



A review of key parameters affecting inorganic scaling in thermal, pressure, and osmosis-driven membranes for produced water desalination

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ABSTRACT

As a byproduct of oil and gas (O&G) extraction, large volumes of produced water (PW) with complex compositions have a potential threat if they are not appropriately managed. Although membrane technologies are promising approaches for PW desalination, the widespread application of membrane desalination technologies is significantly hindered by fouling and inorganic scaling. The research interest in membrane scaling has been proven by the growing number of literature with an average annual growth rate of 17 %. Inorganic scaling was impacted by various factors, mainly including operation conditions, membrane properties and pretreatments. Thus, the review summarized the latest research on inorganic scaling during PW treatment using membrane distillation (MD), forward osmosis (FO) and nanofiltration/reverse osmosis (NF/RO), focusing on these influencing factors. Moreover, FO showed the best resistance performance of inorganic scaling in PW treatment by comparing MD, FO and RO/NF processes. Finally, the trade-off between scaling mitigation behaviors and permeate flux, exploring the effect of fouling on scaling, and studying the unknown risks of chemical agents on desalination could be regarded as future research directions. It is beneficial for the large market prospects of membrane processes in PW treatment by addressing membrane scaling.

1. Introduction

Produced water (PW) is a byproduct from the extraction of fossil fuels, such as oil and gas (O&G) [1]. With the increasing demand for fossil energy, PW generation has risen significantly, resulting in a global volume reaching approximately 250 million barrels per day [2]. PW contains a mixture of constituents, including organics (e.g., straight-chain/branched aliphatic hydrocarbons, single-ring/polycyclic aromatic hydrocarbons, phenols, and organic acids), inorganics (with total dissolved solids (TDS) ranging from 1,000–250,000 mg/L), and microorganisms (e.g., microbes and extracellular polymer substances (EPS)) [3,4].

To mitigate environmental issues and alleviate water stress, various physical, chemical, and biological treatments for PW have been developed [5,6]. Membrane technologies have the advantages of compact modules, enhanced permeate quality, reduced labor, and minimal chemical reagent requirements [7]. Moreover, integrated membrane technologies, such as forward osmosis (FO)-membrane distillation (MD) and FO-reverse osmosis (RO) have been used to desalinate saline PW and improve water recovery [8].

However, various types of fouling, including inorganic fouling (scaling), organic fouling and biofouling occurred in membrane desalination processes, affecting desalination performance for PW [9]. Inorganic fouling, or mineral scaling, is a significant issue in PW due to high

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Nomenclature	
PW	Produced water
MD	Membrane distillation
FO	Forward osmosis
NF	Nanofiltration
PP	Polypropylene
RO	Reverse osmosis
PE	Polyethylene
TDS	Total dissolved solids
MER	Membrane electrochemical reactor
TSS	Total suspended substance
CTA	Cellulose triacetate
ECP	External concentration polarization
VMD	Vacuum membrane distillation
SEM	Scanning electron microscopy
CA	Cellulose acetate
EDTA	Ethylene diamine tetraacetic acid
HA	Humic acid
OAE	Oleic acid ethoxylate
SA	Sodium alginate
PVDF	Polyvinylidene fluoride
DCMD	Direct contact membrane distillation
MDC	Membrane distillation crystallization
PTFE	Polytetrafluoroethylene
MF	Microfiltration
UF	Ultrafiltration
ED	Electrodialysis
EC	Electrocoagulation
GAC-EC	Carbon-electrocoagulation
COD	Chemical oxygen demand
PEM	Proton exchange membranes
DS	Draw solutions
ICP	Internal concentration polarization
RSF	Reverse solute flux
AGMD	Air gap membrane distillation
PA	Polyamide
TFC	Thin film composite
CCD	Closed circuit desalination
DPE	Dodecyl phenol ethoxylate
BSA	Bovine serum albumin

TDS levels and scaling-related ions (e.g., Fe^{2+} , Fe^{3+} , Ca^{2+} , Mg^{2+} and Ba^{2+}) [1,10,11]. Moreover, during membrane desalination, the ions exceeding solubility caused by the increased salt concentration on the feed side and concentration polarization can induce scaling on the membrane surface or within the membrane pores [12]. Common substances responsible for inorganic scaling include CaSO_4 , CaCO_3 , SiO_2 and MgSO_4 [13]. Furthermore, the presence of organic matters might form organic-inorganic complexes, aggravating membrane scaling [14]. Scaling can cause negative impacts on membrane performance, such as weakening the membrane rejection properties, increasing hydraulic resistance and energy consumption [15]. Therefore, various methods, such as pretreatments, optimization of operation conditions, and development of anti-scaling membranes, have been developed to mitigate inorganic scaling in membrane desalination processes [16].

Previous reviews focused on membrane performance and desalination applications in PW treatment scaling mechanisms and prevention strategies for specific membrane types. Chang et al. [10] evaluated the pollutant removal performance of a range of membrane processes for shale gas PW treatment. Shahrim et al. [2] reviewed the stand-alone and hybrid systems of three membrane processes (RO, FO and MD) for PW treatment. These reviews provided an overview of the purification performance and mechanisms of various membrane processes for PW treatment, but the challenge of inorganic scaling in PW desalination was not sufficiently addressed. Matin et al. [17] reviewed the mechanisms of salt precipitation and deposition in the RO process and evaluated control strategies. Lin et al. [18] reviewed mechanisms of different types of fouling as well as cleaning and control strategies for scaling mitigation in FO systems. Similarly, Naji et al. [19] reviewed mechanisms and mitigation strategies for wetting, mineral scaling and fouling in MD systems.

All in all, the scaling issue in various membrane desalination technologies for treating PW has not been systematically evaluated in previous reviews. This review aims to provide a comprehensive roadmap for addressing inorganic scaling in membrane desalination technologies for O&G PW treatment. Initially, a bibliometric analysis is conducted to understand the current research landscape of membrane scaling during PW treatment. The mechanisms of inorganic scaling and key influencing factors are summarized. Subsequently, studies on inorganic scaling in PW treatment using MD, FO and NF/RO are critically reviewed, focusing on the impacts of operation conditions (e.g., temperature, pressure and flow rate), membrane characteristics (e.g., roughness, hydrophilicity/hydrophobicity and functional groups), and pretreatments (e.g.,

conventional pretreatments and membrane pretreatments). A comparative analysis of scaling in MD, FO and RO processes is then performed. Finally, the challenges of controlling membrane scaling are summarized, and future research directions for reducing scaling in membrane processes are proposed.

2. Research trends involving inorganic scaling in membrane desalination

2.1. Research trends using bibliometric analysis

The development of membrane technologies in PW treatment over the past two decades was visualized using bibliometric methods. There was a significant increase in the number of articles regarding membrane desalination for PW treatment (Fig. S1, Supporting Information). Based on the data of 393 publications in the Web of Science database from 2010 to 2023 with detailed information listed in Section S2 (Supporting Information), the development trends in research involving membrane scaling for PW treatment were revealed. In membrane desalination processes, scaling related studies increased significantly with an average annual growth rate of 17 % and the cumulative number of publications were fitting by an exponential model ($R^2 = 0.994$) (Fig. 1a). As depicted in Fig. 1a, studies on inorganic scaling in MD, FO and NF/RO developed rapidly, in particular for the domains of MD and FO, due to their efficacy in treating saline PW. Moreover, FO usually exhibited a lower scaling propensity than other desalination membranes, while MD could harness the waste heat from PW to recover the draw solution. Consequently, the integration of FO-MD processes garnered significant research attention for their efficiency and stability in PW treatment.

Research trends and hotspots of scaling in membrane desalination over time are shown in Fig. 1b. Keywords were categorized into four nodes (before 2018, 2018, 2019 and after 2020). Before 2018, most researches focused on pressure-driven membrane desalination processes. In 2018, attention shifted to the desalination performance of membrane technologies, such as “cost”, “performance”, “desalination” and “recovery”. Then, a gradual increase in research interest in FO and MD processes was observed in 2019 (Fig. 1a). Moreover, growing research interests were directed towards exploring the mechanisms of scaling and its influencing factors in desalination processes. Based on the analysis of wordcloud (Fig. S2, Supporting Information), inorganic scaling was impacted by various factors, mainly including operation

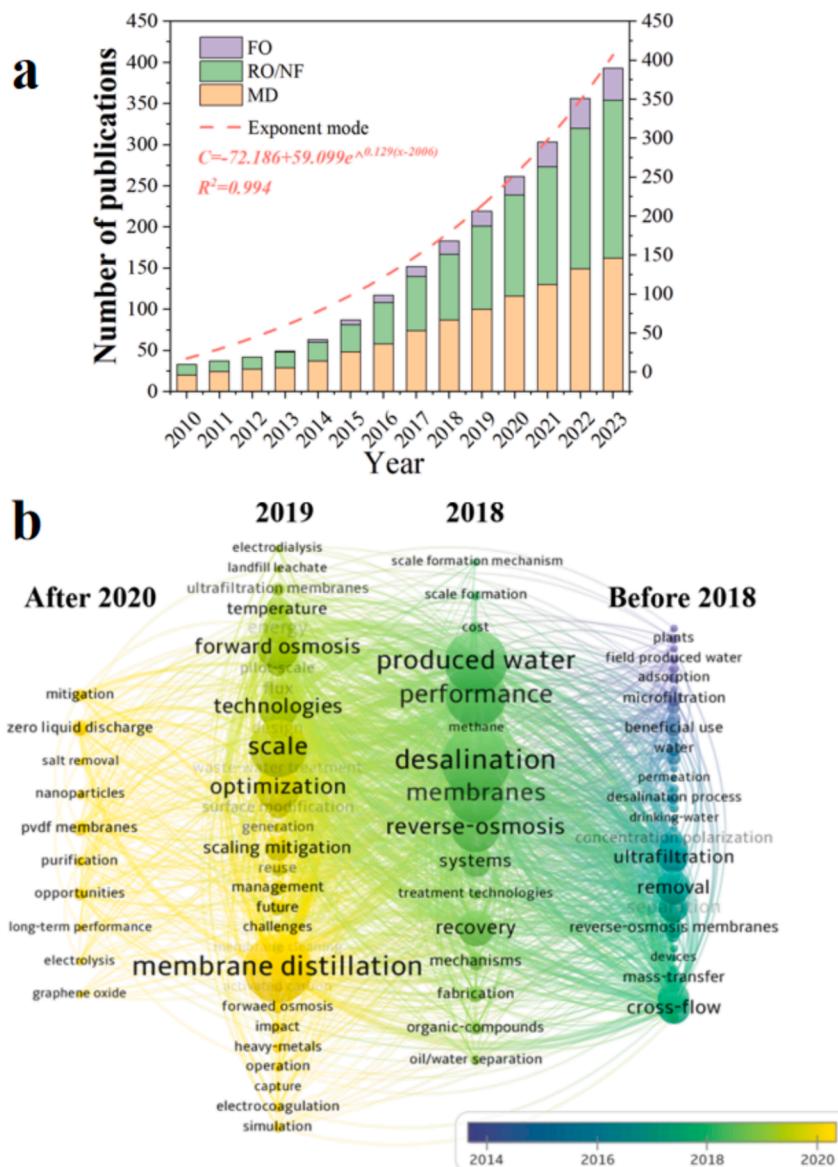


Fig. 1. Research trends on inorganic scaling in membrane desalination based on the bibliometric analysis: (a) The number of publications on scaling research while using membrane technologies for treating PW, (b) time trends of keyword. Data were collected from the Web of Science on May 5, 2024. The keywords was “oil produced water or gas produced water or gas and oil produced water”, “reverse osmosis or nanofiltration or membrane distillation or forwards osmosis” and “inorganic scaling”.

conditions, membrane properties and pretreatments. Since 2020, “zero liquid discharge”, “salt removal” and “long-term performance” appeared, indicating increased attention to the industrial application of membrane desalination processes.

