Chapter 3

Recent Developments of Membrane Technology for Wastewater Treatment

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Abstract

The worldwide one major and important issue is the increasing shortage of freshwater. Water is polluted by various category of pollutant such as heavy metal, organic toxic chemical, dyes and others. In such situation providing better solutions for water treatment is a major challenge for researchers. Various techniques have been used in wastewater treatment applications but among those techniques the membrane technology is the most promising technology. This chapter contains recent progress of membrane technology for advanced wastewater treatment, is systematically summarize. This review includes introduction about different membrane technology such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO). Current status of each membrane separation techniques, membrane cleaning techniques, challenges and promising solutions for various wastewater treatment have been discussed.

Keywords

Membrane Technology, Wastewater, Pollutant Removal, Membrane Fouling, Membrane Cleaning

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1. Introduction

The throughout worldwide contamination of ground and surface water has become a major environmental issue for the last decades. The anthropogenic activities are the main reason of water pollution. That include, various industrial waste, sewage and fertilizers. The major sources of wastewater are industrial and municipal applications [1]. The various industries include tannery [2], dye [3], petroleum [4], electroplating [5], pharmaceutical [6], chemical industries [7], agrochemical [8], pesticide [9] and many more industries producing wastewater and it is harmful to environment, human and animal life.

The various technologies already in used for wastewater treatment, including membrane filtration [10,11], adsorption [12,13], coagulation [2], chemical precipitation [14], electrocoagulation [14], ion exchange [15], capacitive deionization [1,16–17], biosorption and bioaccumulation [18], advanced oxidation process [19]. In the past few decades, membrane separation processes have received significant attention for wastewater treatment due to its high energy efficiency and low environmental impact [20].

This book chapter presents recent progress of membrane technology for advanced wastewater treatment in last three years. This chapter review includes introduction about different membrane separation process namely, microfiltration (MF), ultrafiltration (UF),

nanofiltration (NF), and reverse osmosis (RO). The current status of each membrane separation techniques, challenges and promising solutions for various wastewater treatment have been discussed.

2. Information about different pressure driven membrane processes

2.1 MF process

MF is one of the well-known oldest pressure driven membrane process used in water treatment [21]. It has been used to remove micrometer sized matter (suspended particles, large bacteria, major pathogens, yeast cells and proteins) based on the principle of physical separation [22]. MF membranes have 0.1 –5 μm range of pore diameters. The MF membranes need low hydrostatic pressures to reject a high contaminant [23–25]. The MF process also used as a pre-treatment for RO or NF membrane processes to decrease the fouling potential [26].

2.2 UF process

The UF membrane process can remove compounds ranging between 0.05–10 μm successfully and this process is operated between MF and RO [27]. UF membranes are impressively noticeable water filters to remove macromolecules, suspended matter, pathogenic and microorganisms among others with low energy consumption [28]. However, UF membrane process has some limitations such as incompetence to reject any dissolved organic substances from the water and needs consistent cleaning to achieve high water pressure flow [29].

2.3 NF Process

NF process operated between UF and RO membranes process. The NF membrane pore size is normally of the order of 1 nm. The molecular weight cut-off (MWCO) range for NF membrane is 100–5000 Da. NF membranes work at moderate applied pressures with no phase change and it is capable of successfully removing small organic molecules and multivalent salts. The NF is a highly attractive process for selectivity and is cost effective compared to other traditional processes. Thus, the NF process has extensive applications in water and wastewater treatment [30].

2.4 RO Process

The RO membrane process is the most attractive and globally used technology for desalination and wastewater treatment. It has been used for production of clean water for different application purpose such as drinking, agriculture, and industries. The RO process

efficiency depends on the operating parameters and characteristics of the feed water. In this process external pressure is applied to the feed water stream to overcome osmotic pressure [31,32]. The RO membranes have <1 nm pore size [32, 33]. The clean water is obtained by rejecting salt and other minerals when applied pressure is higher than the osmotic pressure ($\Delta p > \Delta \pi$). The salt rejection and water flux have significant role in the performance evaluation of RO membranes [31,32].

3. Current status of MF, UF, NF and RO membrane processes

3.1 MF current progress discussion

This section includes the recent progress on MF reported in the last three years. MF process are used as single as well as combined or hybrid process.

Ba and co-authors investigated the efficiency of a hybrid bioreactor (HBR) and hollow fiber MF membranes to remove a mixture of 14 PPs from municipal wastewater. The results show that complete removal of PPs from wastewater was obtained by using a HBR and MF membrane process [34]. Goswami and his team reported a study on the treatment of three different industries wastewater by biodegradation followed by a MF membrane process. This integrated system performance indicated very high COD removal. This integrated system also labels as cost-effective approach for different industrial wastewaters with vision of valuable resource recovery [35]. Jafari and co-authors developed the tubular ceramic MF membranes by using activated carbon for the treatment of oily wastewater. The outcome of the experiment indicates the permeation flux and rejection of total organic carbon improved due to existence of activated carbon and natural zeolite in the membrane structure. The TOC rejection was observed up to 99.99% with Mullite-Zeolite-Activated carbon membrane [36]. Saja and co-workers reported their work on flat ceramic MF development by using natural perlite and applied for treatment of industrial wastewater. That membrane was prepared by a uniaxial pressing process and sintering at optimized temperature. The prepared membrane shows 52.11% porosity, 1.70 μm average pore size, 21.68 MPa mechanical resistance and also it has good chemical resistance. The turbidity retention was found to be 97 and 96% for agro-food and tannery effluents respectively from experiments [37]. Pan and co-authors reported work on a novel electrochemical MF process. In that process the coal-based carbon membrane was used for the removal of bisphenol A (BPA) from wastewater. The membrane plays the roles of filter and anode simultaneously. The electrochemical MF process shows the excellent performance for BPA removal because of the synergistic effect between electrochemical oxidation and membrane filtration [38]. Beqqour et al. prepared the low-cost ceramic MF pozzolan membrane using micronized phosphate by dry uniaxial pressing process and sintering. The

