ISSN: 2710-0588

Study on the Removal of PFAS by Functionalized Porous Organic Polymers

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Abstract

Per- and polyfluoroalkyl substances (PFAS) have emerged as a globally significant new class of persistent organic pollutants due to their exceptional chemical stability, environmental persistence, and pronounced bioaccumulative toxicity. Among existing remediation technologies, adsorption is widely recognized as the most promising approach for treating trace and low-concentration PFAS in water bodies due to its high removal efficiency, low operational energy consumption, and ease of operation. Porous organic polymers (POPs), as an emerging platform material, offer an ideal molecular engineering platform for efficient PFAS capture due to their exceptional high specific surface area, precisely tunable pore size distribution, and outstanding physicochemical stability. This paper systematically reviews the design and synthesis strategies for representative POPs, including conjugated microporous polymers (CMPs), hypercrosslinked polymers (HCPs), intrinsically microporous polymers (PIMs), porous aromatic frameworks (PAFs), and covalent organic frameworks (COFs). It focuses on analyzing the intrinsic mechanisms by which functionalization with key groups such as amino and fluorine groups enhances adsorption capacity and selectivity, and delves into the synergistic enhancement processes of electrostatic forces, hydrophobic interactions, and fluorine-affinity interactions in complex aqueous environments. Furthermore, addressing current technical challenges such as the difficulty in removing short-chain PFAS and interference from competing ions in complex real-world aqueous matrices, this paper objectively evaluates the potential and limitations of POPs in practical engineering applications.

Keywords

PFAS, Porous Organic Polymers, Synthesis Mechanism, Functionalization, Adsorption Removal.

1. Introduction

Per- and polyfluoroalkyl substances (PFAS) are a class of man-made aliphatic compounds that are increasingly drawing global attention due to their impacts on the environment and human health[1]. PFAS consist of chains with varying carbon lengths, where at least one or all hydrogen atoms bonded to the carbon chain in the non-fluorinated portion have been replaced by fluorine atoms. Their chemical structure also includes a charged functional group, typically a carboxylic acid or sulfonic acid group attached at one end[2].Based on their terminal functional groups, PFAS can be classified into two types: perfluoroalkyl carboxylic acids (PFCAs) and perfluoroalkyl sulfonic acids (PFSAs)[3].Perfluoroalkyl substances such as perfluorooctanoic acid and perfluorooctanesulfonic acid (PFOS) exhibit persistence in typical environmental degradation processes and are ubiquitous in water[4]. They are difficult to degrade in the environment and may persist long-term in water, soil, and living organisms. Scientific research indicates that these compounds accumulate within living organisms and may pose potential

ISSN: 2710-0588

hazards to ecosystems and human health[5]. Currently, these substances are widely used in consumer goods and industrial applications. Certain specific uses may be critical to health, safety, or the functioning of modern society, and no alternatives have been developed to date[6]. Currently, most treatment methods for perfluoroalkyl substances suffer from poor degradation effectiveness and low degradation efficiency, necessitating highly efficient degradation approaches.

Among various technologies for removing PFAS from contaminated water-such as oxidation, ultraviolet irradiation, sonochemical methods, and electrochemistry-adsorption is highly favored for its simplicity and high efficiency[7]. Traditional PFAS adsorbents primarily include activated carbon (AC), ion exchange resins, minerals, molecularly imprinted polymers (MIPs), biosorbents, carbon nanotubes (CNTs), metal-organic frameworks (MOFs), and others[8]. However, these adsorbents typically exhibit certain limitations, including low capacity, prolonged equilibrium time, weak binding affinity, poor water/chemical stability, and low selectivity toward natural organic matter (NOM), which restricts their practical application in treating contaminated water [9].Considering these factors, porous organic polymers (POPs) can serve as a platform for constructing PFAS adsorbents due to their extremely high surface area, tunable framework structure, adjustable pore size, easily functionalized pore walls, and excellent hydrochemical stability[10].

