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# Advances and Prospects in Multifunctional Composite Fibrous Materials Utilizing Porous Organic Polymers

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**Porous organic polymers (POPs) offer exceptional surface area, tunable pore sizes, and versatile chemical functionality, making them attractive for a range of advanced applications. However, their conventional powdered form limits processability, structural integration, and practical deployment. Integrating POPs into fibrous matrices through electrospinning, a scalable and versatile technique for producing nonwoven fibers, helps overcome these limitations and enables the creation of new material architecture. The resulting POP-incorporated fibrous materials (POP-FMs) combine the intrinsic advantages of POPs with enhanced mechanical integrity, tailored surface properties, and improved mass transport characteristics. These features expand the potential of POP-FMs in areas such as catalysis, environmental remediation, sensing, and biomedical fields. This review discusses recent progress in the design and synthesis of electrospinning-compatible POPs, strategies for fabricating POP-FM composites, and the structure–property relationships that govern their performance. Key challenges and future directions are also explored, underscoring the potential of POP-FMs as next-generation functional materials.**

## 1. Introduction

### 1.1. Structure–Property Relationships in Porous Organic Polymers

Nature provides numerous examples of advanced materials with intricate structures and functions, such as hexagonal honeycomb cells, neuronal networks, feathers, lotus leaves, and caterpillar hairs.[1–7] These natural architectures serve as profound sources of inspiration for material design across various disciplines. The hierarchical structures inherent in these materials play a crucial role in enhancing their performance. Unlike conventional materials with uniform structures, composite materials featuring hierarchical pore structures can achieve synergistic effects across different organizational levels.[8–11] This synergy leads to improvements in properties such as high surface area, efficient mass transfer, responsiveness to stimuli, mechanical strength, and physicochemical stability. These enhanced characteristics expand their potential applications in areas such as adsorption, separation, catalysis, bioengineering, and energy-related processes. In the case of biomimetic natural composite materials with multilayer hierarchical pore structures, maintaining uniformity while preserving porosity is essential for optimizing their functionality.

Porous organic polymers (POPs) have attracted significant attention due to their intrinsic properties, including high Brunauer–Emmett–Teller (BET) surface area, tunable pore sizes, and customizable surface chemistry. These attributes enable the precise tailoring of functionalities to suit specific applications.[12–16] Recent advancements have broadened the spectrum of POPs, categorizing them into soluble polymers, such as polymers of intrinsic microporosity (PIMs).[17,18] and insoluble polymers, including hypercross-linked polymers (HCPs),[19,20] conjugated microporous polymers (CMPs),[21–23] porous aromatic frameworks (PAFs),[24–26] and crystallizable covalent organic frameworks (COFs).[27–30] The diversity of these materials, coupled with their self-assembly capabilities, positions POPs as versatile candidates for various applications. However, despite their diverse nano- and microscale morphologies, assembled POPs typically form heterogeneous solid powders, which pose challenges in handling transportation, and surface accessibility, potentially limiting their practical applications.

Within this framework, a key objective in recent research is the precise construction of POPs with controlled morphologies. These structures range from nanoscale objects, such as nanosheets, nanoparticles, nanospheres, and nanofibers, to macroscale films and freestanding objects, unlocking new possibilities for customized applications. Particularly noteworthy is

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the 1D nanofibrous morphology, which not only preserves beneficial nanoscale effects but can also be assembled into macroscale films or composite structures.[31,32] Furthermore, 1D nanofibers offer advantages such as ease of preparation, lightweight characteristics, high surface contact area, and efficient diffusion into their interior.[33]

To address these challenges, integrating bottom-up and top-down approaches can significantly enhance the structural and functional properties of materials. Material molding technology plays a vital role in large-scale production by ensuring uniformity and consistency. Additionally, it facilitates the integration of various materials, enhancing their synergistic properties and resulting in unique characteristics. Among the various methods for fabricating such nanostructured materials, electrospinning has emerged as a highly versatile and widely adopted technique. Based on our literature review, while melt-blowing, wet-spinning, and 3D printing have also been reported for fabricating porous nanofibers, electrospinning remains the predominant technique for fabricating POP fibers. For instance, Ellison et al. demonstrated the use of melt-blown porous fibers for co-continuous polystyrene/high-density polyethylene polymer blends.[34] Separately, Ke et al. reported the 3D printing of homogeneous and heterogeneous covalent organic frameworks.[35] Each alternative method offers distinct advantages: melt-blowing is valued for its high throughput and low cost, wet-spinning excels at producing strong, continuous filaments for textiles, and 3D printing allows for unparalleled macroscale geometric complexity and mechanical strength. However, due to the clear predominance of electrospinning in POPs fiber nanofabrication, this review will focus specifically on electrospun POP-based composite fiber materials.

### 1.2. Electrospinning: A Key Technique in Morphological Transformation to Functional POP-Fiber Materials

Electrospinning, a textile technique, utilizes electrostatic forces to produce fibrous materials with diverse morphologies in a single-step and straightforward manner.[4,36,37] While electrospinning has been established for many years, recent advancements have expanded material selectivity, diversified fiber structures, and improved both equipment and processes.[38,39] The field of electrospinning has advanced significantly since its inception in 1938, with commercial applications like Donaldson Company's air filters emerging in the 1980s. Currently, numerous organic polymers can be electrospun into complex nanostructures (e.g., core–sheath, hollow, aligned), and the technology has even been scaled for introductory production (**Figure 1**). However, conventional electrospun solid polymeric fibers often suffer from limitations such as low BET surface area, constrained structural hierarchy, and reduced functional performance. Consequently, exploring the electrodynamic processability of POP-based fiber materials is essential for improving their properties and broadening their potential applications.[40,41]

Recent interest in POP-FMs has surged due to their versatile synthesis methods, including electrospinning, extrusion shaping, and post-forming technologies.[42,43] These techniques significantly enhance the applicability of POP-FMs across various fields. For example, the inherent porosity of POPs facilitates drug loading; however, their powdered form restricts their biomedical use, such as in wound dressings.[44,45] Electrospinning can transform these properties into a more suitable format for such applications. In catalysis and adsorption applications, the surface-functionalized groups and inherent porosity of POPs offer significant advantages. However, their powdered form hinders recyclability and reuse. Incorporating POPs into electrospun composite fiber membranes not only improves recyclability but also mitigates clogging issues. Furthermore, the development of multifunctional smart separators is crucial for advancing fast-charging and safe battery technologies.[46] Traditional fiber membranes often lack the necessary hierarchical pore structures, but POP-FMs offer improved regulation of lithium-ion transport,[47] thereby enhancing battery performance.

Given the rapid evolution of technologies related to POPs and electrospinning, a systematic review of recent advancements is both timely and essential. In this review, we first introduce POPs and emphasize their potential integration with electrospinning technology. We then discuss the benefits of electrospinning as a reliable supporting technology for composite design, highlighting how it facilitates the creation of materials with controlled morphologies and enhanced properties. Following this, we explore the emerging fabrication methods for achieving morphologically controlled POP-FMs. These methods have produced POP-FMs with tunable pore sizes, varied morphologies, lightweight characteristics, adjustable surface functionalities, good surface contact, and reduced diffusion resistance. We investigate the importance of pre-selection processes that align with the processing capabilities of POPs and the selection of appropriate electrospinning methods to optimize the properties and applications of the resulting composites. Furthermore, we examine the significant applications of POP-FMs in fields such as biomedicine, energy, catalysis, environmental protection, and gas sensing. Additionally, we discuss emerging research areas, including the development of POPs with enhanced properties, the exploration of emerging applications beyond traditional uses, and the potential for creating multifunctional composite materials for advanced applications. These aspects underscore the continuous innovation and expanding potential of POP-FMs.

## 2. Benefits of Porous Organic Polymers (POPs)-Based Nanofiber Materials

### 2.1. Brief Summary of POPs Materials

POPs can be broadly classified into two main categories based on their structural organization: amorphous and crystalline (Figure 1a). In the amorphous category, the lack of long-range ordered structures, as seen in HCPs, CMPs, PIMs, and PAFs,[12,48,49] results in porosities generated by the random arrangement of polymer chains, forming a network of pores. While polymerization in these materials typically does not produce a regular, repeating structure, each class exhibits distinct structure and unique functional characteristics. For example, HCPs consist of highly cross-linked rigid polymers,[19,20] CMPs feature conjugated structures,[21] PIMs exhibit unique solubility properties, and PAFs boast a BET surface area exceeding 6000 $m^2\ g^{-1}$.[25] Conversely, COFs, a type of POPs, have garnered significant research interest due to their highly ordered, repeating structures


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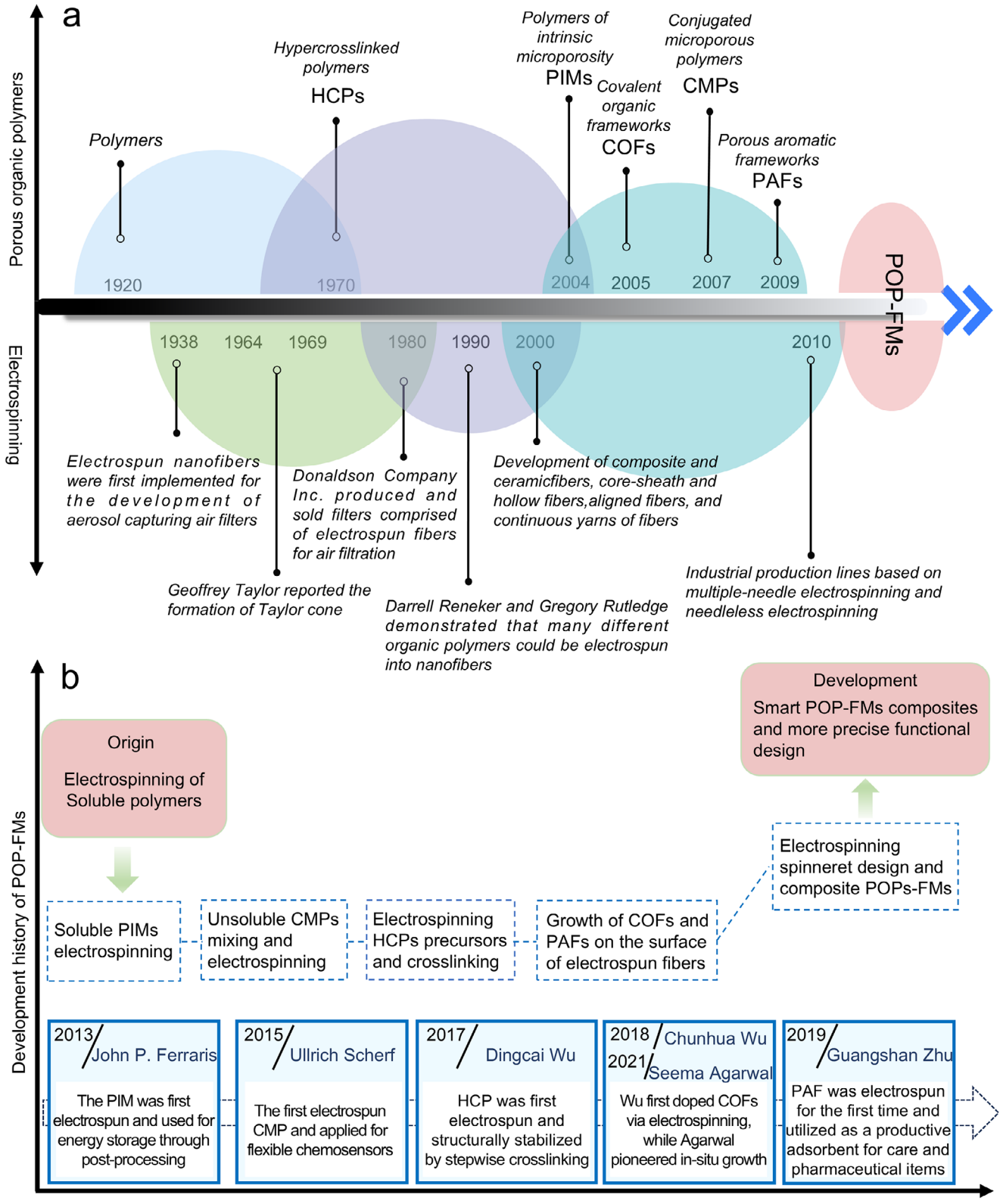


**Figure 1.** a) An outline of the evolution of representative POPs and the historical development of electrospinning. b) The development history of POP-FMs.



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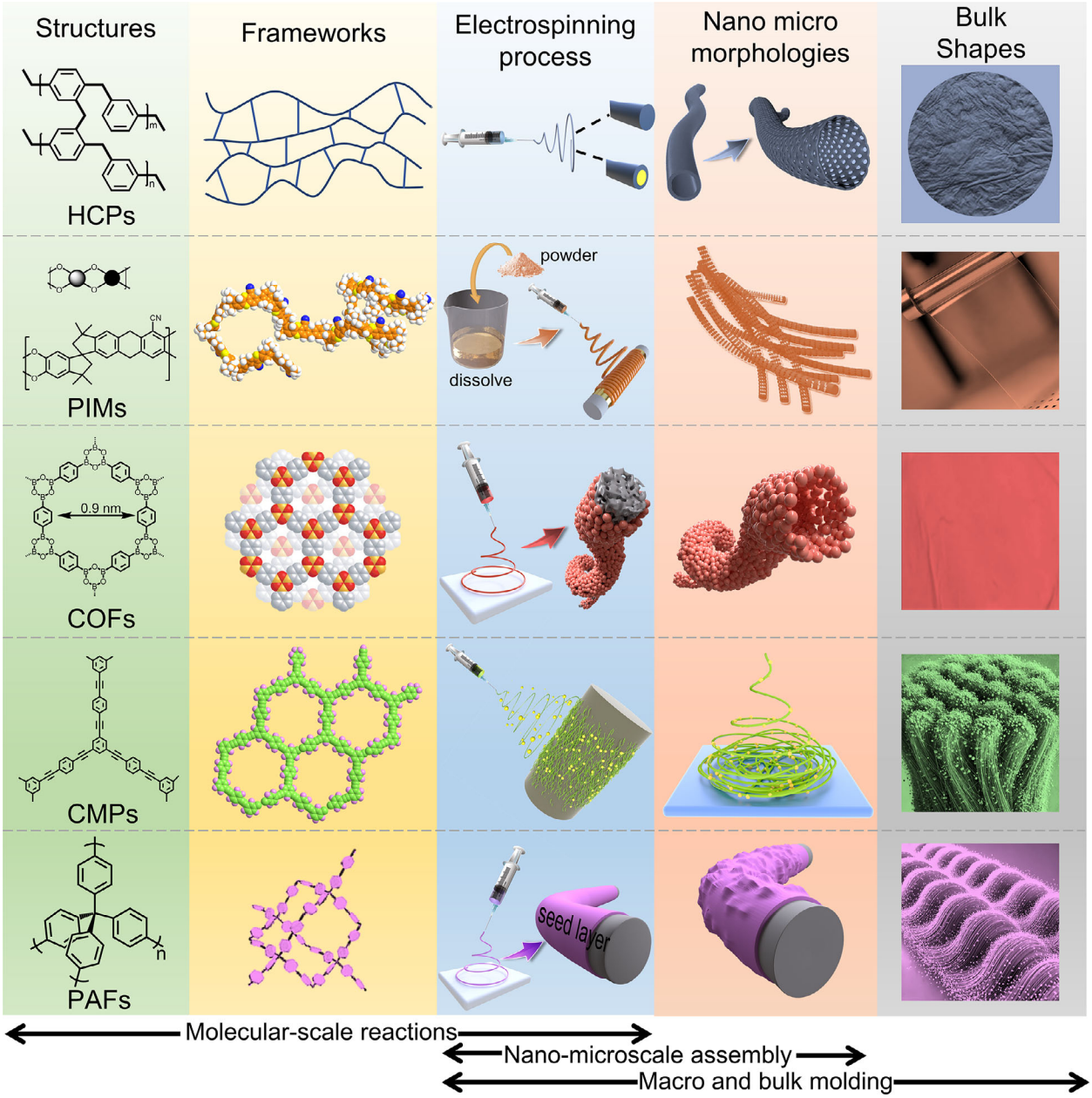


**Figure 2.** Molecular-scale reactions, nano- to micro-scale assembly through chemical reactions, and macro/bulk molding via electrospinning for the fabrication of representative POP-FMs.

at the molecular level.[50] These distinct characteristics enhance their compatibility with electrospinning technology,[4] offering a broad range of strategies to tailor the components, compositions and deposition patterns for targeted functional applications (Figure 1b).

