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Membrane Cleaning Technologies for Water Treatment: A Review

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Abstract: Membrane fouling persists as a critical bottleneck limiting the sustainability of membrane-based water treatment processes. Consequently, the deployment of robust cleaning strategies is paramount for restoring permeate flux and prolonging membrane longevity. This review critically evaluates the mechanisms and efficacy of established physical and chemical cleaning protocols. A comprehensive synthesis is provided, covering conventional techniques—such as hydraulic backwashing and Chemically Enhanced Backwash (CEB)—alongside emerging innovations including ultrasonic cleaning, Advanced Oxidation Processes (AOPs), and hybrid physical-chemical systems (e.g., micro-nano bubble and electrochemically-assisted methods). Comparative analysis reveals that while physical cleaning offers operational simplicity with minimal risk to membrane integrity, its utility is predominantly restricted to reversible fouling mitigation. Conversely, chemical cleaning exhibits superior proficiency in removing irreversible foulants, frequently achieving Flux Recovery Rates (FRR) exceeding 90%, although it introduces concerns regarding membrane degradation. Notably, AOP-based and integrated approaches demonstrate exceptional remediation efficiency, highlighting a promising trajectory for future fouling management strategies.

Keywords: Water Treatment; Membrane Separation; Membrane Fouling; Physical Cleaning; Chemical Cleaning.

1. Introduction

Membrane separation technology has established itself as a pivotal solution for chemical recovery, cell harvesting, and water treatment, owing to its high-efficiency retention of particles, colloids, and microorganisms without requiring chemical additives. However, the inevitable occurrence of membrane fouling during filtration precipitates severe flux decline and escalates operational costs, persisting as the primary impediment to the widespread industrial implementation of this technology [1].

To mitigate fouling, diverse strategies have been employed, ranging from feed pretreatment and the fabrication of anti-fouling materials to the optimization of operational parameters[2-4]. Despite these preventive measures, foulant accumulation remains unavoidable during long-term operation, necessitating periodic cleaning to purge deposits and restore flux. While chemical cleaning typically yields superior flux recovery compared to physical methods, it presents significant drawbacks; prolonged exposure to aggressive agents can compromise membrane integrity and degrade separation performance. Conversely, although physical cleaning offers operational simplicity and avoids secondary pollution, it frequently fails to eradicate irreversible fouling sequestered on the membrane surface or within internal pores [4].

In response to these challenges, this review systematically critically examines the fundamental principles, efficacy, and limitations of physical, chemical, and emerging membrane cleaning technologies. Beyond the mechanisms, it investigates the impact of key operating parameters on cleaning efficiency and evaluates the specific applicability of distinct techniques (summarized in Table 1). Ultimately, this work aims to provide a theoretical foundation for the optimal design of membrane cleaning protocols in sustainable water treatment applications.

2. Principles and Classification of Membrane Fouling

Membrane fouling is defined as the phenomenon whereby foulants—encompassing suspended particulates, colloids, organic matter, microorganisms, and inorganic salts—adsorb onto the membrane surface, form deposits, or occlude membrane pores during filtration. This accumulation precipitates a decline in permeate flux, an escalation in transmembrane pressure (TMP), or alterations in membrane selectivity [5].

The degradation of membrane flux is primarily governed by two mechanisms: external fouling and internal fouling [6]. External fouling denotes the deposition of particles, colloids, and macromolecules on the membrane surface, commonly referred to as the "fouling layer." This layer typically manifests in two distinct forms: a cake layer, formed by the accumulation of retained solids, and a gel layer, resulting from the concentration polarization and precipitation of macromolecular organics, colloids, and inorganic solutes [7]. Conversely, internal fouling arises from the adsorption and entrapment of solutes and fine particles within the membrane substructure. Specifically, this manifests as pore wall adsorption, pore narrowing, or complete pore blockage, involving complex interactions driven by steric effects, covalent bonding, and non-covalent forces [8].

Based on their physicochemical characteristics, foulants are categorized into inorganic, organic, and biological types:

1) Inorganic fouling is predominantly caused by calcium sulfate, calcium carbonate, calcium phosphate, metal oxides, hydroxides, and colloidal matter [9]. These constituents precipitate or scale on the membrane surface, thereby reducing the effective pore size and impeding flux.

2) Organic fouling comprises compounds such as proteins, lipids, carbohydrates, and humic substances. These materials,

often hydrophobic in nature, tend to adsorb onto the membrane surface or become entrapped within the pores, inducing severe fouling [10].

Table 1. Comprehensive comparison of different cleaning strategies for membranes in drinking water treatment

Cleaning Technology	Primary Mechanism	Target Pollutants	Flux Recovery Rate (FRR)	Main Advantages	Limitations	Technology Maturity
Physical Cleaning	Hydraulic shear forces loosen the cake layer	Loose pollutants deposited on the membrane surface	70% ~ 85%	No chemical reagents; operational simplicity; low cost; no membrane damage	Ineffective against pore blocking and strongly adsorbed foulants; requires frequent operation	Commercialized
Conventional Chemical Cleaning	Hydrolysis, saponification, complexation, electrostatic repulsion	Organic adsorption within pores, biofilms, inorganic scaling	90% ~ 98%	Thorough cleaning efficiency	Generation of secondary pollution; high reagent costs; shortens membrane lifespan; long downtime	Commercialized
Advanced Oxidation Processes (AOPs) Cleaning	Strong oxidative degradation by free radicals (•OH, $SO_4^{\bullet-}$)	Recalcitrant organic gel layers, organic adsorption in pores, extracellular polymeric substances (EPS)	95% ~ 99%	Rapid reaction rate; capable of mineralizing organics; near-complete flux recovery	High risk of membrane aging (especially for polyamide membranes); requires strict condition control; high operational costs	R&D / Pilot Scale
Micro-nano Bubble Assisted Chemical Cleaning	Bubble collapse shockwaves, enhanced mass transfer, interfacial adsorption	Mixed pollutants on the surface and in shallow pores	90% ~ 96%	Significantly reduces reagent dosage (>50%); enhances permeability of cleaning solution; environmentally friendly	High energy consumption of bubble generators; difficult control of bubble stability	R&D Phase
Electrochemical Assisted Cleaning	In-situ generation of gas scouring, electrostatic repulsion, electro-oxidation	Biological/organic fouling on conductive or charged surfaces	85% ~ 99%	In-situ online cleaning; high degree of automation; eliminates transport/storage of hazardous chemicals	Risk of by-products (DBPs); electrode corrosion issues; primarily limited to conductive membrane materials	Laboratory Phase

