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Covalent Organic Framework Membranes for Selective Ion Separation

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ABSTRACT

The escalating global urgency for clean water and critical mineral recovery necessitates separation technologies that transcend the inherent permeability-selectivity trade-offs of conventional polymeric membranes. Covalent Organic Frameworks (COFs), distinguished by their precise reticular chemistry, tunable pore architectures, and modular functionality, have emerged as a transformative platform for next-generation ion separation. This review systematically evaluates recent breakthroughs in the engineering of COF-based membranes, focusing on the nexus between fabrication strategies and application performance. We critically examine diverse synthesis methodologies, including interfacial polymerization, in situ growth, and electrophoretic deposition, highlighting how these techniques control membrane crystallization and defect minimization. Furthermore, the review details the deployment of these materials across three pivotal sectors: seawater desalination, where they achieve superior salt rejection; lithium resource extraction, enabling high-precision Li^+/Mg^{2+} differentiation; and the purification of drinking water from heavy metals and radionuclides. The discussion concludes by identifying the critical bottlenecks in scalability and long-term stability that must be addressed to transition COF membranes from laboratory prototypes to industrial implementation.

KEYWORDS

Covalent organic frameworks; Membranes; Ion separation; Desalination; Lithium extraction

1. INTRODUCTION

The global demand for clean water and the effective removal of ionic contaminants have emerged as urgent challenges at the intersection of environmental protection and public health [1, 2]. Driven by rapid urbanization, industrial expansion, and the increasing utilization of nuclear technology, there is an urgent need for efficient and energy-saving separation technologies capable of eliminating salts [3, 4], heavy metals [5, 6], organic contaminants [7, 8], and radiotoxic species [9, 10] from complex aqueous environments.

Membrane separation is widely recognized as an energy-efficient and scalable strategy for water purification and ion removal. However, conventional membrane materials—such as polyamide reverse osmosis [11, 12] and polysulfone ultrafiltration membranes [13, 14] continue to suffer from inherent limitations, including the well-known permeability selectivity trade-off, insufficient chemical and mechanical stability under harsh operating conditions, and vulnerability to fouling and chlorine-induced degradation. These challenges have inspired the design of advanced membrane materials that offer precise molecular sieving, tunable chemical functionality, and long-term structural robustness. Over the past decade, covalent organic frameworks (COFs) have emerged as a highly promising class of porous and crystalline polymers for next-generation membrane-based separations [15, 16]. Constructed from modular organic building blocks connected by robust covalent bonds, COFs possess several characteristics that make them particularly attractive for ion separation [17, 18]. These include ordered and tunable pore channels [19, 20], high specific surface area [21, 22], low framework density, and the ability to incorporate well-defined functional groups at precisely predetermined positions within the pore walls [23], enabling tailored interactions with target ions. These features allow for rational pore-size engineering from the sub-nanometer to nanometer scale, the targeted incorporation of charged or coordinating sites to enhance ion affinity, and the design of anisotropic transport pathways. Together, these capabilities can be leveraged to overcome the conventional flux–selectivity trade-off and to achieve selective separation of specific ionic species.

Despite their numerous advantages and the growing number of promising laboratory demonstrations, COF membranes still face three interrelated challenges that hinder their transition from proof-of-concept studies to practical ion separation technologies. First, achieving continuous and defect-free COF membranes that simultaneously possess atomic-level order and sufficient mechanical strength remains a significant challenge. Many synthetic routes yield polycrystalline powders that are difficult to transform into thin, large-area films, while interfacial and vapor-phase synthesis methods capable of producing high-quality membranes still face obstacles in terms of scalability and substrate compatibility. Second, although COFs can be functionalized to selectively bind or repel target ions, achieving high selectivity in realistic multicomponent aqueous environments—such as seawater or nuclear wastewater containing high ionic strength, competing cations/anions, and organic contaminants—remains challenging. Complex interactions among competitive binding, Donnan effects, and pore confinement phenomena occur in intricate and often interdependent ways that have yet to be fully understood or quantitatively described. Third, under harsh operating conditions—including high salinity, wide pH ranges, oxidizing agents and ionizing radiation encountered in nuclear wastewater—the long-term chemical, mechanical, and radiation stability of COF membranes has not been thoroughly investigated and remains a critical bottleneck for their industrial implementation.

In the context of ion separation, three particularly timely and demanding application domains have shaped the focus of this review: (i) seawater desalination and brine treatment, which require sub-nanometer pore control and high water permeability to effectively exclude hydrated salt ions; (ii) purification of drinking water and industrial wastewater, where selective removal of heavy metals and toxic cations (e.g., Pb^{2+}, Hg^{2+}, Cd^{2+}) as well as oxyanions must be achieved in the presence of natural organic matter and hardness ions; and (iii) removal of radioactive contaminants (e.g., U(VI), Th(IV), Cs^{+}, Sr^{2+}, TcO_4^{-}), where the combination of trace concentrations, high ionic strength, and specific coordination chemistry demands membrane materials capable of both molecular sieving and strong selective binding. Therefore, this review systematically investigates the recent progress of COF-based membranes in these three application areas, focusing on structure-property relationships, ion transport mechanisms, and emerging design strategies aimed at achieving selective and robust ion separations under realistic conditions. COFs can be selectively functionalized with groups such as amidoxime, phosphonate, thiol, and pyridyl moieties to provide tailored coordination environments suited to the chemical characteristics of actinides and fission products. Early studies have demonstrated that COFs exhibit excellent adsorption capacity and selectivity, and can be further processed into membrane forms or incorporated into mixed-matrix composite materials. This review focuses on COF membranes for ion separation, with the explicit goal of bridging advances in material synthesis, fundamental transport mechanisms, and application-driven performance requirements. We emphasize ion-specific design principles and the interplay among three core separation mechanisms—spatial (size-exclusion) effects, electrostatic (Donnan) interactions, and specific chemical coordination, and illustrate how these mechanisms are applied in seawater desalination, lithium extraction from salt lakes, drinking water purification, and the remediation of radioactive nuclides.