Generally, the bibliometrics methods revealed the rapid development of scaling research using membrane desalination technologies for O&G PW treatment. The key influencing factors for membrane scaling included operation conditions, membrane properties and pretreatment methods.

2.2. Factors influencing inorganic scaling in membrane desalination

The operation conditions, membrane properties and pretreatment methods involving inorganic scaling in specific desalination membrane processes have been extensively studied in previous literature, with key influencing factors summarized in Fig. S3 (Supporting Information). With respect to operation conditions, an optimum flow velocity enhanced shear on the membrane surface [20], thereby mitigating the

propensity for scaling. A reduction in flow velocity resulted in flux decline and promoted heterogeneous scaling due to the extended exposure of the membrane to the feed [21]. Conversely, an excessively high flow velocity improved flux, probably leading to accelerated crystal collisions, which could cause scaling and reduce the size of scale deposits. Moreover, as flow velocity increased, the scaling layer became dense and secondary nucleation was initiated [22], leading to severe scaling. An increase in temperature accelerated molecular motion, thereby increasing the probability of collisions among inorganic salt particles and extending the nucleation time [23]. Besides, temperature could also alter material diffusion properties [24] and the solubility of scaling substances [25], thereby affecting the scaling tendency. In NF/ RO processes which are driven by hydraulic pressure, a large hydraulic pressure resulted in increased flux and concentration polarization [26].

With respect to membrane properties, a rougher membrane surface could provide more contact positions for ions and would reduce shear force on the membrane surface [27]. Additionally, surface roughness would also increase surface free energy, facilitating adhesion and crystal

entrapment. The hydrophilicity of NF/RO and FO membranes induced the formation of hydrogen bonds with water molecules, attracting a layer of tightly bound water molecules and preventing direct interactions between foulants and membrane surface [28]. This process elevated the energy barrier for the nucleation or deposition on the membrane surface, thereby benefiting for mitigating scaling [29]. The hydrophobic properties of MD membranes suspended the solution on the membrane surface, reducing the likelihood of ion and membrane contact [30]. Moreover, the change in the functional groups on the membrane surface could modify the membrane zeta potential and influence ion adsorption [31].

As for pretreatment methods, several types of membrane filtration were capable of eliminating a substantial amount of organic matters and inorganic ions. Thereby, the likelihood of ion saturation and the reduced interactions between organic matters and inorganic ions were decreased, ultimately lowering the propensity for scaling. Traditional pretreatment methods, such as pH adjustment, coagulation and advanced oxidation process, were commonly utilized to adjust the composition of feed solutions [32,33]. pH adjustment altered the forms of weak acid ions and metal ions, as well as the membrane zeta potential, thereby influencing the scaling tendency [34]. Coagulation removed a significant portion of organics and a minor quantity of scaling ions from the solution, reducing the synergistic effects between organic matters and inorganic ions, thereby decreasing the scaling propensity [35].

3. Current researches on inorganic scaling in desalination membrane for PW treatment

3.1. Inorganic scaling in MD for PW treatment

3.1.1. Operation conditions for mitigating scaling in MD during PW treatment

The operation conditions, primarily including temperature and feed/permeate velocity, are key factors influencing inorganic scaling in PW treatment. MD scaling is highly influenced by temperature because MD is driven by the temperature difference between the feed and permeate. Compared to low-temperature operation, elevated feed temperatures in the MD system could cause more severe scaling, forming a deeper [36] and denser scaling layer [37] which was more difficult to remove by common cleaning [38]. Nawaz et al. [39] compared the MD performance at different temperatures (50–70 °C) for PW treatment, and found that MD flux dramatically decreased from 11.19, 16.56 and 23.20 L/(m²·h) (LMH) in the initial to 7.97, 12.26 and 0.20 LMH after 20-h operation at 50, 60 and 70 °C, respectively (Fig. 2a). Similarly, when MD was used for PW treatment, Cho et al. [40] observed the normalized flux decline at 50 °C (by 5.9 %) was less than 30 °C (by 14 %), although the decline in MD flux was faster at 50 °C (by 0.042 LMH) than at 30 °C (by 0.011 LMH) (Fig. 2b). This discrepancy at different temperatures could be attributed to the initial high flux at 50 °C which was 2.5 times higher than that at 30 °C. Moreover, elevated temperatures expedited the formation of scaling crystals deep within the membrane pores, intensifying scaling and wetting. Edwie et al. [36] found that when the feed temperature increased from 60 °C to 70 °C, the scaling depth within the membrane increased from 5 μm to 30 μm. Furthermore, temperature

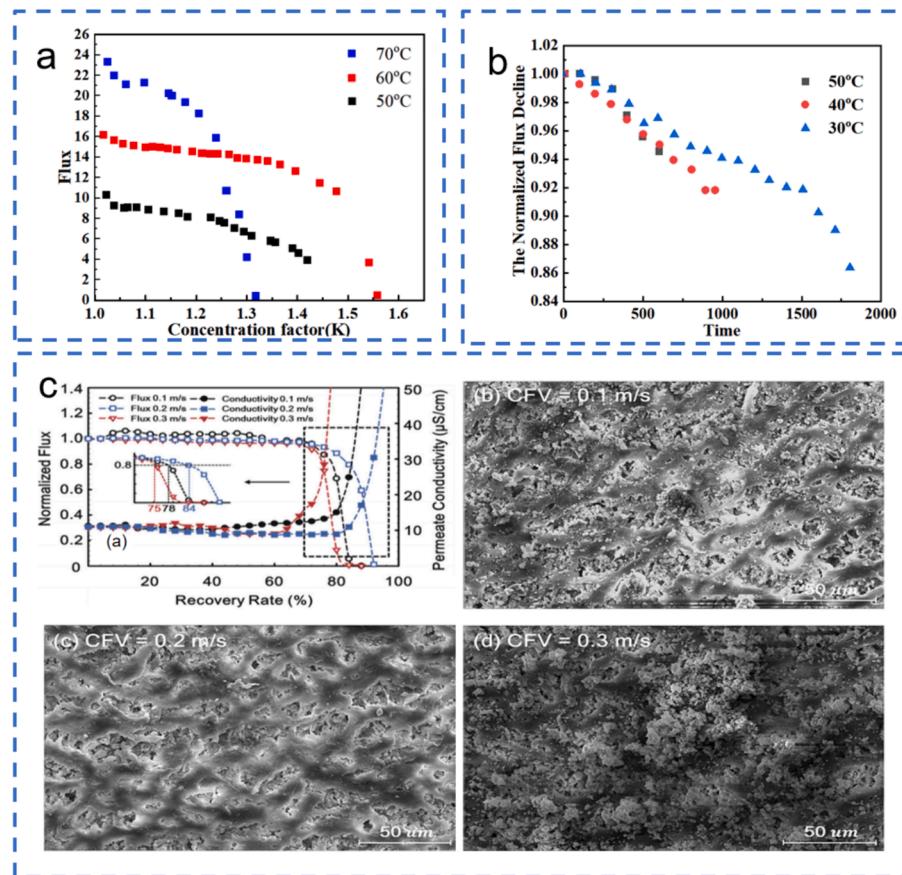


Fig. 2. Variation of MD scaling in treating PW under different operation conditions: (a) Change of flux with feed temperature [39], Copyright (2021) Elsevier B.V. (b) The normalized flux decline at different temperatures [40], Copyright (2018) Elsevier B.V. (c) Comparison of flux and scanning electron microscopy (SEM) images under different flow velocities [38], Copyright (2018) Elsevier B.V. The data are extracted from those figures and replotted.

also influenced the solubility of salts (e.g., directly or inversely proportional to temperature), and thus the extent of inorganic scaling in MD systems. Generally, a rise in temperature would result in an increase in initial flux, severe temperature polarization and intensified scaling [13]. Increasing temperatures would influence the distribution and dimensions of pore apertures, thereby altering the scaling propensity [41].

In addition to temperature, the velocities of feed and permeate sides in the MD process also influenced concentration polarization and temperature polarization, thereby affecting flux and scaling [42]. Moreover, the increase in flow velocity reduced the scaling growth rate [43], while the flux did not continuously increase with the increase in velocity, indicating the existence of the optimum velocity [44]. As reported by Ali et al. [45], an increase in the feed flow velocity from 150 to 250 mL/min, the growth rate of scaling in PW treatment reduced significantly from 0.16 to 0.05 $\mu\text{m}/\text{min}$. However, with an excessive increase in velocity, scaling might be accelerated because secondary nucleation might occur. During the treatment of PW using MD, Kim et al. [38] demonstrated that the least scaling on the membrane surface was found at a feed flow velocity of 0.2 m/s compared to those at 0.1 and 0.3 m/s (Fig. 2c). At the optimum flow velocity (i.e., 0.3 m/s), scaling substances including calcium carbonate and sodium chloride were observed, and the NaCl scaling was predominantly associated with calcium carbonate [38].

Currently, the integration of MD with crystallizers, and the application of ultrasonic crystallization or magnetic fields) [32] have been reported to mitigate scaling on the membrane surface by decreasing the concentration of scaling ions. To be specific, magnetic fields could prevent the entrance of small crystals into membrane pores and promote the precipitation of inorganic compounds with low solubility, effectively alleviating the tendency for scaling on the membrane surface [46]. Compared to the conditions with the magnetic field, Kargari et al. [7] demonstrated that magnetic fields substantially changed membrane scaling behaviors, reducing the contents of Na^+ , Ca^{2+} and Mg^{2+} on the membrane surface by 64.47 %, 41.15 % and 62.43 %, respectively. Moreover, in MDC (i.e., MD integrated with crystallization), in-situ solution crystallization in a crystallizer diminished the concentration of scaling ions in the solution and mitigated the scaling propensity on the membrane surface. In the study reported by Kim et al. [38], MDC was used to treat shale gas PW, and the water recovery (62.5 %) was nearly double that of the traditional MD process (32 %). The MDC system not only mitigated membrane fouling but also promoted the recovery of fresh water and minerals from the PW.

Therefore, during the treatment of PW using MD, appropriate temperatures and velocities could increase membrane flux and mitigate inorganic scaling, while severe scaling might occur under excessive temperatures and flow velocities. The MD flux, water recovery and scaling behaviors under different temperatures and flow velocities are listed in Table S1 (Supporting Information). In addition, innovative devices that improve operation conditions could effectively reduce MD scaling during the treatment of PW.

3.1.2. Membrane properties of MD membranes for mitigating scaling during PW treatment

MD membranes were typically hydrophobic and made of common polymeric materials such as polyvinylidene fluoride (PVDF), polypropylene (PP) and polytetrafluoroethylene (PTFE) [47]. Thus, the MD membranes had distinct properties, such as pore sizes, wettability and roughness, leading to different scaling behaviors. Superhydrophobic membranes, omniphobic membranes and Janus membranes have been developed to enhance scaling resistance through membrane modification [47,48]. Additionally, novel and cost-effective membrane materials with excellent properties have also been proposed for PW treatment. Studies on the effects of membrane characteristics on scaling during PW treatment using MD are summarized in Table S1 (Supporting Information).