3) Biological fouling (biofouling) is driven by the proliferation of microorganisms—including bacteria, viruses, and fungi—on the membrane surface. These organisms secrete substantial quantities of extracellular polymeric substances (EPS) to establish biofilms, which significantly aggravate fouling, reduce permeate flux, and heighten the risk of membrane biodegradation [11].

Furthermore, membrane fouling is classified into reversible, irreversible, and permanent categories based on the binding affinity between foulants and the membrane, and the specific cleaning protocol required for restoration. As illustrated in Fig. 1 [12], reversible fouling originates from loosely attached foulants and can be readily mitigated via physical cleaning. In contrast, irreversible fouling stems from pore blocking and strongly adhered foulants, necessitating chemical cleaning to restore permeability. Permanent fouling refers to residues that exhibit resistance to standard chemical protocols; their gradual accumulation over extended operation ultimately dictates the membrane's service lifespan. Generally, reversible fouling is attributed to cake layer formation, whereas irreversible and permanent fouling are associated with internal pore blockage and strong surface adsorption [12].

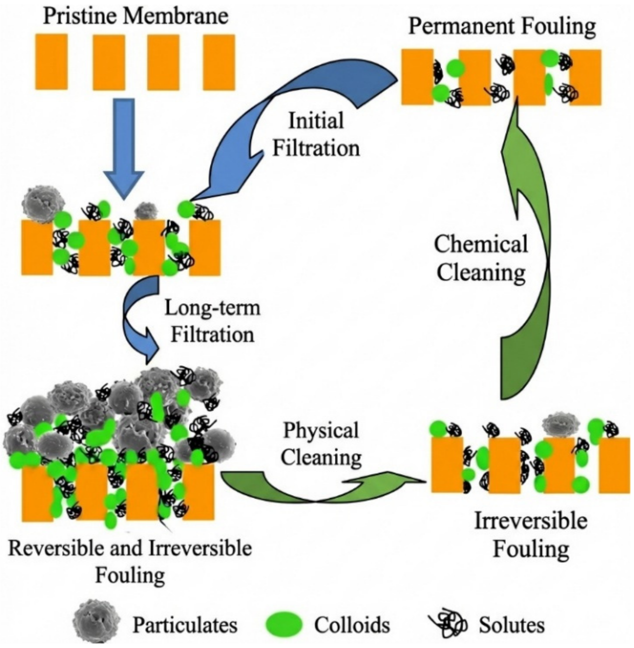

Fig 1. Schematic illustration of the formation and removal of reversible, irreversible, and permanent fouling during membrane filtration.

3. Physical Cleaning

Physical cleaning is defined as the removal of foulants through the disruption of binding forces between pollutants and the membrane surface via mechanical mechanisms—such as hydraulic shear, backwashing, or air-water scouring—without the introduction of chemical reagents. The primary objective of this approach is the mitigation of reversible fouling, specifically the loosely adhered cake or gel layers on the membrane surface. Conversely, physical cleaning is generally considered ineffective for the complete remediation of irreversible fouling that penetrates deep within the membrane pores or exhibits strong adsorption affinity to the membrane matrix.

Conventional physical cleaning strategies encompass air sparging, sponge ball scrubbing, backwashing, and back-pulsing. Among these, air sparging and sponge ball scrubbing are recognized as the most operationally straightforward modalities [4]. Air sparging employs a multifaceted mechanism for fouling control: micro- or nano-scale bubbles coalesce to form a dense barrier near the membrane interface, effectively inhibiting direct contact between foulants and the surface. Simultaneously, larger bubbles or slugs (with diameters exceeding 10 mm) induce turbulence and wake instabilities during their ascent, generating shear forces that actively scour foulants, particularly those constituting the cake layer [13, 14].

3.1. Hydraulic Backwashing

Hydraulic backwashing is implemented by applying a reverse pressure gradient to drive clean filtrate from the permeate side back to the feed side. The cleaning mechanism leverages physical drag and shear forces generated by this reverse flow to loosen and detach the cake layer deposited on the membrane surface. Simultaneously, the reverse hydraulic flow scours the membrane pores, expelling a fraction of the pore-blocking foulants. While this method effectively removes reversible surface foulants and significantly restores permeate flux, its efficacy is constrained against adsorptive irreversible fouling sequestered deep within the pores. Typically, backwashing is automated to occur periodically (e.g., 30–60 seconds every 15–60 minutes of filtration), with the backwash intensity maintained at 1.5–2.5 times the operating flux. This technology is predominantly applicable to hollow fiber and tubular membrane modules (ultrafiltration/microfiltration) that possess self-supporting structures, serving as the fundamental strategy for fouling control in water treatment engineering.