### 2.1.1. Hypercross-linked Polymers (HCPs)

As shown in **Figure 2**, HCPs are a class of nanoporous materials with diverse practical applications, first introduced by Davankov and Tsyurupa in the early 1970s (Figure 1a).[6,51] Compared to other POPs, HCPs are primarily microporous and possess a high surface area, making them particularly suitable for small molecule adsorption and separation applications. They often exhibit good thermal and chemical stability due to their highly cross-linked structure. Moreover, HCPs offer several additional advantages, such as diverse synthetic methods, easy functionalization, low reagent price, and mild operating conditions.[52,53] By carefully selecting the length of the monomer, the appropriate cross-linking agent, and optimizing reaction conditions, a well-developed polymer framework with an adjusted porous topology can be produced.[54] To date, HCPs have predominantly been synthesized using Friedel-Crafts chemistry, employing


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post-cross-linking precursors, one-step self-condensation, or external cross-linking strategies.[55–58]

The production of electrospun HCP fibers via stepwise cross-linking was first demonstrated by Dingcai Wu et al. in 2017 (Figure 1b).[59] The stepwise cross-linking strategy for electrospun nanofibers typically involves multiple steps: initial electrospinning of a precursor solution, followed by the introduction of additional cross-links—either via reactive sites in the polymer or through a loosely pre-cross-linked nanostructure. Subsequent solution polymerization or heat treatment enhances cross-linking density and refines pore structure, improving material stability and mechanical properties.[19,60–62] HCPs nanoparticles also can be incorporated into electrospun fibers as functional fillers or main components. Dispersed in the polymer solution, they are electrospun into nonwoven membranes, introducing hierarchical porosity and tunable performance depending on particle type and content. However, conventional powdered HCPs tend to aggregate, limiting processability and performance. This issue is especially pronounced in HCPs due to their highly cross-linked nature, which often leads to irreversible aggregation, reduced surface area, and pore accessibility. Electrospinning mitigates these problems by enabling rapid solidification and aligned integration of HCPs within the fibers, enhancing structural control and improving adsorption and catalytic performance.[63,64]

#### 2.1.2. Polymers of Intrinsic Microporosity (PIMs)

PIMs represents a class of polymer materials with a unique microporosity structure, distinct from the network structures found in other POPs (Figure 2).[65] The concept of PIMs originated from Neil B. McKeown's research on the synthesis of phthalocyanine and porphyrin-based porous network polymers as potential heterogeneous catalysts.[66] In 2004, McKeown's group demonstrated that covalent bond networks are not necessary for the formation of microporous organic materials (Figure 1a). Described as "Polymers of Intrinsic Microporosity (PIM)",[67,68] PIMs form microporous solids simply due to their rigid, nonplanar, and randomly structured skeleton, which prevents tight packing. This inherent structural characteristic creates microporosity at the molecular level.[69] By reducing the area of interchain contact, PIMs disrupt cohesive interactions, allowing them to dissolve freely in organic solvents at ambient temperatures. Their inherent micropores further facilitate solvent penetration into the polymer's interior, aiding dissolution, which distinguishes them from other POPs. As a result, PIMs can be easily processed using solvent-based techniques.[70,71] Ferraris et al. first demonstrated the use of electrospun PIM-1 as a precursor for energy storage applications (Figure 1b).[72] Owing to their excellent physical stability, solubility, and intrinsic microporosity, PIMs are particularly well-suited for fabrication of separation membranes.[73] In electrospinning, PIMs can not only be electrospun independently,[74,75] but also be synthesized and then mixed with other polymer solutions.[76,77] The solubility of PIMs in organic solvents facilitates the regulation of fiber morphology and diameter during electrospinning.[78–82] Although many PIMs exhibit high solubility, certain monomer combinations can produce insoluble, non-network PIMs. Which may not perform as well as PIM-1.[83–85]

#### 2.1.3. Covalent Organic Frameworks (COFs)

COFs are a distinct class of crystalline porous polymers formed through reversible covalent reactions among organic building blocks.[86] These frameworks meticulously organize organic units into highly ordered structures via robust covalent bonds.[87,88] A unique aspect of their polymerization is the growth of polymer scaffolds through polycondensation, supplemented by non-covalent interactions to construct a framework structure within a single reaction system.[89–91] Since the Omar M. Yaghi group pioneered the synthesis of the first COF in 2005 (Figure 1a),[92] including 2D COF-1 ($S_{BET}$ = 711 $m^2$ $g^{-1}$) and COF-5 ($S_{BET}$ = 1590 $m^2$ $g^{-1}$), these frameworks have demonstrated promising potential in applications such as adsorption,[93–95] separation,[96–99] heterogeneous catalysis,[100–102] and energy storage.[103–107] This versatility is attributed to their well-defined structural characteristics and the tunability of their pore environments. Notably, the first 3D COF,[108] also developed by the Omar M. Yaghi group, features fully accessible pores, providing exposure to all edges and surfaces of the molecular units used in the framework.[109]

Electrospinning is a prominent method for producing continuous polymer nanofibers and porous membranes, prized for their scalability, high surface area, porosity, and flexibility. Polyacrylonitrile (PAN) nanofibers are particularly notable for their excellent thermal and chemical stability, owing to nitrile groups in their polymer chains. Chunhua Wu's group first reported the blending of COFs with PAN to fabricate electrospun fibers,[110] whereas Agarwal et al. pioneered the in-situ growth of COFs on PAN membrane surfaces (Figure 1b).[111] As shown in Figure 2, surface modifications have been used to introduce functional groups or alter the nitrile base, enhancing PAN's suitability as a support for in situ COF growth. PAN can also act as a sacrificial template, maintaining structural integrity upon removal to yield self-supporting porous fibers. Furthermore, presynthesized COFs can be directly incorporated into electrospun fibers via binary precursor solutions, facilitating integration with filament-forming matrices.[112,113] Nevertheless, the small diameter and limited surface area of electrospun fibers restrict COF loading capacity. Excessive COF nanoparticle accumulation can also cause structural defects. Optimizing the composite formulation is therefore essential to improve COF dispersion, preserve functionality, and maintain the structural integrity of the fiber matrix.

#### 2.1.4. Conjugated Microporous Polymers (CMPs)

CMPs represent a unique class of POPs characterized by their extended $\pi$-conjugated structures and permanent nanopores (Figure 2).[21,22,114] In 2007, Cooper et al. reported the first instance of a network of organically conjugated poly(arylacetylene) polymers, designating these materials as "Conjugated Microporous Polymers"(Figure 1a).[115] Since then, CMPs have attracted considerable attention and interest from scientists worldwide, leading to a remarkable increase in the number of publications on CMPs over the past decade.[116–120] In contrast to crystalline COFs, although CMPs are structurally amorphous, their pore structure can be precisely controlled, exhibiting excellent


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thermal and chemical stability.[121–126] Liao et al. synthesized novel hexabromobenzylbenzene (HBB)-based CMPs (HCMP-1, 2, 3, 4) by a Buchwald–Hartwig coupling reaction using HBB as the core building unit and different aryl diamines as the connecting building units.[127] This synthesis expanded the family of CMPs. The degree of conjugation of the entire system was controlled by varying the molecular chain length of the diamine linkers, allowing for the modulation of specific surface area, gas adsorption, and redox activity in HCMPs. As a result, CMPs have been extensively developed and applied across a wide range of fields, including chemical sensing,[128,129] gas adsorption/separation,[130,131] catalysis,[132–314] electrical energy storage/conversion,[135–138] and biomedical applications.[139]

CMPs typically feature highly cross-linked network structures formed through C—C or C—N coupling reactions. They can be synthesized by a variety of methods, including Sonogashira–Hagihara coupling,[140,141] Suzuki–Miyaura coupling,[142–144] Yamamoto coupling,[145,146] Buchwald–Hartwig coupling,[147,148] Schiff base condensation,[149–152] cyclotrimerization,[153–155] phenazine fusion reactions,[150] and oxidative coupling.[122,157–159] In 2015, Ullrich Scherf's group first incorporated insoluble CMPs into electrospun fibers for use in flexible chemosensors.[160] Although CMPs offer a rich variety of building units and diverse connections, their highly conjugated and rigid backbones often lead to the formation of insoluble and infusible powders. This limited processability remains a major barrier to their broader utilization in practical applications. As a result, the processing of CMPs into 1D nanofibers or 2D thin films has emerged as a key research focus in recent years.[161] The fabricated nanofibrous membranes exhibited high flexibility, high porosity, and a high surface area-to-volume ratio. The CMP-based nanofibrous membranes have been employed as sensitive sensors for detecting nitroaromatic and benzoquinone vapors, as well as oxidized metal ions.

#### *2.1.5. Porous Aromatic Frameworks (PAFs)*

Inspired by the stable tetrahedral structure of diamonds, Zhu's group tried to replace the carbon–carbon covalent bonds in diamond with a rigid benzene ring, thereby fully exposing the benzene rings and increasing the internal surface area (Figure 2). In 2009, PAFs materials made their scientific debut, setting a surface area record at the time[162] ($S_{BET}$ = 5640 $m^2$ $g^{-1}$) (Figure 1a). They are distinguished by a stiff aromatic open-framework structure composed of covalent bonds.[163–165] The framework can be tailored through different coupling reactions or the use of various building element types.[166,167] For its functionalization strategy, post-modification of PAFs with pre-anchoring sites is typically employed. Like with most POPs, a common challenge is designing their form and shape for specific applications. It has been demonstrated that when PAFs are integrated with nanofibers by electrospinning (Figure 1b),[168] they not only retain the high porosity and specific surface area of the original PAFs but also provide additional active sites, which could further broaden their application range.[169–172] Meanwhile, designing new synthetic reactions for PAFs is crucial for practical use, particularly given that organic metal catalysts are often expensive and difficult to remove from the framework.[173]

### 2.2. Advanced Morphological Control of POP-Integrated Fiber Materials Formed Via Electrospinning

POPs can be engineered into diverse nano- and microscale morphologies, significantly expanding their application potential and enhancing performance (**Figure 3**). Nanofibrous architectures are particularly attractive because they provide distinctive properties compared to other morphologies such as powders, films, or bulk structures.[174] When organized into macroscopic fibrous membranes, POPs benefit from improved accessibility, mechanical integrity, and ease of integration into practical devices.[175] Conventional processing routes, including lift-off coating and interfacial polymerization can produce POP-based membranes, but they often suffer from limitations in scalability, manual handling requirements, and interfacial instability, which hinder reproducibility and uniformity across large areas.[177]

Electrospinning provides a powerful means to overcome these challenges by directly generating continuous and homogeneous fibers at micro- and nanoscales.[178–180] This method allows precise control over fiber morphology and alignment by optimizing parameters such as nozzle design, applied voltage, flow rate, collector configuration, and environmental conditions (e.g., temperature, humidity). Post-treatment strategies such as cross-linking or thermal annealing further expand the structural diversity achievable with electrospun fibers.[181] As a result, electrospun POP-FMs combine the structural regularity and tunability of electrospun fibers with the inherent porosity and functional versatility of POPs.

Electrospun nanofibers are versatile and can be fabricated from a wide range of materials, including synthetic, natural, blended, and composite polymers.[7,182] This versatility facilitates the integration of POPs into stable, continuous films, thereby improving their handling and enabling practical deployment in flexible devices, coatings, and membranes. The fibrous form also alleviates one of the primary drawbacks of powdered POPs, which is difficulty in direct processing, by transforming them into scalable and easily manipulated macroscopic structures. In addition, electrospun nanofibers are inherently lightweight, a property that aligns with the low density and metal-free composition of most POPs. This combination offers significant advantages for applications in energy storage, filtration, and catalysis, where both weight reduction and high active surface area are critical. Unlike traditional porous nanofiber fabrication methods that often require post-pyrolysis or activation steps to achieve high BET surface areas, POP-FMs intrinsically maintain large surface areas and tunable porosity. Their metal-free nature further distinguishes them from MOF-derived fibers, offering specific benefits for biomedical, electronic, and catalytic systems where metal-free materials are desirable.[183,184]

Beyond their nanofibrous morphology, POPs contribute substantial chemical and structural tunability. By varying monomers and linkers, the pore size, shape, and chemical environment of POPs can be precisely controlled, enabling applications in adsorption, drug delivery, gas storage, and catalysis.[185–190] For example, functional groups can be grafted or doped into the POP framework, tailoring the material to capture specific molecules or facilitate targeted reactions.[191–193] Stability is another critical advantage. While certain polymer nanofibers exhibit limited resistance to harsh conditions, the incorporation of POPs,


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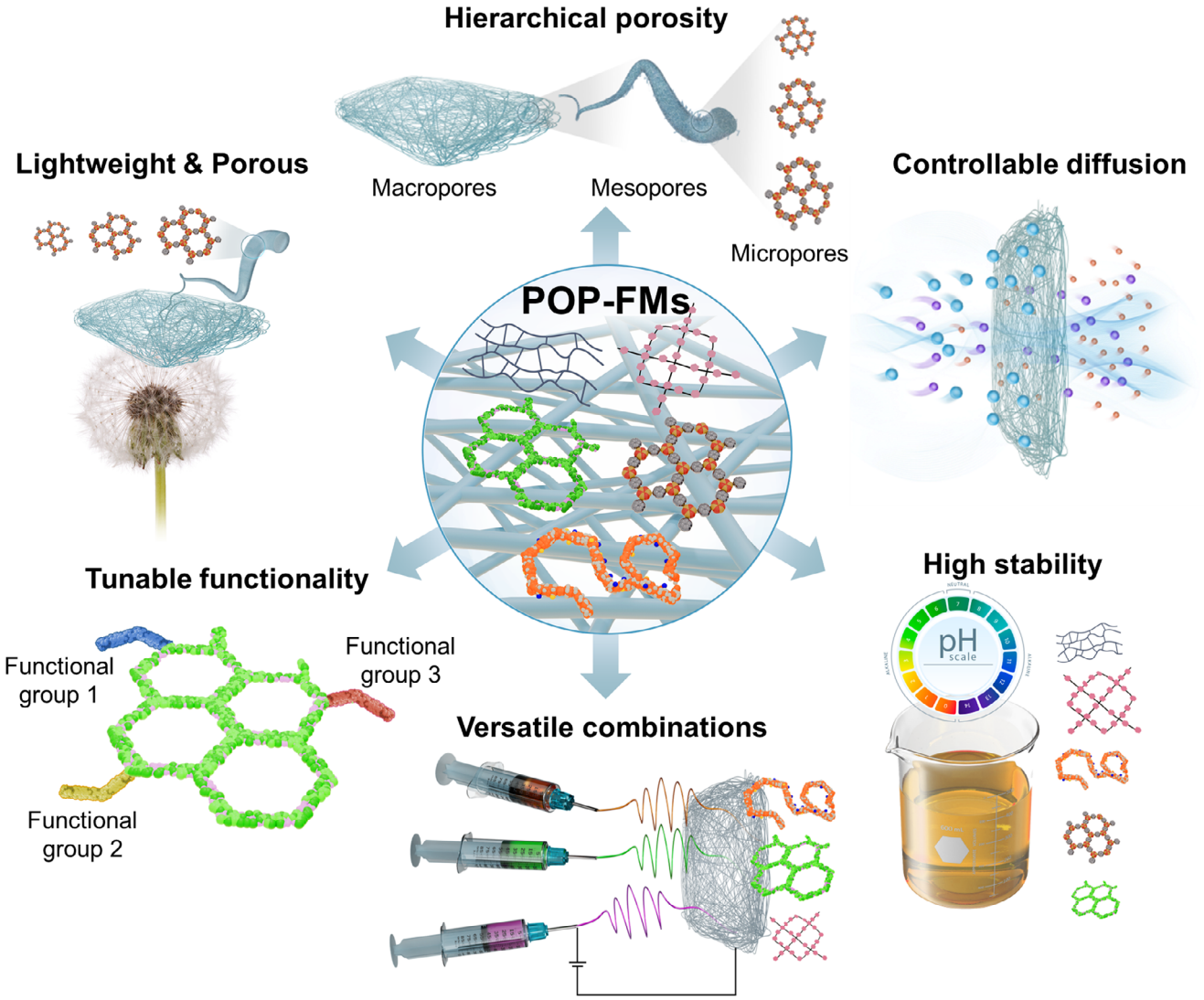


**Figure 3.** Characteristics of POP-FMs combined with electrospinning techniques, highlighting nanofiber-based versatility, lightness, POP-based functionality, stability, diffusion, and hierarchical porosity of POP-FMs.

particularly hypercross-linked polymers (HCPs), provides enhanced stability against acidic or alkaline environments, organic solvents, and wastewater streams.[180] This improvement in durability extends the practical use of POP-FMs in pollutant adsorption, water treatment, and chemical separations. Additionally, POP-FMs often demonstrate superior recyclability compared to powdered POPs, lowering environmental impact and minimizing risks of secondary contamination.[194]

The unique performance of POP-FMs is further amplified by the engineering of hierarchical porosity across multiple length scales. At the molecular level, POP backbones such as PIMs, CMPs, PAFs, and COFs enable precise control of microporosity through polymerization degree, conjugation, and topology.[195–197] At the fibrous scale, electrospinning conditions including voltage, additives, and fiber diameter regulate mesopore formation, while embedding POPs into nanofibers facilitates hybrid microporous–mesoporous structures.[180,198–200] At the network scale, assembling electrospun POP fibers into 3D architectures introduces macroporosity, thereby generating hierarchical pore systems that enhance mass transport and adsorption efficiency.[202–204] Such engineered multiscale porosity not only improves molecular diffusion and accessibility but also maximizes the utilization of active sites, leading to superior performance in catalysis, gas storage, pollutant removal, and biomedical applications.