results indicate that improvement was found in mechanical strength, microstructure and permeability of the membrane on increased temperature. The experimental results showed that the membrane could almost reject all turbidity for tannery effluent and aluminum chloride suspension [39]. Mouiya et al. studied the development of MF ceramic membrane by uniaxially compressing natural Moroccan materials. The natural phosphate can be helpful to porosity development. The prepared MF membrane demonstrated excellent turbidity removal performance for synthetic solution (99.86%), effluent (99.80%), and seawater (99.62%). After the filtration experiments MF ceramic membrane show good mechanical stability and microstructure [40]. Arribas et al. reported work on modification of the structural and morphological properties of polysulfone electrospun nanofibrous membranes by heat post-treatment method to improve filtration performance. The modified membrane shows the higher PWP than commercial membranes with lower pressures requirement and thus it drops energy consumption in filtration [41]. Li and co-authors worked on CuO/carbon membrane fabrication. The electrocatalytic performance of the membrane was improved by uniformly deposition of CuO electrocatalysts on a coal-based carbon membrane. The obtained membrane showed superior removal capability and it showed great potential, high permeability and removal efficiency for the removal of small-sized organic pollutants [42]. Sheikhi et al. studied development of kaolinitic clay-based ceramic membrane for investigation of inline coagulation-MF (IMF) process performance for oily wastewater. The overall results indicated that the IMF system could be reflected as a capable system for filtration performance intensification and membrane fouling mitigation [43]. Hatimi et al. reported work on preparation of low-cost MF membrane from natural clay and pyrrhotite ash solid waste. They made changes in the surface morphology and improved the hydrophobicity of the membrane. The prepared membrane was used for the treatment of industrial wastewaters (dairy and tannery). The membrane performance indicates the almost complete turbidity, oil and fat reduction even at high flows [44]. Manni et al. reported a study on the preparation of low-cost ceramic MF membrane made from natural magnesite. The experimental results clearly demonstrated that magnesite membrane effectively clarifies textile wastewater. The result shows that 99.9 and 69.7% removal of turbidity and COD achieved respectively [45]. Zhang et al. developed diatomite hybrid MF carbon membranes (MFCMs) for removal of oil from oily wastewater. The maximum oil rejection was found to be 98.2% for 200 mg/L of oily wastewater. The permeation flux could be almost completely recovered after regeneration of the MFCMs by simple water backwashing [46]. Homem et al. modified the polyethersulfone MF membranes with polyethilenimine (PEI) and graphene oxide (GO). The best result indicates the 97.8% rejection of Blue Corazol dye and $99.4 \text{ L m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$ pure water permeability. The membrane shows $> 80\%$ flux recovery ratio after cleaning [47]. Mouiya and co-authors reported clay-based ceramic membrane development. In that

corresponding support was prepared using a clay/banana peels mixture followed by sintering. The MF membrane was used for the treatment of industrial wastewater. The Experimental results show high turbidity retention and dye rejection [48]. Liu et al. reported work on the fabrication of membranes from polyvinyl alcohol and polyacrylonitrile by electrospinning approach. The membrane demonstrated good adsorption capacity for Cd (II) ions. This study also found regeneration efficiency of above 90% after three recycle operations. Thus, this membrane could be reused for several times [49]. Abadikhah et al. reported the work SiO₂ nanoparticles deposition on the outer surface of Si₃N₄ hollow fiber membrane through the sol-gel process. It results found that the reduction in the surface roughness and hydrophilicity of the membrane was enhanced. Experimental outcomes show that high separation of oil performance was achieved [50]. Chen and co-authors report work on a waste-to-resource strategy for rational preparation of low cost ceramic membranes, which can be applied for the treatment of heavy metal-laden sludges and the separation of oil-in-water (O/W) emulsions. Outcomes indicate this membrane exhibited high rejection and high flux [51]. In the study reported by Plakas et al. catalytic membrane reactor consisting of a tubular porous alumina membrane with embedded iron oxide nanoparticles was used for pharmaceutical diclofenac oxidation. Significant diclofenac oxidation/mineralization was obtained by the catalytic membrane reactor [52]. The recent progress on MF performance in wastewater treatment are systematically shown in table 1.

3.2 UF current progress discussion

This section includes the recent progress on UF membrane processes reported in the last three years. Bhattacharya and co-authors developed the ceramic UF membrane by using green synthesized zinc oxide nanoparticles. The obtained UF membrane was used for atenolol and ibuprofen drugs removal from a synthetic solution. The Results showed that removal of atenolol and ibuprofen were found to be 96% and 99% respectively by single a step UF process [53]. A fungal bioactive UF membrane from filamentous fungi *Aspergillus carbonarius* M333 was developed by Isik and co-authors. The prepared membrane was used for textile industry wastewater treatment. The fungal bioactive ultrafiltration membrane experimental performance indicate that decolourization of wastewater and COD reduction were obtained 91% and 73.2% respectively [54]. Bhattacharya et al. developed the high flux ceramic UF membrane and it was applied to ciprofloxacin removal from synthetic feed solution. In that study the membrane was prepared with copper oxide and TiO₂ nanoparticles as composite. The obtained UF membrane performance showed effective removal of ciprofloxacin up to 99.5% [55]. Derouich et al. prepared a new composite membrane by polypyrrole layer deposition on a flat porous support based on a natural geomaterial. This obtained membrane was tested for

dye textile effluent treatment. At optimized conditions, the prepared UF membrane removed Congo red dye up to 98% [56]. Li et al. fabricated a hybrid UF membrane and that hybrid UF membrane demonstrated high water flux and outstanding nickel ion adsorption capacity was obtained in the treatment of high-salinity synthetic wastewater [57]. Shakak and co-author reported a study on the preparation of nanocomposite UF membrane from Polysulfone, Polyvinylpyrrolidone and SiO₂ using the phase inversion method. The investigation of membrane performance presented that removal performance of amoxicillin increased from 66.52% to 89.81% by increasing SiO₂ nanoparticles from 0 to 4 wt% respectively. The addition of SiO₂ nanoparticles also enhanced the membrane resistance against fouling [58]. Ye et al. fabricate the tight UF membranes from the PDA/polyethylenimine co-deposition triggered by ammonium persulfate. It was used for highly saline textile wastewater treatment.

