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Collagen and Leather

REVIEW **Open Access**

Research recap of membrane technology for tannery wastewater treatment: a review

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Abstract
As a highly complex aqueous effluent, tannery wastewater from leather industry should be treated appropriately before discharging into the environment. Membrane technology has been shown to be a promising approach for tannery wastewater treatment as it may achieve "Zero Liquid Discharge (ZLD)". This work, as the state-of-the-art, attempts to review the world-wide research trends of membrane technologies, the technical recapitulation and recent advances of such technology for tannery wastewater treatment. Generally, manufacture membrane, membrane-based integrated process, MBR, NF, UF and RO are the hotspots in this field. Details of different membrane technologies configured for tannery wastewater treatment, such as membrane materials, scale, membrane modules, operating conditions and removal efficiency of pollutants, are also summarized. It should be noted that membrane fouling is still a major challenge in the membrane technology during tannery wastewater treatment. Therefore, process coupling, either within diverse membrane technologies or between membrane and non-membrane technologies, is considered as a promising alternative to treat the leather tannery wastewater in the future.

Keywords Leather tannery, Wastewater treatment, Membrane technology

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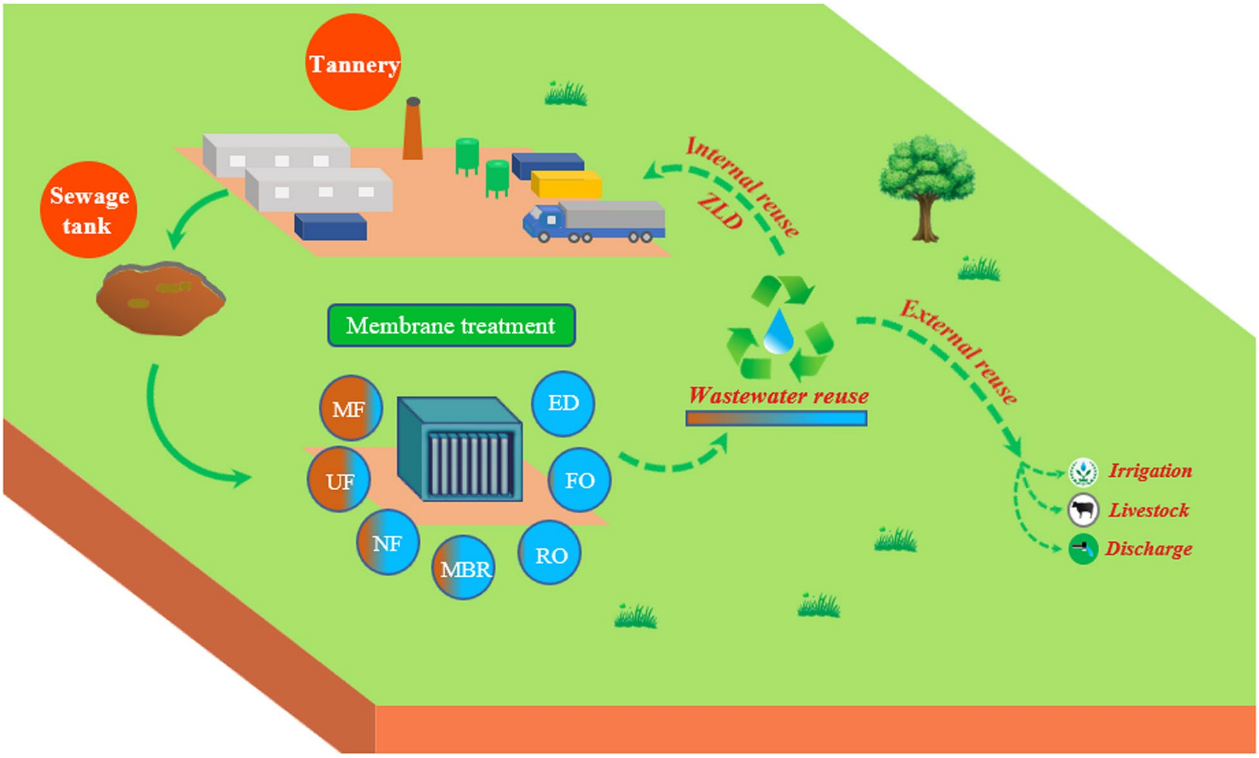
Graphical abstract



1 Introduction

The leather industry and its products play an important role in the world economy. However, a large amount of water is used in the tanning process and about 90% of it is discharged as wastewater [1]. Direct discharge causes serious environmental pollution and is harmful to human health [2, 3]. With the increasing scarcity of water resources, the reuse of wastewater has drawn extensive attention [4, 5]. The complexity of tannery wastewater is caused by the use of hard-to-degrade and even toxic chemicals in different sections of the leather industry, especially the post-tanning process, and the combined tannery wastewater shows high concentrations of Chemical oxygen demand (COD), Biochemical oxygen demand (BOD_5), suspended solids (SS), sulfide, total chromium (Cr), etc. Therefore, the treatment of tannery wastewater is a matter of concern.

Various technologies have been employed to treat tannery wastewater such as gravity separation, air flotation, coagulation, flocculation and biological treatment. According to the literature, these technologies show certain disadvantages such as low efficiency in the removal of inorganic salts, which results in high salt and COD concentrations in the treated wastewater [1, 6, 7]. Consequently, these conventional technologies hardly meet the effluent discharge standards, which are becoming increasingly stringent [8].

Membrane technology has been considered as a prominent alternative for the secondary tannery wastewater advanced treatment thus has significantly developed over the last 20 years because of its potential efficiency, cost-effectiveness and eco-friendliness [9–12] and its ability to achieve "Zero Liquid Discharge (ZLD)". Until now, the membrane-based processes used to treat tannery wastewater include ultrafiltration (UF), microfiltration (MF), nanofiltration (NF), reverse osmosis (RO), forward osmosis (FO), electrodialysis (ED) and membrane bioreactors (MBR). There have been numerous researches reporting various and distinctive findings, while there is no specific review on this topic. Therefore, this work summarizes the performance of different membrane processes with regard of tannery wastewater treatment, which provides guide information for further investigations and applications.

2 Research trends

The investigation of membrane separation technology for the treatment of tannery wastewater was implemented on the Web of science. In the primary search, the following subject terms were considered in the title, abstract and keywords: "tanning wastewater" AND "membrane", "tannery wastewater" AND "electrodialysis", "tannery wastewater" AND "membrane". The search interval was from January 1, 2000 to October 1, 2022, and the search results only include papers.

The search results show that there are more than 110 papers reporting membrane separation for the treatment of tannery wastewater. As shown in Fig. 1, the most investigated topics include MBR, manufacture membrane and NF, while membrane-based integrated process, UF and RO are also the hotspots in this field. As the emerging technologies, electrodialysis and forward osmosis await further investigation.

Figure 2 shows the number of papers and citation frequency per year related to membrane separation for tannery wastewater treatment. Two research peaks were revealed, among which the first one appeared in 2008 and the later one appeared in 2020. The peak of 2008 mainly corresponded to the studies on MBR, while the work of Karahan et al. [3] was the most frequently cited one. This study developed a scientific association between the particle size distribution and biodegradability of tannery wastewater through sequential filtration/ultrafiltration, respirometric analysis and model evaluation. The other notable research peak, appeared in 2020, connected with the studies mainly focusing on manufacturing of new functional membranes to treat the tannery wastewater.