2. PREPARATION METHODS OF COF MEMBRANES

The design and fabrication of COF membranes must comprehensively consider the type of covalent linkage, pore architecture, and monomer geometry in order to achieve an optimal balance between separation performance and structural stability. The most common covalent linkages in COFs are boronate ester bonds and imine bonds. The former are typically formed through the condensation reaction between boronic acids and diols, exhibiting a high degree of reversibility that facilitates the formation of COFs with high crystallinity, uniform pore size, and excellent structural periodicity. Consequently, boronate ester–linked COFs were widely employed in early two-dimensional COF systems. However, the boron–oxygen (B–O) linkage is highly sensitive to moisture and tends to undergo hydrolysis in humid or acidic environments, which can lead to structural collapse and a deterioration of membrane performance [24, 25]. In contrast, imine linkages (C=N) are formed through the condensation reaction between aldehydes and amines, exhibiting superior chemical stability that allows the framework to maintain its structural integrity even under harsh pH conditions or in various organic solvent systems. Although imine linkages have weaker reversibility and slightly lower film crystallinity, it is possible to obtain stable COF membranes with good structural order under mild conditions by optimizing the catalytic system, solvent environment, and reaction temperature.

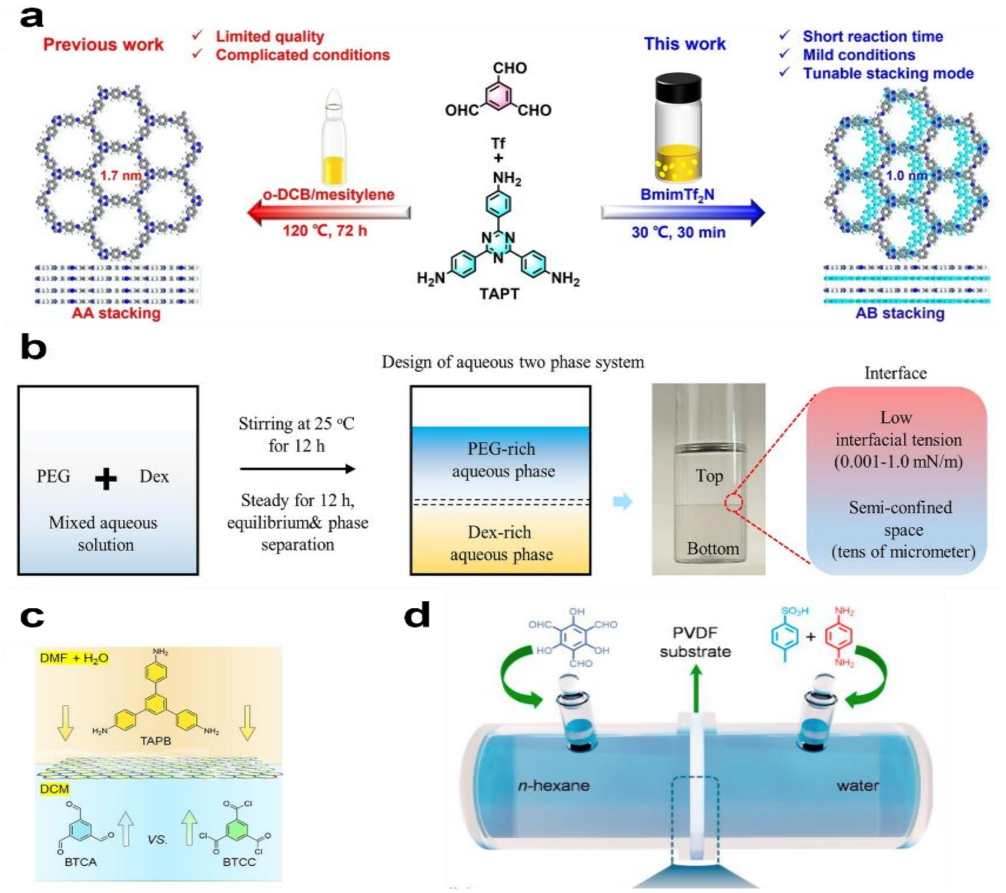

Figure 1. (a) Schematic of two different routes to prepare COF-TAPT-Tf with different stacking modes. (b) Designing process for ATPS. (c) Synthetic strategy and procedure of the representative AmCOF-1 membrane. (d) Schematic illustration of diffusion cell for TpPa growth by UDS

2.1. Interfacial Polymerization

Interfacial polymerization is one of the primary methods for fabricating COF membranes, especially in applications such as nanofiltration and reverse osmosis. In this approach, the membrane structure is generated through monomer polymerization confined at the interface between two immiscible phases, ensuring controlled film growth and structural uniformity. Common interface configurations include liquid–liquid, gas–liquid, and solid–gas interfaces. During interfacial polymerization, the reaction is restricted to the interfacial region, which plays a critical role in controlling the structural order of the COF framework and ensuring uniform membrane growth. Typically, the monomers are distributed between two immiscible phases, with amines dissolved in the aqueous phase and aldehydes or boronic acids in the organic phase. During the reaction, the monomer diffusion rate and reaction kinetics significantly influence the thickness, crystallinity and uniformity of the resulting membrane To enhance the membrane's mechanical strength, COF membranes are commonly formed on top of a porous substrate for better support. By adding certain functional groups to the substrate surface, the interaction between the COF layer and the substrate can be strengthened, which helps improve the membrane's structural integrity and long-term stability during operation.

Liquid–liquid interfacial polymerization confines the polymerization reaction to the interface between two immiscible liquids. Such systems can involve various biphasic combinations, including organic/aqueous, organic/organic, or aqueous/aqueous phases (figure 1a-c) [26, 27, 28]. For instance, Zhu and co-workers successfully prepared a 3D sulfonic acid-functionalized COF membrane by reacting sulfonic acid-functionalized aldehyde monomers and tetra (4-phenyl) methane monomers in a dual acid-containing organic and aqueous two-phase system. The 3D SCOF membrane obtained has fully extended, highly interconnected channels, subnanometer (<1 nm) pores, and a high density of sulfonate groups. These features allow rapid proton and cation transport while maintaining strong selectivity for anions. Under 100% relative humidity at 90°C, the membrane achieves a proton conductivity of 843 mS cm^{-1} [29]. Wang et al. employed an interfacial polymerization method directly on a porous polysulfone (PSF) ultrafiltration substrate. They simultaneously added an aqueous solution of phenylenediamine (Pa) and a hexane solution of 1, 3, 5-triformylphloroglucinol (Tp) to either side of a diffusion cell. During the reaction, Pa molecules could penetrate the PVDF substrate immobilized on the diffusion cell and react with Tp molecules, producing a TpPa COF membrane composited with the substrate (figure 1d). The TpPa/PVDF membrane prepared using this method exhibited excellent performance, surpassing other reported membrane materials for dye separation [30]. Shi's team successfully fabricated robust three-dimensional COF membranes containing interconnected nanochannels using oligomers pre-synthesized at the organic-aqueous interface. Due to the amphiphilic nature and suitable size of the oligomers, they are easily captured at the interface. After maturation at the interface, these oligomers form a crystalline, self-supporting 3D COF membrane with controllable thickness. The resulting membranes exhibit uniformly interconnected pores with pore sizes ranging from approximately 0.8–1.5 nm, which can be tuned by selecting different monomers [31]. Xian et al. introduced an innovative approach for fabricating covalent organic framework (COF) membranes. Using acid-catalyzed liquid–liquid interfacial polymerization, a vertical protonation gradient of the imine bonds was established during synthesis, enabling spatially controlled deprotonation. This process facilitates the formation of heterojunctions both within and between membrane layers, enhances interlayer hybridization, and generates a strong internal electric field under illumination, thereby improving light absorption and suppressing charge recombination. As a result of these structural optimizations, the membranes exhibited remarkable ion transport under extreme concentration gradients (2000:1), achieving a flux of approximately 3.2×10^{12} ions·s^{-1}·cm^{-2} and reducing ion concentrations to ppb levels, demonstrating significant potential for high-precision seawater desalination [32].

