIP derivatives showed catalytic activity towards the conversion of epoxide to cyclic carbonate with high selectivity and yields.

Porous cationic polymers were reported by Buyukcakir et al. They used a Pd-catalyzed Sonogashira–Hagihara approach for their preparation [173]. Essentially, tetrakis(4-ethynylphenyl)methane was reacted with 1,10-bis(4-iodophenyl)-[4,40-bipyridine]-1,10-diium salts (dichloride, ditetrafluoroborate, or dihexafluorophosphate) in presence of a Pd source at 100 °C. The obtained PCP-Cl polymer exhibited a CO_2 capture capacity of up to 1.4 mmol g^{-1} at 298 K and 1 bar (Figure 10). In addition, the PCP derivatives also showed a catalytic activity for the conversion of epoxides to cyclic carbonates. Dani et al. also reported the synthesis of porous cationic polymers [174]. Starting from tetrakis(4-aminophenyl)methane, formaldehyde, and methyl glyoxal were added, and the mixture was reacted immediately according to a Debus–Radziszewski imidazolium synthesis. Anion exchange was then carried for different salts such as sodium tetrafluoroborate, bis(trifluoromethane)sulfonimide lithium salt, potassium hexafluorophosphate, and sodium trifluoromethanesulfonate leading to several derivatives. Click-based porous cationic polymer CB-PCP-1 exhibited a CO_2 adsorption capacity of 2.05 mmol g^{-1} at 273 K and 1 bar (Figure 10).

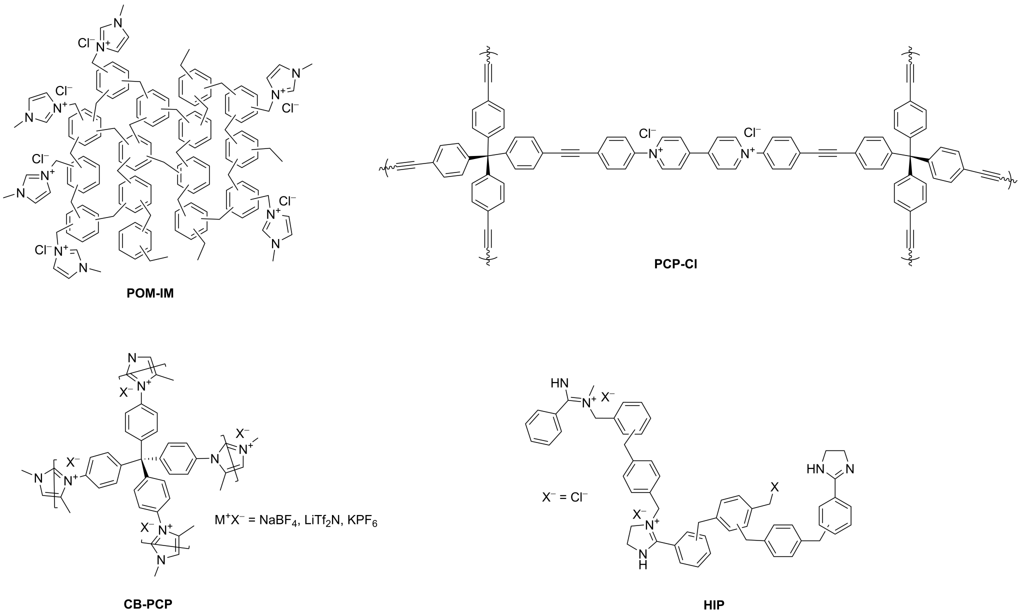


Figure 10. Structures of several ionic polymers [170,172–174].