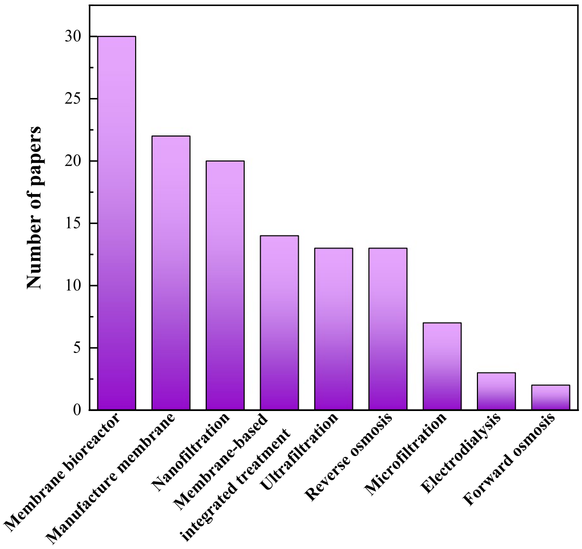


Fig. 1 Topics distribution of the published papers related to "membrane separation" and "tannery wastewater"

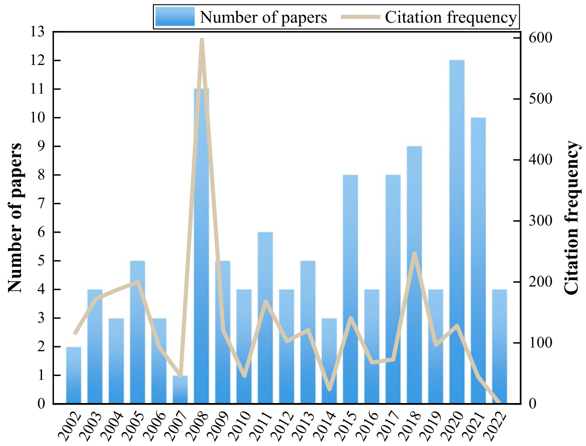


Fig. 2 Number and citation frequency of the published papers per year

3 Research technical recapitulation

3.1 Key parameters of membrane technology

A summary of the extensive literature reveals that the main membrane technologies currently used to treat tannery wastewater are UF, NF, RO and MBR. Membrane material, membrane pore size and operating conditions are crucial factors in membrane technology for treating tannery wastewater. Therefore, in this section, the common configurations of those key parameters were also summarized from the related works. As shown in Fig. 3, the main membrane materials used for tannery wastewater treatment are polyether sulfone (PES) and polyamide since the two membranes show good heat resistance, pressure resistance and corrosion resistance. Meanwhile, the mainly choices of membrane pore sizes for UF, NF and RO are 50/100/150 kDa, 150–300 Da and < 100 Da respectively. The spiral-wound is the most commonly used membrane module for all these three membrane technologies. Operating pressures in descending order are RO, NF, UF. As for MBR, hollow fiber membrane is the most used module due to its high compacity and resistance to fouling. Hydraulic retention time (HRT) usually ranges from 12 to 70 h and sludge retention time (SRT) usually ranges from 30 to 150 days, depending on the production scale. Temperature is usually maintained at the optimum temperature for sludge microorganisms growth: 25–37 °C.

3.2 Pollutants removal efficiency

The main components of the integrated tannery wastewater are shown in Table 1, including COD, BOD, SS, Cr, sulfide and chroma. The concentration of pollutants varies depending on the tannery process. The wastewater quality of the different tanning sections is shown

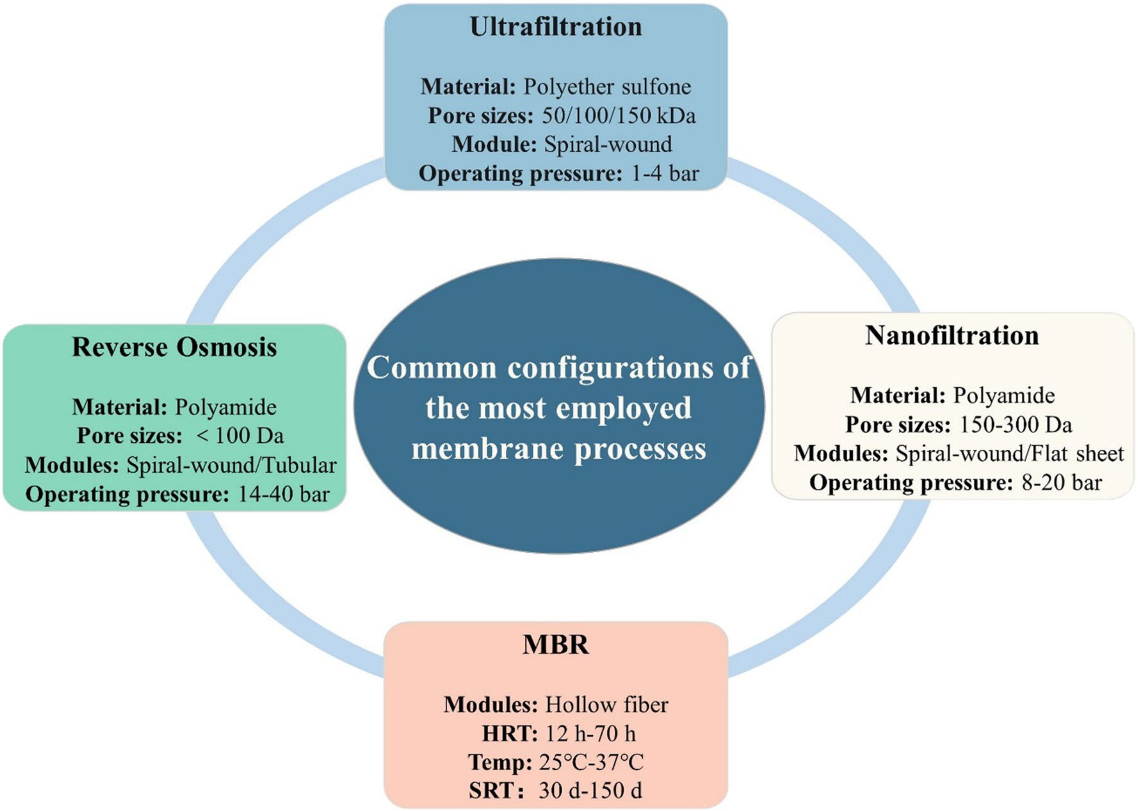


Fig. 3 Common configurations of the most employed membrane processes

Table 1 The typical composition of the integrated tannery wastewater

Index	COD	BOD_5	Suspended solids	Sulfide	Total Cr	Chroma (times)
Concentration (mg/L)	3000–4000	1500–2000	2000–3000	50–100	60–100	600–3500

Table 2 Wastewater quality of different tannery sections (Adapted from Dixit et al.[73])