Membrane scaling behaviors are greatly impacted by the surface morphologies (e.g., pore sizes, roughness and wettability) of MD

membranes. Compared to the common hydrophobic membrane (with a contact angle of about 120°), a slippery membrane could provide a higher surface hydrophobicity ($>150^\circ$) and lower surface energy, thereby forming a higher energetic barrier for scaling [49], as shown in Fig. 3d. MD membranes with larger pore sizes resulted in higher fluxes but more severe polarization and scaling occurred [50]. As reported by Ali et al. [45], the scaling growth rate of the PP membrane ($0.15 \mu\text{m}/\text{min}$) was higher than that of PVDF membrane ($0.03 \mu\text{m}/\text{min}$) during the treatment of PW. The higher growth rate of the PP membrane was attributed to the sponge-like structure of the outer layer, which served as an anchor point for crystal growth and adhesion, in contrast to the smooth surface of the PVDF membrane. Furthermore, Gryta et al. [51] and Cho et al. [40] found that PP membranes with larger pores facilitated more crystal deposition within the pores than membranes with smaller pores. Wettability states of membranes, including Wenzel and Cassie-Baxter wetting states, were the key parameters in membrane scaling [49]. In the Wenzel state, the membranes have relatively low contact angles, allowing water to closely interact with the membrane surface. This interaction led to a negative slip and indicated a static dynamic state, providing sufficient time for crystal deposition [30]. Conversely, membranes in the Cassie-Baxter state exhibited high contact angles, causing water to beat up on the surface, which reduced the chances for crystal aggregation and resulted in a positive slip [30]. This dynamic state of surface fluid velocity reduced interaction time, significantly inhibiting heterogeneous nucleation and preventing homogeneous nucleus deposition [52]. Xiao et al. [52] found that the superhydrophobic CF_4 -MP-PVDF membrane in the Cassie-Baxter state had a slip length of $62 \mu\text{m}$, and a very stable flux and the clean surface were maintained. In contrast, the C-PVDF and MP-PVDF membranes, which were in the Wenzel state with a slip length of $200 \mu\text{m}$, experienced a rapid decrease in flux (by 50 %), as shown in Fig. 3a.

The superhydrophobic, omniphobic and Janus membranes exhibited excellent anti-scaling properties, limiting scaling to shallow depths near the membrane surface and leading to outstanding cleaning efficiency. Superhydrophobic membranes typically had high contact angles ($>150^\circ$) and low sliding angles ($<10^\circ$), hence, they often exhibited Cassie-Baxter wetting characteristics and superior scaling resistance for gypsum and silica [55]. Compared to the PVDF membrane, the flux of the superhydrophobic membrane (PVDF-SINP-FAS) decreased slowly [55], and the superhydrophobic membrane had better scale inhibition for calcium sulfate compared to silica (Fig. S4a). Omniphobic membranes exhibited better performance during the treatment of PW containing salts and oils [56]. As reported by Li et al. [56], the normalized fluxes were 0.50 after 300 min and 0.85 after 600 min for PVDF membrane and omniphobic FZnO-PVDF membrane, respectively, and the omniphobic membrane showed only minor CaSO_4 crystal and less scaling compared to PVDF membrane. The low sliding angle (10.6°) and high contact angle (164°) of omniphobic membranes provided the non-zero fluid velocity at the membrane surface, as confirmed by Xiao et al. [57]. Omniphobic braid-reinforced membranes were covered by fewer scaling substances, and the deposited scaling was easier to clean, as shown in Fig. S4b (Supporting Information) [58]. Generally, Janus MD membranes have a dense hydrophilic surface layer, and the hydrophilic layer could diminish the impact of pollutants such as oil in PW. Du et al. [53] discovered that at a water recovery rate of 53 %, the normalized fluxes of Janus membranes and superhydrophobic membranes were 0.77 and 0.67, respectively, during filtration of real PW, as shown in Fig. 3b. The development of new membrane materials would enhance anti-scaling performance. Du et al. [54] prepared a poly (2,6-dimethyl-1,4-phenylene oxide) (PPO) membrane with a considerably smooth surface, and the resultant membranes exhibited significantly enhanced anti-scaling performance [57]. Unlike the hydrophobic PVDF membranes which were covered with CaSO_4 , there was minimal scaling on PPO membranes (Fig. 3c). Additionally, compared to the membranes modified using fluorinated polymers, PPO membranes offered distinct advantages due to their easy availability and straightforward

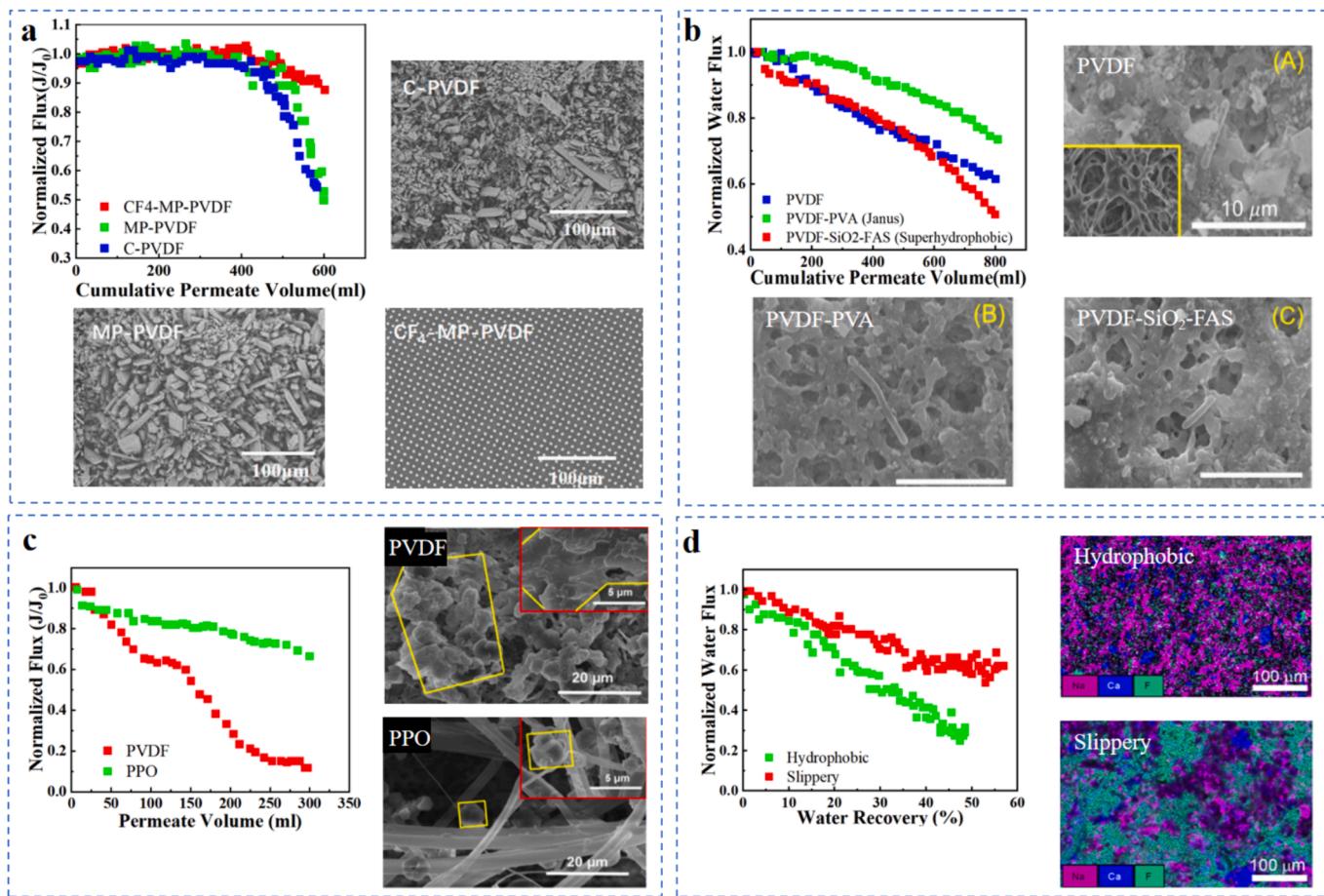


Fig. 3. Variation of MD scaling and flux with different membranes properties: (a) Wettability states including, including Wenzel (MP-PVDF and C-PVDF) and Cassie-Baxter wetting (CF₄-MP-PVDF) [52], Copyright (2019) Elsevier B.V. (b) Comparison of hydrophobic, superhydrophobic and Janus membrane [53], Copyright (2018) Elsevier B.V. (c) Comparison of membrane materials (PPO and PVDF) [54], Copyright (2019) Elsevier B.V. (d) Comparison of slippery and hydrophobic membrane [49], Copyright (2018) Elsevier B.V. The data are extracted from those figures and replotted.

processing.

All in all, higher roughness and larger pore sizes of MD membranes led to more severe scaling during the treatment of PW. The Cassie-Baxter state was effective in reducing membrane scaling. Compared to hydrophobic membranes, superhydrophobic, omniphobic and Janus membranes were likely to offer excellent anti-scaling performance, making scaling not only less frequently occur but also easier to clean.

3.1.3. Pretreatment approaches for mitigating scaling in MD during PW treatment

Appropriate pretreatments are used to reduce the concentration of scaling ions in PW, thereby alleviating membrane scaling tendencies. Conventional pretreatments (coagulation, pH adjustment, electro-coagulation, advanced oxidation process etc.) [33,59] and membrane filtration pretreatments (microfiltration (MF), ultrafiltration (UF), NF, electrodialysis (ED), membrane electrochemical reactor (MER), FO, RO, MD, etc.) [33,60] have been employed to mitigate scaling in MD during PW treatment.

Coagulation was effective in removing organics from PW rather than many inorganic ions (e.g., Ca²⁺, Mg²⁺ and Na⁺), while better inorganic ion removal was reported by softening (e.g., increasing pH) [35]. Moreover, approximately 90 % of iron ions were removed by poly-aluminium chloride [61], and aluminum salts (i.e., Al₂(SO₄)₃·14H₂O and Na₂Al₂O₄) could also efficiently remove silica [62]. Compared to coagulation, electrocoagulation (EC) is more attractive due to its lower dosage of chemical reagents, lower sludge production and simple operation, and combining softening and electrocoagulation can

effectively remove scaling ions. Lobo et al. [63] used granular activated carbon- EC to treat PW and found that EC removed 99 % of total suspended solids (TSS), but EC had limited effect on chemical oxygen demand (COD) (15 % removal) and TDS, as also reported by Sardarp et al. [64]. Moreover, Gao et al. [65] demonstrated that the normalized flux after 175 min for PW without EC pretreatment declined rapidly to 0.85, while the normalized flux of 0.90 with EC pretreatment was achieved. The EC pretreatment was also very effective in increasing the flux of the FO-MD system by roughly 30 % by removing the main scaling ions (calcium and sulfur) [66]. However, as reported by Esmaeilirad et al. [67], the removals of divalent ions in a combined softening-EC process (Ca²⁺=90 %, Mg²⁺=70 %, Ba²⁺=90 %, Si²⁺=70 %) were higher than those in an EC-softening process (Ca²⁺=45 %, Mg²⁺=40 %, Ba²⁺=65 %, Si²⁺=40 %). The removals of divalent ions in EC-softening primarily relied on the softening process, and the solid crystals produced by pre-softening improved the softening effect during EC [67,68]. Furthermore, adjusting the pH of PW can effectively mitigate scaling characteristics. Zhang et al. [69] observed that NaOH softening (at pH = 10) decreased the ion concentrations in PW, such as 48 % of Ba²⁺, 46 % of Ca²⁺, 19 % of Mg²⁺, 32 % of SiO₂ and 20 % of Sr²⁺, thus reducing the potential for scaling. Yan et al. [70] demonstrated that the thickness of the membrane scaling layer at pH = 5 was approximately half that at pH = 9. A combination of different pretreatment methods is used to enhance MD scaling mitigation. As reported by Zhang et al. [69], when the precipitation softening pretreatment method was used, the water flux was reduced by 18 % at a water recovery of 82.5 %, while the flux decline was only 10 % at the same water recovery when a combined

pretreatment method of precipitation softening and walnut shell filtration was used, as shown in Fig. 4g.

