The Application of Graphene-based Membrane Materials in Seawater Desalination

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Abstract: The seawater desalination membrane technology is confronted with the bottleneck of being unable to balance permeability and selectivity. Two-dimensional materials represented by graphene and its derivatives, with their unique nanostructures and excellent physical and chemical properties, offer new ideas for the development of next-generation separation membranes. This paper mainly adopts the literature research method to systematically review the research progress of graphene-based membrane materials in the field of seawater desalination, focusing on two major technical routes: nanoporous graphene membranes and layered graphene oxide membranes. It elaborates on their structural design, preparation process and separation mechanism, summarizes key performance indicators such as water flux (e.g., 22.39 L/m2·h·bar for GO-CD membranes) and rejection rate (>99%), and analyzes core challenges such as swelling, fouling and large-scale production in the transition from laboratory research to practical application. Finally, this paper compares and prospects the industrialization prospects of different technical routes, with the aim of providing references for the future direction of this field.

Keywords: Graphene Film; Desalination of Seawater; Graphene Oxide; Interlayer Spacing Regulation; Desalination Performance

1. Introduction

1.1 Research Background and Significance

1.1.1 Global water crisis and demand for desalination technology

Freshwater shortage is one of the most serious challenges facing human society in the 21st century. It is predicted that two-thirds of the world's population will face a shortage of fresh water by 2025. Desalination, as an "open source" technology for producing fresh water from seawater, has become a strategic measure to address the water crisis[1]. Since the first desalination plant was built in the 1960s, reverse osmosis technology, with its relatively mature process and high water production efficiency, has gradually dominated the global desalination market and currently accounts for about 50% of the world's desalination capacity[1].

1.1.2 Limitations of traditional reverse osmosis membranes and opportunities for graphene-based membranes

Traditional polyamide reverse osmosis membranes face an inherent contradiction between permeability and selectivity in long-term operation, are vulnerable to chemical degradation and biological contamination, and performance improvement is close to the theoretical limit[2].Graphene is a two-dimensional atomic crystal formed by the tight binding of a single layer of carbon atoms in an sp^2 hybridization manner. It has excellent mechanical properties and stability and can be used as a basic material to construct various macroscopic two-dimensional separation membranes with different properties[3].Theoretical calculations suggest that ultrafast transport of water molecules and precise sieving of salt ions can be achieved by introducing sub-nanopores or constructing interlayer nanochannels in the graphene lattice, providing a new path to break through traditional technical bottlenecks[4].

1.1.3 Theoretical significance and practical Value of this study

From a theoretical perspective, the study of mechanisms such as nanoconfined mass transfer and ion sieving in graphene-based membranes helps to deepen the understanding of ultrafine selective separation processes. Gao et al. systematically summarized the selective permeability mechanism of graphene-based films and pointed out that precise regulation of the size and functional groups of nanopores or nanochannels is the key[4]. From a practical perspective, the development of high-performance graphene-based membranes is expected to significantly reduce the energy consumption and cost of seawater desalination. Seo et al. prepared graphene membranes with inherent nanochannels using ambient air chemical vapor deposition and achieved stable operation for 72 hours when treating Sydney Harbour seawater, demonstrating their feasibility[5].

1.2 Research Objectives and Content Framework

1.2.1 Research objectives

This study aims to systematically review and evaluate the research context and current development status of graphene-based membrane materials in seawater desalination. By comparing and analyzing the design principles, performance and engineering challenges of different technical approaches, clarify the current research focus, core challenges and future breakthroughs, thereby providing valuable references for academic research and technological development in this field.

1.2.2 Paper structure arrangement

This paper first classifies graphene-based films based on their structural characteristics and elaborates on their preparation techniques and regulation strategies. Secondly, the core performance indicators of the two types of membranes and the challenges they face in practical application scenarios are systematically evaluated. Again, a comprehensive analysis of their industrialization potential and key technical obstacles. Finally, summarize the full text and look forward to future development trends.