Pollution load	Processing operation (load kg/ton of raw hide/skins)				
	Soaking	Unhairing/liming	Deliming and bating	Chrome tanning	Post-tanning
TSS	11–17	53–97	8–12	5–10	6–11
COD	22–33	79–122	13–20	7–11	24–40
BOD	7–11	28–45	5–9	2–4	8–15
Cr	–	–	–	2–5	1–2
Sulfides	–	3.9–8.7	0.1–0.3	–	–
NH_3-N	0.1–0.2	0.4–0.5	2.6–3.9	0.6–0.9	0.3–0.5
TKN	1–2	6–8	3–5	0.6–0.9	1–2
Chlorides	85–113	5–15	2–4	40–60	5–10
Sulfates	1–2	1–2	10–26	30–55	10–25

in Table 2. Various available membrane technologies have been applied to treat tannery wastewater, and they were shown with different removal efficiencies for tannery wastewater pollutants. Figure 4 recapitulates the removal efficiencies of major pollutants in tannery wastewater for different membrane technologies based on the data in Tables 3, 4 and 5, which was summarized from the literature. Specifically, MF is not efficient in removing COD (54.7%) and BOD (66.7%), but it can deliver a fairly good removal efficiency for SS and Tan, with 85% and 75%, respectively. As for UF, the removal efficiency of COD, SS and vegetable tannins is close to that of MF, and the removal efficiency of Cr and sulfates is only 42% and 33.1% respectively. Both NF and RO have high removal efficiencies for Cr (>95%) and sulphate (>97%). In addition, RO is much more efficient at removing COD (97%) and BOD (BDL) than NF. The removal efficiency of MBR was 83.5% for COD and 87.8% for BOD. However, the removal of Cr is not as efficient as NF or RO. UF–NF–RO was used to assess the efficiency of the integrated process, resulting in high pollutant removal efficiencies of more than 95%. As for ED, the removal efficiency for ion species is above 98.5% and this process is generally used for the final treatment of wastewater.

3.3 Adaptable cases for leather tannery process effluent treatment

This section summarizes the main pollutants in the wastewater from the different tannery sections, then provides technical recommendations for the treatment of wastewater from the different sections based on the technical characteristics of different membrane processes.

The pollutants in the effluent from each tannery section are varied, thus appropriate membrane technologies are required for the removal of these pollutants. As shown in Fig. 5, MF and UF membranes have relatively large pore sizes and are generally used for pretreatment of the secondary tannery wastewater to retain the large molecules, such as suspended solids, fats and proteins. They are suitable for bating, deliming, degreasing and tanning sections effluent treatment. NF and RO retain small molecules and are highly efficient at removing pollutants from tannery wastewater, but they require higher operating pressures than MF and UF, which are suitable for the treatment of ammonium salts, tannins, chromium salts, aldehydes and dyes produced in bating, deliming, pickling, tanning, dyeing sections. FO and ED enable further desalination, concentration, separation and purification of tannery wastewater, but the draw solution for FO needs to be regenerated, e.g. by using membrane distillation technology. Alternatively, high salinity water that needs to be desalinated can be used as the draw solution, e.g. seawater. ED requires high raw water purity, so pre-treatment of tannery wastewater is necessary before entering into this process. FO and ED are suitable for the treatment of inorganic salts such as ammonium and chromium salts produced in bating, deliming and tanning sections. As for MBR, such process has advantages of high quality and stable effluent quality, small residual sludge production, small footprint and easy automation control in the treatment of tannery wastewater. It also

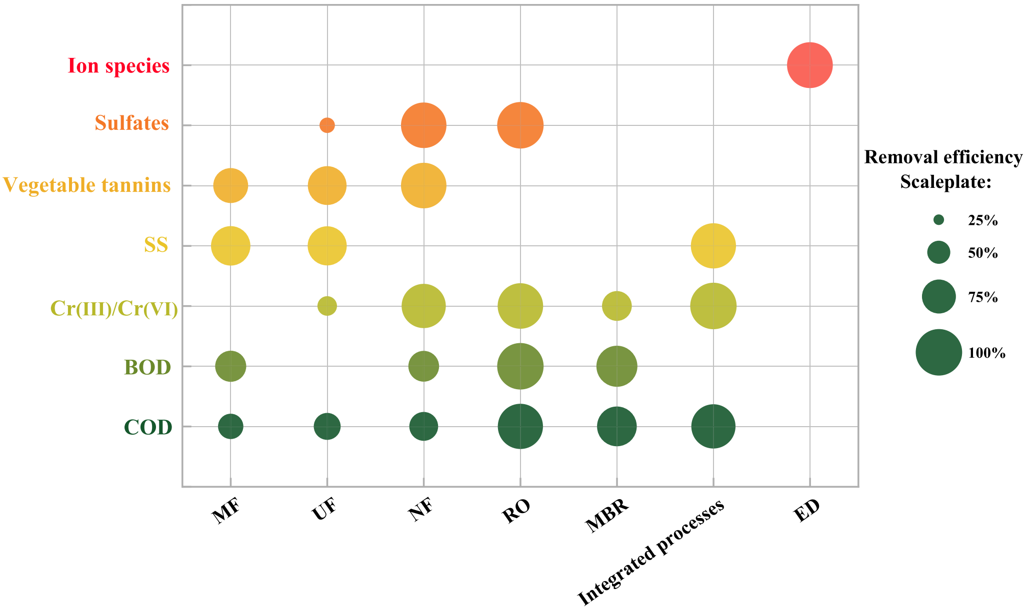


Fig. 4 Removal efficiency of different membrane separation technologies for major pollutants

Table 3 Applications of MF and UF for tannery wastewater treatment

Type	Membrane Material	Scale	Membrane module	Operating condition	Main parameter (s)	Removal efficiency	Reference
Microfiltration	Polyether sulfone	Laboratory	—	Temp = 20 °C TMP = 1.5 bar	COD, TN*	COD (44.5%), TN (29%)	[15]
Microfiltration	Ceramic	Laboratory	–	Temp = 20 °C TMP = 1.6 bar	SS, Turbidity COD, BOD	SS (92%), Turbidity (98%) COD (57.5%), BOD (66.7%)	[13]
Microfiltration	Polyvinylidene fluoride	Pilot	Tubular	TMP = 2.2 bar	Vegetable tannins, Total solids	Vegetable tannins (75%), Total solids (78%)	[39]
Microfiltration	Ceramic	Laboratory	Tubular	TMP = 1.0 bar	COD	62%	[38]
Ultrafiltration	Polyether sulfone	Laboratory	Cartridge	HRT = 4 h Temp = 20 ± 0.5 °C TMP = 10 bar	COD, Cr(III)	COD (39%), Cr(III) (34%)	[17]
Ultrafiltration	Polyether sulfone Poly phenyl sulfone	Laboratory	–	Temp = 25 °C TMP = 3 bar	Dye	99.65%	[74]
Ultrafiltration	Polysulphone	Laboratory	Spiral-wound	Temp = 25 °C TMP = 1.7 bar	COD, Cr(III) SS, Fat	COD (9.5%), Cr(III) (2.2%) SS(84%), Fat (70.5%)	[16]
Ultrafiltration	Polyether sulfone Poly phenyl sulfone	Laboratory	—	TMP = 1 bar	Dye	96.62%	[75]
Ultrafiltration	Polyether sulfone	Laboratory	Flat sheet	Temp = 25 °C TMP = 1.0 bar	COD, Fat	COD (48.2%), Fat (94.5%)	[14]
Ultrafiltration	Polysulfone	Laboratory	–	HRT = 4 h TMP = 5 ba	Vegetable tannins	83%	[76]
Ultrafiltration	Polyethersulphone	Laboratory	—	TMP = 8 bar Temp = 20–25 °C	TOC	58%	[77]
Ultrafiltration	Cellulose-triacetate	Pilot	Spiral-wound	TMP = 3.8 bar	COD, Cr, Sulfate	COD (67%), Cr (50%) Sulfate (33.1%)	[39]
Ultrafiltration	Polyether sulphone	Laboratory	Cartridge	TMP = 2 bar Temp = 25 °C	COD	15.7%	[40]
Ultrafiltration	Ceramic	Laboratory	Tubular	Temp = 20 ± 2 °C TMP = 1, 2, 4 bar	COD	58–90%	[78]

* Total nitrogen

should be noted that MBR has high investment costs and is more difficult to clean when the membrane is contaminated by organic or inorganic pollution. MBR is suitable for the treatment of wastewater in the sections with high ammonia nitrogen content, such as bating and deliming. Otherwise, the wastewater in most of the tanning sections show high levels of COD and BOD, which can be effectively removed by RO or MBR.

