

Review

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Review

# Research Status of Metal Organic Framework in the Field of Membrane Distillation

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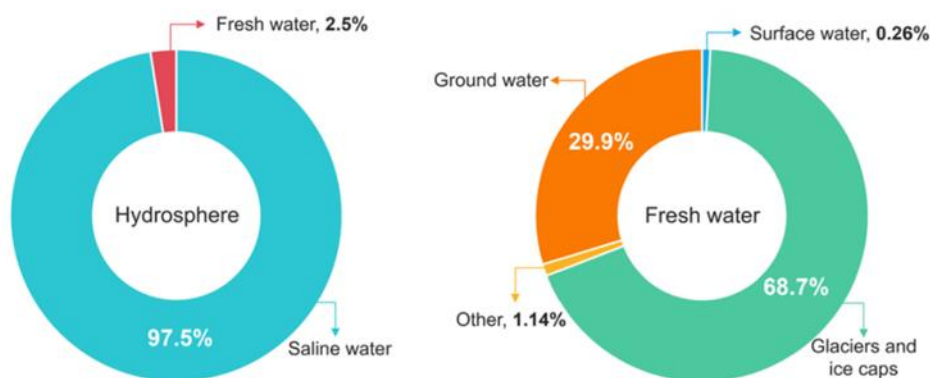
## Abstract

Membrane Distillation (MD) is a heat-driven seawater desalination technology that uses a hydrophobic microporous membrane as its core component. Due to its low energy consumption, high separation efficiency, and ability to handle high-concentration saline wastewater, it has become an effective solution to the shortage of freshwater resources. Nevertheless, issues such as membrane wetting, membrane fouling, and low membrane flux severely limit its large-scale application. Composite membranes prepared using metal-organic framework (MOF) materials as fillers have become a research hotspot due to their advantages, such as permeable microporous channels, customizable pore structures, and modifiable active sites. These properties enable them to effectively reduce temperature polarization and concentration polarization phenomena. This article describes the characteristics of metal-organic framework materials and their current applications in the field of membrane distillation. Comparative analysis of the applicability of MOF polycrystalline membranes and MOF composite membranes in membrane distillation. Discussed the working principle of MOFs in enhancing the performance of membrane distillation. Finally, the problems and challenges associated with the use of MOFs in membrane distillation applications were analyzed. Aims to provide theoretical guidance for the application of metal-organic framework materials in the field of membrane distillation seawater desalination.

**Keywords:** membrane distillation; metal-organic framework membranes; membrane separation; desalination; current research status

## 1. Introduction

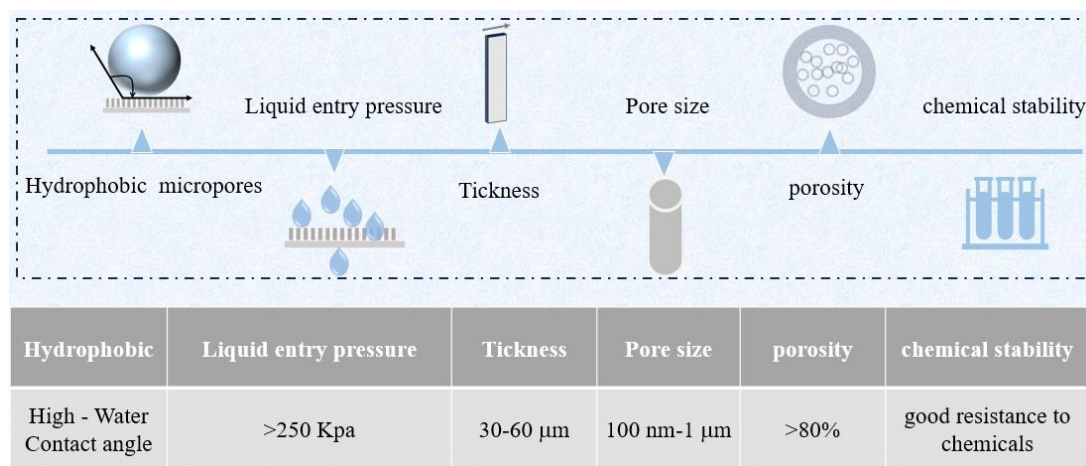
Water resources, as a fundamental resource for life, are facing an unprecedented imbalance between supply and demand. As shown in Figure 1, the fresh water available for human use on Earth accounts for only 0.26% of the total water volume, and its distribution is extremely uneven[1]. According to a 2018 United Nations report, approximately 4 billion people worldwide lack access to clean and safe water, and this situation is likely to worsen in the coming years[2]. In this context, wastewater reuse[3] and desalination technology[4] serve as stable, economical, and sustainable solutions that provide an important means to alleviate the global water crisis. Desalination refers to the process of removing dissolved minerals and salts from seawater to produce drinking water[5]. Since desalination doesn't depend on river flow, reservoir levels, or climate change, and since seawater accounts for 97% of the world's total water resources, this technology has become a key approach in the global water market to meet freshwater demands[6]. Although reverse osmosis (RO) desalination technology has been industrialized, it still faces practical challenges such as high energy consumption and the generation of large amounts of highly saline wastewater. Therefore, there is an urgent need to find an economical and environmentally friendly alternative to address the global water shortage problem.



**Figure 1.** Distribution of water in the Earth's hydrosphere[1].

Traditional seawater desalination methods are mainly divided into membrane-based and thermal processes[7]. The thermal distillation process includes three types: multi-stage flash distillation, multi-effect distillation, and steam compression evaporation. However, high energy consumption and costs require continuous attention to issues related to brine disposal and energy use[7]. Compared to thermal distillation processes, membrane processes have gained widespread attention due to their advantages such as lower energy consumption, more compact equipment, and higher modularity[8]. The most common membrane technologies include Forward Osmosis (FO), Reverse Osmosis (RO), and Membrane Distillation (MD)[9]. Compared to reverse osmosis technology, membrane distillation (MD) offers a higher salt rejection rate, lower operating pressure, and the ability to process high-salinity brines.

Membrane distillation is a new separation technique that utilizes the vapor pressure difference across the membrane to drive the directional migration of water molecules. This technology cleverly combines membrane separation with evaporation processes and is currently in the research and development phase for industrial application. It offers a new solution to alleviate the global shortage of freshwater resources. The core of membrane distillation is the creation of special membrane materials with micron-sized pores and a hydrophobic surface, effectively prevents liquid penetration while allowing water vapor to pass through. However, the MD membrane exhibits progressive wetting and low permeation flux, which significantly limits its practical applications[10]. Membrane wetting can lead to a decrease or even loss of the membrane's ability to reject salts, while low flux results in a deterioration of fresh water production. To achieve good wetting resistance, the MD membrane must have high hydrophobicity, small pore sizes, and no macroscopic pores in its structure to prevent water from penetrating. At the same time, to increase water flux, MD membranes need to have a high porosity, low tortuosity, and an extremely thin thickness. Figure 2 lists the characteristic parameters of the MD membrane. It is necessary to possess key properties such as low surface energy, high hydrophobicity[11], high liquid entry pressure[12], high porosity, optimal membrane thickness[13], and appropriate pore size. These properties are crucial for enhancing steam penetration and salt rejection.



