



Electrospun Metal–Organic Framework Nanofiber Membranes for Energy Storage and Environmental Protection

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Abstract

Metal–organic frameworks (MOFs) are attractive in many fields due to their unique advantages. However, the practical applications of single MOF materials are limited. In recent years, a large number of MOF-based composites have been investigated to overcome the defects of single MOF materials to broaden the avenues for the practical applications of MOFs. Among them, MOF-based hybrid nanofiber membranes fabricated by electrospinning combine the advantages of polymer nanofibers and inorganic porous materials, receiving extensive attention and development in energy storage and environmental protection. This review systematically summarizes the recent progress of MOF-based hybrid nanofiber membranes prepared by electrospinning from the perspectives of preparation and application. Firstly, two main methods for preparing MOF/polymer nanofibrous membranes are discussed. Next, the applications of MOF/polymer nanofibrous membranes in energy storage and environmental protection are summarized at length. Finally, to fully tap the potential of MOF-based nanofiber membranes in more fields, the current challenges are proposed, and future research directions are discussed.

Keywords Metal–organic framework · Electrospun · Energy storage · Environmental protection

Introduction

Global population, economic growth, and energy consumption have led to a marked increase in carbon dioxide and other gas emissions [1–6]. Additionally, the problem of water pollution cannot be ignored. Pollutants from wastewater, such as heavy metals, dyes, etc., can bring out severe damage to humans and even endanger lives [7–10]. Based on the above problems, it is particularly imperative to develop materials with excellent performance for energy storage and environmental protection [11–13]. In this connection, various technologies have been developed to realize the devices with high performance for energy storage and environmental protection [14–17]. Common functional materials with porosity, such as zeolites, porous polymers, active carbon and MOFs, have made significant contributions to energy storage and environmental protection [18–23].

Among the above-mentioned functional materials, MOFs are considered to possess great potential for energy and environmental applications owing to their large specific surface area, flexible skeleton and tunable pore sizes [24–31]. MOFs are a new type of porous crystalline materials composed of organic ligands and inorganic metal ions [32–36]. Compared with conventional porous materials, MOFs have a remarkable feature of controlling their structure and composition by employing appropriate metal ions and organic ligands [37–40]. Hence, MOFs possess the characteristics of various designs, controllable structures, simple fabrication, adjustable functions, flexible frameworks, and so on, all of which are attributed to the promising precursors for the preparation of advanced materials with outstanding properties [41–50]. Due to the merits above, MOFs have become candidates for applications in many fields, including supercapacitors, batteries, wastewater treatment, sensors, air pollution filtration, and so forth [51–61]. However, due to the inherent defects of MOFs (such as traditional morphologies and processing difficulties), single MOF materials can hardly satisfy the requirements of practical applications [62–64]. To solve these problems, researchers have done many studies on MOF composites. MOF composites formed by combining MOFs with other functional materials (such as polymers,

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nanoparticles, carbon substrates, conductive substrates) possess physico-chemical properties that the pristine MOFs cannot provide, such as excellent electrical conductivity, mechanical stability, and catalytic performance. In recent years, a large number of MOF composites have exhibited outstanding stability and excellent physico-chemical properties under harsh environment (highly acid/highly alkali). By introducing suitable polymers and necessary molding techniques, MOF composites are expected to play an important role in energy storage and environmental protection [65]. MOFs are usually synthesized by traditional solvothermal or hydrothermal methods. At the same time, polymers as adhesives can not only enhance the mechanical flexibility of composites, but also ensure the chemical stability of materials. The membrane materials formed by combining MOFs with polymer-carriers have the characteristics of integrity, porosity and flexibility, which can effectively expand the practical applications of MOFs thanks to the combination of the advantages of porous nanoparticles and polymers [66–68].

Electrospinning is a proven and effective method for the direct and continuous preparation of polymer nanofibers [69–75]. Due to the advantages of stable performance, excellent quality, comprehensive spinnable materials and controllable process, it has an extensive application prospect in energy storage and environmental protection [76–90].

In recent years, MOF-based electrospun polymer nanofiber membranes have been widely used in gas sensing, gas separation, air pollution filtration, energy storage, and wastewater treatment [13, 91–101]. MOF/polymer electrospun nanofiber membranes exhibit the following advantages: (I) owing to the outstanding permeability and excellent mechanical strength of polymer electrospun nanofibers, they are considered the ideal frame structure to support MOF particles. These features facilitate the transformation of MOF nanoparticles into self-supporting MOF-based nanofibrous membranes; (II) MOF/polymer composite fibers combine the merits of MOF nanoparticles with various crystal structures as well as synthetic physicochemical properties and electrospun nanofibers, e.g., outstanding flexibility, controllable morphology and lightweight, which are beneficial to commendably realize the synergistic effect and thoroughly expand the applications of the two materials [102–107]; (III) the combination of MOF and electrospinning nanofibers can mitigate the misgivings that particles are easy to agglomerate and difficult to recycle [56, 65, 108, 109]. Consequently, MOF/polymer nanofiber membranes are considered as available functional materials to expand the applications of MOFs, such as environmental protection and electrochemical energy storage [64, 110–116]. Hitherto, two main routes to fabricate MOF-loaded electrospun nanofiber membranes have been developed, namely “direct electrospinning” and “surface in situ growth.” The direct electrospinning route requires only a simple three-step process: synthesis of MOF

powder nanoparticles, preparation of slurry by adding MOF nanoparticles into polymer solutions, and direct electrospun of the slurry. The prerequisite for the successful preparation of nanofiber membranes through this strategy is the chemical compatibility between MOF, polymer and solvent components. The preparation of MOF/polymer nanofiber membranes by surface in-situ growth strategy requires two steps: electrospun of a polymer nanofiber layer and evolution of MOF crystals in the open pores and on the surface of the nanofiber layer. This approach requires the use of relatively stable polymer nanofibers that can endure the preparation conditions of target MOF particles, which may need to consider the use of corrosive solvents or relatively high pressures and temperatures (solvothermal or hydrothermal conditions).

This review systematically summarizes the latest progress of MOF-based electrospun nanofiber membranes. Although there have been many reviews in this research field, the summary of the fabrication routes of MOF-based nanofiber membranes is still not very detailed. In this review, the preparation routes of MOF/polymer nanofiber membranes are illustrated, and the differences, advantages as well as disadvantages of direct electrospinning and surface in situ growth are introduced in detail, which is obviously different from other reviews. Next, the main applications of MOF/polymer nanofiber membranes in energy storage and environmental protection are discussed at length. Finally, the key challenges in the above fields are proposed, and some relevant ideas for future research are put forward. This review will provide essential insights into the rational design of electrospinning fibers for energy and environmental applications in the future and facilitate the development of more advanced functional materials in various industries.

Routes for the Fabrication of MOF/Polymer Nanofiber Membranes

MOF/polymer nanofiber membranes are generally acquired by electrospinning. Electrospinning is a unique nanofiber manufacturing process in which polymer solution systems are jet-spun under the action of high-voltage static electricity [117–120]. Numerous polymer solution systems have been widely used in electrospun, such as polyvinylpyrrolidone (PVP) in ethanol (EtOH), polyacrylonitrile (PAN) in *N,N*-dimethylformamide (DMF), polystyrene (PS) in tetrahydrofuran (THF). The nanofibers prepared by electrospinning possess the features of large specific surface area, high porosity, easy size control and surface functionalization (such as surface coating and surface modification). Thanks to these advantages, the prepared nanofibers exhibit excellent application value in many fields. Two main ways have been excavated for the fabrication of MOF/polymer fiber

membranes based on electrospun, which are direct electrospinning [121] and surface in situ growth [122]. Figure 1 displays the MOF/polymer nanofiber membranes prepared by these two routes. This section discusses the two routes for preparing MOF/polymer nanofiber membranes. The information on fabrication routes is summarized in Table 1.

Direct Electrospinning

MOF/polymer nanofiber membranes are generally prepared by direct electrospinning with a simple three-step preparation: synthesis of MOF powder nanoparticles, preparation of slurry by adding MOF nanoparticles into polymer solutions, and direct electrospun of the slurry [123]. The prerequisite for the successful preparation of nanofiber membranes is chemical compatibility between MOF, polymer and solvent components. Furthermore, the polymer/solvent system, the slurry performance and the electrospinning conditions are

optimized according to the specific application of the target MOF and polymer system [124, 125]. Therefore, MOF/polymer nanofiber membranes with different properties are fabricated.

MOF/polymer nanofibrous membranes can be successfully prepared by electrospinning of MOF/polymer slurries obtained by mixing MOF particles with different polymer/solvent systems. For instance, the supported microporous membranes prepared seminally by direct electrospinning were used for gas separation for the first time [123]. Well-grown and defect-free zeolitic imidazolate framework-8 (ZIF-8) membranes were successfully prepared using ZIF-8 nanoparticles to cover macroporous silica (SiO_2) support tubes with MOF/polymer fiber layers. The direct electrospun process of a mixed slurry of ZIF-8/PVP is shown schematically in Fig. 2a. Figure 2b displays the top-view of the ZIF-8/PVP nanofiber membranes. It shows that ZIF-8 crystals have a rhombic dodecahedron morphology, completely

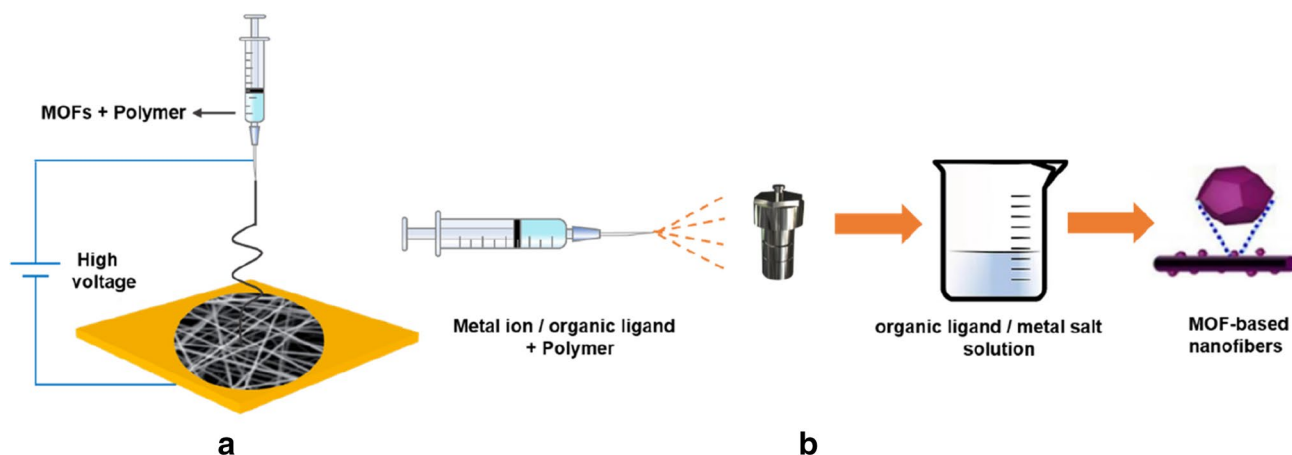


Fig. 1 **a** Direct electrospinning; **b** electrospun metal ion/polymer or ligand/polymer nanofibers followed by surface in situ growth of MOF particles

