

Biological nanofiltration membrane: Electrostatic steering for breaking ion selectivity-permeability trade-off in $\text{Mg}^{2+}/\text{Li}^{+}$ separation

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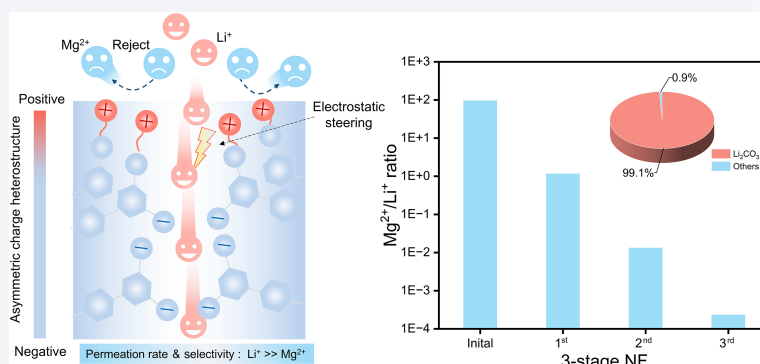
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ABSTRACT: Lithium (Li) is indispensable for low-carbon energy systems, yet its large-scale extraction from high-Mg salt-lake brines is fundamentally constrained by the insufficient selectivity of existing separation technologies. Traditional nanofiltration (NF) membranes suffer from an intrinsic selectivity-permeability trade-off, arising from the strong coupling between electrostatic interactions and ion transport within uniformly charged nanopores. Here, we present a biological ion channel-inspired nanofiltration (BICNF) membrane with an asymmetric charge heterostructure, fabricated by grafting positively charged quaternary ammonium salts onto a negatively charged polyamide nanopore. The as-prepared membrane effectively breaks the ion selectivity-permeability trade-off in $\text{Mg}^{2+}/\text{Li}^{+}$ separation. Unlike uniformly charged NF membranes, the asymmetric charge heterostructure of the BICNF membrane spatially decouples ion pre-screening from transmembrane transport. Moreover, molecular dynamics simulations reveal that the bioinspired charge configuration establishes a directionally biased electrostatic steering effect, which actively guides Li^{+} ions through the nanopores while effectively repelling Mg^{2+} . As a result, the BICNF membrane achieves a high $\text{Mg}^{2+}/\text{Li}^{+}$ selectivity (~ 75) with a high Li^{+} permeability rate of $\sim 0.65 \text{ mol} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$. Using a simulated salt-lake brine, the BICNF membrane enables crystallization of Li_2CO_3 with a high purity of 99.1%. Thus, this work provides a bioinspired strategy to advance highly efficient ion separation for lithium extraction.



KEYWORDS: ion channel, nanofiltration, heterostructure, electrostatic steering, $\text{Mg}^{2+}/\text{Li}^{+}$ separation

1 Introduction

As a critical strategic resource for green energy storage, lithium (Li) plays a fundamental role in modern battery technologies that support the transition toward low-carbon energy systems [1, 2]. Driven by the rapid growth of electric vehicles and large-scale renewable energy deployment, global Li demand has increased sharply in recent years and is projected to rise nearly sevenfold by

2040 [3]. Currently, the majority of the world's identified Li resources are concentrated in salt-lake brines; however, their sustainable exploitation is severely impeded by the absence of efficient $\text{Mg}^{2+}/\text{Li}^{+}$ separation technologies. In high-Mg salt-lake brine, conventional extraction processes fail to effectively discriminate Li^{+} from competing Mg^{2+} ions [4, 5]. This poor selectivity not only compromises the purity and grade of recovered Li but also necessitates additional purification steps, thereby increasing process complexity, cost, and environmental burden. Therefore, overcoming this $\text{Mg}^{2+}/\text{Li}^{+}$ separation bottleneck is crucial for securing high-quality Li supply, and supporting the development of the clean energy field [6, 7].

Nanofiltration (NF) membranes have achieved substantial

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improvements in $\text{Mg}^{2+}/\text{Li}^+$ selectivity; however, their intrinsically negative pore charge creates a fundamental selectivity–permeability trade-off, thereby constraining practical Li^+ extraction [8–11]. To address these limitations, various novel NF membranes based on advanced material engineering, such as metal organic framework (MOF) [12–14], covalent organic framework (COF) [15–17], polyamide (PA) [10, 11, 18, 19], and two-dimensional (2D) composites (e.g., graphene oxide [20] or MXenes [21, 22]), have been developed to enhance ion selectivity performance. Interestingly, these NF membranes have demonstrated notable

breakthroughs in ion selectivity or permeability by leveraging tailored pore chemistry, ordered nanochannels, and tunable electrostatic interactions. Nevertheless, achieving a balanced optimization of Li^+ selectivity and permeability, which is an essential requirement for high-yield Li production, remains a significant challenge for current NF membrane technologies.

Biological ion channels are distinguished by their highly specialized pore architectures and intrinsically asymmetric charge heterostructures, which enable precise regulation of ion selectivity and permeability. In biological K^+ channels (Fig. 1(a)), a positively