2.2. In Situ Growth

In situ growth is an effective method for constructing COF membranes directly on substrate surfaces. This method eliminates the need for membrane transfer and ensures a strong bond between the membrane and the substrate during the reaction. By manipulating reaction time, temperature, and precursor concentration, the membrane thickness and morphology can be precisely tuned. In situ-grown COF membranes typically exhibit high crystallinity and an ordered pore structure. Furthermore, this method exhibits excellent adaptability and scalability to a variety of substrates, making it widely used in the construction of high-performance separation membranes. Subsequently, the BDD and TPA solutions were mixed, and Al_2O_3 along with the substrate was immersed in the mixture, followed by treatment under vacuum for 30 minutes. Next, the mixture with the Al_2O_3-loaded substrate was heated at 160°C for the reaction. Through this method, the researchers successfully achieved the in situ growth of CTFs on the α-Al_2O_3 matrix, ultimately forming a stable composite membrane (figure 2c). The resulting M-CTFs composite membrane demonstrated great potential in dye desalting applications [33]. Pan's team fabricated continuous COF-based membranes by growing COF layers on polyacrylonitrile substrates. They immersed the modified polyacrylonitrile substrate in a monomer solution and in situ synthesized the COF-based membrane at room temperature via a Schiff base reaction. Thanks to the unique microporous channels of the membrane, efficient retention of dye molecules was effectively achieved [34]. The TpEB generated in situ in the PAN solution is uniformly distributed within the polymer matrix, forming a composite structure with good crystallinity and interface compatibility. The resulting membrane exhibits a uniform pore structure and good mechanical stability, demonstrating excellent separation performance. Using the same strategy, COF-PAN composite membranes containing different COF units can also be constructed, further expanding the applicability of this method [35].

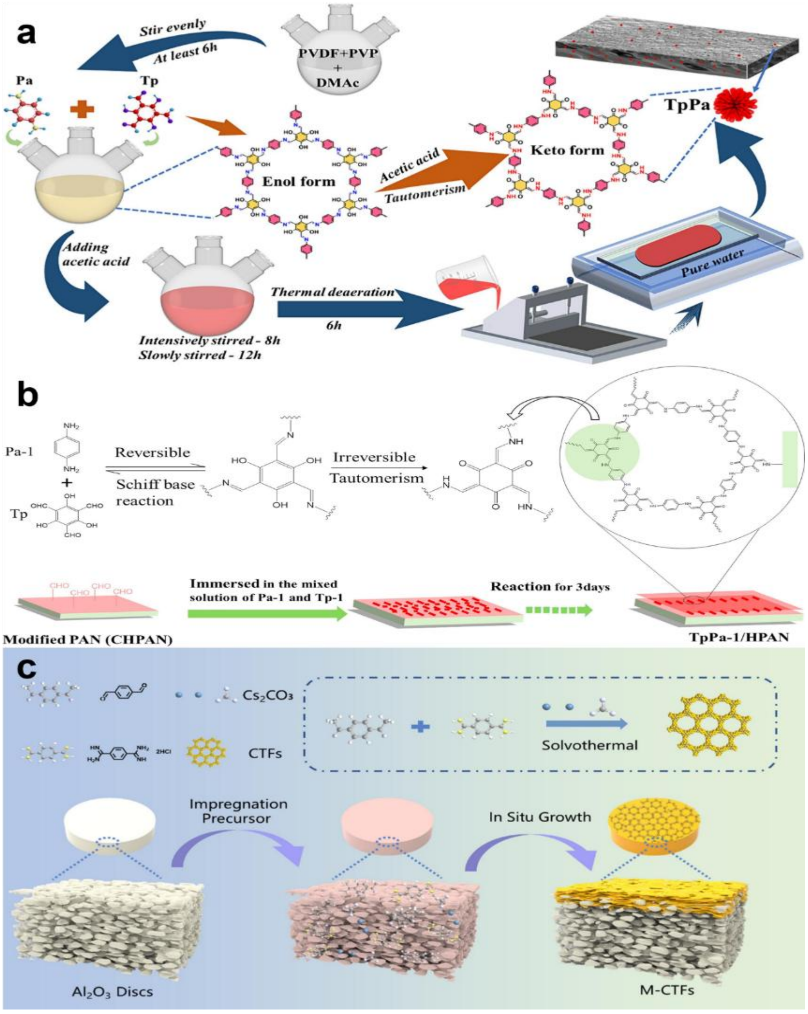

Figure 2. (a) Preparation process of TpPa/PVDF membrane. (b) Scheme of the preparation process of the TpPa-1/HPAN membrane. (c)Preparation scheme of covalent triazine frameworks membranes (M-CTFs) through in-situ growth method