Table 1 Information about the fabrication routes of MOF/polymer electrospun fiber membranes

MOF	Metal center	Ligand	Composite nanofibers	Route	References
ZIF-8	Zn	2-methylimidazole	ZIF-8/PVP	Direct electrospinning	[123]
ZIF-8	Zn	2-methylimidazole	ZIF-8/PI	Direct electrospinning	[126]
ZIF-8	Zn	2-methylimidazole	ZIF-8/PAN	Surface in situ growth (I)	[133]
ZIF-67	Co	2-methylimidazole	ZIF-67/PAN	Surface in situ growth (I)	[134]
ZIF-8	Zn	2-methylimidazole	ZIF-8/PAN	Surface in situ growth (II)	[105]
MIL-100	Fe	Trimesic acid	MIL-100/PAN	Surface in situ growth (II)	[135]
UiO-66-NH ₂	Zr	Amino-terephthalic acid	UiO-66-NH ₂ /PAN	Surface in situ growth (II)	[136]
ZIF-8	Zn	2-methylimidazole	ZIF-8/PI	Surface in situ growth (III)	[137]
ZIF-8	Zn	2-methylimidazole	ZIF-8/PU	Surface in situ growth (III)	[138]
ZIF-8	Zn	2-methylimidazole	ZIF-8/ESF	Surface in situ growth (III)	[139]
ZIF-67	Co	2-methylimidazole	ZIF-67/ESF	Surface in situ growth (III)	[139]
UiO-66-NH ₂	Zr	Amino-terephthalic acid	UiO-66-NH ₂ / PAN	Surface in situ growth (III)	[140]

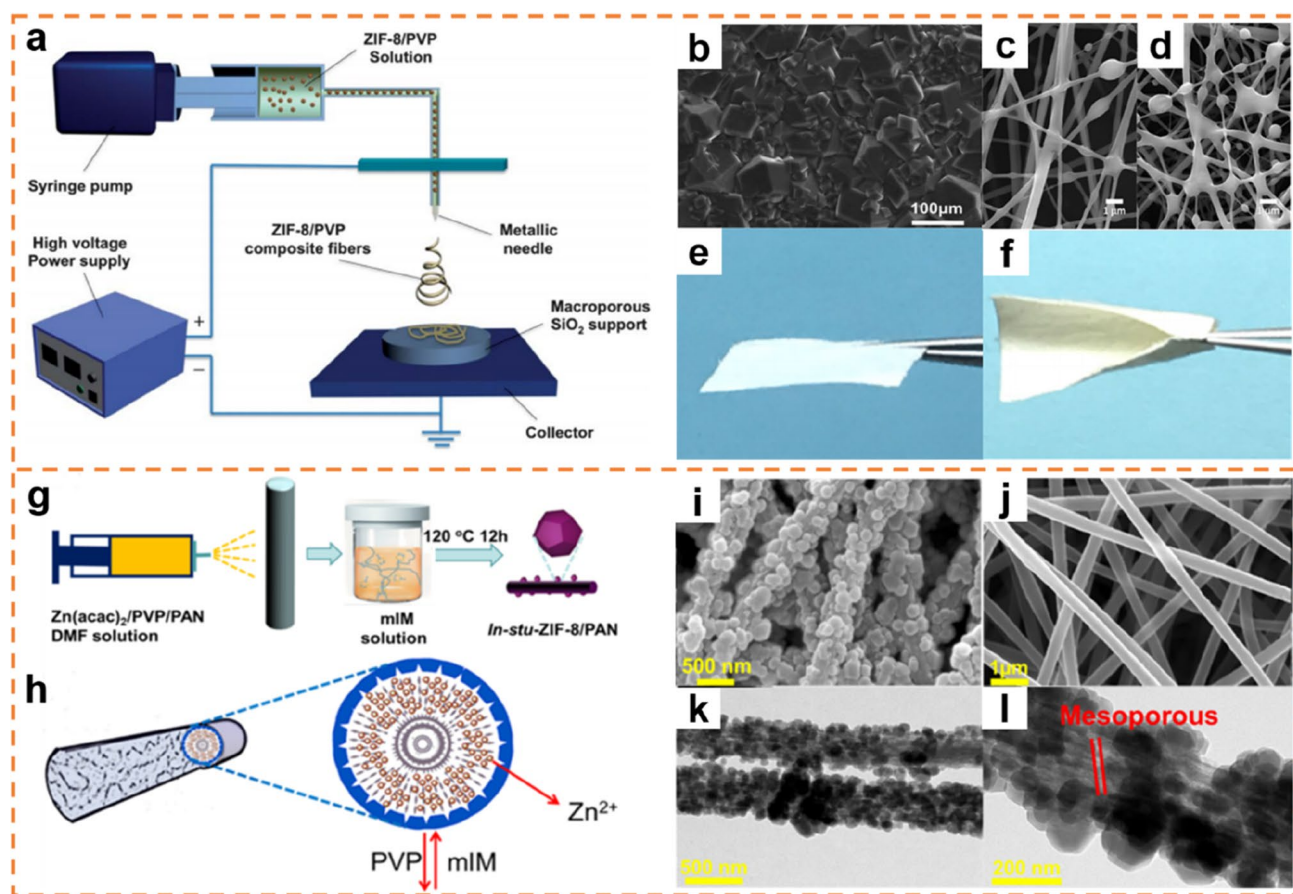


Fig. 2 **a** Schematic diagram of direct electrospinning process for ZIF-8/PVP fiber layers on macroporous SiO₂ support tubes; **b** top-view of the ZIF-8 membranes containing ZIF-8/PVP fibers. Reproduced with permission from Ref [123]. Copyright 2012, the Royal Society of Chemistry. SEM pictures of **c** PI and **d** ZIF-8/PI membranes after filtering PM_{2.5} in the air; exhibition of ZIF-8/PI membrane **e** stiffness and **f** flexibility. Reproduced with permission from

Ref [126]. Copyright 2019, American Chemical Society. **g** Schematic diagram of the in situ growth of ZIF-8 on electrospinning nanofibers; **h** formation mechanism of the in situ ZIF-8/PAN nanofibers; SEM images of **i** the in situ ZIF-8/PAN nanofibers and **j** PAN; **k**, **l** TEM images of the in situ ZIF-8/PAN nanofibers. Reproduced with permission from Ref [133]. Copyright 2018, American Chemical Society

coating the support surface and growing well, which proves that the composite membranes were defect-free. Similarly, the ZIF-8/polyimide (PI) nanofiber membranes were successfully prepared by direct electrospinning [126]. Figure 2 c and d reveal the microstructures of PI and ZIF-8/PI fiber membranes after filtering particulate matter with an aerodynamic diameter of less than 2.5 μm (PM_{2.5}), respectively. The ZIF-8/PI membranes exhibit excellent deformation resistance and independence owing to the high modulus in the initial stage of deformation (Fig. 2e). Simultaneously, the membranes with increased stiffness still maintain sufficient flexibility (Fig. 2f).

Surface In situ Growth

The activity and controllability of MOF-based nanofibrous membranes prepared by direct electrospinning are often limited in practical applications. Details are as follows. (I)

Because the MOF particles are embedded in the polymer nanofibrous membranes, the internal pores of the pristine MOF crystals tend to be coated by the polymer matrix, which greatly limits the activity of MOFs in most applications. (II) Excessive MOF crystals may affect the conductivity of the electrospinning solution, resulting in electrospinning failure. Therefore, the controllability of the amount of MOF nanoparticles introduced is very critical in practical applications.

The route of surface in situ growth can ideally overcome these drawbacks [127, 128]. The brilliance of this approach is that the internal and the surface pores of original MOF particles are not coated by polymer matrix and are commendably controllable. Moreover, the physicochemical properties of the MOF particles remain unchanged. MOF/polymer nanofiber membranes were fabricated initially by surface in situ growth with a simple two-step process: electrospun of a polymer nanofiber layer and evolution of

MOF crystals in the open pores and on the surfaces of the nanofiber layer [129, 130]. There are three main ways of surface in situ growth. They have similar growth paths and features, that is, polymer nanofiber membranes need to be immersed in the corresponding precursors. (I) Metal ions are brought into the polymer nanofibers by electrospun, and then the metal ions/polymer electrospinning nanofiber membranes are soaked in the organic ligand solution to load the target MOFs by surface in situ growth; (II) ligands are introduced in the polymer fibers by electrospun, and then the ligands/polymer electrospinning fiber membranes are soaked in the metal salt solution to load MOF nanocrystals, and (III) MOF crystals are grown directly on the surface of polymer electrospun nanofiber membranes containing some nucleation sites [131, 132]. These three in situ growth routes are not only feasible and simple, but also effectively avoid the problem of poor compatibility between MOFs and polymers. However, these three paths also have limitations. For example, (I) the relatively stable polymer nanofibers are supposed to be used to ensure that they can withstand the preparation conditions of the target MOF particles, which may require consideration of the employment of aggressive solvents or relatively high pressures and temperatures (solvothermal or hydrothermal conditions); (II) the nucleation of MOF crystals is difficult to control, resulting in the shedding of the grown MOFs and the difficulty in controlling the crystal thickness; (III) the reaction temperature, reaction time and concentration of the precursor solution are supposed to be appropriate for MOF nanoparticles growth. These three growth paths are discussed in detail below.

Metal Ion/Polymer Electrospun

The metal ion is brought into polymer nanofibers by electrospun, and then MOF nanocrystals load on fibers by immersing metal ion/polymer into ligand solution. There are many works to fabricate nanofibers based on this approach, which pave the way for the great development of MOF-based fiber membranes. For example, the ZIF-8-based polymer nanofiber (ZIF-8/PAN) membranes fabricated seminally by surface in situ growth have been reported, and the preparation process is illustrated in Fig. 2g [133]. Firstly, the mixture of PVP and PAN was added into the $\text{Zn}(\text{acac})_2$ solution to obtain the spinning precursor. Then, the $\text{Zn}(\text{acac})_2$ /PVP/PAN nanofibers fabricated by electrospinning were soaked into a 2-methylimidazole (mIM) solution to obtain ZIF-8/PAN nanofiber membranes. PVP is easier to dissociate from composite nanofibers under hydrothermal conditions because PVP is more hydrophilic than PAN (Fig. 2h). In situ growth of a homogeneous ZIF-8 crystal layer on the nanofibers' surface was confirmed by scanning electron microscopy (SEM) characterization, and the surface of the fibers with ZIF-8 crystals (Fig. 2i) was rougher than that

of the fibers without ZIF-8 crystals (Fig. 2j). Furthermore, transmission electron microscopy (TEM) characterization demonstrated that ZIF-8 crystals grew not only on the surface of the nanofibers, but also inside the PAN fibers by in situ growth (Fig. 2k and l). And the high magnification TEM picture brought the mesoporous structure of ZIF-8/PAN fibers into sight (Fig. 2l). Similar research was pointed out by Bian et al. [134] They prepared $\text{Co}(\text{AC})_2$ /PAN fiber membranes by electrospun and then immersed the composite fiber membranes into mIM solution dissolved in ethanol at room temperature to incorporate ZIF-67 crystals into the $\text{Co}(\text{AC})_2$ /PAN electrospun membranes by surface in situ growth. As depicted in Fig. 3a, ZIF-67/PAN electrospun fiber membranes were finally obtained. Interestingly, a layer of evenly distributed ZIF-67 crystals was observed on the surface of the nanofiber membranes by SEM (Fig. 3b–g) and TEM (Fig. 3h–j), which made the surface of the fiber rougher. Introducing metal ions into nanofibers by electrospinning and then obtaining uniformly dispersed MOF particles by in situ growth is simple and easy to operate. It plays an important role in the preparation of polymer nanofibers.

