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나노여과 기반 용량성 탈이온화의 진전

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Progress in Nanofiltration-Based Capacitive Deionization

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요 약: 최근 연구는 역삼투압(RO), 나노여과(NF) 및 전기투석(ED)과 같은 막 공정에서 고급 용량성 탈이온화(CDI) 및 막 변형(MCDI)을 포함하는 광범위한 담수화 및 수처리 방법을 탐구합니다. 비교 분석은 저염도 시나리오에서 ED의 비용 효율성을 보여주는 반면 하이브리드 시스템(NF-MCDI, RO-NF-MCDI)은 향상된 염 제거 및 에너지 효율성을 보여줍니다. 새로운 이온 분리 방법(NF-CDI, NF-FCDI)은 향상된 효율성과 에너지 절감을 제공합니다. 이러한 연구는 또한 다양한 산업에 특정한 복잡한 폐수를 처리하는 데 있어 이러한 방법의 효율성을 강조합니다. 환경 영향 평가는 시스템 선택의 지속 가능성의 필요성을 강조합니다. 또한 마이크로 제작된 센서를 멤브레인에 통합하면 실시간 모니터링이 가능하여 기술 개발이 진전됩니다. 이러한 연구는 새로운 담수화 및 수처리 기술의 다양성과 가능성을 강조합니다. 이는 효율성 향상, 에너지 사용 최소화, 산업별 문제 해결 및 기존 방법 한계를 능가하는 혁신을 위한 귀중한 통찰력을 제공합니다. 다양한 응용 분야에서 효율성 향상, 환경 영향 최소화 및 적응성 보장에 초점을 맞춘 지속적인 발전으로 지속 가능한 수처리의 미래는 밝습니다.

Abstract: Recent studies explore a wide array of desalination and water treatment methods, encompassing membrane processes such as reverse osmosis (RO), nanofiltration (NF), and electrodialysis (ED) to advanced capacitive deionization (CDI) and its membrane variant (MCDI). Comparative analyses reveal ED's cost-effectiveness in low-salinity scenarios, while hybrid systems (NF-MCDI, RO-NF-MCDI) show improved salt removal and energy efficiency. Novel ion separation methods (NF-CDI, NF-FCDI) offer enhanced efficacy and energy savings. These studies also highlight the efficiency of these methods in treating complex wastewater specific to various industries. Environmental impact assessments emphasize the need for sustainability in system selection. Additionally, the integration of microfabricated sensors into membranes allows real-time monitoring, advancing technology development. These studies underscore the variety and promise of emerging desalination and water treatment technologies. They provide valuable insights for enhancing efficiency, minimizing energy usage, tackling industry-specific issues, and innovating to surpass conventional method limitations. The future of sustainable water treatment appears bright, with continual advancements focused on improving efficiency, minimizing environmental impact, and ensuring adaptability across diverse applications.

Keywords: *reverse osmosis (RO), nanofiltration (NF), electrodialysis (ED), capacitive deionization (CDI), MCDI*

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1. Introduction

The escalating demands for water resources, driven by population growth, urbanization, and industrial expansion, have created an imbalance between supply and demand. This challenge is further complicated by the ramifications of climate change, intensifying the ongoing threat of widespread water stress and scarcity. Urgent and sustainable solutions are essential to navigate these critical issues[1-6]. This study highlights the central roles of wastewater treatment and desalination in preventing the depletion of freshwater resources. Desalination, particularly, emerges as a key strategy to ease water stress by converting seawater, saline groundwater (SGW), drainage water, and treated wastewater into fresh water. The utilization of membrane technology in these processes has gained significant attention, offering promising avenues to meet the growing global demand for clean drinking water and foster economic growth[7].

This review aims to critically evaluates recent advancements in membrane-based desalination technologies, focusing on the evolution of membrane materials and innovative approaches like nanomaterial incorporation and surface modifications. Additionally, it focuses into emerging membrane-based separation techniques, particularly emphasizing membrane capacitive deionization (MCDI) and its integration into membrane systems. While traditional pressure-driven membrane technologies like reverse osmosis (RO) have long been recognized for their effectiveness, ongoing advancements in membrane materials and advanced separation processes, especially integrated with nanofiltration (NF) membrane, are reshaping industrial waste water treatment scenarios[8-10].