Combining different membrane processes can create a high-performance MD hybrid process, reducing the impact of inorganic scaling and increasing water recovery rates. Typical hybrid processes include ED-NF-MD [72], MF/UF-MD [40], FO-MD [75], RO-MD [76] and MER-MD [74], as shown in Fig. 4a-4f. It was found that ED could effectively separate cations and anions into different chambers, producing scaling-free flow for MD unit [77]. Zhao et al. [72] used ED to treat PW by separating cations (Na^+ , Ca^{2+} and Mg^{2+}) from anions (Cl^- and SO_4^{2-}), and only a small amount of scaling was observed at a water recovery rate of 99.8 %. Pressure-driven membrane filtration can remove part of scale ions and mitigate scale formation, including NF [64], UF [61] and MF [40]. RO-MD hybrid process could improve the water recovery rate and showed low scaling tendency due to the addition of anti-scalants to RO brine. Duong et al. [73] found that the RO-MD hybrid process maintained a stable distillate production rate of 10 L/h without scaling, but silica and calcium scaling occurred in MD during long-term operation. In the FO-MD process, MD could concentrate draw

solutions to maintain the stability of FO process, and a low scaling tendency could be expected. Zhang et al. [75] demonstrated the feasibility of the FO-MD process in treating simulated PW, a flux of 5.7 LMH was maintained at a water recovery of 90 %. Three distinct designs for the FO-MD system have been reported. The first one (Fig. S5a) where FO and MD shared the draw solution tank was the most commonly used [78], but such integration might not be very economical on a commercial scale [79]. The second design (Fig. S5b) was proposed as a compact system for the hybrid FO-MD [80]. Compared the first one, this compact design was more economical, less footprint and more easy to achieve the equal water transfer rates between the FO and the MD process. The third system (Fig. S5c) added an isolation barrier to the compact FO-MD system [81]. The system with an isolation barrier not only had the advantages of compact design, but also decreased the effect of temperature of MD feed on FO membrane [82]. Thus, the compact FO-MD system with an isolation barrier is the best choice for PW treatment. For a stable and successful FO-MD operation, the water transfer rates between the FO and MD should be equal [83]. However, due to the asymmetric structure of FO membrane, severe external and internal concentration

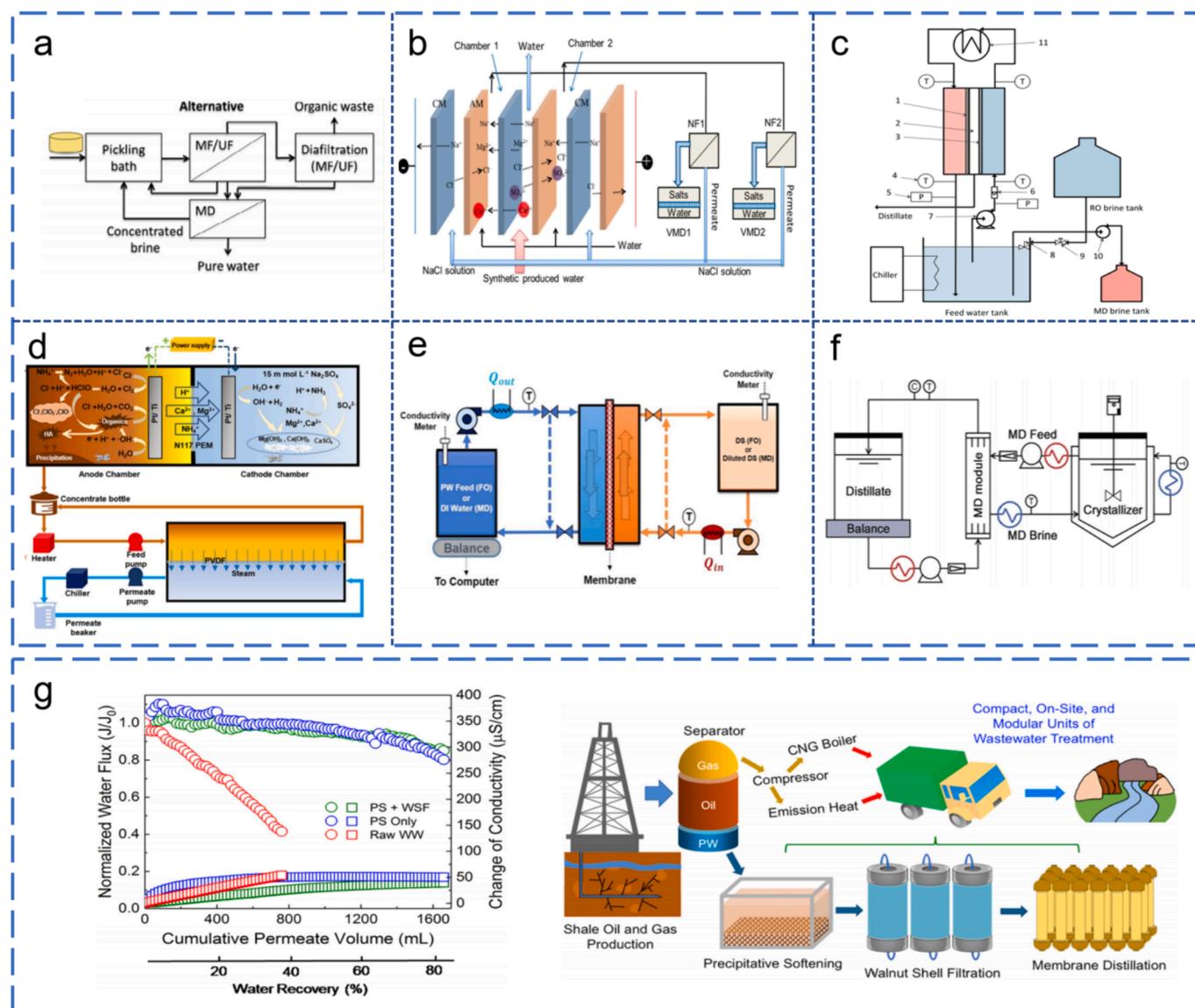


Fig. 4. Schematic diagram of typical MD hybrid processes using different pretreatments: (a) MF/UF-MD process [71], Copyright (2018) Elsevier B.V. (b) ED-NF-MD process [72], Copyright (2020) Elsevier B.V. (c) RO-MD process [73], Copyright (2015) Elsevier B.V. (d) MER-MD process [74], Copyright (2023) Elsevier B.V. (e) FO-MD process [64], Copyright (2019) Elsevier B.V. (f) MDC process [38], Copyright (2018) Elsevier B.V. (g) Process diagram for the combined treatment process [69], Copyright (2019) Elsevier B.V.

polarization had a negative effect on water transfer rates. Thus, the symmetric membranes are more suitable for FO-MD than the asymmetric membranes. For instance, in the study conducted by Li et al. [84], the lab-fabricated symmetric FO membrane performance was superior to commercial TFC or CTA FO membranes to treat textile wastewater. Furthermore, the FO membrane should be capable of functioning smoothly at high temperatures. The selectivity of the FO membrane would be compromised when operating at higher temperatures due to the increased reverse salt flux. Among commercially available options, Porifera membranes stand out as they are particularly well-suited for operation under MD temperature conditions [85]. For MD membranes in the FO-MD system, the characteristics of hydrophobicity, low conductivity, thermal stability, high porosity with a narrow and uniform pore size distribution, low tortuosity, and chemical resistance, are typically constructed from materials such as PTFE, PVDF, and PP. Although decreasing the temperature of DS (i.e., feed temperature of MD) could reduce the damage of FO membrane, the water recovery rate of the MD would be significantly lower than that of the FO membrane, so it is necessary to utilize MD membranes with larger pore sizes and larger membrane areas. Membrane scaling was significantly inhibited by using membrane electrochemical reactor (MER) as an MD pretreatment, through the combined effects of hardness removal (61 %) and feed acidification ($\text{pH} = 2$) [74]. To reduce membrane cost and electricity consumption, the proton exchange membranes (PEM) in MER pretreatment could be replaced with pressure-driven membranes [86].

Moreover, MD scaling mitigation has been reported by the removal of organic matters and inorganic ions using various pretreatments, including pH adjustment, coagulation, granular filtration and membrane filtration (e.g., UF, NF, RO and FO). Moreover, the efficiency and stability of water recovery can be greatly improved by the hybrid RO-MD and FO-MD process. The application of different pretreatment processes for treating PW is also listed in Table S1 (Supporting Information).

In summary, the MD process operated at higher temperatures had higher initial flux but also experienced more severe scaling, while

appropriate flow velocity and innovative devices (e.g., crystallizers, ultrasound and magnetic fields) could effectively reduce scaling. MD membranes with better hydrophobicity, lower roughness and smaller pore sizes led to less scaling, and superhydrophobic, omniphobic and Janus membranes with anti-scaling properties have been developed. Appropriate pretreatments (e.g., traditional pretreatment approaches or membrane filtration) could effectively reduce scaling ions in MD for PW treatment.

3.2. Inorganic scaling in FO for PW treatment

3.2.1. Draw solutions and pretreatments in FO for mitigating scaling

FO process, driven by the osmotic pressure differential across the membrane, is significantly influenced by both the draw solution (DS) and the feed solution, which can profoundly impact concentration polarization (CP) (i.e., internal concentration polarization (ICP) and external concentration polarization (ECP)) and reverse solute flux (RSF), thus altering membrane scaling performance. Severe CP could lead to ions over-saturating and crystallizing within the membrane support layer and on the membrane surface, thereby impacting the FO flux significantly [87].