The performance of backwashing is contingent upon multiple variables, including membrane material properties, feed water matrix, and the specific duration and frequency of the cleaning cycle. Regarding membrane composition, ceramic membranes exhibit superior hydraulic fouling reversibility compared to their polymeric counterparts. For instance, Alresheedi et al. [15] demonstrated that the backwashing efficiency of ceramic ultrafiltration membranes is 1.5–2 times higher than that of polymeric variants. Research by Ye et al. [16] indicated that extending the filtration interval from 20 to 60 minutes failed to increase the fraction of reversible fouling removable by backwashing; moreover, prolonging filtration to 90 minutes significantly aggravated irreversible fouling. Conversely, increasing the backwash duration from 10 to 30 seconds reduced the initial transmembrane pressure (TMP) of the subsequent filtration cycle by over 50% and decreased the fouling rate by half.

Generally, the flux recovery rate (FRR) of this method can approach 80%–90% when foulants are dominated by loose cake layers, particulates, and colloids. However, when fouling is driven by organic adsorption, biofilms, or inorganic scaling (irreversible fouling), the recovery rate typically diminishes to below 60%.

3.2. Back-pulsing

Back-pulsing is a cleaning technique characterized by the application of high-frequency, extremely short-duration reverse pressure pulses. The fundamental cleaning mechanism leverages instantaneous pressure fluctuations to generate intense fluid shockwaves and reverse shear forces within both the membrane pores and the surface boundary layer. These forces induce a slight elastic expansion of the membrane pores, thereby rapidly disintegrating and ejecting dense cake layers and pore blockages [17]. Compared to conventional backwashing, back-pulsing exhibits a significantly higher energy density, enabling it to eradicate strongly adhered reversible fouling more thoroughly and effectively mitigate membrane flux decay. Defined by its "high frequency and short duration," the pulse interval typically spans merely 0.1 to 1 second. However, the generation of instantaneous high pressure necessitates membrane materials with robust mechanical integrity; consequently, this technology is predominantly applicable to inorganic ceramic membranes or reinforced hollow fiber membranes. It is particularly advantageous in filtration scenarios involving feed solutions with high solid content or elevated viscosity. The performance efficacy of back-pulsing is illustrated in Fig. 2 [17].

Implementation of back-pulsing occurs primarily via two modalities: hydraulic back-pulsing and gas back-pulsing. Hydraulic back-pulsing functions by driving clean water reversely into the membrane module, whereas gas back-pulsing involves introducing compressed gas directly into the permeate side to displace the liquid retained in the filtration chamber and membrane pores. Under identical operating parameters, the efficacy of gas back-pulsing is generally inferior to that of its hydraulic counterpart, and it carries the risk of inducing embrittlement in certain polymeric materials [17]. Ma et al. [18] conducted a comparative analysis of these methods against conventional filtration; results indicated that after a 1-hour filtration cycle, hydraulic back-pulsing enhanced the flux by a factor of 3.7, whereas gas back-pulsing achieved a 3.2-fold increase. The efficiency of back-pulsing is contingent upon multiple variables, including the amplitude of the transmembrane pressure (TMP) per pulse, pulse duration, and frequency.

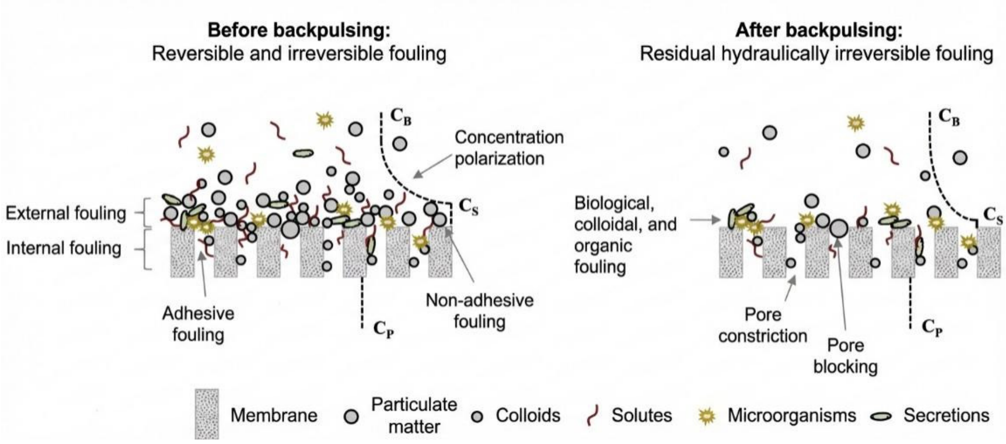

Fig 2. Overview of membrane before and after backpulsing in processes. Where C_B, C_S and C_P are the solute concentration in the bulk, near the membrane surface and in the permeate, respectively

The flux recovery rate (FRR) for this method typically ranges from 60% to 95%. Standard operational protocols involve pulse durations of 0.1–1 s, with a reverse flow rate approximately 2–10 times the operating flux, or an instantaneous reverse pressure differential reaching 70%–100% of the membrane module's maximum permissible limit. A typical cleaning cycle comprises 1–5 consecutive pulses separated by intervals of 1–5 seconds.