**Figure 2.** Summary of Required Characteristics for MD Membrane.

Research shows that membrane materials affect membrane structures, which in turn influence membrane properties. Therefore, selecting the appropriate material for preparing MD membranes is crucial. Polymers are currently the preferred membrane material due to their low raw material and production costs, ease of processing, and suitability for mass production[14–16]. Inorganic membranes have attracted widespread attention as promising membranes for gas and liquid separation, thanks to their excellent thermal and chemical stability, good operational stability, and impressive separation performance[17]. In recent years, due to the inherent “trade-off” characteristics of polymer membranes, the preparation and development of hybrid matrix membranes by dispersing nanoparticles as fillers within a polymer matrix have become popular among researchers. The reasons why mixed matrix membranes have superior separation performance compared to pure polymers are mainly: Firstly, the addition of inorganic nanoparticles can reduce the entanglement of polymer molecular chains, decrease molecular chain aggregation, increase the free volume of the membrane, and thereby enhance the molecular permeation rate. Secondly, the pores in the inorganic particles themselves reduce mass transfer resistance while providing additional pathways for molecular transport. In most nanofillers, MOFs, as an organic-inorganic hybrid material, are considered one of the candidate materials for modifying traditional polymer membranes due to their high porosity, low thermal conductivity, and good compatibility with polymeric materials. Table 1 summarizes the different types of membranes used in membrane distillation.

**Table 1.** Different types of membranes used in membrane distillation.

Membrane Type	Common Materials	Advantages	Insufficient	Ref
Inorganic membrane	Ceramics ( $\text{Al}_2\text{O}_3$ , $\text{ZrO}_2$ , $\text{TiO}_2$ ), metals (stainless steel, titanium alloy), glass	High-temperature resistant, acid and alkali corrosion-resistant, high mechanical strength, good chemical stability, uniform pore size distribution, strong resistance to contamination, and long service life.	High preparation costs, brittle membrane that breaks easily, difficult processing, high costs for large-scale application, and complex membrane module design.	[18,19]
Polymer membrane	Polyvinylidene fluoride (PVDF), Polytetrafluoroethylene (PTFE), Polypropylene (PP), Polysulfone (PSF)	Simple preparation process and low cost; good flexibility and easy to shape; suitable for mass production; flexible membrane module design.	Poor high-temperature resistance (usual operating temperature $<100^\circ\text{C}$ over long periods), prone to swelling in organic solvents, weak resistance to	[20–22]

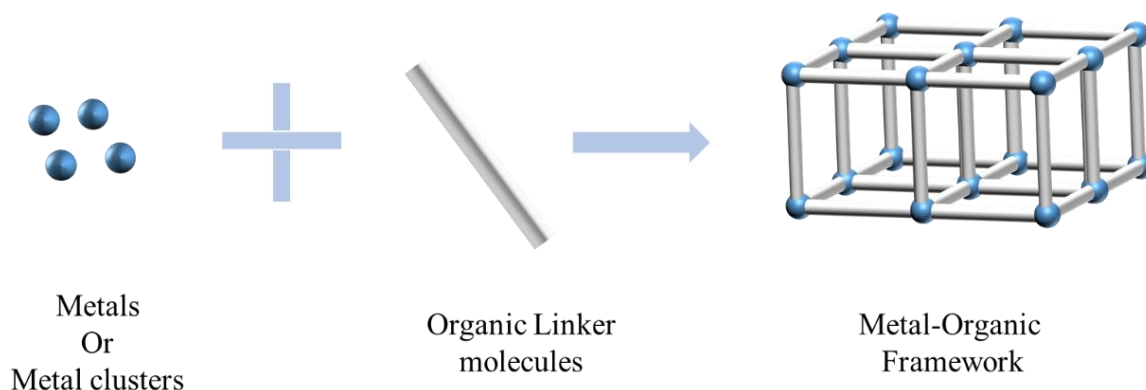
Mixed Matrix Membrane	Polymer substrates (PVDF, PTFE, etc.) + Inorganic fillers (graphene, MOFs, ceramic particles, carbon nanotubes, etc.)	Combines the flexibility and processability of polymer membranes with the advantages of inorganic fillers, such as high-temperature resistance, pollution resistance, and high porosity. Results in better flux and retention rates compared to single polymer membranes.	contamination, and rapid aging and flux degradation with prolonged use. Inorganic fillers have poor compatibility with polymer substrates, tend to agglomerate, require complex preparation processes, are difficult to disperse within the polymer, and result in higher costs compared to pure polymer films. [20,23,24]
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## 2. Metal-Organic Framework Materials (MOFs)

Metal-organic frameworks (MOFs) are porous crystalline materials formed by organic ligands connecting inorganic building units, as shown in Figure 3. MOF materials were first discovered by Robson in 1990, through the bonding of discrete metal clusters. They were officially named MOFs in 1995. Yaghi first reported the synthesis of MOF-5, a material whose crystalline porous framework is constructed with  $Zn^{2+}$  as metal nodes and terephthalic acid as ligands. Modular design of porous materials was achieved (BET specific surface area of  $2900 \text{ m}^2/\text{g}$ ), laying the structural foundation for the development of MOFs[25–27].

MOFs exhibit three significant advantages in the field of membrane distillation for seawater desalination. Firstly, the excellent porosity of MOFs (94%) gives them a unique advantage in the field of separation. Secondly, the structural units of MOFs combine with various metal ions/clusters through coordination bonds in different coordination modes, thereby endowing them with high structural diversity. Thirdly, the organic ligands in MOFs typically possess long-chain structures and  $\sigma$ -bond characteristics, and the vast majority of MOF materials exhibit significant flexibility[28]. In summary, MOFs' advantages such as their extremely high specific surface area, adjustable pore structure, controllable chemical properties, and excellent structural stability have made them a revolutionary class of materials in the fields of materials science and chemistry. They hold great potential for applications across various disciplines[29–32]. Research shows that the addition of MOFs can significantly improve material properties, enhancing the membrane's permeability and selectivity[33]. Unfortunately, the powder-like nature of MOFs limits their application in the field of water treatment. First, MOF powders not only tend to aggregate but can also cause blockages that lead to secondary pollution. Second, powdered MOFs are prone to loss during practical applications, which not only shortens their lifespan but also increases operating costs. Third, immature production methods, unstable production cycles, and poor biocompatibility limit the widespread application of MOFs. Therefore, combining MOFs with regular pore structures with polymers that possess processability to create mixed matrix membranes(MMMs) is highly favored by researchers.