## 3. Electrospinning-Based Strategies for POP-FMs Fabrication

Electrospinning is a highly useful method for generating POP-FMs with composite structures.[205,206] For effective electrospinning, the engineered synthesis of POPs and their properties are crucial. Specifically, since PIMs are the only soluble POPs, they can be directly dissolved to prepare electrospinnable solutions. For other POPs, it is necessary to control their size, dispersibility, and hydrophilic–hydrophobic balance to prepare suitable


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working fluids. To achieve composite formation through electrospinning, careful selection of strategies is required. While PIMs can be directly electrospun, blending with a polymer is the most common method for other POPs. Additionally, advanced methods such as post-formation, direct growth after electrospinning, and self-forming processes have been developed. Once POP-FMs are generated, post-functionalization methods must be carefully chosen, particularly for carbon-based and non-carbon-based POP-FMs.

### 3.1. Engineered Synthesis of POPs Suitable for Electrospinning

Except for PIMs, which can be directly electrospun, blending with polymers is the most straightforward method for obtaining POPs suitable for electrospinning. The construction of these composites can be achieved using covalent or non-covalent interactions, depending on the designed target system. Furthermore, for various types of constructions, post-modifications have also been achieved to improve functionality or tailor properties for specific applications. PIMs are a type of POPs that form microporous solids due to the low packing efficiency of their rigid and contorted macromolecular chains. Unlike other POPs, PIMs do not contain a cross-linked covalent bond network, which allows them to dissolve in organic solvents and be processed into robust films, coatings, or fibers. The size of the particles is critical for homogeneous dispersion in the electrospinnable fluid.[207] Compared to microscale-sized POPs particles, nanoscale POPs usually exhibit excellent properties and good processability. For example, the rod-shaped HOF-101-H nanocrystals, approximately 60 nm in length, exhibit good processability for electrospinning and can be well-confined within polyvinylidene fluoride (PVDF) fibers with a diameter of 300 nm.[208] While crystallinity in other POPs can hinder the electrospinning process and limit its scalability, certain 2D COFs can be processed via aqueous exfoliation to yield fewer-layered 2D COFs. These exfoliated COFs can be stabilized by lipophilic polymers or surfactants, which prevent $\pi$–$\pi$ stacking and aggregation of the exfoliated COFs, enabling stable COF dispersions in water. Additionally, polyethylene glycol (PEG) functionalization can further enhance dispersibility. For example, Jia et al. reported that PEG-modified monofunctional curcumin (CUR) derivatives and amine-functionalized COF-1 were coupled to create the water-dispersible polymer-COF-1 nanocomposites.[209] Wu et al. synthesized hairy microporous polymeric nanospheres that were pH/temperature sensitive, carbonizable, and water dispersible. Based on the hypercross-linking methods, the surfaces of hairy functional block can be further modified via surface-initiated atom transfer radical polymerization, allowing the introduction of other functional groups.[210] This approach enables precise tuning of surface chemistry, facilitating easy regulation of hydrophilicity and hydrophobicity.

### 3.2. Electrospinning-Based Methods for POP-FMs Fabrication

After achieving precise control over the size, dispersibility, and hydrophilic-hydrophobic balance of POPs, electrospinning emerges as a key technique for producing POP-FM composites. For most of the POPs, straightforward electrospinning can be achieved by mixing POPs with a filament-forming polymeric matrix. However, due to the structural and chemical diversity of POPs, it is essential to select the most suitable precursors and consider potential post-treatment processes. Based on these fabrication strategies, the electrospinning-based techniques for POP-FMs production can be divided into five categories, as illustrated in **Figure 4** and **Table S1** (Supporting Information).

#### *3.2.1. Direct Dissolution of POPs for Electrospinning*

The direct dissolution of the POPs in the organic solvent, along with the filament-forming polymers, represents a classical approach to electrospinning technique. In contrast to other POPs, PIMs are typically soluble in organic solvents at ambient temperature without the need for additional solubilizing groups. This distinctive property of PIMs enables them to be processed from a co-dissolved solution into solid fibrous membranes for applications such as gas separation, composites and hollow fiber membranes, thin films for sensors, and 3D printing. Moreover, functional groups can be easily introduced during direct electrospinning (Figure 4aI), or PIMs can be electrospun with other polymers to form layered composites (Figure 4aII). Compared with other types of POP, PIM is not composed of a cross-linked covalent network. The good solubility of PIMs is promoted by the same design features that ensure inefficient packing and the generation of intrinsic microporosity, with the rigid and contorted chain structure also reducing inter-chain cohesion and encouraging the absorption of solvent to aid dissolution. For example, Tamer Uyar's group reported that PIM-1 could be dissolved in the 1,1,2,2-tetrachloroethane at a concentration of 23% (w/v). To ensure the homogeneity, the PIM-1 solution was stirred at 60 °C for 1 h and then left at room temperature overnight. Using PIM-1 with a molecular weight of 89 kDa (Mw) and a narrow polydispersity index of 1.78, fibers with an average diameter of 2.07 ± 0.52 µm were successfully generated.[74] In another report, the modified polyimides of intrinsic microporosity (PIM-PIs) were synthesized. These POPs were found to be soluble in the dimethylformamide (DMF) at various concentrations, with the high BET surface areas ranging from 270 to 506 $m^2 g^{-1}$. The authors demonstrated that adding tetraethylammonium bromide salt significantly improved conductivity and spinnability, resulting in thinner fibers.[211] Apart from the PIM-1, PIM-2, synthesized using various fluoroarenes, can also be electrospun after being dissolved in tetrachloroethane at a concentration of 43%. The resulting fiber mat, with an average diameter of 5.5 ± 1.5 µm, demonstrates notable microporosity and a BET surface area of approximately 600 $m^2 g^{-1}$. Furthermore, this represents the first evidence of the electrospinning process successfully creating a self-standing PIM-2 fiber membrane.[212] Directly dissolving PIMs is a simple and efficient method, but its limitation is evident, as it applies to only a few PIM-related polymers.

#### *3.2.2. Electrospinning Molding After Mixing*

Electrospinning after mixing involves preparing a blended fluid by dissolving or dispersing various materials into a solvent or a solvent mixture before subjecting it to the electrospinning process. This approach allows for the creation of fibers with tailored


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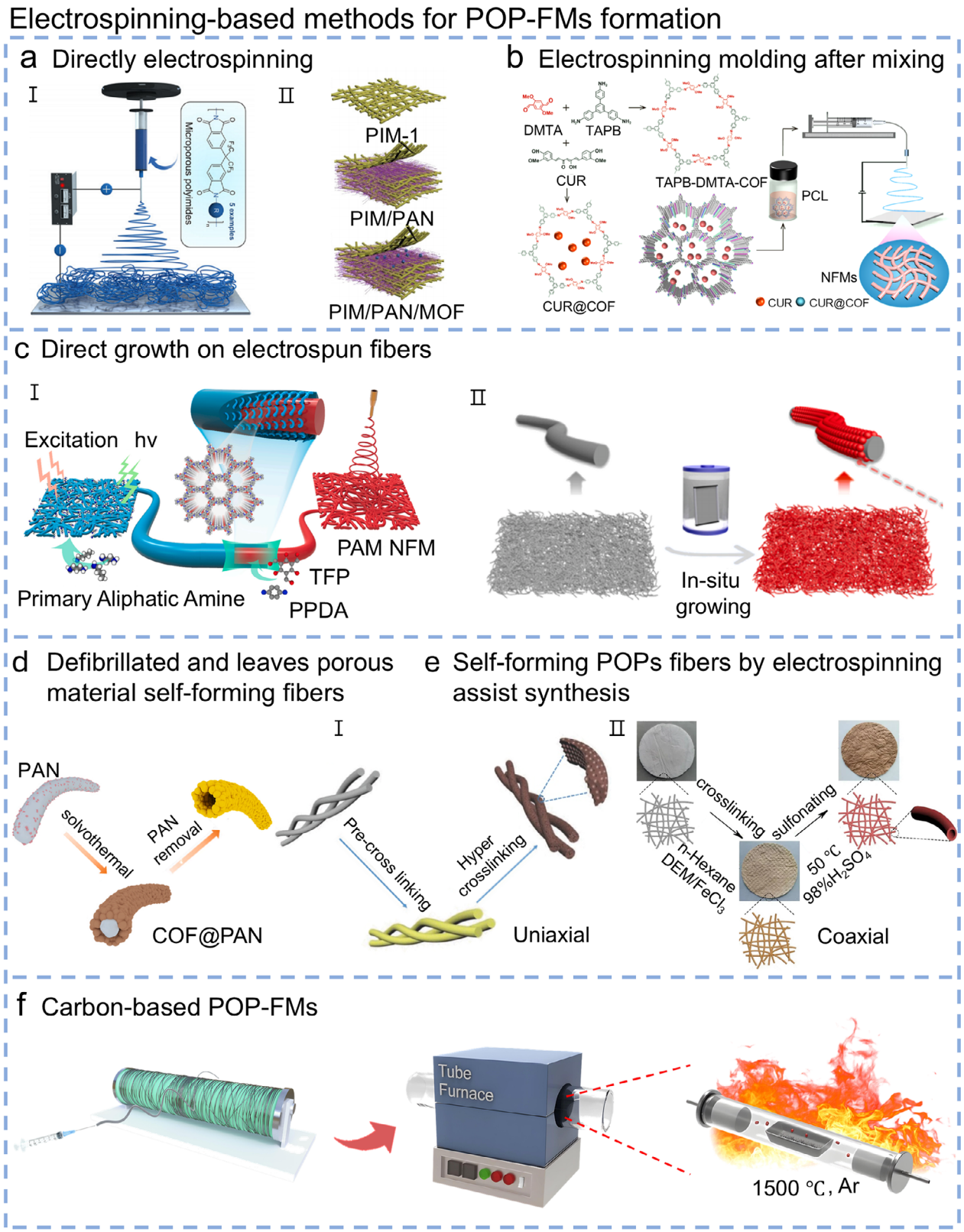



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properties by combining different polymers or nanoparticles in an electrospun uniaxial fiber (Figure 4b). It ensures uniform dispersion of components, either on a nanoscale or micrometer scale, preventing possible phase separation and aggregation.[213] The process typically includes the following steps: first, the POPs are synthesized through the conventional liquid phase polymerization. Then pre-quantified POPs are dispersed in the organic solution and further mixed with the electrospinnable polymeric solutions. To ensure homogeneity, sonication is usually necessary to completely blend the mixture solution. Finally, electrospinning is conducted by carefully selecting the needle tip and optimizing parameters such as voltage, flow rate, and collector distance. In some cases, particularly for energy-related applications, a pyrolysis process is also essential. This method often generates fibers with a filament–granule composite morphology, where the POPs particles appear as granules, and the polymer forms the filaments. In certain special circumstances, particle aggregation may occur, resulting in the formation of different spindle-shaped thicknesses.[214] For example, the Ullrich Scherf group demonstrated the electrospinning of CMP/PLA mixtures to create light-emitting nanofibrous films. The CMP, with an average diameter of around 80 nm. was used to synthesize electrospun fibers with a diameter ≈500 nm. The resulting nanofibrous sheets exhibited high porosity, surface-area-to-volume ratios, and flexibility.[160]

### 3.2.3. Direct Growth on Electrospun Fibers

Due to poor dispersion of POPs in the electrospun solution, the number of POPs that can be mixed into the solution is often limited. Additionally, when POPs are incorporated inside the fibers, their porous sites are difficult to expose. Growing POPs directly on electrospun fibers offers an alternative option, increasing the number of POPs on the surface of nanofibers.[215] This method typically involves preparing precursor solutions and spinning them into fibers using electrospinning techniques, followed by chemical or physical deposition directly onto these fibers. This process introduces porosity while maintaining the fiber morphology, allowing POPs to be synthesized directly on the surface or inside the spun fibers (Figure 4c). Common growth methods include: 1) In situ polymerization, where a porous layer is formed by immersing the fibers in a solution of monomers and cross-linkers, followed by polymerization under appropriate conditions; 2) Surface modification, where specific functional groups are introduced onto the fiber surface to facilitate chemical reactions with polymer monomers; 3) Sol–gel processing, in which POPs are transformed into porous polymers by coating the precursor solution and post-treatment; and 4) Vapor deposition, where precursor gases are deposited onto the fiber surface and polymerized. The key advantage of these techniques lies in the ability to tune the properties of the porous structure by controlling growth conditions such as temperature, time, and precursor concentration, enabling fine control over the material properties. In addition, the direct growth method enables the strong adhesion of the porous material to the fiber substrate,[216] which is crucial for improving the mechanical stability and functional integration of the composite.

Given the rich pore structure and abundant proton acceptor groups (C≡N) on the electrospun PAN membrane, hydrogen bonds can form with proton donors (amino groups ($-NH_2$) and aldehyde groups (–CHO)) in the imine COFs. Ma et al. reported that by using electrospun PAN as the raw material, COFs with exposed spherical structures could grow on the PAN fibers, resulting in a morphology where spheres are attached to the fibers. After formation, the BET surface area significantly increased from 13 $m^2 g^{-1}$ for the PAN membrane to 197 $m^2 g^{-1}$ for the PAN/COF composite.[217] Furthermore, by modifying the monomer used for generating the COFs, the surface morphology of the fiber could also be optimized. Herath et al. reported a similar process using PAN fiber as the substrate and growth of guanidinium-based ionic COFs. This process generated rough surface morphologies, rather than spherical morphologies, with the fiber diameter of 287 nm for the raw PAN fiber and the fiber diameter of 440 nm for COF-PAN composite fiber.[43]

In addition to COFs that can grow on the surface of PAN, PAFs-based POPs have also been reported to grow on the surface of PAN fiber. In this approach, the PAN fibers are first electrospun as a template, and then the polyaniline (PANI) is grown on the surface of PAN fibers. Following this, PAF-45 modification is in situ synthesized in the presence of the PANI-PAN composite fiber. The resulting composite fiber exhibited satisfactory tensile strength of 6.67 MP and elongation at break of 8.14%, along with an enhanced BET surface area of 262.4 $m^2 g^{-1}$, making it suitable for practical adsorption applications.[168]

### 3.2.4. Defibrillated and Self-Forming Porous Material Fibers

Defibrillation, which allows the porous material to form fibers on its own, is usually involved in the preparation of fiber templates by electrospinning. Generally, polyvinyl alcohol (PVA) or PAN electrospun fiber template is placed in the precursor solution of POPs, where growth occurs on the fiber surface or within the fibers. Then the fiber template is removed by using

**Figure 4.** Electrospinning-based methods for composite formation. aI) Direct electrospinning with solution processability. aII) Alternating electrospinning realizes superposition of fiber membrane. b) POPs dispersed in organic solvents are electrospun by mixing with other substances. cI) On the electrospun fiber, the seed layer grows from the inside out. cII) Seed layer on the electrospun fiber grows on the surface. d) Electrospun fibers serve as sacrificial templates, leaving the porous material on the surface to self-form fibers after removal. eI) Hypercross-linking is performed on the pre-cross-linked construction unit. eII) In the low temperature cross-linking system for cross-linking electrospinning fiber membrane. f) The electrospinning fibers carbonization post-processing. Panel (aI) Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International license (https://creativecommons.org/licenses/by/4.0).[211] Copyright 2021, published by Elsevier. Panel (aII) Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International license (https://creativecommons.org/licenses/by/4.0).[230] Copyright 2020, published by Elsevier. Panel b) Reproduced with permission.[213] Copyright 2022, published by American Chemical Society. Panel (cI) Reproduced with permission.[327] Copyright 2022, published by Elsevier. Panel (cII) Reproduced with permission.[215] Copyright 2023, published by Elsevier. Panel d Reproduced with permission.[260] Copyright 2024, published by Elsevier. Panel (eI) Reproduced with permission.[59] Copyright 2017, published by Wiley. Panel (eII) Reproduced with permission.[57] Copyright 2022, published by Elsevier.