An innovative hybrid approach, the fusion of capacitive deionization (CDI) with NF in FCDI-NF systems, has recently garnered attention for its potential in energy-efficient desalination. Studies have indicated a 16% reduction in energy consumption compared to traditional BWRO desalination units, achieving stringent drinking water standards. However, practical implementation and feasibility assessments of NF-MCDI hybrid systems remain limited, necessitating deeper exploration of their operational viability[11-13]. This study aims to bridge this gap by proposing an energy-efficient NF-MCDI hybrid desalination system and determining optimal operational conditions for commercially available NF and CDI modules. The primary objective is to surpass conventional RO processes in specific energy requirements while meeting strict drinking water standards. In conclusion, this comprehensive review and experimental study serve as a crucial step in harnessing membrane-based technologies, especially NF-MCDI hybrid systems, to address the imminent global challenge of water stress and scarcity.

2. Capacitive Deionization

MCDI is a promising technology for brackish water treatment, yet it is often compared to pressure-driven methods like RO and NF, leaving out electrodialysis (ED), which is renowned for its cost-effectiveness in low salinity brackish water desalination[14]. To address this gap, this study directly compares MCDI and ED in the desalination of model sodium chloride solutions with concentrations ranging from 0.7 to 1.0 g/dm^3. The study shows that MCDI consumes notably more energy than ED, especially when generating 0.5 g/dm^3 of product. MCDI's energy consumption ranged from 0.428 to 0.499 kWh/m^3, while ED demonstrated greater energy efficiency, requiring only 0.034 to 0.230 kWh/m^3. This difference varied depending on the feed concentration and the specific membrane used. Typically, MCDI has been evaluated against pressure-driven techniques, but this comparison underscores the cost-effectiveness of ED in low-salinity brackish water desalination. ED offers numerous advantages over (M)CDI, including its effectiveness in handling scaling-prone waters and well-documented fouling mechanisms of porous carbon electrodes. Moreover, ED allows for straightforward module capacity expansion, a feature less achievable in (M)CDI. The long-term performance of installations also remains a consideration. While some suggest that the frequent polarity changes in MCDI might prolong membrane life compared to ED, the endurance of porous carbon electrodes in (M)CDI is still an area requiring further research. An advantage for MCDI lies in the absence of electrode reactions, a common occurrence in ED, where hydrogen, oxygen, and sometimes chlorine are evolved at electrodes, particularly under high voltage conditions.

3. Removal of Monovalent Ion

Biro *et al.* explored the use of MCDI to treat both model water samples and real wastewater from the textile industry[15]. To prevent membrane and electrode scaling and fouling during MCDI, nanofiltration was integrated as a pre-treatment for wastewater. Various conditions were tested in MCDI to optimize desalination efficiency and reduce conductivity. The research effectively lowered the conductivity of high-salinity wastewater to the target level (below 1.5 mS cm^{-1}), accomplishing desalination rates of up to 95%. The efficiency of the desalination process was influenced by the initial conductivity and the specific ion composition present in the water samples. Furthermore, the study utilized chemometric analysis to investigate relationships among several factors, such as the presence of ions (Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, Br^-, F^-, SO_4^{2-}, NO_3^-), desalination efficiency, water recovery, phase duration, and solution flow rate. By employing multiple linear regression, a predictive model for desalination rates was created, closely aligning with experimental results, and exhibiting differences of less than 1%. The research demonstrates the successful application of MCDI for the treatment of high-salt wastewater, achieving significant desalination rates.

Jeong *et al.* highlights an innovative approach to address the energy-intensive nature of typical RO systems when treating brackish water[16]. It introduces a novel hybrid system that combines NF and MCDI to enhance both salt removal efficiency and energy efficiency. The research evaluated the NF-MCDI hybrid system's performance through a comprehensive series of experiments and modeling conducted across diverse operational scenarios. This investigation meticulously examined the influence of critical factors, including feed concentration, NF recovery rate, and MCDI flow rate, on both salt removal efficiency and energy consumption. Remarkably, the research findings revealed that NF in isolation proved inadequate for substantial salt removal in saline water. However, when integrated with MCDI, the system demonstrated a remarkable enhancement, achieving an impressive 95% salt removal rate. Moreover, the investigation unveiled an optimized setup for the NF-MCDI system, showcasing exceptional effectiveness, especially when handling feed concentrations of below 10 g/L. This finely tuned configuration not only outperformed traditional RO systems in terms of energy efficiency but also met the rigorous criteria for drinking water standards, maintaining salt concentrations at or below 0.5 g/L. In summary, the NF-MCDI hybrid system presents a promising solution to address the energy-intensive issues related to brackish water treatment. Utilizing the synergies between NF and MCDI, this innovative system not only achieves outstanding salt removal capabilities but also significantly reduces energy consumption.