4. Biobased Polymers

Bioderived polymers and their related compounds offer abundant and sustainable options for CO_2 fixation, chemisorption, and physiosorption [175–177]. Biobased polymers are defined as a type of polymer derived from renewable biological sources, such as plants, animals, or microorganisms, rather than from fossil fuels. These polymers can either be biodegradable or non-biodegradable. They are characterized by their low carbon dioxide footprint and are associated with sustainable practices [178]. For instance, the polysaccharides alongside polynucleotides and polypeptides, are the most prevalent naturally occurring biopolymer [179]. They are constituted of sugar monomers bound together by O-glycosidic bonds. By utilizing a catalytic system (i.e., DBU), epoxides, together with CO_2, can be efficiently converted into cyclic carbonates, demonstrating high conversions and selectivity. As described in the upcoming paragraphs, biomass can be employed adequately to enhance the capture of CO_2 [180] and its subsequent conversion into valuable compounds [181]. In their review, Aghel and coworkers detailed a variety of activated carbons such as cherry stones, corncob, cotton pulp, and wheat straw to name a few, with their corresponding adsorption data [182].

Guo et al. reported the preparation of a bio-based polybenzoxazine through carbonization and activation [183]. Starting from paraformaldehyde, fufurylamine, and 4,4′-thiobisphenol, a TBP-fa intermediate was obtained. The latter was then cured at a high temperature (230 °C) followed by carbonization at 500 °C. The obtained material was finally activated at 800 °C in the presence of KOH to give a series of NSOPC derivatives (Figure 11). Among them, the NSOPC-1 derivative showed a CO_2 uptake of 3.88 mmol g^{-1} at 1 bar and 298 K.

A fully bio-based epoxy thermoset (HMF-GU-Ep/DFA) has been reported as an efficient CO_2 capture adsorbent by Nabipour and coworkers [184] (Figure 11). They prepared the epoxy resin by curing 5-hydroxymethyl-2-furaldehyde-3, 5-diamino-1,2,4-triazole with 5,5′-methylenedifurfurylamine. They were able to reach a CO_2 uptake of 2.15 mmol g^{-1} at 298 K. Interestingly, the obtained epoxy thermoset was also effective as a flame retardant agent.