Table 1. Recent progress on MF performance in wastewater treatment

Process	Membrane details	Wastewater type	Name pollutants	Feed Concentration	Removal % / efficiency/ output	Ref.
Hybrid hybrid bioreactor (HBR) and hollow fiber MF	Polysulfone hollow fiber MF membrane	municipal wastewater	Mixture of 14 pharmaceutical products	13 spiked pharmaceutical products at 10 mg/L and native acetaminophen at 35.5 mg/L	90%	[24]
biodegradation followed by MF membrane filtration.	low-cost ceramic membrane	A: dairy wastewater B: paper and pulp wastewater C: biomass gasification wastewater	COD removal	A: 2234 mg L ⁻¹ B: 937 mg L ⁻¹ C: 2157 mg L ⁻¹	A: 92.7%, B: 87.6% C: 88.2%	[35]
MF	hybrid ceramic/carbon microfiltration membrane (Mullite-Zeolite-Activated carbon membrane)	oily wastewater treatment	A: Oil B: TOC	A: 500/1000/1500 mg/L B: 1500 mg/L	A: 98.5/ 98.2/97.4% B: 99.99%	[36]
MF	ceramic membrane based on Moroccan natural perlite	agro-food (A) and tannery effluents (B)	Turbidity (NTU)	A: turbidity: 224 NTU B: turbidity: 36 NTU	A: 97% and B: 96%.	[37]
Electrochemical MF process	coal-based carbon membrane	bisphenol simulated wastewater	bisphenol A and COD	BPA (50ppm)	97% and 90%,	[38]

MF	ceramic microfiltration pozzolan membrane	aluminum chloride suspension (A) and tannery industry wastewater(B)	Turbidity	A: turbidity: 1743 NTU B: turbidity: 201NTU	A: 99% B: 97%	[39]
MF	ceramic MF membrane (based on natural clay and Moroccan phosphate)	Beamhouse effluent (A), raw seawater (B), and aluminium chloride(C)	Turbidity	A: (turbidity: 760 NTU) B: (turbidity: 120 NTU) C: (turbidity: 725 NTU)	A: (99.80%), B:(99.62%) C: (99.86%).	[40]
MF	polysulfone electrospun nanofibrous membranes	Synthetic wastewater	humic acid	15 mg/L	Separation factor percentage 77.6	[41]
electrocatalytic MF	electrocatalytic microfiltration CuO/Carbon membrane	Synthetic wastewater	A: Rhodamine B (RhB) B: COD C: TOC	RhB: 300 mg/L	A: 99.96%, B: 71.82% and C: 64.29%	[42]
Inline coagulation-MF (IMF) process	kaolinitic clay-Based ceramic membrane	oily wastewater	Oil	921 mg/L	98.06%	[43]
MF	natural clay and pyrrhotite ash solid waste based MF membrane	A: Tannery effluent B: Dairy effluent	A1: Turbidity A2: Conductivity A3: Cr B1:Turbidity B2:Conductivity B3:Oil and fat	A1: 595.00 NTU A2: 39.40 ms/cm A3: 48.31 mg/l B1: 789.00 NTU B2: 4.35 ms/cm B3: 150.02 mg/L	A1: 96% A2: 3% A3: 8% B1: 100 % B2: 3% B3: 94 %	[44]
MF	low-cost ceramic microfiltration membrane made from natural magnesite	Textile wastewater.	Turbidity	144.5 ± 0.6 NTU	99.9%.	[45]
MF	Diatomite hybrid microfiltration carbon membrane	oily wastewater	Oil	200 mg/L	98.2%	[46]

MF	polyethersulfone microfiltration membrane surface modified with graphene oxide	Textile industries wastewater.	Blue Corazol (BC) dye rejection	10 mg/L	97.8%	[47]
MF	ceramic membrane from clay and banana peel powder	tannery wastewater(A) textile wastewater (B)	A1:Turbidity A2: COD B1:Turbidity B2: COD	A1: 554 NTU A2: 2413 mg L ⁻¹ B1: 250 NTU B2: 536 mg L ⁻¹	A1: 99.99% A2: 90.38% B1: 99.98% B2:85.72%	[48]
MF	Nanofibrous PAN-PVA composite membranes	Contaminated water	Nanoparticles and heavy metal ions A:Cr (VI), B:Cd (II)	A: 40-120 mg/L B: 20-60 mg/L	adsorption capacity A: 66.5 mg/(g membrane) B: of 33.6 mg/(g membrane) rejection ratios higher than 99.3%	[49]
MF	SiO ₂ nanoparticles modified Si ₃ N ₄ hollow fiber membrane	oily wastewater	Oil	1000 ppm	95%	[50]
MF	Spinel-based ceramic membranes	Oily wastewater	Oil	500 mg/L	99.5%	[51]
catalytic membrane reactor	tubular porous alumina (α -Al ₂ O ₃) membrane	synthetic solutions	Diclofenac	200 μ g/L	~ 65%	[52]

The remarkable fractionation of dye/salt mixtures are attained. These bio-inspired tight UF membranes are a good alternative for textile wastewater treatment among other possible UF practices [59]. Isawi prepared the vermiculite nanoparticles incorporated polyvinylidene fluoride flat sheet membrane. The outcomes of that study clearly revealed that the application of vermiculite nanoparticles in membranes was beneficial to the enhancement of membrane performance, antifouling and mechanical properties, compared to other membranes at the same condition [60]. Yang et al. successful prepared a novel

