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Investigations of Nanomaterial-Based Membranes for Efficient Removal of Contaminants from Wastewater via Membrane Distillation: A Critical Review

ABSTRACT

The requirement for wastewater treatment is paramount in ensuring environmental sustainability and safeguarding public health. As industrialization and urbanization accelerate, the volume of wastewater generated continues to increase, containing a diverse range of pollutants and contaminants. Untreated wastewater poses serious threats to ecosystems, water bodies, and human communities, leading to pollution, waterborne diseases, and ecological imbalances. Effective wastewater treatment becomes essential to mitigate these adverse effects by removing or reducing pollutants before discharge into natural water sources. This process helps to preserve water quality, protect aquatic life, and maintain the overall health of ecosystems. Membrane distillation (MD) has emerged as a promising technology for wastewater treatment, offering an innovative approach to address the challenges associated with conventional treatment methods. In MD, a hydrophobic membrane serves as a selective barrier, allowing water vapor to pass through while preventing the passage of contaminants. This paper offers an extensive overview of the latest advancements in nanotechnology and membrane distillation applied in wastewater treatment. We will delve into different types of nanomaterials that have been used to enhance the properties of MD membranes, such as nanocomposites, nanoparticles, and nanofiber membranes. We also explore the mechanisms by which these nanomaterials improve the separation efficiency, anti-fouling properties, and durability of MD membranes. Additionally, we highlight the potential of hybrid membranes that combine different types of nanomaterials for further improving the performance of MD in wastewater treatment. We provide examples of recent studies that have investigated the use of hybrid membranes, including carbon nanotube-graphene oxide hybrid membranes, nanocomposite nanofiber membranes, and silver nanoparticle-embedded membranes. We also identify some areas for future research and development, such as the scale-up and commercialization of nanotechnology-based MD systems. In summary, this review paper highlights the potential of nanotechnology to enhance the performance of MD in wastewater treatment, leading to improved water quality and a cleaner environment.

Keywords: *Nanocomposite; Nanofibers; Wastewater treatment; Membrane Distillation*

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Graphical Abstract:

INTRODUCTION

Indeed, chemical contamination of water has gained significant attention from the public sector, society, and, most significantly, the whole industrialized globe [1]. There are two main sources, point and non-point. The initial description pertains to pollutants originating from a singular origin, such as industrial emissions directly entering water bodies. On the other hand, the second refers to pollutants discharged from various sources. Water pollution stems from various contributors, including energy consumption, radioactive waste, urban expansion, sewage discharge and treatment, industrial effluents, mining operations, as well as the use of pesticides and chemical fertilizers. Every activity that uses water, whether it be residential, agricultural, or industrial, will cause pollution since the effluent will contain undesired contaminants, some of which may be dangerous [2]. Nanotechnology has great potential for improving the efficiency and effectiveness of wastewater treatment. It involves the use of nanoscale materials and processes to remove contaminants from wastewater [1,2]. By utilizing membranes with nanoscale pores, Nanofiltration and Reverse Osmosis processes offer efficient and effective treatment solutions, ensuring the production of clean and safe water for various applications. Nanofiltration membranes can remove particles down to 1 nm in size, while reverse osmosis membranes can remove particles down to 0.1 nm in size [3,4]. Nanoparticles can also be used to adsorb contaminants from wastewater. Overall, the use of carbon nanotubes, graphene oxide, and metal oxide nanoparticles in wastewater treatment holds great promise for addressing the global water pollution crisis. These advanced materials, with their unique properties and exceptional pollutant removal capabilities, offer a sustainable and efficient solution for the purification of wastewater [5-7]. Nanoparticles can also be used as photocatalysts to break down contaminants in wastewater. Titanium dioxide nanoparticles are commonly used for this purpose, as they can be activated by UV light to generate reactive oxygen species that can degrade organic compounds [8]. Another one is Nanobubble Technology, according to which nanobubbles are small bubbles (less than 100 nm in diameter) that can be used to enhance aeration and mixing in wastewater treatment. They can also help to remove contaminants by attaching to them and carrying them to the surface of the water [9]. Figure 1 presents some of the technologies employed in water treatment, where nanoparticle and nanotechnology are extensively used.