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appropriate solvents or heat treatment methods, resulting in a self-supporting porous structural material (Figure 4d).

Inspired by the methods mentioned above, defibrillated and loose porous materials can yield pure POPs while maintaining fiber chain-like morphologies. Seema Agarwal's group demonstrated that PAN can also serve as a sacrificial template. To obtain the self-standing porous COF membranes, PAN and p-phenylenediamine were first electrospun. After in situ growth of COFs, the PAN was removed by using a Soxhlet extractor and DMF. The resulting pure COF membranes exhibited a large surface area of 1153 $m^2$ $g^{-1}$, along with good mechanical stability, excellent thermal stability, and flexibility.[111]

Besides COFs, CMPs have also been reported to use the polyvinyl pyrrolidone (PVP) fiber as sacrificial templates. For example, Lee et al. reported using the Sonogashira–Hagihara coupling reaction to grow CMPs on a PVP electrospun membrane, resulting in the formation of a hollow CMPs fiber membrane. Upon removal of the PVP nanofibers, the surface area increased to 758 $m^2$ $g^{-1}$ due to the formation of the hollow tubular structure.[218]

In a recent study, Chong et al. provided a feasible method for replacing precious metals for developing platinum-free oxygen evolution reaction (OER) catalysts. Co-zeolitic imidazolate framework (ZIF), with high inherent porosity and network structure, was selected as the catalyst precursor. This precursor is converted into interlinked hollow metal oxide particles by low-temperature oxidation. La- and Mn-doped Co-ZIF were prepared in solution and then suspended in a PAN solution for electrospinning. Finally, the resulting material was activated for 6 h in circulating air at 360 °C to remove organic components and form porous fibers.[219] This work also offers an alternative solution for achieving doped porous polymers or composites with unique morphologies.

#### 3.2.5. Self-Forming POPs Fibers by Electrospinning Assist Synthesis

The synthesis of self-forming POPs fibers aided by electrospinning typically involves dissolving the precursors of POPs in appropriate solvents. Electrospinning is then used to spin the fluid into a fiber membrane, which serves as a precursor of POPs. This approach is primarily related to POPs based on HCPs, where chemical or physical cross-linking is performed in the pre-solidified aromatic linear polymers, resulting in a 3D network structure (Figure 4eI). The resulting POPs fiber not only retains the morphological characteristics of the template but also exhibits stability after cross-linking and a specific pore structure, thereby enhancing their potential for wide applications in various fields.

Polystyrene, a readily available polymer containing aromatic hydrocarbons, is also a major source of waste plastics.[220] However, due to solubility issues, polystyrene tends to dissolve readily in conventional cross-linking solutions such as dichloroethane, making it challenging to preserve fiber morphologies while achieving effective cross-linking. Glacial acetic acid (HAc) was identified as a highly suitable solvent due to its strong ability to slow down the Friedel–Crafts reaction and its limited solubility in polystyrene. The formation of a coordination link between the carboxyl group in HAc and the catalyst $FeCl_3$, significantly reduced the efficiency of the Friedel–Crafts reaction, allowing the fibrous morphologies to be preserved and further retained even after the subsequent hypercross-linking process.[59] Cyclodextrins (CDs), cyclic oligosaccharides with a partially hydrophobic cavity interior and a truncated cone shape, can combine organic micropollutants to form complexes. By electrospinning highly concentrated CD solutions with a naturally occurring graphitic acid linker, nanofibrous hypercross-linked cyclodextrin network membranes were created. This cross-linking process enables the porous network to maintain its structure in both organic and aqueous solvents.[58] Unlike stepwise cross-linking, electrospun fiber membranes can be directly immersed in a low-temperature cross-linking system for rapid cross-linking,[57] effectively retaining the pre-designed morphological structure of the fiber membrane (Figure 4eII).

#### 3.2.6. Other Approaches

In addition to the above-mentioned methods, physical approaches can also be employed in the fabrication of POP-FMs. For example, Tang's group demonstrated that cationic conjugated microporous polymers (CCMPs) could be in situ polymerized on the pristine polypropylene (PP) fabrics, such as those used in medical masks, through a simple spray process.[221] The reaction solution for generating CCMPs was homogeneously sprayed onto the PP fibers and subsequently incubated under 80 °C. The in situ polymerization of CCMPs on the surface of PP fibers, along with the resulting network structure, contributed to enhanced adhesion stability of CCMPs coated on the fibers, as demonstrated in this study. Unlike approaches that grow adherent coatings directly on fiber surfaces, Yi's group electrosprayed pre-synthesized POP fillers as charged microdroplets or particles onto the substrate and then cross-linked the composite membrane to immobilize the fillers, producing a selectively functional layer with controlled filler distribution and increased surface roughness.[222] This strategy is more effective at controlling microstructure, reducing filler aggregation, and substantially improving membrane flux and mass-transfer efficiency. Coating processes are also widely applied to the construction of POP-FMs, and they demonstrate excellent controllability and adaptability. Li's group opened a new pathway for the mass production of POPs membranes by exploiting the mildness and scalability of click chemistry.[223] They used a coating technique to induce in situ polymerization on a kapok fiber substrate, thereby fabricating a POPs composite membrane whose hierarchically engineered pores solved the fixed pore-size limitation of traditional filter materials. Similarly, Zhu's group first proposed an interfacial polymerization strategy using hydroxyl-containing POP, innovatively recasting POP from its traditional role as an inert filler into the structural motif of the membrane separation layer, thereby maximizing its intrinsic porosity and functional-group utility.[224]

### 3.3. Post-Functionalization of POP-FMs

#### 3.3.1. Carbon-Based POP-FMs

POP-FMs can also be pyrolyzed in an inert atmosphere to generate carbon materials by selecting non-degradable polymers


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(Figure 4f). Carbon materials are particularly indispensable for energy-related applications, such as supercapacitors, batteries, and fuel cells. Derived carbon materials derived from POPs have been reported in various forms, including fibers, sheets, and even ortho-cubes beyond the traditional spherical structures. Electrospinning technology allows for the synthesis of carbon fibers with adjustable pore size, heteroatom doping, and composite morphologies by selecting suitable polymers, employing effective pore expansion methods, and optimizing the carbonization pathway. By effectively combining bottom-up and top-down technologies, the unique advantages of POPs and electrospinning can be integrated. For instance, Jong Hak Kim's group reported carbon nanofibers derived from hydrogen-bonded organic frameworks (HOFs) with a distinctive "flower-on-fiber" morphology.[214] To ensure heteroatom doping and structural design, flower-like melamine cyanurate was first synthesized. These HOFs were then added into a DMF solution, followed by the addition of PAN, which was completely dissolved to create an electrospinnable blended solution. The resulting PAN@melamine cyanurate (MCA) carbon nanofiber (c-PAN@$MCA_{0.25}$) exhibited a high nitrogen content of 13.5 at% high specific surface area of 403.1 $m^2 g^{-1}$.[214] HCPs have also been reported as an ideal resource for generating electrospinning carbon materials. Wu et al. demonstrated that the stepwise cross-linking of electrospun HCPs can produce fibrous carbon. This method produced carbon fibers with a BET surface area of 1615 $m^2 g^{-1}$. Unlike other composite fiber materials, the hypercross-linked process enables the rapid thermal synthesis of the porous carbon fibers resulted from the electrospinning of cross-linkable polymer precursors.[59] Ding Yuan's group demonstrated that combining electrospun PAN with MOF/COF hybridization strategy can produce a 3D highly open catalyst with a hierarchical structure.[225] The resulting materials featured the formation of metal–Nx (M–Nx, where M = Co and Zn) moieties and uniform Co nanoparticles with high-density active sites. This was achieved through the integration of the MOF/COF hybridization method with direct pyrolysis and electrospinning process. The well-designed branch-leaf nanostructures facilitated the formation of numerous homogeneous active sites on the carbon frameworks derived from MOFs and COFs, resulting in a synergistic effect from multiple active sites.

#### *3.3.2. Non-Carbon-Based POPs-FMs*

POP-FMs can not only be pyrolyzed to generate carbon materials with important functions for specific target applications, but also non-carbon-based POPs-FMs can be post-functionalized for expanding their application scope. With advancements in coaxial electrospinning, hollow cross-linked aromatic polystyrene fibers can be successfully synthesized. This hollow morphology is achieved by using polyvinylpyrrolidone (PVP) as the core material and polystyrene (PS) as the shell. After the hypercross-linking process, the hollow fibers with a diameter of 2-3 µm are obtained. Furthermore, through sulfonation process, the aromatic rings of the cross-linked PS can be functionalized. Notably, the contact angle changes from 90° to 0°, indicating excellent hydrophilicity, which is essential for dye removal applications.[57]

In addition to HCPs, PIMs also offer reactive sites for post-functionalization. Tamer Uyar's group reported the creation of an amine-modified PIM-1 fibrous membrane by reacting a PIM-1 fibrous membrane with borane dimethyl sulfide complex. The amine modification did not affect the fiber diameter, which remained at 2.3 ± 0.3 µm.[226] The resulting material's insolubility, chemical stability, and amine functionality make it suitable for various specialized applications.

Furthermore, the MOFs can also be functionalized on the surface of COFs-based POP-FMs. Ma et al. demonstrated this by using a PAN/*p*-phenylenediamine electrospun membrane as the precursor. COF polymerization was initiated using the 1,3,5-triformylphloroglucinol (Tp) monomer, and subsequent removal of PAN resulted in a self-standing nanofiber membrane. By replacing the traditional supports, such as inorganic discs, metal meshes, and polymer membrane, the COF membrane served as a tailored substrate for the confined growth of ZIF-8. This approach enabled the synthesis of a COF-MOF composite membrane with enhanced functionality.[227]

### 3.4. Advanced Design of POPs Via Electrospinning: Key Findings and Opportunities

Electrospinning, as a versatile fabrication technique, has enabled the development of a wide range of composite nanofibers with enhanced functionality and performance. The following subsections will explore the integration of porous organic polymers (POPs) with various matrices and functional groups, highlighting their emergent properties and broad applicability. To facilitate a comprehensive exploration of each area, they were purposefully separated into three distinct categories 1) POPs with polymer composites, 2) POPs in MOF-based nanofibers, and 3) POPs functionalized with catalytic groups (**Figure 5a**). This structure reflects the diverse applications of electrospun fibers and illustrates how the integration of POPs with other functional materials enables the tailoring of properties across different fields. Each subcategory represents a significant area of research that highlights the versatility of electrospinning technology in enhancing the performance of composite materials, particularly through the incorporation of POPs. Lastly, it is noted that some advanced electrospinning techniques may offer even greater potential for POP-FMs.

Integrating POPs with other polymers in electrospun fibers represents unique properties surpassing those of individual materials, enabling diverse energy,[228] environmental, and biomedical applications.[229,230] These composites benefit from enhanced mechanical strength, flexibility, and surface area, which improve adsorption capacities, catalytic activities, and stability in complex environments. Hierarchical structuring achieved through electrospinning further optimizes functionalities such as gas sensors,[231,232] antimicrobial efficacy,[233,234] and controlled molecular transport,[235,236] making POP-polymer composites integral to the advancement of next-generation functional materials. This synergistic integration of materials shows new frontiers in fields such as energy,[237] environmental,[238] and biomedicine,[239] positioning POP-incorporated polymeric nanofiber composites as a cornerstone in the development of next-generation functional materials. Recent studies underscore


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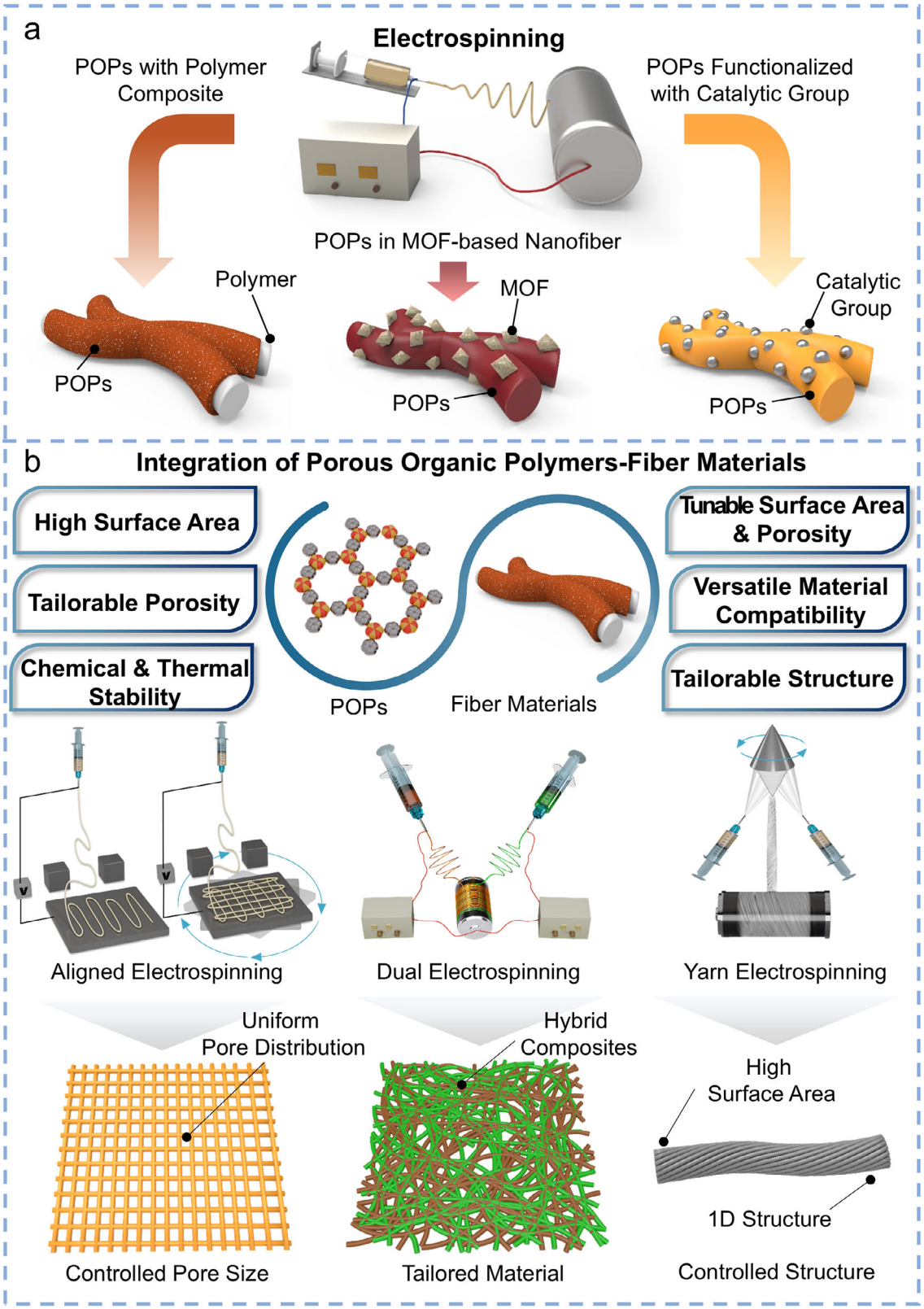


**Figure 5.** a) Schematic illustration of nanofibrous POPs-based composite using electrospinning technology. b) Advantages of POP-FMs and future directions of emerging various electrospinning techniques that aligned electrospinning, dual electrospinning, and yarn electrospinning.



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how electrospun POP composites can be tailored to tackle challenges in large-scale, high-performance materials. For example, nanofiber-based POP composites are advancing air filtration technology, as seen in Lijuan et al. catalytic conjugated microporous polymer membranes, which exhibit over 99.5% efficiency in removing particulate matter.[240] These findings encourage new designs for air purification systems that integrate structural resilience with active pollutant degradation. Emphasizing multifunctional design through electrospinning reveals the transformative potential of POP-based composite. They demonstrate how tailoring nanofiber morphology and material composition can meet the evolving demands in industrial, environmental, and biomedical applications, offering readers insights into the versatility of electrospinning as a cornerstone technology in advanced materials design.

POPs combined with MOFs within electrospun nanofibers offer synergistic properties that surpass the limitations of each material. POPs provide chemical stability and versatility, while MOFs contribute high surface area and tunable porosity.[241–245] Together, these materials form hierarchical structures with enhanced mass transport and increased exposure of active sites, essential for catalysis, energy storage, and environmental remediation. The interconnected micro-, meso-, and macroporous architectures facilitate superior ion diffusion, electron transfer, and adsorption capabilities, significantly improving functional performance across diverse applications.[246–251] MOFs integrated into polymer nanofibers have introduced new opportunities for enhancing mass transfer in sorbent materials. For instance, Longlong et al. created a novel bifunctional electrocatalyst by combining MOF and COF into a 3D hierarchical structure featuring branch-leaf nanostructured carbon nanofibers.[225] This hierarchical structure supports effective oxygen reduction and evolution reactions, resulting in high power density, mechanical flexibility, and long-term stability, making it ideal for wearable energy storage applications. This case study reveals the transformative potential of POP-MOF hybrid nanofibers by strategically combining the chemical robustness of POPs with the high surface area and active site accessibility of MOFs. Researchers are paving the way for multifunctional materials that can address key challenges in catalysis, water treatment, and energy storage. The hierarchical design afforded by electrospinning enables tailored transport properties, making these composites highly versatile and inspiring further innovation in advanced material development.