3.3. Ultrasonic Cleaning

Ultrasonic cleaning is executed by introducing acoustic energy into the liquid medium surrounding the membrane module. Its fundamental mechanism leverages the cavitation effect, which induces vigorous convection (acoustic streaming) accompanied by micro-jets, shockwaves, and localized heating, thereby facilitating high-efficiency foulant removal. Ultrasonic waves propagate through gaseous, liquid, and solid media via alternating cycles of adiabatic compression and expansion, creating oscillating regions of high and low pressure. During the rarefaction phase, the medium is subjected to negative pressure, triggering cavitation; conversely, during the compression phase, the violent collapse of cavitation bubbles generates transient "hot spots" characterized by extreme conditions (localized temperatures reaching 5,000 K and pressures up to 1,000 atm in aqueous solutions). The energy released by this collapse disrupts the physicochemical interactions between foulants and the membrane surface, effectively detaching the fouling layer and inhibiting further deposition [19]. In operational practice, parameters such as frequency (typically 20–50 kHz) and power density require precise regulation, with intermittent operation often employed to mitigate heat accumulation. Given that intense mechanical vibrations can compromise membrane integrity, this technology is predominantly restricted to robust inorganic ceramic or metallic membranes, while its application to organic polymeric membranes remains strictly limited.

Table 2. Physical membrane cleaning strategies and efficiencies

Cleaning Strategy	Membrane	Foulants	Key Operating Conditions	Flux Recovery (%)	Ref.
Hydraulic Backwashing	PES (MWCO 70 kDa)	Natural Organic Matter (NOM)	Backwashing for 1–2 min after 7 h of filtration	85	[23]
	PSF (MWCO 25 kDa)	Algae (Reservoir water)	Backwashing only, duration 30 min	68	[24]
	PSF (MWCO 25 kDa)	Algae (Reservoir water)	Forward flushing only, duration 30 min	59	[24]
	PSF (MWCO 25 kDa)	Algae (Reservoir water)	Backwashing + Forward flushing, duration 30 min	84	[24]
	PVDF (0.04 μm)	Seawater organic matter	Backwashing for 30 s at 110 LMH	85	[16]
Back-pulsing	Ceramic membrane (0.2 μm)	Chromium hydroxide suspension	Pulse duration 0.5 s, amplitude 170 kPa	100	[25]
	Ceramic membrane (0.35 μm)	Primary sewage effluent	Frequency 1 pulse/30 s; pulse duration 0.3 s	65	[26]
Ultrasonic Cleaning	PSF (0.1 μm)	Wastewater organics	35 kHz, ultrasonic bath	57	[27]
	PVDF (0.45 μm)	Protein solution	20 W power, water bath, 60 min	43	[28]
	PTFE (0.6 μm)	Humic acid (HA)	15 W power, probe-type, 25 min	45	[29]

The efficacy of ultrasonic cleaning is contingent upon critical variables, principally frequency and acoustic intensity. A study investigating ceramic membranes fouled by sulfate polystyrene latex particles [20] demonstrated that at high frequencies (>200 kHz), full flux restoration necessitates a power density exceeding 1.05 W/cm² and a treatment duration of over 30 seconds; conversely, lower frequencies achieve comparable results with reduced power input. Furthermore, Ohl et al. [21] proposed that a tandem frequency approach (combining high and low frequencies) yields superior cleaning outcomes compared to single-frequency modes, attributed to the increased proliferation of micro-bubbles on the membrane surface. Similarly, ultrasonic intensity enhances cleaning efficiency by amplifying acoustic amplitude and fluid turbulence. Maskooki et al. [22] confirmed via flat-sheet membrane experiments that permeate flux exhibits a linear correlation with ultrasonic power, a phenomenon ascribed to the intensified cavitation effect. However, it must be noted that excessive power intensity significantly heightens the risk of structural damage to the membrane.

In summary, flux recovery rates (FRR) exceeding 90% are achievable for surface-deposited cake layers, whereas efficacy for pore blockage remediation typically ranges between 70% and 80%. Standard operational protocols involve frequencies of 20–45 kHz, power densities controlled between 0.1–0.5 W/cm², cleaning durations of 5–20 minutes, and maintenance of the cleaning solution temperature at 30–50°C.

The efficiencies of various physical cleaning methods are presented in Table 2 [4].

4. Chemical Cleaning

Chemical cleaning is defined as the restoration of membrane separation performance through the application of specific chemical reagents. This process relies on distinct reaction mechanisms—including oxidation, solubilization, chelation, and hydrolysis—that occur between the agents and the foulants. These chemical interactions actively disrupt both the structural integrity of the foulants and their adhesive binding to the membrane surface. Consequently, this method is recognized as a pivotal strategy for mitigating the flux decline associated with prolonged operation. The primary objective of chemical cleaning is the remediation of irreversible fouling that remains recalcitrant to physical cleaning methods; this specifically encompasses organic adsorbates sequestered deep within membrane pores, tenacious biological biofilms, colloidal deposits, and inorganic scales crystallized on the membrane surface. The operational protocol typically involves immersing the membrane elements in a chemical solution, followed by a thorough rinsing phase to eliminate residual reagents and detached foulants [30].

4.1. Acid/Alkali Cleaning

Acid cleaning exploits chemical interactions between acidic solutions and inorganic scales—specifically carbonates or metal oxides precipitated on the membrane—to dissolve and eliminate these deposits. Consequently, this technique serves as the principal method for mitigating inorganic fouling. Frequently employed acidic agents include hydrochloric, citric, nitric, and phosphoric acids, with the specific selection contingent upon the distinct foulant composition adhering to the membrane surface [30].

In contrast, alkali cleaning operates by utilizing alkaline solutions to react with adsorbed organic matrices and biological foulants, such as proteins, thereby inducing their solubilization and detachment. Accordingly, this approach is regarded as the preeminent strategy for the remediation of organic fouling. Typical alkaline reagents encompass hydroxides, carbonates, and perborates. While sodium hydroxide (NaOH) is known to significantly enhance cleaning efficacy, recent investigations indicate that it induces accelerated aging and the degradation of chemical constituents in polyvinylidene fluoride (PVDF) membranes [31], potentially leading to the generation of microplastics in the effluent [32].