2. Classification and Preparation Techniques of Graphene-based Films

2.1 Nano-Porous Graphene Membranes

2.1.1 Structural characteristics and separation mechanism

Nano-porous graphene membranes achieve sieving by creating uniform-sized pores smaller than 1 nanometer on a complete graphene lattice. The separation mechanism is mainly based on the strict size exclusion effect: the diameter of a water molecule is about 0.32 nanometers, while the hydration diameter of common salt ions in seawater is usually about 0.7 nanometers. Therefore, when the pore size of the nanopore is larger than the diameter of the water molecule and smaller than the hydration diameter of the salt ion, the size screening effect can allow the water molecule to pass through while blocking the ion[4]. Molecular dynamics simulations predict that for monolayer graphene with etched pore diameters of 0.45 nanometers, the desalination rate can reach 100% [1].

2.1.2 Review of Pore-forming techniques and methods

Achieving controllable and uniform sub-nanometer pore-forming is the greatest challenge. Yuan's team designed a centimeter-scale self-supporting, mechanically strong nano-porous graphene membrane made of mesoporous silica film, using short pulses of oxygen plasma to bore holes within 1.2 nanometers in diameter. The membrane blocked 85% of sodium chloride and up to 98% of macromolecular solutes and could withstand pressures up to 10 megapascals [1]. However, the grain boundaries reduce the mechanical strength of graphene and damage the structural integrity of graphene, making it difficult to increase the area of the graphene layer. Growing graphene layers through chemical vapor deposition makes the cost of nano-porous graphene films much higher than that of polymer films, and large-area graphene layers are more prone to oversized pores and defects [1].

Wan et al. pointed out that porous graphene films drilled into single-layer graphene have poor mechanical properties and are prone to damage during use, so the researchers enhanced their stability by constructing composite films. In addition, defect-free graphene achieves the purpose of blocking by extending the path of gas molecules through the film. By taking advantage of this property, the gas barrier effect [3].

2.1.3 Research Progress and Representative Works at Home and Abroad

Research in this field is still in the basic exploration stage. Gao et al. reviewed the research progress of introducing nanopores into graphene layers by physical or chemical methods, pointing out that physical methods can precisely control the morphology and size of nanopores by controlling the irradiation dose, while chemical methods can not only control the size of nanopores by controlling the etching time, but also insert different types and quantities of charged functional groups at the edges of nanopores Cationic or anionic selective permeability[4]is produced. However, in the practical application of large-area graphene films, the intrinsic defects introduced by the CVD process and the cracks generated during the transfer process significantly reduce ion selectivity, resulting in a substantial decrease[4]. Although nano-porous graphene films have extremely high desalination potential in theory, their complex preparation process and high cost limit their large-scale application. In contrast, layered graphene oxide (GO) membranes have become a current research hotspot due to their simple preparation and tunable structure.

2.2 Layered Graphene Oxide-based Films

2.2.1 Structural regulation strategies (interlayer spacing regulation, crosslinking modification, etc.)

Layered graphene oxide films are composed of a large number of GO nanosheets stacked with surfaces rich in oxygen-containing functional groups, and the core of their separation performance depends on the size of the nanochannels between the sheets. Gao et al. pointed out that GO undergoes swelling in water and the interlayer spacing gradually expands, resulting in a decrease in selectivity. Therefore, while precisely regulating the interlayer spacing, it is necessary to ensure its continuous selective permeability [4].

Feng Bo et al. prepared graphene oxide-doped polyimide hollow fiber membranes using phase conversion. XRD characterization revealed that when water molecules entered the interlayer, they combined with oxygen-containing functional groups to form a water film, with the remaining space in the middle being 0.33nm, which was between the size of water molecules and hydrated salt ions, thus having a good desalination effect [6].

Jin Lejia et al. reviewed the research progress of cross-linked graphene oxide composite films and pointed out that covalent cross-linked GO composite films with stability in aqueous solution could be produced by the reaction of cross-linking agents such as diamines and dicarboxylic acids with oxygen functional groups on GO. The cross-linked structure gives the membrane better stability and proton conductivity, resulting in a composite membrane with better separation performance [7].

Wei Yangyang et al. introduced hydrophilic carbon nanotubes and alternately sprayed multiple layers of GO and carbon nanotubes on polydopamine-modified base membranes to prepare multi-layered composite desalination membranes. The tube space of the carbon nanotubes and the assembly gap of MLGO-CNTs introduced effective permeation channels to the composite membrane. When the mass per unit area of MLGO-CNTs was 72mg/m², the salt retention rate of the composite membrane to the $Na2\ SO4$ solution was up to 92.8% [8].

