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Progress in Preparation of Hydrophilic Polymer Brush-Grafted UiO-66-NH_2 Mixed Matrix Membranes and Their Antifouling Seawater Desalination Performance

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Abstract. Nowadays, the primary dilemma facing humanity is the gradual depletion of freshwater resources. In recent years, global warming and the rapid growth of population and industrialization have intensified this contradiction, and seawater desalination is undoubtedly the optimal solution to this problem. Currently, the most mainstream seawater desalination technology worldwide is reverse osmosis. According to data from the Global Water Intelligence, seawater reverse osmosis accounts for more than 70% of global seawater desalination capacity. Membrane fouling is one of the main limitations of reverse osmosis, including organic fouling, inorganic scaling, and colloidal fouling. Fouling leads to flux decline, increased energy consumption, shortened membrane lifespan, and requires frequent use of chemical cleaning agents, further increasing costs and environmental burden. This paper reviews membrane preparation methods and compares the performance of modified MMMs and TFNs with their respective pristine membranes. From the data, MMMs and TFNs demonstrate superior performance and are fully capable of serving as primary membrane materials for seawater desalination. Research has found that polymers are undoubtedly the most suitable for seawater desalination research. The general process involves first synthesizing UiO-66-NH_2 nanoparticles, then performing polymer brush grafting modification, followed by preparation into mixed matrix membranes and thin-film nanocomposite membranes, and then conducting performance tests on the two different membranes. Polymer brush-grafted UiO-66-NH_2 shows significant advantages in antifouling and desalination. However, long-term stability and cost control remain the core challenges facing the industrialization of this technology.

Keywords: MMMs membrane, TFNs membrane, permeability, seawater desalination, UiO-66-NH_2

1. Introduction

Nowadays, more than 1.8 billion people face freshwater resource scarcity, and polymer membranes are the most mainstream membrane separation materials today [1]. However, traditional membrane separation materials are limited by the "trade-off" effect between permeability and selectivity, which restricts their application in seawater separation [2]. To break through the above bottleneck, MOFs,

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with their surface area and tunable pore channels, are widely regarded as the most suitable materials for water treatment membranes, and UiO-66-NH_2 is one of the most concerned nanofillers in the water treatment field. Currently, the synergistic effect of UiO-66-NH_2 with hydrophilic polymer brushes has demonstrated excellent performance in both antifouling and water flux. Surface-initiated reversible addition-fragmentation chain transfer (RAFT) polymerization has been proven to be an effective method for precisely grafting functional polymer brushes onto the surface of UiO-66-NH_2 [3]. Even with today's highly advanced science and technology, applying UiO-66-NH_2 mixed matrix membranes to industrialization is not an easy task. The first problem to overcome is the lack of large-area, defect-free UiO-66-NH_2-based mixed matrix membrane scale-up preparation, and there is still no long-term operation data for UiO-66-NH_2 mixed matrix membranes in real water bodies. Moreover, there is no mainstream international method for judging performance quality. This paper systematically explores the preparation methods and performance characteristics of UiO-66-NH_2 mixed matrix membranes. It can provide reference basis for subsequent related research and process preparation in this field.

2. Preparation process of UiO-66-NH_2 modified membranes

The method for manufacturing UiO-66-NH_2 mixed matrix membranes is to use solvothermal synthesis with $ZrCl_4$ + 2-aminoterephthalic acid, DMF as solvent, to synthesize UiO-66-NH_2 nanoparticles [4]. Then surface-initiated reversible addition-fragmentation chain transfer (RAFT) polymerization: a chain transfer agent (CTA) was first anchored onto the surface of UiO-66-NH_2, followed by polymerization reaction to grow polymer brushes [5].