The selection of DS is a critical factor influencing scaling in the FO process. The primary DS included monovalent salt solution, divalent salt solution, salt solution containing anti-scalants and acid solution. When DS contained divalent salts, such as Ba^{2+} , Ca^{2+} , Mg^{2+} , SO_4^{2-} and CO_3^{2-} , accelerated membrane scaling could be observed, due to the diffusion of these ions to the feed side [88]. Lee et al. [89] demonstrated that using $(\text{NH}_4)_2\text{CO}_3$ as the DS led to increases in the concentrations of HCO_3^- and NH_4^+ in the feed solution (i.e., PW) by 0.06 mol/L and 0.44 mol/L, respectively due to the diffusion effect, thus resulting in a significant flux decline (Fig. 5a). Concurrently, scaling ions from the feed solutions could also permeate to the DS through forward solute flux, leading to the occurrence of scaling on both sides of the FO membrane [90]. However, the FSF had a minimal effect on scaling due to the restriction imposed by RSF [91]. Fertilizer driven FO (i.e., FDFO) which employs fertilizers as

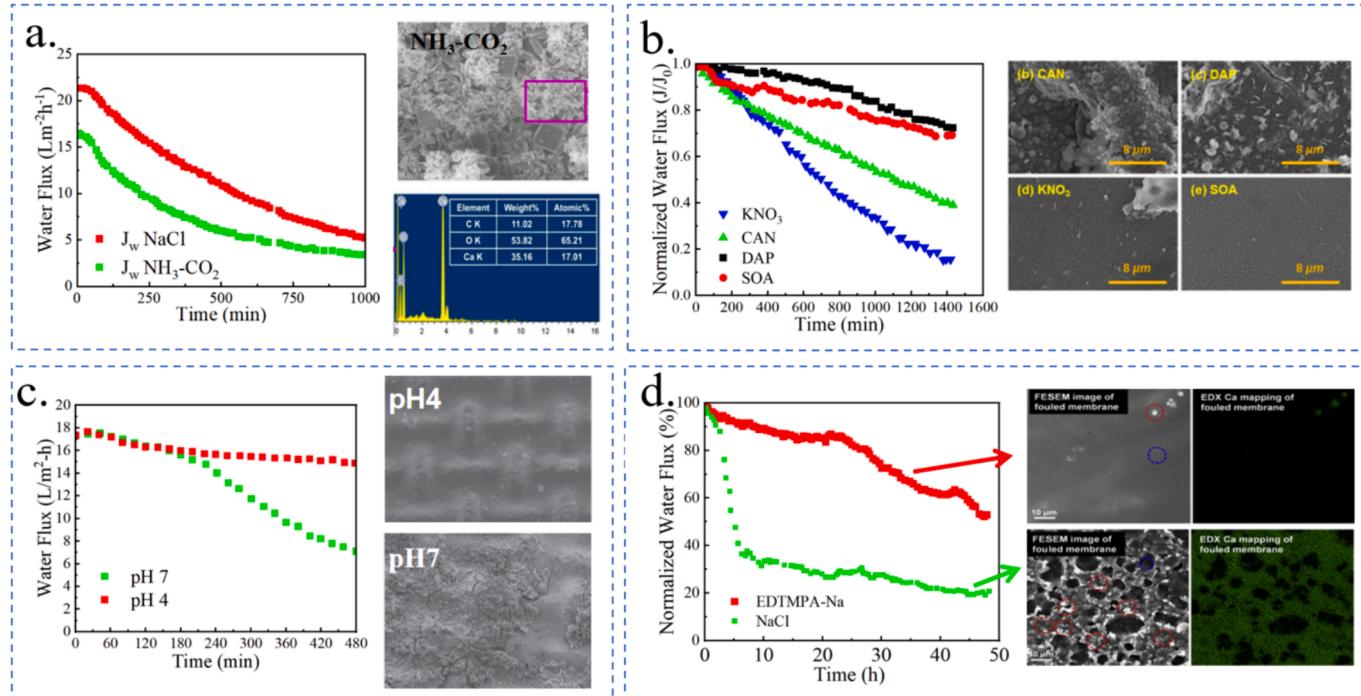


Fig. 5. Comparison of flux variation and membrane integrity under various draw and feed solutions in FO: (a) divalent salts ($\text{NH}_3\text{-CO}_2$) [89], Copyright (2017) Elsevier B.V., (b) fertilizers (DAP, SOA, KNO_3 and CAN) [90], Copyright (2017) Elsevier B.V., (c) acid solution [98], Copyright (2017) Elsevier B.V. and (d) adding anti-scalants (EDTMMPA-Na) [95], Copyright (2020) Elsevier B.V..

an effective DS in FO, has the benefits of without additional separation steps (thus being cost-effective), while this process may result in severe scaling problems due to the presence of scaling ions. Kim et al. [90] used chemical fertilizers as DS for treating coal seam gas PW and observed a significant reduction in flux with using KNO_3 as DS, due to a large RSF. Conversely, using calcium nitrate and di-ammonium phosphate (DAP) as DS also resulted in severe flux reduction due to the presence of scaling ions in calcium nitrate and DAP (Fig. 5b). However, divalent salts (e.g., MgCl_2 , CaCl_2 and MgSO_4) which had larger ion sizes and lower diffusivity than monovalent salts which could reduce the RSF [92]. Additionally, the diffusion of monovalent ions would be inhibited by multivalent ions due to complex ion interactions or interactions between ions and the membrane surface, such as steric hindrance, ionic shielding, and electrostatic interactions [93]. NaCl solution was frequently utilized as DS due to the advantages of high solubility, convenient recycling, and negligible scaling risk [94]. The incorporation of anti-scalants into the DS can effectively reduce membrane scaling and RSF and the higher concentrations of anti-scalants, the better anti-scaling performance. Ding et al. [95] explored the role of anti-scalants in the DS in scaling performance and found that using ethylenediamine tetra sodium salt (EDTMPA-Na) as DS resulted in a slower flux decline and less scaling compared to NaCl solution (Fig. 5d). Furthermore, Gwak et al. [96] found that the RSF for NaCl was approximately double that of an antiscalant-blended DS containing a mixture of NaCl and poly (aspartic acid sodium salt). In addition, antiscalants from DS with the peculiarity of lower diffusion hindered the diffusion of monovalent ions, weakening the RSF for monovalent ions in DS [97]. While anti-scalants mitigate scaling, their recovery from the solutions presents a critical challenge. The pH of the DS also influences membrane scaling characteristics. The H^+ by RSF into the DS could change localized pH on the membrane surface without significantly altering the pH of PW, thereby changing the equilibrium of weak acid salts and mitigating membrane scaling tendencies. Zhang et al. [98] found that minimal scaling was observed at pH = 4 compared to that at pH = 7 and 8.5, as confirmed by the membrane images (Fig. 5c). DS using MgCl_2 manifested substantial scaling because Mg(OH)_2 was the primary scaling substance when pH exceeded 9, and MgCl_2 solutions showed low scaling propensity by maintaining optimal pH levels [99].

The FO performance is also affected by feed solutions (i.e., PW characteristics), which are associated with the osmotic pressure gradient and scaling trend. Pretreatments using UF and pH adjustment reduce the concentration of scaling precursors in the feed solution, thus decreasing the scaling tendency. Chen et al. [100] found that UF pretreatment delayed flux decline to 75 % for 15–20 min, whereas the flux immediately decreased to 75 % without pretreatment. However, UF pretreatment could not completely prevent scaling formation. EC prior to FO could significantly reduce membrane organic fouling and inorganic scaling by removing significantly TOC and TSS by 70 %, leading to an increase of water recovery by 21 % [101]. Moreover, Jeong et al. [34] found that the higher pH values (2, 4, 6 and 8) of feed solutions could lead to more severe scaling.

Therefore, using scale inhibitors and acidic solutions as DS for treating PW could greatly mitigate inorganic scaling, but DS using divalent ions (Mg, Ca, etc.) might aggravate the formation of scaling due to the transport of the scaling ions from DS to FS. For feed solution, pretreatments such as UF and pH adjustment could remove the scaling ions, thereby alleviating FO scaling. The effects of drawing solutions and pretreatments on scaling while using FO for treating PW are summarized in Table S2 (Supporting Information).

3.2.2. Membrane properties of FO membranes for mitigating scaling

The scaling propensity in FO processes is greatly affected by the orientation, structure, and properties (e.g., roughness, functional groups and solute rejection) of FO membranes. The orientation of the FO membrane, including the active layer-facing feed solution (AL-FS) and the active layer-facing draw solution (AL-DS), would affect the scaling

distribution within membrane pores or on the membrane surface. In AL-DS mode, a considerable amount of scaling ions primarily gathered in the membrane pores [102], while scaling ions primarily accumulated on the membrane surface in AL-FS mode [103], leading to scaling within the pores and on the membrane surface, respectively. Compared to scaling on the membrane surface, scaling within the membrane pores was more difficult to remove [15]. Moreover, the pore structure could be severely changed due to the occurrence of scaling within membrane pores, affecting membrane performance [104]. Furthermore, the ICP of AL-FS with a compensation mechanism can maintain the stable flux during the treatment of hyperhaline oilfield PW with a high scaling tendency. Zhang et al. [104] demonstrated a flux reduction was 50 % and 10 % for the AL-DS and AL-FS, respectively, indicating the intense impact of scaling within pore structure in AL-DS. ICP, spanning the entire interface, was particularly challenging to mitigate by adjusting operation conditions [31]. The properties of the support layer, including porosity, thickness, and tortuosity were key determinants of ICP [105]. The structural parameter S, indicating the solute diffusion within the support layer, was given by $S = \tau l/\epsilon$, where τ represented tortuosity, l stood for thickness, and ϵ for porosity, with a smaller S value correlating with lower ICP [106,107]. The functional groups on the membrane surface can change its scaling characteristics by altering the membrane potential. Mi et al. [108] found that the flux reduced to 10 % and 50 % for polyamide (PA) and cellulose acetate (CA) membranes, respectively, verifying more severe scaling in PA membranes (Fig. 6a). This was attributed to the negatively charged carboxyl groups on the PA membrane surface, which adsorbed positively charged calcium ions, leading to the aggregation of calcium ions and saturation crystallization [109]. What's more, Euvrard et al. [110] found that hydroxyl and carboxyl functional groups could form hydrogen bonds with silica particles, producing a high-strength Si-O-Si anhydride bond by dehydration and polymerization, making silica scaling difficult to clean [110]. Additionally, the more negative potential produced by a greater number of functional groups on the membrane surface could attract more Ca^{2+} ions, enhancing the scaling potential [111]. Ding et al. [95] found that the polysulfone (PSf) thin film composite polyamide (TFC) membrane with more functional groups, generated a higher surface potential compared to the Aquaporin TFC, thus leading to significant scaling and a severe flux decline (Fig. 6b). Moreover, the cleaning efficiency for scaling is significantly hindered by the rough membrane surface. Mi et al. [112] found that a flux recovery was nearly 100 % and 60 % for smoother CA membranes and rougher PA membranes, respectively. Solute rejection in the FO process played a crucial role due to the influence of RSF. Compared to TFC membranes, CTA membranes, with poorer solute rejection and a greater propensity for RSF decreasing the concentration of DS, suffered a more severe flux decline (20 %) [98]. Moreover, RSF also can elevate the risk of scaling as scaling ions migrate from the DS to the feed side. Membrane modification can mitigate the tendency of scaling by reducing roughness. Chen et al. [100] modified a TFC membrane with polyethylene glycol to reduce roughness for treating PW, and demonstrated that the modified membrane exhibited less scaling than the common TFC membrane, as shown in Fig. 6c.

In general, during the treatment of PW using FO membrane, the appropriate orientation of FO membrane (AL-FS), with the advantages of scaling on the membrane surface and compensatory mechanism, could keep flux stability. However, functional groups and the potential on the FO membrane surface could aggravate scaling by attracting scaling ions in PW. Although the roughness of the FO membrane surface led to serious scaling and difficulty in cleaning, the surface modification could reduce the roughness, thus mitigating scaling. As listed in Table S2 (Supporting Information), different membrane characteristics result in different scaling and performance during the treatment of PW using FO membranes.