Han Hongda prepared GOCD supramolecular composite membranes by covalently bonding cations - β -cyclodextrins to the surface of GO sheets. By increasing the interlayer distance of the membrane, the GOCD membrane has a stable interlayer distance in both dry and wet states and exhibits excellent anti-swelling properties. Compared with pure GO membranes, the flux of GO-CD membranes increased from 3.73 L/ (m^2·h·bar) to 22.39 L/ (m^2·h·bar), with a rejection rate of 99.22%[9].

2.2.2 Preparation process and scaling progress

GO membranes were mainly prepared using solution methods such as vacuum filtration, spin coating, spray coating, dip coating, and layer self-assembly. Wan Wubo et al. systematically reviewed the preparation methods of graphene-based films such as vacuum filtration, spin coating/spray coating, layer self-assembly, and blending. The vacuum filtration method is simple to operate, offers a wide range of substrate membrane options, and the thickness of the membrane can be regulated by solution concentration. The resulting films have good mechanical properties and excellent separation performance; Spin coating requires simple equipment structure and controllable conditions, and the membrane area and thickness can be adjusted to a certain extent; Spray coating is simple and efficient, with a wide range of substrate options, and can be used to prepare graphene films over large areas; The layer-by-layer self-assembly method is simple to operate, and the film-making process is not bound by the shape and size of the substrate, and is mainly used to construct complex multilayer film structures; The blending method allows for the functional modification[3].

To achieve industrialization, the focus of research has shifted to large-scale continuous preparation. Wei et al. fabricated PDA-MLGO-CNTs layered composite desalinated membranes on polydopamine-modified base membranes by alternating spraying, and achieved precise regulation[8]. However, achieving uniformity, defect-free and strong bonding to the support on large areas remains a major engineering bottleneck at present.

2.2.3 Representative Studies and Research Trends of GO Membranes

Both at home and abroad have achieved remarkable results in the field of GO membranes. Cheng et al. pointed out that graphene oxide films can achieve the function of seawater desalination. By effectively controlling the spacing of the membrane layers, it can achieve the screening of pure water and salt ions, improve the desalination efficiency, and graphene desalination membranes are durable and have a low cost [2]. Zheng Jiajia et al. reviewed the research progress on the removal of antibiotics from water by GO films, and pointed out that GO has good wettability and surface activity, and the oxygen-containing functional groups it contains make it easy to functionalize, which can directly electrostatically adsorb or bond with the medium material, making the obtained composite structure [10].

The domestic team has performed well in membrane structure regulation for practical applications, anti-swelling properties and large-scale preparation. Feng et al. modified the surface and internal structure of hollow fiber membranes by doping GO to increase the permeation channels [6]. By introducing carbon nanotube intercalation, Wei et al. improved the stability [8] Jin Lejia et al. systematically summarized the performance characteristics [7]. Han Hongda achieved efficient retention [9].

3. Performance Evaluation of Graphene-based Membranes in Seawater Desalination

3.1 Analysis of Desalination Performance Indicators

3.1.1 Water flux and desalination Rate

In the experiment, the optimized graphene-based membrane showed certain potential. The following table presents some examples and illustrates the performance advantages.

Table 1. Water Flux and Salt Rejection Rate of Representative Graphene-based Membranes

Membrane materials	Water flux (L/m^2·h·bar)	Retention rate (%)	References
GO-PI hollow fiber membrane	14.99	> 99.7%	[6]
PDA-MLGO-CNTs	4.8	Up to 92.8%	[8]
GO-CD	22.39	99.22% (G250)	[9]

As shown in Table.1, the GO-PI hollow fiber membrane prepared by Feng Bo et al. had a permeation flux of 14.99kg/(m^2·h) to 3.5% sea salt water at 90°C and a desalination rate higher than 99.7%. This is due to the hydrophilicity of GO, and the addition of PI enhances the hydrophilicity of the membrane, changes the surface and cross-sectional structure of the hollow fiber, reduces surface defects and increases the permeation channels of water molecules[6]. Note that in the original reference, the value and unit of water flux were 14.99 kg/(m^2·h). Convert it here:

Mass to volume conversion: For pure water or dilute brine solutions, the density ρ is very close to 1 kg/L at room temperature and high temperature. Therefore, 1 kg of water ≈ 1 L of water.

