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An integrated study of dual membrane techniques for the removal of disinfection by-products in drinking water

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Abstract. In the context of escalating global water resource pressures and rising populations, ensuring the quality and safety of water supplies has become paramount. Natural Organic Matter (NOM) and Disinfection By-Products (DBPs) are prevalent contaminants in numerous water sources. Their presence compromises water's sensory qualities, but certain DBPs have been substantiated to be associated with diverse health implications, including carcinogenic risks. Hence, developing efficient, cost-effective, and reliable technologies for their removal is of critical significance. Dual-membrane filtration, an emergent water treatment methodology, demonstrates profound potential in addressing NOM and DBPs.This article offers a comprehensive review of the applications of dual-membrane filtration across various water treatment paradigms, underscoring its pivotal role in safeguarding potable water quality and addressing future water quality challenges. This study reveals that the MF-NF membrane achieves a removal rate of disinfection by-products of approximately 70%-80%. The UF-NF membrane presents an overall UV254 removal efficiency of over 90%. The NF-RO membrane demonstrates removal efficiencies exceeding 89% for trichloromethane and halogenated acetic acid formation potential. Future research should focus on optimizing the performance of dual-membrane systems, investigating novel membrane materials, and assessing the removal efficiency for emerging DBPs.

Keywords: Disinfection By-Products, Natural Organic Matter, dual-membrane filtration, MF-NF membrane, UF-NF membrane, NF-RO membrane.

1. Introduction

In modern society, drinking water's safety and hygiene are among the essential issues of concern for people. Many countries and regions use disinfectants to eradicate harmful microorganisms in water. However, during the use of disinfectants, NOM in the water source reacts with the disinfectants (such as chlorine) to form disinfection DBPs, including trihalomethanes (THMs), haloacetic acids (HAAs), ketones, aldehydes, and other compounds. These DBPs have been proven to pose potential risks to human health, such as carcinogenicity, reproductive disorders, and chronic diseases. Toxicological studies have shown that most identified DBPs have cytotoxic, neurotoxic, and genotoxic effects. Epidemiological studies have also shown a correlation between long-term consumption of chlorinated drinking water and bladder cancer, colon cancer, and miscarriage in pregnant women. Therefore, removing DBPs from drinking water has become one of the current research hotspots [1].

Ensuring the safety of drinking water is crucial for maintaining public health. Therefore, it is necessary to develop effective methods to remove DBPs to improve the quality and hygiene of drinking water, which can help reduce the incidence of diseases and enhance people's quality of life. Conventional treatment processes have made significant contributions to providing drinking water for residents and ensuring their water safety. However, the increasingly severe water pollution has led to a growing number of pollutants in water, making conventional processes such as chlorination and activated carbon adsorption less effective in removing organic compounds from water. The total organic carbon and permanganate index removal rates are between 20% and 30%. At the same time, conventional processes show significant removal of turbidity, with the effluent turbidity ranging from 0.73 to 0.96 NTU. However, excessive dissolved organic matter in the water hinders the destabilization of colloids, resulting in a significant decrease in the effectiveness of conventional treatment processes in removing raw water turbidity. Therefore, researchers have started exploring new technologies and methods to address this issue [2].

Membrane filtration technologies, such as UF and RO, can reduce the concentration of DBPs through physical barriers and selective removal [2]. UF membranes can remove suspended solids, bacteria, and larger organic molecules, thereby reducing the formation of DBPs. Reverse osmosis membranes, on the other hand, can selectively remove dissolved DBPs, providing a more thorough removal effect. Compared to traditional treatment methods, dual membrane filtration technologies have many advantages, such as high removal efficiency, low energy consumption, and reduced generation of secondary pollutants.

Despite some existing studies demonstrating the effectiveness of membrane filtration technologies in removing disinfection byproducts (DBPs), further research and improvement are still required regarding membrane selection, design, and optimization of operational conditions. This paper focuses on dual membrane filtration technology and comprehensively reviews the removal of natural organic matter (NOMs) and DBPs in various water treatment processes. The primary emphasis is placed on evaluating the effectiveness and current application status of different removal methods while exploring their feasibility and sustainability. Ultimately, this research aims to provide scientific evidence and recommendations to support the development of the drinking water industry and the protection of public health.