4.2. Advanced Oxidation

Advanced oxidation cleaning is characterized as an intensive remediation strategy wherein strong oxidizing radicals, generated via Advanced Oxidation Processes (AOPs), are deployed to eliminate membrane fouling. AOPs function by directly degrading organic foulants adsorbed onto the membrane; concurrently, they modify the physicochemical properties of these foulants—specifically their functional groups, hydrophobicity, and surface charge—through chemical oxidation. These alterations attenuate the adhesive interactions between the foulants and the membrane surface, thereby facilitating foulant detachment and effectively restoring filtration efficiency [33]. This method exhibits exceptional efficacy in removing irreversible organic fouling that remains recalcitrant to conventional chemical agents, leading to significant restoration of membrane flux. Given that strong oxidation may induce polymer aging, this technology is predominantly applied to systems utilizing oxidation-resistant materials, such as PVDF and ceramics, and is frequently leveraged for fouling control in the treatment of high-concentration refractory organic wastewater.

Innovative applications of this technology have been demonstrated in recent studies. Li et al. [34] developed a sodium percarbonate-manganese dioxide (SPC-MnO_2) effervescent tablet. Upon hydration, the tablet releases oxygen bubbles and hydroxyl radicals, which efficiently eradicated HA-induced membrane fouling within 5 minutes. This protocol increased the terminal membrane flux from 50% to 95% and reduced irreversible fouling resistance by over 90%. Further advancements were reported by Xiao et al. [35], who employed a peroxymonosulfate/hydroxylamine/ferrous iron (PMS/NH_2OH/Fe(II)) system noted for its high radical yield. Mediated by hydroxyl radicals (HO·), this system successfully restored the flux of membranes fouled by HA, bovine serum albumin (BSA), and SA to 96%, 97%, and 103% of their initial values, respectively, in dead-end filtration mode. After multiple cleaning cycles, the flux increased to 181%–219% of the initial value while maintaining stable rejection performance, thereby achieving a synergistic breakthrough in both selectivity and permeability. Additionally, Xu et al. [36] proposed a novel strategy utilizing membrane foulants themselves as driving agents. Through a foulant-driven permanganate/peroxymonosulfate (PM/PMS) process (FDPP), they achieved simultaneous micropollutant degradation and fouling control. Results indicated that membranes fouled by BSA, HA, and SA were effectively cleaned in just 6 minutes, with flux recovery rates reaching

99.9%, 97.6%, and 89.4%, respectively. Notably, the flux recovery rate remained above 96% even after 20 repeated fouling-cleaning cycles.

Typically, advanced oxidation cleaning achieves flux recovery rates (FRR) of 90%–100%. Standard operational protocols entail the addition of oxidants—such as hydrogen peroxide, ozone, or persulfate—to the cleaning solution. Activation of these oxidants is facilitated by UV irradiation, ultrasonic assistance, or transition metal catalysts, generating highly oxidizing hydroxyl or sulfate radicals *in situ* on the membrane surface. These radicals directly oxidize macromolecular foulants, inducing chain scission, degradation, and ultimate mineralization.

4.3. Combined Physical and Chemical Cleaning

4.3.1. Chemically Enhanced Backwashing (CEB)

Chemically Enhanced Backwashing (CEB) constitutes an on-line maintenance strategy wherein low-concentration chemical agents are injected into the backwash stream, driven reversely through the membrane pores, and subjected to a brief static soaking period. The fundamental principle synergizes the dual effects of physical hydraulic shear and chemical reactivity: hydraulic action loosens the sediment layer, while chemical agents (e.g., sodium hypochlorite, acids, or alkalis) disrupt the adhesive binding forces between foulants and pore walls via oxidation, sterilization, or solubilization mechanisms. CEB effectively remediates irreversible fouling that remains recalcitrant to simple physical cleaning, significantly mitigating the escalation of transmembrane pressure (TMP) and reducing the frequency of shutdowns required for intensive offline cleaning. Operational protocols typically entail periodic automatic cycles (e.g., daily or weekly). Reagent concentrations are maintained at levels substantially lower than those used in offline cleaning (typically 200–500 mg/L), accompanied by a short contact time of 5–20 minutes. This technology is extensively implemented in Membrane Bioreactors (MBR) and large-scale ultrafiltration/microfiltration systems for municipal water treatment, serving as a pivotal process for sustaining long-term operational stability.

In a hybrid system employing PVDF membranes for coagulation-ultrafiltration treatment, H_2O_2 was intermittently dosed into the feed tank during the cleaning cycle. Over a 60-day operational period, this method significantly suppressed bacterial proliferation within the membrane tank and reduced the accumulation of extracellular polymeric substances (EPS)—specifically proteins and polysaccharides—on the membrane surface by more than 50%. The study further confirmed that the milder oxidative potential of H_2O_2 does not compromise the structural integrity of the membrane [37].