Porous organic polymers (POPs) are a class of multidimensional porous network materials constructed through strong covalent bonds between organic building blocks of varying geometric shapes and topological structures. They represent an emerging field in recent porous material research[11]. Porous organic polymers offer advantages such as lightweight properties, high inherent porosity, excellent stability, and the ability to pre-design and tune their structure and functionality[12]. These materials hold immense application potential in gas storage/separation, multiphase catalysis, optoelectronic conversion, chemical and biosensing, energy storage, and conversion. Porous organic polymers primarily encompass hypercrosslinked polymers (HCPs), intrinsically microporous polymers (PIMs), conjugated microporous polymers (CMPs), and porous aromatic frameworks (PAFs), while crystalline persistent organic pollutants are represented by covalent organic frameworks (COFs)[13]. Functionalized porous organic polymers represent a class of highly structured and controllable materials. Their distinctive architecture is achieved through chemical synthesis by assembling specific organic molecules via chemical bonding into network or framework structures[14]. The pores in these materials can exhibit varying sizes and shapes at the nanoscale to micrometer scale. This tunable pore structure endows the materials with an enormous specific surface area[15]. The increase in specific surface area enhances adsorption performance, enabling outstanding performance in gas adsorption, separation, and catalytic reactions[16]. Due to their structural flexibility and tunability, functionalized porous organic polymers are regarded as cutting-edge materials with promising applications in energy, environmental, and medical fields[17]. With ongoing research into their preparation methods and properties, these materials are expected to play an increasingly significant role in the future.

2. Porous Organic Polymers

2.1. Conjugated Microporous Polymers

Conjugated microporous polymers (CMPs) represent a novel class of porous materials featuring an amorphous organic framework[18]. The unique feature of cmp lies in its high flexibility in component molecular design and pore parameter control[19]. It possesses a nanoscale pore structure, with these micropores providing an extremely large surface area that facilitates gas adsorption, separation, and catalytic reactions[20]. Polymer molecules contain conjugated structures internally, enabling free electron transfer within the material. This

ISSN: 2710-0588

property facilitates applications in electronic devices such as sensors and optoelectronic devices. The structure of CMPs can be tuned through various synthesis methods, including controlling pore size and increasing or decreasing conjugated structures, thereby exhibiting high chemical tunability[21]. Due to the high surface area and microporous structure of covalent microporous polymers, they can be used for the adsorption and storage of gases such as hydrogen and carbon dioxide, offering potential applications in gas separation. [22].CMPs exhibit excellent activity and selectivity in catalytic reactions and can be applied in fields such as organic synthesis and catalytic conversion[23].