2. Types and control of disinfection by-products in drinking water treatment

2.1. Types of disinfection by-products

DBPs are a type of secondary pollutants generated during the disinfection process when disinfectants react with precursors such as natural organic matter (NOM), anthropogenic pollutants, and halide ions present in water.

Since the discovery of THMs in chlorinated disinfection water in 1974, there has been an increasing focus on and research into DBPs in drinking water. Common DBPs can be classified into organic and inorganic types. In organic DBPs, cx3r-type DBPs have become a typical category due to their high toxicity, high concentration, and frequent detection. Further investigations have led to the identification of DBPs with even greater toxicity, including haloacetamides, halobenzoquinones, et al. Additionally, since 1987, inorganic DBPs generated by alternative disinfectants, such as chlorite, chlorate, and bromate, have also been discovered. Overall, over time, the types and quantities of DBPs in drinking water have continued to increase, and the related field has experienced rapid development [1].

2.2. Current approaches to control disinfection byproducts

Methods for controlling DBPs in drinking water can be categorized into four main groups: source control, process control, end-of-pipe control, and coordinated control. Source control involves reducing the concentration of DBP precursors in water through source protection or water treatment processes before adding the disinfectant. Source control includes source protection, conventional processes, enhanced conventional processes, pre-treatment, and advanced treatment processes. Process control is aimed at

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minimizing the formation of DBPs or altering the types of DBPs generated by modifying disinfection methods and optimizing disinfection processes. End-of-pipe control involves using physical or chemical methods to eliminate a range of already-formed DBPs. Coordinated control involves the integration of source, process, and end-of-pipe control methods to effectively manage multiple types of DBPs [1].

Among these control methods, membrane filtration is primarily applied in the enhanced conventional source and end-of-pipe control processes. Conventional treatment processes have become increasingly challenging to improve drinking water quality fundamentally. Therefore, selecting membrane processes for advanced treatment facilities is crucial in enhancing the removal efficiency of dissolved organic compounds, notably trace organic substances [3].

3. Removal of precursors and disinfection byproducts from water through membrane filtration

3.1. Reverse osmosis membrane

Reverse osmosis (RO) technology is a separation method in which water molecules permeate through a membrane under higher solution osmotic pressure, while the membrane retains solute molecules. Due to microscopic pores, RO membranes can retain various pollutants present in water, such as salt ions, emerging organic contaminants, and others. Consequently, RO technology is effective in removing DBPs from drinking water [4].

The mechanism responsible for removing DBPs involves steric hindrance, electrostatic exclusion, and membrane adsorption. Steric hindrance occurs due to the small average pore diameter of the RO film, which is less than 1 nm [5]. Consequently, under external pressure, the steric resistance effect efficiently prevents a significant portion of large-sized DBPs from passing through. It is logical to assume that DBPs with higher molecular weights are more effectively trapped by the RO membrane, resulting in a higher removal rate. Regarding electrostatic exclusion, the RO membranes can either facilitate or impede the interception of ionic DBPs depending on the influence of functional groups affected by pH levels resembling water. Membrane adsorption also plays a role in removing uncharged DBPs that exhibit greater hydrophobicity [5].

Research has shown that when the concentration of pollutants is not excessively high, the RO system can serve as a suitable alternative for the removal of volatile organic compounds (VOCs), trihalomethanes (THMs), several pesticides, and solvents [3]. Additionally, some studies have indicated that this technology is the most effective water treatment method for simultaneously removing bromides, iodides, and their respective organic and inorganic DBP precursors.

3.2. Ultrafiltration membrane

UF is a membrane separation process that utilizesUF membranes with a pore size range of 0.01~0.1μm. These membranes consist of a fibrous or spongy fouling support layer and an epidermis layer that acts as a filter. The pressure-driven process enables the permeation of solvents and small molecules through the membrane, while macromolecular solutes are left behind. This technology is characterized by UF membranes with high flux, high-temperature resistance, and strong oxidation resistance, as well as membrane modules that facilitate water reuse and protein separation [6].