Experimental investigations [38] indicate that backwashing with a CO_2-saturated solution effectively restores fouled membrane flux. Maximization of CO_2 saturation is achieved by lowering the solution temperature and elevating the pressure. Dissolved CO_2 exerts a simultaneous physical and chemical cleaning effect: chemically, it generates a localized acidic environment upon dissolution; physically, the rapid depressurization during the cleaning phase triggers the nucleation of CO_2 bubbles, which enhance shear forces and scour foulants from the membrane surface. Qi et al. [39] demonstrated via constant flux filtration experiments that Supersaturated CO_2-Enhanced Backwashing (SCEB) restores transmembrane pressure more effectively than deionized water backwashing, achieving energy savings of 6.5%, 2.7%, and 6.9% for ceramic membranes treating lake water, activated sludge, and slaughterhouse wastewater, respectively. Another study utilized supercritical carbon dioxide (sCO_2) to clean reverse osmosis membranes. Results indicated that cleaning efficiency for various foulants stabilized at 82%–87%; notably, for sodium alginate (SA) fouling, sCO_2 cleaning efficiency reached 89.7%, markedly surpassing deionized water rinsing (12.2%), pH 5 solution (6.2%), liquid CO_2 (9.3%), and gaseous CO_2 (7.2%) [40]. Consequently, backwashing with CO_2-saturated solutions represents a promising alternative to conventional methods, effectively extending membrane service life while reducing operational costs.

In summary, CEB typically yields a flux recovery rate (FRR) of 80%–95%. For the remediation of organic or biological fouling, sodium hypochlorite (NaClO) is generally dosed at concentrations of 200–500 mg/L (as available chlorine). For inorganic scaling, hydrochloric acid (HCl), sulfuric acid (H_2SO_4), or citric acid is utilized, with the pH adjusted to 2–3. For the removal of oil fouling or organic matrices, alkaline agents such as sodium hydroxide (NaOH) are employed. Following reagent injection into the membrane module, a static soak phase of 10–30 minutes is mandated to allow for complete reaction between the agents and the foulants.

4.3.2. Ultrasonic-Assisted Chemical Cleaning

Ultrasonic (US)-assisted chemical cleaning is defined as a synergistic remediation protocol wherein an ultrasonic field is integrated with chemical agents—such as acids, alkalis, oxidants, or chelators—to execute cyclic rinsing or static immersion of the membrane module. The fundamental mechanism is predicated on the micro-jets and shockwaves generated by ultrasonic cavitation. These physical forces actively intensify mass transfer within the boundary layer, accelerate reagent penetration into fouling layers and membrane pores, and disrupt the structural integrity of cake or gel formations. By weakening foulant adhesion, this process facilitates more comprehensive chemical reactions. Consequently, this hybrid approach typically outperforms standalone chemical cleaning, yielding superior flux recovery rates and reduced cleaning durations; it proves particularly efficacious against organic fouling and biofilms. Operational parameters—specifically frequency, power density, temperature, and duration—require rigorous control to mitigate the risk of membrane damage arising from excessive acoustic intensity. The scope of application is predominantly focused on ceramic membranes, metallic substrates, and specific reinforced polymeric membranes engineered to withstand mechanical vibrations, making it a frequent choice for the remediation of irreversible fouling [19].

Empirical studies underscore the advantages of this hybrid method. Popovic et al. [41] reported that a 30-minute treatment combining 35 kHz ultrasound with chemical agents achieved a flux recovery rate (FRR) of 87% ± 3% for 200 nm ceramic membranes; a subsequent ultrasonic cycle further elevated the recovery to 96.3% ± 0.4%. Similarly, Jin et al. [42] demonstrated that coupling a 2 g/L citric acid solution with ultrasound restored 81% of the initial flux for PVDF hollow fiber ultrafiltration membranes. In stark contrast, a 7-hour static soak in citric acid yielded only 73.3% recovery, while standalone ultrasonic treatment achieved a mere 56.2%. Furthermore, Thombre et al. [43] observed that a brief 4-minute cleaning cycle using 1.0 M NaOH combined with ultrasound restored 90% of the permeate flux, a performance that significantly surpasses that of either ultrasonic or chemical cleaning in isolation.

In summary, US-assisted chemical cleaning typically attains exceptional flux recovery rates, ranging from 90% to over 99%. Standard operational protocols involve maintaining a solution temperature of 30–50°C and incorporating appropriate chemical agents (e.g., 0.2%–2% NaOH, HCl, EDTA, or surfactants). Low-frequency ultrasound (20–45 kHz) is employed to maximize penetration depth, while power density is strictly regulated between 0.1–0.5 W/cm², with cleaning durations generally strictly limited to 10–30 minutes.

4.3.3. Micro-Nano Bubble Assisted Chemical Cleaning

Micro-Nano Bubble (MNB) assisted chemical cleaning is defined as an advanced technique wherein a specialized generator produces a proliferation of micro- and nano-scale bubbles, which are subsequently integrated into a chemical solution to rinse the membrane. The underlying mechanism exploits the synergy between chemical reagents and the unique physicochemical properties of the bubbles. Specifically, phenomena such as surface charge adsorption, the localized generation of extreme temperature and pressure upon bubble collapse, and radical-mediated oxidation actively strip both loose and dense fouling layers while accelerating the degradation of organic matrices. Consequently, this method significantly enhances the removal rates of refractory organic fouling and concurrently reduces the requisite dosage of chemical agents. This technology is deemed particularly suitable for polymer membrane systems susceptible to damage and is regarded as a promising strategy for the remediation of biofilms and gel layers.

Research highlights the efficacy of this approach in mitigating material degradation. In a study addressing PVDF ultrafiltration membranes fouled by anionic polyacrylamide, Lu et al. [44] determined that while a 0.1% (v/v) concentration was optimal for standalone sodium hypochlorite (NaClO) cleaning, it induced significant material erosion and performance decay. However, coupling this agent with air Micro-Nano Bubbles (MNBs) enabled a 70% reduction in NaClO dosage without compromising cleaning efficiency, thereby effectively retarding membrane aging. Furthermore, Liu et al. [45] evaluated the performance of ozone-coupled micro-nano bubbles (O_3-MNB) against traditional tap water backwashing, MNB backwashing, and ozone water backwashing. The results demonstrated that the irreversible fouling resistance associated with O_3-MNB backwashing was markedly lower—constituting only 4.8%, 10.0%, and 23.3% of the resistance observed in tap water, MNB, and ozone water protocols, respectively. Additionally, the O_3-MNB treatment effectively eradicated viable bacterial cells and eliminated the majority of protein and polysaccharide foulants from the ceramic membrane surface.