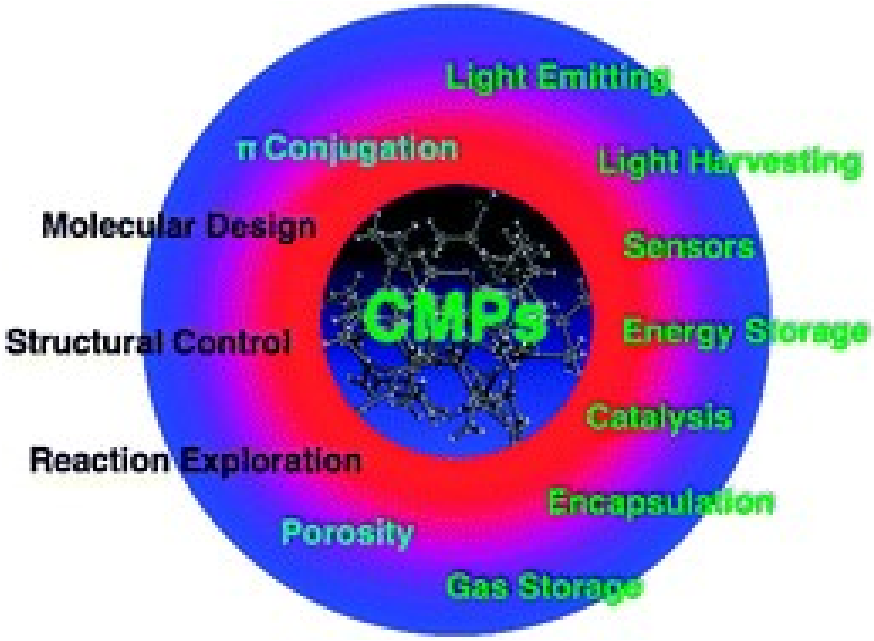

Figure 1. Design mechanism of CMPs

The first step in synthesizing CMPs is selecting appropriate monomers. These monomers typically comprise organic small molecules featuring double or multiple bonds, capable of forming polymer structures either independently or through reactions with other monomers. Monomers generally require polymerization via ligand chemistry, with one critical synthesis step being the formation of a microporous structure[24]. This is typically achieved by introducing voids or pores to form structural or functional groups, thereby endowing the polymer with pore sizes that increase its surface area and gas adsorption capacity[25]. The unique structure and properties of conjugated microporous polymers have made them a hotspot in materials science and energy research. Through synthetic design, researchers continue to explore new CMPs, hoping to unlock broader applications across various fields. Research has synthesized FCMP with large surface area, high porosity, and excellent chemical stability[26]. This marks the first application of the SPE method for PFASs. FCMP demonstrated higher recovery rates and adsorption capacity than CMP-2, indicating that fluorine enhances the concentration efficiency of PFAS in CMPs. Under optimal conditions, SPE coupled with HPLC-MS/MS exhibited a broad linear range, low detection limits, and outstanding accuracy and precision. Tighe et al. [27] synthesized a microporous polymer, PolyHIPE, which offers the advantage of high versatility and can be easily tailored for specific applications. Furthermore, PolyHIPE exhibits remarkably high adsorption capacity, reaching 300 mg/g at the highest analytical concentration of PFOA. This performance surpasses that of various PFAS remediation technologies, including activated carbon, metal-organic frameworks (MOFs), molecularly imprinted polymers, and other amino-based polymer adsorbents.

ISSN: 2710-0588

2.2. Hyper-crosslinked Polymers

Hyper-crosslinked polymers (HCPs) are a type of microporous polymer prepared by extensively crosslinking linear or mildly crosslinked precursor polymers[28]. They can be obtained through a two-step process or a one-pot method, starting from several aromatic monomers. Supercrosslinked polymers exhibit extremely high surface area and porosity, low density, outstanding adsorption properties, and high chemical and thermal stability[29]. The crosslinking degree in supercrosslinked polymers is exceptionally high, resulting in materials with high porosity and surface area. Due to their abundance of micropores and mesoporous structures, these pore systems facilitate the adsorption and storage of gases, solutes, and other substances[30]. Moreover, due to their porous structure, supercrosslinked polymers typically exhibit high adsorption capacity and can be used for adsorbing and separating molecules, organic substances, and other materials[31]. Under certain conditions, hypercrosslinked polymers can exhibit excellent chemical stability, enabling stable operation across diverse environments. Due to their high surface area and porous structure, these polymers are commonly employed in gas separation and storage applications. They also serve as effective adsorbents for removing organic pollutants, heavy metals, and other contaminants from water[32].

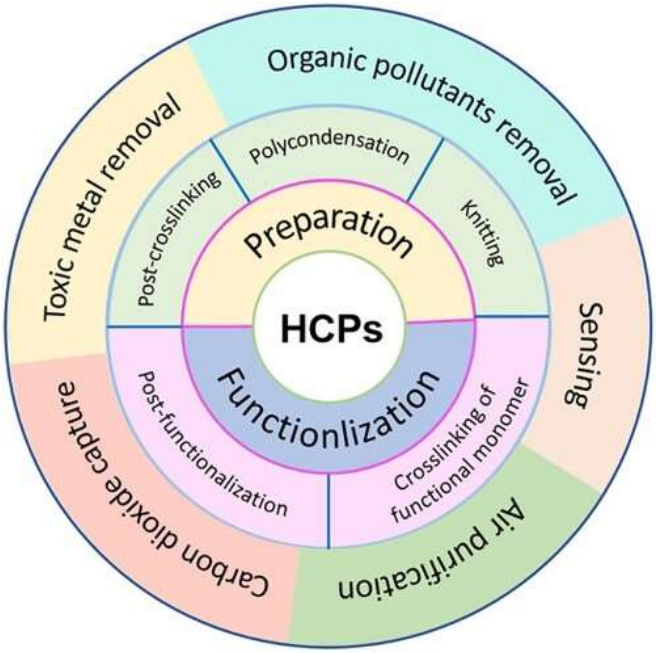

Figure 2. Preparation, Functionalization, and Applications of HCPs

The synthesis of HCPs primarily relies on Friedel–Crafts chemistry, which enables rapid kinetics to form strong bonds, resulting in highly crosslinked networks with dominant porosity. HCPs are mainly prepared through three approaches: (1) post-crosslinking of polymer precursors, (2) direct one-step polycondensation of functional monomers, and (3) weaving rigid aromatic building blocks with external crosslinking agents. The diversity of building blocks, coupled with expanded synthetic approaches, positions HCPs as a valuable platform for exploring novel organic porous materials with significant potential for addressing energy and environmental challenges. Post-synthesis characterization of the hypercrosslinked polymers, such as nitrogen adsorption-desorption, is essential to confirm their pore structure and specific surface area. Subsequently, they can be applied in fields including gas adsorption, separation, and catalysis. He et al. [33] synthesized and characterized four PANI crosslinked polymers with varying PFA ratios. Among these, PANI_PFA_2.4 exhibited the highest adsorption capacity and