Ligand/Polymer Electrospun

Immersing ligand/polymer substrates prepared by electrospinning into a metal salt solution to grow MOF particles by in situ generation has been widely used to fabricate electrospun fiber membranes. Blending mIM solution in polymer nanofibers and then soaking the composite fibers into the solution of $\text{Zn}(\text{OAc})_2 \cdot 2\text{H}_2\text{O}$ dissolved in methanol to load MOF nanocrystals by in situ growth has been reported in detail by Ma et al. [105]. Figure 3k illustrates the synthesis procedures of ZIF-8/PAN fibers. From the microstructure of the composite (Fig. 3l), it can be concluded that ZIF-8 crystals grew along the nanofibers and adhered to the PAN substrate, which brought about stronger adhesion [123] and ideally overcame the difficulty of nanofiber surface pretreatment. Furthermore, the SEM images (Fig. 3m, n) intuitively demonstrated that the surface of the PAN substrate was coated with a layer of dense and homogenous ZIF-8 crystals [104], indicating that the ZIF-8/PAN electrospun fiber membranes were successfully prepared. Additionally, Zhao et al. immersed trimesic acid (H_3BTC)/PAN blend electrospun fiber membranes prepared by electrospinning into FeCl_3 solution dissolved in pure water [135]. They found that trimesic acids from H_3BTC /PAN fiber membranes acted as the nucleation sites for MIL-100(Fe) grown in situ on electrospun fibers during the hydrothermal reaction, and a compact and continuous layer of MIL-100 particles was grown in situ on the surface of the fiber membranes (Fig. 4a). As depicted in Fig. 4b, c, the PAN surface was coated entirely by the MIL-100 crystals, which grew along the nanofibers and were well-aligned after a two-step process. According

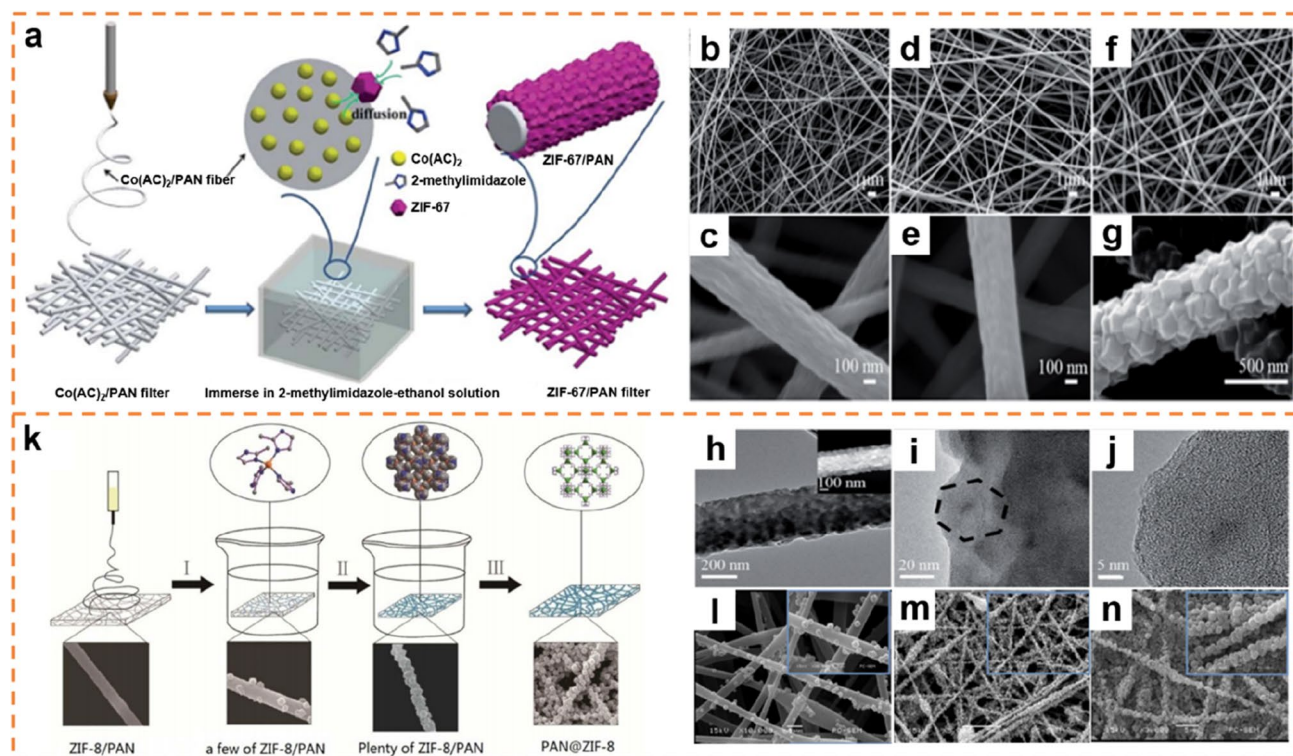


Fig. 3 **a** Schematic diagram of the in situ growth of ZIF-67 on electrospinning fibers; SEM photographs of **b**, **c** the pure PAN fibers, **d**, **e** $\text{Co}(\text{AC})_2/\text{PAN}$ fibers, and **f**, **g** ZIF-67/PAN fibers; **h** dark-field and bright-field TEM photographs of the ZIF-67/PAN fibers; **i**, **j** high-resolution TEM photographs of the ZIF-67/PAN fibers. Reproduced

with permission from Ref [134]. Copyright 2018, the Royal Society of Chemistry. **k** Schematic diagram of the fabrication procedures of PAN@ZIF-8 electrospinning fibers; SEM pictures of **l** ZIF-8 on the PAN and **m**, **n** the ZIF-8/PAN nanofibers. Reproduced with permission from Ref [105]. Copyright 2016, the Royal Society of Chemistry

to Liu et al., PAN electrospun fiber membranes blended with amino-terephthalic acid (ATA) ligands were soaked into a solution of ZrCl_4 dissolved in acetone, and a uniform layer of UiO-66-NH_2 particles was evenly grown in situ on the surface of fiber membranes with ATA ligands acted as the nucleation sites under solvothermal conditions, as shown in Fig. 4d–g [136].

MOF/Polymer Electrospun

Due to the distinct chemical properties of MOF and polymers, it is difficult to achieve direct bonding between MOF and polymer surfaces. To solve this problem, serving polymer nanofiber membranes containing nucleation sites (such as carboxyl) as the substrate, MOF crystals are grown in situ on the surface of nanofiber membranes through the interaction between nucleation sites from electrospinning nanofiber membranes and metal ions from MOFs, which can provide outstanding load efficiency for MOFs. The driving forces for the growth of MOF crystals mainly come from the interaction between specific functional groups. The ideal in situ growth of ZIF-8 crystals can be achieved on the surface of PI electrospun fiber membranes [137]. The surface of PI fiber

membranes contains a large number of carboxyl groups, which can form a hydrogen bonding interaction with the imino groups from the ligand (mIM) of ZIF-8. It can be seen from SEM that ZIF-8 crystals were uniformly loaded on PI electrospun membranes. Zhou et al. took polyurethane (PU) electrospun fiber membranes with concentrated chromic anhydride and sulfuric acid-functionalized surface as the growth substrate of ZIF-8 (Fig. 5a) [138]. The functionalized fiber surface increased the binding sites, which was favorable for the fibers to capture Zn^{2+} . Figure 5b–e reveals the SEM images for in situ growth of ZIF-8 on PU. A strategy to fabricate electrospun-silk-nanofiber (ESF) membranes containing a layer of dense and uniform ZIF-8 or ZIF-67 crystals was proposed by Li et al. [139] Compared with the conventional polymer fiber membranes, the density and amount of MOF crystals loaded on the surface of fibers were obviously improved owing to the arrangement of binding and nucleation sites on the ESF membranes. In other findings, the electrospinning PAN fiber membranes soaked into NaOH and HCl solutions sequentially were engaged in the solution of UiO-66-NH_2 for in situ generations [140]. The surface of PAN fiber membranes was covered with a layer of homogeneous UiO-66-NH_2 particles by the interaction

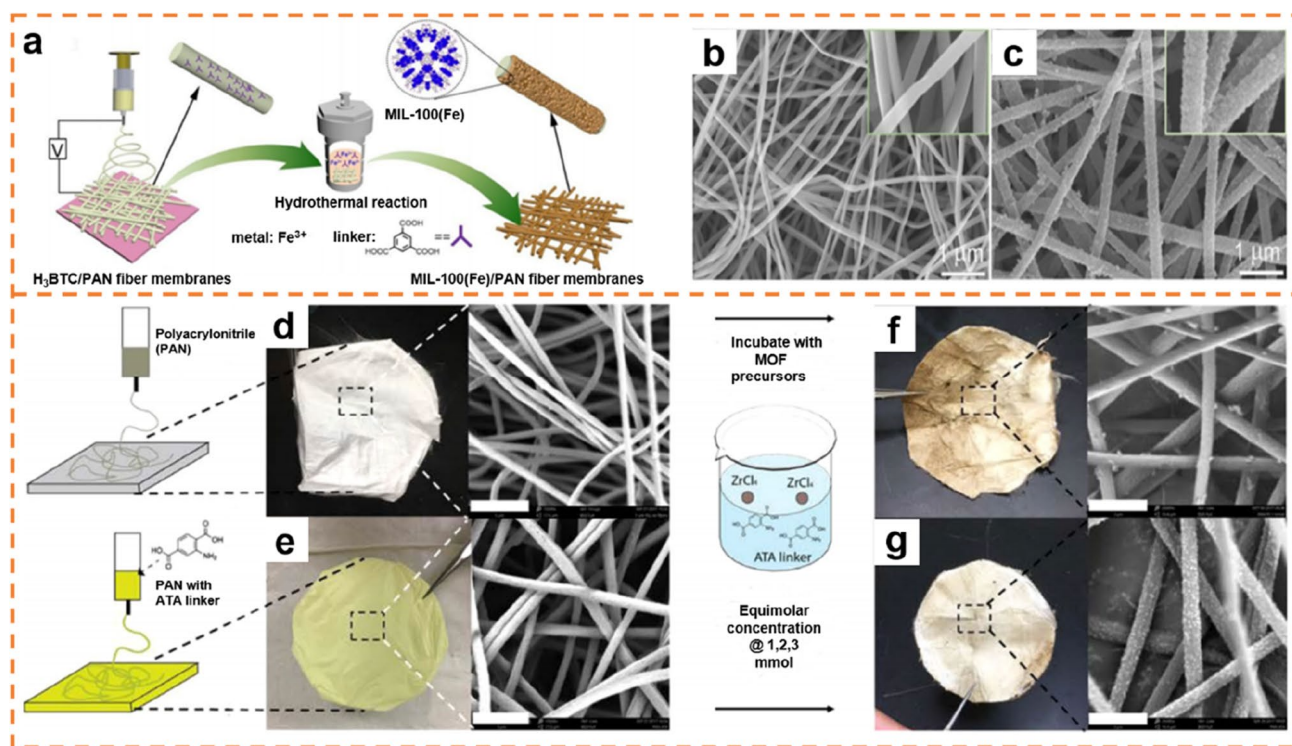


Fig. 4 **a** Schematic diagram of the fabrication of MIL-100/PAN fiber membranes. SEM pictures of **b** H₃BTC/PAN fiber membranes and **c** MIL-100/PAN fiber membranes. Reproduced with permission from Ref [135]. Copyright 2019, the Royal Society of Chemistry. Schematic diagram of the preparation of UiO-66-NH₂/PAN fiber mem-