3.2.3. Cleaning methods in FO for mitigating scaling

Although FO membranes tended to experience less scaling than other

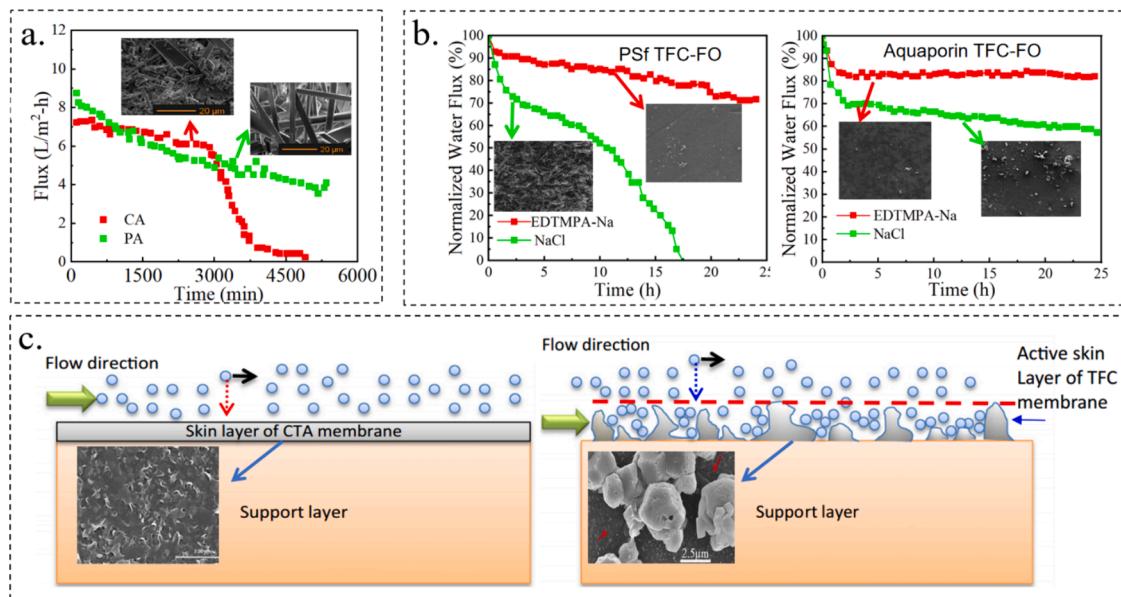


Fig. 6. The effects of FO membrane properties on flux performance and scaling behaviors during PW treatment: (a) Types of functional groups (PA-carboxyl group and CA-hydroxy group) [108], Copyright (2010) American Chemical Society, (b) the number of functional groups (PSf-large functional group and Aquaporin-little functional group) [95], Copyright (2020) Elsevier B.V., (c) modification membrane [100], Copyright (2015) Elsevier B.V..

membrane processes (e.g., NF/RO) [113], appropriate membrane cleaning approaches were crucial for the continuous desalination of PW by reducing scaling. Most of the scaling in FO systems could be removed by simple physical cleaning (osmotic backwashing, hydraulic flushing, etc.) [108], while chemical cleaning (EDTA, citric acid, NaOH, etc.) and combined cleaning approaches were utilized for alleviating irreversible scaling. Alamoudi et al. [114] demonstrated that 70 %–90 % of flux recovery was achieved by osmotic backwashing, and 30 min osmotic backwashing of showed the highest flux recovery and less specific RSF among the investigated cleaning. Moreover, osmotic backwashing has been found to be more efficient than hydraulic flushing. Kim et al. [115] discovered that osmotic backwashing and hydraulic flushing recovered the flux to 100 % and 97.6 %, respectively in 24 h operation, and osmotic backwashing was superior to hydraulic flushing in 72 h operation because the water flux was fully recovered with a cleaner membrane surface. The water flux recovery of only 22 % was achieved due to the CaCO_3 scaling rigidly attaching to the membrane surface, but EDTA cleaning could recover the initial membrane flux, as reported by Lee et al. [89]. Moreover, the combination of air scouring, hydraulic flushing and osmotic backwashing demonstrated an excellent cleaning efficiency (Fig. 7a) [115]. Acid solutions (e.g., HCl and citric acid) which could dissolve inorganic scaling, were more effective than physical cleaning [115]. In comparison, alkaline cleaning could remove organic matter which might associated with inorganic salt crystals on the

membrane surface, and ethylene diamine tetraacetic acid (EDTA) as a cleaning agent could chelate Ca^{2+} to clean the scale. Kim et al. [90] found that citric acid cleaning was more efficient than EDTA and NaOH solutions (Fig. 7b), because citric acid not only chelated with Ca^{2+} like EDTA, but also acted as a weak acid to dissolve inorganic scaling, achieving better cleaning efficiency with increasing concentrations.

In general, during PW treatment using the FO process, anti-scalants and acidic solutions as DS significantly reduced the formation of scaling, while divalent salts (e.g., Mg^{2+} and Ca^{2+}) as DS might aggravate scaling and reduce RSF. For feed solutions, pretreatments could reduce and delay scaling formation. Chemical cleaning (e.g., EDTA, citric acid and NaOH) and combined cleaning methods were superior to physical cleaning methods (e.g., osmotic backwashing and hydraulic flushing) in removing scaling, ensuring the stable operation of the FO process for treating PW. Although the AL-FS mode of FO processes could maintain stable flux, FO membranes with specific functional groups altered the membrane surface potential, attracting scaling ions and accelerating scaling. Membrane surface roughness increased the difficulty of cleaning, but it could be reduced by modifying the membrane surface.

3.3. Inorganic scaling in NF/RO for PW treatment

3.3.1. Operation conditions in NF/RO systems for mitigating scaling

Driven by high hydraulic pressure, the performance of NF/RO is

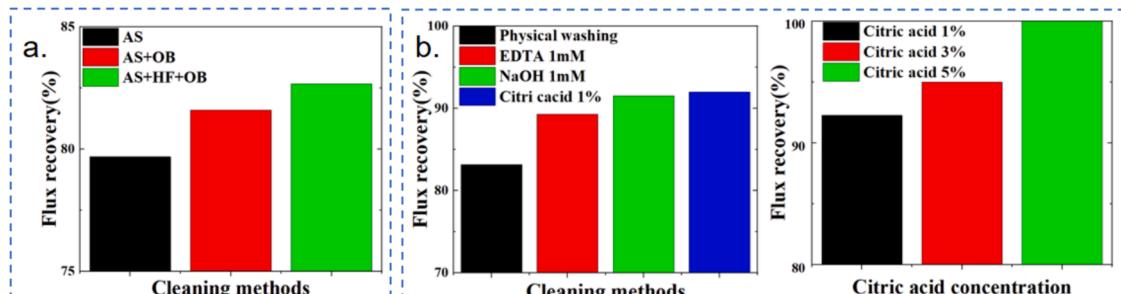


Fig. 7. Comparison of inorganic scaling under various cleaning methods in FO process: (a) Cleaning efficiency of combined cleaning methods [115], Copyright (2020) Elsevier B.V. (b) Cleaning efficiency of chemical agents (EDTA, NaOH, and citric acid) and efficiency of concentrations of citric acid (1%, 3%, and 5%) [90], Copyright (2017) Elsevier B.V. The data are extracted from those figures and replotted.

significantly impacted by operation conditions, particularly hydraulic pressure. Increasing pressure could improve flux and concentration polarization, leading to changes in scaling form and aggravating the degree of scaling. However, there was a limiting flux that did not increase with rising pressure which was related to the quality of the PW [116]. Research by Schäfer et al. [26] revealed a 40 % greater flux reduction in an NF membrane under high hydraulic pressure of 10 bar compared to 5 bar due to scaling formation. Moreover, Lee et al. [117] found that surface crystallization was predominant at higher pressure, while bulk crystallization was more typical at lower pressure, with the higher pressure contributing to an increased growth rate of the scaling layer mass. What's more, the solubility product of CaCO_3 remained relatively constant, while that of CaSO_4 decreased with increasing temperature, leading to homogeneous crystallization [25]. Apart from temperature and pressure, changes in flow velocities also influence the form of scaling. Lower flow velocities lead to higher, concentration polarization on the membrane surface compared to that in the bulk in the bulk, whereas higher flow velocity increases shear rate, reducing solute concentration at the membrane surface and affecting scaling behaviors. Lee et al. [117] reported that bulk crystallization (homogeneous) mainly occurred at higher flow velocities, while surface crystallization (heterogeneous) predominantly occurred at lower flow velocities.

In addition to undergoing direct optimization, operation conditions can also be optimized through device innovation to mitigate scaling formation. Vibrating RO membranes could increase flow velocities, thus reducing concentration polarization and membrane scaling. Li et al. [118] found that when vibrations at 20, 53 and 55 Hz were applied, the permeate flux declined after approximately 7-h operation was reduced to 20 %, 16 % and 15 % respectively, compared to a 29 % decline without vibration. Reversing the flow before the induction time of scaling can replace the supersaturated brine with the unsaturated feed flow, effectively resetting the induction clock by "zeroes the elapsed nucleation time". Pomerantz et al. [119] found while the RO flux began to drop after 0.5–1 h under normal conditions, scaling was not observed after 22 h by switching the direction every 0.5 h. What's more, Riley et al. [120] demonstrated that using closed circuit desalination (CCD) to treat PW resulted in minimal scaling and organic fouling on the membrane surface, indicating that CCD could be a promising method for facilitating PW reuse. Lee et al. [121] discovered that rotating RO membranes generated vortices which promoted large crystal formation, thereby reducing the deposition of small particles on the membrane surface. Additionally, higher rotation speeds could enhance shear forces and vortices, thereby improving the anti-scaling behavior.

In summary, an increase in hydraulic pressure and flow velocity could enhance the water flux, but it would also alter the form and tendency of scaling. Innovative flow promotion devices (rotating membrane, vibrating membrane, changing the flow direction and CCD) could significantly mitigate scaling formation by indirectly altering operation conditions.

3.3.2. Membrane properties of NF/RO membranes for mitigating scaling

The desalination performance of high-pressure membrane processes is significantly affected by membrane properties including hydrophilicity, roughness and pore sizes. A high-pressure membrane could be modified into a hydrophilic membrane to reduce scaling by adding nanomaterials and hydrophilic substances [122,123]. For example, Ashraf et al. [124] incorporated nanomaterials into membranes and found that the modification resulted in smaller water contact angles, higher porosity, and larger average pore sizes, thus improving hydrophilicity. In addition, the hydrophilicity of RO membranes was improved by the addition of graphene oxide, which increased the number of hydroxyl and carboxyl groups on the membrane surface [125]. Furthermore, Al-Ghouti et al. [29] demonstrated that the flux decline was 22 % and 15 % for the common RO membrane and the graphene oxide-modified RO membrane, respectively, and the minimal

scaling was observed in the modified membrane (Fig. 8d). The roughness of high-pressure membranes significantly affects scaling. Vrijenhoek et al. [27] demonstrated that, compared to other operation conditions, the roughness of the membrane surface had the greatest impact on scaling. Xu et al. [126] found that the flux decline was more severe for the rough membranes (NF-90, TFC-S and XLE) than the smooth membranes (TFC-HR, TFCULP, ESNA and TMG-10) (Fig. 8a). What's more, the relationship between membrane surface roughness and flux variation is shown in Fig. 8a, with rougher membranes leading to greater flux decline. In addition, the initial flow rate decreased steeply due to "valley clogging", which means that the particle preferentially accumulated in the valley. Different degrees of salt rejections and scaling were observed for NF/RO membranes with different pore sizes. Riley et al. [120] found that the BW30 membrane exhibited higher TDS rejection and more severe scaling than the ECO membrane with larger pore sizes. Kong et al. [127] treated PW using RO membrane (NESA1) and NF membrane (NF90) and observed a significant decline in water flux and severe scaling on the NESA1 membrane (Fig. 8b). Thus, scaling in NF/RO membrane could be decreased by modifying membranes characteristics, including enhancing hydrophilicity, increasing pore size and reducing roughness.