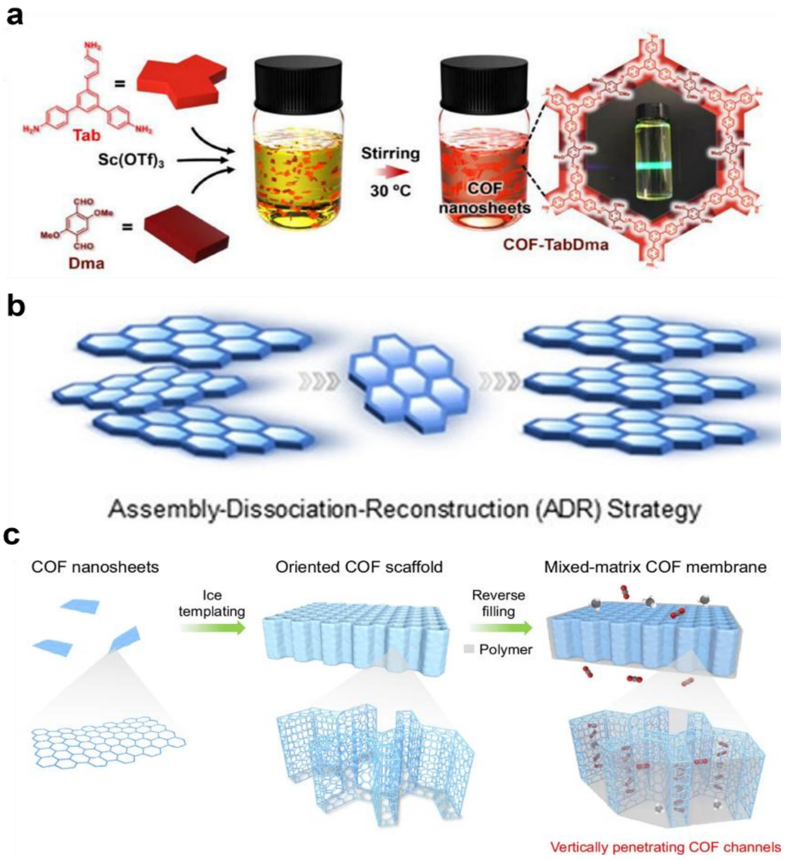

Figure 3. (a) Schematic illustration of the synthesis of COF-TabDma nanosheets; inset shows Tyndall light-scattering effect in the suspensions. (b) Schematic illustration of the assembly-dissociation-reconstruction (ADR) method for the synthesis of COF membrane. (c)Schematic illustration of reverse-filling approach to fabricating MMMs with continuous and vertically penetrating COF channels for CO_2/CH_4 separation

2.3. Post-Assembly

Han et al. synthesized COF nanosheets composed of 1, 3, 5-tris (4-aminophenyl) benzene and 2, 5-dimethoxyterephthalaldehyde using a single-phase solution method.The resulting COF material demonstrated excellent hydrophobicity, chemical stability, and a mesoporous structure, all of which contribute to enhanced mass transfer efficiency (figure 3a). The formation of nanosheets was confirmed by Tyndall light scattering observed in the suspension. The uniform nanoscale pores of the PAN membrane promoted efficient deposition and delamination of the nanosheets. The membrane bioreactor was fabricated using a vacuum-assisted layer-by-layer stacking technique on a polyacrylonitrile (PAN) ultrafiltration membrane [36]. Wu and his team dissolved Tp in 1.5 mL of meslin and Pa in a mixed solvent of acetonitrile and dioxane. They then added 3 M COOH and allowed the reaction to proceed at room temperature for 36 hours, resulting in a red precipitate, which was subsequently washed and dried. At the same time, in order to improve the binding force between PAN substrate and COF, hydrolyzed PAN substrate was prepared in advance. TpPa thin films were prepared using a solvent-induced interfacial polymerization strategy. A suitable amount of Pa was dissolved in the aqueous phase, while Tp was dissolved in the organic phase for the reaction. The resulting COF film was collected and washed to remove any residual chemicals. A similar method was applied to further synthesize TpPa thin films on HPAN substrates, yielding a film with an effective synthesis area of approximately 5.7 cm² [37]. Zhang et al. designed double-node and triple-node building blocks for the assembly of COFs, first conducting a condensation reaction in the temperature range of 80-100°C to synthesize COF nanosheets. Next, the COF nanosheets were reassembled in aqueous solution in a water vapor environment at 60°C, and the resulting reconstructed film had higher crystallinity than the original COF nanosheets(figure 3b). By comparing the experimental data with the simulation results, it was found that the optimized AB stacking structure can effectively construct one-dimensional (1D) channels while reducing the pore size in COF [38].

2.4. Mixed Matrix Membranes

Anionic TpPa-SO3H COF nanosheets were synthesized using a previously established single-solution phase method. First, a COF nanosheet suspension was cast onto a flat substrate and frozen perpendicular to the casting solution under a temperature gradient. The freezing of water induced phase separation in the aqueous nanosheet suspension. As ice crystals slowly formed, COF nanosheets were expelled from the ice and spontaneously accumulated, progressively organizing into an ordered structural template between the vertically growing ice crystals(figure 3c). Finally, the ice template was removed via freeze-drying, yielding an Ori-COF membrane with exceptional thermal and solvent stability [39]. To combat the deteriorating climate, the fabrication of highly permeable and selective membranes for CO2 enrichment and separation is of great value. In this study, zirconium-based metal-organic frameworks (Zr-MOFs) containing defective structures (defective UiO-66-NH2, D-UN) with high volumetric yield (12 g L^{-1} in a single batch) were prepared for the first time at room temperature using a simple green synthesis strategy. After modification with pentafluorobenzaldehyde, defect-engineered UiO-66-NH_2 (D-UN) nanoparticles were transformed into fluorine-containing F-g-UN, exhibiting localized and densely distributed fluorine on the particle surfaces. Fluorine-functionalized mixed matrix membranes exhibited both high permeability and selectivity, resulting from the synergistic effects of enhanced CO_2 affinity and improved filler dispersion conferred by the fluorination [40].