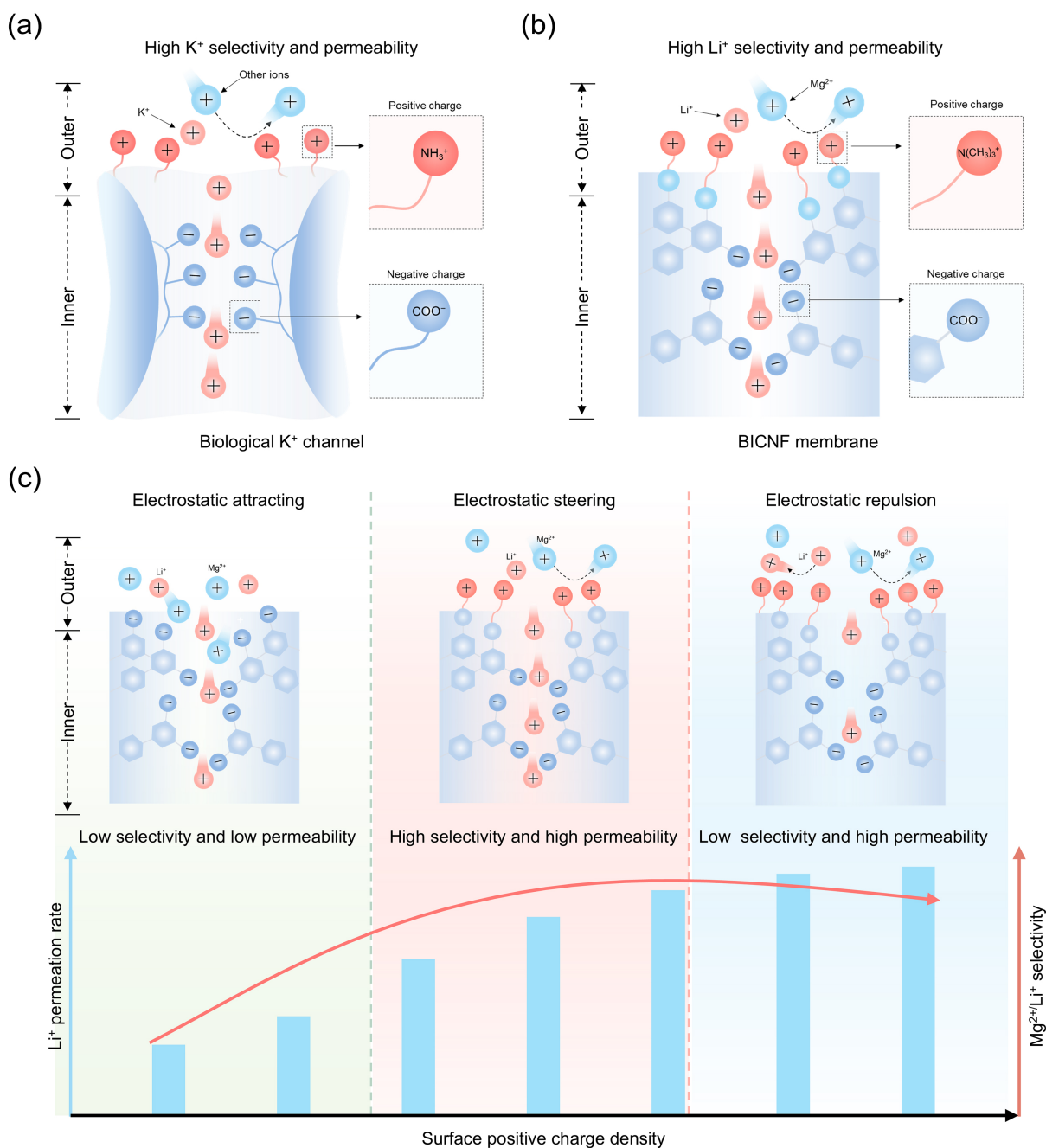


Figure 1 Bioinspired design for enhanced $\text{Mg}^{2+}/\text{Li}^+$ separation. Schematic of (a) biological K^+ ion channel and (b) BICNF membrane. (c) Influence of surface positive charge density of the BICNF membrane on $\text{Mg}^{2+}/\text{Li}^+$ separation by regulating electrostatic interactions. For intermediate charge density (pink background), electrostatic steering preferentially guides Li^+ into the nanochannels, enabling high selectivity and high permeability simultaneously. In contrast, weak electrostatic attraction from low charge density (green background) results in low selectivity and low permeability, while strong electrostatic repulsion from excessive charge density (blue background) impedes ion transport and leads to low selectivity and high permeability.

charged outer vestibule enriched with Lys⁺/Arg⁺ residues is spatially coupled with a negatively charged inner selectivity filter composed of Glu⁻/Asp⁻ residues, establishing a directional electrostatic steering along the nanochannel pathway. Notably, the positively charged outer surface acts as a critical regulatory module, governing ion pre-screening and directional guidance during transmembrane transport [23–26]. This asymmetric charge heterostructure gives rise to a finely orchestrated electrostatic steering effect, whereby ion migration is no longer governed by passive diffusion alone but is actively guided along the permeation pathway by spatially organized electrostatic fields [27–29]. As a result, K⁺ channels simultaneously achieve extraordinary ion selectivity (exceeding 10³) and ultrafast permeation rates (approaching the theoretical diffusion limit) [30–33]. Therefore, this biological charge-engineering strategy may provide a promising avenue for breaking the inherent ion selectivity–permeability trade-off in Mg²⁺/Li⁺ separation.

In this work, we present a biological ion channel-inspired nanofiltration (BICNF) membrane featuring a positive outer surface and negative inner pore, overcoming the inherent trade-off between Li⁺ selectivity and permeability in Mg²⁺/Li⁺ separation (Fig. 1(b)). This asymmetric charge heterostructure establishes an interface-derived, directionally biased electrostatic steering effect that pre-ori-ents Li⁺, accelerates their transmembrane permeation, and simultaneously rejects Mg²⁺ by modulating the surface positive charge density (Fig. 1(c)). Molecular dynamics (MD) simulations further corroborate the presence of this electrostatic steering effect, revealing a preferential Li⁺ transport pathway and suppressed Mg²⁺ migration arising from the asymmetric electrostatic potential within the nanopores. By leveraging this spatially electrostatic regulation strategy, the BICNF membrane overcomes the intrinsic trade-off between Li⁺ selectivity and permeability, achieving a high selectivity of ~ 75 with rapid Li⁺ permeation rate (~ 0.65 mol·m⁻²·h⁻¹). Furthermore, the Li⁺ extraction performance is validated in a simulated brine separation process, producing Li₂CO₃ crystallization with a high purity of 99.1%. Thus, these findings collectively highlight the great practical potential of the BICNF membrane for Li extraction.

2 Experimental

2.1 Preparation of BICNF membrane

The aqueous phase was prepared by dissolving 0.3 g piperazine (PIP) and 0.5 mL triethylamine in 100 mL deionized water, and adjusting the pH to 9.8 with 1 M H₃PO₄. The organic phase was prepared by dissolving 0.2 g trimesoyl chloride (TMC) in n-hexane. Polyether sulfone (PES) membranes were pre-soaked in water for 12 h, and immersed in the aqueous solution for 5 min. After that, the excess solution was removed with a rubber roller. Interfacial polymerization was initiated by contacting the membranes with the organic phase for 1 min at room temperature, producing a thin cross-linked polyamide layer. After that, the membranes were rinsed with n-hexane and air-dried. For the *in situ* sequential grafting reaction, polyethyleneimine (PEI) (0.2 g) and epoxypropyl trimethyl ammonium chloride (EPTAC) (0.15 g) were dissolved in 100 mL deionized water containing 0.02 g NaOH. The mixture was stirred until complete dissolution. Then, the membranes were immersed in this solution at 60 °C for 10 min. The modified membranes were thoroughly rinsed and stored in deionized water

prior to characterization to avoid structural deformation, and designated as BICNF membranes.