ISSN: 2710-0588

PFOA affinity, both determined by the crosslinker's balancing effect in increasing specific surface area (SSA) and reducing surface charge. In the presence of multicomponent PFAS mixtures, both PANI and PANI_PFA_2.4 exhibited enhanced adsorption capacity for perfluorosulfonates compared to perfluoroacetic acids (containing the same fluorocarbon content), with adsorption increasing with longer fluorocarbon chain lengths. However, crosslinked PANI_PFA_2.4 exhibits lower removal efficiency for all PFAS species in multicomponent mixtures compared to others. This is attributed to the narrower pores in the crosslinked polymer being blocked by preferentially adsorbed long-chain PFAS.

2.3. Polymers of Intrinsic Microporosity

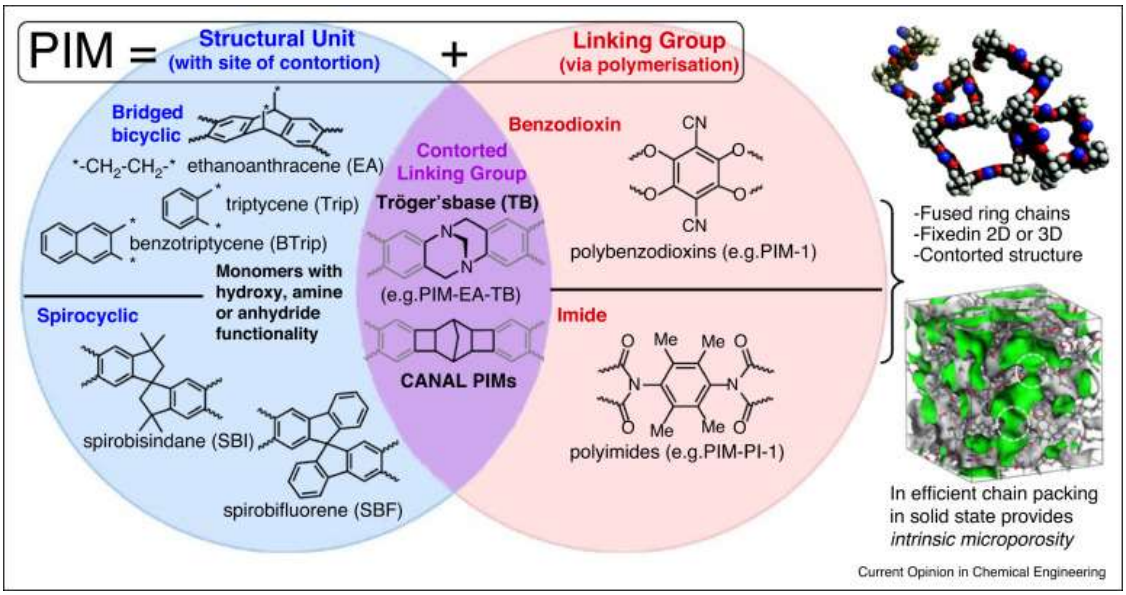

Figure 3. Main Structure and Preparation Mechanism of PIM

Polymers of intrinsic microporosity (PIMs) Microporous materials are defined as solids containing interconnected pores smaller than 2 nanometers. The formation of stable micropores within polymers requires overcoming cohesive forces between different chains (or adjacent segments of the same chain) to prevent their collapse [34]. Utilizing monomer rigidity and distorted spatial configurations (such as screw-ring structures) prevents polymer segments from packing tightly, thereby creating "awkward" non-efficiently filled spaces that generate inherent free volume. Most polymers possess sufficient flexibility and rotational freedom to rearrange, maximizing cohesive interactions and preventing micropores[35]. Therefore, they possess large and accessible surface areas, typically ranging from 300-1500. $m^2 \cdot g^{-1}$[36]. Over the past several decades, international efforts have focused on optimizing traditional microporous materials such as zeolites (aluminosilicates) and activated carbon for specific applications in adsorption, separation, and heterogeneous catalysis. Newly cast films of intrinsically microporous polymers represent the only polymeric materials without a network structure that have been identified as microporous materials, exhibiting exceptionally high gas permeability [37]. The permeability of aged PTMSP membranes is significantly lower than that of freshly prepared membranes, with ultimate losses reaching up to 90% of the initial permeability. This indicates that the microporosity of PTMSP is transient rather than an inherent property of the polymer[38]. Research confirms that, like all vitreous polymers, the gas permeability of PIM-1 (and all other PIMs) films also decreases with aging, although at a slower rate than PTMSP[39]. However, the formation of PIM micropores is clearly intrinsically linked to its macromolecular structure. The inherent micropores of polymers are defined as interconnected, continuous networks of molecules, whose formation is a direct result of the shape and rigidity of the constituent macromolecules[40]. Generally, polymers fill space to