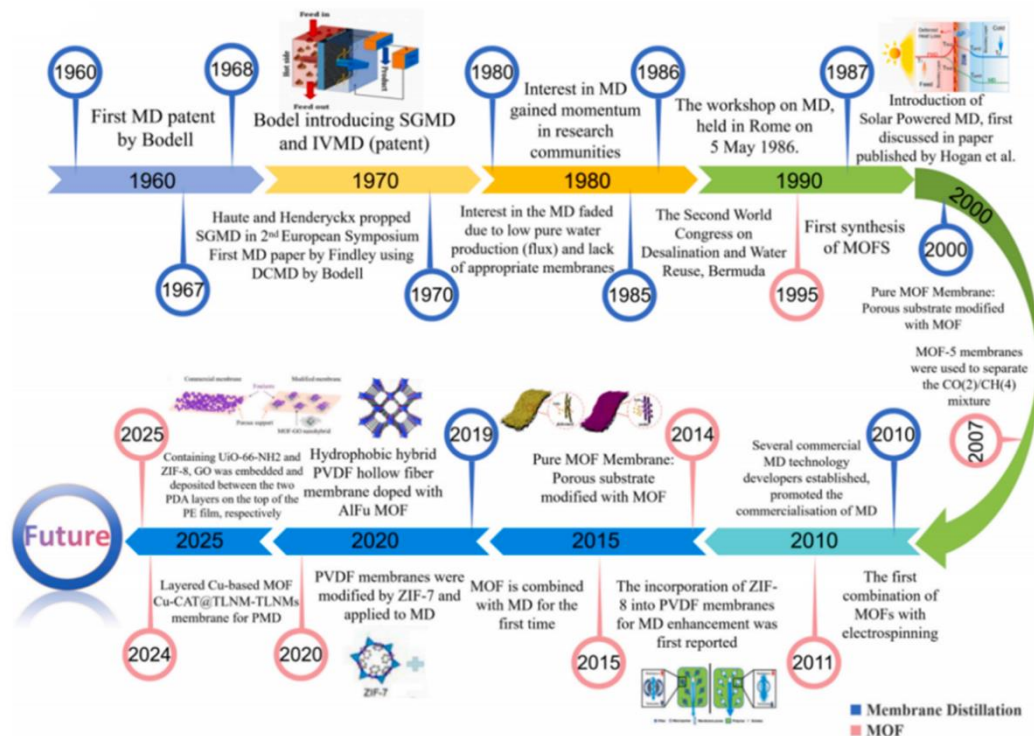
As fillers for mixed matrix membranes (MMMs) and thin-film nanocomposite membranes, MOFs can not only effectively remove oil stains and trace pollutants, but also eliminate heavy metal ions[34] and purify organic dye contamination[35]. They also demonstrate excellent performance in the treatment of brackish water and seawater desalination[36]. Thus, they have become key materials for advancing membrane distillation technology towards greater energy efficiency, higher performance, and larger-scale application. Section 3 will systematically review the current research status of MOF-modified membrane distillation membranes.



**Figure 3.** Schematic diagram of metal-organic framework (MOF) structure.

### 3. Research Status of MOFs-Modified Membrane Distillation Membranes

Membrane distillation technology was introduced in the 1960s[37]. In 2007, MOF-based membranes were used for gas separation[38]. That same year, membrane distillation technology became commercially viable. Subsequently, various membranes suitable for membrane distillation were developed[39]. In 2015, MOFs and membrane distillation technology were combined for the first time[40]. Figure 4 summarizes the application potential and development history of MOFs in membrane distillation technology. In general, MOF-based membrane design strategies can be divided into two main categories. This section will focus on: (1) MOF polycrystalline membranes; (2) MOF-based composite membranes.



**Figure 4.** Illustrated History of Membrane Distillation and MOF Development[33].

#### 3.1. MOF Polycrystalline Film

MOF polycrystalline film is a membrane structure in which MOF crystals that grow well together serve as the selective layer[17]. The permeation selectivity and overall properties of the

membrane are determined by the nature of the building units, pore size, and porosity of the MOFs. Therefore, it exhibits excellent performance in the field of molecular separation[41]. However, preparing dense and defect-free MOF polycrystalline films on porous substrates still presents many challenges. It typically exhibits low mechanical strength, which limits its long-term stability and commercial applicability. Over the past decade, researchers have made significant efforts in developing methods for preparing MOF polycrystalline films, thereby advancing research in this field. Currently, MOF polycrystalline films are mainly prepared by in-situ growth, secondary growth, and layer-by-layer growth methods.

### 3.1.1. In-Situ Growth Method

In-situ growth is an effective method for preparing MOF membranes. The core concept is to mix MOFs with the supporting substrate in a single precursor solution. In MOF crystals, nucleation, growth, and co-growth on the substrate surface occur simultaneously. The key advantage of this preparation method is that MOFs can be grown in situ on the surface of the substrate. Liu et al. [42] were the first to report on the preparation of MOF-5 membranes on porous alumina substrates, laying the foundation for subsequent research in this area. Chuang et al.[40] co-grew MOF crystals on alumina tube substrates and modified the surface of the MOF membrane by introducing perfluorinated molecules. Research has found that the modified membrane material not only retains the high stability and high water permeability of the alumina substrate, but its surface modification also endows it with excellent hydrophobic properties. This has opened up entirely new research directions for designing next-generation high-performance membrane distillation membranes for seawater desalination.

In terms of material surface modification, Huang and his research team[43] used bio-inspired polydopamine to modify the substrate surface, successfully synthesizing ZIF-8 membrane with varying degrees of crystallinity. Polydopamine modification significantly enhanced the interfacial binding between the substrate and ZIF, thereby promoting the nucleation and in-situ growth of ZIF-8. The prepared ZIF membrane performed excellently in seawater desalination applications, with an ion rejection rate of up to 99.8%. In addition, Huang et al.[38] successfully prepared dense and pure-phase UiO-66-NH<sub>2</sub> membranes on aminopropyl-propoxyethoxysilane (APTES)-modified macroporous Al<sub>2</sub>O<sub>3</sub> tubes. As the feed temperature increased, the water flux rose from 1.5 kg·m<sup>-2</sup>·h<sup>-1</sup> to 12.1 kg·m<sup>-2</sup>·h<sup>-1</sup>, while the retention rate remained at 99.7%. Rahman et al.[44] synthesized pure UiO-66 crystals in situ on alumina hollow fiber membranes. After modification via fluorosilane grafting, the modified membranes achieved a water flux of 14.95 L·m<sup>-2</sup>·h<sup>-1</sup> and a NaCl rejection rate of 99.9% during direct contact membrane distillation. Li et al.[45] prepared a pure-phase UiO-66 polycrystalline membrane on alumina hollow fibers using in-situ solvothermal synthesis. This membrane exhibited excellent polyvalent ion retention properties and good permeability. Liu et al.[46] prepared a continuous aluminum MOF-303 membrane on an  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> substrate using in-situ hydrothermal synthesis. This film exhibits excellent retention efficiency for divalent ions while maintaining high permeability.