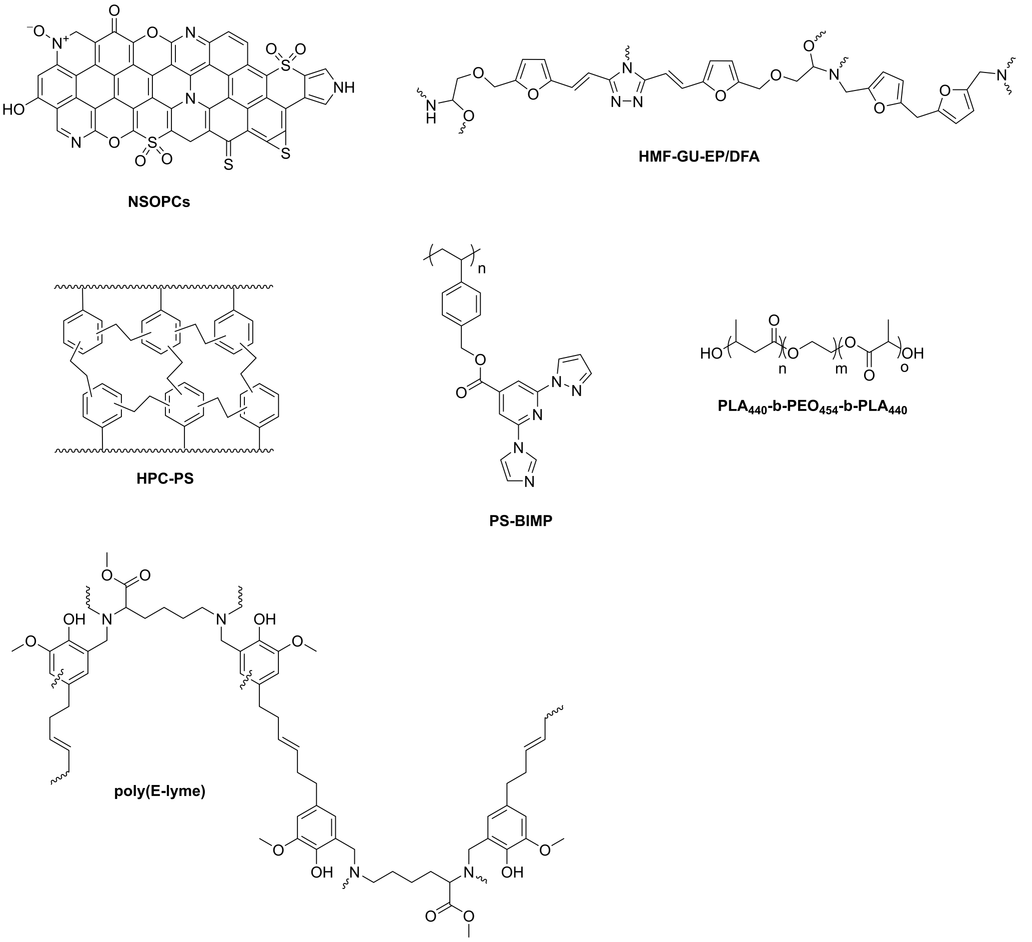


Figure 11. Bio-based polymers for CO_2 capture [183–188].

Aerogels made from sustainable materials like starch, cellulose, alginate, chitosan, and lignin have also been used for CO_2 capture due to their high porosity [189]. For instance, maximum CO_2 adsorption has been reported for a series of cellulose-based aerogel composites such as cellulose-crosslinked polyethyleneimine (2.32 mmol g^{-1}) [190], cellulose-derived porous carbon (0.257 mmol g^{-1}) [191], cellulose nanofibril grafted with amino silane (1.91 mmol g^{-1}) [192], and cellulose silica composites (3.68 mmol g^{-1}) [193].

The utilization of bio-based sorbents has retained attention as a cost-effective alternative to more traditional sorbents such as zeolite 13X [194,195]. First, biomass waste materials are typically carbonized (600 °C) under an inert atmosphere. Then, activation is carried out by chemical means (alkaline or acid solution), physical activation (steam or other gases at 800 °C), or a self-activation technique (i.e., utilization of the gases evolved during the carbonization phase as activating agents) [196,197]. For example, Tangsathitkulchai and coworkers have reported the use of coconut shell chars activated with a thermal annealing step at 450 °C, leading to a CO_2 adsorption of 3.60 mmol g^{-1} at a temperature of 272.85 K at 2.5 bars [198]. Similarly, cherry stones were used as bio-based sorbent through a physical activation method (steam) giving a CO_2 uptake of 2.50 mmol g^{-1} at 322.85 K at 2.5 bars [199]. At last, Vargas et al. studied the CO_2 capture of palm shell activated chemically by $ZnCl_2$ leading to an uptake of 17.44 mmol g^{-1} at 272.85 K at 20 bars [200].

Plastic waste can also offer opportunities as starting materials for CO_2 capture. A recent review from Teo and coworkers highlighted the main utilization of commodity plastics for the preparation of functional materials for CO_2 capture [201]. One of those, polystyrene (PS), is a versatile material for creating polymeric adsorbents due to the susceptibility of its aromatic groups to electrophilic functionalization: its π-electron-rich phenyl groups enable induced-dipole interactions with CO_2 molecules. Hypercrosslinked polymers (HCPs), synthesized via Friedel–Crafts alkylation from waste-expanded polystyrene (EPS) with crosslinkers, like carbon tetrachloride, exhibited high CO_2 adsorption capacity and selectivity. For instance, Fu and coworkers reported the preparation of an HCP PS network by using a Friedel–Craft approach (dichloroethane, $AlCl_3$) leading to a CO_2 capture capacity of 1.987 mmol g^{-1} at 273 K and 1.13 bars [185] (Figure 11). Hybrid HCPs, combining commercial PS with an octavinylsilsesquioxane (OVS) crosslinker, demonstrated tunable porosity and a CO_2 uptake of 1.12 mmol g^{-1} at 298K and 1.01 bars [202]. The latter were obtained by Friedel–Craft reaction between PS and OVS in the presence of $AlCl_3$. Another example showcases a two-step synthesis involving the nitration of PS to give polynitrostyrene followed by a reduction in HCl/Sn condition to furnish PSNH2 polymer as reported by Merchán-Arenas et al. [203]. The PSNH2 polymer showed a CO_2 uptake of 1.05 mmol g^{-1} at 273 K and 1 bar. At last, functionalizing chloromethylated PS with 2,6-bis-imidazo-1-yl-pyridine-4-carboxylic acid and imidazole at high temperature (403 K), led to the formation of the PS-BIMP polymer [186] (Figure 11).