The semiconductor industry's rapid growth has raised environmental concerns, particularly regarding freshwater usage and the generation of hazardous wastewater. Semiconductor wastewater often contains the toxic compound Tetramethylammonium hydroxide (TMAH), requiring effective removal methods before discharge [17](Fig. 1 and 2).

However, affordable TMAH removal technologies are scarce. Consequently, this study sought to assess and compare the suitability of MCDI, RO, and NF for treating semiconductor wastewater containing TMAH. Bench-scale experiments were conducted to evaluate the removal efficiencies of TMAH, total dissolved solids (TDS), and total organic carbon (TOC) in semiconductor wastewater. The results highlighted that both MCDI and RO were highly effective in removing TMAH, surpassing NF when operating under the same

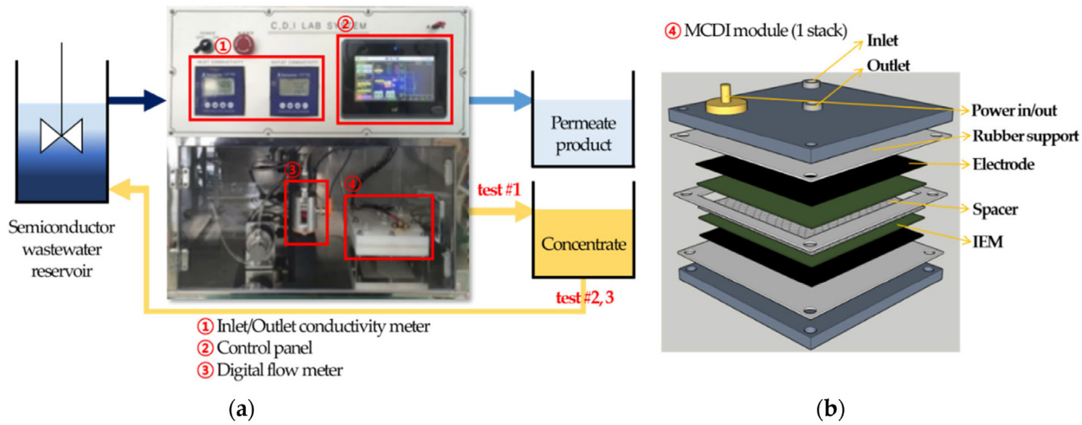

Fig. 1. Schematic diagram of lab-scale processes: (a) MCDI test procedure scheme; (b) Detailed design of ion-exchange module (Reproduced with permission from Lee *et al.*[17], Copyright 2023, MDPI).