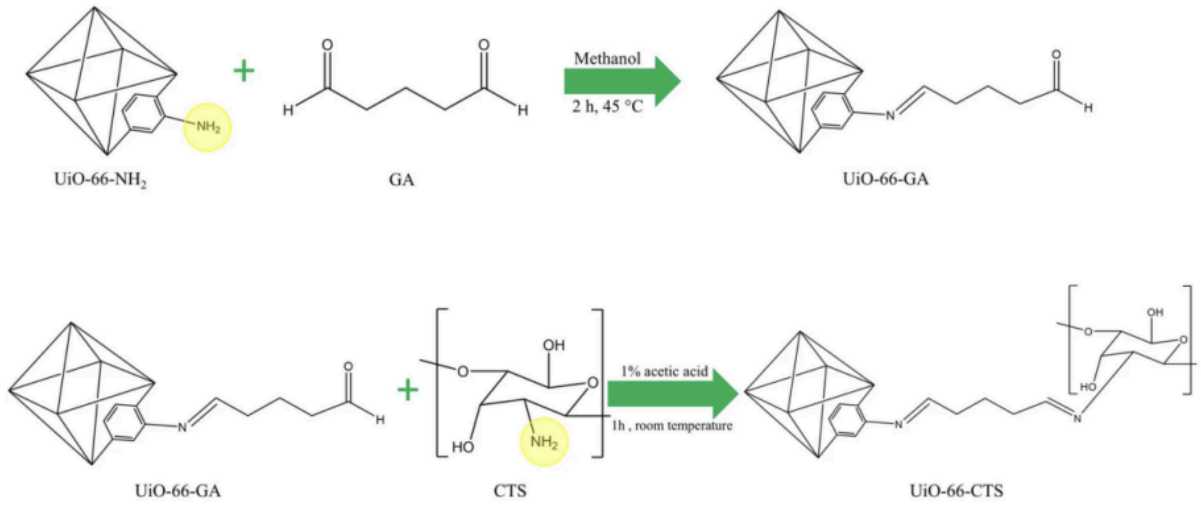


Figure 1. Reaction schematic diagram of stepwise modification preparation of UiO-66-CTS from UiO-66-NH_2 [6]

Figure 1 shows the grafting of chitosan polymer brushes onto the surface of UiO-66-NH_2, finally generating UiO-66-CTS. The prepared polymer brush-functionalized UiO-66-NH_2 is then used to prepare mixed matrix membranes (MMMs) via non-solvent induced phase separation (NIPS) [6]. Meanwhile, thin-film nanocomposite membranes (TFNs) are generated via interfacial polymerization (IP). After preparing these two membranes, they are compared with pristine membranes in terms of water flux/permeability, salt rejection, long-term operation stability, and scale-up preparation potential [7].

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3. Comparative analysis of basic separation performance of MMMs and TFNs membranes

Table 1. Comparative analysis of basic separation performance of MMMs and TFNs membranes [8-11]

Comparison Dimension	MMMS	TFNS
Water flux/permeability	2.31 times improvement (compared to pristine membrane)	About 50% (compared to TFC membrane)
NaCl rejection	>99.9%	Maintains same rejection as TFC
Na2SO₄ rejection	98.20%	97.80%
Long-term operation stability	Structurally stable after 100 hours continuous operation	Performance almost unchanged after 120 hours operation

Table 1 shows the improvement of modified MMMs and TFNs compared to pristine membranes. TFN membranes have higher water flux values, while MMMs have better salt rejection. The relationship between the two membranes is more complementary than competitive, each exerting its own advantages. Hydrophilic polymer brush-grafted UiO-66-NH_2 as a nanofiller can enhance salt rejection and water permeability in both membrane types.

4. Optimal process performance and comprehensive advantage assessment of TFN modified membranes

Table 2. Comparison of flux recovery ratio (FRR) among various nanofillers for mixed matrix membranes [6]

Filter	FRR (%)
Zn: Al_2O_3	98.4
Amino-functionalized TiO_2 nanotube	90
Dodecylamine-functionalized GO	89.1
UiO-66	95.62
UiO-66-NH_2	91.74
UiO-66-CTS	95.14

Table 2 shows the effect of different nanofillers on membrane antifouling performance (FRR, flux recovery ratio), aiming to illustrate which filler modification effect is best through comparison. Within the UiO-66 series internal changes, although the introduction of amino groups (-NH_2) increases hydrophilicity, it may also change the surface charge of MOF or the interfacial compatibility with the polymer matrix, leading to slight decrease in antifouling performance. However, UiO-66-CTS forms hydrophilic polymer brushes on the surface, effectively inhibiting pollutant adsorption through the formation of a dense hydration layer, compensating for the performance loss caused by amination, and restoring FRR to near the level of pristine UiO-66, indicating that the polymer brush grafting modification strategy indeed improves the antifouling performance of the membrane.