In addition to polystyrene (PS), waste polyvinyl chlorides (PVCs) are also a viable source for developing polymeric adsorbents for carbon capture and utilization (CCU) [201]. In this regard, Sneddon et al. synthesized PVC-based/silica composites by introducing the aminated PVCs on mesoporous silica [204]. The ethylenediamine-treated PVC/SBA-15 composite exhibited an adsorption efficiency of 0.5 mmol g^{-1} at 298 K. Moreover, these composites demonstrate increased hydrophobicity, indicating suitability for operation under humid conditions without deactivation by moisture. Beyond CO_2 capture, recent research shows the potential of PVC for CO_2 reduction and simultaneous dechlorination [205]. In a simulated aqueous environment containing HCO_3^- to replicate captured CO_2 in an alkaline solution, polyvinyl chloride (PVC) demonstrated the capability to efficiently convert hydrogen carbonate into formate at 300 °C. The mechanistic studies suggest that PVC experiences dechlorination and actively engages in redox reactions, leading to an impressive formate conversion of up to 16% and an almost complete dechlorination efficiency nearing 100%.

At last, polyethylene terephthalate (PET) is commonly utilized to produce activated carbon adsorbents due to the well-established infrastructure for recycling PET drink bottles. PET's high carbon content allows effective carbonization, and the presence of oxygen atoms creates polar sites in the resulting porous carbon materials, facilitating CO_2 physisorption. Activation with potassium hydroxide (KOH) is necessary to generate micro and mesopores in PET for efficient CO_2 adsorption at ambient conditions [206]. The activation process involves PET decomposition at a high temperature, releasing CO_2 and CO, which then react with KOH to form K_2CO_3. This compound further reacts with carbon precursors, enlarging the pores and improving CO_2 binding by introducing oxygen-containing groups. Activated PET adsorbents, analyzed for elemental composition, show increased oxygen content and enhanced CO_2 uptake capacity with a higher KOH-to-carbon mass ratio [207]. Nitrogen-containing groups, incorporated through urea during the KOH activation process, have been shown to further enhance CO_2 adsorption and selectivity in porous carbons derived from PET. These functionalized materials exhibit promising CO_2 adsorption capacities of 1.31 mmol g^{-1} at 303 K and 1 bar, with easy regenerability [207].

Literature data also point to poly(L-lactide) as a candidate for CO_2 capture as reported by Stelitano et al. [208]. Three PLA samples were prepared, differing in morphology commercial pellet (cPLA), powder (pPLA), and flakes (fPLA). The fPLA showed a CO_2 uptake of 3.59 mmol g^{-1} at 333 K and 15 bars. The enhanced CO_2 adsorption performance of the fPLA sample can be linked to its more disordered crystalline structure (pseudo-orthorhombic α' structure) and increased porosity.

Mohamed and coworkers reported the use of a high molecular weight PLA-b-PEO-b-PLA triblock copolymer for CO_2 capture [187] (Figure 11). The material served as the sole template for generating large mesoporous carbons, utilizing a resol-type phenolic resin as the carbon source. By subjecting the material to thermal curing and carbonization processes, large mesoporous carbons (>50 nm) were produced as well as a substantial surface area (>600 m^2g^{-1}). This outcome was attributed to the preference of the hydroxyl (OH) units in phenolics to interact with the poly(ethylene oxide) (PEO) block rather than the poly(lactic acid) (PLA) block. The polymer exhibited a CO_2 uptake of 5.22 mmol g^{-1} at 298 K and 1 bar.

Polybenzoxazines prepared from amino acids have been shown to possess CO_2 adsorption capacity [188]. The selected amino acids (γ-aminobutanoic acid, 6-aminohexanoic acid, L-lysinate, phenylalanine) were readily available and inexpensive. The first step was the preparation of the diphenolic acid methyl ester (MPD) by reacting the diphenolic acid with trimethoxymethane in the presence of *p*-toluenesulfonic acid as a catalyst. The amino acid methyl esters were prepared by reacting the chosen amino acid with TMSCl. The targeted polymers were obtained by reacting MDP with the amino acid intermediate in the presence of Et_3N and paraformaldehyde affording the monomers. These were then thermally polymerized at 120–240 °C. One of the polymers prepared this way, poly(E-lyme), showed a CO_2 adsorption of 5.8 mmol g^{-1} at 273 K and 1.06 bars (Figure 11).