ISSN: 2710-0588

maximize attractive interactions between constituent macromolecules, thereby minimizing the amount of void space. Most polymers possess sufficient conformational flexibility to rearrange their shapes, maximizing intermolecular cohesion and efficiently filling the available space[41]. The method for maximizing intrinsic microporosity involves designing polymers with highly rigid and twisted molecular structures that create "awkward" macromolecular shapes incapable of efficiently filling spaces. Specifically, due to their fused ring structures, PIMs lack rotational freedom along the polymer backbone. This ensures macromolecular components cannot rearrange their conformations, thereby locking their highly twisted shapes during synthesis [42]. Ghanem[43] et al. reported the synthesis and characterization of intrinsically microporous network polymers derived from tert-butoxypropane monomers with alkyl groups attached at bridgehead positions. They investigated the hydrogen adsorption capacity of these polymers. Among PIMs based on tert-butoxypropane, those containing methyl or isopropyl substituents exhibited hydrogen adsorption capacities at the highest level among pure organic materials with low to moderate adsorption strengths.

2.4. Porous Aromatic Frameworks

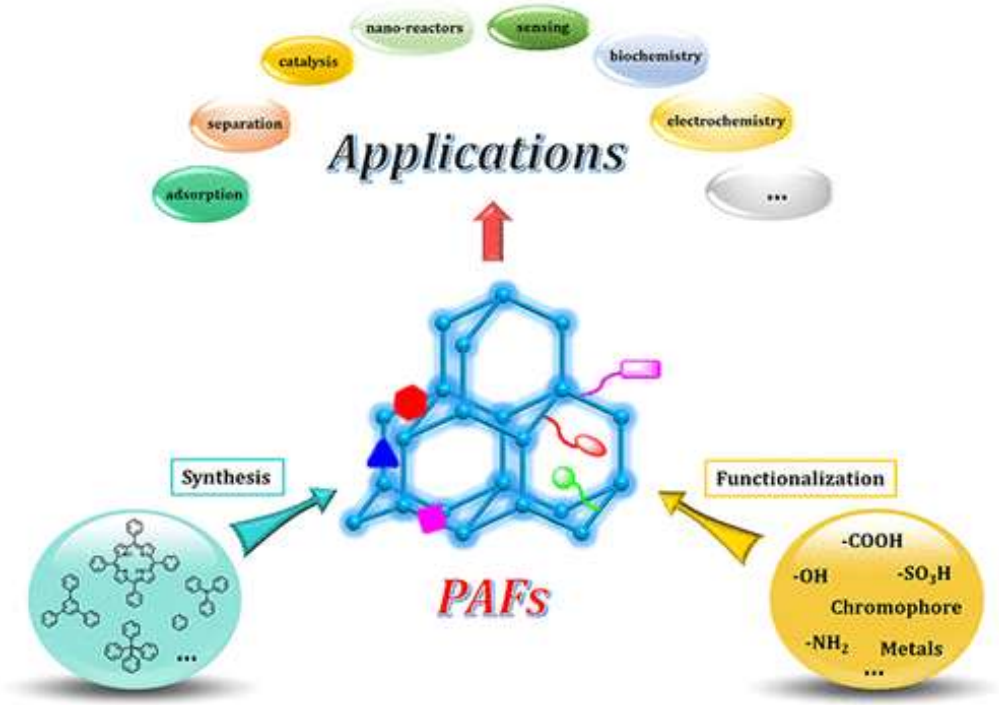

Figure 4. Functionalization of PAFs and Their Applications

Porous aromatic frameworks (PAFs) represent a significant class of porous solids. PAFs feature rigid frameworks and exceptionally high surface areas. Moreover, uniquely, they are composed of aromatic building blocks connected by carbon-carbon bonds [44]. Various functionalities can originate from the intrinsic chemical properties of their building blocks or be achieved through post-modification of aromatic motifs using established reactions. Notably, strong carbon-carbon bonds enable PAFs to remain stable under harsh chemical treatments. Consequently, PAFs exhibit chemical and functional specificity compared to traditional porous materials such as zeolites and metal-organic frameworks. The unique properties of PAF enable it to withstand harsh environments and facilitate functionalization through demanding chemical treatments. Over the past decade, the field of PAF research has advanced rapidly. As a promising porous material, it has garnered significant attention in recent years due to its high surface area and excellent mechanical properties. Several advanced and efficient methods have been developed for constructing PAFs, including boronic acid condensation reactions, dibenzodioxane

ISSN: 2710-0588

formation reactions, palladium-catalyzed Sonogashira-Hagihara cross-coupling reactions, palladium-catalyzed Suzuki cross-coupling reactions, nickel(0)-catalyzed Yamamoto-type Ullmann cross-coupling reactions, and the trimerization of aromatic nitrile compounds. However, their application is constrained by limited pore volume and broad pore size distribution[45].