thermally stable polyimide (PI) and it was designated as characteristic candidate due to its excellent thermal stability. This PI-based UF membrane showed a rejection of 98.65% direct red 23 dye [61]. Liu and team developed the CNT/PVDF/stainless steel mesh conductive UF membrane. The new obtained membrane showed good electrical conductivity and high mechanical strength. The results showed efficient Cr(VI) removal under the application of voltage [62]. Algamdi and co-authors prepared hybrid polyethersulfone membranes incorporated with GO via a nonsolvent induced phase separation method. The GO improved membrane antifouling capabilities and membrane reusability [63]. Saja et al. fabricated the low-cost UF bentonite membrane deposited on ceramic perlite support. The bentonite layer was made by spin-coating process followed by sintering. The result showed that 97.0 and 80.1 % rejection of Direct Red 80 and Rhodamine B respectively [64]. Arefi-Oskoui et al. reported work on modification of polyethersulfone UF membrane using ultrasonic assisted functionalized MoS₂. The obtained membrane performance evaluated against treatment of oil refinery wastewater. The optimum nanocomposite UF membrane showed maximum average surface roughness and subsequently highest efficient area for filtration than other nanocomposite membranes. The experimental result of optimum nanocomposite UF membrane show COD and the turbidity were decreased by 40.8 mg/L and 2.1 NTU for the feed wastewater of 240 mg/L and 12.2 NTU respectively [65]. Ibrahim et al., fabricated polysulfone blend UF membrane using poly[styrene-alt-(N-4-benzoylglycine-maleamic acid)] cumene terminated via the phase inversion method. The membrane indicate excellent performance against removal of heavy metal ions (Pb²⁺ and Cd²⁺). This membrane can be reused by simple acid wash [66]. Huang et al. reported work on Al³⁺-doped TiO₂ (AT) tight UF membrane with stable anatase phase made by a modified solgel process using butyl titanate (as the precursor) and aluminum chloride (as aluminum source). The membrane showed 96.9% rejection for Alizarin Red-S for feed concentration of 250 ppm [67]. Benkhaya and team prepared composite UF membranes from blend of polysulfone and polyetherimide with flat pozzolan support. The optimized membrane showed promising performance against rejection of 96.1% and 75.8% for acid orange 74 and methyl orange respectively [68]. Yu et al. reported work on modification of Poly(vinylidene fluoride) UF membranes with poly(vinyl pyrrolidone) to improve the antifouling properties. The pore modification and the surface modification improved the membrane flux and membrane hydrophilicity respectively. The modified membranes show improved anti-fouling abilities and such membranes are appropriate for low-level radioactive wastewater treatment [69]. Benkhaya et al. prepared new organic UF membranes based on the physical blends of Polysulfone/Polyetherimide by phase inversion process. The prepared membrane was found effective for dye rejection [70]. Ahmad et al. reported work on polyvinyl chloride (PVC) membrane for improvement of surface morphology, hydrophilicity and antifouling performance by using bentonite. The

oil rejection by using prepared membrane were 97.0% and 92.5% from oily-wastewater with zero salinity and 35000 ppm salinity respectively [71]. The recent progress on UF membrane application in wastewater treatment are systematically shown in table 2.

Table 2. Recent progress on UF performance in wastewater treatment

Process	Membrane details	Wastewater type	Name pollutants	Concentration of Feed	Removal % / efficiency/ Output	Ref.
UF	ZnO UF membrane	synthetic wastewater	A: atenelol B: ibuprofen	A: 505 ppb B: 1000 ppb	A: 96% B: 98%	[53]
Biodegradation and UF	Bioactive ultrafiltration membrane	textile wastewater	A: Basic Red 18 (BR 18) dye B: Remozal Brilliant blue R (RBBR) C: COD	A: 10 mg/L B: 10 mg/L C: 1372 mg/L	A: 96% B: 95% C: 73.2%	[54]
UF	CuO/TiO ₂ ceramic ultrafiltration membrane	synthetic solution	Ciprofloxacin	500 µg/L	99%	[55]
UF	Polypyrrole/sintered pozzolan ultrafiltration membrane	synthetic solution	Congo red dye	600 ppm	98 ± 1 %	[56]
UF	PAA/ZIF-8/PVDF hybrid ultrafiltration	high salinity wastewater	nickel (II)	[Ni(II)] ₀ =2 mg/L, [Na ⁺] ₀ =15000 mg/L	maximum adsorption capacity 219.09 mg-Ni(II)/g	[57]
UF	Nanocomposite ultrafiltration membrane (PSF/PVP/SiO ₂)	aqueous solutions	amoxicillin	30 mg/L	90%	[58]
UF	bio-inspired tight ultrafiltration membrane	highly-saline textile wastewater	A: reactive dyes (reactive blue 4) B:salts(NaCl/Na ₂ SO ₄ mixture)	A: 2.0 g/L B:30.0 g/L	A: > 98.2% B: > 97.0%	[59]
UF	PVDF membranes incorporated with the vermiculite NPs	Wastewater	humic acid	1 g/L	94.56%.	[60]

UF	Polyimide ultrafiltration membrane	aqueous dye solution	direct red 23 dye	100 ppm	98.65%	[61]
electrochemical membrane system	CNT/PVDF/stainless steel mesh conductive Ultrafiltration membrane	aqueous solution	hexavalent chromium	1 mg/L	95.2%	[62]
UF	Graphene oxide incorporated polyethersulfone hybrid ultrafiltration membranes	aqueous solution	humic acid	10 –100 ppm	94.5%	[63]
UF	Ultrafiltration bentonite membrane	aqueous solution	Direct Red 80 and Rhodamine B	100 ppm	97.0 and 80.1 %	[64]
UF	MoS ₂ /polyethersulfone nanocomposite membrane	oil refinery wastewater	A: COD B: turbidity	A: 240 mg/L B:12.2 NTU	A: 40.8 mg/L (83 %) B: 2.1 NTU	[65]
UF	Polysulfone blend ultrafiltration membrane	Wastewater	heavy metal ions (A: Pb ²⁺ and B: Cd ²⁺)	10 ppm	A: 91.5%, B: 72.3%	[66]
UF	Al ³⁺ -doped TiO ₂ tight ultrafiltration membrane	aqueous solutions	Alizarin Red-S	250 ppm	96.9%	[67]
UF	Polysulfone (PSf) and polyetherimide (PEI) composite ultrafiltration membrane	aqueous dye solutions	A: acid orange 74 (AO74) B: methyl orange (MO)	50 ppm	A: 96.1 B: 75.8%	[68]
UF	Poly(vinyl pyrrolidone) modified poly(vinylidene fluoride) ultrafiltration membrane	radioactive wastewater	A: nuclide ions and B: SDBS (anionic surfactant sodium dodecyl benzene sulfonate)	A: 0.1 mg/L B: 200 mg/L	A: 98% of divalent ions Sr(II) and Co(II) B: SDBS were lower.	[69]
UF	Polysulfone/Polyetherimide	aqueous dye solutions	A: acid orange 74 (AO-74) and	50 ppm	A: 68.5% B: 64.7%	[70]