branes by in situ growth of UiO-66-NH₂ on electrospinning fibers: **d** control PAN sample; **e** PAN with ATA; **f** control PAN post-synthesis; **g** PAN with ATA post-synthesis. Reproduced with permission from Ref [136]. Copyright 2017, American Chemical Society

between the amino groups from the MOFs and the C=O bond from carboxyls on the surface of the fibers, as depicted in Fig. 5f.

Applications of MOF Nanofiber Membranes

MOF nanofibrous membranes play a significant role in energy [141–143] and environmental [144–148] applications. This section reviews the applications of MOF nanofiber membranes in energy storage and environmental protection, and summarizes such information in Table 2.

Batteries

Lithium-ion batteries (LIBs), lithium-sulfur batteries (LSBs) and sodium-ion batteries (SIBs) are the most widely used energy storage devices in our lives, which play an important role in mobile phones and computers [149]. Generally, the properties of the battery are determined by the electrode materials. Consequently, abundant researches have been conducted to explore excellent electrode materials with acceptable rate performance, high specific capacitance and outstanding cycle performance [150–154]. Thanks to the

large specific surface area, hierarchical porous structure and exceptional conductivity of MOF derivatives (such as carbonized MOF nanofibers), they are considered as potential electrode material candidates.

Lithium-Ion Batteries

Carbonized MOF nanofibers are generally used as self-templates or precursors to develop porous metal oxides for electrode materials [155–159]. These materials can not only improve the pore size distribution and the specific surface area of the contact area between the electrolyte and the electrode, but also supply extra active centers to effectively reduce the diffusion path of lithium ions, bringing about the enhancement of the electrochemical properties [160–162].

Advanced anode materials for lithium storage have been intensively studied. For example, inspired by the high nitrogen (N) content and the porous structures of MOF derivatives, a strategy combining electrospinning and the subsequent hydrothermal reaction was designed to prepare Molybdenum disulfide (MoS₂) based nanofibers with a layered structure, in which thin MoS₂ nanosheets were vertically assembled on the N-doped porous carbon nanofibers (PCNFs) derived from PAN-MOF [115]. As visualized in

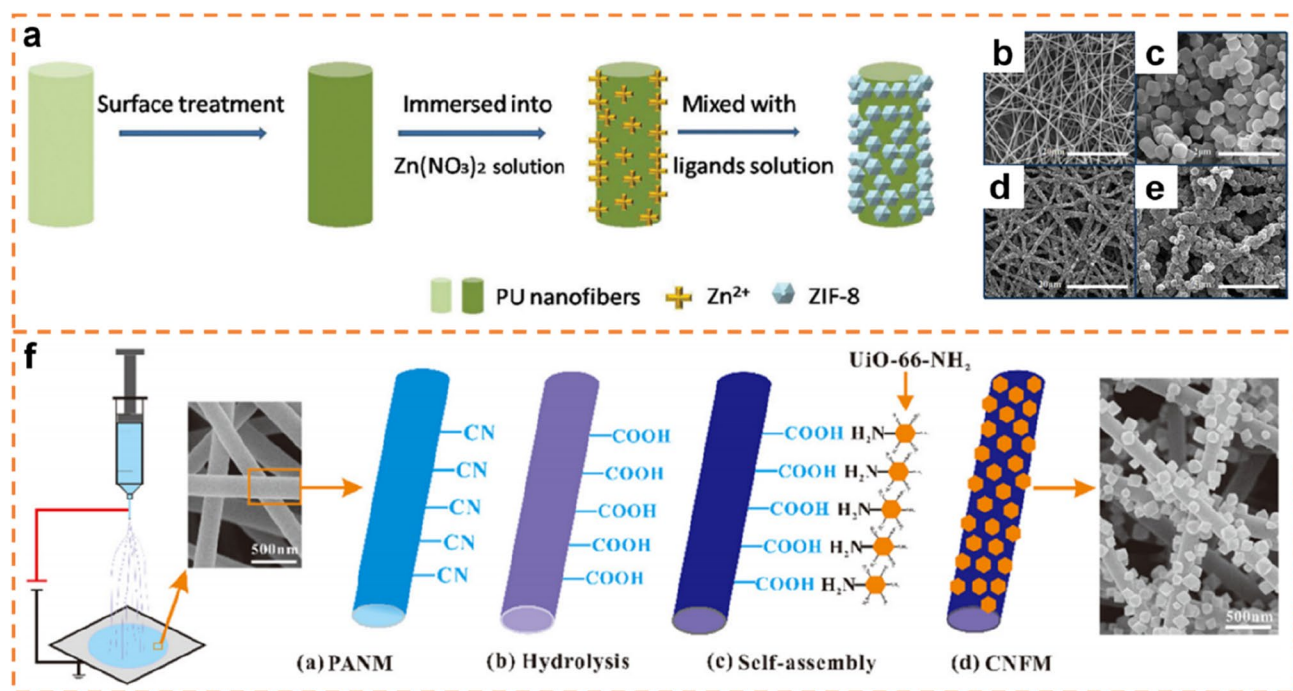


Fig. 5 a Schematic sketch for in situ growth of ZIF-8 crystals on PU fibers; SEM pictures for in situ growth of ZIF-8 on PU: **b** PU; **c** ZIF-8; **d**, **e** ZIF-8/PU. Reproduced with permission from Ref [138]. Copyright 2014, the Royal Society of Chemistry. **f** Schematic sketch

of the fabrication of PAN fiber membrane covered with UiO-66-NH₂. Reproduced with permission from Ref [140]. Copyright 2019, Elsevier

Table 2 Information about the MOF/polymer electrospun fiber membranes for energy and environmental applications

MOF	Composite nanofibers	Route	Application	References
ZIF-8	ZIF-8/PAN	Direct electrospinning	Lithium-ion batteries	[115]
ZIF-67	ZIF-67/PAN	Surface in situ growth (I)	Lithium-ion batteries	[163]
ZIF-67	ZIF-67/PAN	Surface in situ growth (I)	Lithium-sulfur batteries	[175]
ZIF-67	ZIF-67/PAN	Direct electrospinning	Lithium-sulfur batteries	[176]
ZIF-8/ZIF-67	ZIF-8/ZIF-67/PAN	Surface in situ growth (II)	Sodium-ion batteries	[193]
MIL-88A(Fe)	MIL-88A/PAN	Surface in situ growth (III)	Sodium-ion batteries	[194]
MOF-808-EDTA	MOF-808-EDTA/PAN	Direct electrospinning	Heavy metal ions, oils	[205]
ZIF-67	ZIF-67/CA	Surface in situ growth (II)	Heavy metal ions	[206]
bio-MOF-1	bio-MOF/PAN	Direct electrospinning	Dyes	[212]
ZIF-8	ZIF-8@GO/PLA	Surface in situ growth (III)	Dyes	[213]
MIL-53(Al)-NH ₂	MIL-53-NH ₂ /PAN	Surface in situ growth (III)	PM, SO ₂ , O ₃	[220]
ZIF-8, UiO-66-NH ₂ , MOF-199, Mg-MOF-74	ZIF-8/PAN, UiO-66-NH ₂ , MOF-199/PAN, Mg-MOF-74/PAN	Direct electrospinning	PM _{2.5} , PM ₁₀ , SO ₂	[221]
ZIF-8	ZIF-8/PI	Direct electrospinning	PM _{2.5}	[126]
HKUST-1	HKUST-1/PAN	Direct electrospinning	CO ₂	[225]

Fig. 6a, firstly, the PAN@ZIF-8 fiber precursor was prepared by electrospun, and then the PAN@ZIF-8 nanofiber membranes were calcined to acquire N-doped PCNFs. Finally, MoS₂ nanosheets were evenly assembled on the surface of PCNFs under hydrothermal conditions to obtain PCNF@MoS₂, whose microstructure is shown in Fig. 6b,

c. The resulted PCNF@MoS₂ hybrids were employed as anode materials for LIBs with large capacity, excellent cycle (Fig. 6d) and rate (Fig. 6e) properties. Additionally, Chen et al. demonstrated a valid multi-step route to prepare a hierarchical tubular structure composed of carbon nanotubes (CNTs) and Co₃O₄ hollow nanoparticles