3.3.3. Anti-scalants and pretreatments in NF/RO systems

Although the NF/RO processes often lead to the formation of a dense scale layer on the membrane surface, the scaling layer can be effectively mitigated and stable flux can be maintained by using anti-scalants and pretreatment methods. There are a series of pretreatment methods to control NF/RO scaling including pH adjustment, coagulation and NF filtration. Rahardianto et al. [129] found that the saturation index ($\text{SI} = \text{IAP}/\text{K}_{\text{sp}}$, where IAP was the ion activity product and K_{sp} was the solubility product for the mineral salt of CaCO_3) decreased steadily from 25 to 0.002 as the pH was reduced from 8 to 5. In high-pressure membrane process, coagulants can remove scaling ions and reduce the tendency to scale. However, excessive concentration of coagulants would lead to the formation of a fouling layer on the membrane surface and low concentrations might not significantly impact membrane performance [128]. Kim et al. [130] found that after 24-h operation, the flux of PW treated with coagulation-flocculation-sedimentation decreased to 78% of the initial flux, while the flux of untreated PW decreased to 20% of the initial flux. Moreover, Millar et al. [131] found that 70.5 % of silicates were removed from PW by coagulation with aluminum chlorohydrate (ACH), which reduced the tendency for scale formation [132], with the higher amounts of ACH leading to the higher removal rates. Chang et al. [133] found that the alkalinity (i.e., CO_3^{2-} and HCO_3^-) in shale gas PW was significantly reduced by adding $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ and the SI of CaCO_3 decreased to -0.41, resulting in less scaling. However, excessive use of $\text{Al}_2(\text{SO}_4)_3$ could introduce large amounts of SO_4^{2-} , which led to sulfate scaling on NF membranes [133]. Furthermore, some studies have explored using NF as a pretreatment for the RO process [134]. Liu et al. [135] used a dual NF system for pretreatment before RO, achieving a total water recovery rate of 65.7 %. In the first NF stage, most SO_4^{2-} and a small portion of Ca^{2+} were retained, while in the second NF stage, most of the remaining Ca^{2+} was retained. This process effectively separated SO_4^{2-} and Ca^{2+} into different streams, reducing both NF and RO scaling tendencies and decreasing energy consumption.

Scaling in NF/RO systems can be effectively mitigated by using scale inhibitors. Anti-scalants have been reported to delay crystal growth by adsorbing on the active growth sites of scaling crystals, thereby altering the crystal structure [136], and preventing crystal deposition on the membrane surface [137]. Meantime, Lyster et al. [138] suggested that anti-scalants delayed the nucleation rate of crystals by increasing the surface energy of the nuclei. Al-Mhyawi et al. [128] used RO process to treat PW and found that scaling and flux decline were significantly inhibited by adding anti-scalants dodecyl phenol ethoxylate (DPE), while the effect of anti-scalants oleic acid ethoxylate (OAE) was not obvious after 100 min, as shown in Fig. 8c. Scaling could also be

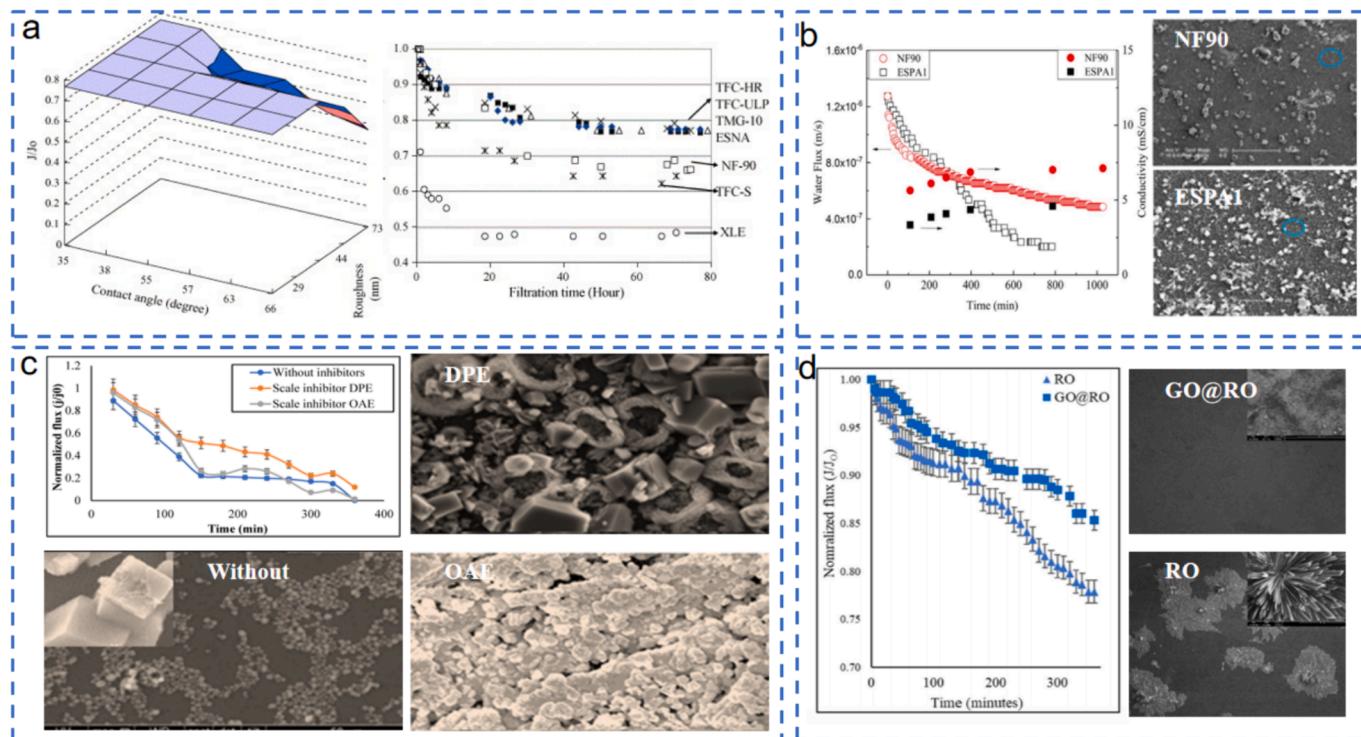


Fig. 8. The influences of membrane properties (roughness, hydrophilicity, and pores size) and anti-scalants on membrane flux and scaling in NF/RO process: (a) Different roughness (rough membrane (NF-90, TFC-S, XLE) and smooth membrane (TFC-HR, TFCULP, ESNA, TMG-10)) [126], Copyright (2006) Elsevier B.V., (b) pores size (ESPA1-small pores and NF90-large pores) [127], Copyright (2018) Elsevier B.V., (c) Anti-scalants (DPE and OAE) [128], Copyright (2021) American Chemical Society, and (d) Hydrophilicity (GO@RO-Hydrophilic membrane and RO-common membrane) [29], Copyright (2020) Elsevier B.V.

prevented by the repulsion between anti-scalants accumulated on the membrane surface and calcite ions [139]. Furthermore, different types of anti-scalants, scale inhibition mechanisms, and their applications to control scaling in the RO process have been reviewed by Yu et al. [16].

In summary, developing new devices and modified membranes have proven to be effective strategies to mitigate scaling in NF/RO membranes wastewater treatment. Although pretreatments and the addition of anti-scalants were crucial in reducing scaling, it was also essential to optimize the appropriate dosages of these chemical agents. Additionally, the scaling tendency was also affected by membrane properties and operation conditions, especially hydraulic pressure. The application and performance of high-pressure membrane processes for treating PW are presented in Table S3 (Supporting Information).

4. Comparison, challenges, and future outlook

4.1. Comparison of inorganic scaling for primary membrane desalination technologies

Based on the different driving forces and membrane characteristics of NF/RO (hydraulic pressure), FO (osmotic pressure) and MD (vapor pressure), the scaling behaviors of those membrane processes varied significantly. In MD, the high temperature of the feed produced vapor pressure driving the process, and could reduce the nucleation induction time for supersaturation, thus accelerating scaling [140]. However, the elevated temperature also increased the diffusion coefficients, which hindered concentration polarization and mitigated scaling [141]. FO process is driven by the osmotic pressure difference between the feed and draw solutions. The high concentration of draw solutions can lead to severe ICP in AL-FS mode, but the compensation mechanism of ICP can maintain a stable flux. Compared to MD, the effect of hydraulic resistance on scaling was more pronounced in NF/RO and FO systems [142]. Tow et al. [143] compared membrane processes under similar initial

fluxes and found that the resistance to calcium sulfate scaling decreased in the order of FO, MD, and RO, as shown in Fig. 9a. Moreover, Jang [144] evaluated the behaviors of silica fouling and organic fouling in different membrane processes, as shown in Fig. 9b. The FO process demonstrated excellent scaling resistance, while MD process showed superior fouling resistance.

The various properties of membranes, tailored to each process, significantly affect their scaling inhibition capabilities. Compared to FO and NF/RO membranes, MD membranes were more hydrophobic and had lower surface energy, thus resulting in a higher barrier (ΔG^*) for heterogeneous nucleation and leading to longer induction times [145]. Moreover, many large crystals could not enter the small pores of NF/RO and FO membranes [143]. In contrast, MD membranes could experience a rapid flux decline due to the accumulation of scaling within their large membrane pores [146]. The pore size of membranes influences not only scaling performance but also cleaning efficiency. It was found by Jang et al. [144] that scaling on the MD process was more difficult to remove using physical methods, compared to FO and RO, as shown in Fig. 9c.

Therefore, despite the shared mechanism of scaling, the scaling properties of different types of desalination membranes (NF/RO, FO, and MD) varied significantly, with FO exhibiting excellent scaling resistance. The large pore sizes of MD membranes led to scaling within the membrane pores, making it difficult to clean, their hydrophobicity also resulted in longer induction times mitigating scaling. The primary metrics (i.e., anti-scaling, anti-organic, energy consumption, scale-up, cleaning efficiency, pretreatment demand, effluent quality and salinity limit) of these desalination membrane technologies for treating PW are systematically evaluated (Fig. 10a).

4.2. Challenges and future outlook

In recent years, there has been growing interest in the study of scaling in membrane processes during the treatment of PW, with