To systematically elucidate the role of charge regulation in ion transport, a series of BICNF membranes were fabricated by varying the EPTAC concentrations while keeping all other synthesis parameters constant. Specifically, EPTAC concentrations of 0, 0.5, 1.0, 1.5, 2.0, and 2.5 g·L⁻¹ were employed, and the resulting membranes were denoted as BICNF0, BICNF1, BICNF2, BICNF3, BICNF4, and BICNF5, respectively. By jointly considering permeability and ion selectivity, the BICNF3 membrane exhibits optimal performance and is therefore selected for subsequent comparison with the polyamide nanofiltration (PNF) membrane.

3 Results and discussion

3.1 Design of the BICNF membrane

Figure 2(a) shows that the BICNF membrane is fabricated via a bioinspired surface charge-engineering strategy, in which positively charged EPTAC-PEI was grafted onto an interfacial polymerized PA layer by an epoxy-amine nucleophilic ring-opening reaction [34]. To verify the successful formation of this chemically and electrostatically asymmetric architecture of the BICNF membrane, both Fourier transform infrared (FTIR) spectroscopic and X-ray photoelectron spectroscopy (XPS) characterizations were subsequently performed. As evidenced by the C=O–N– stretching vibration band at 1617 and 1650 cm⁻¹ in Fig. S1 in the Electronic Supplementary Material (ESM), the PA layer was successfully formed on the PES membrane [33, 35]. After sequential modification with EPTAC-PEI in the BICNF membrane, a distinct peak at 1034 cm⁻¹ and a new band at 3273 cm⁻¹ were ascribed to the quaternary ammonium group [8] and amino groups [9], respectively. This finding confirms the occurrence of a reaction between amino and epoxy groups, which consequently facilitated the effective formation of positively charged functionalities on the membrane surface. Subsequently, the N 1s spectrum of the BICNF membrane can be deconvoluted into three peaks at 399.3, 400.4, and 401.6 eV, assigned to –C–N(H)–, –C=O–N–, and –C–N(CH₃)₃⁺, respectively (Fig. 2(b)) [36]. Meanwhile, the characteristic peak corresponding to –C–N(CH₃)₃⁺ is also observed in the C 1s spectrum in Fig. 2(c), confirming the successful surface modification of the membrane. In contrast, the high-resolution N 1s and C 1s spectra of the pristine PA layer (Fig. S2 in the ESM) show no detectable signals associated with quaternary ammonium group, confirming that these features originate exclusively from the EPTAC-PEI grafting process in the BICNF membrane. Thus, the characteristic peak of quaternary ammonium salt is the key evidence for the successful grafting of EPTAC-PEI onto the surface of the BICNF membrane. Furthermore, increasing the EPTAC concentration in the BICNF membrane modification progressively enhances the content of the quaternization functional groups on the membrane surface from 0 to 39.8% (Fig. S3 in the ESM). The BICNF membrane was further analyzed by XPS depth etching to probe the depth-dependent distribution of charge-bearing functional groups across the membrane (Fig. 2(d)). Subsequent depth-profiling XPS analysis reveals that quaternary ammonium groups are highly enriched at the membrane surface (27.3% of the surface composition), but its content decreases sharply within the first ~ 20 nm from the surface. At greater depths (20–40 nm), the quaternary ammonium content is substantially lower than that at the outer surface. This gradient distribution forms a surface-