Figure 1: Innovations of Nanotechnology in water treatment

Nanotechnology and membrane distillation

By leveraging the unique properties of nanomaterials, researchers can develop advanced membranes with improved flux rates, higher selectivity, and reduced fouling. Furthermore, nanotechnology enables the integration of membrane distillation with other treatment technologies, opening up new possibilities for more efficient and sustainable wastewater treatment processes. Membrane distillation entails a separation technique employing a hydrophobic membrane to isolate impurities from water via vaporization and subsequent condensation. The simplest and mostly employed configuration of MD is Direct Contact Membrane Distillation (DCMD). Figure 2 presents the DCMD (simplest configuration). Top of FormThe process operates at low pressure and low temperature, making it energy-efficient and suitable for treating wastewater [10]. Recent Researches reported some ways that nanotechnology can be used to improve membrane distillation for wastewater treatment [11-14]:

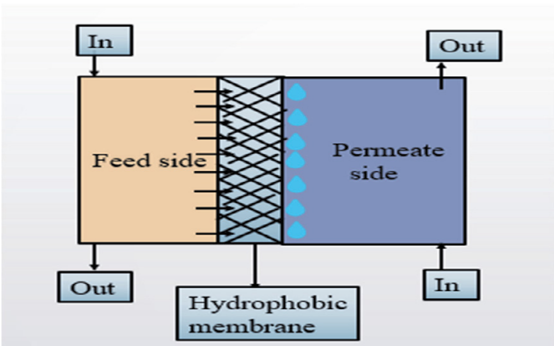

Figure 2: Schematic diagram of DCMD Configuration

- **Nanocomposite Membranes**: Membranes made of nanocomposites can have improved hydrophobicity and stability, leading to better separation efficiency and durability. For example, adding carbon nanotubes or graphene oxide to the membrane matrix can enhance its thermal conductivity and make it more resistant to fouling.
- **Nanofiber Membranes**:

Top of Form

The extensive surface area of electrospun nanofiber membranes offers benefits across different fields such as filtration, tissue engineering, and energy storage applications. In filtration applications, the large surface area allows for efficient capture of particles and contaminants, enabling improved separation efficiency. The narrow pore size distribution of these membranes further enhances their filtration capabilities, as it restricts the passage of larger particles while allowing smaller molecules to permeate through. These nanofiber membranes can be functionalized with nanoparticles or nanocomposites to enhance their hydrophobicity and anti-fouling properties. Here are some examples: Polyvinylidene fluoride (PVDF) nanofiber membrane, Polyacrylonitrile (PAN) nanofiber membrane, Polysulfone (PSf) nanofiber membrane etc.

- **Hybrid Membranes**: Hybrid membranes that incorporate different types of nanomaterials, such as carbon nanotubes and graphene oxide, offer numerous advantages over traditional single-component membranes. These membranes demonstrate superior separation efficiency, anti-fouling properties, thermal stability, and mechanical strength. With their potential for enhanced performance in various applications, hybrid membranes are poised to revolutionize the field of membrane-based separations [15]. Nuthenya et al. synthesized PVDF membrane embedded with SiO_2 nanoparticles for MD. They found that the embedded nanoparticles enhanced the hydrophobicity of the membrane surface, resulting in higher water vapor flux and reduced fouling [16]. Zhang et al. (2018) reported that a PVDF membrane embedded with TiO_2 nanoparticles exhibited excellent thermal stability and anti-fouling properties [17]. In addition, various studies have investigated the effect of different types of nanoparticles on MD performance. For instance, Alkhouzaam et al. investigated the use of graphene oxide (GO) nanoparticles embedded in a PVDF membrane for MD. They found that the embedded GO nanoparticles improved the membrane's hydrophobicity and thermal stability, resulting in higher water vapor flux and reduced fouling [18].