The high porosity of the MOF polycrystalline membrane prepared by in-situ growth provides additional pathways for the transport of water vapor molecules. However, due to the slow membrane growth process and stringent synthesis conditions, this approach remains challenging for large-scale production of large-area MOF polycrystalline membrane. At the same time, MOF polycrystalline membrane are prone to crack defects and insufficient mechanical strength when subjected to stress, making them unable to meet the requirements for long-term operational stability.

### 3.1.2. Secondary Growth Method

The secondary growth method consists of two steps. In the first step, a layer of MOF nanocrystals is uniformly and densely deposited on the surface of the porous substrate in advance. In the second step, the seed-treated substrate is placed in the MOF precursor solution. Under solvothermal conditions, the seeds nucleate and grow preferentially, ultimately resulting in a continuous MOF

polycrystalline membrane with no major defects. Compared to the in-situ growth method, this approach allows for better control over crystal orientation, resulting in a crack-free, dense, and continuous membrane layer. Jin et al. successfully developed a ZIF-300(Zn) membrane on an alumina substrate using the secondary growth method for the first time. This membrane exhibited high and stable performance: its permeability was  $39.2 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$ , while its retention rate for  $\text{CuSO}_4$  was 99.21%[47]. The quality of the seed layer is crucial for the membrane structure, and the seeds must be firmly anchored to the support. Crosslinking agents can effectively enhance the interaction between MOF seeds and porous substrates. Polyethyleneimine (PEI) can form hydrogen bonds with two organic linker groups in the MOF seeds as well as with free functional groups on the substrate surface, such as hydroxyl groups[48].

The secondary growth method allows for the production of MOF membranes with uniform thickness, good orientation, and significantly improved membrane properties. However, its process is more complex than the in-situ growth method. Typically, nanoscale MOF crystals are required to obtain a satisfactory seed layer. Using this method directly to produce high-quality, large-area membranes remains challenging. Currently, there are few applications of membranes prepared by this method in the field of seawater desalination using membrane distillation.

### 3.1.3. Layer-by-Layer Growth

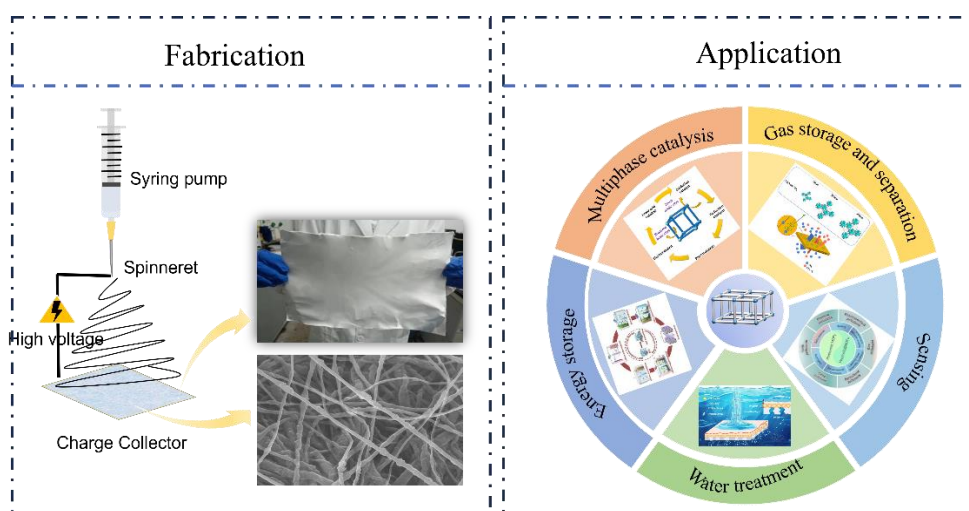
The layer-by-layer growth method, also known as liquid-phase epitaxial growth, is a self-assembly technique in which ultra-thin, uniform, and continuous MOF membrane are constructed layer by layer through alternating adsorption/reactions on the substrate surface. This method precisely adjusts membrane growth parameters (such as precursor concentration and number of growth cycles), thereby enabling precise control over membrane thickness. Shekhah et al. prepared the first MOF-based thin film containing HKUST-1(Cu) using a layer-by-layer deposition method[49]. The layer-by-layer growth method is one of the most promising approaches for achieving large-scale production of MOF polycrystalline membrane. Although this method has advantages such as strong interface bonding, few defects, and ease of scaling up, it consumes a large amount of organic solvent during membrane growth, increasing production costs. Additionally, it has limitations including a long preparation time and low single-layer growth yield.

## 3.2. MOF-Based Composite Membranes

MOF-based composite membranes are prepared by embedding MOF particles as porous fillers within a polymer matrix. Combining the excellent properties of MOFs with the mechanical stability of polymers, this approach has been widely reported in the field of membrane distillation. Research has found that it is possible to improve the trade-off between selectivity and permeability in traditional polymer membranes, while also enhancing the membrane's thermal stability and mechanical strength. This section will systematically review the current research status of MOF-based composite membranes, focusing on their various preparation methods.

### 3.2.1. Electrospinning

The principle of electrospinning is an advanced technique for fabricating fiber membranes by stretching a charged polymer solution or melt in a high-voltage electric field to form nanofibers[50]. Figure 5 shows the preparation and applications of electrospun nanofiber membranes. In this device, a high-voltage power source causes the polymer solution to form a charged jet. Under the electric field generated between the capillary and the collection plate, the jet deforms into a Taylor cone. When the deformation is sufficient to overcome the surface tension of the liquid, the solution jet is ejected onto the collection plate in a spiral shape. The polymer fibers then deposit on the collection plate, while the solvent evaporates, ultimately resulting in the formation of nanofibers[51].