As shown in Figure 2, in the performance evaluation of TFN membranes, by optimizing interfacial polymerization conditions and MOF addition amount, TFN-2 membrane demonstrated excellent comprehensive performance. Its water permeability reached as high as ~39 $L \cdot m^{-2} \cdot h^{-1} \cdot bar^{-1}$, and Na_2SO_4 rejection maintained at ~95%, successfully breaking the "trade-off" effect between permeability and selectivity of traditional membranes (Figure a). Meanwhile, the membrane

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exhibited excellent separation selectivity for divalent and monovalent salts (Figures c, d), and maintained highly stable performance during 120 hours of continuous operation (Figure e). In benchmark comparison with other advanced membranes (Figure f), this TFN membrane is located in the upper right region of the performance distribution map, indicating that its separation performance is at the international leading level.

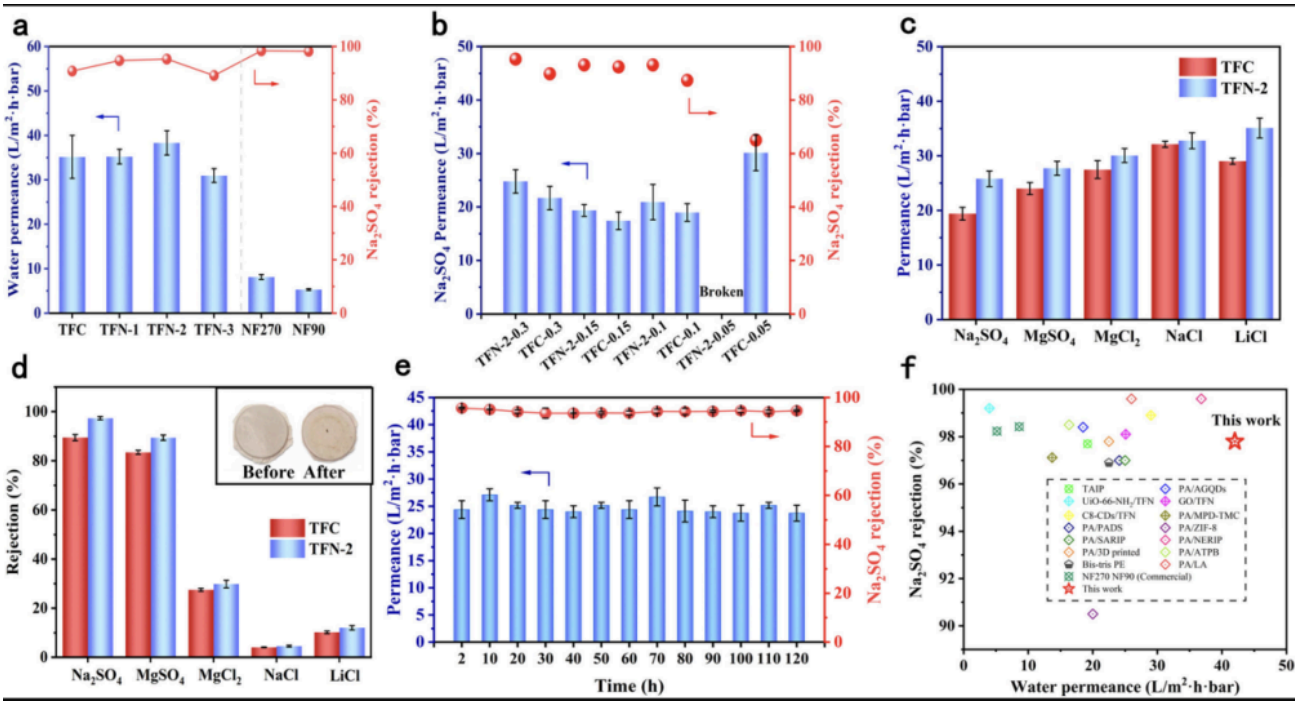


Figure 2. Optimal process performance and comprehensive advantage assessment of TFN modified membranes [9]

5. Existing challenges and development trends

Despite its promise, this technology still faces three major challenges on the path to industrialization. First, the scale-up preparation of large-area, defect-free UiO-66-NH_2-based mixed matrix membranes remains a bottleneck. Second, there is a lack of long-term operation data in real water bodies. Finally, there is a lack of unified international performance evaluation standards, and the synthesis cost of UiO-66-NH_2 is relatively high, requiring the development of green, low-cost synthesis routes while maintaining performance [2].