5. Performances Comparison of Polymeric and Crystalline Materials

In order to give the reader a comparative overview of the polymeric materials discussed previously, Table 1 summarizes the performances of the materials discussed in Sections 3 and 4.

Table 1. Summary of polymer material performances.

Name	Type	T (K)	P (bar)	CO_2 Uptake (mmol/g)	Reference
TMU-42	MOF	—	—	7.29	[90]
COF-103	COF	—	35 bars	26.8 mmol CO_2	[112]
ZIF-69	ZIF	273	1 atm	83 L of CO_2/L	[104]
PEI-PDVB	PEI	298	—	3.70	[127]
TMPTA	PEI	348	—	0.685	[124]
ZN-1500	PANI	298	1.0	3.54	[125]
PIL	PILs	298	1.0	0.3	[129]
HPILs	PILs	298	1.0	1.47	[130]
BpP6-OTf-CMP-2	CMP	273	1.0	1.83	[133]
LKK-CMP-1	CMP	273	1.0	2.27	[134]
CMP-(bpy)20-Mn	CMP	298	1.0	1.06	[135]
CMP-YA	CMP	298	1.0	1.27	[136]
CTFs	CTF	273	1.0	3.00	[150]
cCTFs	CTF	273	1.0	0.93	[151]
CTF-FUM	CTF	298	—	2.55	[152]
Tt-POP-2	POP	273	1.0	1.79	[153]
TPOP-3	POP	273	1.0	4.09	[159]
PPN-81	POP	295	1.0	1.92	[154]
NUT-1	POP	273	1.0	1.87	[160]
NOP-53	NOP	273	1.0	2.27	[164]
NAP-1	NOP	298	1.0	2.25	[163]
NOP-51A	NOP	273	1.0	4.22	[165]
HAzo-POP	POP	273	1.0	1.68	[162]
BILB-101	POP	298	0.15	1.00	[161]

Table 1. *Cont.*

Name	Type	T (K)	P (bar)	CO_2 Uptake (mmol/g)	Reference
DHTP-Ea BZ	POP	298	1.0	3.29	[166]
CE-BZ-TPE-POP	POP	298	1.0	4.39	[167]
NP-NHC	NHC	298	1.0	0.78	[168]
NUT-21	NHC	273	1.0	8.3	[169]
POM-IM	IP	273	1.0	3.68	[170]
HIP	IP	298	1.0	2.1	[172]
PCP-Cl	IP	298	1.0	1.4	[173]
CB-PCP-1	IP	273	1.0	2.05	[174]
NSOPC-1	Biobased	298	1.0	3.88	[183]
HMF-GU-Ep/DFA	Biobased	298	—	2.15	[184]
Coconut shell	Biobased	272.85	2.5	3.60	[198]
Cherry stones	Biobased	322.85	2.5	2.50	[199]
HCP-PS	Biobased	273	1.13	1.987	[185]
PSNH2	Biobased	273	1.13	1.987	[203]
EDA-PVC/SBA-15	Biobased	298	—	0.5	[204]
PET	Biobased	303	1.0	1.31	[207]
fPLA	Biobased	333	15	3.59	[208]
PLA-b-PEO-b-PLA	Biobased	298	1.0	5.22	[187]
poly(E-lyme)	Biobased	273	1.06	5.8	[188]

Similarly, Figure 12 shows the performance of the most promising MOF materials in terms of their adsorption pressure and capacity, as discussed in Section 2.3.