**Figure 5.** Preparation and Applications of Electrospun Nanofiber Membrane.

Since the first application of electrospun nanofiber membranes in the field of membrane distillation in 2008[39]. Its high specific surface area, high porosity, adjustable fiber diameter, interconnected structure, excellent thermal stability, and ease of producing superhydrophobic surfaces make this technology the preferred choice for developing ideal materials for membrane distillation. MOF nanofiber membranes have a wide range of applications in gas separation and storage, catalysis, air pollutant filtration, and water treatment[52]. However, feed liquids in the water treatment field often contain surfactants and hydrophobic pollutants, leading to issues with membrane pore wetting and contamination in electrospun nanofiber membranes (ENMs). Therefore, researchers are focused on developing hybrid matrix membranes composed of organic-inorganic fillers and polymer nanofibers, thereby combining their advantages to improve the membrane's physicochemical properties and separation performance. In particular, research on how MOFs, when combined with nanofibers as organic-inorganic fillers, endow the resulting MOF-nanofiber membranes with excellent properties is ongoing. This section will systematically review the current research status of MOF nanofiber membranes in the field of membrane distillation. Table 2 also summarizes the relevant research findings.

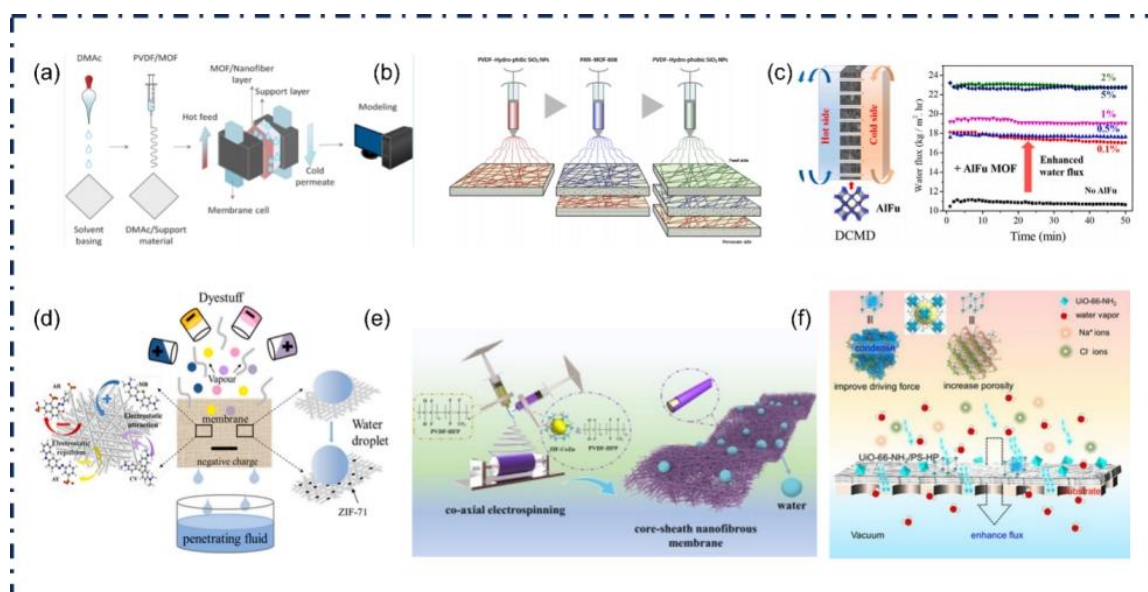
**Table 2.** Electrospun MOF Nanofiber Membranes for Membrane Distillation.

MOF membrane	MOF type	MD configuration	Feed	Temperature (°C)	WCA (°)	Flux (L·m <sup>-2</sup> ·h <sup>-1</sup> )	Ref
MOF-F300/PVDF	MOF-F300	DCMD	3.5% NaCl	48	138.06± 2.18	2.87	[53]
PAN-MOF	MOF-808	DCMD	35 g L <sup>-1</sup>	46 ± 1.5 °C	140.8	4.4	[54]
P/AlFu-2	AlFu MOF	DCMD	3.5% NaCl	40	135 ± 0.3	22.8	[55]
ZIF/PcH	ZIF-71	DCMD	35 g L <sup>-1</sup>	60	134 ± 1.2	20	[56]
PVDF/MAF-4	MAF-4	DCMD	35 g L <sup>-1</sup>	60	/	27.9	[27]
ZIF-CoZn@PVDF-HFP	ZIF-CoZn	DCMD	35 g L <sup>-1</sup>	60	144	21.8	[57]
UiO-66-NH <sub>2</sub> /PS	UiO-66-NH <sub>2</sub>	VMD	3.5% NaCl	70	/	137.6	[58]

This section will provide a review of research achievements related to MOF nanofiber membranes in the field of membrane distillation, organized according to the timeline of publication of relevant literature. In 2018, Yang et al.[53] prepared superhydrophobic polyvinylidene fluoride nanofiber membranes loaded with iron 1,3,5-benzenetricarboxylate MOF at a concentration of up to 5 wt% on nonwoven substrates using electrospinning technology. The membrane remained stable during 5 hours of operation. Compared to the original PVDF membrane (1.86 L·m<sup>-2</sup>·h<sup>-1</sup>), the PV-5 membrane achieved flux of 2.87 L·m<sup>-2</sup>·h<sup>-1</sup> at 48 °C. This represents a 54% increase in flux. Additionally, the NaCl rejection rate was as high as 99.99%(Figure 6a); In 2020, Rana et al.[54] developed a three-