Wastewater Treatment via Membrane Distillation

Sangeetha (2022) conducted a study investigating the effectiveness of dual-layered omni phobic electrospun nanofibrous membranes (ENFMs) within the realm of direct contact membrane distillation (DCMD). These dual-layered membranes were created using polyvinylidene fluoride (PVDF) and a fluoropolymer known as perfluorohexane (PFH) through the process of electrospinning. The study included testing the dual-layered ENFMs with simulated seawater, and the results showcased their exceptional vapor performance. Being tested with simulated seawater, the dual-layered ENFMs had outstanding vapor flux of 8.5 L/m^2h and salt rejection of greater than 99.99%. They also revealed a high porosity and water contact angle (WCA) of 143˚. When tested with synthetic wastewater containing pharmaceutical substances, the constructed ENFMs (Electrospun Nanofiber Membranes) exhibited excellent permeate quality. The permeate showed a low organic content of 1 ppm (parts per million) and had a conductivity of 30 S/cm (siemens per centimeter). These results indicate that the ENFMs are highly effective in removing pharmaceutical substances from wastewater [19]. Jingjee Ju's (2020) study investigated the fabrication and performance of hierarchically structured superhydrophobic polytetrafluoroethylene (PTFE)/polyhedral oligomeric silsesquioxane (POSS) nanofibrous membranes for MD. The membrane fabrication procedure included electrospinning, succeeded by surface modification employing a fluorinated silane agent. The main focus of the study was to optimize the PTFE/POSS nanofibrous membrane, specifically the membrane labeled as #POSS-2. This particular membrane exhibited a remarkable three-dimensional (3D) superhydrophobic property, with a WCA measured at 151 ± 4°. The vinyl-POSS concentration was varied to identify the optimal concentration that would result in the desired membrane performance. Similarly, different temperatures and salt concentrations were tested to understand their influence on the membrane's performance. The study conducted on #POSS-2 in the field of membrane distillation has yielded promising results. Specifically, it has shown a remarkably competitive water flux of 40 ± 2 L/m^2h during the DCMD process. This achievement was obtained when the feed temperature was maintained at 60 °C, while the permeate temperature was kept at 20 °C.