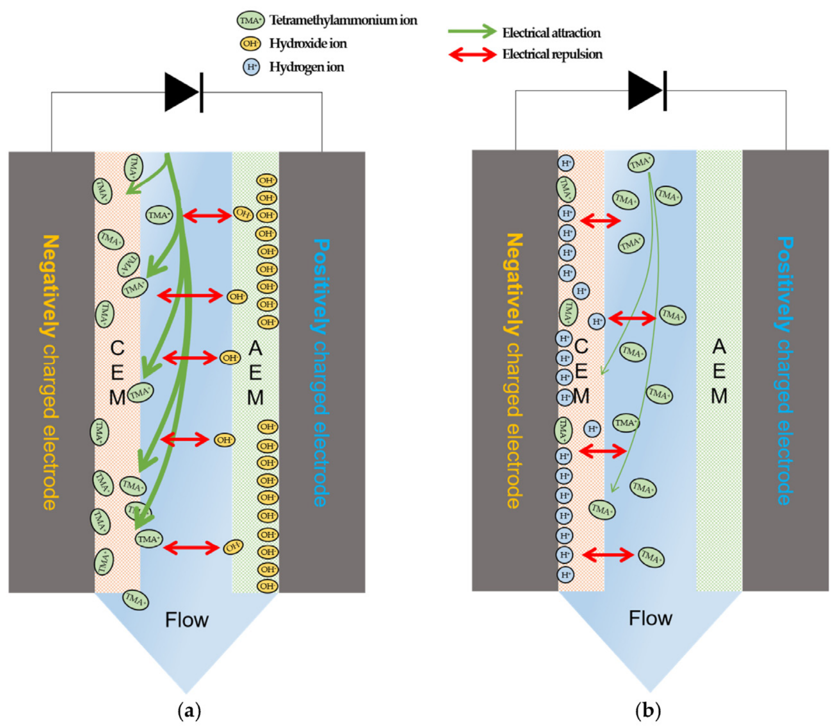

Fig. 2. Electrosorption mechanism model of TMA ions in the MCDI process according to the solution pH: (a) High pH conditions (pH 10); (b) Low pH conditions (pH 3) (Reproduced with permission from Lee *et al.*[17], Copyright 2023, MDPI).

recovery conditions. MCDI exhibited superior performance in eliminating monovalent ions such as TMA+ in comparison to divalent ions. Additionally, it was observed that MCDI achieved better TMA+ removal rates in basic solutions, showcasing its potential for treating semiconductor wastewater containing TMAH. This research compared the efficiency of RO, NF, and MCDI processes in removing TMAH from semiconductor wastewater. NF proved inadequate due to its limited molecular weight cut-off for TMAH treatment. RO effectively removed TDS (~23.9 mg/L) and TMAH (~3.0 mg/L) but only accounted for 56% of TOC removal. MCDI demonstrated significant removal capabilities for TDS, TOC, and TMAH; MCDI excelled in removing monovalent ions like TMA ions, outperforming multivalent ions due to their ionic hydrate radii. MCDI's TMAH removal efficiency improved with pH adjustment, particularly in higher pH semiconductor wastewater.

Mao *et al.* did integration of NF membranes within CDI cells to desalinate both mono- and divalent salt solutions [18]. This concept was theoretically and experimentally investigated and introduces a novel method for separating mono- and divalent ions. Effective separation relies on the variance in diffusion coefficients among these ions. When combined with the suitable adsorption period (1.5 hours), it becomes feasible to enhance the concentration ratio of NaCl to $MgSO_4$ threefold, even when initially dealing with an equal mixture of the two salts. In this proof-of-principle work, the combination of CDI and NF membranes is proposed for mono- and divalent ion fractionation. Both experimental and theoretical findings demonstrate that NF membranes significantly impede ion migration, especially divalent ions, owing to their lower diffusion coefficient values within the membrane. This mechanism can be effectively leveraged to separate NaCl and $MgSO_4$ from an initial 1:1 mixture of these salts, with an optimal adsorption duration of 1.5 hours. During desorption, there was a remarkable threefold increase in the Cl^-/SO_4^{2-} ratio, and further enhancements are possible with improved desorption times. Notably, NFCDI exhibits superior energy efficiency, consuming approximately 0.2 kWh/m^3, and displays enhanced fractionation capabilities when compared to NF operating in isolation.

Nativ *et al.* presents the selective separation of monovalent (Na^+) and divalent (Mg^{2+}) cations, as well as anions (Cl^- and SO_4^{2-}), from aqueous solutions through the Flow Electrode Capacitive[19]. Deionization (FCDI) process. By employing nanofiltration (NF) and ion-exchange membranes in the FCDI module, the researchers investigated various settings, including different applied cell potentials (0.6, 0.8, and 1.23 V) and initial molar concentration ratios (1, 10, and 20). The study found that the NF270 membrane had limited success in effectively separating sodium and magnesium, as reflected by permselectivity values ranging from 0.69 to 1.04. In contrast, when it came to separating Cl^- and SO_4^{2-}, the NF-FCDI method demonstrated significant potential, with permselectivity values varying from 1.28 to 7.03. This suggests that the NF-FCDI approach is particularly promising for selectively removing anions from the solution. The NF-FCDI process proved highly efficient in separating chloride and sulfate ions. However, it was less successful in achieving selectivity for sodium and magnesium separation using the NF270 membrane. Further investigation is required to understand why cation separation efficiencies differ between NF-based electrochemical and pressure-driven processes.