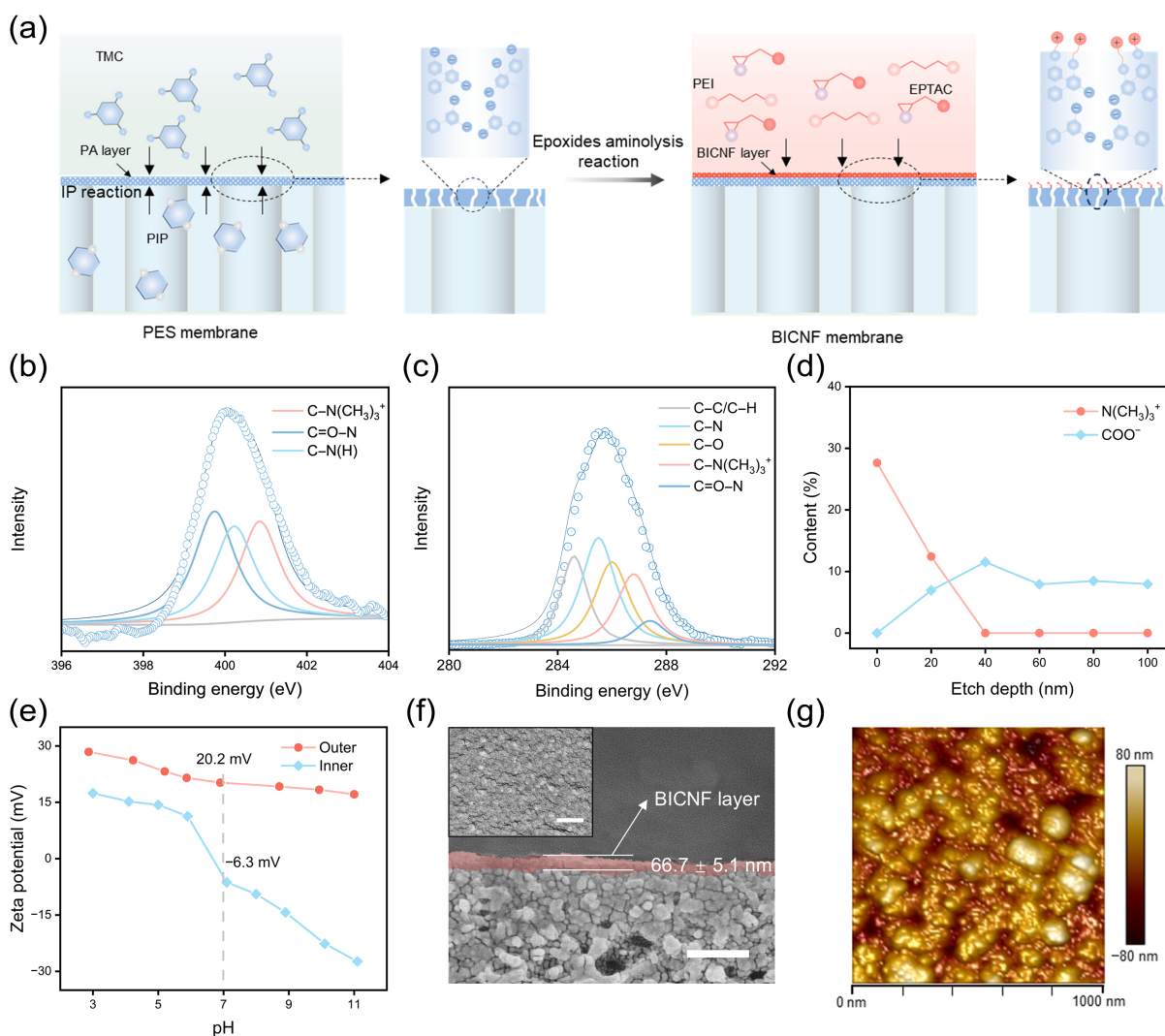


Figure 2 Characterization of the BICNF membrane. (a) Schematic of the fabrication process of the BICNF membrane. (b) N 1s spectra and (c) C 1s spectra of the BICNF membrane. (d) Depth-dependent distribution of $\text{N(CH}_3)_3^+$ in N 1s spectra and COO^- in C 1s spectra of the BICNF membrane. (e) Zeta potential of the outer surface and inner pore of the BICNF membrane. (f) SEM images of the cross-section and surface (inset) of the BICNF membrane (scale bar is 200 nm, and scale bar in the inset is 1 μm). The stained areas represent the BICNF (red) layer. (g) AFM image of the BICNF membrane.

dominant positively charged layer, which arises directly from the kinetically favorable crosslinking reaction between the epoxy groups of EPTAC and the amine groups of PEI. In contrast, the slower reaction with carboxyl groups restricts the incorporation of quaternary ammonium groups within the nanopore [37].

Next, zeta potential measurement was further employed to corroborate the surface charge distribution inferred from XPS analysis. Depth-dependent analysis reveals an asymmetric charge heterostructure in the BICNF membrane, characterized by a positively charged outer surface and negatively charged inner nanopores (Fig. 2(e)). This structure arises from the surface enrichment of quaternary ammonium groups introduced by EPTAC modification, whereas carboxyl groups remain predominantly confined within the subsurface polyamide nanopores. Such spatially separated charge domains establish a directional electrostatic landscape across the membrane thickness, which is expected to play a critical role in regulating ion transport under nanoconfinement. Notably, the surface positive charge progressively increases with increasing EPTAC modification concentration compared with the PNF membrane, confirming the

effectiveness of charge regulation (Fig. S4 in the ESM). After PEI deposition, a high density of protonated amine groups under neutral pH was introduced to the surface of the BICNF0 membrane. These protonated amine groups dominate the interfacial positive electrostatic potential (+7.54 mV for BICNF0), effectively offsetting the contribution of underlying carboxyl groups and thereby inducing charge reversal [16, 38]. Subsequent EPTAC modification further increases the surface positive charge density, thereby establishing a well-defined asymmetric charge heterostructure across the membrane. Based on these results, the combined XPS and zeta potential analyses provide mutually reinforcing evidence for a well-defined asymmetric charge architecture within the membrane. The depth-resolved XPS (Fig. 2(e)) reveals a clear gradient of fixed charge-related groups, with surface enrichment of positively charged amine/quaternary ammonium species and increasing carboxyl groups toward the interior. This chemical distribution directly governs the local charge density, while the zeta potential reflects the resultant macroscopic electrokinetic behavior. The consistency between surface chemical composition and positive zeta potential establishes a direct

correlation between charge distribution and electrostatic characteristics, further verifying the formation of an asymmetric charge structure.

To elucidate the structural feature of the membranes, scanning electron microscopy (SEM) and atomic force microscopy (AFM) were utilized. Notably, the BICNF membrane exhibits a significantly reduced functional layer thickness of 66.7 ± 5.1 nm with a roughness of 3.2 ± 1.5 nm in Figs. 2(f) and 2(g). In contrast, the PNF membrane exhibits pronounced nodular structure with a functional layer thickness of 90.5 ± 12.4 nm and a roughness of 14.3 ± 3.3 nm in Fig. S5 in the ESM, which originates from the rapid polyamide formation during the interfacial polymerization reaction. As a result, the BICNF membrane exhibits a smoother surface compared to the PNF membrane. This distinction can be attributed to the bioinspired interfacial charge regulation which moderates the local reaction kinetics and suppresses excessive polyamide aggregation, thereby inhibiting the formation of nodular features in the BICNF membrane [37, 38].

Notably, the enhanced network density restricts the lateral growth and swelling of the functional layer during formation, which in turn leads to the development of a thinner selective layer with improved structural integrity and potentially superior separation performance than that of the PNF membrane [39, 40]. Additionally, the BICNF membrane exhibits superhydrophilicity, as evidenced by its rapid decrease in water contact angle (WCA) from ca. 28.5° to $\sim 0^\circ$ within 10 s (Fig. S6 in the ESM). In contrast, the PNF membrane showed an initial WCA of ca. 67.3° , and decreased to ca. 32.3° after 10 s. The superior hydrophilicity of the BICNF membrane is primarily attributed to the presence of quaternary ammonium group, which possess intrinsic hydrophilic characteristics due to their strong electrostatic interactions with water molecules. Consistent with this enhanced hydrophilicity, the BICNF membrane exhibits a substantially higher water flux than the PNF membrane (Fig. S7 in the ESM). Furthermore, the superior water permeability of the BICNF membrane arises from its asymmetric charge architecture, which establishes a directional electrostatic field across the membrane thickness. This charge distribution reduces the interfacial energy barrier for water entry, facilitates electrostatically guided water transport along the thickness direction, and thereby accelerates water migration through the membrane [41–43]. In addition, the synergistic effect between super-hydrophilicity and the ultrathin functional layer endows the membrane with improved water permeability. The enhanced hydrophilicity lowers the interfacial resistance to water adsorption, while the shortened mass transport pathway markedly accelerates the transmembrane migration of water molecules. Collectively, these results confirm that the BICNF membrane features a structurally and electrostatically asymmetric configuration that jointly optimizes interfacial wetting and transport kinetics.