The fundamental structure of PAFs consists of aromatic building blocks that connect in specific ways to form three-dimensional frameworks. This architecture endows PAFs with enormous specific surface area and pore volume, enabling potential applications in gas adsorption, mixture separation, and gas storage[46]. PAFs typically feature tunable pore sizes and chemically functional groups, endowing them with broad application prospects across diverse fields such as gas separation, catalysis, drug delivery, and energy storage[47]. The design and synthesis methods of these materials have garnered significant attention in the fields of materials science and nanotechnology, as their unique structures and properties hold great promise for addressing challenges in energy and environmental sectors. The structure of PAFs can be precisely controlled through adjustments to synthesis methods, including pore size, surface area, and pore distribution[48]. The core advantage of PAFs lies in their high degree of customizability: researchers can leverage the inherent chemical properties of building blocks for fundamental design, or perform post-modification of aromatic units through established chemical reactions, thereby achieving precise control over pore structure, surface charge, and chemical affinity[49]. This highly controllable structural property enables PAFs to be functionally tailored for specific requirements, such as selective separation of particular gases or multiphase catalysis. With their tolerance in extreme environments and outstanding physicochemical properties, PAFs demonstrate immense application potential across multiple fields including gas storage and separation, multiphase catalysis, drug delivery, and energy storage. Furthermore, through the development of sustainable synthesis pathways, PAFs serve as highly efficient alternative materials, offering promising green technological solutions to address current challenges in environmental purification and energy conversion. Overall, as a novel porous material, PAFs have garnered significant attention in materials science and engineering due to their highly controllable structure, outstanding performance, and broad application prospects. Pezoulas et al.[50]employed post-synthetic modification of a porous polymer network solid (PPN-6, also known as PAF-1) by introducing multiple chemical functional groups to synthesize PPN-6-FNDMB. This material exhibits unique binding interactions with PFAS molecules through electrostatic forces, hydrogen bonding, hydrophobic interactions, and fluorophilicity. It demonstrated superior PFAS adsorption performance compared to other PPNs and commercial adsorbents; however, concerns remain regarding the application of fluorinated adsorbents in water treatment systems.

2.5. Covalent Organic Frameworks

Covalent organic frameworks (COFs) represent one of the most significant dynamic members among porous organic materials. They are constructed through reticular chemistry, wherein structural units are linked by covalent bonds[51]. Due to their attractive properties, such as large surface area; structural versatility; ease of surface modification; and high chemical stability[52], catalysis; sensing; adsorption; gas storage; and many other fields where it finds valuable applications. COFs, as crystalline porous organic polymers, are formed through reversible reactions [53], condensation of two-dimensional (2D) and three-dimensional (3D) organic building precursors. They are composed of light elements (such as H, B, C, N, O) and are connected through robust covalent bonds via reticular chemistry.