4 Recent research advancements

In this section, recent advancements on membrane process for tannery wastewater treatment are elaborately described according to the literature. Through the recapitulative details compiled in Tables 3, 4 and 5, these processes can be divided into three general categories: (i) pressure-driven processes, such as: MF, UF, NF and RO; (ii) non-pressure-driven processes, for instance, FO and ED, (iii) Biological-based membrane technology process (MBR) and the integrated or coupled processes of various membrane-based processes.

4.1 Pressure-driven membrane processes

4.1.1 Treatment of tannery wastewater by MF/UF

Compared with NF and RO, the operating pressures are lower and the membrane pore sizes are larger in the process of MF and UF. Under a pressure-driven process, small-sized substances such as water, organic low molecules and inorganic ions in the solution can pass through the micro-pores, while large-sized substances such as bacteria, colloids, particulates and organic macromolecules in the solution cannot pass through the membrane and are trapped.

When subjected to MF and UF, most suspended solids (94%) [13] and fat substances (94.5%) [14] could be removed from the tannery wastewater. Gallego et al. [15] treated the wastewater from the deliming/bating operations by MF, which achieved the removal ratio for COD

Table 4 Applications of NF and RO for tannery wastewater treatment

Type	Membrane material	Scale	Membrane module	Operating condition	Main parameter (s)	Removal efficiency	Reference
Nanofiltration	Polyamide	Laboratory	Cartridge	HRT = 4 h Temp = 18 ± 0.5 °C TMP = 20 bar	COD, Cr(III)	COD (67%), Cr(III) (95%)	[17]
Nanofiltration	Cellulose acetate	Laboratory	Flat sheet	HRT = 3 h Temp = 25 ± 1 °C TMP = 14 bar	Cr(III)	90.2%	[79]
Nanofiltration	Synthetic polymer	Laboratory	Spiral-wound	Temp = 25 °C TMP = 5 bar	COD	71.5%	[80]
Nanofiltration	Polyamide	Laboratory	Spiral-wound	Temp = 25 °C TMP = 14 bar	COD, Cr(III)	COD (51%), Cr(III) (97%)	[16]
Nanofiltration	Polyamide	Laboratory	Flat sheet	Temp = 25 °C	COD, Cr(VI)	COD (67%), Cr(VI) (99.9%)	[81]
Nanofiltration	Polyamide	Laboratory	–	Temp = 25 °C TMP = 14 bar	Cr(III), Cl	Cr(III) (98%), Cl (22%)	[82]
Nanofiltration	–	Laboratory	–	Temp = 25 °C TMP = 20 bar	Vegetable tannins	97.6%	[83]
Nanofiltration	–	Pilot	Spiral-wound	HRT = 24 h Temp = 26 °C	Cr, Sulfate	Cr (99%), Sulfate (97%)	[84]
Nanofiltration	–	Laboratory	Spiral-wound	Temp = 25 °C TMP = 4 bar	Sulfate, Organic matter	Sulfate (98.7%), Organic matter (85.6%)	[85]
Nanofiltration	–	Laboratory	Spiral-wound	Temp = 25 °C TMP = 20 bar	Sulphate	99.5%	[56]
Nanofiltration	Cellulose	Laboratory	–	TMP = 2 bar	Cr(VI)	87%	[86]
Nanofiltration	–	Laboratory	Flat sheet	TMP = 14 bar	Cr, Sulfate	Cr (96%), Sulfate (96%)	[87]
Nanofiltration	Polyether sulfone	–	–	TMP = 4 bar HRT = 30 min Temp = 25 ± 1 °C	Dye	90%	[88]
Nanofiltration	Polyamide	Laboratory	–	TMP = 8 bar Temp = 20–25 °C	TOC	78%	[77]
Nanofiltration	Keratin-poly sulfone	–	–	TMP = 30 psi	COD, BOD	COD (53%), BOD (66%)	[89]
Reverse Osmosis	–	Laboratory	Tubular	HRT = 55 h Temp = 25 °C TMP = 0.82 MPa	Cr(III)	99%	[90]
Reverse Osmosis	–	Laboratory	Spiral-wound	TMP = 20 bar Temp = 25 °C	COD	98%	[40]
Reverse Osmosis	Polyamide	Laboratory	–	TMP = 40 psi	Cr, N	Cr (99%), N (89%)	[91]
Reverse Osmosis	Polyamide	Pilot	–	TMP = 14.65 bar	Cr	84%	[92]
Reverse Osmosis	Cellulose acetate	Pilot	–	TMP = 7 bar	Cr(VI)	99.80%	[93]
Reverse Osmosis	Polyamide	Laboratory	Spiral-wound	TMP = 7 kg/cm^2	COD, BOD	BDL**	[94]
Reverse Osmosis	–	Laboratory	Tubular	TMP = 40 bar	Cr(III)	99.9%	[95]
Reverse Osmosis	Polyamide	Laboratory	Cartridge	HRT = 4 h Temp = 20 ± 0.5 °C TMP = 21 bar	COD, Cr(III)	COD (95%), Cr(III) (100%)	[17]
Reverse Osmosis	Polyamide	Pilot	Spiral-wound	TMP = 14 bar	COD, Cr, Sulfate	COD (95%), Cr (100%) Sulfate (100%)	[39]

**Below the detection limit

(44.5%) and total nitrogen (29%). Similarly, the polysulphone UF membrane treated the wastewater from the chrome tannery stage with a removal efficiency of 9.5% and 2.2% for COD and Cr(III) respectively [16]. Those works indicate that MF and UF are not quite efficient for the removal of COD, total nitrogen and Cr(III).