Rosentreter *et al.* explores the partial desalination of SGW using various membrane desalination technologies and MCDI[20](Fig. 3).

SGW typically contains elevated concentrations of sodium, chloride, sulfate, and nitrate, often surpassing recommended levels for safe drinking and irrigation. This study assessed the desalination capabilities of NF, brackish water reverse osmosis (BWRO), seawater reverse osmosis (SWRO), and MCDI. The evaluation focused on specific energy consumption (SEC) and water recovery rates. The experimental findings indicated that for SGW with a TDS concentration of 1 g/L, both MCDI and NF demonstrated similar SEC values, falling within the range of 0.2 to 0.4 kWh/m³, and achieving water recovery rates between 35% and 70%. However, at higher TDS concentrations ($\geq$ 2 g/L), BWRO and SWRO exhibited the lowest SEC values, ranging from 0.4 to 2.9 kWh/m³, with water recovery rates spanning 40% to 66%. Although MCDI didn't match the competitiveness of pressure- driven membrane desalination technologies at higher salinity levels, it showcased remarkable selectivity for nitrate and displayed substantial potential for versatile desalination applications. The desalination technology's effectiveness depended on the operational approach and its selectivity towards specific ions (Na^+, Cl^-, NO_3^-, and SO_4^{2-}). When aiming for low salt rejection, both MCDI and NF performed admirably, with MCDI ach-

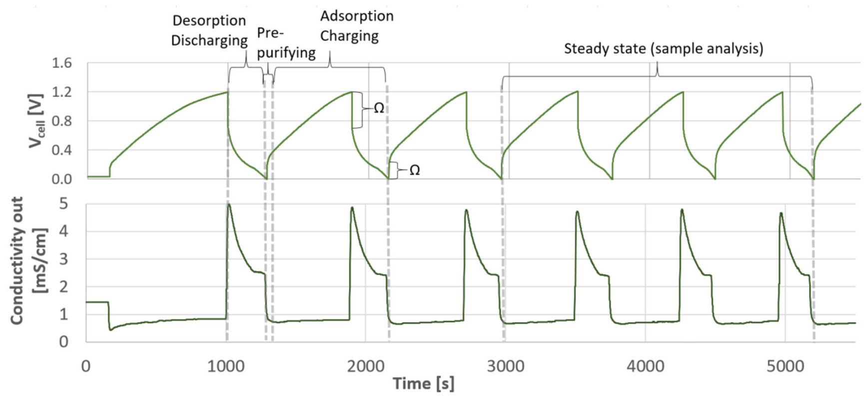

Fig. 3. Cell voltage (V_{cell}) and conductivity of the outflow of the MCDI module during desalination of slightly SGW at 14 A (Ω: ohmic resistance) (Reproduced with permission from Rosentreter *et al.*[20], Copyright 2021, MDPI).

ieving the lowest SEC values for slightly SGW (TDS = 1 g/L) within the recommended concentrations for irrigation and drinking water. However, when higher salt rejection was essential, NF surpassed MCDI due to its superior water recovery capability. BWRO and SWRO consistently exhibited excellent desalination performance for TDS concentrations (≥ 2 g/L). Notably, MCDI excelled in treating NO^{3-} rich groundwater and proved adaptable to varying operating conditions, underscoring its versatility and potential.

Zhang *et al.* addresses the critical need for water reuse and salt resource recycling in high-salt wastewater from the coal chemical industry, aiming for sustainable development[21]. Traditional methods like the RO-NF-crystallization process consume significant energy, especially during heat-driven crystallization. In response, this study proposes an innovative solution: the RO-NF-MCDI system, designed for salt classification and recovery from coal chemical industrial wastewater. In this system, sludge-derived biochar serves as the MCDI electrode, providing a stable desalination capacity for source water with NaCl concentrations up to 300 mM, equivalent to a TDS level of 18,000 mg · L^{-1}. This capability makes it well-suited for desalination following RO-NF salt separation. This research also presents a two-channel architecture within the MCDI system. This design allows for uninterrupted operation by seamlessly transitioning the inlet and outlet between the concentration and desalination channels during the charging and discharging phases. This development underscores the practicality and potential of the RO-NF-MCDI system for effectively classifying and recovering salt from coal chemical industrial wastewater. The biochar electrode derived from sludge demonstrates remarkable stability, while the MCDI system efficiently desalinates solutions containing both monovalent and divalent ions. The inventive two-channel design enables concurrent desalination and concentration processes, thereby improving the overall efficiency of the system. The introduced RO-NF-MCDI system, utilizing sludge-derived biochar and a two-channel design, presents a promising avenue for salt classification and recovery in coal chemical industrial wastewater.