In summary, micro-nano bubble assisted chemical cleaning typically yields exceptional flux recovery rates, ranging from 90% to 98%. Standard operational protocols entail the generation of high-density micro-bubbles with diameters spanning from hundreds of nanometers to tens of micrometers. These bubbles are combined with low-concentration chemical agents, such as 0.1%–0.5% acid, alkali, or sodium hypochlorite; alternatively, the utilization of ozone micro-bubbles provides enhanced oxidative potential. The cleaning process is typically executed via circulation or immersion at ambient to mild temperatures (25–40°C) for a duration of 30 to 60 minutes.

4.3.4. Electrochemical Assisted Cleaning

Electrochemical Assisted Membrane Cleaning (EAMC) is defined as a specialized technique wherein an electrode system is integrated into the membrane module, utilizing an applied electric field in conjunction with a liquid medium to remediate fouled membranes. Specifically, the application of an electric field amplifies the surface charge density, thereby inducing electrostatic repulsion between the membrane and foulants of identical charge. This repulsion effectively mitigates the deposition of contaminants—such as dyes, water-soluble natural organic matter (NOM), and particulate matter—onto the membrane surface [10].

Several studies demonstrate the efficacy of this mechanism. Sun et al. [46] augmented the negative surface charge density of a graphene hydrogel membrane (GHM) via an external electric field. By exploiting electrostatic repulsion, they successfully detached negatively charged bovine serum albumin (BSA), achieving a maximum flux recovery rate of 99.0%. Similarly, subsequent research [47] revealed that after a 100-minute treatment, applying a voltage of 3V reduced transmembrane pressure by 33.7%, while an increase to 5V yielded a 51.1% reduction. Fan et al. [48] employed a positive potential on CNTs/Al_2O_3 membranes to inhibit fouling by positively charged silica particles. Their results indicated that a 1.5V potential enhanced the flux and removal efficiency of latex particles, phenol, and NOM by factors of 1.6, 4, 5.7, and 3, respectively, compared to control conditions without an electric field. Furthermore, backwashing coupled with an electric field completely restored membrane flux. However, inherent limitations persist: the Joule heat generated by the electric field may compromise membrane integrity or catalyze the decomposition of chlorine-containing compounds into disinfection by-products (DBPs). To ameliorate these drawbacks, the implementation of alternating current (AC) in lieu of direct current (DC) has been proposed [49, 50].

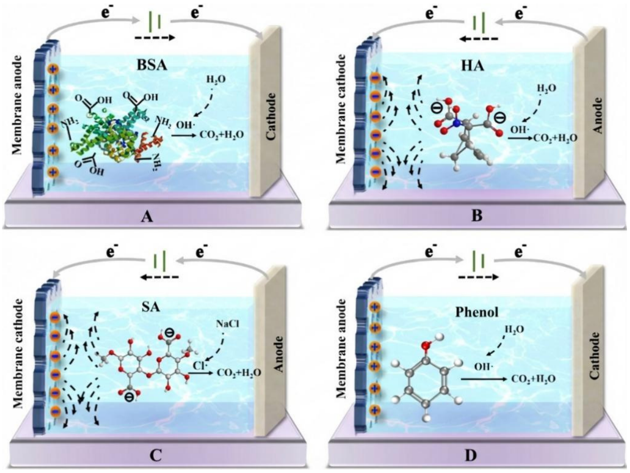

Fig 3. Mechanism of organic fouling removal: (A) BSA removal by indirect oxidation, (B) HA removal by the combined effects of electrostatic repulsion and indirect oxidation, (C) SA removal by the combined effects of electrostatic repulsion and indirect oxidation, (D) phenol removal by indirect oxidation.

Beyond electrostatic repulsion, electrochemical oxidation serves as a prevalent mechanism for membrane cleaning, categorized into direct and indirect oxidation pathways. Direct oxidation entails the initial adsorption of foulant molecules onto the membrane surface, followed by electron transfer processes wherein the molecules lose electrons. These foulants subsequently undergo oxidative degradation, decomposing into CO_2, H_2O, and other low-molecular-weight byproducts. Vecitis et al. [51] utilized multi-walled carbon nanotube (MWNT) filters as anodic materials to degrade methylene blue (MB) and methyl orange (MO) via direct oxidation. Under a voltage of 2–3V, over 90% of the MB and MO were eliminated within a residence time of 1.2 seconds.

Conversely, indirect oxidation generates highly reactive radicals (e.g., $\cdot OH$, $\cdot Cl$, and $SO_4^{\cdot -}$) through the electrolysis of H_2O, Cl^-, and SO_4^{2-} present in the solution. These reactive species subsequently oxidize surface foulants, producing mineralized products that are removed via membrane separation (see Figure 3 for the schematic mechanism [10]). The synergy between indirect oxidation and membrane filtration significantly amplifies foulant removal, ensuring high-efficiency cleaning.