Table 5 Applications of integrated processes, ED and MBR for tannery wastewater treatment

Type	Membrane material	Scale	Membrane module	Operating condition	Main parameter (s)	Removal efficiency	References
UF–NF–RO	–	Laboratory	Tubular	TMP = 30 bar	Cr(VI)	99.90%	[96]
UF–NF–RO	PES (UF) Polyamide (NF, RO)	Laboratory	–	pH = 4	COD, Cr(III), SS	COD (95%), Cr(III) (100%), SS (97%)	[17]
NF–RO	TFC (NF, RO)	Laboratory	Tubular Laminar	HRT = 1 h	Dye, COD, BOD, Cl	Dye (100%), COD (99.8%), BOD (99.8%), Cl (96%)	[24]
NF–RO	Polyimide (NF) Polyamide (RO)	Pilot	Spiral-wound	Temp = 20 ± 1 °C	COD, Cr	COD (95.7%), Cr (99.98%)	[35]
UF–NF–ED	PVC(UF)	Laboratory	–	TMP = 2 bar Temp = 26–28 °C pH = 6–7	Inorganic salt	Desalination rate: 61.9%	[30]
FO–NF	Polyamide (NF) Cellulose triacetate (FO)	Laboratory	Flat sheet	$C_{Nacl} = 0.8$ M	COD, Chloride, Sulfate	COD (98.5%), Sulfate (98.2%), Chloride (97.2%)	[37]
MF–NF–RO	Polyamide (NF, RO)	Pilot	Spiral-wound	Temp = 25–30 °C pH = 7.0–8.5	TDS	> 98%	[97]
ED	CMT and AMV	Pilot	–	Potential = 8 V	Ion species	> 98.5%	[31]
ED	Nafion® 324 and AMX	Laboratory	–	pH = 2.5 Current density = 150 A m^{-2}	Separation of Cr(III)	Separated	[32]
MBR (UF)	Polyether sulfone	Pilot	Tubular	HRT = 12–20 h Temp = 25–37 °C TMP = 3 bar	COD, Cr(III)	COD (88%), Cr(III) (74%)	[98]
MBR	Cellulose ester	Laboratory	Flat sheet	HRT = 6.25 h TMP = 0.6 bar Temp = 27–30 °C	COD, BOD, Total-P, Cr	COD (45.8%), BOD (87.8%) Total-P (66.7%), Cr (50%)	[99]
MBR	–	Pilot	Hollow fiber	HRT = 70 h Temp = 20 °C SRT = 150 d	COD, Phenol	COD (79%), Phenol (74.5%)	[34]
MBR	–	Pilot	Hollow fiber	HRT = 70 h SRT = 50 d	COD	79.90%	[100]
MBR (MF)	Polyvinyl difluoride	–	Hollow fiber	TMP = 5 kPa	COD, Cr	COD (90%), Cr (67%)	[101]
MBR	Polyvinylidene fluoride	Pilot	–	HRT = 40 h	COD	90%	[33]
OMBR (FO)	Cellulose triacetate	Laboratory	Flat sheet	Flow rate = 30 L·h^{-1}	COD	98%	[29]
OMBR (FO)	Cellulose triacetate	Laboratory	–	HRT = 2.86–13.78 d	COD	64.53–85.65 ± 5.0%	[102]
MBR	Polyvinylidene fluoride	Laboratory	Hollow-fiber	SRT = 30 d	COD, TN	COD (87 ± 14%), TN (43 ± 10%)	[103]
MBR	–	Laboratory	Hollow-fiber	HRT = 24 h	Ammonium, COD, TN	Ammonium (100%) COD (90%), TN (60–90%)	[104]
MBR (UF)	Polyether sulfone	Laboratory	Tubular	linear velocity = 1.8 m/s	COD, TN, TP	Oxic: COD (87.5%), TN (44%) TP (51%) Anoxic/oxic: COD (91.9%), TN (85.8%) TP (39.0%)	[105]

Membrane pore size and operating conditions have a significant effect on pollutant removal efficiency and membrane flux in tannery wastewater treatment. Berna et al. [17] studied three UF membranes made of the same material but with different pore sizes (20, 50, and 150 kDa), and found that foulants removal efficiency decreased with pore size increasing regardless of the operating conditions. Yang et al. [14] found that an increase in shear rate at the UF membrane surface increased the membrane steady-state flux within a certain range, which implied that increasing the shear rate alleviated membrane fouling. In addition, the authors analyzed the membrane fouling behavior of UF process

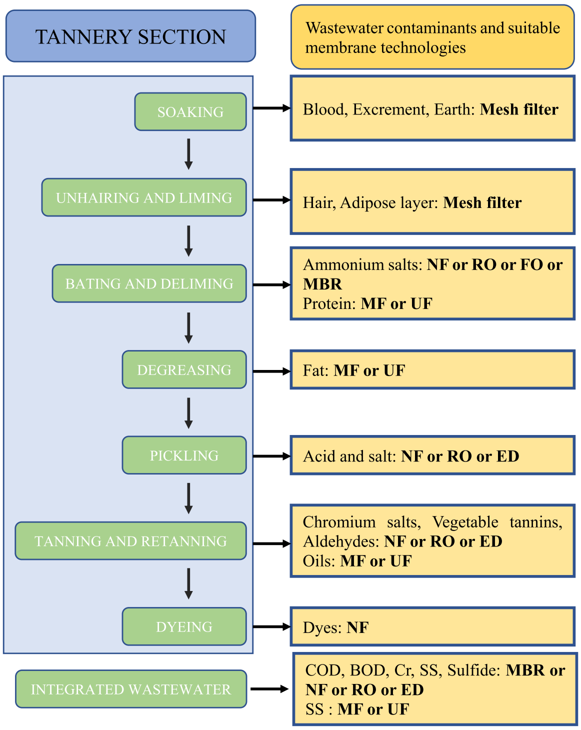


Fig. 5 Contaminants in wastewater from different tannery sections and suitable membrane treatment technologies

for the treatment of tannery wastewater based on the theoretical models.

In fact, membrane surface charge, pore size and morphology are the main factors contributing to membrane fouling during MF/UF. Current MF/UF membranes have a relatively wide pore size distribution (PSD). Membranes with large differences in pore size distribution are more susceptible to contamination, as the largest pores initially carry a disproportionate amount of flow, making them susceptible to clogging [18]. The isoporosity of MF/UF membranes is therefore a key objective of membrane technology. In order to achieve high water permeability, a high pore density (pores/area) is also required. Hydrophilicity usually makes membranes less prone to fouling and easier to clean, therefore, different efforts have been reported to prepare the 'anti-fouling' membranes based on increasing the membrane hydrophilicity [11, 19–21].

4.1.2 Treatment of tannery wastewater by NF/RO

NF has a range of membrane pore sizes between RO and UF membranes and has a high removal performance for divalent and multivalent ions and organic matter with molecular weights between 200 and 1000, while the removal efficiency for monovalent ions and small molecules is lower than that of RO. Furthermore, the operating pressure of the NF process is often lower than that of RO. As one of the most sophisticated membrane separation processes, RO can block all dissolved salts and organic matter with molecular weight greater than 100 but only allows water molecules to pass through. Consequently, the desalination rate of RO membranes is generally higher than 95%.

Berna et al. [17] tested the efficiency of NF and RO for the removal of pollutants from UF-treated chrome tannery wastewater. It was observed that the removal efficiency of NF for COD, Cr(III), Na^+, and SO_4^{2-} was 67%, 95%, 25%, and 92%, respectively, while the removal efficiency of RO for these pollutants was 95%, 100%, 99%, and 100%, respectively. In addition, a comparative cost assessment claimed that chemical precipitation units had higher investment costs but lower operating costs compared to membrane process. Moreover, the quality of the recovered chromium was lower after chemical treatment due to the presence of organic matter, metals and other contaminants. Therefore, membrane technology is considered more feasible in respect of process and quality of recovered chromium than the existing technologies for treating chrome tannins [22, 23].

Similar claim of high removal of pollutants was also reported by the process of NF followed by RO [24]. Those works suggested that RO exhibited extremely high removal efficiency for various pollutants and it often required an upstream treatment of tannery wastewater.