4. Removal of Divalent Ion

Cetinkaya *et al.* focused on copper ion (Cu^{2+}) removal, by using NF membranes and a MCDI system[22]. The research revealed that the most effective Cu^{2+} removal (99%) occurred when these two systems worked together in an integrated setup. MCDI is a purification method employing ion exchange membranes and an electric field between opposing electrodes. The optimal conditions for Cu^{2+} ion removal were found to be a flow rate of 50 mL/min, a direct current voltage of 1.2 V, and an operation time of 15 minutes. An environmental evaluation of the membrane system was conducted using a life cycle assessment (LCA) method to analyze its impact under different operational scenarios. A sensitivity analysis was performed, comparing the environmental effects of various materials employed in both the membrane and MCDI systems. The LCA findings underscored that the MCDI system exhibited notably greater environmental consequences in all aspects, with particular emphasis on material and energy-related impacts. This research emphasizes the importance of conducting a comprehensive system analysis when choosing wastewater treatment methods, considering both environmental and cost aspects. The NF&MCDI system's potential for Cu^{2+} ion removal was explored under different conditions, addressing associated challenges. Although the integrated NF&MCDI system demonstrated superior Cu^{2+} ion removal, its LCA results indicated slightly elevated overall environmental impacts, mainly attributed to electricity usage. To improve the integrated system's environmental performance, efforts should be directed towards reducing electricity consumption. As the integrated NF&MCDI system expands in scale, future research should prioritize the optimization of cell voltage distribution.

Zhang *et al.* presents a novel approach to tackle the challenges posed by concentration polarization in membrane separation processes through the use of microfabricated capacitors to monitor concentration polarization in a cross-flow NF membrane system[23]. Concentration polarization boundary layers (CPBL) often lead to decreased permeate flux and an increased risk of membrane fouling. In this research, microfabricated interdigitated capacitors were developed for in-situ monitoring of CPBL growth in a cross-flow NF membrane system, using calcium sulfate ($CaSO_4$) solution as the feed. These capacitive microsensors respond to changes in solute concentration at varying distances from the membrane surface. The microcapacitor sensors allowed real-time characterization of both transient and steady-state concentration boundary layer polarization behavior. Importantly, these measurements aligned well with theoretical predictions from concentration polarization models. Furthermore, by combining microcapacitor sensors with ultrasonic time-domain reflectometry (UTDR), the study enabled real-time observation of the entire process, from the initiation of concentration polarization to the onset of membrane fouling. And also allows for comprehensive monitoring of the membrane system's response from CP initiation to scaling layer growth. This multisensor approach proves advantageous for understanding membrane phenomena and offers potential for the development of “smart” membranes.

5. Conclusions

In reviewing diverse studies on desalination and water treatment technologies, it's evident that this field is rapidly advancing with various innovative approaches. Membrane-based processes like RO, NF, ED, and emerging methods like CDI and MCDI showcase a spectrum of methodologies. Comparing desalination methods reveals their strengths and weaknesses. While MCDI shows potential in specific scenarios, ED stands out for its cost-effectiveness and robustness, especially in low-salinity desalination. Hybrid systems like NF-MCDI and RO-NF-MCDI combinations demonstrate synergistic benefits, enhancing salt removal and reducing energy consumption. Studies targeting specific industries, such as semiconductor or coal chemical, demonstrate the adaptability of these methods for treating complex industrial wastewater, focusing on pollutant removal like TMAH. Life cycle assessments emphasize the importance of considering environmental consequences in designing wastewater treatment systems. Integration of microfabricated capacitors and sensors into membrane systems allows real-time monitoring, aiding in understanding membrane behavior for more responsive technologies. These studies highlight the potential of emerging desalination and water treatment technologies, offering insights into efficiency, energy reduction, industry-specific challenges, and novel approaches. With ongoing advancements, the future of sustainable water treatment looks promising, aiming to enhance efficiency, environmental impact, and adaptability across various applications.

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