	blends organic ultrafiltration membrane		B: methyl orange (MO)			
UF	mixed-matrix poly(vinyl chloride)-bentonite ultrafiltration membrane	saline oily wastewater	A: Oil B: salt	A: 200 ppm B: 35000 ppm	A: 97.0% B: negligible salinity	[71]
UF	polyethersulfone (PES) and regenerated cellulose (RC) commercial ultrafiltration membranes	poultry processing wastewater (Bird washer[BWW] water, Chiller water[CW])	A: biochemical oxygen demand (BOD), B: chemical oxygen demands (COD), C: total suspended solids (TSS), D: fat, oil and grease (FOG)	A: 400 mg/L B: 1000 mg/L C: 300 mg/L D: 200 mg/L	A: (up to 93%), B: (up to 94%), C: (up to 100%) D: (up to 100%) for both wastewater streams.	[72]
complexation- UF process	composite membrane (made of polysulfone and nanoclay)	aqueous solutions	anionic dye (Acid-Orange 7, AO7)	83 mg/L	99.62%	[73]

3.3 NF current progress discussion

This section includes recent progress on NF membrane processes reported in last three years. Zhao et al. fabricated a loose layered double hydroxides /polymer hybrid layer NF membrane. It tested for the desalination of textile wastewater. The membrane performance shows a sufficient permeability, high rejection of dyes and low salt rejection [74]. Zhao and co-authors prepared a loose NF composite membrane and it was used for treating textile wastewater. The membrane prepared at optimized condition show high permeability and high rejection of methyl blue dye. It also shows excellent antibacterial properties [75]. Zhang et al. prepared a photo-assisted NF membrane build from g-C₃N₄, TiO₂, CNT and GO. Photo-assisted NF membranes show a higher water flux and high methyl orange dye rejection (~100%). The NF membrane combined with photocatalysis shows the efficient removal of bisphenol A, ammonia and antibiotic from water [76]. Maryam et al. tested two types of loose NF membranes for the filtration of selected small molecular weight drugs from synthetic wastewater. The pH effect study show that behaviour of drugs changed with pH. The results showed remarkable treatment of drugs in the order of, DIC > IBU > PARA along with TOC and COD [77]. Tsehay et al. reported a study on the combination of TiO₂ nanoparticles via blending in order to fabricate PES (TiO₂) membranes. For the reference

purpose they used commercial PES NF membranes and Pristine. The results indicate a high water permeability and 94.9% direct red 23 rejection with obtained membrane. Therefore, PES (TiO₂) membrane a capable candidate in industrial wastewater treatment [78]. Kusworo et al. reported work on polymeric membrane modification using TiO₂ nanoparticle to improve its separation and mechanical properties. The prepared membrane was used for treatment of natural-rubber wastewater.

The addition of TiO₂ improved the hydrophilicity and mechanical strength of the membrane. The experimental results show best pollutant removal efficiency for TDS, COD, NH₃, and turbidity rejections with PSf-TiO₂ membrane [79]. Guo and team prepared composite NF membranes with the layer by layer approach using plant polyphenol tannic acid with rich catechol groups and hydrophilic Jeffamine having amino groups. The obtained membrane showed high pure water permeance of 37 Lm⁻²h⁻¹bar⁻¹ and also maintaining rejections higher than 90% for various dyes of molecular weight range of 269 to 1017 g mol⁻¹ [80]. Bandehali et al. developed PEI based NF membranes by incorporation [L-cysteine functionalized POSS nanoparticles]. The prepared membrane demonstrates smoother surface and excellent antifouling properties than pristine PEI membrane [81]. Wang and co-authors fabricated graphene oxide/attapulgit composite membranes for the removal of dyes from wastewater. This composite of membranes showed rough hierarchical microstructure and higher surface hydrophilicity. The obtained composite membrane showed high rejection close to 100% for 7.5 mg L⁻¹ RhB wastewater [82]. PEM-based NF membranes were prepared from poly(allylamine hydrochloride) and poly(acrylic acid) on polyacrylonitrile UF support. This membrane was used for rejection of Diclofenac, Naproxen, 4n-Nonylphenol and Ibuprofen from synthetic secondary treated wastewater [83]. de Souza et al. investigated the performance of two NF membranes for norfloxacin removal from a synthetic pharmaceutical wastewater. Both membranes showed high selectivity to norfloxacin and rejection of the antibiotic persisted between 87 and 99.5% [84]. Wang and team prepared GO-based membranes by grafting zwitterionic polymer. The membrane's antifouling ability improved due to the grafted zwitterionic polymer. The prepared membrane presented high rejection for different dyes namely Congo Red, Orange G and Methyl Orange [85]. Sangeetha and co-authors prepared hybrid NF membrane from chitosan/polyvinyl alcohol and montmorillonite clay. The obtained hybrid NF membrane show excellent overall performance [86]. Qin et al. reported work on preparation of 8 mol% yttria-stabilized ZrO₂ (8YSZ) NF membranes using size-controlled spherical ZrO₂ nanoparticles. This prepared membrane was tested in the treatment of pesticide wastewater. The experimental performance of 8YSZ NF membranes showed the removal rate of carbofuran higher than 82% and whereas the extreme removal rate could reach 89% [87]. Cao et al. investigated separation performances of the dye, the intermediate (H-acid) and

salts (NaCl) by two fabricated NF membranes having different charges and pore structural properties. Both membranes showed long-term stability and rejection of RB-5 was higher than 95%. [88]. Tavangar and co-authors prepared loose NF nanocomposite membranes from Poly(ether sulfone) by incorporating nanoparticles of cerium oxide. The membrane was prepared via a non-solvent induced phase separation approach. The obtained membranes performances were investigated by treatment of aqueous dye solutions and real textile wastewater. The nanocomposites membrane demonstrated notable better antifouling properties than the pristine membrane [89]. The various studies on NF membrane used in wastewater treatment are systematically shown in table 3.