Otherwise, Ortega et al.[25] utilized NF membranes to treat acidic leachates from polluted soils, which exhibited high retention efficiencies, particularly for higher valent ions. This work has certain guiding significance for the treatment of residue produced in leather industry.

4.2 Non-pressure-driven processes: FO and ED

FO is the process during which water transfers across a selectively permeable membrane driving by the gradient of chemical potential (or osmotic pressure). Feed solution (FS) with lower osmotic pressure and draw solution (DS) with higher osmotic pressure are placed on each side of the selective permeability membrane [26].

As an emerging membrane technology, FO technology is presently in the early stage of industrial development. It has attracted significant attention in recent years for treating wastewater [27]. Compared to conventional pressure-driven membrane processes, FO has the advantages of low membrane fouling tendency, low energy consumption and high pollutant retention rate [28]. In the FO system, as depicted in Fig. 6, water passes through the FO membrane and flows from the feed side to the draw side. Pollutants in the tannery wastewater are trapped by the membrane and during dewatering they are retained in the feed solution or on the membrane surface. As a result, the volume of the feed solution is reduced and the

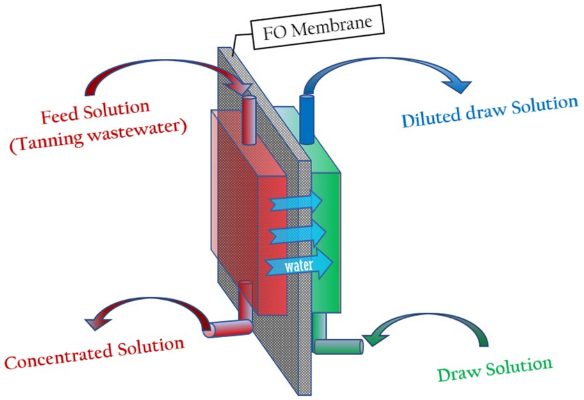


Fig. 6 FO membrane process for the treatment of tannery wastewater

concentration is increased, which facilitates the subsequent treatment, e.g. biological treatment.

As shown in Table 5, relatively few researches on the use of FO technology to treat tannery wastewater has been reported. Lujan-Facundo et al. [29] employed an external cross-flow FO membrane bioreactor (Fig. 7c) for the treatment of tannery wastewater. They used actual wastewater from the ammonia separation absorption column as the draw solution, and focused on reverse salt flux, biomass characteristics, water flux and membrane fouling. They found that COD removal ratio was maintained at around 80% during the first 50 days of operation of the osmotic membrane bioreactors (OMBR) system. In addition, the contact angle of the fouled and virgin FO membranes was evaluated and showed that the presence of microbial residues or contaminants on the membrane surface can alter the membrane properties to make it more hydrophobic. Energy Dispersive X-ray (EDX) analysis was also carried out on the fouling FO membrane, which confirmed that organic contaminants were predominant.

Electrodialysis, a membrane separation operation driving by the potential difference that employs the ion exchange membranes of selective permeability to remove or enrich electrolytes from a solution, began to be applied in the operation of wastewater desalination. Liu et al. [30] applied an integrated UF-NF-ED process for the treatment of tannery wastewater, and electrodialysis desalinated the NF filtered out wastewater, achieving an overall desalination rate of 61.9%. Meanwhile, it has been shown that electrodialysis has an extremely high removal efficiency for all ionic species present in tannery effluents [31]. Lambert et al. [32] used a modified electrodialysis membrane to separate chromium from simulated tannery wastewater. The results showed that the separation of trivalent chromium and sodium ions was feasible. As electrodialysis mainly removes ions, large molecules

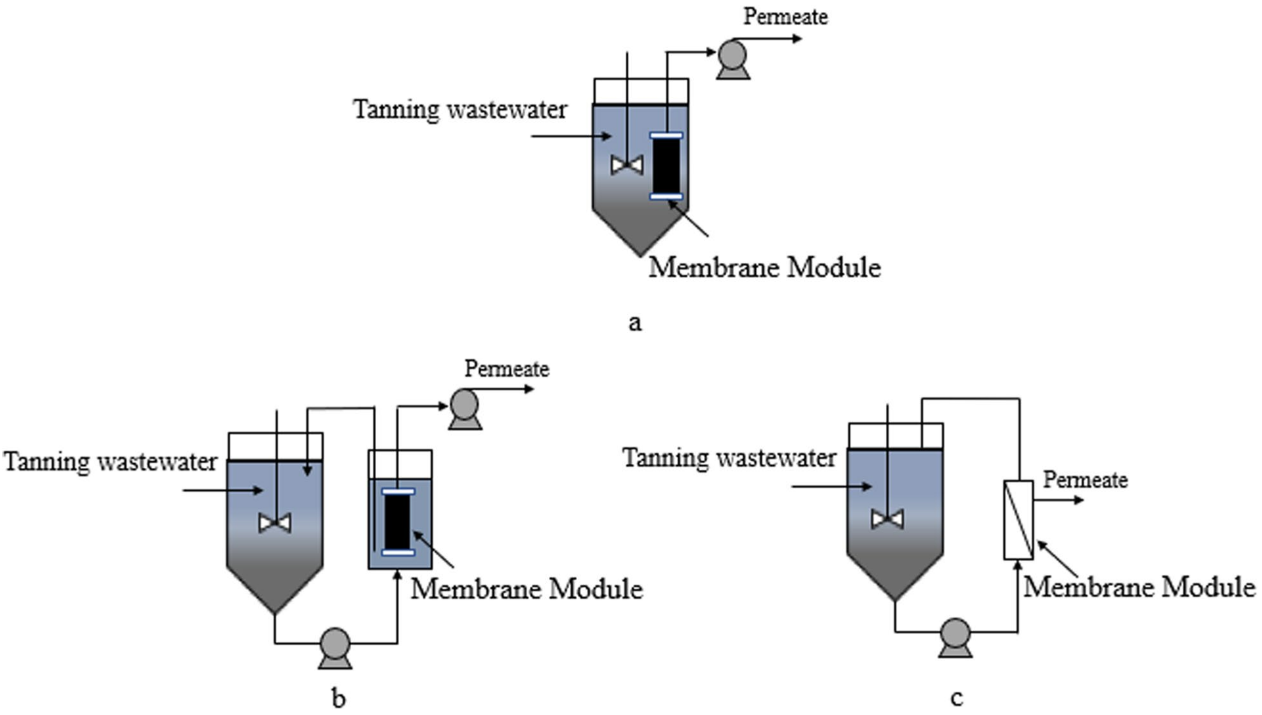


Fig. 7 Membrane bioreactor configuration for the treatment of tannery wastewater: **a** Internal submerged type, **b** External submerged type, **c** External cross flow type

(suspended matters, fats and proteins) may affect the separation efficiency, therefore, it is necessary to pretreat actual wastewater before being imported to such process.

4.3 Membrane bioreactor

MBR is a powerful water treatment technology that combines a membrane separation unit with a biological treatment unit. In such process, the traditional secondary sedimentation tank is replaced with a membrane module where a high concentration of activated sludge is maintained, allowing to reduce the footprint of the wastewater treatment facility and the amount of sludge. Compared with traditional biochemical water treatment technology, MBR has the following main features: high treatment efficiency, good effluent quality; compact equipment, small footprint; easy to achieve automatic control, simple operation and management. As shown in Fig. 1, MBR is currently the most studied technology in relation to membrane technology for the tannery wastewater treatment.