3.2 Highly efficient ion separation

The separation performance of the BICNF membrane was systematically evaluated from ion selectivity and permeation behaviors. Firstly, the pore structure of the membrane was evaluated through polyethylene glycol (PEG) rejection measurements [44–46]. Figure 3(a) demonstrates that the molecular weight cutoff (MWCO) of the BICNF membranes (328 Da) was lower than that of the PNF membrane (574 Da). Based on the MWCO characterization, the pore size distribution of

the membranes was fitted using the rejection curve under the assumption of a log-normal distribution in Fig. 3(b) [11]. The pore size distribution analysis reveals that the average pore diameter of the BICNF membrane was smaller than that of the PNF membrane. Significantly, the pore size of the BICNF membrane falls within the range comparable to hydrated Li^+ radius. This nanoconfinement effect combined with the asymmetric surface charge distribution, contributes to the enhanced ion selectivity of the BICNF membrane. Furthermore, the pore size analysis provides direct structural evidence for the formation of a denser functional layer in the BICNF membrane than PNF membrane.

Importantly, the BICNF membrane exhibits a more pronounced valence-dependent selectivity than the PNF membrane, with low rejection of monovalent cations (e.g., K^+ 22.1%, Na^+ 24.6%, and Li^+ 24.7%) and sharply increased rejection of multivalent ions (e.g., Ca^{2+} 98.5%, Mg^{2+} 98.7%, and Al^{3+} 99.7%) (Fig. 3(c)). Owing to their elevated dehydration energy barriers and enhanced electrostatic ion-membrane interactions, the transmembrane permeation of multivalent ions (Ca^{2+} , Mg^{2+} , and Al^{3+}) is markedly suppressed. These multivalent cations possess large hydrated radii and high charge densities, resulting in strongly bound hydration shells that require substantial energy to partially strip during transport through nano pores. Besides, strong electrostatic interactions with the positively charged surface of the BICNF membrane further increase transport resistance, collectively leading to reduced permeability compared with weakly hydrated monovalent ions. As shown in Fig. 3(d), the rejection behaviors of Li^+ and Mg^{2+} exhibit markedly different dependencies on membrane surface charge density, highlighting the decisive role of electrostatic steering in achieving efficient $\text{Mg}^{2+}/\text{Li}^+$ separation. Initially, with the gradual increase in surface positive charge density, the rejection of Mg^{2+} is enhanced while that of Li^+ is reduced, which can be reasonably attributed to the asymmetric charge distribution that promotes electrostatic steering and preferential ion transport. In contrast, a moderate increase in surface charge density results in a slight decrease in Li^+ rejection. As the surface charge density further exceeds a critical threshold, the ion transport mechanism transitions from electrostatic steering to dominant electrostatic repulsion, leading to a pronounced reduction in Li^+ permeability [47]. This behavior can be further rationalized by Donnan equilibrium theory, where an excessively high fixed positive charge density at the membrane interface generates a strong Donnan potential, which in turn disrupts the balanced ion partitioning and ultimately hinders Li^+ transport across the membrane [48, 49].

Meanwhile, partially hydrated monovalent ions exhibit lower energy barrier hindrance and achieve more facile permeability. When the feed-side $\text{Mg}^{2+}/\text{Li}^+$ concentration ratio was varied from 1 to 1000, the corresponding $\text{Mg}^{2+}/\text{Li}^+$ separation factor (S) remained remarkably stable at 71–77 (Fig. 3(e)). In order to further elucidate ion permeation behavior, we employed a H-type diffusion cell to measure Li^+ and Mg^{2+} permeation rates across the membrane (Fig. 3(f) and Fig. S8 in the EMS). Under controlled conditions (0.1 M LiCl and 0.1 M MgCl_2), the PNF membrane exhibits a Li^+ permeation rate of $0.18 \text{ mol}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$, while the Mg^{2+} permeation rate is $0.097 \text{ mol}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$, indicating a low $\text{Mg}^{2+}/\text{Li}^+$ selectivity. In detail, the negatively charged PNF membrane establishes an interfacial Donnan potential that energetically disfavors the partitioning of Mg^{2+} [50, 51]. Compared with Li^+ , Mg^{2+} experiences a significantly higher electrostatic energy barrier when approaching and entering the charged nanochannels, resulting in a reduced Mg^{2+}