Table 3. Recent progress on NF performance in wastewater treatment

Process	Membrane details	Wastewater type	Name pollutants	Feed Concentration	Removal % / efficiency/ Output	Ref.
NF	loose layered double hydroxides (LDHs) LDHs/polymer hybrid NF membrane.	simulated dye wastewater	A: Dyes (Methyl blue, acid fuchsin) B:salt (NaCl)	A: 500 ppm B: 1 g/L	A: 97.9% and 97.5% B: lower salt rejection (<3%).	[74]
NF	PEGylation of plant polyphenols/polypeptide-mediated loose NF membrane	A: aqueous solutions B: Actual textile wastewater	A1: Dyes (methyl blue) A2: salts(NaCl) B1:COD, B2:NH3-N	A1:100~500 ppm A2:2–10 g/L B1: 459 mg/L B2: 8.95 mg/L	A1: 98.9% A2: 17% B1:89.1% B2:84.5%	[75]
photo-assisted NF	multifunctional graphene-based NF membrane	wastewater	A: salt ions B: antibiotic (sulfamethoxazole). C: bisphenol A D: TOC E: Methyl Orange	A: 2 g/L B: 5 mg/L C: 5 mg/L D:41.33 ±0.13 mg/L	A: 67% (Na ₂ SO ₄) B: 80% C: 82% D: 78% E: ~ 100%	[76]
NF	HYDRACoRe 50 (NF50) Hydranautics (Oceanside, CA, USA)	wastewater	Pharmaceutically Active Compounds (PhACs), A:Diclofenac (DIC), B:Ibuprofen (IBU) and C:Paracetamol (PARA), D:TOC E: COD	A: 0.1g/L B: 0.1g/L C: 0.1g/L	A:99.7% B:81.2% C:49% D:95.3% E:84%	[77]

NF	Polyethersulfone (TiO ₂) composite membranes	Aqueous solution	Direct Red 23	0.5 g/L	94.9%	[78]
NF	nano-TiO ₂ loading in polysulfone membranes	Natural-rubber wastewater treatment.	A:TDS, B:COD, C:N-NH ₃ , D:Turbidity	A: 297 mg/L B: 273 mg/L C: 18.68 mg/L D: 32 NTU	A: 14.03 % B:87.88 % C: 88.79 % D: 99 %	[79]
NF	Tannic acid and Jeffamine /polyacrylonitrile composite NF membranes	Dyes solution	A:Methyl orange (MO) B:Rose bengal sodium salt (RB) C:Methylene blue (MB) D:Congo red (CR) E:Rhodamine B (RhB)	35 µM dye solution	A: 91% B:99.5% C:98% D:99.5% E:99%	[80]
NF	[L-cysteine-co-POSS/PEI] membrane.	Aqueous solutions	A: Na ₂ SO ₄ B: Cr(SO ₄) ₂	A: 1100 mg/L B: 500 mg/L	A: 80 % B:79%	[81]
NF	Graphene oxide/attapulgit (GO/APT) composite membranes	dyes solution	rhodamine B	7.5 mg L ⁻¹	100%	[82]
NF	Polyelectrolyte multilayer (PEM)-based nanofiltration (NF) membranes	synthetic secondary treated wastewater	Micropollutants A:Diclofenac B:Naproxen C:Ibuprofen D: 4n-Nonylphenol	A: 0.5 µg/L B: 2.5 µg/L C: 40 µg/L D: 7 µg/L	A: 81.5% B: 66.6% C:51.6% D:61.7%	[83]
NF	NF 270 and NF 90 (Filmtec – Minneapolis, MN)	pharmaceutical effluent	Norfloxacin	50 mg L ⁻¹	87 –99.5%	[84]
NF	zwitterionic polymer modified GO@PDA/PES membranes	organic dye solutions	A:Congo Red, B:Orange G C:Methyl Orange	100 mg/L	A:100%, B:82% C:67%	[85]
NF	Chitosan/polyvinyl alcohol/montmorillonite clay membrane	synthetic wastewater	Chromium	50 ppm	84 –88.34%	[86]
NF	yttria-stabilized ZrO ₂ nanofiltration membrane	synthetic pesticide wastewater	Pesticide (Carbofuran)	40/80/120/160/200 ppm	82/84/85/87/89 %	[87]
NF	M-PIP and M-PEI	complex dye-wastewater	A:reactive black 5 (RB-5) B:H-acid	A: 1 g/L B: 0.2 g /L	A: > 95% (both membrane)	[88]

					B: 96.0% (M-PEI)	
NF	cerium oxide-loaded loose nanofiltration polyethersulfone membranes	Real textile wastewater.	A:Color, B:COD, C:Turbidity, D:TDS, E:TSS etc.	A: >1100 (Pt-Co) B: 3460 ± 20 mg/L C: 1340 ± 10 NTU D: 6300 ± 10 mg/L E: 250 ± 5 mg/L	A: 374 ± 20 (Pt-Co) B: 710 ± 12 mg/L C: 12.22±1.22 NTU D: 5950 ± 10 mg/L E:18.0 ± 0.5 mg/L	[89]
NF	cerium oxide-loaded loose nanofiltration polyethersulfone membranes	aqueous dye solutions	A:Congo Red B:Direct Red 23 C:Direct Red 243	100 ppm	A: 99.36% B: 99.53% C: 99.78%	[89]
NF	polyamide composite membranes (NE40, NE70) nonpolyamide composite membranes (MPS-34, MPS-36) Polyethersulfone (PES) based membrane (NP010, NP030)	copper-refining sulfuric acid wastewater	metal ions	More than 2000 mg/L of metal ions such as calcium, copper, iron, and magnesium. (Ca: 74 ppm, Cu:1758 ppm, Fe:429 ppm, Mg:125)	greater than 90% metal ion rejection with NE40, NE70, MPS-34	[90]