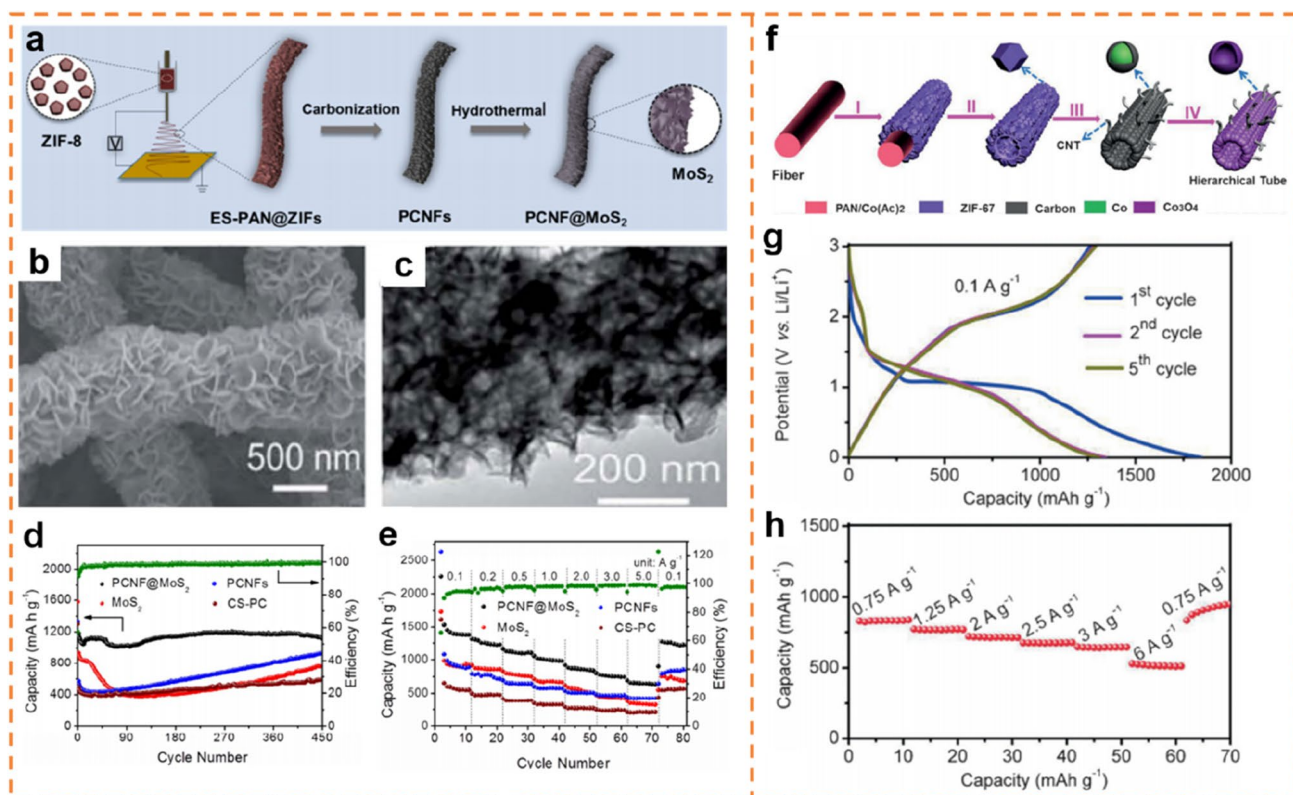


Fig. 6 **a** Schematic diagram for the fabrication process of the PCNF@MoS₂ fibers; **b** SEM and **c** TEM pictures of PCNF@MoS₂; **d** cycle property at 1.0 A·g⁻¹; **e** rate property at different current densities. Reproduced with permission from Ref [115]. Copyright 2019, Elsevier. **f** Preparation of the CNT/Co₃O₄ microtubes; Electrochemi-

cal properties of the CNT/Co₃O₄ microtubes; **g** Charge–discharge voltage profiles at 0.1 A·g⁻¹; **h** rate performance at different current densities. Reproduced with permission from Ref [163]. Copyright 2016, Wiley–VCH

for lithium-ion batteries [163]. They prepared Co(AC)₂/PAN fiber membranes by electrospun, and then soaked the fiber membranes into mIM solution to incorporate ZIF-67 crystals in the Co(AC)₂/PAN electrospun membranes by surface in situ growth. The resulting ZIF-67/PAN fibers could be converted into CNT/Co-carbon composites by a heat treatment in Ar/H₂. As illustrated in Fig. 6f, the CNT/Co₃O₄ microtubes were obtained by further calcinating the composites. The structure of CNTs and Co₃O₄ hollow nanoparticles not only shortened the diffusion distance of lithium ions, but also provided enough contact for fast charge transfer between electrolytes and active materials. The CNTs could accelerate the rapid electron transfer and avert the accumulation of Co₃O₄ particles during cycling. Moreover, the derivative of ZIF-67 could relieve the volume expansion during electrochemical reactions. It is worth mentioning that the resulting CNT/Co₃O₄ composites delivered a high capacity of 1281 mAh·g⁻¹ at 0.1 A·g⁻¹ (Fig. 6g) with phenomenal rate (Fig. 6h) and excellent cycle performance as anode materials for LIBs.

Lithium-Sulfur Batteries

Lithium-sulfur batteries possess seductive theoretical specific capacity [164–166]. However, the theoretical capacity is difficult to achieve due to the poor conductivity of sulfur elements [167]. Simultaneously, the polysulfides produced in the electrochemical reactions are prone to dissolve and move between negative and positive electrodes, resulting in the loss of active substances and the reduction of the performance of LSBs [168–170]. To improve the conductivity of electrode materials and suppress the shuttle of polysulfides, it is considered that the effective way is to develop electrode materials with high porosity, large specific surface area and homogeneous heteroatom doping as sulfur carriers [171]. The emerging MOF-derived porous carbon materials are regarded as potential candidates for LSBs [172–174].

Compared to other carbon nanofiber (CNF) materials, the developed ZIF-67-based CNFs as sulfur hosts can remarkably improve the rate and the cycle performance of LSBs. Thanks to its exceptional structural stability and capacity

reversibility, sulfurized polyacrylonitrile (SPAN) possesses high value in the application of LSBs. Inspired by this, condensed and ultrathin SPAN membranes were designed and fabricated [175]. As visualized in Fig. 7a, ZIF-67 crystals were grown in situ on electrospinning fiber membranes composed of PAN and CNT. Then, surface-anchored CoS_2 nanoparticles were produced by a vulcanization process to acquire CoS_2 -PAN-CNT composites as a cathode for LSBs. Impressively, the layer of CoS_2 nanoparticles on the surface of the fiber membranes efficiently suppressed membrane thickening and fiber expansion. Meanwhile, CoS_2 nanoparticles with high conductivity were conducive to accelerating the redox kinetics of sulfur conversion, so as to realize high-performance LSBs with greatly enhanced areal capacity. This research refreshes the current properties of SPAN batteries with the high capacitance (Fig. 7b), excellent cycle stability (Fig. 7c, d) and outstanding rate performance (Fig. 7e), revealing stupendous prospects for achieving flexible LSBs with large energy density. Additionally, the $\text{C}/\text{Co}_9\text{S}_8$ hybrid carbon nanofibers as sulfur carriers were obtained by electrospinning and self-templating combined with solvothermal and calcination, as depicted in Fig. 7f [176]. The carbon case of the $\text{C}/\text{Co}_9\text{S}_8$ nanofibers derived from mIM in ZIF-67 could enhance the conductivity

of Co_9S_8 , which assisted the transport of ion/electron and inhibited the departure of the sulfur to some extent. The hollow polyhedron structure of the $\text{C}/\text{Co}_9\text{S}_8$ hybrid fibers provided broad sulfur storage space and strong bonding for polysulfide. As a cathode for LSBs, the $\text{C}/\text{Co}_9\text{S}_8$ -C@S hybrid fibers displayed an initial specific capacity of $1013.7 \text{ mAh}\cdot\text{g}^{-1}$ at 0.1 C, exhibiting a capacity of $694.9 \text{ mAh}\cdot\text{g}^{-1}$ after 150 cycles (Fig. 7g). Excitedly, the specific capacity reached $894.7 \text{ mAh}\cdot\text{g}^{-1}$ at 1 C, and the capacity attenuation of each cycle was about 0.116% over 500 cycles (Fig. 7h).

Sodium-Ion Batteries

Sodium in sodium-ion batteries (SIBs) is abundant and inexpensive on Earth [177–179]. Unfortunately, the electrode materials used in LIBs cannot meet the ideal reversible capacity and cycle performance of SIBs because the radius of sodium ions (1.02 \AA) is larger than that of lithium ions (0.76 \AA) [180–183]. Therefore, the structural stability and reaction kinetics of SIBs are affected [184, 185]. Fortunately, the one-dimensional carbon electrospun nanofibers derived from MOF with a high specific surface area have good conductivity and structural stability, which could not only produce some spacious channels for the transmission of

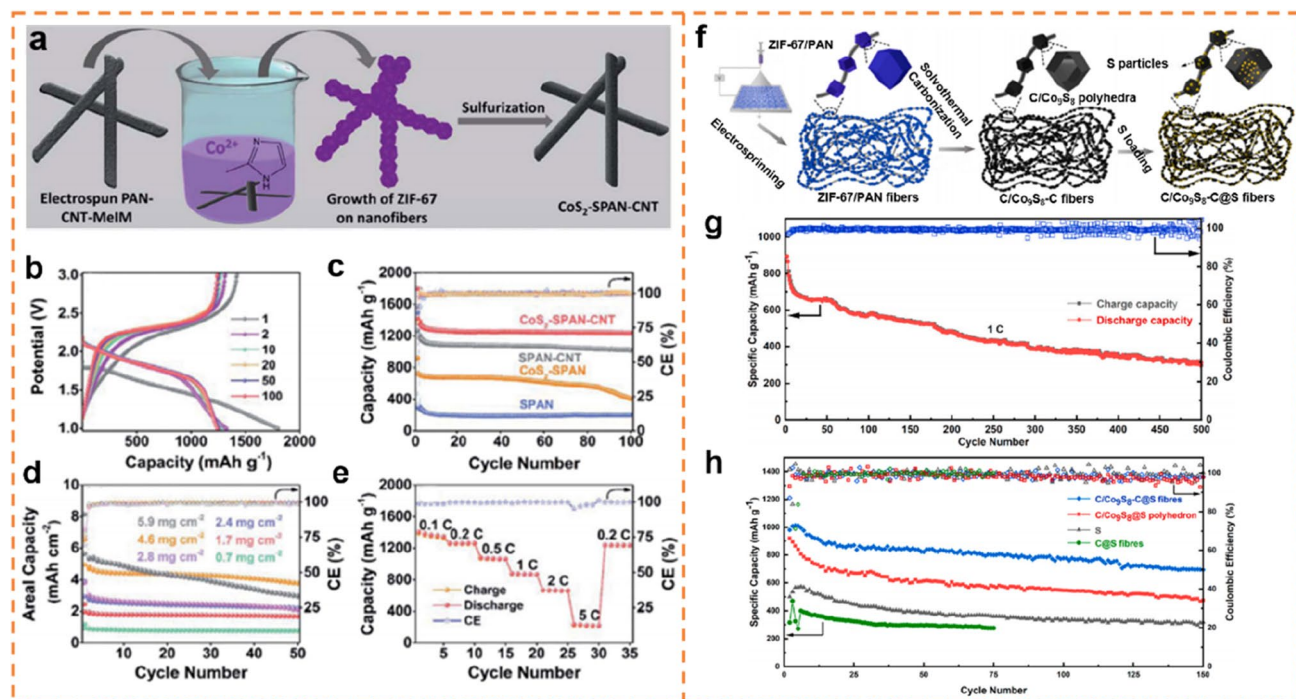


Fig. 7 a Schematic diagram of the fabrication of the CoS_2 -SPAN-CNT electrode; b charge/discharge profiles at a sulfur loading of $2.4 \text{ mg}\cdot\text{cm}^{-2}$ at 0.2 C; c cycle properties of the CoS_2 -SPAN-CNT and control samples at 0.2 C; d cycle properties of CoS_2 -SPAN-CNT with areal sulfur loadings at 0.2 C; e rate capability from 0.1 to 5 C. Reproduced with permission from Ref [175]. Copyright 2020, the

Royal Society of Chemistry. f Schematic diagram of the fabrication route for $\text{C}/\text{Co}_9\text{S}_8$ -C@S nanofiber; g cycle properties and coulombic efficiency of the $\text{C}/\text{Co}_9\text{S}_8$ -C@S nanofibers at 1 C; h cycle properties and coulombic efficiency of $\text{C}/\text{Co}_9\text{S}_8$ -C@S nanofibers, $\text{C}/\text{Co}_9\text{S}_8$ @S polyhedron, C@S nanofibers and the sulfur cathode at 0.1 C. Reproduced with permission from Ref [176]. Copyright 2021, Elsevier

ion and electron in SIBs, but also relieve the volume expansion during the cycle of SIBs [186–190]. Furthermore, entangling one-dimensional carbon nanofibers into a three-dimensional conductive matrix implanted with heterogeneous nanoparticles can satisfy faster diffusion kinetics and shorten the diffusion distance of ions [191, 192].