Conversion results: 14.99 kg/(m^2· h) = 14.99 L/(m^2· h)

Pressure unit conversion: 1 MPa = 10 bar.

In Feng Bo et al. 's experiment, the driving force for pervaporation was a vacuum of 0.1 MPa (1 bar).

Since the driving force of the experiment was exactly 1 bar, numerical division was not necessary:

14.99 L/ (m^2· h) present 1 bar = 14.99 L /(m^2· h· bar)

The PDA-MLGO-CNTs composite membrane prepared by Wei Yangyang et al. achieved a maximum salt retention rate of 92.8% for 1 g/L Na_2SO_4 solution under an operating pressure of 5bar, and a water flux of 4.8 L/ (m^2·h·bar), which was 71.4% higher than that of the PDA-MLGO membrane. As the mass ratio of MLGO/CNTs decreased, the salt rejection rate of the composite membrane decreased slightly, and the water flux increased significantly[8]. In this salt rejection process, the size exclusion mechanism dominated.

The GOCD membrane prepared by Han Hongda had a rejection rate of 99.22% for G250 and a flux of 22.39 L/ (m^2·h·bar) when the loading was 0.1 mg. The flux of GO-CD membranes was significantly higher than that of pure GO membranes. Go-CDs membranes have a rejection rate of over 99% for negatively charged dye molecules and 62.56% for rhodamine B, a result[9].

3.1.2 Anti-contamination and long-term stability

Long-term stability

Is at the core of GO membrane applications. Gao et al. pointed out that when water acts on the GO membrane, the GO nanosheets become negatively charged due to the ionization of some oxygen-containing functional groups, and the electrostatic repulsion between layers gradually expands the interlayer spacing, causing swelling and resulting in a decrease in selectivity[4]. Jin Lejia et al. pointed out that the practical application of GO as a water treatment membrane is limited due to its hydrophilicity and electrostatic repulsion in the water environment, which can easily lead to the disintegration of the membrane structure in practice[7]. Introducing covalent bonds that connect graphene oxide layers, etc., may enhance the stability of the membrane.

Wei Yangyang et al. investigated the performance stability of PDA-MLGO-CNTs films in 400 hours of continuous experiments. The salt rejection rate of the composite membrane showed high stability. After 400 hours, the salt rejection rate of the composite membrane with different MLGO/CNTs mass ratios remained in the range of 82.8% to 89.9%. The water flux decreased from 2.3 to 4.8 L/ (m^2·h·bar) to 1.8 to 3.3 L/ (m^2·h·bar) but was still higher than that of PDA-MLGO membranes. The attenuation rate of the PDA-MLGO-CNTs water flux was in the range of 21.7% to 31.4%, much lower than the 50.0% of the PDA-MLGO membrane, due to the less force effect on the space inside the rigid CNTs tubes, which suppressed the compression[8].

Han Hongda studied the stability of the GO-CD membrane in a 12-hour continuous separation experiment, and the results showed that the GO-CD membrane maintained a retention rate of more than 98% for BBG molecules while the flux loss was no more than 2 L/ (m^2·h·bar). XRD results showed that the GOCD membrane had a very stable interlayer distance in both dry and wet states and demonstrated excellent anti-swelling properties[9].

In terms of anti-pollution performance, Cheng et al. pointed out that graphene itself is lipophilic and is widely used in the treatment of oil and organic solvents, etc. Graphene aerogel and others can increase adsorption capacity and improve adsorption rate, and can be used for emergency treatment[2]. Seo et al. used permeable graphene-based membranes for membrane distillation desalination, and in salt water mixtures containing contaminants such as oil and surfactants, the retention rate of water vapor flux and the retention rate of salt were significantly better than those of commercial distillation membranes. Calculations show that the interaction between graphene and pollutant molecules belongs to the type of weak physical adsorption, and the kinetic energy brought by the continuous inflow of water overcomes[5].