Alongside its notable water flux performance, #POSS-2 has showcased exceptional long-term stability throughout a continuous DCMD operation lasting for 200 hours.Top of Form This indicates that the material is capable of maintaining its effectiveness and efficiency over an extended period of operation [20]. In their study, Hadi Attia et al. (2017) documented the effectiveness of a superhydrophobic electrospun membrane for removing heavy metals through Air Gap Membrane Distillation (AGMD). The membrane was prepared by combination of electrospinning technique and surface modification with a fluorinated silane agent. The research conducted revealed important findings regarding the manufacturing of an immaculate beadless membrane mat. Through experimentation, it was determined that a polymer concentration of 15 wt.%, combined with a cationic surfactant concentration of 0.05 wt.%, produced the desired results. In terms of the solvent mixture, a ratio of 6:4 of dimethylformamide (DMF) to acetone was found to be optimal for the membrane mat production. Additionally, an electric field strength of 14 KV was applied during the manufacturing process. The performance of the membrane mat was evaluated based on various parameters. The lead rejection rate achieved was 72.77%, indicating the membrane's ability to effectively remove lead from the solution. The liquid entry pressure was measured to be 17 psi, suggesting the membrane's ability to withstand pressure differentials and prevent liquid penetration. Furthermore, the WCA was determined to be 132°, indicating the hydrophobic nature of the membrane. This property is significant as it allows for the efficient separation of water from other substances. Composite PVDF membranes containing 11 wt.% and 20 wt.% functionalized alumina (Al_2O_3) membranes performed better than composite PVDF membranes with 18° WCA and 27 psi liquid entry pressure, with 99.36% heavy metal rejection and 5.9% higher permeate flow [21]. Jiaxin Cui et al. (2020) focuses on the application of electrospun nanofiber membranes for wastewater treatment. Electrospun nanofiber membranes exhibited high porosity and specific surface area, providing an efficient platform for various filtration and separation processes. The nanofiber membranes showed excellent mechanical strength and stability, which can be further improved by optimizing the electrospinning parameters such as polymer concentration and solvent type. The performance of electrospun nanofiber membranes can be enhanced by incorporating functional materials such as nanoparticles and biopolymers. Electrospun nanofiber membranes demonstrated superior adsorption and removal efficiency for various pollutants in wastewater, including heavy metals, dyes, and organic compounds. The application of electrospun nanofiber membranes in wastewater treatment can significantly improve the efficiency and reduce the cost compared to conventional technologies. Overall, the study provides quantitative evidence for the potential of electrospun nanofiber membranes as an efficient and cost-effective platform for wastewater treatment [22]. Mohamed Shaban (2015) aimed to investigate the effect of incorporating titanium dioxide (TiO_2) nanotubes into a mixed matrix polyethersulfone (PES) membrane on its performance in terms of water flux and rejection of various pollutants. The VMD mathematical model was used to calculate the best membrane performance under optimal conditions. The experimental findings showed that by employing 0.53% TiO_2NTs in the membrane preparation solution, it was able to generate a high-performance NF/RO mix membrane with a salt rejection percentage of 97% and a permeate flow of 18.2 L/m^{2}h [23]. Junghyun Kim et al. (2022) presented a novel membrane modification method using multi-layered single-wall carbon nanotubes (SWCNTs) and polyvinylidene fluoride (PVDF) to improve the performance of membrane distillation (MD). The research aimed to explore how electrical repulsion influences membrane performance and fouling prevention. Quantitative findings revealed that among the membranes tested, the SWCNT/PVDF membrane with the greatest number of SWCNT layers (10 layers) demonstrated the highest water flux (up to 43 L/m² h) and the lowest contact angle (29°).The fouling resistance of the modified membranes was evaluated by using humic acid as a model foulant, and the results demonstrated that the SWCNT/PVDF membrane with 10 layers exhibited the highest fouling resistance with a flux decline of only 6% after 6 hours of operation. The study concluded that the incorporation of SWCNTs in the PVDF membrane can improve the membrane performance in terms of water flux and fouling resistance by inducing electrical repulsion between the membrane surface and the foulants. Moreover, increasing the number of SWCNT layers can further enhance the membrane performance, making it a promising approach for the development of high-performance MD membranes [24]. Navya Thomas's (2021) study investigated the use of antiscaling 3D printed feed spacers coated with nanoparticles for membrane distillation (MD) applications. The 3D printed feed spacers were coated with silica nanoparticles using a facile dip-coating method. A scale-inducing aqueous solution of calcium sulphate was used as the feed of a direct contact MD process to evaluate the antiscaling effectiveness of uncoated and FS coated spacers. The FS coated spacer had a scalant (Ca^{2+}) attachment of 0.24 mg cm^2, which is 74% less than the uncoated 3D spacer's attachment of 0.95 mg cm^2. Additionally, 60% less scaling on the membrane surface was seen when the antiscaling FS coated spacer was used. Microscale roughness-induced hydrophobicity and decreased surface-free energy, which impaired the scalant's interaction with the spacer surface, were the main variables that lowered scaling with FS coating. The results showed that the coated spacers exhibited a higher water vapor flux and salt rejection rate compared to the uncoated spacers [25]. Teoh et al. (2022) aimed to investigate the effectiveness of using DCMD for the simultaneous recovery of water and nutrients from aquaculture wastewater. The final membrane has surpassed the standard for super hydrophobicity. It attained a high 153.3° WCA and a low 8.4° contact angle hysteresis. A slight and consistent decrease in flow rate by 1.4 L/m^{2}h was observed during the continuous separation and treatment of fish farm effluent, indicating reduced fouling of the surface-printed membrane. The water recovery in the batch feed concentration process reached 86.3%. The initial concentrations of ammonia (ranging from 16.4 to 82.2 mg/L), phosphate (from 18.0 to 99.8 mg/L), and potassium (between 68.0 to 384.8 mg/L) were all at least five times higher in the retentate concentration of fish farm water. This concentrated feed could potentially serve as the primary nutrient source in liquid fertilizer production. Except for ammonia (>86%), all chosen inorganic compounds in the feed concentration process are rejected at a rate higher than 99%. Overall, the results suggest that the DCMD system has great potential for the sustainable management of aquaculture wastewater [26]. Table 1 presents the recent research trends in DCMD incorporating nanoparticles: Insights from Literature. Figure 3 presents correlation between Feed Temperature and WCA and Flux in Nanoparticle-Embedded Polymer Membrane. The Michal Bodzek (2019) titled "Nanotechnology in water and wastewater treatment. Graphene – the nanomaterial for next generation of semipermeable membranes" reviews the potential of graphene-based materials for water and wastewater treatment. The author concludes that graphene-based materials have the potential to revolutionize the field of membrane technology due to their unique properties, such as high mechanical strength, thermal stability, and exceptional water transport properties. Graphene-based membranes have shown promising results for desalination, water purification, and wastewater treatment applications. However, the paper also highlights that more research is needed to address the challenges related to the scalability and cost-effectiveness of these membranes [27].