Combining simple electrospinning and subsequent annealing treatments, a one-dimensional N-doped PCNF anchoring amorphous and graphite carbon-encapsulated CoS₂ particles (AGC-CoS₂@NCNFs) was successfully fabricated as an anode for SIBs. It exhibited superior electrochemical performance [193]. The fabrication process and microstructure of the composite are displayed in Fig. 8a–c. Remarkably, the rationally designed composite possessed improved Na-storage performance. It is worth mentioning that the electrode material shows outstanding rate properties at different current densities (Fig. 8d), a large specific capacity of 876 mAh·g⁻¹ at 100 mA·g⁻¹ and an exceptional cycle performance of 148 mAh·g⁻¹ at 3.2 A·g⁻¹ over 1000 cycles (Fig. 8e). The research indicated

that the enhancement of electrochemical performance was attributed to the one-dimensional N-doped PCNF structure and the graphitic carbon covering, which could cleverly restrain aggregation of CoS₂ particles and alleviate the volume changes in electrochemical reactions to some extent. Additionally, a three-dimensional Fe₇S₈ particles/N-doped carbon nanofibers (Fe₇S₈/N-CNFs) composite was designed and prepared via facile electrospinning and subsequent calcination treatment by Wang et al. [194]. The preparation process of Fe₇S₈/N-CNFs is illustrated in Fig. 8f. As depicted in Fig. 8g and h, Fe₇S₈ particles were encapsulated in the three-dimensional interconnected Fe₇S₈/N-CNFs with a unique structure. When the Fe₇S₈/N-CNFs composite was employed as a negative electrode for SIBs, a high capacity of 649.9 mAh·g⁻¹ was retained after 100 cycles at 0.2 A·g⁻¹ (Fig. 8i) and excellent cycle stability was realized (Fig. 8j). The acquired composite exhibited the following advantages: (I) Fe₇S₈ particles originated from MIL-88A(Fe) could reduce the diffusion distance of sodium ions; (II) the unique three-dimensional structure enhanced the conductivity and

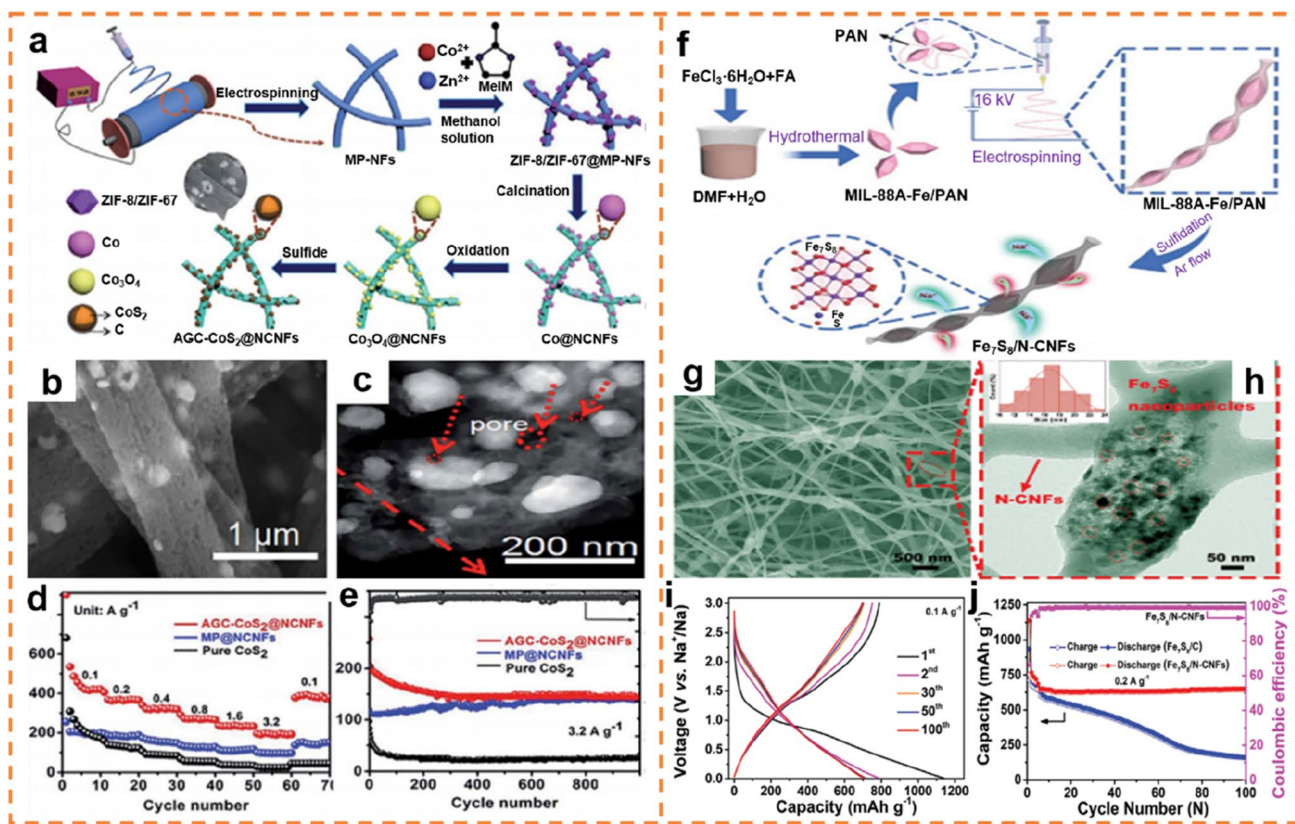


Fig. 8 a Schematic illustration of the fabrication of AGC-CoS₂@NCNFs; b SEM and c TEM images of AGC-CoS₂@NCNFs; d rate performance of AGC-CoS₂@NCNFs; e long-term cycle performance at a 3.2 A·g⁻¹ current density. Reproduced with permission from Ref [193]. Copyright 2020, Elsevier. f Schematic diagram of the synthetic process of Fe₇S₈/N-CNFs; g FESEM picture of Fe₇S₈/N-CNFs;

h HRTEM picture and nanoparticle size distribution (the inset) of Fe₇S₈/N-CNFs; i charge/discharge curves of the Fe₇S₈/N-CNFs electrode for the first, second, 30th, 50th and 100th cycles at 0.1 A·g⁻¹; j cycle performance and coulombic efficiency of the Fe₇S₈/N-CNFs electrode at 0.2 A·g⁻¹. Reproduced with permission from ref [194]. Copyright 2021, Wiley–VCH

alleviated the volume changes and self-aggregation of Fe₇S₈ nanoparticles; and (III) abundant active sites for sodium ions adsorption and paths for electrolyte diffusion were provided. These researches shed light on the development of anode materials for SIBs with superior electrochemical performance.

Water Treatment

Presently, shortage of drinking water and wastewater pollution have given rise to strong demand for water purification via cost-efficient and energy-savings ways [195]. Unfortunately, conventional single filtration and adsorption technologies are unable to simultaneously remove pollutants of different sizes from wastewater, especially heavy metals, dyes and bacteria [67, 196, 197]. Consequently, the development of energy-savings, cost-effective and advanced wastewater purification materials has been widely regarded as the most critical research segment in this field. Emerging MOF materials can adsorb pollutants from wastewater owing to their large surface area, high porosity and easy modification. However, they are powder materials that are easy to agglomerate, resulting in a decrease in adsorption capacity. It is difficult to efficiently recover these powder materials from wastewater, which is easy to cause secondary pollution [198, 199]. Recently, numerous researches on MOF-based nanofibrous membranes have been implemented for the applications of wastewater treatment, indicating that MOF-based nanofibrous membranes possess an extraordinary potential in removing pollutants from wastewater [200, 201]. This section systematically summarizes the application of MOF-based nanofiber membranes for the removal of heavy metals and dyes from wastewater.

Heavy Metal Ions

The residual heavy metals in water pose a severe threat to the life and health of human beings [202–204]. It is imperative to effectively remove them from wastewater. In an excellent research, modified-MOF (MOF-808-EDTA) nanoparticles loaded PAN membranes were designed and fabricated through solvothermal treatment and electrospun to simultaneously adsorb heavy metal ions and separate oil–water [205]. The preparation process of MOF-808@PAN electrospun fiber membranes and the mechanisms of heavy metal ions adsorption and oil-in-water emulsions separation are revealed in Fig. 9a. Interestingly, the composite membranes exhibited superoleophobicity in water and could effectively separate oil-in-water emulsions (Fig. 9b). Meanwhile, the powerful adsorption capacity of MOF-808-EDTA allowed the nanofiber membranes to capture heavy metal ions (Pd²⁺, Cu²⁺, Cr²⁺ and Cr³⁺) at different concentrations within a short period (Fig. 9c), indicating outstanding adsorption

capacity for removing heavy metal ions from wastewater. It is worth mentioning that the membranes also exhibited outstanding corrosion resistance and recyclability. The ZIF-67-based “pearl-necklace-like” hybrid fiber membranes were synthesized by in situ growth on the surface of mIM/cellulose acetate (mIM/CA) electrospinning nanofibers, as illustrated in Fig. 9d [206]. When ZIF-67 crystals were approximately saturated, the surface area of the integrated composite nanofiber membranes was much higher (463.1 m²·g⁻¹), which was dozens of times that of the primitive mIM/CA electrospinning fibers (6.9 m²·g⁻¹). Attractively, the composite nanofiber membranes exhibited a remarkable adsorption capacity of Cr⁶⁺ (14.5 mg·g⁻¹) and Cu²⁺ (18.9 mg·g⁻¹), as displayed in Fig. 9e and f. Moreover, the adsorption mechanisms of the composite membranes were analyzed. It was concluded that the ions exchange and electrostatic adsorption made a contribution to the excellent adsorption capacity for Cr⁶⁺ and Cu²⁺. Additionally, the appearance of Cr³⁺ ions indicated that the Cr⁶⁺ ions partially converted to Cr³⁺ during the adsorption. This work demonstrates that the composite membranes were potential environmental materials for the removal of heavy metals from wastewater.