Compared to conventional processes, UF has several advantages, including a high turbidity removal rate, no need for toxic processes that involve chlorine, and low energy consumption. Additionally, the process requires a small area, is easy to automate control, and saves workforce and land resources [4]. UF is commonly used for reducing turbidity, suspended solids, and particles. However, UF is not as effective in separating humic substances, which have a high potential for forming THMs (trihalomethanes) and HAAs (haloacetic acids). In contrast, NF is more successful in removing precursors of THMs. UF membranes are highly effective in removing DBP precursors during laboratory-scale experiments. However, it has been observed that organic carbon, specifically AOC (assimilable organic carbon), cannot be successfully removed through this treatment method [3].

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3.3. Nanofiltration membrane

The separation mechanisms of NF membranes primarily encompass the steric hindrance effect, Donnan effect, and dielectric exclusion effect [7]. The steric hindrance effect, also known as a sieving effect, refers to the selective separation of substances based on the size of membrane pores. The Donnan effect arises from the charged surface of NF membranes, which results in the repulsion of ions and makes it difficult for charged ions to pass through the membrane pores. The dielectric exclusion effect occurs due to the disparity between the dielectric constant of water and the dielectric constant of the membrane material, leading to the exclusion of ions at the membrane surface. Additionally, in certain circumstances, adsorption may also influence the removal efficiency of NF membranes for pollutants [7].

For haloacetic acids, NF membranes have a better removal effect due to the influence of electrostatic repulsion. The primary removal mechanism of nitrosamines is the resistance effect. Therefore the removal effect for this type of substance could be better, especially for NDMA due to its smaller molecular weight, resulting in a removal rate of only about 20%. Due to the electrostatic repulsion effect, the NF membrane has a better removal effect on the precursors of nitrosamines, with a removal rate generally above 90%. Therefore, the removal of these DBPs should focus on the removal of their precursors. The main removal mechanism for HKs, HANs, and their precursors is the resistance effect. NF has a poor control effect on the precursors but has some removal effect on the DBPs [7]. Different NF membranes show significant differences in the removal effect of trichloroacetaldehyde-like substances, which are greatly influenced by the properties of the membrane. Therefore, suitable NF membranes should be selected for their removal.

4. Research status and advantages of the double membrane method in drinking water treatment

The double-membrane method is a frequently employed water treatment technique, extensively utilized in areas such as drinking water, industrial wastewater treatment, and seawater desalination. Research has found that NF membranes are commonly used for drinking water treatment in the double membrane method. These membranes are known for their capability to retain medium and small molecular weight organic pollutants in drinking water and effectively reduce the hardness of the water. The combined process of NF membranes with MF, UF, and RO membranes can be used for advanced treatment of drinking water to meet different practical needs.

4.1. Microfiltration / Nanofiltration

Liu et al. [8] were the first to apply the “pressed-pot MF with NF” water treatment process domestically. They successfully adapted and enhanced this process for large-scale engineering projects involving deep drinking water treatment. They confirmed the practicality of using a combination of pressure tank MF and nanofiltration in engineering applications.

In the application, NF membranes can effectively remove medium and small molecular weight organic pollutants from drinking water. They also reduce water hardness and selectively remove Ca2+, Mg2+, SO2-4, NO-3, and other trace elements. However, the NF system may experience fouling over time, negatively affecting its performance. Using pressure tank MF as a pretreatment process for NF proves to be effective in meeting the requirements for NF inlet water. The NF system demonstrates stable operation at high flux and exhibits excellent membrane performance. The project achieves an overall high recovery rate in the NF system, with TOC and COD removal rates exceeding 90%. DBPs removal rates range from 70% to 80%, while dye substance removal rates surpass 70%. However, the overall desalination rate is relatively low.

4.2. Ultrafiltration / Nanofiltration

The combination process of UF and NF as a purely physical process significantly reduces the dosage of disinfectants and partially solves the problem of DBPs. Li’s preliminary study on Taihu Lake water utilized the UF-NF dual membrane combination process [9]. A 13-month continuous on-site pilot study demonstrated the stable operation of the UF-NF combination process.