layer nanofiber membrane structure. The top layer consisted of 5 wt% hydrophobic SiO<sub>2</sub>-PVDF nanofibers, the middle layer contained 1.5 wt% MOF-PAN nanofibers, and the bottom layer was made up of 1 wt% hydrophilic SiO<sub>2</sub>-PVDF nanofibers. The membrane flux increases with the increase in the amount of MOF added. A flux of 4.40 L·m<sup>-2</sup>·h<sup>-1</sup> was achieved during 5 hours of direct membrane distillation operation, while excellent retention performance was maintained (Figure 6b); In 2021, Wu et al.[55] incorporated AlFu MOF into polyvinylidene fluoride-co-hexafluoropropylene (PVDF-HFP) nanofiber membranes. The addition of AlFu MOF had a positive effect on improving the membrane's performance. Compared to the PVDF-HFP fiber membrane (10.6 kg·m<sup>-2</sup>·h<sup>-1</sup>), the nanofiber membrane with 2% AlFu MOF added exhibited a flux of 22.8 kg·m<sup>-2</sup>·h<sup>-1</sup>, representing an 114% increase in performance. This demonstrates its excellent membrane distillation capabilities (Figure 6c); In 2022, Huang et al.[56] incorporated ZIF-71 into PVDF-HFP (PcH) to prepare hydrophobic electrospun nanofiber membranes. Compared to the PcH membrane's flux of 14 L·m<sup>-2</sup>·h<sup>-1</sup>, the ZIF-71/PcH membrane exhibited a significantly higher flux of 20 L·m<sup>-2</sup>·h<sup>-1</sup>, representing an increase of 42.8% (Figure 6d); In the same year, Wu et al.[27] prepared PVDF and MAF-4 composite membranes using electrospinning. Compared to the original PVDF nanofiber membrane (17.5 L·m<sup>-2</sup>·h<sup>-1</sup>), the flux of the PVDF/MAF-4 composite membrane was 27.9 L·m<sup>-2</sup>·h<sup>-1</sup>. The high-porosity MAF-4 provides additional steam transport pathways and excellent thermal insulation properties; In 2024, Jiang et al.[57] prepared core-shell structured ZIF-CoZn@PVDF-HFP composite nanofiber membranes using coaxial electrospinning. The flux reached 21.8 L·m<sup>-2</sup>·h<sup>-1</sup>, a 70% increase compared to the pure PVDF-HFP nanofiber membrane (Figure 6e); In 2025, Yao et al.[58] prepared UiO-66-X/PS nanofiber membranes using electrospinning and hot-pressing techniques. The membrane achieved a flux of 137.6 L·m<sup>-2</sup>·h<sup>-1</sup> and a desalination efficiency of 99.95% at 70°C and a vacuum pressure of -85 kPa, outperforming commercial PTFE (28.9 L·m<sup>-2</sup>·h<sup>-1</sup>) as well as most reported polymer membranes (Figure 6f).

The synergistic effect of the high porosity of the electrospun nanofiber membrane and the additional molecular transport pathways provided by the MOFs significantly enhances the flux of the MOF-based composite membrane. The key to improving the performance of fiber membrane distillation lies in the compatibility between MOFs and the polymer matrix, the structure of the MOFs themselves, and the amount of MOFs added.



**Figure 6.** (a) Schematic diagram of Iron-BTC/PVDF nanofiber membrane; (b) Schematic diagram of MOF/SiO<sub>2</sub>-PVDF nanofiber membrane; (c) Schematic diagram of AlFu MOF/PVDF-HFP nanofiber membrane; (d) Schematic diagram of ZIF-71/PVDF-HFP nanofiber membrane; (e) Schematic diagram of ZIF-CoZn@PVDF-HFP composite nanofiber membrane; (f) Schematic diagram of UiO-66-X/PS nanofiber membrane.

### 3.2.2. Dip Coating Method

the surface of plate-shaped, cylindrical, or irregularly shaped objects in order to create a film. The polymer concentration in the impregnation solution, impregnation time, and cross-linking agent concentration are the key process parameters of this method. These parameters affect the thickness of the coating material, the pore size of the membrane, and the integrity of the membrane structure[59]. Rahimpour et al. [53] coated a polyvinylidene fluoride substrate with an ultra-thin ZIF-8/chitosan composite layer, thereby enhancing the membrane distillation performance of the membrane for seawater desalination. Cao et al.[60]successfully synthesized nanoscale and micrometer-sized ZIF-8 particles by systematically studying the rules governing particle size control. After hydrophobic modification using POTS, these particles were deposited on a PTFE substrate via spraying. This membrane exhibits superior permeate flux compared to commercial PTFE membranes during the membrane distillation process. This research not only provides an innovative strategy for the preparation of superhydrophobic coatings but also expands the application potential of MOF materials as such coatings. Niu et al.[61] developed TFMOF/PDMS Janus membranes. Through in-situ growth of polyaniline, hydrophilic modification with polydimethylsiloxane (PDMS) was performed, followed by deposition of zirconium-based metal-organic framework (TFMOF) particles using a dip-coating process. This approach combines asymmetric wettability structures with photocatalytic membrane distillation to provide a solution for coal chemical wastewater treatment.

Although the dip-coating process is simple and easy to operate, with low equipment requirements and low costs, it is also suitable for large-scale production. However, there are issues such as poor interfacial compatibility between MOFs and the substrate, as well as a decline in membrane permeability and mechanical properties under high loading conditions. To study the synergistic control strategies of developable substrate pretreatment (plasma, surface grafting) and dip-coating parameters (rotation speed, solution concentration, temperature). By combining in-situ growth techniques, directed loading of MOFs within the pores of the substrate can be achieved, thereby enhancing the practical application potential of this technology.

### 3.2.3. Other Methods

Phase transition method is one of the core techniques for preparing MOF hybrid matrix membranes. This method is based on the thermodynamically driven phase separation principle, converting a homogeneous polymer solution (or composite precursor solution) into a membrane material with a specific porous structure. First, the polymer is dissolved in a solvent to prepare a homogeneous polymer solution. Subsequently, a film with a thickness of 100–200  $\mu\text{m}$  was prepared. Finally, the prepared polymer film is immersed in a coagulation bath using water as the non-solvent, causing the polymer to precipitate out of the solution[57]. Rahimpour et al.[62] developed a new type of thin-film composite (TFC) membrane by coating a ultra-thin layer of zeolite imidazolate framework (ZIF-8)/chitosan on the surface of a PVDF substrate. The film was treated by immersing it in a non-solvent bath using the phase transition method. The results showed that the ZIF-8/chitosan modified membrane had a NaCl retention rate of over 99.5%.Xie et al.[63]successfully prepared a novel hydrophobic hybrid PVDF hollow fiber membrane doped with aluminum fumarate metal-organic framework (AlFu MOF) using the dry spray-wet phase transition method. The AlFu MOF additive has a positive effect on improving membrane performance.