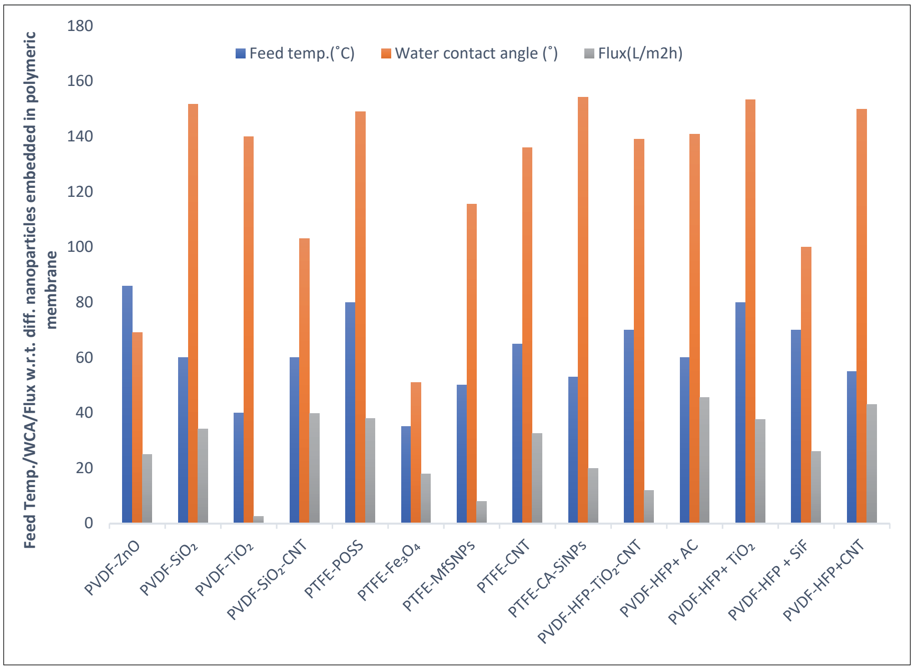

Figure 3: Variation in Water Contact Angle (WCA) and Flux with Changing Feed Temperature in Polymer-Embedded Nanoparticle Membrane

Table 1: Exploring Nanoparticle Integration in DCMD: Overview of Recent Investigations.

S. No.	MD Configuration	Polymer -Nanoparticle Membrane Material	Feed temp. (˚C)	Water contact angle (˚)	Liquid entry pressure (kPa)	Membrane Thickness (μι)	Pore Size (μι)	Porosity (%)	Flux (L/m²h)	Salt Rejection (%)	References
1	DCMD	PVDF-CNT	60	150.4	40.5	88	1.12	89.4	NA	NA	28
2	DCMD	PVDF-ZnO	86	69	900	NA	NA	14	25	99	29
3	DCMD	PVDF-SiO_2	60	151.7	72.3	135	1.27	79.9	34.2	≥99.9	30
4	DCMD	PVDF-TiO_2	40	140	NA	84.5	0.41	44.43	2.5	97	31
5	DCMD	PVDF-SiO_2-CNT	60	103	590	NA	0.22	63.6	39.77	99	32
6	DCMD	PTFE-POSS	80	149	16	NA	0.5	70	38	99.99	33
7	DCMD	PTFE-Fe_3O_4	35	51	379	221	NA	60.7	18	NA	34
8	DCMD	PTFE-MfSNPs	50	115.5	NA	NA	NA	NA	8	99.1	35
9	DCMD	PTFE-CNT	65	136	483	NA	0.292	NA	32.49	99.9	36
10	DCMD	PTFE-CA-SiNPs	53	154.2	NA	303	0.47	50.6	19.92	NA	37
11	DCMD	PVDF-HFP-TiO_2-CNT	70	139	81	73	0.46	86	11.85	NA	38
12	DCMD	PVDF-HFP+ AC	60	140.9	1.36	200	0.805	91.3	45.6	99.99	39
13	DCMD	PVDF-HFP+ TiO_2	80	153.4	90.5	99	0.76	89.8	37.6	99.99	40
14	DCMD	PVDF-HFP + SiF	70	100	250	142	0.23	59	26	NA	41
15	DCMD	PVDF-HFP+CNT	55	150	136	NA	0.715	NA	43	99.8	42