3.4 RO current progress discussion

The recent efforts taken by researchers on progress on RO membrane processes are summarized as follow. Pang and Zhang prepared thin film nanocomposite RO (TFN RO) membranes using hydrophilic nanoparticles. The experimental results show salt rejection was improved and it increased up to 98.6% with fluorinated silica nanoparticles at optimal 0.12% (w/v) [91]. Fujioka et al. study evaluated the variable rejection of a N-nitrosodimethylamine (NDMA), produced by fouling in the RO membrane. The fouling substances characterization in the NF treated wastewater found that fouling matters included tryptophan or tryptophan-like matters can increase NDMA rejection [92]. Ghaseminezhad et al. prepared nanocomposite RO membrane from cellulose acetate and graphene oxide with polyester fabric support. The results presented that the graphene oxide concentrations would affect the membrane's porosity, morphology, mechanical strength, hydrophilicity, roughness, and thermal stability. CA membrane modified with 1 wt% GO

reject salt from synthetic seawater was 1.8 times greater compare to pristine CA membrane at 25 bar [93]. Fujioka et al., modified three commercial and one prototype RO membrane using heat treatment to achieve over 90% removal of N-nitrosodimethylamine (NDMA). The experimental outcomes show the use of heat treatment to a prototype membrane lead to high removal of 92% (1.1-log) of NDMA [94]. Sahinkaya et al., studied a pilot scale RO process (using a spiral wound membrane element) with three different operational modes to collect more information. The result showed that the permeate (conductivity 150 $\mu\text{S}/\text{cm}$) produced in the RO process can be recycled in the dyeing process as the feed conductivity was around 5500 $\mu\text{S}/\text{cm}$, at 70% water recovery [95]. Qi and co-authors fabricated the polymersomes-based RO membranes which offered excellent water permeability and water/salt selectivity mainly under feed conditions with high salt concentrations.

This study provides a standard shift in developing new-generation RO membranes for energy and cost-effective desalination processes [96]. Lopera et al. reported a study on the removal of theophylline, amoxicillin, caffeine, theobromine, and penicillin G from WWTP wastewater by RO membrane (BW30-2540). This method attained 100% contaminants removal [97]. Bastos et al., reported work evaluates the rejection of phenols from refinery wastewater by RO. The result show rejections of up to 98% of phenols and 99% of both COD and TOC by RO membrane [98]. The various studies on RO membrane used in wastewater treatment are systematically shown in table 4.

Table 4. Recent progress on RO performance in wastewater treatment.

Process	Membrane details	Wastewater type	Name pollutants	Feed Concentration	Removal % / efficiency/ Output	Ref.
RO	fluorinated silica-polyamide thin film nanocomposite RO membranes	Aqueous solution	NaCl rejection	2000 ppm	98.6%	[91]
RO	ESPA2 Nitto/Hydranautics (Osaka, Japan) and BW30, Dow Chemical Company (Midland, MI, USA),	Secondary wastewater effluent of a municipal wastewater treatment plant	N-nitrosodimethylamine (NDMA),	500 ng/L	< 70%	[92]
RO	Cellulose acetate (CA)/Graphene oxide (GO)	synthetic seawater	NaCl	25000 ppm	90	[93]

	nanocomposite membrane					
RO	heat-treated RO membrane	Aqueous solution	N-nitrosodimethylamine (NDMA)	1000 ng/L	92%	[94]
RO	RO pilot scale unit	biologically-treated textile wastewater	conductivity	5500 μ S/cm	Less than 150 μ S/cm and 70% water recovery	[95]
RO	Polymersomes-based RO membrane	Aqueous solution	NaCl	32000 ppm NaCl	98.9%	[96]
RO	DOW FILMTEC BW30-2540 membrane	Aqueous solution	emerging contaminants (including stimulants and antibiotics)	approximately 1 mg/L for each pollutant	100%	[97]
RO	RO membranes, BW30 and SW30	refinery wastewater	A: Phenol B: COD C: TOC	A: 128 ppm B: 1605 mgO ₂ /L C: 399.4 ppm	A: 98% B: 99% C: 99%	[98]
RO	tangential-flow RO bench-scale membrane plant	olive-oil washing wastewater	COD	188.7 $\pm$ 37.9 mg/L	1.5 mg/L	[99]

4. Challenges and promising solutions in MF, UF, NF and RO membrane processes

The third major research area in MF is membrane fouling. The unwanted deposits create the problems of pore blockage which decrease membrane permeability and selectivity [22]. Majority of the fouling studies involved MF membrane modifications. Gu et al. studied graphene oxide quantum dots (GOQDs) effects on the surface properties of ceramic membranes. The result of GOQDs modified ceramic membranes indicate improved hydrophilicity, water permeability with remain a porous structure and reduced membranes resistance [100]. Ghalamchi and co-authors developed the cost-effective blended MF membranes ZnAlCu-NLDH/polyethersulfone (PES) and gC₃N₄/PES nanocomposite membranes. These two membranes showed significant flux recovery ratio than the pristine PES membrane [101]. Kose-Mutlu and team modified the polymeric MF membranes by adsorption bismuth chelate (BisBAL), which have a high anti-bacterial effect on various microorganisms, on the membrane surface. The modified membranes showed strong resistance to biofouling [102]. Electric membrane bioreactor has significantly attracted interest for wastewater treatment as well as in water purification due to its outstanding

performance in fouling mitigation. The application of ionic liquids in pyrrole oxidative polymerization deals a novel and effective strategy in conductive membrane preparation and helps as an excellent scheme in fouling mitigation in membrane bioreactor [103]. Gu et al. modified membrane surface with alumina presented significant changed in better antifouling properties than in pristine membranes [104]. Zhang et al. developed silver-decorated biomimetic silica nanopollens (SNPs) to modify a polyvinylidene fluoride MF membrane. The modified membrane confirmed compelling antibiofouling performance than the pristine membrane [105]. Zhang and co-authors fabricated a highly antibiofouling membrane. The excellent biofouling resistance was obtained by grafting quaternary ammonium compounds (QAC) onto a silica-decorated membrane using alkoxysilane polycondensation reaction. This membrane showed comparable water permeability, surface roughness, hydrophilicity with the pristine membrane [106].