Electrochemical Advanced Oxidation Processes (EAOPs) represent a quintessential implementation of indirect oxidation. This technology generates radicals *in situ* to degrade foulants without necessitating external chemical additives, rendering it more environmentally benign than traditional Advanced Oxidation Processes (AOPs). The Electro-Fenton (EF) process exemplifies a specific EAOP mechanism wherein H_2O_2 is generated *in situ* via the two-electron oxygen reduction reaction (ORR) at the cathode. This approach obviates the safety risks associated with the transport and storage of H_2O_2 inherent to traditional Fenton processes [52]. Jiang et al. [53] employed the EF process to degrade the antibiotic florfenicol via *in situ* oxidation by hydroxyl radicals ($\cdot OH$). Following continuous dehalogenation, defluorination, and desulfurization, the antibiotic was mineralized into CO_2 and inorganic ions (F^-, Cl^-, SO_4^{2-}), effectively alleviating antibiotic-induced membrane fouling.

In summary, distinct from the chemical cleaning methods previously detailed, EAMC exploits electrons as the primary reaction reagent to eliminate foulants. This versatile technology is not constrained by specific membrane configurations or materials and is applicable across various separation processes, including ultrafiltration, nanofiltration, and reverse osmosis, utilizing either inorganic or organic polymer membranes. EAMC offers distinct advantages, including superior cleaning efficiency (with flux recovery rates reaching 90%–99%), minimal chemical consumption, the elimination of secondary pollution, and high automation potential. These attributes have garnered increasing scholarly and industrial attention for EAMC in recent years.

Each cleaning method is presented in Table 3.

Table 3. Chemical membrane cleaning strategies and efficiencies

Cleaning Strategy	**Membrane**	**Foulants**	**Key Operating Conditions**	**Flux Recovery (%)**	**Ref.**
Alkaline Cleaning	Tubular ceramic membrane (0.1 μm)	Whey protein	1.0 wt% NaOH, 30 min	80	[54]
Acid Cleaning	PVDF (0.01 μm)	NOM	Steam (90∘extC90∘*extC*), 1 M HCl, 30 min	83	[55]
Advanced Oxidation	PES (MWCO 30 kDa)	HA	SPC-MnO_2 effervescent tablet, 5 min	95	[34]
	PES (MWCO 30 kDa); PVDF (MWCO 100 kDa)	HA	0.5 wt% H_2O_2 solution with MnO_2 particles, 5 min	95	[56]
	PES (MWCO 100 kDa); PVDF (MWCO 50 kDa)	HA	Co_3O_4-NR (1.6 g/L) and PMS (2 mM), 15 min	98	[57]
	PES/PVDF (MWCO 100 kDa)	HA and Ca^{2+}-complexed organic fouling	SPC, CA, and Fe (II), 1 h	99	[58]
	PES (MWCO 100 kDa)	HA, BSA, SA	PMS / NH_2OH / Fe(II), 1 h	96	[35]
	Al_2O_3 ceramic membrane (0.05 μm)	HA, BSA, SA	Permanganate (PM) / PMS, 6 min	96	[36]
	PES (MWCO 50 kDa)	HA, Ca^{2+}	PMS (10 mM) / Cl^- (15 mM) solution, 1 h	94	[59]
Chemically Enhanced Backwashing	Ceramic membrane (0.1 μm)	Lake water, activated sludge, wastewater	Backwash enhanced by CO_2-supersaturated water, 5 min	90	[39]
	Polyamide RO membrane	SA	50 mL/min supercritical CO_2 (sCO_2), 30 min	90	[40]
Micro-Nano Bubbles	PVDF flat-sheet (0.02 μm)	400 mg/L Anionic polyacrylamide	0.03% NaClO-AMNBs, 120 min	97	[44]
	α*α*-Al_2O_3 ceramic membrane (0.1 μm)	WTP carbon filter backwash water	O_3-MNBs, 1 min	97	[45]
Electrochemical Assisted	Conductive GHM (0.1 μm)	BSA	*In situ* electro-oxidation (GHM cathode), bias voltage -1 V	99	[46]

5. Conclusion

(1) Physical cleaning is regarded as the foundational approach for managing reversible membrane fouling. This method offers distinct advantages, including operational simplicity, the elimination of chemical additives, reduced costs, and minimal risk of membrane material degradation. Techniques such as hydraulic backwashing, back-pulsing, and ultrasonic cleaning effectively detach loose cake layers adhering to the membrane surface, thereby restoring a portion of the membrane flux. However, the efficacy of physical cleaning is constrained by its limited penetration depth, rendering it ineffective against irreversible foulants entrapped within membrane pores. Consequently, physical cleaning is typically deployed as a routine maintenance protocol or integrated into hybrid cleaning strategies.

(2) Chemical cleaning constitutes the core strategy for mitigating irreversible membrane fouling. By leveraging chemical agents—such as acidic/alkaline reagents and oxidants—this method effectively decomposes and eliminates recalcitrant foulants located both within the pores and on the surface, generally achieving flux recovery rates exceeding 90%. The incorporation of advanced oxidation processes further amplifies cleaning efficiency and specificity. Nevertheless, the limitations of chemical cleaning are substantial: the operational procedures are relatively complex, and operating costs are elevated. Furthermore, exposure to agents with strong oxidative properties or extreme pH levels may induce aging in polymeric membrane materials, compromise hydrophilicity, and potentially trigger the release of microplastics.

(3) Combined physical-chemical methods and emerging technologies represent a pivotal trajectory in the advancement of membrane cleaning. Chemically Enhanced Backwashing (CEB) synergizes hydraulic shear with chemical reactivity, significantly prolonging the intervals between intensive deep cleanings. Similarly, Micro-Nano Bubble (MNB) assisted chemical cleaning sustains exceptional flux recovery rates while substantially curtailing chemical dosage. Additionally, Electrochemical Assisted Membrane Cleaning (EAMC), which exploits electrons as the primary reaction reagent, offers distinct benefits such as superior cleaning efficiency, minimal chemical consumption, the elimination of secondary pollution, and a high potential for automation.

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