3.2 Performance Challenges in Practical Applications

3.2.1 Performance degradation in complex water quality

The composition of real seawater is extremely complex, containing components such as dissolved organic matter, microorganisms, colloidal particles, in addition to the main salts. The vast majority of current studies use simplified saltwater models, and there is insufficient research on the long-term evolution of membrane performance in complex real water bodies. Although GO membranes show excellent separation performance in laboratory conditions, their performance often deteriorates significantly in actual seawater due to the coexistence of multiple ions, the presence of organic matter and microorganisms. Seo et al. in the Sydney Harbour desalination test, oil contaminants in the seawater caused severe wetting problems in the polytetrafluoroethylene based membrane, resulting in significant contamination of the polytetrafluoroethylene-based membrane and a continuous decline in water vapor flux Permeable graphene-based membranes maintained a stable high water vapor flux (50 liters per square meter per hour) and a 100% salt rejection rate for 72 hours[5].

3.2.2 Selectivity issues in multi-ion coexistence

Multiple ions coexist in seawater such as Na^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, SO_4^{2-}. The separation of GO membranes is the result of the combined effect of size sieving and electrostatic repulsion. Gao et al. pointed out that multivalent ions such as Ca^{2+} and Mg^{2+} may form coordination with oxygen containing functional groups on the GO surface, causing local contraction or expansion of the membrane structure and thereby affecting ion selectivity[4]. Cheng et al. pointed out that the pH value affects the adsorption capacity of graphene for metal ions, and an increase in pH value enhances the adsorption of Pb^{2+} and Cd^{2+}, which is superior to other traditional adsorption materials[2]. Han Hongda's research shows that GOCD membranes have retention rates of 16.47%, 22.39%, and 52.47% for NaCl, $MgSO_4$, and Na_2SO_4, respectively. According to the Donnan repulsion effect, Na^+ can be attracted by negatively charged GOCD membranes, and this process is accompanied by the permeation of Cl^- to balance the charge. This leads to the transmembrane transport of NaCl[9].

3.2.3 Mechanical strength and durability

Macroscopic GO membrane interlayer bonding mainly relies on van der Waals forces, and interlayer slippage may occur under high-pressure reverse osmosis conditions. It can be prepared as a thin-film composite membrane, but the interfacial bonding strength is key. Feng et al. doped GO into polyimide and utilized the strong covalent bond formed between the oxygen-containing functional groups on GO and the PI resin to alter the surface and internal structure of the hollow fiber membrane, generating a double-layer finger pore structure and increasing the water molecule permeation channel. The water flux and rejection rate remained basically unchanged at 75 ° C for 120 hours of operation, demonstrating high stability[6]. Jin et al. pointed out that the cross-linked GO composite membrane, by introducing chemical bonds such as covalent bonds connecting the GO layer, has a strong hydrogen bond[7].

4. Industrialization Prospects and Key Technical Issues of Graphene-based Membranes

4.1 Comparison of Technical Routes and Analysis of Industrialization Potential

4.1.1 Comprehensive Evaluation of Performance - Cost - Scale

Nano-porous graphene membranes have the highest theoretical performance limit, but their preparation process is extremely complex and costly, making it difficult to scale up and not commercially viable in the short term. Gao et al. pointed out that the synthesis of large-area graphene films mostly relies on the CVD process, which not only has large defects but also inevitably introduces a small number of intrinsic defects into the graphene, making the film unable to effectively prevent the solute from passing through, and it also introduces additional defects in subsequent processing, which leads to a significant decrease in the mechanical[4]. Layered GO films, based on solution processing, have relatively easy access to raw materials and are highly compatible with existing film preparation processes, making them the most promising path for industrialization at present. Cheng et al. pointed out that the REDOX method is the most commonly used method for preparing graphene. Preparing graphite by this method has low cost, simple operation and stable quality, and is suitable for large-scale production[2]. Jin Lejia et al. systematically compared GO film preparation techniques such as vacuum filtration, dip coating, spin coating and layer-by-layer self-assembly, and pointed out that each of these methods has its own advantages. Among them, vacuum filtration is a widely used method for preparing graphene oxide[7].