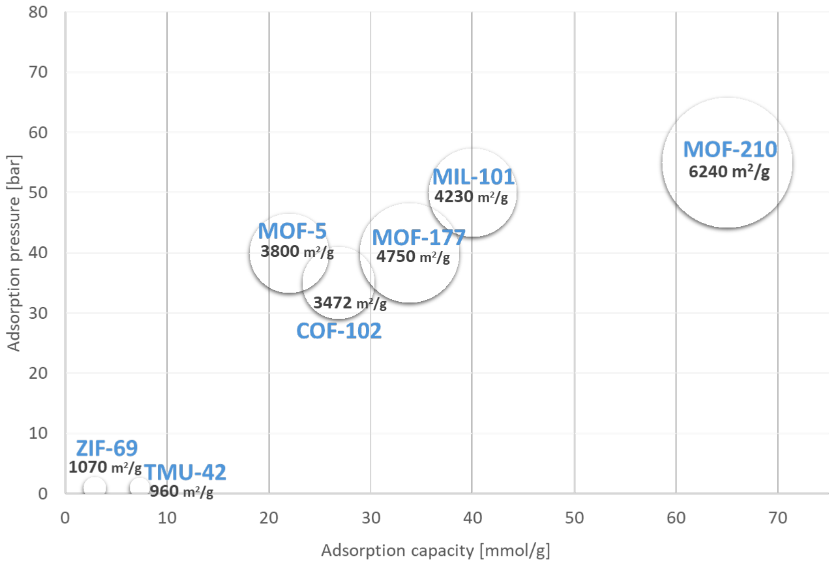


Figure 12. Carbon dioxide adsorption capacity measured at different CO_2 pressures and corresponding BET surface area of some promising candidates among metal–organic frameworks, covalent–organic frameworks, and zeolitic imidazolate frameworks [75,77,83,89,90,104,112].

6. Techno-Economic Comparison of CO_2 Capture Technologies

When developing a techno-economic analysis, the key aspects to take into consideration are (1) the principal obstacle to de-risk and enable the large-scale implementation of the technology, (2) the current cost drivers, and (3) the cost comparison of available technologies [209]. Herein, Table 2 presents the techno-economic comparisons of some of the key available technologies such as (1) amine scrubbing (monoethanolamine), post-combustion, (2) O_2/CO_2 recycle combustion (oxyfuel combustion), (3) Selexol, pre-combustion, (4) MOF-based mixed-matrix membranes for pre-combustion capture, and (5) MOF-based mixed-matrix membranes for post-combustion capture [210–214]. As seen, the TRL level of these technologies is mainly at 8–9, except for the MOF-based approaches. The energy demand [kWh/T CO_2] is similar across the different technologies, whereas the price per ton of captured CO_2 [US $/T] varies from 16.9 to 55.

Table 2. Techno-economic analysis comparison of CO_2 capture technologies.

Technology Name	TRL *	Energy Demand [kWh/T CO_2]	Price per ton of Captured CO_2 [US $/T]	Reference
Amine scrubbing (monoethanol amine), post-combustion	9	400–500	29–55	[210]
O_2/CO_2 recycle combustion (oxyfuel combustion)	9	100–200	35	[211]
Selexol **, pre-combustion	8–9	200–300	5–28.8	[212,213]
MOF-based mixed matrix membranes, pre-combustion capture	4	150	16.9	[214]
MOF-based mixed matrix membranes, post-combustion capture	4	240	30	[214]

* TRL stands for technology readiness level and measures the maturity of a technology from initial concept (TRL 1) to full deployment (TRL 9). ** Selexol processes use a physical solvent (Selexol) made of dimethyl ethers of polyethylene glycol to selectively remove H_2S/carbonyl sulfide and remove CO_2 in bulk from gas streams.

In terms of scalability, demonstrations at higher TRLs (6 and above) must be successful in order to de-risk the technology [215]. At the moment, only a limited number of technologies can operate outside the laboratory level (e.g., CO_2 mineral sequestration, post-combustion CO_2 capture, mesoporous silicas for adsorption processes) as shown in Table 2 [216–223]. On a practical level, several factors must be taken into account. First, it is important to point out that components used in the pilot plant for commercial set-up are general-purpose equipment or very specialized custom-built solutions [224]. Therefore, it is critical to develop and create specialized and integrated solutions between different unit operations, processes, and material supply chains. Attention must be put towards the material's performance (i.e., CO_2 capture, stability) in comparison to its cost. For each material and capture process, models must be generated considering parameters (performance, production cost, end-of-life) and be rigorously optimized, including investment and operating considerations.