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Li investigated the removal efficiency of various processes on UV254. Humic substances in surface water mainly represent UV254, and their value is not only related to the dissolved organic carbon (DOC) content in water but also has a good correlation with color and DBPs. The research showed that the conventional process achieved a removal rate of around 60% for UV254. At the same time, direct filtration through UF had a deplorable removal effect, with a removal rate of around 50%. The combination process mainly relies on the NF membrane to remove UV254, and the overall removal rate of UV254 by the entire process is above 90%, reducing the UV254 content in the raw water to below 0.01cm-1. The high turbidity removal rate of UF creates favorable inlet conditions for NF. The combined process of UF-NF ultimately results in improved effluent turbidity, average CODMn, and average DOC compared to single membrane data. From the results, it can be seen that the UF-NF combination process has a good removal effect on organic matter in water.

The study also found that in the experiment of removing odor compounds using the combination process, UF-NF exhibited effective removal efficiency for 2-methylisoborneol and geosmin, with a removal rate of approximately 77.2%. The combination process can significantly reduce the problem of water odor caused by odor compounds during summer algal blooms in Taihu Lake, and it also reduces the potential formation of trichloromethane in water, further ensuring the safety of water quality.

4.3. Reverse osmosis / Nanofiltration

Xie et al. [10] addressed the issue of organic matter that was difficult to remove in a water treatment plant in Zhejiang Province by using an NF system with a treatment scale of 12 m3/h. They also investigated the feasibility of using RO membrane to treat the NF concentrate and increase the system's water production rate. Four types of NF membranes were studied, and the results showed that the CODMn of the produced water by all four types of NF membranes was stably below 0.45 mg/L, with an average removal rate of over 78.0%. The average removal rate of TOC was approximately 90.0%, while the produced water UV254 consistently remained below 0.004 cm-1, with an average removal rate of around 98.5%. Furthermore, using O3/BAC can eliminate unoxidized or unabsorbed organic matter. By treating the NF concentrate with the RO membrane, the desalination rate was over 98%, and the water production rate of the NF system was increased from 90% to 97.5% while ensuring that the water quality met the requirements. The system's effluent did not detect any of the 16 disinfection by-products specified in the "Drinking Water Hygiene Standards" (GB 5749-2022), and the removal rates of trichloromethane and haloacetic acid formation potential were above 89%.

5. Conclusion

This paper focuses on dual membrane filtration technology and comprehensively reviews the removal of natural organic matter (NOMs) and DBPs in various water treatment processes. The primary emphasis is placed on evaluating the effectiveness and current application status of different removal methods while exploring their feasibility and sustainability.

Studies show the toxic effects of by-products and a link between chlorinated drinking water and cancer. Removing by-products from water is a crucial research area. According to the literature review, RO, UF, and NF can all, to some extent, remove DBPs from drinking water. In practical applications for removing DBPs from drinking water, the dual-membrane method often utilizes NF as the primary technique. In the combination process of MF and NF, MF can effectively remove insoluble substances in water, reduce membrane fouling in the NF system, and provide a long filter lifespan, stable filtration precision, and low investment cost. In the combination process of UF and NF, the effluent from UF is comparable to that of conventional processes in terms of water quality indicators, and subsequent NF treatment can further remove small impurities in water. The dual-membrane process can effectively remove organic matter from water, with removal rates for CODMn, DOC, and UV254 all exceeding 80% while reducing the potential formation of trihalomethanes, thus ensuring water quality safety. In the NF-RO combination process, treating NF concentrate with a RO membrane can increase the water recovery rate from 90% to 97.5%. Additionally, it can effectively remove soluble microbial metabolites and fluorescent organic compounds, such as humic acids, thereby reducing the risk of disinfection by-product formation. In the case of treating concentrate, NF-RO may be a preferable choice.

Although dual membrane filtration technology has demonstrated favorable performance in removing DBPs, there are still limitations and areas for future improvement. Future studies should focus on optimizing the performance of dual membrane systems, exploring novel membrane materials, and developing advanced pre-treatment strategies to enhance DBP removal efficiency. Furthermore, addressing the long-term stability and fouling control of dual membrane systems is necessary to ensure their sustainable operation. With ongoing research and development, dual membrane filtration technology holds significant potential for improving water quality and addressing the challenges associated with DBP removal in water treatment processes.

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