4.1.2 Exploration of the most promising technical paths

In general, GO composite films are most likely to be the first to achieve commercial application. The GO-PI hollow fiber membrane prepared by Feng Bo et al. has shown excellent desalination performance[6]. The PD-MLGO-CNTs layered composite desalination membrane prepared by Wei et al. by alternating spray method maintained good separation performance during 400 hours of continuous operation[8]. The GOCD membrane prepared by Han Hongda through covalent bonding of cyclodextrin derivatives significantly improved water flux and anti-swelling performance while maintaining a high rejection rate[9]. The CVD graphene membrane developed by Seo et al. achieved stable operation for 72 hours when treating Sydney Harbour seawater, demonstrating the potential for practical application[5].

4.2 The Transition Path from Theory to Application

4.2.1 Establishment of a standardized testing System

Current research lacks a unified and standardized performance testing standard. There are differences in test pressure, water quality, membrane area, flux reporting units, etc. used in different literatures, making it difficult to compare data and objectively assess the progress of the technology. Therefore, establishing a set of standardized test standards that are close to actual operating conditions is the foundation for promoting the healthy development of this field.

4.2.2 Current Situation and Bottlenecks of Large-scale Preparation Technology

There are two major challenges in the large-scale production of GO films: one is the mass production of high-quality, stable graphene oxide; The second is the continuous coating technology of large-area, uniform, high-performance film layers. Wan et al. pointed out that the films produced by the vacuum filtration method are prone to damage during the separation from the base membrane, making it difficult to maintain their integrity; Spin coating is difficult to produce on a large scale and has the problem[3]. The alternate spraying method adopted by Wei Yangyang et al. provides a new idea for large-scale production, achieving precise regulation[8].

4.2.3 Key Obstacles and Countermeasures for engineering application

The key obstacles to engineering application include: long-term operational reliability verification, dedicated membrane element development, full industrial chain cost control, etc. The solution lies in strengthening industry-university-research cooperation and promoting innovation throughout the entire chain from material design, membrane preparation to system integration. Jin et al. pointed out that most of the current preparation methods are at the laboratory research stage, and further research is needed to assess the cost-effectiveness of large-scale membrane preparation and monitor its long-term stability in practical applications[7]. Gao et al. also stressed the need to improve the quality of existing CVD[4].

5. Conclusions and Prospects

5.1 Conclusions

Graphene-based membrane materials, especially layered graphene oxide membranes, show great application potential in seawater desalination. Existing research has made significant progress in the precise control of interlayer spacing, enhanced structural stability, functionalized modification, and membrane design for specific processes. Nano-porous graphene membranes theoretically have the highest performance ceiling, but due to many factors, it is difficult to achieve large-scale application in the short term[1][4]. Layered GO membranes are based on solution processing, and the raw materials are relatively easy to obtain. Through certain strategies and techniques, effective regulation of the interlayer spacing and stabilization of the membrane structure can be achieved[3][6-9].

To achieve large-scale application of this technology, the following core issues must be addressed: long-term stability in complex real water quality and continuous optimization of anti pollution performance; And low-cost, high-throughput, high-consistency large-scale manufacturing technology.

5.2 Prospects

Future research will present the following trends: A. Combining molecular dynamics simulations and advanced characterization techniques to deeply reveal mass transfer and contamination mechanisms in complex systems. Seo et al. combined DFT calculations and molecular dynamics simulations to study the permeation mechanism of water vapor through graphene overlapping grain boundaries, providing a theoretical tool for understanding water transport[5]. B. Develop new two-dimensional material hybrid membranes, smart responsive membranes and more environmentally friendly and efficient stabilization strategies. The idea of regulating the interlayer spacing of GO films by covalently bonding cyclodextrin derivatives by Han et al. is worthy of further expansion[9]. C. Promote the establishment of standard testing methods and conduct long-term research based on real seawater. Seo et al. 's research on 72-hour desalination tests using Sydney Harbour water provides an example for engineering validation[5]. D. Strengthen in-depth collaboration between materials engineers, chemical engineers and enterprises to establish a comprehensive R&D system.

With the continuous optimization of the preparation process and the establishment of a standardized testing system, graphene-based films are expected to gradually move from the laboratory to pilot-scale applications within the next decade.

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