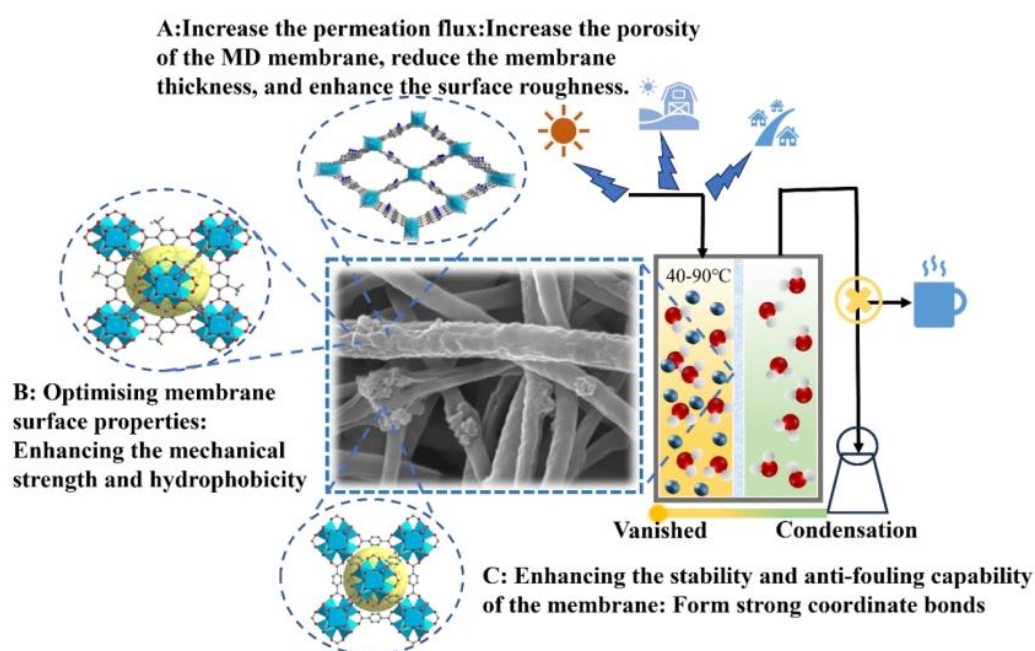
Solution casting is a technique in which a solution is applied to a substrate, and then the solvent is evaporated to form a thin film. Xie et al.[64] prepared hydrophilic/hydrophobic bilayer membranes by solution casting on a hydrophobic microporous polytetrafluoroethylene (PTFE) substrate. Composed of a dense layer of polyvinyl alcohol (PVA) doped with hydrophilic aluminum fumarate AlFu MOF. With the adjustment using nano-porous AlFu MOF, the water vapor flux reached 45.1  $\text{L}\cdot\text{m}^{-2}\text{h}^{-1}$ , while a 99.9% retention rate was maintained in the DCMD process.

Vacuum filtration is a classic and efficient method for preparing metal-organic framework membranes. Using the vacuum negative pressure difference as a driving force, the suspension containing MOF nanoparticles (or precursors) is passed through a porous support. Enable MOF

particles to rapidly accumulate and form a continuous, dense film layer on the surface or within the pores of the support material. This method is simple to operate, has a short preparation time, and allows for easy control of membrane thickness. Chen et al.[65] modified UiO-66-NH<sub>2</sub> using polyvinyl alcohol (PVA) as a crosslinking agent. The modified UiO-66-NH<sub>2</sub> was then immobilized on a PTFE membrane through vacuum filtration, resulting in the development of a new type of Janus composite membrane. In the 24 hour DCMD experiment, a desalination rate of 99.99% was achieved, with a flux of 21.3 L·m<sup>-2</sup>·h<sup>-1</sup>. The research team also used a crosslinking agent to fix GO and UiO-66-NH<sub>2</sub> onto the PTFE hydrophobic membrane. Compared to the original PTFE membrane, the Janus membrane exhibits superior moisture resistance. The flux was maintained at 21.2 L·m<sup>-2</sup>·h<sup>-1</sup>, and the retention rate reached 99.9% after 48 hours of continuous operation[66]. Moriones et al.[67] deposited the MOF@GO nanohybrids onto two layers of polydopamine (PDA) positioned on top of a commercial polyethylene (PE) film using vacuum filtration. Compared to the GO membrane (5 L·m<sup>-2</sup>·h<sup>-1</sup>), the MOF@GO membrane has a higher flux (13 L·m<sup>-2</sup>·h<sup>-1</sup>). In summary, the addition of MOFs not only increases membrane flux but also enhances the membrane's water-repellent properties, demonstrating significant potential applications in the field of membrane distillation for seawater desalination. In summary, MOF polycrystalline films possess desirable properties due to their customizable pore structures, excellent hydrophobic/superhydrophobic control capabilities, high chemical stability, and good thermal stability. It provides a new material option to address the bottleneck issues in traditional membrane distillation processes, such as membrane fouling, low permeate flux, and low thermal efficiency. The MOF composite membrane has significant advantages over MOF polycrystalline membranes in terms of scalability and cost control, demonstrating strong commercial potential.

#### 4. The Working Principle of MOF in Membrane Distillation

Metal-organic frameworks possess high porosity, extremely large specific surface area, and customizable pore structures. By regulating the pore structure, hydrophobicity, thermal stability, and anti-fouling properties of the membrane, introducing it into the membrane distillation system for the preparation of MOF-based membranes can significantly enhance the flux and long-term operational stability of membrane distillation. This section explores the working principles of MOFs in membrane distillation. The key mechanisms include enhancing permeate flux, optimizing membrane surface properties, and improving membrane stability and anti-fouling capabilities, as illustrated in Figure 7.



**Figure 7.** Schematic diagram of the working principle of metal-organic frameworks in membrane distillation.

#### 4.1. Increasing Permeate Flux

MOFs enhance permeation flux by increasing the porosity of MD membranes, reducing membrane thickness, and increasing surface roughness[68]. The pores in commercial hydrophobic membranes (such as PVDF and PTFE membranes) are typically large, disordered pores, resulting in long diffusion paths for water vapor with high resistance. Increasing the porosity of the MD membrane enhances the effective surface area available for water vapor evaporation, thereby providing more pathways for transport[69]. Cheng et al.[63] prepared hollow fiber hydrophobic membranes doped with AlFu MOF. AlFu MOF makes the originally dense PVDF membrane structure porous. Increase porosity while enhancing the mass transfer pathways of the membrane. The increase in effective porosity not only enhances the membrane's pore size but also reduces the tortuosity of the membrane pores[70]. Reducing the thickness of the MD membrane shortens the actual distance that water vapor must travel through the membrane, thereby helping to decrease mass transfer resistance. Guo et al.[71] incorporated copper-based metal-organic framework materials (Cu-CAT) into hydrophilic/hydrophobic polyvinylidene fluoride nanofiber membranes, creating photothermal-responsive Janus nanofiber membranes. This approach reduced the thickness of the photothermal layer by 15  $\mu\text{m}$ . In terms of increasing roughness, MOFs create a uneven, rough surface structure by forming interconnected pore networks with the membrane material[67]. Geng et al.[57] prepared a series of ZIF-CoZn@PVDF-HFP composite nanofiber membranes with core-shell structure using coaxial electrospinning. The functional nanoparticles were uniformly and stably dispersed on the surface of the nanofibers, resulting in secondary roughness. In addition, the permeate flux of the membrane is proportional to its porosity and inversely proportional to its thickness and curvature[72]. Too large an aperture will affect the solute retention efficiency, while too thin a membrane thickness will impair its mechanical properties. Therefore, an ideal MD membrane needs to achieve a balance among retention rate, mechanical strength, and permeate flux in order to fulfill its potential for practical applications.