The functionalization of POPs with catalytic groups using electrospinning methods, such as metal nanoparticles and metal oxides, enhances their versatility, resulting in multifunctional materials with high surface area and advanced reactivity. These hybrid materials leverage the structural tunability and high surface area of POPs, while the incorporated catalytic groups enable advanced functionalities like selective adsorption,[252,253] catalytic degradation,[254,255] and pollutant removal.[256,257] Combining the structural properties of POPs and the catalytic activity of metals creates new material functionalities that are highly sought after for environmental and energy applications.[258,259] Catalytically functionalized POPs in nanofibers offer reusability, enhanced mass transport, and increased interaction between pollutants and catalytic sites due to their hierarchical porous structures. These properties make POP-catalysts composite strong candidates for next-generation remediation and energy storage systems. For example, Sachin et al. further expanded POP applications with COF hollow fibers functionalized with magnetic nanoparticles, which act as both dye-degradation catalysts and adsorbents, enhancing recyclability and reducing iron leaching in pollutant removal.[260] This versatility underscores the potential of catalytic POP composites in addressing complex environmental challenges.

To further expand the functional potential of POP-based composites, additional research opportunities arise through advanced electrospinning techniques, such as aligned electrospinning, dual electrospinning, and yarn electrospinning (Figure 5b). These methods show promises for diversifying POP structures, allowing for precise control over fiber alignment, diameter, and composite layering. Aligned electrospinning could enable anisotropic properties tailored to specific applications, such as directional strength in POP-FMs or enhanced flow properties in filtration.[261,262] Dual electrospinning supports the co-fabrication of POPs with synergistic materials, optimizing pore architecture, and catalytic functionality.[263,264] Additionally, yarn electrospinning offers the potential for robust, flexible POP composites that maintain high porosity and enable mechanical integration into larger, structurally complex systems.[265,266] These advancements offer a pathway for POP-based materials to reach broader applications, from flexible electronics[267] to regenerative medicine, where the scalability and customizable nature of these composites can significantly enhance performance.

## 4. Innovative Applications of POP-FMs Enabled by Unique Fibrous Morphology

By leveraging these unique properties, POP-FMs with fibrous morphology are propelling innovations across diverse fields, including environmental technology, healthcare, and energy systems. As shown in **Figure 6** and Table S2 (Supporting Information), their potential applications extend to the fields of biomedicine, energy, catalysis, environment, gas sensing, and more.

### 4.1. Biomedical Applications

Electrospinning is a straightforward and adaptable method for creating polymeric fiber scaffolds that replicate the structure of the extracellular matrix, making it highly suitable for a variety of biomedical applications. Accordingly, numerous studies have reported the significant potential of electrospun fibers for biomedical use. To closely mimic the hierarchical architectures, integrating POPs with electrospinning technologies offers a promising approach for creating POP-FMs composite membranes tailored for biomedical applications. Importantly, the porosity of POPs is crucial for enhancing air permeability, facilitating efficient drug loading within the porous framework, and enabling controlled release profiles in response to specific stimulus. Additionally, electrospun fibers often serve multifunctional roles, including adhesion, synergistic pro-healing, wound dressing, and drug delivery. For example, slow wound healing remains a significant challenge in clinical treatment. Dai's group developed a new kind of medicated fibrous films by dispersing enrofloxacin, flunixin meglumine, and a $\beta$-COF complex into a thermoplastic polyurethane


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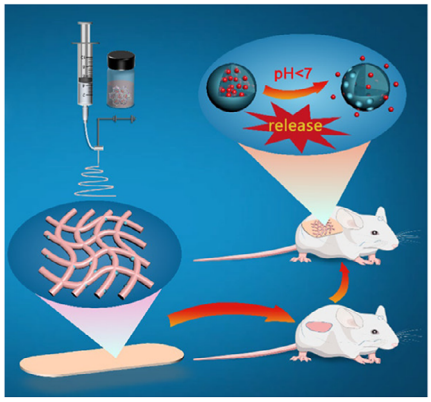
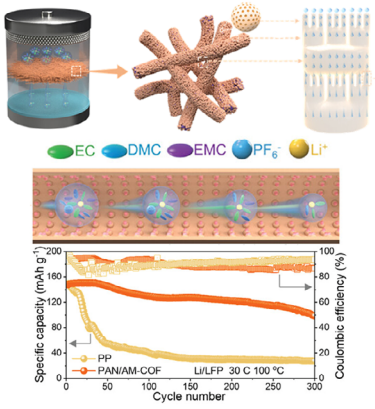
# Innovative applications of POP-FMs

## a Biomedical applications



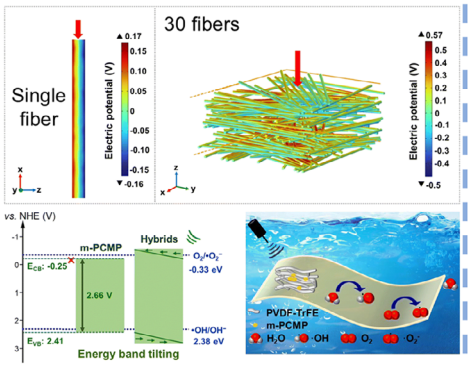

Wound dressing

## b Energy applications



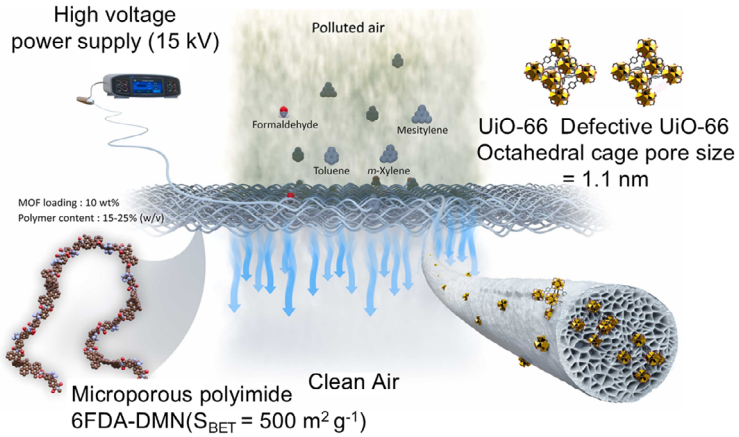

Smart battery separators

## c Heterogeneous catalysis applications



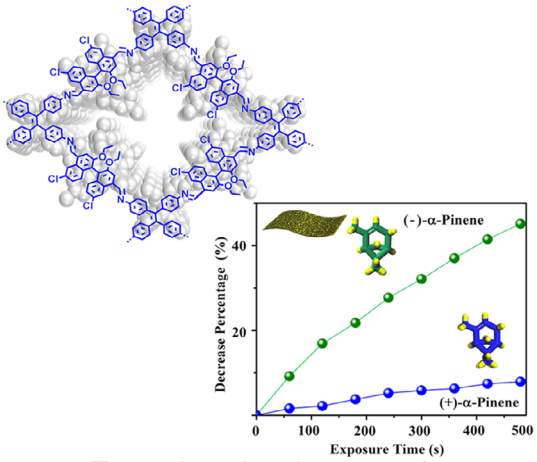

Piezo-photocatalysts

## d Environmental applications




Capturing volatile organic compounds

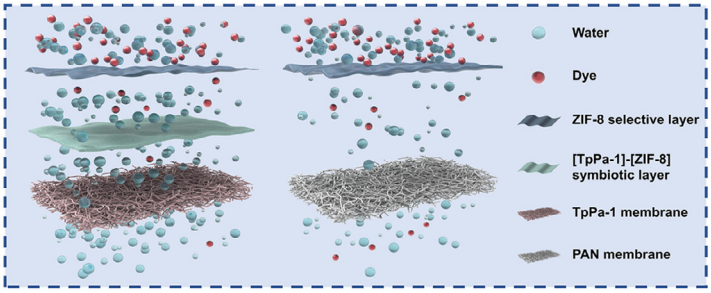
## e Sensor applications




Enantioselective sensing

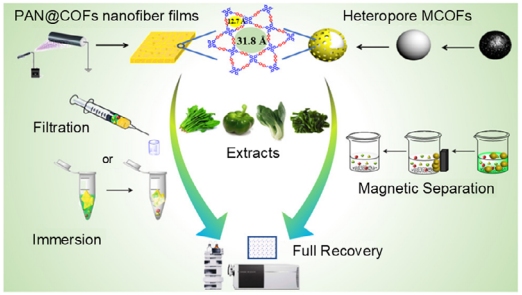
## f Other applications

I




Molecular eparation

II




Nontargeted analysis of food safety



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solution. This fibrous film demonstrated effective slow-release characteristics and exhibited strong antibacterial activity against *Staphylococcus aureus* and *Escherichia coli*.[213] achieving a bacteriostatic rate of 99% over 50 h. More importantly, in a mouse full-layer skin defect model, the fibrous membrane achieved a good ability to promote wound healing by down-regulating inflammatory factors such as tumor necrosis factor-$\alpha$ and interleukin-1$\beta$, while up-regulating growth factors like vascular endothelial growth factor and epidermal growth factor. Furthermore, the use of mild and natural antimicrobial ingredients is increasingly preferred to reduce the risk of drug resistance caused by antibiotics. CUR and L-borneol are both natural antimicrobial ingredients, but with different antibacterial mechanisms. CUR directly kills the bacteria, while L-borneol can reduce bacterial colonization. The CUR can be loaded as high as 27.68% by using COFs formed via the Schiff base reactions. Similarly, L-borneol can be loaded onto the surface of hollow HCPs spheres through an esterification reaction. The CUR-loaded COF can be further electropsun to generate polycaprolactone PCL/COF nanofibers for wound healing applications (Figure 6a). PH responsibility was observed on the PCL/COF fiber, under low pH circumstances, typically found in tumor-infiltrated and bacteria-infected tissues, which exhibit an acidic extracellular microenvironment. The high concentration of $H^+$ caused the protonation of nitrogen atoms in imine groups and C≡N, reducing the hydrophobic interaction between CUR and COF. As a result, CUR can be easily released at the wound site, and the electrospun fiber morphologies further enhance the thermal ability, mechanical properties, and biocompatibility. Although L-borneol has low antibacterial activity on its own, its antimicrobial effectiveness is enhanced after being grafted onto the surface of hollow HCPs spheres. When combined with fibers in the home-made syringes,[268] the HCPs demonstrated the expected absorption and antibacterial properties.

To address the problem of drug-resistant bacteria, reactive oxygen species (ROS) are effective and eco-friendly bactericidal agents commonly used in antibacterial and medical applications without inducing resistance.[269] Hydrogen-bonded organic frameworks (HOFs) are a kind of photoactive material that can produce strong ROS under light irradiation. Wang et al. showed that the HOFs, with a size of ≈60 nm, could be encapsulated in PVDF-hexafluoro-propylene nanofibers. Even at just 0.5 wt.%, these fibers were able to generate singlet oxygen ($^1O_2$) and kill pathogens, including viruses, bacteria, and fungi, within 30 min under ambient light conditions at 520 nm.[208] Similarly, the Landfester group proposed a light-induced disinfection strategy by embedding photosensitive conjugated microporous polymer (CMP) nanoparticles into polyvinyl alcohol hydrogel nanofibers via colloidal electrospinning, followed by cross-linking the membrane to render it insoluble in water. Based on the production of active singlet oxygen from the nanoparticles under visible-light irradiation, complete elimination of *Escherichia coli K-12* and *Bacillus subtiles* was achieved in 120 and 60 min, respectively.[270] Furthermore, the membrane's good mechanical properties allowed for easy removal after use and prevented the nanoparticles from leaking into the wound.

POP-FMs are primarily used in antibacterial and wound healing applications, but they are also finding emerging uses in biosensors.[271] In these applications, accurate quantitative assessment of small molecule metabolites in biological samples is critical. Zhang's group devised a unique electrospinning winding approach for directly producing non-powder COFs and developed a solid phase microextraction fiber coating with a mechanically stable macromolecular barrier.[272] In conjunction with gas chromatography, a simple and effective quantitative analytical approach for hydroxyl polycyclic aromatic hydrocarbons was developed. The test features a low detection limit (0.008–0.050 ng·L), a broad linear range (0.02–1000 ng·L), and high reproducibility (1.0%–7.3%). The coated fiber is resistant to matrix interference in complex biological substrates (2.5%–16.7%) and has been effectively applied to analyze metabolic byproducts in human urine and plasma.

### 4.2. Energy Applications

The adjustable pore structure and multifunctionality of POPs render them a promising candidate for applications in electrochemical energy storage, including supercapacitors,[273–275] lithium secondary batteries (lithium-ion batteries,[276,278] lithium–sulfur batteries,[279–281] lithium-metal batteries[282,283]), and osmotic energy storage.[284] Nevertheless, the practical application of rigid POP nanoparticles is limited by their poor processability and inherently low electrical conductivity.[23,285] In contrast, 1D nanofibrous materials such as carbon nanotubes (CNTs) and carbon nanofibers (CNFs) exhibit excellent electrical conductivity and mechanical strength.[286,287] Therefore, integrating POPs into nanofibrous architectures offers a promising solution to overcome these limitations and expand their applicability, particularly in energy-related fields.

**Figure 6.** Innovative applications of POP-FMs. a) Nanocomposite can effectively support the drug in situ, and further incorporate it into PCL nanofibers by electrospinning, which has a pH intelligent response of drug release in acidic extracellular microenvironment. b) Schematic diagram of ion transport pathway and desolation of $Li^+$ using separator with hierarchical porous channels for lithium metal batteries and long-term cycling performance of Li||LFP cells under 100 °C conditions at 30 °C. c) Electric potential of PVDF-TrFE single fiber and 30 randomly distributed fibers in a certain area under vertical pressure was simulated by finite element method. A schematic example of m-PCMP's energy band tilting as a result of the PVDF-TrFE piezoelectric potential. A schematic representation of RhB degradation with m-PCMP/PVDF-TrFE nanofibril hybrid films. d) Diagram illustrating the electrospinning process of microporous polyimide nanofibrous membranes implanted in metal organic frameworks for air filtration purposes, specifically focusing on volatile organic compounds. e) Molecular formula of chiral COFs and decrease percentage upon exposure to $\alpha$-pinene for 7@PVDF. fI) Separation of dye aqueous solution by the [TpPa-1]-[ZIF-8] (left) and PAN-[ZIF-8] membranes (right) is shown schematically. fII) Preparing heteropore COF-based magnetic nanospheres and nanofiber films for vegetable sample pretreatment. Panel (a) Reproduced with permission.[213] Copyright 2022, published by American Chemical Society. Panel (b) Reproduced with permission.[46] Copyright 2024, published by Wiley. Panel (c) Reproduced with permission.[306] Copyright 2022, published by Royal Society of Chemistry. Panel (d) Reproduced with permission.[76] Copyright 2022, published by Elsevier. Panel € Reproduced with permission.[112] Copyright 2019, published by American Chemical Society. Panel (fI) Reproduced with permission.[227] Copyright 2024, published by Wiley. Panel (fII) Reproduced with permission.[113] Copyright 2020, published by American Chemical Society.