7. Challenges and Future Perspectives

For critical industrial processes, such as gas separation and post-combustion capture, MOF-based membrane technologies offer a promising alternative to traditional energy-intensive unit operations. This is attributed to their remarkable adsorption capacity, thermal and hydrolytic stability, and chemical robustness. However, this technology requires further maturation and scaling up. Currently, the gas capture and separation markets are dominated by commercial polymeric materials and traditional scrubbers, most of which were developed before the 1990s. This highlights a stagnation in the development of new materials necessary to meet the increasing demands in this field. Metal–organic frameworks constitute exhibit significant potential to address the performance and stability limitations observed in many polymers under industrially relevant conditions [225,226]. Smith et al. brilliantly concluded that over the past 50 years, nearly 100,000 new MOF structures have been discovered, with 50% of these identified in the last 7 years alone, underscoring the remarkable and rapid advancement of this technology. [36]. Researchers are increasingly drawn to MOFs due to their precise tunability of pore dimensions, the ability to covalently decorate and exchange ligands, and their chemical and thermal stability [225]. These features enable exploration of the diverse architectural and chemical variability of MOFs, allowing for membrane performance metrics that surpass previously reported standards. Zeolitic imidazolate frameworks are particularly notable for their consistently high performance and excellent thermal stability, making them promising candidates for further investigation and large-scale applications [227].

Despite the numerous advantages of MOFs, the transition from laboratory research to industrial-scale application faces several significant challenges. Regardless of the discovery of nearly 100,000 MOF structures, scale-up precedents are scarce, hindering the broader adoption of MOF technologies. Cost efficiency remains a critical hurdle, as current production methods are not economically viable for large-scale operations [228]. Additionally, the fabrication of defect-free, continuous MOF films is still not well understood, limiting their practical use as a functional membrane. Furthermore, Guiver et al. highlight that the separation performance of COF and MOF membranes in the presence of detrimental impurities remains unexplored [229,230]. Hybrid MOFs and ZIFs, such as mixed-linker or mixed-metal MOFs, present an exciting area of research due to their potential for enhanced performance. However, the structure–activity relationships in these materials are poorly understood, complicating their design and optimization.

Concerning polymeric materials, the main challenge lies in the following aspects: selectivity and permeability, aging, thermal, and chemical stability, as well as scalability and cost [69,231]. It is important that the polymeric membrane can achieve a high selectivity for CO_2 over other gases such as N_2, CO, and CH_4 while maintaining a relative permeability [232]. In terms of aging, the polymeric membrane must be stable to light, moisture, and oxidation as it will decrease its performance over time [233]. Likewise, the thermal and chemical stability of the polymer is a critical parameter as it is required for industrial application, especially under harsh conditions [234]. At last, the most important parameters, the cost of the membrane fabrication and its corresponding production at a large scale, remain a huge barrier [235]

In spite of these challenges, a lot of efforts are put into the development of polymers. One interesting aspect that was not discussed in this review is the mixed-matrix membranes. They consist of MOFs/ZIFs or other inorganic material incorporated into polymer matrices in order to enhance their overall performance [35,236–239]. A second aspect is the recent improvement of new enhanced fabrication techniques (i.e., additive manufacturing, such as 3D printing) that can likely lead to the production of polymeric membranes with more precise and consistent properties [240,241]. Finally, the use of post-treatment methods such as cross-linking or thermal annealing to modify the membrane (surface) properties is another option to further increase its CO_2 capture capacity [242–244]. Addressing these challenges will necessitate a concerted effort in research and development to fully realize the potential of MOFs and polymeric materials in industrial applications.

8. Conclusions

In summary, this review provided an overview of the recent polymers (petroleum-based and bio-based) for the capture of CO_2. In particular, amine-based, ionic polymers, conjugated porous polymers, and porous polymer networks offer a variety of performances for CO_2 adsorption. In the same vein, bio-based polymers propose greener alternatives with comparable adsorption capacity.