The fouling remains a major barrier limiting UF applications in treating various waters. Thus, research on UF membrane modification is a growing sustained area of attention. Liu and co-authors prepared PVDF/PFSA-g-GO UF membranes. The prepared membranes (PVDF/PFSA-g-GO) showed greater permeability and antifouling capacity than PVDF membrane [107]. He and co-authors prepared γ -alumina double-layered UF membranes and single-layered UF membranes. The results show both single- and double-layered UF membranes exhibited comparable performance in particle size retention. The double-layered membrane presented better water flux, resistance and anti-fouling properties than the single-layered UF membrane [108]. Zhang and co-authors prepared a supported Ti^{3+} self-doped anatase TiO_2 UF membrane and its photocatalytically enhanced anti-fouling in surface water treatment [109]. Huang et al. prepared UF membranes using magnetic nanoparticles for emulsion treatment. The application of magnetic nanoparticles improved ultrafiltration performance and flux significantly [110]. The biofouling is also a critical issue in the UF membrane technology. To resolve the complex biofouling problem various useful studies were reported in recent years. Khoerunnisa and co-worker reported on the preparation of chitosan/ polyethylene glycol / multi-walled carbon nanotubes / Iodine composite membranes via phase inversion technique. This composite membrane exhibited superior antibacterial activity against *Escherichia coli* (*E. coli*) and *Staphylococcus aureus* (*S. aureus*). Also, significant changes were observed in membranes properties such as the improvement of membrane hydrophilicity, porosity and mechanical strength [111]. Ahmad et al. prepared amine-functionalized zeolitic imidazole framework -8-decorated GO for UF hollow fibre membranes via the phase inversion method. These prepared membranes showed remarkable antifouling properties with a greater flux recovery ratio than the unmodified PES membranes. It also showed antibacterial competence against *Escherichia coli* and *Staphylococcus aureus* [112]. Sathya and Nithya prepared a polyetherimide

composite membrane with tungsten oxide as anti-bacterial agent and pectin as blending additive. The result show that biofouling properties were significantly improved by the addition of metal oxide [113]. Zhang et al. prepared the mixed matrix UF membranes from guanidyl functionalized graphene/polysulfone. This membrane showed superior permeability and prominent antifouling properties, and excellent antimicrobial activity [114].

Research on NF membrane development for improving antifouling properties are the global attention of researcher. Pandey et al. developed fouling-resistant mixed-matrix NF membrane from $Ti_3C_2T_x$ (MXene)/cellulose acetate (MXene@CA) composite. The optimal membrane result indicated a significantly improved hydrophilicity and good antifouling properties [115]. Mi et al. developed effective antifouling and dye desalting NF membrane from zwitterionic functionalized monomer. The membrane exposed excellent antifouling performance with a good flux recovery ratio [116]. Koulivand and co-workers developed a novel mixed matrix NF membrane by using carbon dots blended with a polyethersulfone (PES) matrix. The PES/CDs membrane also presented significant antifouling properties. The carbon dots could be superb membrane modifier for promising wastewater treatment [117].

Shang et al. fabricated a nanostructured NF membrane with using sodium bicarbonate on a porous substrate. This obtained membrane improved fouling resistance was a result of the improved ridge-and valley nanostructure [118]. Vatanpour and Haghghat prepared the polyvinyl chloride NF membranes from Multiwalled carbon nanotubes modified with triethylenetetramine via the phase separation method and immersion precipitation technique. The result indicated the significant reduction in the rate of membrane fouling with prepared membrane [119]. Zhu et al., fabricate a thin-film composite (TFC) NF membrane with incorporation of calcium bicarbonate. The prepared membrane demonstrated outstanding antifouling performance and satisfactory flux recovery after physical cleaning for the optimized membrane [120].

Biofouling is a serious difficulty in RO membrane processes. Li and co-authors prepared thin film nanocomposite RO membranes from GOQD and silver phosphate (AP). GOQD as ideal nanofiller to enhance membrane permeability because its rich hydrophilic groups and AP has higher bactericidal effect than silver particle usually have in membrane applications. The prepared membrane demonstrated strong bactericidal property, good stability and outstanding antifouling performance [121]. The modified a polyamide RO membrane using a polyampholyte composed of cationic [2-(acryloyloxy) ethyl] trimethyl ammonium chloride (TMA) and anionic 2-carboxyethyl acrylate (CAA) via surface-initiated atom transfer radical polymerization. The optimized RO membrane showed an excellent anti-biofouling performance [122].

5. Membrane cleaning techniques and cleaning agents

The various techniques used in membrane cleaning applications are chemical, mechanical, electrical hydraulic, and ultrasonic methods [123–126]. Each of these techniques has advantages and limitations, and should be useful under appropriate conditions [127]. The chemical cleaning method is the most effective approach to improve permeability of membrane and membrane cleaning [128]. Various chemical cleaning agents includes Caustic soda (e.g. sodium hydroxide), Oxidants (e.g. hydrogen peroxide, sodium hypochlorite), acids (e.g. hydrochloric acid, sulphuric acid, citric acid) are used to remove foulants from the membrane surface and to improve the membrane efficiency [129–132].

Conclusions

Membrane separation technology is a capable technology in the field of wastewater treatment. This chapter provides systematic information of recent progress in membrane technology for wastewater treatment for the last three years. This comparative and details review provides the application of different membrane process like MF, UF, NF and RO can be applied in effectively industrial wastewater treatment. The review also includes the problems and challenges associated with the process such as membrane fouling, sensitivity, biofouling and flux recovery which are the main limitation of the current membrane technology. Researchers from worldwide are taking continuing efforts to solve these issues. From the literature, the different materials incorporation in membranes such as in graphene oxide quantum dots, carbon dots, metal oxides, nanomaterials, nanocomposite, calcium bicarbonate, sodium bicarbonate, zwitterionic functionalized monomer, among others were found effective for improved membrane performances. Thus, membrane technology will remain the most effective method for wastewater treatment.

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