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COFs are predominantly found in powder form on a macroscopic scale. However, their microscopic morphology is remarkably diverse, and it has been demonstrated that COFs with nanofiber structures can be effectively combined with CNTs to create high-quality continuous films. For instance, Zhang et al. reported a unique olefin-connected COF nanofiber thin-film electrode for flexible micro-supercapacitors (MSCs).[288] The fibrous morphology of such COF is attributed to the planar triazine molecules in the COF layers. The abundance of triazine molecules significantly strengthens the $\pi$–$\pi$ stacking interactions, which facilitates the orderly growth of the 2D COF layers in a specific direction. This controlled growth ultimately results in the formation of fibrous morphology and good dispersion in certain polar solvents. The resulting MSCs based on fibrous COF films exhibit high area capacitance, wide operating voltage output, high bulk energy density, and excellent cycling capability. Furthermore, some 1D COFs can form dendritic core–shell structures, a property that is not observed in conventional 2D or 3D COFs. The application of these COFs in lithium-ion battery electrode materials enhances the contact area between the active sites and the electrolyte solution. This, in turn, improves the utilization of the active sites and significantly increases the specific capacity of the electrode materials. Concurrently, the robust $\pi$–$\pi$ interaction between the COFs and CNTs facilitates the uniform dispersion of COFs on the surface of CNTs, thereby enhancing the material's rate capability.[289,290]

The performance of electrochemical energy storage devices is closely linked to the electrical conductivity of the materials used. However, POPs exhibit relatively poor electrical conductivity. One of the most prevalent methodologies for enhancing the electrical conductivity of POPs is to cultivate them in situ on conductive materials as a substrate using a hard template on the substrate material.[291] This in situ polymerization method offers several key advantages. First, during polymerization process, reinforcing materials such as carbon nanotubes form noncovalent bonding connections with the polymer, improving interfacial strength. Second, the method avoids particle agglomeration while maintaining particle dispersion. Third, the in-situ method enables the formation of a cross-linked structure, ensuring the structural stability of the material in solution.[292] For the first time, Lyu *et al.* chemically grafted carbon molecular polymers onto CNFs via an as-grown strategy to prepare CNF@CMP hybrid fibers with a specific capacitance of 671.9 mF $cm^{-2}$ for a single fiber.[273] The fully solid-state symmetric fibrous supercapacitor (FSC), obtained through assembly, exhibited a high areal capacitance of 398 mF $cm^{-2}$, an energy density of 18.33 µWh $cm^{-2}$, and a maximum operating voltage of 1.4 V. Furthermore, it demonstrated remarkable flexibility and mechanical stability, indicating significant potential for use in wearable electronic devices. Gao et al. prepared a $\beta$-ketoallylamine-connected diamino-9,10-phenanthrenequinone (DAPQ)–COF*X* composite (where *X* represents the wt.% of CNT) with a tubular core–shell structure and a COF layer tightly grown on the CNT surface via in-situ polycondensation.[293] The synergistic effect of the dual structure of COF and CNT enhanced the utilization of active sites and improved charge transfer efficiency. DAPQ-COF50 exhibited a capacitance of 162 mAh $g^{-1}$ at a current density of 500 mA $g^{-1}$, demonstrating excellent rate performance and cycling stability. Similarly, Liu et al. developed conjugated microporous/mesoporous polymer that was polymerized in situ on CNTs as nanotentacles, using squaric acid (denoted as S) and 1,3,5-Tris(4-aminophenyl) benzene (denoted as T) as monomers.[294] The cross-linked CNT network not only enhanced electron transport in the active material but also addressed the significant drawback of sluggish redox kinetics of polysulfides. The obtained S/ST-CMP@CNT composites exhibited a high initial specific capacity of 1307 mAh $g^{-1}$ at 0.2 C, with a capacity decay rate of only 0.047% per cycle after 500 cycles. These findings suggest significant potential for advancing the development of high-performance lithium-sulfur batteries.

Electrospinning is a technique that applies high voltage to a polymer solution of specific viscosity, overcoming surface tension to emit, elongate, solidify, and collect continuous nanofibers.[295,296] Nanofibrous POPs exhibit high porosity and flexible molecular designability, which enables the creation of a larger contact area between active sites and guest molecules, thereby enhancing the interaction between them. For instance, Liao *et al.* developed a novel nanofiber separator based on COFs with hierarchical porous channels by electrospinning (Figure 6b).[46] The objective of this study was to enhance the ionic conductivity and increase the $Li^+$ transfer number of fast-charging LMBs. The experimental results demonstrate that the assembled Li||LFP full cell exhibits excellent cycling stability, with over 1000 cycles at 10 °C. Moreover, the cell exhibits excellent thermal stability, with the capacity remaining at 97.2 mAh $g^{-1}$ after 300 cycles at 100 °C. These advantages render this LMB fully equipped for normal use in hot climates and broaden the applicability of nanofiber-like separators in energy storage devices. Shortly thereafter, a series of imine-based COFs with varied core structures was designed as cathode, separator, and anode materials, enabling the assembly of an all-nanofiber COF-based battery.[297] This system exhibited a high energy density of 518 Wh $kg^{-1}$ at an ultra-high-power density of 9771 W $kg^{-1}$, with an exceptionally fast charging time of just 56 seconds. Moreover, the device demonstrated a low-capacity decay rate of 0.56% per cycle at 5 A $g^{-1}$ and 100 °C, highlighting its significant potential for electric vehicle applications. A recent breakthrough by Hu's group demonstrated a solid polymer electrolyte (SPE) fabricated via coaxial electrospinning, which achieved a record-high ionic conductivity of $5.48 \times 10^{-4}$ S·$cm^{-1}$ at 30 °C and $3.4 \times 10^{-4}$ S·$cm^{-1}$ at −20 °C, effectively overcoming the low-temperature limitations of conventional polyethylene oxide-based SPEs.[298] This performance was enabled by the intrinsic porous architecture and surface conduction sites of the PAFs framework, which collectively established a continuous 3D $Li^+$ transport pathway. Moreover, the lithium salt-grafted PAFs immobilized the anions within the rigid skeleton, ensuring that $Li^+$ served as the sole mobile charge carrier.

On the other hand, in the context of the increasing demand for portable, flexible electrochemical energy conversion and storage systems, an efficient 3D bifunctional nanofiber electrocatalyst developed by Yuan's group exhibits excellent electrocatalytic activity for both oxygen reduction reaction (ORR; half-wave potential of 0.82 V) and oxygen evolution reaction (OER; 343 mV at 10 mA $cm^{-2}$). It is worth noting that water-based rechargeable Zn–air batteries (ZAB) achieves a favorable power density of 203.63 mW $cm^{-2}$, an outstanding specific capacity of 802.91 mAh $g_{Zn}^{-1}$, in addition to satisfactory mechanical flexibility and


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device stability in the assembled all-solid-state flexible ZAB.[225] In a similar vein, Tang's group developed a solar thermal fuel by attaching azobenzene to polymers of intrinsic microporosity (PIMs), which shows great promise for adaptable solar energy conversion and storage. Its strong energy density ($ED_g$)/enthalpy change ($\Delta H$) is accompanied by a reasonably substantial temperature difference (6.6 °C) observed during the macro-scale heat release performance.[299] This is very important for applications using intelligent temperature control fabrics. Additionally, Zhang's group integrated flexible PEG fragments using a bottom-up self-assembly method into a framework that exhibits high ionic conductivity (0.153 mS $cm^{-1}$ at 30 °C) and can rapidly transport $Li^+$ in a rigid 2D COF architecture through its segmental motion.[47] The pouch cells, as well as the Li|Li and Li|LFP cells assembled at 60 °C, exhibited superior electrochemical performance.

The osmotic energy generated by the osmotic pressure difference between river water and seawater represents a promising renewable energy source.[300] In a seminal study, Pan et al. reported the first instance of a COF dispersion being processed into continuous macro- and meter-scale pure COF fibers using wet-spinning.[284] The fiber-forming ability of the obtained COF, synthesized from 2,5-diaminobenzenesulfonic acid (DABA), 4,4′-diaminostilbene-2,2′-disulfonic acid (DASD), and 1,3,5-triformylphloroglucinol (TFP), and referred to as DABA/DASD-TFP-COF, can be attributed to the presence of numerous sulfonic acid groups in the polymer backbone, which promote excellent dispersion in polar solvents. DABA and DASD serve as amine-functionalized building blocks, each containing sulfonic acid ($–SO_3H$) groups that impart hydrophilicity and ionic conductivity, enabling efficient ion transport in aqueous environments. TFP acts as a tri-aldehyde linker that facilitates the formation of an extended $\pi$-conjugated framework through Schiff-base condensation. The resulting DABA/DASD-TFP-COF fibers exhibited power densities of 70.2 and 185.3 W $m^{-2}$ under 50-fold and 500-fold salt gradients, respectively. These findings underscore the significant potential of COF fibers in blue osmotic energy conversion. Moving forward, electrospinning may be explored to fabricate COF fibers with enhanced functional properties, such as smaller diameters, larger surface areas, and higher porosity, compared to those produced via wet-spinning.

### 4.3. Heterogeneous Catalysis

POPs are an ideal platform for the catalytic application due to their well-defined pore structure, high BET surface area, and diversity of their physicochemical environments. Similarly, electrospinning has been extensively studied in recent years for catalytic applications. Some electrospun nanofibers can act as effective catalysts on their own, but the majority are employed as support components for active catalysts, such as oxide nanoparticles, metals, and enzymes. Generally, catalysts are either encapsulated within the nanofiber structure or deposited on the surface of fiber. The catalyst–support interaction is significantly enhanced by the large surface area and tailored surface chemistry of electrospun nanofibers, leading to improved catalytic activity, selectivity, stability, and reusability. For example, Omar K. Farha's group showed that PIM-1 can serve as a matrix for anchoring pure inorganic Keggin-type polyoxometalates ($H_3PW_{12}O_{40}$). In this approach, the amidoxime functionality of PIMs successfully anchored polyoxometalates particles.[77] The results indicated that the conversion efficiency toward oxidation of 2-chloroethyl ethyl sulfide by $PW_{12}$@PIM-1- amidoxime composite fibers, in the presence of peroxides, was comparable to that of the composite in powdered form. More importantly, the selectivity of the composite fibers was remarkably high (≈86%). Additionally, the recycled composite fiber catalyst maintained a high selectivity of over 72% even after three uses. In addition, atomic layer deposition (ALD) coating is also an effective approach for depositing catalysts on the surface of supporting materials for catalytic applications. Patil et al. demonstrated successful deposition of NiOOH/Ni $(OH)_2$ on the surface of pyrolyzed PIMs fiber. The resulting non-noble metal catalyst on the flexible carbon fibers showed superior performance comparable to state-of-the-art noble metal catalysts.[301] Specifically, it exhibited a low onset potential ($\eta$HER = −40 mV and $\eta$OER = 290 mV versus reversible hydrogen electrode), a small overpotential at $\eta$10 (OER = 390.5 mV and HER = −147 mV), excellent kinetics (Tafel slopes of 50 mV $dec^{-1}$ for OER and 41 mV $dec^{-1}$ for HER), and high stability (over 16 h) in water splitting within an alkaline medium (0.1 M KOH).

Photocatalytic technology has attracted significant attention due to its ability to use solar energy for clean energy conversion and pollutant degradation. Conjugated microporous polymers (CMPs) form a 3D network structure through their $\pi$-$\pi$ conjugated system, which can effectively capture photons and generate electron-hole pairs, thus promoting the photocatalytic reactions.[302,303] Compared to traditional inorganic photocatalysts, CMPs offer adjustable electronic structure and bandgaps, enabling accurate regulation of light absorption range and catalytic activity through molecular design and optimized synthesis methods.[304] In addition, CMPs can also enhance their photocatalytic performance by introducing different functional groups,[305] which improve the separation efficiency of photogenerated carriers and extend their lifetime. CMPs featuring delocalized $\pi$-conjugation bonds represent a distinctive class of photocatalytic materials. For example, as illustrated in Figure 6c, piezoelectric PVDF-co-trifluoroethylene (PVDF–TrFE) can be combined with pyridine-based CMPs for producing hybrid materials. Hybrids containing 50 wt.% modification of the pyridine-based CMP (PCMP) with PVDF–TrFE demonstrated excellent photodegradation efficiency of 97% for rhodamine B (RhB),[306] with a high pseudo-first-order kinetic constant ($k$) of 0.0289 $min^{-1}$. This performance is comparable to that of neat m-PCMP (0.0304 $min^{-1}$) and nearly 15 times greater than the kinetic constant of pure PVDF–TrFE (0.0019 $min^{-1}$). Even after ten repeated cycles, the degradation efficiency remained high at 85%. However, the pores constructed by POPs are prone to being buried within nanofibers during processing. To prevent this issue, adjusting the pore sites in electrospun fibers is essential to preserve their functionality and accessibility. Lee et al. demonstrated that PAN fibers containing polyvinylpyrrolidone (PVP) can be combined with CMPs. After electrospinning, the PVP can be easily removed by washing with distilled water, leaving the fibers with abundant pore sites,[200] These composite fibers, with numerous exposed pore sites, can be used not only for photocatalysis of rhodamine B but also for organic synthesis, such as the oxidization of various benzylamine derivatives. More importantly, the fibers can be recycled through a simple washing process.


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### 4.4. Environmental Applications

POPs have natural porosity and relatively high surface area, offering distinct advantages for environmental applications. However, the powder form of POPs is challenging to recover and reuse effectively. To address these limitations, electrospun POPs-FMs offer effective solutions. In addition, main pollutants, such as dyes, toxic organic substances, gas vapors, and particulate matter, can be more effectively enriched, collected, and recycled using these fiber membranes. With the global rise in water resource consumption driven by population growth and industrial development, water pollution has become an essential issue, often involving dyes, organic compounds, and metals, which seriously affect human health and environmental safety. POP-FMs have demonstrated the ability to absorb dyes or degrade them via photocatalysis. For example, the Uyar group has reported several POP-FMs with effective dye removal capabilities.[307,308] The maximum adsorption capacity of fully hydrolyzed PIM-1 fibers was found to be 157 ± 16 mg $g^{-1}$ for Methylene Blue and 4 mg $g^{-1}$ for Congo red when the adsorption was conducted by 20 mg $L^{-1}$ dye solution without using any dilution. A unique advantage is that these membranes can be used for the direct separation of dye effluents. Selectivity toward harmful cationic pollutants remains a challenging task for environmental applications. To achieve this, decorating the surface of POP-FMs with metal oxides has proven to be a highly suitable method. The Uyar group also showed that the hydrolyzed PIMs combined with ZnO composites can selectively adsorb cationic dyes and degrade them by producing the reactive oxygen species under photoirradiation. Importantly, this membrane remains effective after ten cycles of use. The interaction between dyes and POP-FMs are very important parameters for effective adsorption, Lu et al. demonstrated that the electrostatic interaction of HCPs-based tubular POP-FMs is the main driving force, while the other non-covalent interaction such as $\pi$–$\pi$ stacking, also plays an important role.[57]

Organic pollutants are another form of pollutants, unlike dyes, they often exist in a more insidious form. Li et al. employed an in-situ growth method to cultivate flower-like COFs on the surface of PAN fiber membranes for the adsorption of contaminants in water. The prepared membrane has a specific surface area of 205 $m^2$ $g^{-1}$ and can reach adsorption equilibrium within 30 min.[309] Tests have shown that even after 10 cycles, the membrane maintains a 98% adsorption efficiency for aflatoxin. Additionally, the membrane exhibits good thermal stability up to 300 °C, it also possesses good mechanical properties, with a Young's modulus of 68.5 MPa. The reported PAFs loaded within electrospun PAN fiber have the maximum adsorption capacities for ibuprofen (IBPF), chloroxylenol (CLXN), and *N,N*-diethyl-meta-toluamide (DEET), based on Langmuir models, corresponding to 613.50 mg $g^{-1}$, 429.18, and 384.61 mg $g^{-1}$, respectively.[168] The adsorption comparison experiment showed that the adsorption capacity of PAFs-based POP-FMs primarily originates from PAFs. However, their unique advantage lies in their capability to be separated from the adsorption solution while exhibiting excellent reusability. Even trace amounts of oil pollutants in water can cause significant environmental damage. To address this issue, dip-coating methods have been applied to combine COF with PAN nanofibers.[310] This membrane demonstrates high separation efficiency for different kinds of oils, such as vacuum pump oil and paraffin oil etc. Heavy metal pollutants are harmful and highly toxic even at low concentrations. More importantly, they persist in the environment, bioaccumulate in the food chain, and pose serious risks to health and ecosystems. Herath et al. in situ polymerized guanidine-based ionic covalent organic frameworks on electrospun PAN fiber membranes to prepare hybrid nanofiber membranes.[43] These fiber membranes, due to their positive charge, exhibited a high adsorption rate for the anion Cr (VI) at pH levels below 4, with a maximum adsorption capacity of 173 mg $g^{-1}$ at pH 3. The highest adsorption efficiency for Cr (VI) was achieved at 55 °C. After five cycles, SEM images of the fiber membranes revealed smooth fiber surfaces, indicating excellent regeneration performance.

However, threats to wastewater do not originate solely from organic contaminants. Inorganic radionuclides with high mobility and significant biotoxicity pose an equally severe challenge to water safety. Ma's group developed an ionic porous aromatic framework (iPAF-P67) with exceptional adsorption performance and combined it with a polyethersulfone matrix. Through electrospinning technology, they fabricated continuous nanofiber membranes by stretching the blended solution under a high electric field.[311] The team employed a gravity-driven H-shaped membrane cell design in their process, which enabled the treatment of low-concentration (50 ppb) $ReO_4^-$ solutions. Remarkably, the residual concentration was reduced to 0.123 ppb after nanofiber membrane treatment, meeting the World Health Organization's drinking water standard requirement of 0.159 ppb.