Future UiO-66-NH_2 membrane materials will not only be antifouling but will develop toward multifunctional directions such as antibacterial and self-cleaning. By combining antibacterial nanoparticles (such as Ag@UiO-66-NH_2) with hydrophilic polymer brushes, dual functions of antibacterial and antifouling can be achieved [3]. The photocatalytic activity of UiO-66-NH_2 can also be utilized in combination with polymer brushes to develop membranes with self-cleaning capability [2].

Moreover, future UiO-66-NH_2 membranes will have the ability to dynamically adjust their surface properties according to environmental changes, achieving on-demand separation, such as smart membranes targeting emerging pollutants like microplastics.

Looking ahead, research on UiO-66-NH_2 membrane materials will gradually move from the laboratory to complex environments such as real seawater or high-salinity wastewater for long-term

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(months or even years) operation testing. For example, some studies have extended the application feasibility assessment of membranes to natural seawater and industrial high-salinity wastewater [2].

Ultimately, the application of UiO-66-NH_2 membrane materials is expected to expand from single water treatment to cross-disciplinary applications. Such membranes have been applied to the treatment of complex wastewater systems such as textile printing and dyeing wastewater, oily wastewater, and laundry wastewater [2]. On the basis of removing pollutants, recovery of salts or specific organic compounds can also be achieved, which is also an important development direction in the field of sustainable water treatment.

6. Conclusion

This review systematically summarizes the preparation methods, performance advantages, and current core challenges of hydrophilic polymer brush-grafted UiO-66-NH_2 (amino-modified UiO-66) in mixed matrix membranes (MMMs) and thin-film nanocomposite membranes (TFNs), and draws the following conclusions:

First, UiO-66-NH_2 is an ideal nanofiller for constructing high-performance water treatment membranes. Its excellent water stability (strong Zr-O bonds, structural integrity under extreme pH conditions), high specific surface area, and tunable pore structure have attracted widespread attention in the fields of water purification and seawater desalination. Incorporating UiO-66-NH_2 into polymer membrane matrices can significantly improve membrane hydrophilicity, construct additional water transport channels, and achieve synergistic enhancement of water flux and salt rejection.

Second, polymer brush grafting modification is an effective strategy to solve the problems of MOF agglomeration and interfacial compatibility. By grafting hydrophilic polymer brushes (such as chitosan, PEG, etc.) onto the surface of UiO-66-NH_2 through methods such as surface-initiated RAFT polymerization, a "molecular bridge" can be formed to enhance MOF-polymer interfacial interactions and eliminate interfacial defects. Data from Figure 2 show that the FRR of chitosan-grafted UiO-66-CTS reached 95.14%, significantly better than unmodified UiO-66-NH_2 (91.74%), compensating for the antifouling performance loss caused by amination, proving that the polymer brush grafting strategy indeed improves the antifouling performance of the membrane.

Furthermore, both MMMs and TFNs can achieve significant performance improvement through the introduction of UiO-66-NH_2, and the two are complementary rather than competitive. In the TFN membrane system, by optimizing interfacial polymerization conditions and MOF addition amount, the water permeability of TFN-2 membrane reached as high as ~39 $L \cdot m^{-2} \cdot h^{-1} \cdot bar^{-1}$, Na_2SO_4 rejection maintained at ~95%, and performance remained highly stable during 120 hours of continuous operation. In the MMMs system, the UiO-66-NH_2-based membrane achieved NaCl rejection of >99.9%, and the structure remained stable after 100 hours of continuous operation. TFN membranes have higher water flux values, while MMMs have better salt rejection. The two membrane configurations each exert their own advantages and can be selected according to specific application scenarios.

In summary, polymer brush-grafted UiO-66-NH_2 demonstrates tremendous potential in breaking through traditional membrane performance bottlenecks in the fields of water purification and seawater desalination. Future research should focus on scale-up preparation process optimization, long-term operation verification in real water bodies, and development of multifunctional integrated membrane systems, promoting the transition of such membrane materials from laboratory to practical application.

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