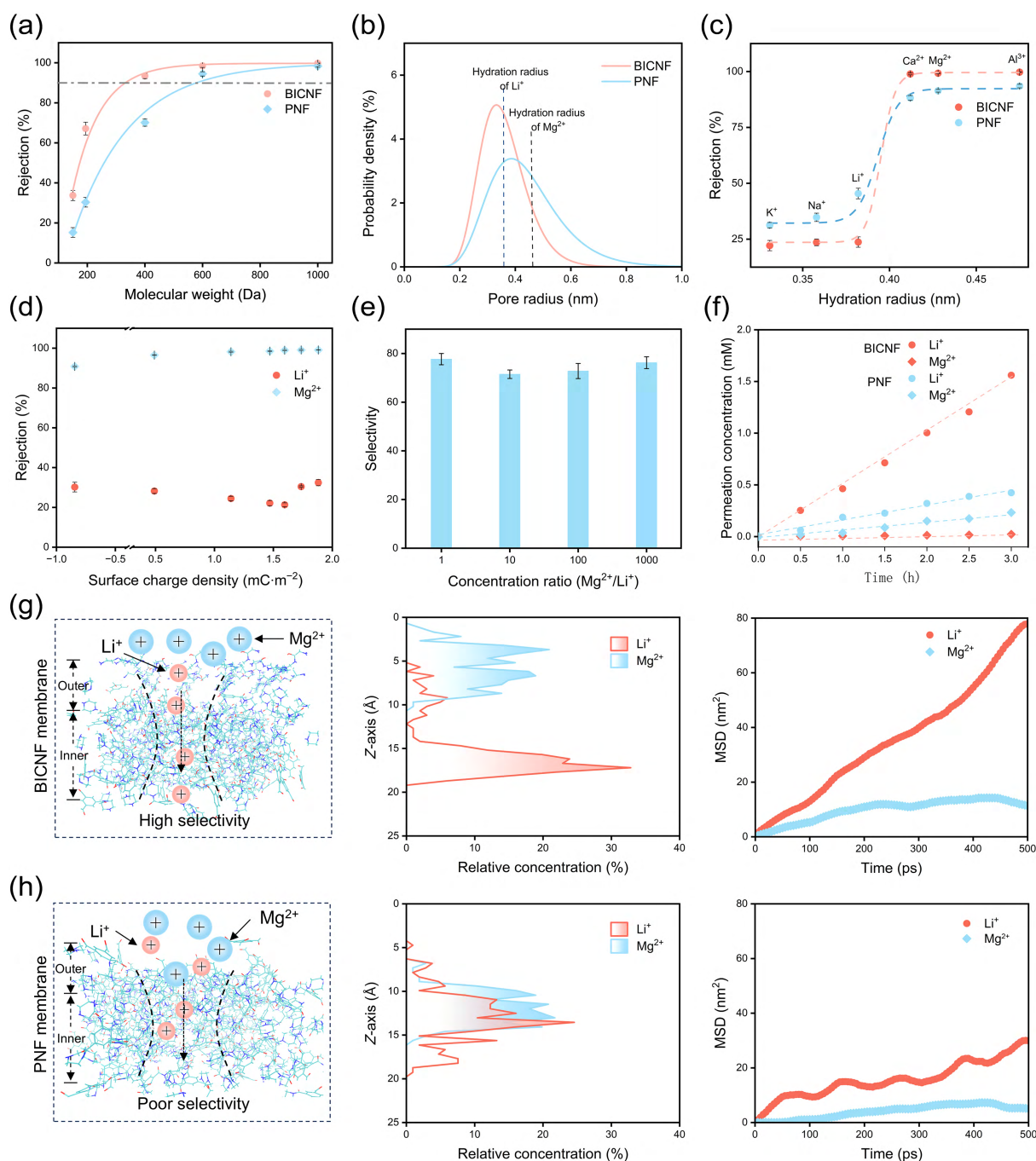


Figure 3 Separation capability of BICNF and PNF membranes. (a) Rejection of polyethylene glycol with different molecular weights and (b) pore size distribution of the BICNF and PNF membranes. (c) Rejection of various ions as a function of hydration radius for BICNF and PNF membranes. (d) Mg^{2+}/Li^{+} rejection of the membranes as a function of surface charge density. (e) Selectivity of the BICNF membrane under different Mg^{2+}/Li^{+} concentration ratios. (f) Mg^{2+}/Li^{+} permeation concentration of the BICNF and PNF membrane. MD simulation of amorphous cells, ion distribution and MSD of (g) the BICNF membrane and (h) the PNF membrane.

concentration within the membrane. In addition, Mg^{2+} possesses a large hydrated radius and high hydration energy, resulting in an effectively enlarged hydrated ionic size; therefore, its transport through the membrane is significantly hindered, leading to a reduced permeation rate [52, 53]. In contrast, the BICNF membrane exhibits markedly enhanced separation performance, delivering a Li^{+} permeation rate of $0.65 \text{ mol} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$ while suppressing the Mg^{2+} flux to as low as $0.0081 \text{ mol} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$. These results conclusively demonstrate that the BICNF membrane exhibits exceptional Mg^{2+}/Li^{+} separation performance, featuring

simultaneously ultrahigh Li^{+} permeability. Overall, the superior Mg^{2+}/Li^{+} selectivity of the BICNF membrane originates from its asymmetric charge architecture. This unique structure synergistically strengthens electrostatic exclusion, ion dehydration resistance, and steric confinement, thereby establishing a highly selective transport pathway that facilitates Li^{+} permeation while efficiently restricting Mg^{2+} passage. The I - V characterization further reveals the critical role of the asymmetric charge configuration in enabling electrostatic steering effect within the BICNF membranes (Fig. S9 in the ESM). Compared with the PNF membrane, the

BICNF membrane exhibits a significantly higher current density with the current increasing from 50.1 to 75.6 μA at an applied potential of 1.0 V. This current-enhancing effect is gradually amplified with increasing EPTAC concentration. The resulting I - V curve not only shows higher current under the same applied bias but also displays an increasingly pronounced asymmetry favoring cation transport in the forward direction, consistent with ion rectification behavior observed in biological K^+ channels. The calculated current rectification ratios further highlight the strengthening of directional ion permeability with increasing modification concentration. Upon incorporation of the bioinspired asymmetric-charge configuration into the BICNF membrane, the emergence of ionic transport asymmetry becomes evident even at the initial stage of surface charge regulation, resulting in a gradual increase in the rectification ratio to approximately 20%. In contrast, the PNF membrane exhibits a nearly unity ratio about 1.0, confirming symmetric conduction expected for a uniformly negatively charged nanopore. Thus, for the BICNF membrane with spatially nonuniform charge distribution, the internal electrostatic field displays distinct responses to oppositely oriented driving forces, thereby enabling direction-dependent ion transport.

Meanwhile, MD simulation was employed to compare ion permeability behaviors in the BICNF membrane and PNF membrane. Such theoretical modeling enables direct visualization of ion distribution, diffusion trajectories, and energy barriers within the nanopores, thereby elucidating the structure-selectivity relationship at the molecular scale. As shown in Fig. 3(g), Mg^{2+} predominantly accumulate near the positively charged outer surface of the BICNF membrane and are effectively rejected due to strong electrostatic repulsion. In contrast, Li^+ preferentially localizes within the negatively charged inner channels, where favorable electrostatic attraction guides their entry and promotes rapid diffusion along the nanopore. Meanwhile, the permeation rate of Li^+ ($0.14 \text{ nm}^2\cdot\text{ps}^{-1}$) is significantly faster than that of Mg^{2+} ($0.03 \text{ nm}^2\cdot\text{ps}^{-1}$) within the membrane. This strong discrimination originates from the asymmetric charge structure of the BICNF membrane, in which a positively charged outer surface prescreens incoming ions and strongly rejects Mg^{2+} via coulombic interactions, while the negatively charged pores preferentially attract and accelerate Li^+ permeability. This distinct spatial distribution highlights a pronounced electrostatic steering effect, whereby ions are selectively guided rather than indiscriminately adsorbed. While molecular dynamics simulations capture ion distribution and electrostatic steering at the nanometer scale, the predicted selectivity reflects nanoscale transport behavior.