Dyes

The dyes released into the water are toxic and even carcinogenic. They are difficult to degrade spontaneously, which cause great harm to human health and the green environment [207, 208]. To solve the problem of dyes pollution, a variety of strategies have been performed to capture organic dyes from wastewater [209–211]. The bio-MOF/PAN nanofibrous filter was cleverly prepared by co-electrospun of bio-MOF-1 (Zn₈(Ad)₄(BPDC)₆O·2(NH₂(CH₃)₂)⁺, 8DMF, 11H₂O) with an anionic skeleton and PAN for selective separation and capture of cationic dyes from an aqueous solution, as illustrated in Fig. 9g [212]. Due to the existence of the free NH₂(CH₃)₂⁺ cations from the channels of bio-MOF-1 and the nucleophilic groups from PAN, bio-MOFs-1 and PAN possessed outstanding adsorption capacity for cationic dyes. Owing to the synergistic interaction of PAN and bio-MOF-1, the bio-MOF/PAN filter displayed a quick adsorption rate for methylene blue (MB⁺) (Fig. 9h) and achieved the rapid capture of MB⁺ from the solution of mixed dyes. It is worth mentioning that the bio-MOF/PAN filter could be efficiently regenerated via ion exchange. Remarkably, it still maintained stable adsorption values for MB⁺ after the five cycles of filtration-regeneration (Fig. 9i), manifesting that the bio-MOF/PAN filter had the ability to retain a long service life. Additionally, Dai et al. proposed an experimental scheme to coat ZIF-8@GO on the surface of polylactic acid (PLA) electrospinning

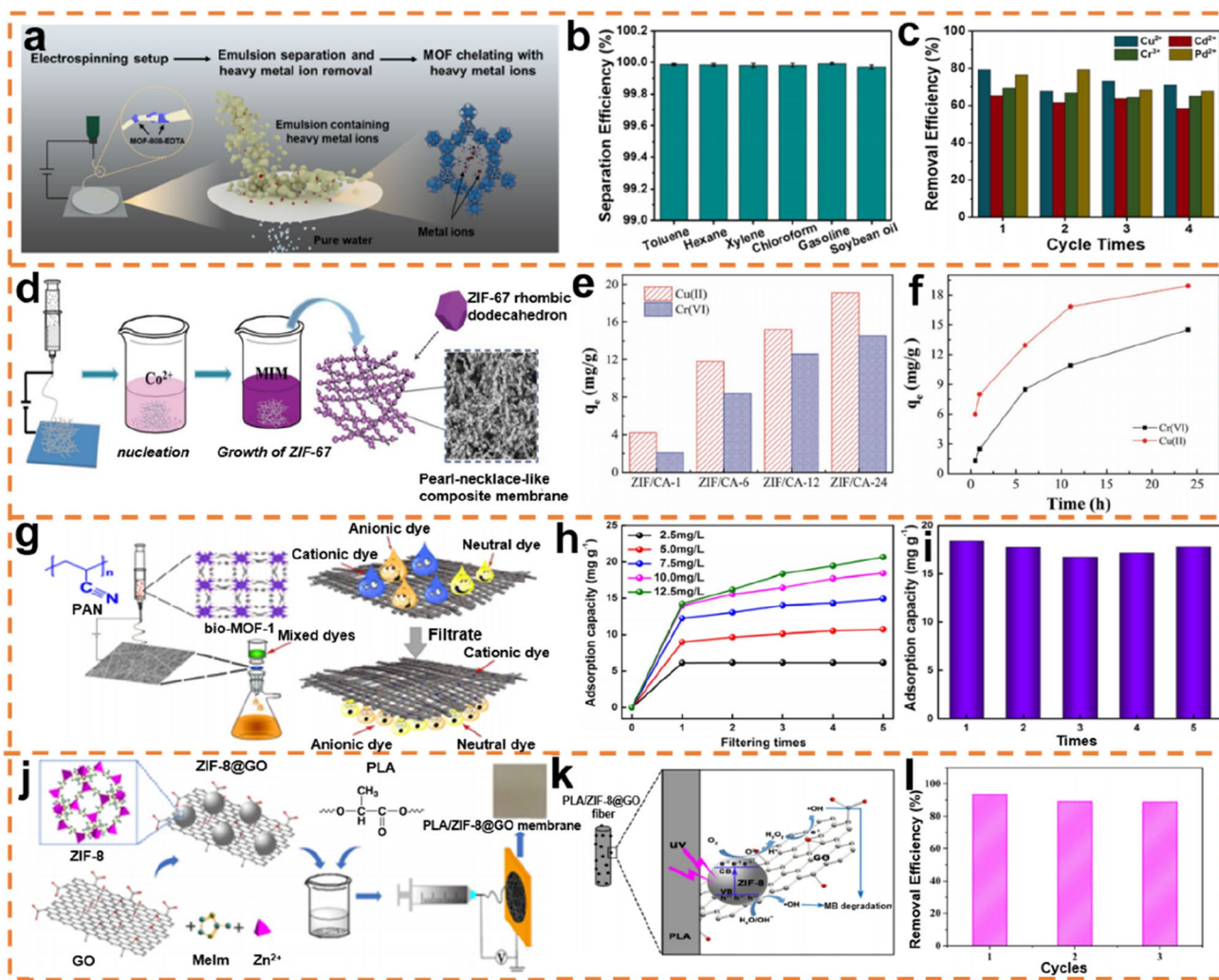


Fig. 9 **a** Preparation of the MOF-808@PAN nanofiber membranes and mechanisms of heavy metal capture and oil-in-water emulsions separation; **b** separation efficiency of different oil-in-water emulsions; **c** cycle test of the adsorption capacity of the MOF-808@PAN nanofiber membranes for different heavy metal. Reproduced with permission from Ref [205]. Copyright 2020, American Chemical Society. **d** Schematic diagram of the fabrication of pearl-necklace-like composite fiber membranes; **e** adsorption capacity of all samples for Cr⁶⁺ and Cu²⁺; **f** effect of contact time on the adsorption of Cr⁶⁺ and Cu²⁺ on the ZIF/CA-24 (experimental condition: at room temperature, pH ≈ 6.5, without stirring). Reproduced with permission from

Ref [206]. Copyright 2018, Wiley–VCH. **g** Preparation of bio-MOF/PAN membrane and its adsorption of cationic dyes; **h** adsorption capacity of bio-MOF/PAN filter for MB⁺ with different original concentrations; **i** adsorption capacity of bio-MOF/PAN filter for MB⁺ in five cycles. Reproduced with permission from Ref [212]. Copyright 2020, Elsevier. **j** Fabrication Process of the PLA/ZIF-8@GO composite nanofiber membranes; **k** mechanisms of photocatalytic degradation of MB on PLA/ZIF-8@GO nanofibers; **l** removal efficiency of MB on PLA/ZIF-8@GO nanofibers with three cycles. Reproduced with permission from Ref [213]. Copyright 2018, American Chemical Society

nanofiber membranes via in situ growth for photocatalytic degradation and adsorption MB [213]. As shown in Fig. 9j, k, the fabrication process of the ZIF-8@GO/PLA fiber membranes and the mechanisms of photocatalytic degradation are revealed, respectively. During the reaction, PLA acted as an adsorbent and catalyst carrier, and ZIF-8 produced holes and photogenerated electrons under illumination, which facilitates a series of reactions. Simultaneously, GO served as a paralysis of electron–hole pairs recombination, which could enhance the performance of

the hybrid fiber membranes. The results demonstrated that the degradable ZIF-8@GO/PLA electrospun nanofiber membranes delivered higher tensile strength than pure PLA. Satisfactorily, the composite nanofiber membranes exhibited excellent MB removal efficiency by photocatalytic degradation and adsorption. In the case of a shallow ZIF-8@GO concentration (0.06 mg·mL⁻¹), the removal efficiency of MB could still reach over 90% after three cycles (Fig. 9l).

Air Pollutant Filtration

With the development of worldwide industrialization, atmospheric pollutants in the form of toxic gases and delicate particulate matter (PM) have become a severe worldwide environmental issue [214, 215]. Past studies have demonstrated that adsorbents such as zeolites, activated carbon, resins, biomass and metal oxides can capture these pollutants from the atmosphere [210, 216, 217]. Unfortunately, the high regeneration cost and limited adsorption capacity of these traditional materials have forced researchers to develop novel materials and technologies to remove air pollutants [218]. Researchers found that the nanofibers prepared by electrospinning possess the advantages of controllable pore size, large specific surface area and high air permeability [94]. Furthermore, the composites fabricated by loading MOF crystals on nanofibers have significantly improved removal ability, so they are considered candidates for rapid and effective capture of pollutants from the air [219].

Toxic Gases

Sulfur dioxide (SO₂) and ozone (O₃), the primary pollutants in the air, pose a serious threat to the biological system and human health. Fortunately, MOF-based electrospun nanofibers can effectively capture SO₂ and O₃ from the air. For example, MIL-53(Al)-NH₂/PAN hybrid fiber membranes prepared by in situ growth of MIL-53(Al)-NH₂ crystals onto the surface of PAN polymer nanofibers via electrospinning

technology and subsequent hydrothermal treatment exhibited outstanding adsorption capacity for air pollutants, especially PM, sulfur trioxide (SO₃) and O₃ [220]. The preparation process is shown in Fig. 10a. Remarkably, the PM removal efficiency of the obtained composite nanofiber membranes could reach 99.99%. More interestingly, the MIL-53(Al)-NH₂/PAN composite fiber membranes could selectively and efficiently remove SO₂ and O₃ pollutants without being affected by carbon dioxide (CO₂), oxygen (O₂) and other gases. The composite nanofiber membranes could not only reduce the SO₂ concentration from 7300 to 40 ppb (Fig. 10b), but also rapidly drop the O₃ concentration from 3000 to 7 ppb (Fig. 10c), which is far lower than the national air quality standard (81 ppb). This work indicated that the great potential of the composite membranes prepared by simple and cheap electrospinning technology as candidate materials for air purification and environmental protection.