2.5. Other Methods

Electrophoretic deposition (EPD) is a widely used colloidal-based approach for rapid thin-film fabrication. In this process, charged particles dispersed in a stable suspension migrate toward an electrode under an applied electric field, where they accumulate to form a uniform and dense coating. Wang et al. demonstrated that H_3 can serve as a model COF probe for the synthesis of ionic COF nanosheets, which were successfully fabricated via interfacial polymerization in water. The ionization of sulfonic acid groups (–SO_3H) confers a negative charge to the COF backbone, which drives the slow interfacial migration of aldehyde monomers dissolved in n-octanoic acid toward aqueous amine monomers. Under the combined influence of diffusion and solvent-mediated effects, this controlled interfacial polymerization produces uniform ionic COF nanosheets (iCON). This study highlights the feasibility and tunability of interfacial polymerization for constructing ionic COF thin-film structures [41]. Zhang et al. utilized electrophoretic deposition (EPD) to assemble composite films comprising MXene nanosheets and COF nanoparticles. Uniform COF nanoparticles (COF NPs) were first synthesized from TAPB and PDA precursors. MXene nanosheets and COF NPs were then alternately deposited onto a polyamide (PA) microfiltration substrate using a layer-by-layer electrophoretic deposition (LBL-EPD) strategy, yielding a MXene/COF composite membrane with a characteristic sandwich-like structure (figure 4a). This approach enables precise control over the interfacial assembly by carefully regulating the electric field direction and deposition time. Compared with conventional solution self-assembly or vacuum filtration, the layer-by-layer electrophoretic deposition (LBL-EPD) process offers advantages including operational simplicity, high film uniformity, and tunable thickness, providing an efficient and controllable strategy for constructing high-performance, multifunctional COF membranes [42]. The fabrication of conventional COF membranes is often hindered by the rigid framework and the rapid aggregation and precipitation of nanoparticles during network formation, making it challenging to obtain crystalline membranes with continuous and self-supporting structures. To overcome this limitation, a sol–gel method has been developed for the preparation of free-standing crystalline COF films. By introducing a small amount of a linear, flexible co-crosslinker into the COF synthesis system, interlayer interactions between nanocrystals are enhanced, promoting their orderly counter-diffusion growth and enabling the formation of continuous, dense, and structurally intact COF films.In this process, solvent selection is critical. Protic solvents, which stabilize charged intermediates and facilitate oligomer formation, can markedly enhance the stability of the gel system and its ability to form continuous COF membranes (figure 4b). In a model system based on a two-dimensional Schiff base COF constructed from triacylphloroglucinol (TP) and p-phenylenediamine (PDA), the introduction of polypropylene glycol bis (2-aminopropyl ether) (PPG-NH_2) as a flexible molecular chain enabled controlled nanocrystal assembly and the sol–gel transformation, providing a strategy to form continuous and uniform COF films. This strategy effectively suppresses excessive particle growth and agglomeration, yielding a precursor sol with uniform particle size and high stability, which ultimately allows the formation of continuous, free-standing TpPa-COF films [43]. Guo et al. prepared a composite membrane (NCOFM) based on amino-functionalized COF nanosheets (NCOFN) using a vacuum filtration method. First, NCOFN nanosheets with good dispersibility and adjustable component ratios were synthesized via an oil-water-oil three-phase interfacial reaction strategy. In this system, amine and aldehyde monomers spontaneously diffuse under acidic conditions and undergo an imine condensation reaction at the interface, enabling the directional growth of COF nanosheets. The resulting NCOFN nanosheet dispersion is then deposited onto a polyacrylonitrile (PAN) substrate via vacuum-assisted filtration, forming a continuous, dense, and defect-free COF layer. Due to the high aspect ratio of the nanosheets and the well-defined interlayer electrostatic interactions, the resulting membranes exhibited excellent structural integrity and tunable thickness. Furthermore, free-standing, exfoliatable NCOF membranes were obtained by filtration through a hydrophobic polyethersulfone (PES) membrane(figure 4c). These membranes retained excellent crystallinity and a well-ordered nanochannel structure, while exhibiting excellent thermal and chemical stability [44]. Wang and his team used a vapor-assisted synthesis strategy to prepare two-dimensional covalent organic framework (COF) films with ultrahigh conductivity. This method achieves the growth of uniform and continuous COF films by utilizing solvent vapor to promote the controlled diffusion and interfacial condensation of reactive monomers in a closed system.Using metal-containing fluorinated copper phthalocyanine (CuPcF_{16}) and redox-active 2, 3, 6, 7-tetrahydroxyanthraquinone (THAQ) as building blocks, the two form a CuPc-AQ-COF film with excellent conductivity through nucleophilic substitution reaction. This design utilizes the 18π conjugated system of the metal center to provide an electron channel (figure 4d). At the same time, the strong electron-withdrawing effect of the fluorine atom enhances electron delocalization, and the anthraquinone unit further promotes charge transfer, thereby significantly improving the conductivity performance [45].

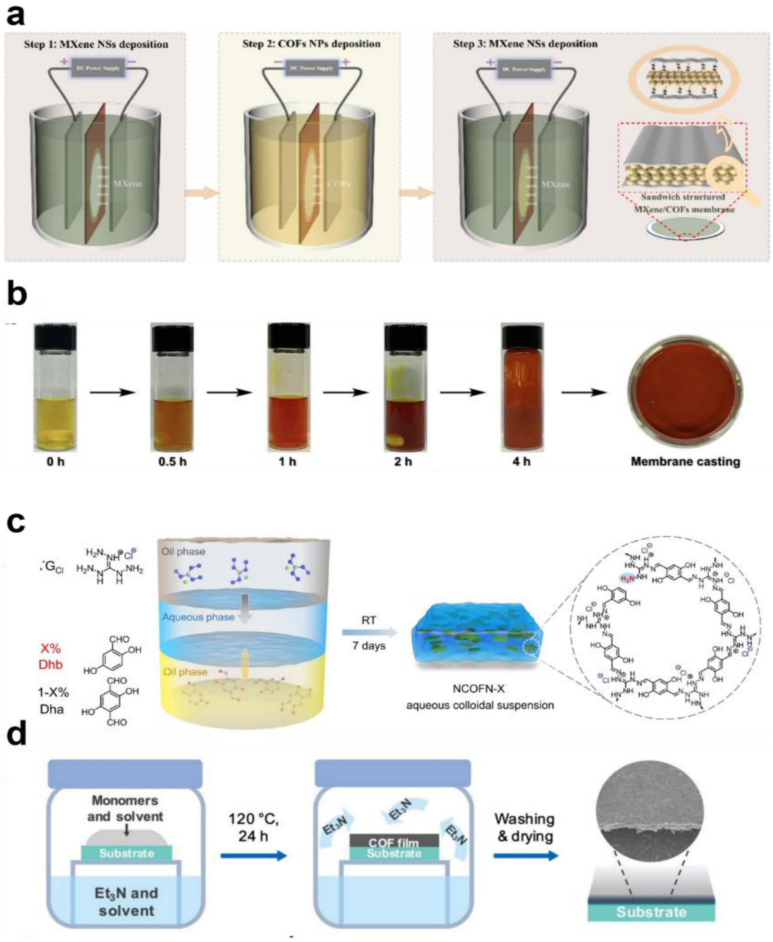

Figure 4. (a) Scheme of a sandwich structured MXene/COFs membranes fabricated by LBL EPD. (b) Photographs showing the transformation of the COF membrane precursor solution during the heating process. (c) Schematic illustration of the synthesis process of NCOFN nanosheets by the oil-water-oil triphase method. (d) Schematic of the preparation of CuPc-AQ-COFs (films) through vapor-assisted synthesis