In macroscopic filtration experiments, these nanoscale steering effects are collectively amplified across the membrane, resulting in a higher $\text{Mg}^{2+}/\text{Li}^+$ separation performance than that in the simulations. In contrast, the PNF membrane exhibits a fundamentally different ion distribution pattern. As illustrated in Fig. 3(h), both Mg^{2+} and Li^+ tend to accumulate at the membrane surface, driven by strong electrostatic attraction between the cations and the negatively charged polyamide matrix. Thus, the PNF membrane shows a permeation rate of $0.04 \text{ nm}^2\cdot\text{ps}^{-1}$ for Li^+ and $0.03 \text{ nm}^2\cdot\text{ps}^{-1}$ for Mg^{2+} , reflecting their comparable diffusion within the membrane. This nonspecific electrostatic adsorption leads to similar surface concentrations and closely matched permeation rates for Mg^{2+} and Li^+ , indicating the absence of directional ion guidance or selective transport pathways. Collectively, both experimental measurements and theoretical simulations

demonstrate that the BICNF membrane achieves more efficient $\text{Mg}^{2+}/\text{Li}^+$ separation and accelerated Li^+ permeation than the PNF membrane. This performance enhancement arises from an electrostatic steering effect, thereby breaking the conventional trade-off between Li^+ selectivity and permeability.

3.3 Extraction of Li ion in simulated salt-lake brine

To further assess the practical applicability for Li recovery, the Li^+ extraction performance of the BICNF membrane was evaluated in complex ionic environments. Specifically, pH-dependent boric acid separation and multistage Li^+ purification at practical wastewater concentrations were investigated to demonstrate the membrane's highly efficient separation performance. In salt-lake brines, boron typically exists as boric acid or weakly dissociated borate species, which are difficult to remove due to their small molecular size, weak charge, and high mobility in aqueous media [54, 55]. During lithium extraction and concentration, boron can be enriched together with Li^+ , leading to severe contamination in subsequent lithium carbonate or lithium hydroxide products. Then, the rejection rates of all three ions increased gradually with rising pH, with this effect being most prominent for borate species (Fig. 4(a)). In detail, Li^+ rejection increased from $24.8\% \pm 1.2\%$ at pH = 3 to $27.6\% \pm 1.4\%$ at pH = 10, while Mg^{2+} rejection increased from $96.7\% \pm 0.5\%$ to $99.7\% \pm 0.2\%$ over the same range. In the case of boron-containing species, the rejection rate increased sharply above pH = 9, coinciding with the acid dissociation constant ($\text{pK}_a = 9.24$) of boric acid. Below this pK_a value, boron predominantly exists as neutral $\text{B}(\text{OH})_3$, which can pass through the membranes due to its small molecular size ($\sim 0.23 \text{ nm}$) and lack of charge. When the pH values are above the pK_a , deprotonation produces the monovalent anion $\text{B}(\text{OH})_4^-$, and further increases in pH promote the condensation of borate into polyborate species. These multi-charged species possess significantly larger hydrated diameters (up to $\sim 0.66 \text{ nm}$) and stronger electrostatic interactions with the charged membrane surface, leading to a marked increase in rejection, which exceeds 90% at pH > 9. Therefore, the BICNF membrane achieves superior boric acid separation by combining precisely regulated nanopore confinement with asymmetric charge heterostructure, which enhance interfacial interactions and effectively suppress boric acid permeability. In addition, the long-term stability of the BICNF membrane in $\text{Mg}^{2+}/\text{Li}^+$ separation was evaluated over a continuous 7-day separation operation in Fig. 4(b). Throughout the entire testing period, the BICNF membrane maintained nearly constant $\text{Mg}^{2+}/\text{Li}^+$ rejection and water flux, demonstrating excellent structural stability. The excellent long-term stability observed in Fig. 4(b) is closely associated with the structural robustness of the asymmetric charge heterostructure in the BICNF membrane. Specially, the charge distribution across the membrane thickness establishes a persistent directional electrostatic field, which not only promotes electrostatically guided water transport but also stabilizes ion-transport pathways during continuous operation. Because this asymmetric charge architecture is intrinsically embedded within the crosslinked polymer network rather than relying on labile surface modifications, the resulting electrostatic field remains stable under prolonged exposure to high-Mg brines.

In contrast to reported materials (Fig. 4(c) and Table S1 in the ESM), the BICNF membrane enhances ion selectivity and permeation rate, achieving a high S of ca. 75 together with a high Li^+ permeation rate of $0.65 \text{ mol}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$. This outstanding