There are three types of membrane bioreactor configurations for tannery wastewater treatment (Fig. 7): internal submerged, external submerged, external cross-flow, which are defined by the relative position of the membrane to the bioreactor. Internal submerged is an alternative to external membrane bioreactors to reduce energy costs: by immersing the membrane in the bioreactor, the energy required to recirculate the feed solution is eliminated. However, internal membrane bioreactor fouling is a major challenge compared to external configurations. For external cross-flow mode, the membrane is connected to a bioreactor and the feed solution is circulated between the two vessels driving by pumps. This mode is usually used for continuous product recovery. Umaiyakunjaram et al. [33] investigated the treatment performance of internal submerged anaerobic membrane bioreactor on high suspended solids raw tannery wastewater, which achieved high COD removal rate (90%) and biogas yield. External submerged mode obtained high removal efficiency of COD (79%) and phenol (74.5%) [34]. Luján-Facundo et al. [29] concluded that external cross-flow mode was efficient for COD removal from tannery wastewaters.

4.4 Coupled membrane processes

As precedently described, the removal efficiency of pollutants varies considerably among the different membrane technologies. MF and UF have higher retention efficiency for large molecules due to their larger pore size, but not for small molecules. NF is more effective than MF and UF in removing small molecule contaminants such as inorganic salts and tannins. RO has high retention efficiency for the majority of pollutants, but it often needs an upstream treatment. Therefore, the coupling of different membrane processes is often adopted for the treatment of tannery wastewater.

A summary of the literature (Table 5) shows that the most commonly studied membrane coupling technology is NF–RO [24, 35, 36]. Stoller et al. [35] utilized NF–RO for tannery wastewater treatment and found that the content of contaminants in the RO permeate was under the discharge limit. The membrane coupling technology was also compared with conventional biological processes from the technical and economic aspects, which showed that NF–RO reduced the total cost by 21% under optimized conditions [35].

Parimal et al. [37] used the FO-NF integrated system for the pilot treatment of tannery wastewater and found that the removal efficiency of pollutants (COD, chloride, sulphate) was higher than 97%. The economic viability of the system has also been confirmed and it is considered promising for industrial scale wastewater treatment.

In addition, membrane coupling technologies that have been reported in the literature include UF–NF–RO [17], MF–UF [38], MF–UF–RO [39], UF–NF–ED [30], UF–RO [40]. Kiril et al. [17] used an integrated UF–NF–RO system to treat chrome tanning wastewater and found that the UF process removed 72%, 39% and 34% of SS, COD and Cr respectively. 91%, 67% and 95% of SS, COD and Cr were removed by the NF process and 97%, 95% and 100% of these three pollutants were removed by the RO process. Moreover, the permeate from this integrated process can be directly discharged or reused in the tannery process. Liu et al. [30] utilized an integrated UF-NF-ED system to treat tannery wastewater and found that the UF process achieved 96.5%, 53.7% and 45.8% removal of turbidity, chroma and COD respectively. 90% recovery of fresh water was achieved by the NF process and 61.9% desalination was achieved by the ED process. The results show that the coupled process can achieve the reuse of the treated tannery wastewater.

The coupled membrane process allows the advantages of different membrane technologies to be fully exploited, not only for high overall removal of pollutants, but also to reduce the load on the back-end membrane technology. Therefore, the coupling of membrane processes for wastewater treatment is also one of the future trends.

5 Non-membrane-based technologies for the treatment of tannery wastewater

At present, non-membrane-based technologies are the main approaches for tannery wastewater treatment. Generally, non-membrane-based technologies can be divided into advanced oxidation processes (AOPs), biological treatment, adsorption, and coagulation/flocculation. However, it is often difficult for these technologies to achieve ZLD or direct emission standards, therefore, the coupling of them and membrane processes could be a trend in the future. Here below presents several technical approaches which are considered promising as coupled with membrane processes.

AOPs, combined with the use of electricity, light irradiation, catalysts and oxidants, can oxidise and degrade large, non-degradable organic substances in wastewater into small, low or non-toxic substances, or even directly into CO_2 and H_2O, which approaches complete mineralization [41, 42]. However, due to the high complexity of tannery wastewater, there is a potential risk of releasing intermediate compounds that are probably more toxic than the original compounds after treatment with AOPs.

Biological treatment occupies a prominent position among the various wastewater treatment approaches, among which the aerobic process is dominant, especially activated sludge [43]. Moving-bed biofilm reactor (MBBR) and biological contact oxidation have been studied as new methods for the biological treatment of tannery wastewater [44].

Adsorption is the most common method of the physicochemical treatment, which is commonly used to treat heavy metals [45], aromatic compounds and dyes wastewater [46]. A systematic review [47] mentioned that tannery solid waste and sludge from the treatment of tannery wastewater can be used to prepare low-cost adsorbents, which can effectively improve the adsorbent's performance on chromium, dyes and other pollutants in tannery wastewater by adjusting temperature, pH and adsorbent dosage.

Coagulation/flocculation is one of the most widespread methods for the pretreatment of tannery wastewater [48, 49], and chemical flocculants are mostly used in wastewater treatment. The use of some chemical flocculants causes secondary pollution to water bodies, so research and development of non-toxic and non-hazardous flocculants are necessary.

6 Challenges and future perspective

6.1 Challenges

Although membrane separation technology for the treatment of tannery wastewater has been extensively researched on a laboratory scale, industrial applications still face some major challenges such as membrane fouling.

In the membrane process of tannery wastewater treatment, the membrane was fouled by impurities in the feed solution, which results in the decline of membrane permeability and further impacts membrane service time, filtration time, operating temperature and applied transmembrane pressure (TMP) [50]. In many instances, cake layer formation was considered as the major contributor to membrane fouling in the membrane process [51]. Polysaccharides and proteins are major contributor to formation and growth of the cake layer [27].

The fouling formation could be speculated according to the related reports, depicted in Fig. 8. When the tannery wastewater transits across the membrane as the separation occurs, the foulants smaller than the membrane pore size would smoothly pass through the membrane along with the penetrating fluid [52]. In contrast, the foulants similar or bigger than the surface pore size would cause blocking or adhesion in the membrane pores [51], thereby giving rise to membrane internal fouling. The cake layer develops subsequently. Studies have shown that the formation and growth of the cake layer on the membrane surface could be considered as three