3. ION SEPARATION APPLICATIONS

3.1. Desalination

Covalent organic framework (COF) membranes have emerged as promising candidates for seawater desalination owing to their tunable pore size, periodic nanochannels, and excellent chemical stability. Furthermore, the construction of COF membranes with sub-nanometer pores and intrinsic charge has been demonstrated as an effective strategy to enhance desalination performance. TbTG COF membranes with high crystallinity were synthesized via liquid–liquid interfacial polymerization followed by vacuum-assisted self-assembly, exhibiting an intrinsic pore size of approximately 0.4 nm (figure 5a). The guanidinium groups embedded in the framework endowed the membrane with stable positive charges, thereby strengthening the synergistic effects of molecular sieving and Donnan exclusion. Consequently, the TbTG COF membranes achieved high rejection rates for various salts, including Na_2SO_4 (98.7%), $MgSO_4$ (99.1%), $MgCl_2$ (99.5%), and NaCl (93.3%), and maintained excellent separation performance over a wide range of salt concentrations and pH values, highlighting their strong potential for seawater desalination applications [46]. Wang et al. fabricated an ultrathin COF membrane with a thickness of only 85 nm via an electrochemical interfacial polymerization strategy, in which the synergistic self-repairing and self-inhibiting effects enabled the formation of highly compact selective layers. The resulting TpPa@PAN-4.0 membrane exhibited salt rejection higher than 99.7% in pure water as well as in 7.5 wt% and 15 wt% NaCl solutions, and achieved a high permeation flux of 92 kg m^{-2} h^{-1} at 80 °C in 7.5 wt% NaCl solution(figure 5b), demonstrating excellent potential for high-salinity desalination applications [47]. Beyond pressure-driven desalination, COF membranes have also shown great promise in membrane distillation (MD) processes. A fluorinated COF (TGTf) membrane with intrinsic hydrophobicity and superior anti-wetting properties was reported to maintain a high water flux of 195 L m^{-2} h^{-1} and a salt rejection of 99.80% during continuous operation for 168 h at 65 °C and 12 kPa(figure 5c), representing one of the longest operational durations for COF-based membranes in MD systems [48]. Both experimental and simulation results revealed that the incorporation of fluorine atoms not only enhanced the hydrophobicity of the membrane but also generated an electrostatic barrier within the fluorinated nanochannels, effectively suppressing ion penetration and improving resistance to fouling and scaling. However, conventional COF membranes often suffer from relatively low crystallinity and oversized pores, which limit their ion sieving capability. To address this issue, Ren and co-workers developed a rotaxane-mediated interfacial polymerization strategy by introducing pseudorotaxane linkers, thereby constructing highly crystalline rotaxane-based COF (RCOF) membranes with sub-nanometer channels. These membranes delivered a remarkable water flux of 180.2 kg m^{-2} h^{-1} together with an NaCl rejection of 99.9%(figure 5d), while also exhibiting long-term operational stability and antifouling performance [49].

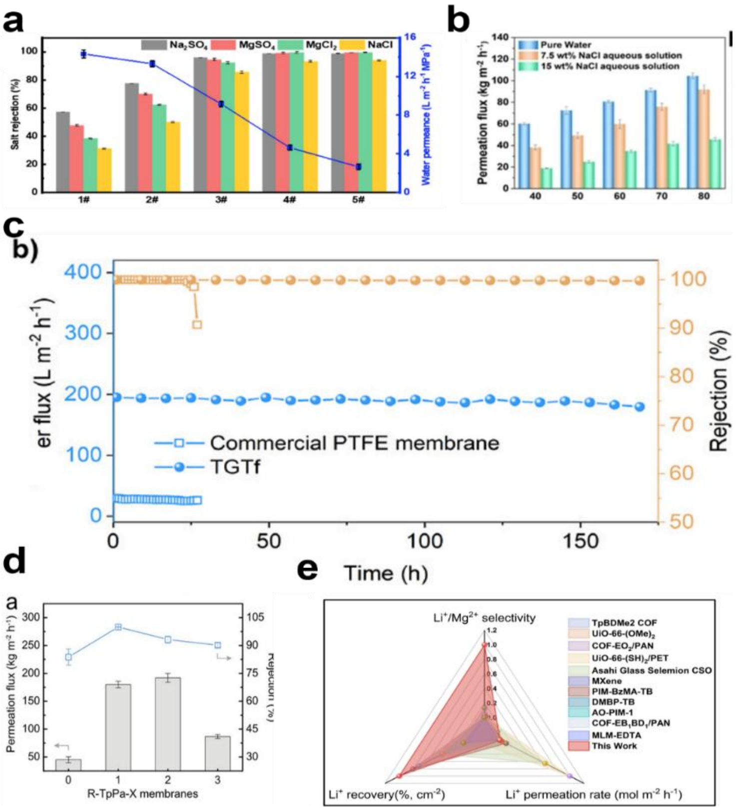

Figure 5. (a) Salt rejection of TbTG COFMs with different thickness. (b) Temperature-dependent permeation flux of the TpPa@PAN-4.0 membrane in different salinity. (c) Long-term MD performance of the TGTf membrane and commercial PTFE membrane. (d) Desalination performance of RCOF membranes with different rotaxane content (salinity, 3.5 wt.% NaCl; operation temperature, 80 °C). (e) Performance comparison of TAT-TP-P with other high performance membrane materials. Li^{+}/Mg^{2+} selectivity, Li^{+} permeation rate, and lithium recovery per unit membrane area were selected as metrics to more comprehensively compare the lithium extraction performance of the membranes

3.2. Lithium Extraction from Salt Lake Brines

Efficient and selective lithium recovery from salt lake brines is essential to satisfy the rapidly increasing global demand for lithium resources. Nevertheless, the development of ion-transport membranes that simultaneously deliver high selectivity and fast ion flux remains challenging because of the similar physicochemical properties of competing ions, particularly Li^+ and Mg^{2+}. Recent studies have demonstrated that covalent organic framework (COF) membranes with well-defined nanochannels and tunable chemical functionalities provide new opportunities for addressing this long-standing problem.

A triazine-based COF membrane functionalized with sulfonate groups (TAT–TP–P) was recently reported for achieving rapid Li^+ transport with exceptionally high Li^+/Mg^{2+} separation selectivity. This separation mechanism originates from the cooperative effects of pore confinement and chemical recognition. Specifically, precise regulation of the sub-nanometer pore size exposes the effective charge of Mg^{2+} while preserving the hydration shell of Li^+, thereby amplifying their charge disparity. Meanwhile, triazine units with strong dipole–dipole interactions, together with sulfonate groups, form an energetic trapping environment for Mg^{2+}, enabling highly selective discrimination between Li^+ and Mg^{2+} within the nanochannels. In addition, the relatively hydrophilic pore environment stabilizes hydrated Li^+ and weakens its interaction with sulfonate moieties, allowing Li^+ to migrate rapidly through a hopping transport pathway along the sulfonate side chains. When evaluated in an electrodialysis system, the membrane exhibited a Li^+ transport rate of 0.56 ± 0.03 mol m^{-2} h^{-1} (figure 5e), while Mg^{2+} remained below the instrumental detection limit, indicating outstanding lithium extraction performance [50].