ISSN: 2710-0588

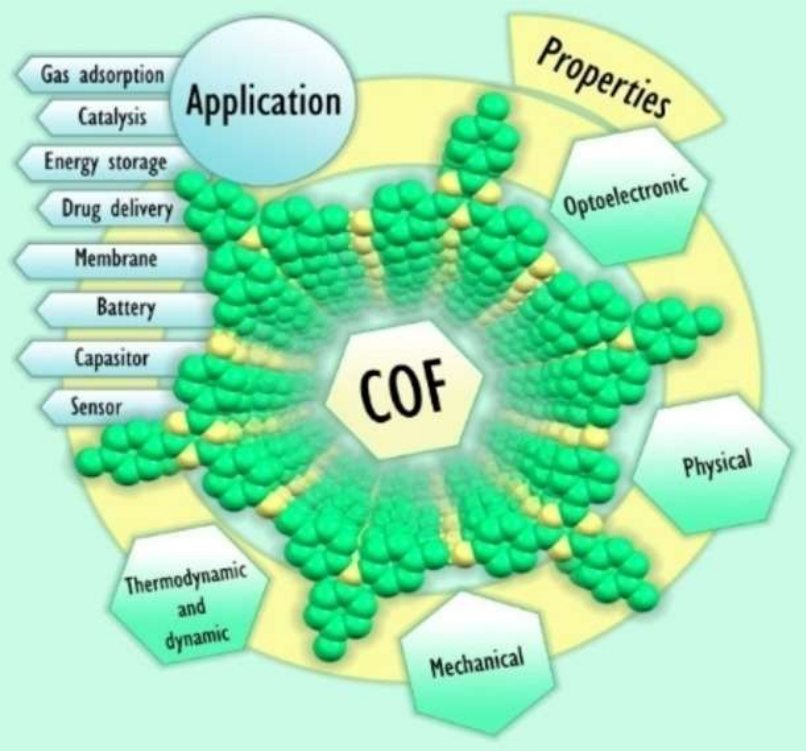

Figure 5. Applications and Advantages of COF

The remarkable progress in COFs is primarily attributed to their self-healing capabilities and the thermodynamically controlled dynamic covalent chemistry involved in their assembly, which results in long-range ordered crystalline structures [54]. COFs typically exhibit low density, thereby demonstrating exceptional stability in organic solvents and even under varying acidic conditions. They can withstand harsh environments while maintaining their ordered structure and crystallinity[55]. This is because the metal-free framework structure is connected via strong covalent bonds rather than the coordination bonds that form MOFs. Furthermore, the presence of hydrogen bonds and p-p stacking interactions within COFs further strengthens the framework and pore structure, protecting them from solvation and hydrolysis effects[56]. Compared to inorganic zeolites and porous silica materials, COFs exhibit higher porosity and larger, tunable pore sizes, accelerating reactant diffusion and product desorption to enhance yield and selectivity. Furthermore, COFs possess ultra-high theoretical specific surface areas and high chemical stability, making them ideal multiphase catalysts [57]. Wang et al.[58] synthesized five COFs with distinct pore sizes and investigated their adsorption performance toward six PFAS with varying C–F chain lengths. The PFAS adsorption capacity on all five COFs increased with longer C–F chain lengths. The optimal pore size range was determined to be 2.5–4.0 times the PFAS chain length for achieving maximum adsorption capacity. Molecular dynamics simulations revealed that larger COF pore sizes enhanced PFAS diffusion into the COF pores. However, excessively large pores weakened the adsorption binding energy between the COF framework and PFAS molecules, leading to reduced PFAS adsorption efficiency. Therefore, achieving efficient removal of PFAS from water by adjusting the pore size and surface hydrophobicity of COFs is highly feasible.Amin et al. [59]synthesized covalent organic framework nanospheres-MELEM-COF and MEL-COF-using anthraquinone. These nanospheres exhibit highly crystalline characteristics and demonstrate exceptional affinity for multiple anionic PFAS compounds, enabling simultaneous removal of various pollutants within 30 minutes. Removal efficiencies of 90.0–99.0% were achieved for all six anionic PFAS compounds, with a maximum adsorption capacity of 2500 mg/g-surpassing all

ISSN: 2710-0588

previously reported COFs. Furthermore, MELEM-COF exhibited rapid exchange kinetics, reaching equilibrium within 30 minutes.