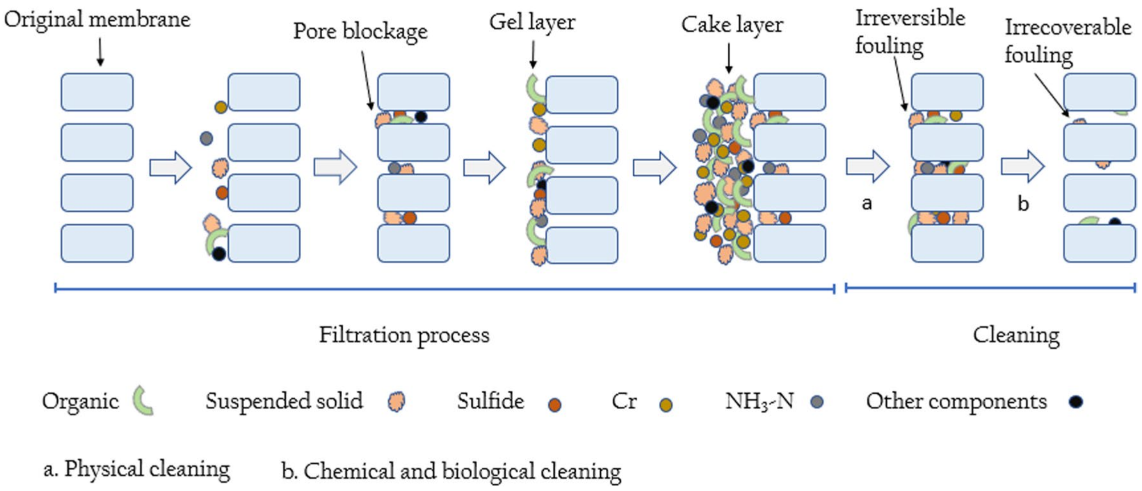


Fig. 8 Membrane fouling process in filter process of tannery wastewater

phases [53]. In the first phase, EPS adhered to the membrane surface, and various substances (e.g. proteins, fats, SS) [54] could be implanted in the cake layer to form bunches. Afterwards, EPS and microorganisms increased rapidly, and the cake layer grows at a rapid rate in this stage. In the end, the biovolume growth rate went down and formed the filter cake. The permeate flux declines and membrane resistance increases with the formation of the cake layer [54, 55]. To summarize, membrane pore blocking and cake layer deposition on membrane surface are the major factors causing membrane fouling.

In order to mitigate the membrane fouling in the membrane process, several methods have been reported, such as the membrane cleaning, the anti-fouling modification, the pretreatment of feed effluents and the optimization of operating conditions, etc. [56–59].

Physical cleaning approaches include ultrasonic cleaning, forward and reverse flushing, backwashing, air flushing, sponge ball cleaning, electrical fields and magnetic fields [60, 61]. However, physical cleaning could only remove most of the membrane surface foulants, but the membrane pore foulants (internal fouling) are hard to be fully removed. Intermittent chemical cleaning could effectively dissolve and leaching-out foulants in the membrane pores, e.g. organic matter and oils (alkalis), insoluble salts (acids), proteins and polysaccharides (enzymes) [62] (Fig. 8). Chemical coagulation is diffusely applied to mitigate the membrane fouling [63–65]. In chemistry cleaning, the optimal choice of the cleaning reagent is crucial, which should not only do no damage to membrane material but also be effective in removing pollutants. [66]. However, chemistry cleaning is often time-consuming [67–69], leading to process interruption and degraded membrane lifespan [70]. Therefore, the physical and chemical methods are often incorporated to effectively rinse the fouled membrane [62].

To sum up, membrane fouling depends on various factors, such as membrane characteristics (pore size, surface property, etc.), the characteristics of the wastewater solution [71] and operating conditions [24]. Therefore, it is necessary to systematically investigate the abovementioned factors to reach an anti-fouling performance due to the complexity of tannery wastewater.

6.2 Future perspectives

Membrane fouling as the “necking problem” of membrane technology has been a hot topic of research. Therefore, the development of anti-fouling membrane materials and optimization of membrane process configurations are the major trends for future research.

At present, there is relatively few researches on coupling non-membrane technology with membrane technology for the treatment of tannery wastewater. By now, only ozone technology has been coupled with membrane technologies (RO and NF) which showed promising results [72]. In the future, the coupling between non-membrane technologies and membrane technologies may offer pertinent reference for further development of the technology of tannery wastewater treatment. For example, owing to the O_3 improvement on the biodegradability of the tannery wastewater, MBR coupled with O_3 pre-treatment, can be more conducive to the degradation of organic matter in the backend MBR, thus enabling the treatment system achieve high productivity along with the pollutants removal. Furthermore, pre-ozonation could be used to treat tannery wastewater by coupling the FO process with fertilizer as the draw solution, which can not only degrade pollutants but also mitigate FO membrane fouling. Moreover, the diluted fertilizer can be used directly for agricultural irrigation etc.

In fact, the non-membrane-based technologies are effective in the removal of organic pollutants, heavy metal ions and inorganic non-metal ions from tannery wastewater. However, they can not completely remove pollutants from tannery wastewater and it is difficult to achieve ZLD or direct discharge standards by themselves alone. Therefore, any one of the non-membrane-based technologies, either AOPs, biological, coagulation/flocculation or adsorption, coupled with an individual membrane technology (MF, UF, RO, FO, MBR, ED) or an already-coupled membrane technology (NF + RO, UF + NF, UF + RO, etc.) could be expected to deliver better performance of tannery wastewater treatment, as summarized in Fig. 9. The future process, probably coupled, is expected to deliver a high removal efficiency for the majority of pollutants and mitigated membrane fouling as well.

7 Conclusions

This work reviews membrane technologies for tannery wastewater treatment in recent years. The research trends, technical recapitulation and recent advancements in this topic are systematically summarized. Appropriate application of diverse membrane technologies in the tannery wastewater treatment enhances the process efficiency, while membrane fouling is still the major challenge of its further development. Moreover, aiming at the characteristics of tannery wastewater with high salt concentration and organic matter, a single-process treatment is not adequate to meet the emission or “ZLD” standard. Therefore, new membranes or new coupling within diverse membrane technologies or between membrane and non-membrane technologies are considered as the key point of future investigation in this field.

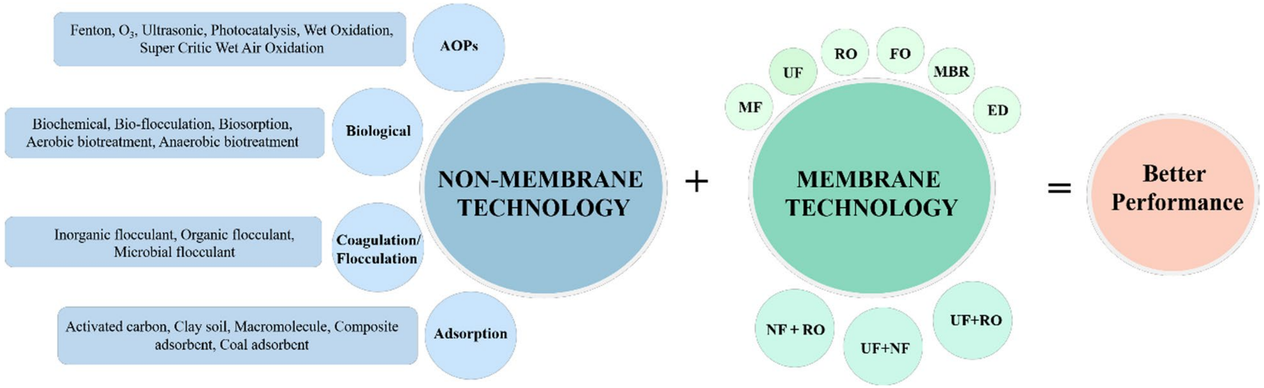


Fig. 9 Future research trends: coupling between non-membrane technologies and membrane technologies, coupling between membrane technologies

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Availability of data and materials
The authors declare that all the data supporting the findings of this study are available within the article.

Declarations

Competing interests
The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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