Nanofiltration (NF) membranes have also emerged as promising candidates for lithium recovery from brines with low Li^+/Mg^{2+} ratios, where precise control of pore structure and surface charge is particularly important. A dual-functional COF-assisted NF membrane was fabricated by assembling quaternary ammonium COF (QACOF) nanosheets onto a polyacrylonitrile (PAN) support as an interlayer. The presence of the QACOF interlayer enriched amine monomers and regulated their diffusion during interfacial polymerization, resulting in a polyamide selective layer with a narrower pore size distribution and enhanced size-sieving capability. Concurrently, the negatively charged membrane surface and the positively charged QACOF interlayer cooperatively promoted Li^+ and Cl^- transport through electrostatic interactions. Consequently, the resulting NF membrane achieved a high water permeance of 25.73 L m^{-2} h^{-1} bar^{-1} and a remarkable Li^+/Mg^{2+} separation factor of 82.58 at an extremely low Li^+/Mg^{2+} mass ratio of 1:100. Furthermore, the membrane maintained rejection rates exceeding 99.0% for divalent salts (Na_2SO_4, $MgSO_4$, and Li_2SO_4) and preserved approximately 95.0% rejection of $MgCl_2$ at a high salt concentration of 6000 ppm, confirming the dominant role of size sieving in Mg^{2+} exclusion [51].

Beyond chemical functionality, regulating the orientation and stacking configuration of COF membranes has been shown to be an effective strategy for enhancing ion selectivity. Self-supporting COF membranes with vertically aligned pore channels were prepared via solvent modulation using identical building blocks. Compared with membranes possessing randomly oriented pores, vertically aligned nanochannels significantly improved ion transport efficiency and selectivity. These membranes exhibited preferential permeation of Li^+ over Na^+ and K^+, achieving Li^+/K^+ and Li^+/Na^+ selectivity ratios of 38.7 and 7.2, respectively. This work highlights the importance of structural alignment and layer stacking in tailoring COF membranes for lithium ion separation. In addition, incorporating functionalized COF nanosheets into microporous polyamide membranes provides an effective route to overcome the traditional permeability–selectivity trade-off in Li^+/Mg^{2+} separation. A cationic COF (TpTGCl) was embedded within the polyamide matrix to construct continuous ion-transport pathways, which not only reduced the internal density of the membrane but also strengthened its positive charge characteristics. The resulting composite membrane delivered a water permeance of 19.6 L m^{-2} h^{-1} bar^{-1} and achieved a Li^+/Mg^{2+} separation factor of up to 21.3 under a high Mg^{2+}/Li^{+} ratio of 30. This strategy demonstrates that charged and nanoporous two-dimensional COF frameworks can serve as effective transport enhancers in microporous membranes, enabling both fast and precise lithium ion separation for practical brine extraction applications [52].

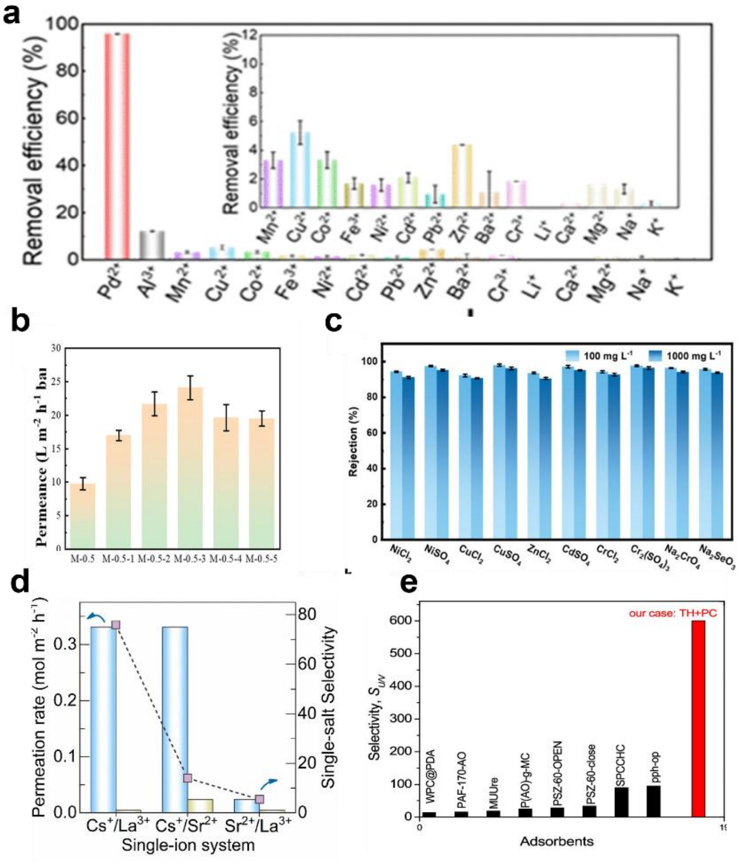

Figure 6. (a) The selectivity of TpPz membranes towards metal ions. (b) Water permeance probability density function curves. (c) Salt rejection of heterocharged membrane at different concentrations. (d) The insert shows the cutoff line. Permeation rates and selectivities of c single-ion. (e) Comparison of U/V selectivity between established adsorbents and our proposed self-assembly approach with TH (5.8mg) and PC (4.0mg)

3.3. Drinking Water Separation

The development of multifunctional COF-based membranes has opened new possibilities for the efficient removal of trace contaminants from drinking water. A dual-functional system was constructed by integrating in situ recovered palladium nanoclusters into a covalent organic framework (COF) membrane. Strong interactions between Pd species and the pyrazine nitrogen atoms in the COF framework promoted the formation of a Pd layer within an ultrathin COF nanofilm with a thickness of approximately 40 nm. To simulate practical electroplating wastewater conditions, the Pd concentration used in the experiments closely matched that encountered in real industrial processes. As shown in Figure 6a, the TpPz membrane exhibited the highest removal efficiency for Pd^{2+} (>95.0%) among 17 competing metal ions, demonstrating excellent selectivity toward trace Pd^{2+}. Moreover, the TpPz membrane achieved its maximum Pd^{2+} adsorption capacity after 24 h of reaction, highlighting its strong affinity and stability for noble metal capture [53].