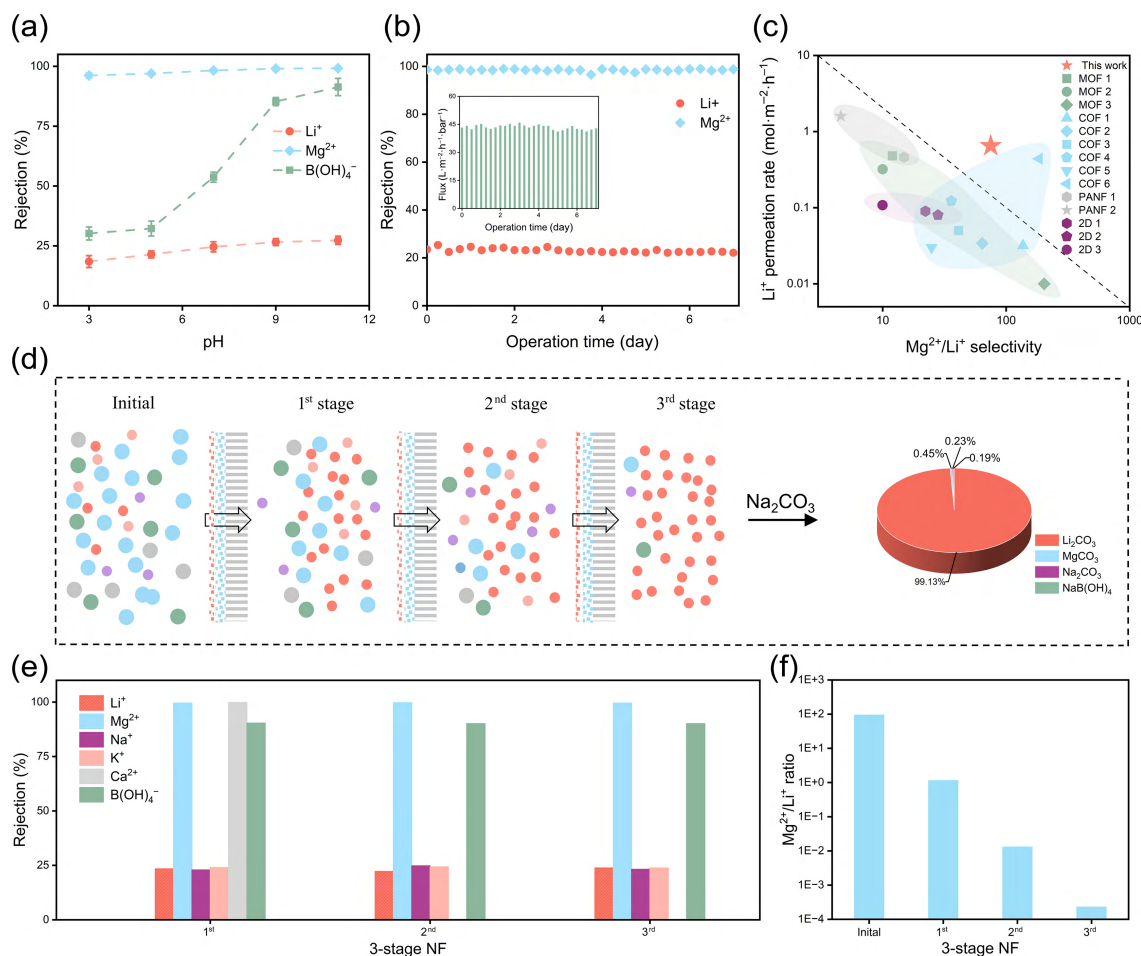


Figure 4 Li extraction performance of the BICNF membrane from simulated salt-lake brine. (a) Ion rejection of the BICNF membrane at different pH conditions. (b) Long-term separation performance of the BICNF membrane (inset is the water flux of the BICNF membrane). (c) Performance comparison of the BICNF membrane with reported NF membranes. (d) Schematic of 3-stage NF process from a simulated salt-lake brine and the corresponding purity of Li_2CO_3 crystallization. (e) Ion rejection and (f) $\text{Mg}^{2+}/\text{Li}^+$ ratio in 3-stage NF process.

performance originates from the bioinspired asymmetric charge heterostructure and precisely regulated surface positive charge density, which decouple ion selectivity from permeability resistance. The positively charged outer surface effectively prescreens and repels Mg^{2+} via the Donnan exclusion, while the negatively charged inner channels selectively attract and accelerate hydrated Li^+ permeation along low-resistance pathways. As a result, selective ion guidance rather than passive size-based filtration governs the separation process, enabling the BICNF membrane to overcome the long-standing limitation between high ion selectivity and permeability that characterizes traditional NF membranes.

The practical Li recovery performance of the BICNF membrane was evaluated using a simulated salt-lake brine test [56]. The brine initially contained about 0.05 M Li^+ along with high concentrations of Mg^{2+} , Na^+ , Ca^{2+} , and B(OH)_4^- , resembling the composition of typical Chinese salt lakes (Fig. 4(d)). Following a 3-stage BICNF separation process combined with sodium carbonate precipitation, the Li^+ concentration in the final product increased to 99.1%, corresponding to battery-grade Li_2CO_3 purity standards (> 99%). Throughout the 3-stages separation, the membrane maintained stable rejection rates for Mg^{2+} (> 99.5%), Ca^{2+} (> 99.8%), Na^+ (< 24.4%), Li^+ (< 23.5%), and B(OH)_4^- (> 90.0%) at pH = 10, demonstrating good selectivity. At this stage, the BICNF membrane

continues to preferentially transport Li^+ while nearly completely excluding residual divalent cations, with Mg^{2+} rejection remaining above 99.4% and Li^+ rejection limited to ~22% (Fig. 4(e)). As a result, the $\text{Mg}^{2+}/\text{Li}^+$ molar ratio is further suppressed from 94.8 to 2.3×10^{-4} , representing a reduction of more than six orders of magnitude relative to the initial brines (Fig. 4(f)). This ultra-low Mg/Li ratio effectively eliminates Mg interference during Li_2CO_3 crystallization, while monovalent ions such as Na^+ and K^+ remain soluble and do not co-precipitate. Consequently, the BICNF membrane enables the production of battery-grade Li_2CO_3 from high- Mg^{2+} salt-lake brines.

4 Conclusions

In summary, we fabricated a BICNF membrane by constructing an asymmetric charge heterostructure that mimics the configuration of biological K^+ channels. A well-controlled interfacial polymerization was adopted to deposit a thin PA layer, followed by surface modification with EPTAC-PEI via a base-catalyzed epoxide-amine coupling reaction. Such design endows the membrane with an asymmetric charge heterostructure, thus establishing a biomimetic K^+ -channel-like architecture with electrostatic steering. Compared with conventional NF membranes, the BICNF membrane exhibits a thinner and smoother selective layer, confirming a well-regulated

interfacial polymerization process. The BICNF membrane achieved a $S_{\text{Mg/Li}}$ of 75 at $\text{Mg}^{2+}/\text{Li}^+$ ratios > 10 and a Li^+ permeation rate of $0.65 \text{ mol}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$, exceeding the conventional NF membranes. It maintained strong rejection of Mg^{2+} , Ca^{2+} , and $\text{B}(\text{OH})_4^-$ during membrane filtration of simulated high-Mg salt-lake brine. When followed by sodium carbonate precipitation, the final Li_2CO_3 product reached a purity of 99.1%, meeting battery-grade standards. Therefore, this study offers a promising pathway for efficient Li extraction and broader ion-separation applications, overcoming the trade-off between ion selectivity and permeation rate.

Electronic Supplementary Material: Supplementary material (experimental section; FTIR images; XPS spectra; zeta potential; SEM image; AFM image; dynamic water contact angles data; water flux data; Li^+ permeation data; current–voltage curve; and separation performance data) is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94908893>.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

X. P. L.: Writing – review & editing, writing – original draft, investigation, data curation. M. Q. Y.: Investigation, data curation. J. X. M.: Writing – review & editing, writing – original draft, supervision, conceptualization.

Use of AI statement

None.

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