#### 4.2. Optimizing Membrane Surface Properties

Composites formed by MOFs and a matrix can enhance the interactions between molecular chains in the membrane material, thereby improving the membrane's mechanical strength and toughness. This allows the membrane to withstand external forces in harsh water environments while reducing the risk of membrane rupture[73]. In addition, the surface chemical properties of MOFs can be manipulated through ligand modification. Introducing fluorine- or alkyl-containing organic ligands into the MOF structure, or performing hydrophobic modifications on the MOFs, can significantly enhance the surface hydrophobicity of the membrane. Chuang et al.[40] synthesized MOF crystals in symbiosis on alumina tube substrates, Then, perfluorinated molecules are introduced onto the surface of the MOF-functionalized membrane. This membrane retains the high water permeability and resistance to high temperatures and pressures inherent in the alumina substrate, while also possessing a hydrophobic surface. Xu et al.[74] developed a novel porous, hydrophobic MOF-801@GO-PDMS membrane. PDMS contributes to the membrane's hydrophobic stability by utilizing its inherent hydrophobicity and the cross-linking of its molecular fragments.

#### 4.3. Improving Membrane Stability and Anti-Fouling Properties

As the operating time of membrane distillation prolongs, solutes gradually accumulate on the membrane surface and clog the pores, resulting in reduced flux and membrane wetting, ultimately leading to damage to the membrane structure[75]. By optimizing the metal/ligand composition and surface modification of MOFs, the stability of the membrane under extreme pH conditions and its tolerance in extreme temperature environments can be enhanced[33,35]. Highly charged metal ions with high charge densities form strong coordination bonds with oxygen-containing ligands such as carboxylate groups. Thus effectively resisting acidolysis caused by attacks from hydrogen ions ( $\text{H}^+$ ) or substitution by hydroxyl groups ( $-\text{OH}$ ). The UIO series, which contains high-valent metals such as

Zr<sup>4+</sup> and Hf<sup>4+</sup>, exhibits strong acid resistance and can operate effectively for extended periods in high-salinity and highly acidic environments. It still maintains good structural integrity and acid resistance, making it a candidate material for numerous research projects currently. In contrast, MOFs formed from low-cost metals have weaker bonding energies and tend to undergo hydrolysis in non-neutral pH environments, leading to the collapse of their crystal structures. In general, by selecting appropriate metals/ligands and modifying the surface, the stability of MOFs under extreme pH conditions can be effectively improved.

## 5. Development Trends and Challenges of MOF-Modified Membranes

### 5.1. Improving the Hydrophobicity of MOF Membranes

Membrane distillation requires the use of superhydrophobic or fully hydrophobic membranes to prevent pore wetting. Pore wetting occurs when the aqueous phase penetrates the membrane, resulting in the loss of its hydrophobic properties[35]. Inspired by natural surfaces with hydrophobic and self-cleaning properties such as lotus leaves, gecko feet, and mosquito eyes, highly hydrophobic membranes can be designed and fabricated to enhance their resistance to wetting. Shi et al.[76] synthesized superhydrophobic and superoleophilic metal-organic framework (MOF) materials by carrying out an amide reaction between amino groups and octadecyl chloride. The introduction of long alkyl chains reduced the surface free energy, enabling the successful preparation of a superhydrophobic zirconium-based metal-organic framework (UiO-66-NH-C18). Liu et al.[77] proposed that thermal activation can be used to remove hydroxyl groups from the framework of MOFs, thereby creating a hydrophobic surface that enhances the membrane's resistance to wetting. The hydrophobic MOF membrane has a water flux of 12.8 L·m<sup>-2</sup>·h<sup>-1</sup>, with a NaCl rejection rate of over 99.9%. Therefore, the preparation of hydrophobic MOF-based membranes is one of the directions for future development.

### 5.2. Achieving Large-Scale Production of MOF Membranes

The key to large-scale production of MOF membranes lies in developing a process that allows for continuous manufacturing. Wang et al.[78] reported a thermally induced phase separation hot-pressing (TIPS-HoP) strategy. Ten different MOF membranes can be prepared using the roll-to-roll method. However, Large-scale production tends to result in defects such as pinholes and grain boundary cracks, thereby directly reducing separation efficiency. At the same time, the interaction between MOF crystals and the support materials (such as Al<sub>2</sub>O<sub>3</sub> ceramics or PVDF polymers) is primarily physical adsorption. This results in poor interfacial compatibility. During scaled-up production, membrane detachment or peeling can occur due to fluctuations in tension and pressure. Zheng et al. used gravity-induced deposition to create a mixed matrix polyacrylonitrile (PAN) substrate with a ZIF-8-rich seed layer, utilizing the gradient distribution of ZIF-8 before the phase transition occurred (ZS-PAN). Subsequently, with the assistance of tannic acid (TA), a dense and defect-free ZIF-8 film was prepared on a flexible polymer substrate, followed by the secondary growth of ZIF-8[79]. While this technology provides a approach for preparing defect-free MOF membranes, large-area, continuous production of such membranes remains a significant challenge[17].

### 5.3. Improve the Operational Stability of MOF Membranes

Most MOFs (such as ZIF-8, UiO-66) are prone to crystal dissolution, structural collapse, or ligand loss under conditions of high humidity, highly polar solvents, or extreme pH values[80]. Therefore, zirconium-based/aluminum-based MOFs with high chemical stability (such as UiO-66-NH<sub>2</sub>, MIL-53) are selected, cross-linking and modification using polydopamine and silane coupling agents to enhance the interconnections between MOF crystals[81], hydrophobic modification of the membrane surface using long-chain alkyl or fluorine-containing reagents to prevent direct contact between

water and the MOF framework, these are all effective strategies for improving material stability. In the future, efforts will still be needed to address issues related to practical applications, such as membrane wetting, membrane fouling, and insufficient long-term stability, in order to promote the industrialization of MOF-based membranes.

## 6. Summary

The ability of MOFs to have regularly structured pores and surface sites that can be easily functionalized makes them excellent candidates for membrane distillation. Although some fundamental research achievements have been made in the field of membrane distillation using MOF polycrystalline membranes and MOF-based composite membranes, industrial application has not yet been achieved. The main challenges include the following three aspects:

(1) scalable film-making equipment has not yet been developed, making it impossible to meet the demands of large-scale industrial production.

(2) the cost issue associated with the processing and synthesis of MOF materials remains a significant challenge.

(3) fundamental issues such as particle agglomeration, poor compatibility, easy degradation, and limited loading capacity of MOFs during film formation require further research.

With the continuous development of interdisciplinary research and technological advancements, MOFs membranes are expected to become key technologies in areas such as high-salinity wastewater treatment and seawater desalination. They will provide important technical support for addressing global water shortages and achieving the “dual carbon” goals.

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