The above classes of porous solid adsorbents propose great promise for CO_2 capture. However, challenges such as stability, scalability, and cost-effectiveness need to be addressed for widespread commercial adoption. Nonetheless, ongoing research and development efforts continue to improve the performance and viability, it is generally accepted that MOFs remain the leading candidates for mitigating greenhouse gas emissions from industrial sources.

In a broader perspective, the carbon capture technologies described in this review could play a critical role in achieving global climate goals and transitioning to a low-carbon future. As mentioned, there is an urgency to reduce carbon dioxide (CO_2) emissions to mitigate climate change, as well as to tackle challenges associated with decarbonizing various sectors, particularly those reliant on fossil fuels like power generation.

Carbon capture technology offers a transformative solution for heavy-emitting industries, such as power, steel, cement, oil, gas, chemicals, and transport, potentially reducing CO_2 emissions by over 95%. These sectors are pivotal in the energy transition, supplying essential resources while striving for lower-carbon outputs, crucial for renewable energy infrastructure like wind turbines and solar panels. Achieving net-zero emissions in these industries is feasible, as indicated by a 2019 IEA report projecting carbon capture technologies to contribute around 27% of necessary emission reductions in, for instance, the cement, iron, steel, and chemical sectors [245].

On the brighter side, these industries are, nowadays, more inclined to support and develop carbon capture initiatives as there are now several tax credit programs to further strengthen the viability of these new initiatives. For example, the United States passed the Inflation Reduction Act in 2022, and several countries, such as Canada, Australia, the Netherlands, the United Kingdom, and the European Union, have joined the effort and proposed similar programs. In summary, carbon capture stands poised to overcome financial challenges and offer a competitive pathway to achieving net-zero targets with robust industry involvement, government backing, and creative supply chain strategies. The outlook for this technology appears brighter than ever before.

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Abbreviations

The following abbreviations are used in this manuscript:

AIBA	2,2′-azobis[2-methylpropionamidine] dihydrochloride
$AlCl_3$	aluminium chloride
BDC	benzene-1,4-dicarboxylate
BTB	1,3,5-benzenetribenzoate
BZ	benzoxazine
CCS	carbon capture and storage
CCU	carbon capture and utilization
CH_4	methane
CO_2	carbon dioxide
CO2EXIDE	CO_2-based electrosynthesis of ethylene oxide
COFs	covalent–organic frameworks
CTFs	covalent triazine-based frameworks
DHPT	dihydroxyterephthalaldehyde
DMF	*N*,*N*′-dimethylformamide
EPS	expanded polystyrene

Et_3N	triethylamine
FCU	face-centered cubic
$FeCl_3$	iron (III) chloride
H_2O	water
HCl	hydrochloric acid
HCPs	hypercrosslinked polymers
HPILs	hypercrosslinked poly(ionicliquid)s
HUMs	hybrid ultramicroporous materials
IEA	international energy agency
Ils	ionic liquids
KOH	potassium hydroxide
MOFs	metal–organic frameworks
MOPs	microporous organic polymers
MPD	disphenolic acid methyl ester
N_2	nitrogen
$NaBH_4$	sodium borohydride
NHC	N-heterocyclic carbene
NOPs	nanoporous organic polymers
OVS	octavinylsilsesquioxane
PAFs	porous aromatic frameworks
PANI	polyaniline
PCN	porous coordination network
Pd	palladium
PDVB	poly(divinylbenzene)
PEI	polyethylenimine
PEO	polyethylene oxide
PET	polyethylene terephthalate
PILs	poly(ionic liquids)
PIMs	polymers of intrinsic microporosity
PLA	polylactic acid
POFs	porous organic frameworks
POPs	porous organic polymers
PPNs	porous polymer networks
PS	polystyrene
PVAm	poly-N-vinylformamide
PVC	polyvinyl chloride
SBUs	secondary building units
Sn	tin
TCM	2,4,6-tris(chloromethyl)-mesitylene
THF	tetrahydrofuran
TMPTA	N1-(3-trimethoxysilylpropyl) diethylenetriamine
TMSCl	trimethylsilyl chloride
TPA	triphenylamine
ZIFs	zeolitic imidazolate frameworks
$ZnCl_2$	zinc chloride

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