CONCLUSION

- Silicon dioxide exhibits compatibility with PVDF, modifying the water contact angle to 151.7˚ with the inclusion of nanoparticles and enhancing flux to 34.2 L/m²h. Likewise, POSS shows good compatibility with PTFE.
- Additionally, the combination of silicon dioxide and carbon nanotubes with PVDF demonstrates a high flux of 39.7 L/m²h.
- Inclusion of CNT to the polymer results in major enhancement in measurement of WCA and Flux.
- Pore size, liquid entry pressure, water contact angle, temperature and salinity of feed water are the key parameters which affects the performance of membranes in terms of flux and wettability.
- Nanofibrous polymer membranes and nano particles embedded polymer membranes have shown high flux and stability for seawater desalination

Despite the remarkable progress in nanoparticle-enhanced polymeric membranes, several challenges remain to be addressed. These include scalability, cost-effectiveness, long-term stability, and environmental impacts associated with nanoparticle synthesis and disposal. Additionally, there is a need for further research to understand the interactions between nanoparticles and polymeric matrices, as well as their effects on membrane performance under real-world conditions. Overall, nanoparticle-modified polymeric membranes hold great promise for advancing the field of wastewater treatment by offering sustainable, cost-efficient, and high-performance solutions. Continued interdisciplinary research and collaboration will be essential in realizing the full potential of these innovative membrane materials for addressing global water challenges and ensuring access to clean water for all.

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IZVOD

ISTRAŽIVANJA MEMBRANA NA BAZI NANOMATERIJALA ZA EFIKASNO UKLANJANJE ZAGAĐIVAČA IZ OTPADNIH VODA PREKO MEMBRANSKE DESTILACIJE: KRITIČKI PREGLED

Zahtevi za tretman otpadnih voda su najvažniji u obezbeđivanju održivosti životne sredine i očuvanju javnog zdravlja. Kako se industrijalizacija i urbanizacija ubrzavaju, količina proizvedene otpadne vode nastavlja da se povećava, koja sadrži raznolik spektar zagađivača. Neprečišćena otpadna voda predstavlja ozbiljnu pretnju ekosistemima, vodnim telima i ljudskim zajednicama, što dovodi do zagađenja, bolesti, koje se prenose vodom, i ekološke neravnoteže. Efikasno prečišćavanje otpadnih voda postaje neophodno za ublažavanje ovih štetnih efekata uklanjanjem ili smanjenjem zagađivača pre ispuštanja u prirodne izvore vode. Ovaj proces pomaže u očuvanju kvaliteta vode, zaštiti vodenog sveta i održavanju opšteg zdravlja ekosistema. Membranska destilacija (MD) se pojavila kao obećavajuća tehnologija za prečišćavanje otpadnih voda, nudeći inovativan pristup za rešavanje izazova povezanih sa konvencionalnim metodama prečišćavanja. U MD, hidrofobna membrana služi kao selektivna barijera, dozvoljavajući vodenoj pari da prođe, dok sprečava prolaz zagađivača. Ovaj rad nudi opširan pregled najnovijih dostignuća u nanotehnologiji i membranskoj destilaciji primenjenoj u tretmanu otpadnih voda. Udubićemo se u različite vrste nanomaterijala koji su korišćeni za poboljšanje svojstava MD membrana, kao što su nanokompoziti, nanočestice i membrane od nanovlakna. Takođe istražujemo mehanizme pomoću kojih ovi nanomaterijali poboljšavaju efikasnost odvajanja, svojstva protiv obraštanja i izdržljivost MD membrana. Pored toga, ističemo potencijal hibridnih membrana koje kombinuju različite vrste nanomaterijala za dalje poboljšanje performansi MD u tretmanu otpadnih voda. Dajemo primere nedavnih studija koje su istraživale upotrebu hibridnih membrana, uključujući hibridne membrane ugljenik nanocevi-grafen oksid, membrane od nanokompozitnih nanovlakna i membrane ugrađene u nanočestice srebra. Takođe identifikujemo neke oblasti za buduća istraživanja i razvoj, kao što je povećanje i komercijalizacija sistema MD zasnovanih na nanotehnologiji. Ukratko, ovaj pregledni rad naglašava potencijal nanotehnologije da poboljša performanse MD u tretmanu otpadnih voda, što dovodi do poboljšanog kvaliteta vode i čistije životne sredine.

Ključne reči: *nanokompozit; Nanofibers; Tretman otpadnih voda; Membranska destilacija*

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