In addition to water pollution, air pollution also presents a major challenge in environmental protection. Water pollutants, which exist in suspended, dissolved, or colloidal states, can be removed through methods like adsorption and filtration. Similarly, air pollutants can be adsorbed and filtered using POPs-based fibrous composite membranes. Electrospun PIM-1 ultrafine fibers have demonstrated the ability to remove aniline from both water and air.[74] Szekely's group developed highly efficient air filtration membranes combining intrinsically microporous PIMs with MOFs, exhibiting excellent adsorption performance. The inherent porosity of PIMs allows for rapid adsorption of air pollutants. The composite fiber membrane can remove polar formaldehyde and non-polar toluene, xylene, and mesitylene from the air,[76] with adsorption capacities of 124, 142, 214, and 201 mg $g^{-1}$, respectively. After four cycles, the removal efficiency of volatile organic compounds (VOCs) only decreased by 5–8%, while fiber morphology remained intact. This study represents the first successful integration of microporous PIMs and MOFs via electrospinning for VOC gas removal (Figure 6d).

Pharmaceuticals and personal care products such as ibuprofen (IBPF), clofibric acid (CLXN), and *N,N*-diethyl-meta-toluamide (DEET) could disrupt hormonal functions and interfere with the endocrine system.[168] Bisphenol A (BPA) is a well-known endocrine disruptor with low biodegradability, causing significant health risks, including cardiovascular diseases. Inspired by the structure of lymphatic vessels, Liao's group designed CMP-based catalytic membranes with a hierarchically porous structure that enables efficient mass transfer and high fluid uptake. These membranes, synthesized using nitrogen-rich poly(tripheneamine) through electrospinning and in situ polymerization, feature a thin layer of cross-linked nanoparticles that activate peroxymonosulfate to generate singlet oxygen,


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effectively degrading over 95% of BPA[37]. Furthermore, the membrane demonstrated high water permeability, achieving a permeance exceeding 2500 L $m^{-2}$ $h^{-1}$ $bar^{-1}$ over 100 h.

Beyond conventional adsorbents, POP-FMs, such as modified PIM-1 fibers, have been reported to absorbed toxic industrial chemicals and chemical warfare agents.[230,312] Recently, HCPs featuring imidazole groups were synthesized using 1,2,4,5-tetrakis(bromomethyl)benzene and 1-benzylimidazole as precursors, resulting in a tetravalent imidazolium monomer. Subsequent hypercross-linking was performed using formaldehyde dimethyl acetal (FDA) as the cross-linking agent. The resulting HCPs demonstrated exceptional affinity and adsorption capacity for the radioactive pertechnetate anion ($^{99}TcO_4^-$).[313] Moreover, these HCPs were successfully incorporated into PAN-based fibers via conventional electrospinning techniques. While the fibrous membranes showed slightly lower adsorption performance compared to the powdered form, they offered significant advantages, including enhanced mechanical stability, recyclability, and ease of handling, making them more suitable for practical applications in radioactive waste remediation. This capability broadens the potential of POP-FMs for environmental applications and even military settings.

Carbon dioxide ($CO_2$), a crucial component of the carbon cycle, plays a significant role as a greenhouse gas, substantially affecting the climate. Parallel to toxicant removal, POP-FMs have been reported to exhibit outstanding $CO_2$ adsorption characteristics. Pan *et al*. demonstrated that amino-functionalized HCPs exhibited an excellent $CO_2$ capture capacity of 35.4 wt.% (8.1 mmol $g^{-1}$).[314] The particle amount and polystyrene segments play an important role in enhancing $CO_2$ adsorption ability. Similarly, PIM-1 can also be electrospun to produce materials with tailored pore sizes, resulting in improved $CO_2$ adsorptive capacity.[315]

### 4.5. Gas Sensor Applications

Gas sensors are crucial in modern industries, ensuring human health and environmental safety.[303] Conventional sensors often utilize inorganic materials due to their low gas detection limits, robust thermal/mechanical properties, and facile mass production. However, inorganic materials, such as semiconducting metal oxide-based gas sensors, have several drawbacks, including high operating temperatures, lack of flexibility, and poor selectivity.[316–318] These issues highlight the advantages of organic materials, particularly polymers, as suitable alternatives.[319–321] Organic polymers offer several benefits, including tunable molecular designs, lightweight and flexible natures, lower operating temperatures, and high selectivity.[322,323] In the POPs-incorporated nanofibers, the hierarchical porous structure has the potential to enhance the reaction kinetics of gas analyte molecules, improving processes such as absorption, desorption, and diffusion.

The COFs are known for promising sensing materials owing to their highly ordered structure, versatile functionalization, and robust characteristics. Thus, their fluorescence properties are attributed to their crystalline porous structures and extended $\pi$–$\pi$ conjugation. As a result, COFs have been employed in various liquid and gas sensing. However, the poor machinability and limited customization of COF powders hinder their practical applications. Electrospun fibers, including COFs materials, can improve the flexibility and customization of chemosensors,[324] core ($TiO_2$)–sheath (COF-136) nanostructured chemiresistor,[325] making them more practical and broadly appealing.

Enantioselective sensing plays a crucial role in asymmetric catalysis, chiral recognition, and optical applications. The Cui group demonstrated that two imine-linked chiral fluorescent COFs can be electrospun with PVDF to produce a free-standing COFs-PVDF composite nanofiber membrane (Figure 6e).[112] With just 10 wt.% of COF powder in PVDF, the membrane exhibited maximum emission at 380 nm, enabling the quantification of decreased percentages. The membrane displayed enantioselectivity toward various chiral vapors, including $\alpha$-pinene, limonene, fenchone, terpinen-4-ol, and carvone. Additionally, the membrane could be easily regenerated by heating at 100 °C under vacuum for ≈1 h.

Chemiresistive gas sensors offer distinct advantages, such as low power consumption, facile fabrication, and compatibility with miniaturization, making them a suitable candidate for next-generation portable gas sensor systems.[326] However, the poor conductivity of POPs limits their application as chemiresistive materials. To overcome this limitation, hybrid structures of POPs are necessary to enable their integration into chemiresistor systems. As an example, the Xu group reported COF-316@$TiO_2$ heterostructure nanowires, which enhance the $NO_2$ sensing performance through a filter-amplifying mechanism.[325] The COF-316 altered the chemical activity of $TiO_2$ by concealing the oxidative sites and amplifying the reductive sites through the photogenerated electrons and holes. This combination of unique COFs properties and heterojunction characteristics, including long-term stability, anti-humidity performance, and the ability to locally concentrate gas analytes, enhances sensing capabilities. In addition, the photoelectronic effect significantly improved sensing performance for both reductive gases (e.g., ethanol) and oxidative gases (e.g., $NO_2$).

Explosives and other hazardous materials pose significant risks in daily life, emphasizing the importance of their accurate detection. The Scherf group reported that CMPs can be electrospun with polylactic acid (PLA) for detecting nitroaromatic explosives. In this study, the resulting nanofibrous film exhibited a high solid-state photoluminescent quantum yield, attributed to aggregate-induced emission effect of the tetraphenylethene monomer. This film showed significant quenching properties toward 1,2-dinitrobenzene (DNB), 1,3,5-trinitrobenzene (TNB), and 1,4-benzoquinone (BQ) vapor.[160] The quenching effect is likely due to the photoinduced electron transfer between tetrakis(4-bromophenyl) ethene based polymer networks (PTBPE) and the electron-deficient analytes, resulting from the offset of their lowest unoccupied molecular orbital energies. Additionally, the system demonstrated good reproducibility. Volatile organic compounds (VOCs), a group of organic chemicals, are known for their ability to vaporize at room temperature. They are commonly present in various industrial and household products. For example, Zhao et al. developed a composite electrospun nanofiber membrane specifically for detecting primary aliphatic amines by processing COFs via electrospinning. COFs, created through a Schiff base reaction and enol-ketone tautomerization, exhibit high crystallinity, stability, and exposed N–H bonds, enhancing interaction with primary aliphatic amines


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and improving fluorescence detection.[327] Under 355 nm excitation, the composite fiber membrane was used for fluorescence sensing of various VOCs gases. The emission signal intensities of *N,N*-dimethylacetamide, acetonitrile, and dimethyl sulfoxide showed slight increases compared to other VOCs, indicating the fiber membrane's distinctive selectivity for diaminopropane. After five regeneration cycles, the nanofiber membrane retained 84% of its fluorescence enhancement, demonstrating excellent structural stability. The composite nanofibers prepared by electrospinning ensured full interaction between VOC molecules with the COF materials, significantly improving sensing performance.

### 4.6. Other Applications

Seawater desalination is a crucial application for producing pure water, particularly in regions suffering from a shortage of safe and clean drinking water. As shown in Figure 6fI, Ma et al. demonstrated that COF-MOF composite fiber materials are promising for desalination processes.[227] In this approach, the COFs nanofiber membrane serves as a porous substrate, while ZIF-8 functions as the ultrathin selective layer. A specialized 2symbiotic layer2 formed between the MOF phase and the COFs nanofibers, combined with their excellent interface compatibility, allows the [TpPa-1]–[ZIF-8] membrane to maintain outstanding selectivity and permeability during 12 h of continuous operation.

Pervaporation is an energy-efficient membrane process used to separate water-alcohol mixtures, a crucial step in biofuel production. Gorgojo's group developed a thin film composite (TFC) membrane consisting of a PVDF nanofiber membrane coated with PIM-1, designed for efficiently recovering n-butanol from water.[328] The synergistic combination of the low mass transfer resistance offered by the electrospun PVDF nanofiber substrate and the microporous PIM-1 layer enabled a high recovery rate of n-butanol. The total permeates fluxes of the PIM-1 TFC membranes exceeded 16.1 kg $m^{-2}$ $h^{-1}$, reaching up to 35.4 kg $m^{-2}$ $h^{-1}$, with n-butanol/water separation factors ranging from 4.8 to 8.

Dimethyl 4-nitrophenylphosphate, a nerve agent simulant, is highly toxic and harmful, even in trace amounts. While MOFs are known to detoxify organophosphorus agents, the integration of MOFs within a porous polymer matrix could enhance their effectiveness in real-world applications as breathable materials. In this context, the Omar K. Farha group demonstrated that PIM-1 can be electrospun and combined with hexa-nuclear zirconium clusters ($Zr_6$). The excellent processability of PIMs facilitates precise control of $Zr_6$ content and prevents cluster aggregation.[329] Notably, with less than 0.63% of the loaded sample, the material achieved hydrolysis half-life of under 1 h. POP-FMs have demonstrated several intriguing properties in food chemistry, particularly as sensors for food safety and quality monitoring.[330,331]

For instance, inorganic arsenic (iAs), a highly toxic contaminant crucial for food safety regulations, has been effectively monitored using these materials. Kang et al. developed a COF synthesized from 1,3,5-trialdehyde phloroglucinol and 3,3'-dinitrobenzidine, which was then combined with PAN via electrospinning, followed by hydride-generation atomic fluorescence spectrometry.[332] The resulting method measured an iAs concentration at 99.3 μg $kg^{-1}$, closely matched the certified value of 92 ± 10 μg $kg^{-1}$. It achieved recoveries ranging from 86.48% to 99.11% and demonstrated a relative standard deviation between 0.40% and 1.18%, highlighting superior accuracy and precision. This approach presents an effective alternative to traditional, more costly methods like high-performance liquid chromatography-inductively coupled plasma mass spectrometry for monitoring contaminants in food. In further research conducted by the Kong group, COFs were co-electrospun into films suitable for immersion operations aimed at removing phytochromes (Figure 6fII).[113] Utilizing just 30 mg of this film proved sufficient to eliminate phytochromes, achieving pesticide recovery rates ranging from 60.52 to 107.35%.

Beyond these environmental applications, Hydrogen storage in MOFs has been a significant research challenge, as the structure of MOFs in forms including particles, fibers, or aerogels greatly affects storage performance. Sonwabo E. Bambalaza and his team enhanced hydrogen storage by co-pelletizing a zirconium-based MOF (UiO-66) with electrospun polymer nanofibers (PIM-1 and PAN), forming composite monoliths.[75] These monoliths retained over 85% of the porosity of the original UiO-66 powder and exhibited a unique microporous-mesoporous structure, unlike the predominantly microporous pristine UiO-66. Notably, the UiO-66/PIM-1 monolith achieved a usable hydrogen capacity of 2.3 wt.%, which is 17% higher than that of the original powder and pellets.

## 5. Concluding Remarks and Future Challenges

### 5.1. Current Status

Based on the VOS (Visualization of Similarities) view analysis, the research hotspots have been redefined and are illustrated in **Figure** 7a. This figure provides a clearer understanding of the relationship between the POPs and electrospinning. The large rectangles represent the major intersections, while the smaller rectangles indicate specific research branches. Interestingly, the HCPs, CMPs, PIMs, COFs, and fibers are positioned at the five corners of the diagram. Notably, these are off-center, with the branches expanding to form a petal-like shape. Meanwhile, this figure does not show the PAFs, which is due to the lack of publications related to PAFs and electrospinning. From this figure, we can also clearly notice that the POP-FMs have been extensively explored in environmental areas such as water treatment, gas separation, dye/heavy metal/oil removal, and particulate matter. The second part is in antibacterial and wound healing applications for biomedical purposes. Composite fibers are often used in catalysis or photocatalysis applications. In the energy area, carbonation is sometimes necessary and often used as the proton exchange membrane in fuel cells. Specifically, for the CMPs, sensors are also an important application direction. Furthermore, several design aspects can also be observed, including integrations with 3D printing and metal–organic frameworks (MOFs). Additional applications include food preservation, chromatographic analysis, and solid-phase extraction.

A variety of features must be considered when developing different types of POPs. While PIMs offer inherent advantages like solubility, their library of available types remains limited. Consequently, creating PIM-based composites is a crucial strategy for enhancing their performance. For instance, PIMs have been mixed with poly(ethylene oxide) (PEO) in acetonitrile and


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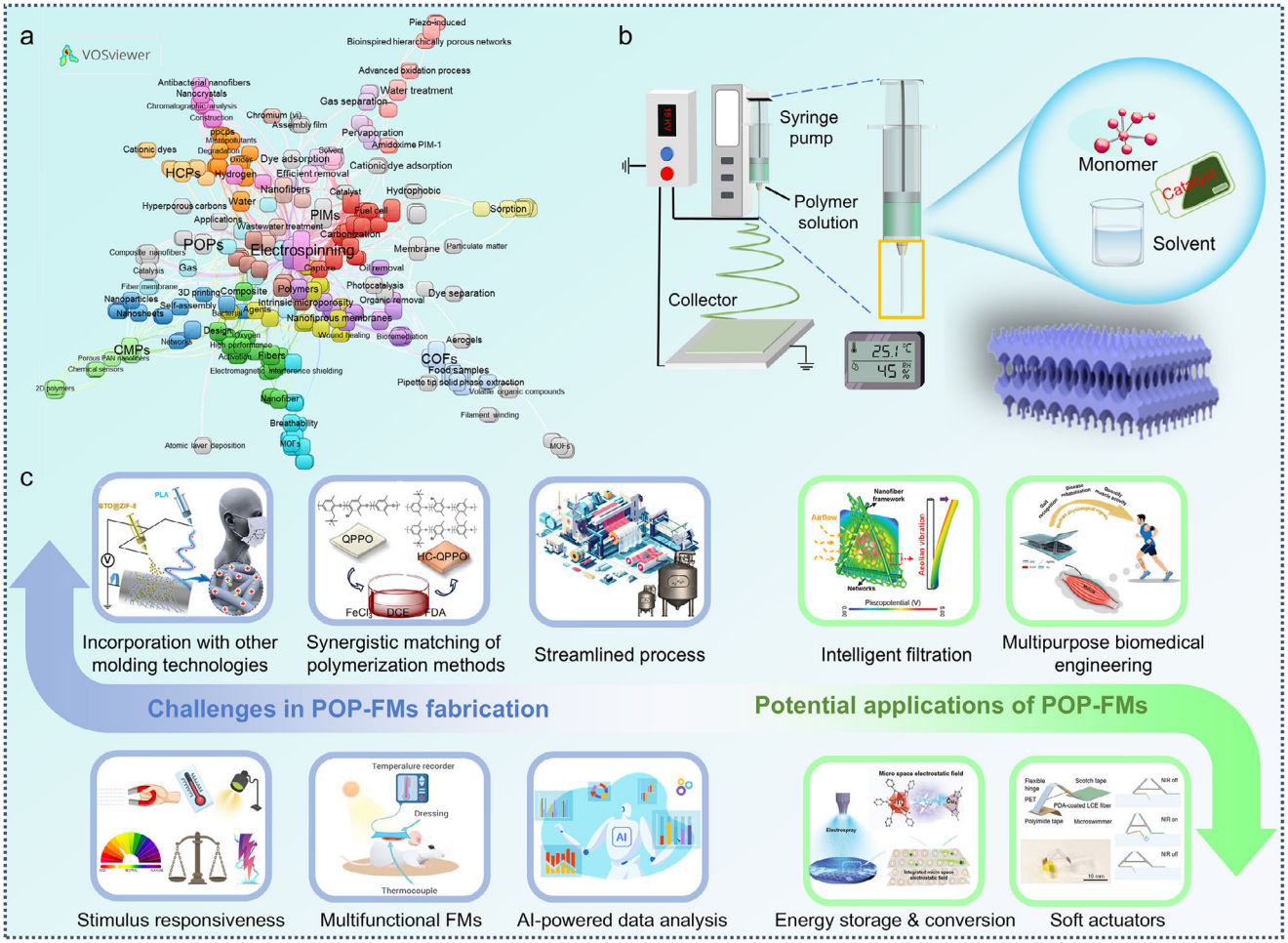


**Figure 7.** a) Visualization analysis diagram of POP-FMs, generated using VOSviewer software, highlights the key hotspots related to POPs and electrospinning. b) Schematic diagram of the preparation model. c) especially the development of preparation challenges and potential applications. Image for "Incorporation with other molding technologies": Reproduced with permission.[337] Copyright 2023, published by American Chemical Society. Image for "Synergistic matching of polymerization methods": Reproduced with permission.[339] Copyright 2024, published by Wiley. Image for "Multifunctional FMs": Reproduced with permission.[358] Copyright 2024, published by Elsevier. Image for "Intelligent filtration": Reproduced with permission.[347] Copyright 2020, published by Wiley. Image for "Multipurpose biomedical engineering": Reproduced with permission.[359] Copyright 2020, published by Elsevier. Image for "Energy storage & conversion": Reproduced with permission.[353] Copyright 2024, published by Wiley. Image for "Soft actuators": Reproduced with permission.[356] Copyright 2021, published by American Association for the Advancement of Science.

processed via conventional casting. The resulting PEO-PIM composite demonstrated significantly improved rate performance, long-term cycling stability, and excellent safety for solid-state Li-S batteries.[333] This underscores the imminent need to find suitable methods, such as electrospinning, for fabricating PIM composites.