Particulate Matter

PM with small size can enter the respiratory system through the respiratory tract, which causes respiratory diseases, such as upper respiratory tract infection, bronchial asthma, bronchitis, pharyngitis, pneumonia. The MOF-based electrospun nanofibers can not only remove SO₂ and O₃ from the air, but also effectively capture PM. As the first work to explore the interaction between MOF-based nanofibrous membranes and PM, Zhang et al. designed four kinds of MOF/polymer nanofibrous membranes (UiO-66-NH₂/PAN, ZIF-8/PAN,

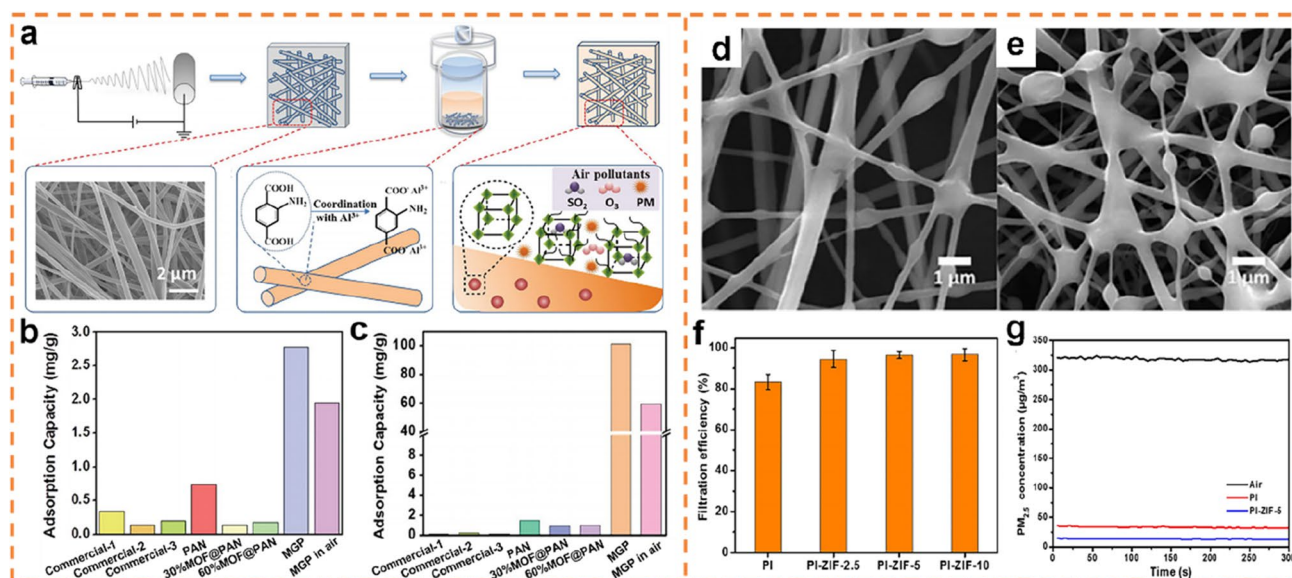


Fig. 10 **a** The fabrication and adsorption processes of MOF-based nanofiber membranes; **b** SO₂ removal capacity of the different fiber membranes; **c** O₃ removal capacity of the different fiber membranes. Reproduced with permission from Ref [220]. Copyright 2019, the Royal Society of Chemistry. SEM images of **d** PI membranes and **e**

ZIF-8/PI membranes; **f** PM_{2.5} removal efficiency of different membranes; **g** the concentration of PM_{2.5} captured by different membranes in harsh conditions. Reproduced with permission from Ref [126]. Copyright 2019, American Chemical Society

MOF-199/PAN and Mg-MOF-74/PAN) with loadings up to 60% [221]. The performance of these four MOF/polymer nanofiber filters was investigated in an actual smog environment ($PM_{10}=720 \mu\text{g}\cdot\text{m}^{-3}$, $PM_{2.5}=350 \mu\text{g}\cdot\text{m}^{-3}$). The results displayed that ZIF-8/PAN exhibited the strongest capacity to remove PM ($89.67 \pm 1.33\%$ for PM_{10} , $88.33 \pm 1.52\%$ for $PM_{2.5}$), and the performance did not decrease obviously after 48 h of continuous exposure. The excellent PM removal capacity of ZIF-8/PAN was attributed to the highest zeta potential value of ZIF-8. Interestingly, these fiber filters could selectively capture SO_2 when exposed to SO_2/N_2 . Commendably, their hierarchical nanostructures allowed the permeation of fresh air at high gas flow rates (pressure drop < 20 Pa). Additionally, a strategy combining electrospinning was designed to prepare ZIF-8/PI composite nanofiber membranes with different loadings of ZIF-8 crystals [126]. SEM characterization demonstrated that the hybrid fiber membranes obtained by introducing ZIF-8 nanoparticles displayed a rougher surface than the pure PI membranes (Fig. 10d, e). As shown in Fig. 10f, compared with the pure PI membranes, the ZIF-8/PI composite membranes exhibited a higher removal efficiency for $PM_{2.5}$, which was attributed to the larger specific surface area of the composite membranes and the excellent properties of ZIF-8, such as exceptional chemical and thermal stability (up to 300 °C). Interestingly, ZIF-8/PI composite nanofiber membranes with ZIF-8 loading of 10% exhibited the highest $PM_{2.5}$ removal rate ($96.6 \pm 2.9\%$). Additionally, the composite membranes could efficiently capture $PM_{2.5}$ in a short time, even under harsh conditions, which is quite remarkable (Fig. 10g).

Gas Storage and Separation

The increase of population and the development of global industrialization have led to the increasing CO_2 emission, which poses a serious threat to human health and the ecological environment [222]. In recent years, MOFs have received increasing attention owing to their large specific surface area, high porosity, tunable pore size and flexible framework structures [223]. Unfortunately, a single MOF material may suffer from blockage of pipelines and difficulty in recycling in practical gas storage and separation work [224]. To address these issues, the researchers constructed MOF materials with porous structures into nanofiber membranes by electrospinning to introduce more efficient adsorption sites. The resulting MOF-based nanofibrous membranes play a major role in scaling up practical applications for gas storage and separation.

It is found that the size, loading amount, morphology and specific surface area of MOF crystals supported on the fibrous membranes greatly affect the properties of MOF/polymer nanofiber membranes. For example, the flexible and self-supporting HKUST-1/PAN nanofiber membranes

with MOF particles loading up to 82% prepared by electrospinning can efficiently and selectively adsorb CO_2 [225]. This is the work with the highest MOF loading rate at present. Surprisingly, the designed HKUST-1/PAN nanofibrous membranes exhibited outstanding CO_2 adsorption capacity ($3.9 \text{ mmol}\cdot\text{g}^{-1}$) and excellent CO_2/N_2 selectivity. It is worth mentioning that the results of 100 adsorption–desorption experiments showed that the CO_2 adsorption capacity hardly changed and remained at 95% ($3.7 \text{ mmol}\cdot\text{g}^{-1}$) of the initial value, revealing the potential of stable recoverability and excellent practical value of HKUST-1/PAN nanofiber membranes. The abundant CO_2 adsorption sites on the nanofibrous membranes are mainly located in the open metal sites. In addition, the designed HKUST-1/PAN nanofibrous membranes possessed ultra-high specific surface area, suitable pore size and porosity, which made a significant contribution to the selective separation of CO_2/N_2 . This work provides a low-cost and scalable production route to convert MOF crystals into flexible and self-supporting nanofibrous membranes, laying the cornerstone for better post-combustion CO_2 capture.

Conclusion and Perspectives

In recent years, MOFs have received increasing attention owing to their large specific surface area, high porosity, tunable pore size and flexible framework structures. However, due to the limitations of MOFs, a single MOF material cannot well satisfy the needs of practical applications. Fortunately, a series of MOF composites and derivatives have been gradually discovered and studied. Among them, MOF/polymer nanofibers fabricated by electrospinning have been intensively reported. The resulting composites not only overcome the limitations of single MOF materials, but also realize co-spinning of multiple materials, significantly improving the diversity of nanofibers and broadening the applications of MOF/polymer nanofibers. Based on this, researchers have conducted a large number of reports on them. This review systematically summarizes the two main routes (direct electrospinning and surface in situ growth) for the preparation of MOF/polymer nanofiber membranes by electrospun combined with other methods, such as hydrothermal and solvothermal reactions. Both routes can successfully prepare nanofibrous membranes with high specific surface area, large porosity and superior air permeability. Next, the applications of MOF/polymer nanofiber membranes prepared by these two routes in lithium-ion batteries, lithium-sulfur batteries, sodium-ion batteries and environmental protection are discussed. Although great progress has been achieved in the applications of MOF-based nanofiber membranes, there are still various inevitable challenges and

problems in future research, as shown in the following six points.

- (1) Although thousands of MOF nanoparticles have been successfully prepared, the MOFs developed as precursors are limited to traditional structures, such as MIL and ZIF series. Consequently, MOF-based nanomaterials generally have similar structures and morphologies, which is not conducive to realizing the structural diversity, but also affects the further improvement of the performance of related materials. Hence, it is necessary to develop a wide variety of MOFs as precursors of MOF-based nanomaterials to exploit the greater potential of materials. Meanwhile, the development of these novel MOFs is beneficial to expanding the raw material sources for MOF/polymer systems, paving the way for obtaining higher-performance MOF/polymer nanofibers.
- (2) Considering the enrichment of nanofiber types and the development of more potential MOF/polymer nanofiber materials, more polymer materials can be investigated to develop diverse MOF/polymer systems. However, these polymers should be guaranteed to withstand harsh MOF synthesis and decoration conditions, such as aggressive solvents and temperatures.
- (3) Post-processing technology has been widely used in the processing of traditional nonwoven fibers in the textile industry. Future research can focus on exploring the synergistic mechanism of MOF crystals and polymer electrospun nanofibers. Then, appropriate post-processing technology is adopted to improve the performance of MOF/polymer nanofibers to expand its practical applications.
- (4) In recent years, the types and structures of MOF/polymer nanofibers have gradually increased. Still, there are few theoretical studies to illustrate the relationship between structure and performance, which is not conducive to achieving controllability of material composition and structure. For instance, porous carbon nanomaterials derived from MOFs obtained by metal ion removal and carbonization treatments may exhibit large structural uncertainties. Because MOF/polymer nanofibers are a relatively new architecture, more theoretical studies on the structural evolution of fibers during the post-processing are required to satisfy the practical needs.
- (5) Although MOF/polymer nanofibers possess obvious advantages in structure and performance, there are also some disadvantages that cannot be ignored. Unfortunately, no commercial products of MOF/polymer fibers have been reported so far. The high cost of semifinished materials and the complicated preparation process may be the reasons that restrain the large-scale

production of the composite in commerce. Some traditional MOFs can be synthesized with cheap metal ions and organic ligands, but low yield usually leads to high cost. Consequently, developing inexpensive ligands with guaranteed high yield is the direction for the further development of MOF/polymer nanofibers. Additionally, improving the related equipment and facilities for the production of MOF/polymer nanofibers is also a significant breakthrough. These improvements are expected to allow MOF-based nanofibers to enter the market of high commercial value for the first time. For example, MOF-based nanofibers can be used to capture contaminants in water.

- (6) MOF/polymer nanofiber membranes have been widely used in energy storage and environmental protection. However, the problems of poor stability, high cost and poor regeneration ability in practical applications limit the further development of fiber membranes. All research results need to be scaled from laboratory to large-scale production and application. Therefore, it is a great challenge and opportunity to design MOF/polymer nanofiber membranes that are both economical and environmentally friendly for commercial applications. In addition, appropriate post-treatment technologies can be applied to the modification of MOF/polymer nanofiber membranes to enhance their stability and feasibility in practical applications.

MOF/polymer nanofiber membranes have been widely used in energy storage and environmental protection. However, the development of MOF/polymer electrospun fiber membranes with fewer defects, higher chemical stability and lower cost brings great challenges and opportunities for future research. In conclusion, MOF-based nanofibers hold the promise of playing a central role in energy storage and environmental protection. Researchers can conduct more in-depth research on MOF-based nanofibers, and ultimately promote the better applications of MOF-based nanofiber membranes in energy storage and environmental protection.

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Declarations

Conflict of Interest The authors declare that they have no conflict of interest.

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