2.6. Macromolecular Polymers--Hydrogels

Hydrogels are three-dimensional (3D) networks of hydrophilic polymers that absorb large amounts of water, forming viscoelastic, crosslinked materials with a network structure induced by crosslinking agents and solvents[60]. Due to their ability to alter shape and volume in response to external stimuli, hydrogels have been extensively studied for diverse applications spanning biology to environmental science. In recent years, hydrogel materials have become ubiquitous in daily life, appearing in various forms tailored to specific functionalities-such as soaps, shampoos, toothpaste, hair gel, and contact lenses-while also finding significant industrial applications in oil extraction, pharmaceuticals, agriculture, textiles, and water treatment[61]. Therefore, gel materials have become one of the most popular materials in our daily lives. Compared to traditional methods, hydrogels offer several advantages[62]. First, they exhibit hydrophilicity, slightly higher than water. Second, monomers with the desired functionality can be readily used for direct synthesis, eliminating the chemical processing required to introduce functional groups in conventional methods. Third, they provide more sites per unit volume by adsorbing pollutants into their 3D networks. Fourth, hydrogels can be synthesized with controlled size, charge, and functional groups. Fifth, polymer hydrogels are environmentally friendly, biocompatible, and easy to handle. Finally, hydrogels are reusable if efficient desorption processes are developed[63]. Huang et al. [64] developed a reusable hydrogel adsorbent for removing long-chain and short-chain perfluoroalkyl acids, as well as 2,3,3,3-tetrafluoro-2-(heptafluoropropoxy)propanoic acid (GenX). Among these, amino-functionalized PEGDA exhibited the highest adsorption capacity for all five PFAS compounds. Both amino-functionalized and bifunctional adsorbents could remove GenX from water, achieving maximum adsorption capacities of up to 98.7 μmol/g.

3. Conclusion

Although functionalized porous organic polymers demonstrate outstanding adsorption capacity in laboratory pure water systems, their application in complex real-world wastewater faces multiple competitive interferences and engineering challenges. In real-world aquatic environments (such as groundwater, surface water, or industrial wastewater), adsorption efficiency is constrained by multiple factors, including competition from inorganic ions and the influence of natural organic matter: High concentrations of coexisting anions (e.g., SO_4^{2-}, Cl^-, HCO_3^-) compete with anionic PFAS for active sites such as quaternary ammonium groups or amino groups on the polymer backbone, resulting in a significant electrostatic shielding effect. Significantly reduces equilibrium adsorption capacity. Macromolecular organic compounds such as humic acid (HA) interfere with adsorption through two mechanisms: pore blockage by occupying microporous structures and competition for nonpolar adsorption sites via hydrophobic interactions. Effective removal of short-chain PFAS represents the greatest challenge currently facing adsorption technologies. Traditional polymer frameworks relying solely on hydrophobic interactions struggle to overcome the desorption tendency caused by the high water solubility of short-chain PFAS.

Functionalized porous organic polymers (F-POPs) have emerged as one of the most promising technological platforms for addressing perfluoroalkyl and polyfluoroalkyl substances (PFAS) contamination in water treatment. This review systematically outlines the design principles of various F-POPs-ranging from hypercrosslinked polymers (HCPs) and covalent organic frameworks (COFs) to functionalized hydrogels-alongside representative materials for PFAS removal. Research indicates that precisely introducing quaternary ammonium cation sites and fluorinated side chains into polymer backbones through molecular engineering can

ISSN: 2710-0588

significantly overcome the bottlenecks of weak adsorption and poor selectivity for short-chain PFAS (e.g., PFHxA, PFBS) inherent in traditional adsorbents. Despite breakthroughs at the laboratory stage, practical application of F-POPs faces constraints: 1) Mass transfer resistance and kinetic bottlenecks, requiring further optimization to balance porosity and swelling rate; 2) Cost pressures from high-performance monomers, limiting large-scale material production.

To advance the practical implementation of functionalized porous organic polymers in PFAS remediation, future research should focus on: 1) Avoiding simple phase transfer of pollutants by utilizing POPs as catalytic centers or carriers. Combining photocatalysis, electrocatalysis, or persulfate activation technologies enables in-situ complete mineralization of C-F bonds after adsorption and enrichment, eliminating secondary pollution risks. 2) Further explore interactions beyond electrostatic and fluorine affinity (e.g., anion-π interactions, multi-hydrogen bond networks) to construct more specific "molecular beacon"-type adsorption sites. 3) Establish a systematic economic and environmental assessment framework to quantitatively analyze the carbon footprint across material synthesis, application, regeneration, and final disposal, while developing more sustainable green synthesis routes (e.g., aqueous synthesis, solvent-free methods).

In summary, functionalized porous organic polymers offer highly competitive tools for tackling global PFAS pollution challenges due to their "customizable" structural properties. With the deep integration of materials science and environmental engineering-particularly through synergistic optimization of diffusion and affinity via hydrophilic/fluorinated bifunctionalization strategies-we have reason to believe that next-generation high-performance POPs adsorbents will play an irreplaceable role in safeguarding global drinking water safety and ecological restoration.

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