In another study, a hydrophilic COF monomer, 1, 3, 5-tris (4-aminophenyl) benzene, was introduced into the aqueous phase during interfacial polymerization to enhance organic–inorganic compatibility within thin-film nanocomposite membranes. The incorporation of COF components into the selective layer effectively loosened the membrane structure, leading to an increased water permeance of 24.6 $L\ m^{-2}\ h^{-1}\ bar^{-1}$. Ion rejection in membrane separation processes is governed by a combination of ion valence, hydration radius, and charge density. Owing to the negatively charged membrane surface, cations such as Mg^{2+} and Na^+ are electrostatically attracted, whereas anions including SO_4^{2-} and Cl^- are repelled. Because divalent Mg^{2+} carries a higher charge density than monovalent Na^+, its electrostatic interaction with the membrane is stronger (Figure 6b). To maintain electroneutrality, associated counter-ions are also excluded, resulting in a higher rejection of NaCl compared with $MgCl_2$ under identical operating conditions [54].

3.4. Heavy Metal Separation

Transition-metal-based homogeneous photocatalysts have created significant opportunities for organic synthesis. However, the most widely used ruthenium (II) and iridium (III) polypyridyl complexes are among the rarest and most expensive metal catalysts. Moreover, immobilizing these valuable photocatalysts for recycling remains challenging, as their opacity may impede light transmission during photochemical reactions. Although conventional polymer membranes have been explored for the recovery of homogeneous catalysts, their practical application is limited by difficulties in precisely tuning pore structures and maintaining stability in polar organic solvents. In this context, covalent organic framework (COF) membranes have emerged as promising alternatives for catalyst recovery. A series of COF membranes with tunable pore sizes and excellent solvent resistance were developed for efficient photocatalyst separation. Ruthenium- and iridium-based photocatalysts were successfully recycled for up to ten consecutive catalytic cycles in various photochemical reactions while maintaining high catalytic activity, recovery efficiency, and permeance. Gram-scale recovery of photocatalysts was achieved (Figure 6c). In addition, cascade separation of iridium photocatalysts and purification of small-molecule products using COF membranes with different pore sizes were demonstrated. These results highlight the considerable potential of COF membrane technology to transform current practices in pharmaceutical and fine chemical synthesis [55].

Although COF membranes have been widely recognized as promising platforms for membrane separation, achieving rapid and highly selective ion transport remains challenging. A novel COF membrane featuring vertically aligned nanorod channels was proposed for efficient separation of rare metal ions. A quaternary ammonium–functionalized monomer was rationally designed and polymerized on porous substrates via interfacial synthesis, forming an asymmetric COF layer. The upper region consisted of vertically aligned nanorods, which increased the effective contact area for water and monovalent ions, while the lower region formed an ultrathin dense layer responsible for high permeability and selectivity. The resulting membrane exhibited exceptional separation performance, with a Cs^+ permeation rate of 0.33 mol m^{-2} h^{-1}, approaching that of the porous substrate, and Cs^+/La^{3+} selectivity values as high as 75.9 in single-ion systems and 69.8 in binary systems (Figure 6d). These findings demonstrate the significant potential of structurally engineered COF membranes for rare metal ion separation [56].

3.5. Radionuclide Separation

The strategic integration of radionuclide-specific ligands into the crystalline channels of COF membranes has opened a high-efficiency pathway for the remediation of nuclear wastewater and seawater uranium extraction. Unlike conventional polymer membranes, the rigid, periodic pores of COFs provide a stable platform for the precise arrangement of chelating sites, such as amidoxime, phosphonate, and nitrogen-rich heterocyclic groups. For instance, an amidoxime-functionalized COF (AO-COF) nanofiltration membrane was recently developed for the selective recovery of uranium (U(VI)) from spiked seawater. By regulating the density of AO groups within the COF skeleton, the membrane achieved an exceptional uranium adsorption capacity of 16.91 mg g^{-1} in natural seawater, with a record-high selectivity for uranium over vanadium (Figure 6e). The ordered 1D nanochannels of the AO-COF membrane facilitate the "size-selective" transport of uranyl ions (UO_2^{2+}), while the strong coordination between the AO groups and theU(VI) center prevents the co-permeation of high-concentration competing ions like Mg^{2+} and Ca Mg^{2+} [57].

In addition to cationic radionuclides, the separation of anionic species such as pertechnetate ($99TcO_4^-$) presents a significant challenge due to its high mobility and long half-life. To address this, cationic COF-based thin-film nanocomposite (TFN) membranes were engineered by incorporating triaminoguanidinium-based COF nanosheets into a polyamide selective layer. The inherent positive charge of the guanidinium units (surface potential≈+38.5 mV) significantly enhanced the Donnan exclusion effect, repelling common cations while selectively attracting TcO_4^- through anion exchange. Experimental results revealed that the optimized cationic COF membrane exhibited a TcO_4^- rejection rate exceeding 98.2% even in the presence of 2.0M $NaNO_3$ background salt, maintaining a high water permeance of 28.4 L m^{-2} h^{-1} bar^{-1}. Furthermore, the covalent linkages within the COF framework ensured the membrane's structural integrity under extreme radiation doses (up to 200 kGy), demonstrating far superior radiation stability compared to commercial ion-exchange membranes [58].

4. CONCLUSIONS AND PERSPECTIVE

This review has synthesized the rapid advancements in Covalent Organic Framework (COF) membranes, illustrating their potential to redefine the standards of ion separation technology. By analyzing the evolution of preparation techniques and their practical utility, several key insights emerge:

(1) Vancements in Membrane Engineering: The field has moved beyond simple synthesis to sophisticated fabrication strategies. Techniques such as interfacial polymerization and solvent-modulated assembly now allow for the production of continuous, defect-free, and ultrathin COF layers on scalable substrates. These methods enable precise control over pore dimensions and chemical environments at the atomic level, which is fundamental to their separation efficiency.

(2) Plication-Driven Performance: COF membranes have demonstrated capabilities that frequently outperform commercial benchmarks. In desalination, they maintain high water permeance without sacrificing salt rejection. More notably, in the strategic area of lithium extraction, functionalized COF channels successfully break the trade-off between flux and selectivity, offering a viable solution for sustainable resource recovery from salt lakes. Similarly, their affinity for specific contaminants proves invaluable for the remediation of complex nuclear and industrial wastewaters.

Future Outlook: While the structural diversity of COFs offers limitless possibilities, the path to commercial viability requires a shift in focus. Future research must prioritize the development of cost-effective, large-area manufacturing processes and the enhancement of membrane robustness against harsh operating conditions, such as extreme pH and high salinity. Bridging the gap between precise molecular design and macroscopic durability will be the defining challenge in establishing COF membranes as a cornerstone of next-generation water treatment and resource recovery infrastructure.

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