The processing of POPs is often constrained by their morphology. For HCPs, morphology becomes fixed post-polymerization, making it essential to utilize their partially cross-linked intermediate state for processing. Previous research has shown that HCP morphology is assembled within the first several ten minutes of polymerization.[52] Extending this cross-linking time is critical for subsequent electrospinning, as fully cross-linked HCPs become difficult to disperse, hindering their shaping. Similarly, CMPs face challenges with insolubility after synthesis. The difficulty in achieving stable dispersion makes post-synthesis redispersion impractical for large-scale applications.

Using pre-designed precursors presents a promising solution. For example, Liao's group recently fabricated a photothermal zwitterionic fibrous membrane from a porphyrin-based CMP precursor and functional antibacterial agents. After electrospinning and polymerization, this bio-inspired membrane effectively disrupted high-salinity gradients to prevent salt deposition, achieving an evaporation rate of 2.64 kg $m^{-2}$ $h^{-1}$ and a photothermal efficiency of 97.6% under 1 kW $m^{-2}$ solar irradiation.[334]

COFs offer another pathway to advanced POP-FMs. COFs can be easily grown on PAN surfaces, enabling the fabrication of free-standing functional membranes. Based on monomer selection, COF-based POP-FMs can be designed as cathodes, separators, or anodes for batteries.[335] As a proof of concept, an assembled battery using such materials delivered a high energy density of 517 Wh $kg^{-1}$, a power density of 9771 W $kg^{-1}$ with a 56-s charging time, and stable high-temperature operation.[297] The application of POP-FMs is highly dependent on their


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morphology: hierarchical foams or gels are ideal for adsorption due to high mass flow and interconnected pores, while membranes and thin films are better suited for photocatalysis, $CO_2$ reduction, and energy devices (e.g., supercapacitors, fuel cells) as they provide high accessibility to active sites and facilitate proton transport.[336]

Finally, the electrospinning of PAFs remains in its early stages. A critical challenge is preserving their extremely high surface area within a fiber matrix. Achieving POP-FMs with a high PAF loading capacity is essential, but effectively dispersing and integrating PAFs into electrospun fibers requires deeper exploration.

### 5.2. Future Challenges in POP-FMs Design

Electrospinning, when applied to POPs, has proven highly effective in fabricating porous films or composites (Figure 7b). The resulting POP-FMs offer several significant advantages, including ease of process, lightweight, unique porous characteristics, tunable surface or doped functionality, controllable diffusion resistance, and enhanced stability through improved surface contact. However, as illustrated in Figure 7c, several challenges persist in both fabrication and potential applications. In particular, comprehensive research into the 3D functionality of POP-FMs is still limited. Moreover, while electrospinning holds great promise, its scalability for mass production on POP-FMs currently falls short compared to the efficiency of traditional wet spinning techniques. To fully harness the potential of POP-FMs, it is essential to advance stimulus-responsive capabilities and integrate multiple functionalities, for mimicking natural materials and enabling multifunctional applications. Although this review provides an in-depth analysis and a comprehensive overview, significant efforts are needed to refine data selection strategies, optimize fabrication process, and tailor POP-FMs for specific target applications. In this context, we propose six key strategies to further advance the development and application of POP-FMs:

1) Incorporation with other material molding technologies, including electrospray and 3D printing.

The fabrication of POP-FMs largely depends on electrospinning, which enables the synthesis of 2D or 3D fibrous composites characterized by hierarchical porosity, comprising inherent polymer porosity, external fibrous surface porosity, and inter-fibrous voids. This process relies not only on the design and functionality of electrospinning itself, including spinneret design, processing parameter optimization, and solvent selection, but also on its integration with other material molding technologies. Combining electrospinning with methods such as electro-spraying or 3D printing offers significant potential for expanding both preparation capabilities and functional applications. For instance, the limited solubility of certain porous polymers in solution often restricts their uniform deposition onto fiber membrane. Simultaneous use of electrospinning and electro-spraying simultaneously can enhance the loading efficiency of POPs onto fibrous surfaces, ensuring more effective incorporation into composite structures.[337] We anticipate that integrating electrospinning with advanced fabrication techniques, such as 3D printing and film coating, will significantly broaden the functional potential of POP-FMs.

2) Synergistic matching of different polymerization methods

Electrospinning can also be effectively integrated with other molding techniques to enable cross-polymerization of POPs or their hybridization with other porous materials. This synergistic approach enhances both performance and efficiency, often delivering enhanced results that surpass the sum of the individual components. For example, incorporating hyper-cross-linked polystyrene as a filler into PIM-1 has been shown to significantly retard aging and improve gas permeability.[338] Additionally, PIMs can be transformed into high-performance ion exchange membranes with well-defined microporous structures through hypercross-linking approaches.[339] COFs and CMPs have also been successfully combined with HCPs.[340–342] Furthermore, hybrid systems that combine POPs with MOFs, silica, and other materials have demonstrated noticeable enhancement in performance.[343] Such composite materials, synthesized through advanced molding and cross-linking techniques, often yield unexpected and beneficial results. Nonetheless, further experimental studies are required to better understand the interplay between multiple materials and polymerization methods and to optimize their integrated functionality.

3) Developing a streamlined process for nanofibers fabrication via electrospinning.

Enhancing the integration of electrospinning with POPs requires a deep understanding of their synthesis, which typically involves liquid-phase polymerization. This process depends on the precise blending of solvents, monomers, and catalysts, along with careful control of reaction conditions such as temperature and agitation. Similarly, successful electrospinning requires the preparation of a well-dispersed polymer slurry mixture under precisely controlled temperature and humidity conditions, as well as fine-tuned processing parameters. To advance both scalability and commercialization of this technology, it is crucial to establish a streamlined process that effectively combines top-down electrospinning techniques and bottom-up polymer synthesis strategies.

4) Stimulus responsiveness: key to enabling optical, magnetic, chiral, pH-responsive, electrical, thermal, and polarity-sensitive functions

Inspired by nature, humans have applied biological principles to develop various functional materials through bionics. Robotics has evolved from traditional rigid structures to soft robots made from flexible materials. Future robots may grow, self-heal, and dynamically alter their shape and function in response to diverse physical and chemical environments. Realizing this vision requires a multidisciplinary approach that combines chemistry, mechanical engineering, materials science, and bioengineering. Smart materials based on POP-FMs are essential for creating artificial systems that seamlessly integrate seamlessly with living organisms, further blurring the line between the synthetic and the biological. Beyond their use in environmental sensors and actuators, these smart materials are crucial in advancing soft robotics and bioelectronics.[344] They are particularly significant in medical innovations, enabling physiological monitoring, minimally invasive surgery, targeted drug delivery, human-computer interfaces, and rehabilitation.

5) Realization of multifunctional fiber materials: essential for enabling diverse, beyond-single-function applications


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For instance, during the pandemic, mask materials were designed to be breathable, stain-resistant, and capable of isolating bacteria and viruses. POP-FMs show immense potential for developing such multifunctional fiber materials. Their unique properties make them suitable for advanced applications such as radiation cooling, atmospheric water harvesting, and optical displays. POP materials combined with electrospinning have proven their efficacy in diverse domains, including biomedical applications, sewage treatment, gas storage, conversion, and energy catalysis. This versatility highlights the need for further integration to fully realize their multifunctional potential. For example, integrating radiation cooling with wound healing through electrospinning technology could accelerate recovery when wounds are exposed to sunlight.[345]

6) Artificial-intelligence-powered data analysis: effective integration with POP-FMs.

Artificial intelligence (AI)-driven data analysis holds great promise for advancing the integration of POP-FMs. Recent data from the Web of Science show tens of thousands of publications in the fields of electrospinning and POPs. Utilizing AI tools, such as machine learning, can significantly enhance parameter optimization. For example, Dai et al. demonstrated that an artificial neural network model can analyze data from existing literature to push the performance boundaries of these technologies.[346] Such approaches enable the identification and preparation of optimal combinations of predicted POPs and electrospinning techniques. By utilizing predicted synthesized data, researchers can design experiments more effectively, thereby enhancing application potential under specifically optimized conditions.

### 5.3. Potential Key Applications

To fully unlock the multifunctional potential of POP-FMs, advancing and integrating their applications across diverse fields is crucial. Strengthening these five key areas—environmental filtration, biomedical engineering, energy, soft actuators, and more—will enable transformative breakthroughs that address current societal and technological challenges.

#### *5.3.1. Intelligent Air/Water Filtration Application*

Healthy lifestyles are closely tied to environmental quality. With global climate change, the demand for clean air and water continues to rise. The development of smart materials for water and air detection, treatment, and recycling, supported by the porous and film-forming characteristics of POP-FMs, offers significant advantages for environmental management. Our investigation highlights the unique potential of these materials in environmental studies. POP-FMs can effectively absorb traditional pollutants such as dyes, organics, and metals while also serving specialized purposes like gas sensing, seawater desalination, particulate matter filtration, and antibacterial applications. This multifunctionality paves the way for developing intelligent air and water treatment materials that can perform a range of tasks,[347] from pollutant adsorption and pathogen elimination to environmental response through color change, signaling the need for replacement.

#### *5.3.2. Multipurpose Biomedical Application*

Drug delivery and antibacterial properties are key research areas in the POP-FMs for biomedical engineering. However, the medical field offers broader applications, including biosensors, bioinformatics, bioprinting, and wearable medical devices. These aspects are crucial for advancing the application of POP-FMs in medicine and remain important areas for further exploration. For instance, POPs have demonstrated effective color response to VOCs,[348] pH changes,[349] and sweat detection,[350] while nanofibers can be utilized for sensing human physiological signals and functioning as antennas for data transmission.[351] These capabilities expand the potential applications of processed POP-FMs. Bio-responsive smart materials can convert biochemical stimulus information into physical or chemical output signals, enabling intelligent medical electronic sensing through intelligent signal reading systems. The precise design of new bio-responsive smart materials and the investigation of their structure-activity relationships are at the forefront of world science and technology. Developing bio-responsive framework materials, such as sensing materials for organic semiconductor sensing chips, can advance the construction of medical electronic devices. By incorporating these materials as functional layers and regulating their chemical composition and micro-nano structures, the researcher can investigate response mechanisms and regulatory methods for medical electronic chips. Monitoring changes in electronic behavior during the stimulation of body fluids and gases, combined with integrating electrospinning technology and POPs, enables the development of effective wearable capabilities and additional functionalities. This approach establishes a mathematical relationship between target substances and signals, facilitating the efficient monitoring of disease biomarkers. Furthermore, creating a database of bio-responsive framework materials and developing AI-assisted design networks could unlock even more possibilities for the biomedical applications of POP-FMs.

#### *5.3.3. Energy Application*

Energy storage and conversion technologies are essential for addressing global challenges such as fossil fuel depletion, environmental pollution, and the uneven distribution of energy resources. Technological advances like supercapacitors, batteries, solar cells, fuel cells, and photocatalysis have had a profound positive impact worldwide. Electrospinning has emerged as a powerful technique for shaping various MOFs into hybrid materials with multiscale porosity and additional functionalities.[352] Although the electrospinning of POPs is still in its early stages, the wide variety of POPs and their compatibility with electrospinning techniques enable diverse applications, making them potentially more versatile than MOFs for energy-related uses.[353] For instance, PIM-1 has been reported to be quantitatively modified with functional groups such as ketones, alcohols, imines, and hydrazones, enhancing its $CO_2$ capture and separation capabilities without affecting solubility.[354]

Additionally, new methods utilizing electrospinning could further optimize PIM-1 for energy applications. COFs have also been incorporated into electrospun PAN nanofibers, creating


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macropores (323 nm), mesopores (7 nm), and micropores (1.6 nm) as separators in lithium metal batteries.[46] Moreover, PS-based block/tri-co-polymers can self-assemble with controlled mesopore sizes; after pre-hypercross-linking steps, these pores can be solidified and converted into porous carbon to support Pt/Fe catalysts.[355] With the continued development of functional POPs and their integration into electrospinning techniques, creating hierarchical pores and enhancing electrolyte transport could significantly improve the performance of energy storage and conversion systems.

#### *5.3.4. Soft Actuator Application*

The stimulus responsiveness of POPs and electrospun fibers imparts exceptional flexibility, enabling them to convert various forms of energy, such as electric fields, light, heat, and magnetic fields, into mechanical motion. These soft actuators are widely utilized in applications, including soft robotics, wearable devices, and haptic/acoustical feedback systems. For instance, Cai et al. demonstrated that liquid crystal elastomers can be electrospun into microfibers that act as actuators.[356] These fibers exhibit a photothermal effect with a polydopamine coating, enabling applications such as micro tweezers, micro swimmers, and microscale impedance pumps. Another study reported that polysulfonated covalent organic frameworks (pS-COFs) can serve as mobile cation hosts in electrochemical soft actuators, with the designed pS-COF-based electrochemical soft actuator showing excellent electrochemical actuation properties and functioning as a soft fluid-switch.[357] Despite these advancements, very few reports on soft robots utilizing POP-FMs currently exist. Integrating photomagnetic responsive polymers into electrospun fibers presents a significant challenge because developing methods to enhance their responsiveness remains an important area for future exploration.

In summary, the future of POP-FMs lies in the seamless integration of advanced fabrication techniques, most notably the versatility of electrospinning, which enables the use of diverse polymer types and precise control over fiber morphology, with the inherent tunability and structural diversity of POPs. This synergistic combination offers unprecedented opportunities for tailoring multifunctional materials across a wide range of applications. As such, a multidisciplinary and holistic approach that integrates material design, structural engineering, and AI-driven optimization will not only accelerate the transition from laboratory-scale innovations to practical applications but also unlock the vast potential of POP-FMs across next-generation technologies, including energy storage, air, and water purification, gas and biosensing, soft actuators, and biomedical systems.

## Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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## Conflict of Interest

The authors declare no conflict of interest.

## Author Contributions

W.S., Y.W., and Y.C. contributed equally to this work. All authors researched data for the article. D.G.Y., G.L., Y.L., and I.D.K. contributed substantially to the discussion of the content. W.S., Y.W., Y.C., X.Z., D.K., and E.S. wrote the article. W.S., Y.W., Y.C., and E.S. crafted all the figures. All authors contributed to the discussion of content and writing of the manuscript, added substantial thoughts, and revised the manuscript in a collaborative manner.

## Keywords

electrospinning, fibrous composite materials, hierarchical porosity, multifunctional applications, porous organic polymers

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