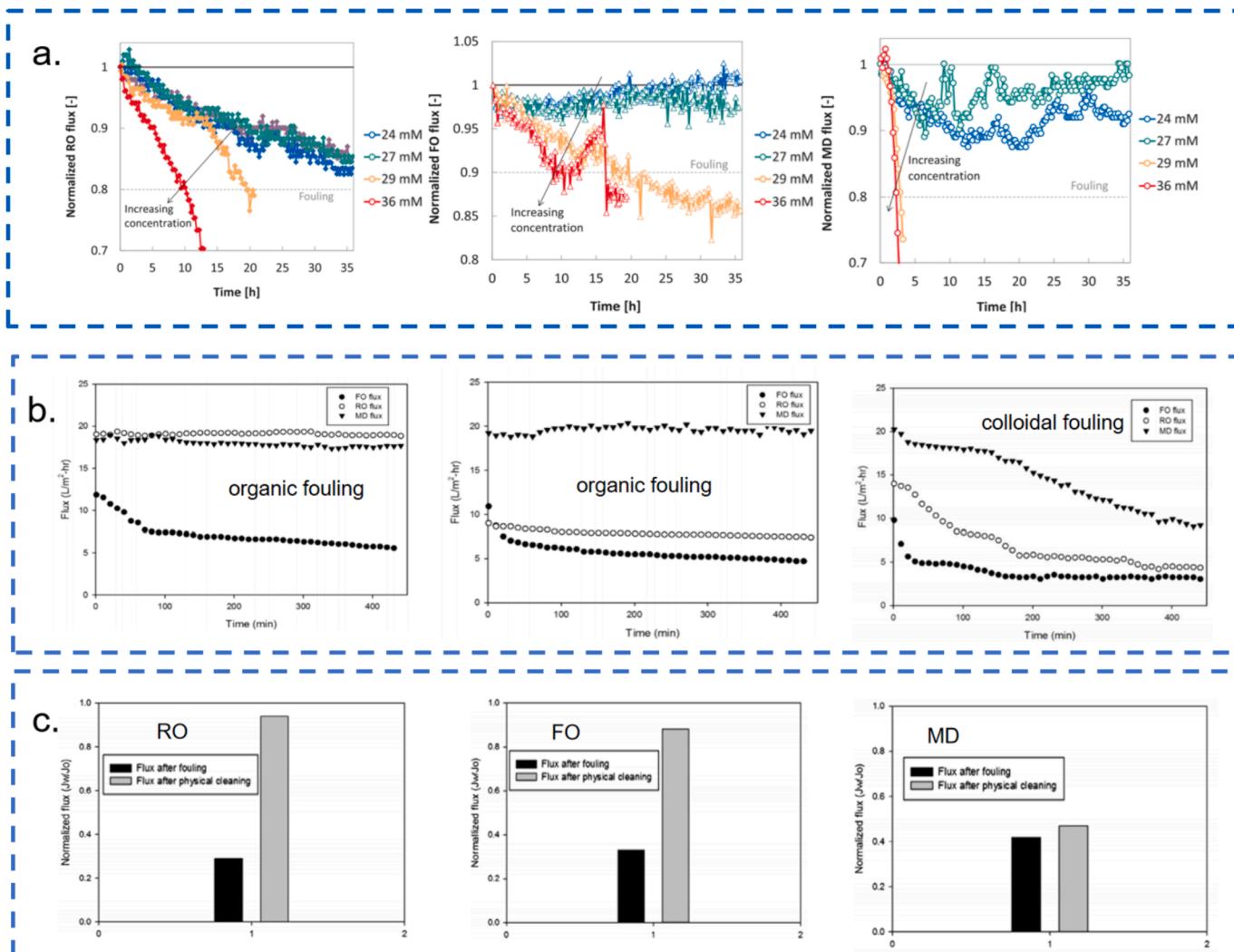


Fig. 9. Comparison of difference of scaling and cleaning efficiency on NF/RO, FO and MD: (a) Anti-scaling [143], Copyright (2018) Elsevier B.V., (b) organic fouling and silica gel fouling [144], Copyright (2016) American Chemical Society, and (c) cleaning performance [144], Copyright (2016) American Chemical Society.

significant progress made in understanding and controlling scaling. However, there are several scientific and technical challenges, which may benefit from the directions suggested in this article (Fig. 10b).

- (1) The inorganic scaling during using MD, FO and NF/RO processes for treating PW can be alleviated by optimizing operation conditions, but the permeate flux will also vary with changing operation conditions. There may be a trade-off between the scaling mitigation behaviors and the flux. For example, the scaling was mitigated by reducing feed temperatures in MD, but the permeate flux also decreased [39]. Moreover, the high permeate flux would lead to severe membrane scaling [37]. However, the development of process devices (e.g., vibration [118], rotation [121] and two-stage membrane distillation [60]) could alleviate the trade-off effect and improve desalination performance by both mitigating scaling tendencies and enhancing permeate flux. Thus, it is essential to analyze the association between the scaling mitigation behaviors and the flux as well as to innovate the new device for inorganic scaling control.
- (2) During desalination processes using MD, FO and NF/RO, the presence of organic matters (or organic fouling) can influence scaling behaviors mainly in three ways (Fig. 11a), namely affecting homogeneous nucleation by intervening scalant-scalant interactions [147], altering the free energy barrier to

heterogeneous nucleation [148] and changing the deposition kinetics [149]. However, inconsistent results existed on the effects of organic foulants on mineral scaling in different desalination processes. Wang et al. [150] found that the gypsum scaling on NF membranes fouled by organic foulants was more severe than that on virgin NF membranes (Fig. 11d). Similarly, in FO process, membrane fouled by humic acid (HA), sodium alginate (SA) and bovine serum albumin (BSA) faster decreased than virgin membrane (Fig. 11b) [151]. However, Wang et al. [152] discovered that organic matters (e.g., HA, SA and BSA) at low concentrations (50 mg/L) could mitigate CaSO_4 scaling in MD (Fig. 11c). Thus, it is urgent to develop novel organic detection methods [153,154] due to complex organic matters of PW, and it is essential to further study the mechanism of organic fouling on scaling to mitigate the effect of organic fouling on scaling in the actual desalination process.

- (3) Various chemical agents have been used to mitigate and clean membrane scaling, but chemical agents with inappropriate dosages may pose some risks to desalination performance. Excessive dosages would change membrane properties (e.g., alkaline cleaning [155]) and arouse organic fouling and inorganic scaling (e.g. aluminum coagulation [133]), while slight dosages could diminish the efficiency of mitigating scaling [156]. Thus, it is vital to select the appropriate chemical agents with optimized

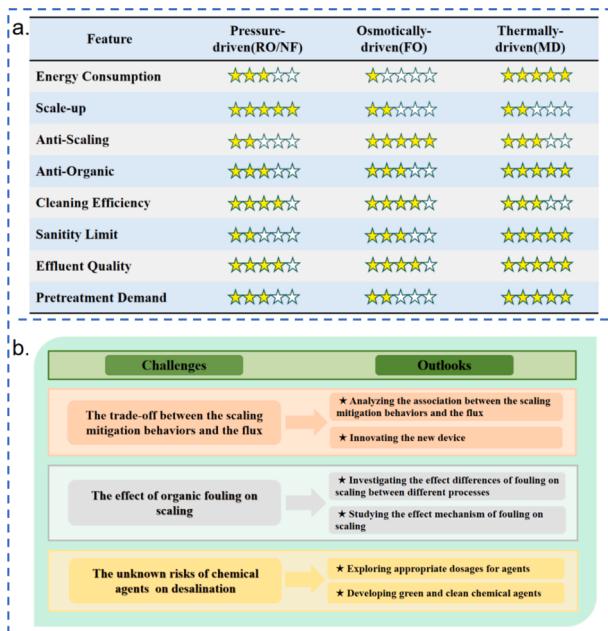


Fig. 10. Comparison of (a) primary metrics and (b) challenges and corresponding outlook of desalination membrane technologies for treating PW.

dosages. Future studies should focus on exploring the appropriate dosages for each chemical agent as a guide to desalination processes and experimental studies. Moreover, adding anti-scalants had also the risks of scaling [157] and biofouling [158], thus, there also is a need to develop green and clean chemicals to prevent secondary fouling.

5. Conclusion

As the interest in treating O&G PW using desalination membrane processes grows, the issue of scaling within these processes has garnered

increasing attention. Recent studies on scaling in the treatment of PW using NF/RO, FO and MD membrane technologies are systematically reviewed in this paper, and the main concluding remarks are drawn.

- (1) Research on scaling during membrane desalination for PW has significantly increased, with an average annual growth rate of 17 %, particularly in MD processes. Research trends and hotspots of scaling in membrane desalination have gradually shifted from focusing on a specific membrane process to highlighting practical applications. Scientists have shown particular concern for factors influencing scaling, including operational conditions, membrane characteristics and pretreatments.
- (2) Inorganic scaling in MD could be regulated by adjusting operation conditions, membrane properties and pretreatments. Adjusting operation conditions (median flow velocity and low temperature) could effectively mitigate scaling in MD. Developing new materials (e.g., PPO), modifying membranes (super-hydrophobic, omniphobic and Janus membranes) and changing membrane properties (e.g., reducing pore sizes and membrane roughness) could benefit from alleviating scaling. The types of pretreatments including pH adjustment, coagulation and membrane filtration were mainly used to remove organic matters and inorganic ions to mitigate MD scaling.
- (3) In addition to membrane roughness and pretreatment, the scaling tendency was mainly impacted by draw solutions, membrane properties and cleaning methods in FO. Although DS (scale inhibitors and acidic solutions) could greatly mitigate inorganic scaling in FO, DS containing divalent salts might exacerbate scaling due to RSF. The scaling behaviors could be changed by the membrane properties including AL-FS orientation which affected scaling on the membrane surface and functional groups which could alter membrane potential. Compared to physical cleaning, FO scaling could be more effectively cleaned by chemical cleaning and combined cleaning.
- (4) The form of scaling (bulk crystallization and surface crystallization) and scaling tendency in NF/RO were significantly affected by hydraulic pressure. Developing new devices (e.g., rotating membrane, vibrating membrane, changing the flow direction,

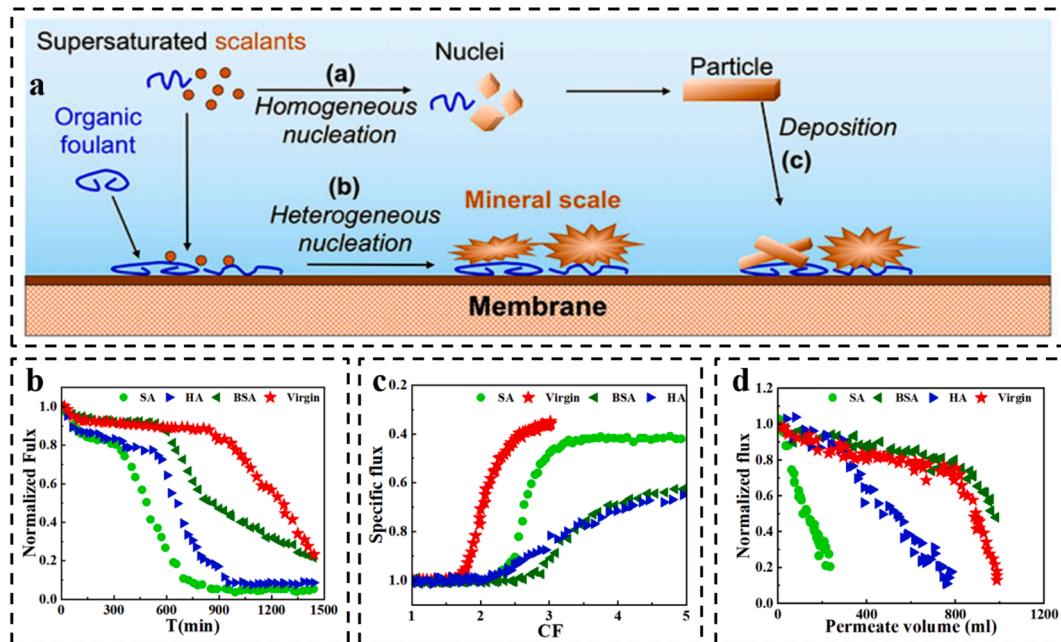


Fig. 11. The effects of fouling on scaling behaviors: (a) Potential mechanism by which organic foulants impact the nucleation and growth of mineral scales [14], Copyright (2023) American Chemical Society. Scaling on membrane surface influenced by organic matter in (b) FO [151], Copyright (2014) Elsevier B.V., (c) MD [152], Copyright (2021) Elsevier B.V., and (d) NF [150], Copyright (2016) Elsevier B.V. The data are extracted from those figures and replotted.

- and closed circuit desalination) and changing membrane properties (increasing hydrophilicity and enhancing pore sizes) could significantly mitigate scaling. Anti-scalants could also effectively mitigate scaling by increasing the surface energy of nuclei and altering the crystal structure.
- (5) The scaling behaviors in MD, FO and NF/RO for PW treatment varied significantly due to their different driving forces. Compared to NF/RO and MD processes, the FO process with the compensation mechanism demonstrated the best resistance to inorganic scaling. Moreover, the small pore sizes (FO and NF/RO membranes) and hydrophobicity (MD) could influence scaling and cleaning performance. To address the existing challenges, future research directions were proposed, including trade-off of scaling mitigation behaviors and the permeate flux, exploring the effect of fouling on scaling, and solving the unknown risks of chemical agents on desalination.

Declaration of competing interest

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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary material

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