Organic Fouling

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Organic fouling in the forward osmosis process is complex and influenced by different parameters in the forward osmosis such as type of feed and draw solution, operating conditions and type of membrane. In this article, we reviewed organic fouling in the forward osmosis focussing on wastewater treatment applications. Model organic foulants used in the forward osmosis literature were highlighted, followed by the characteristics of organic foulants when real wastewater are used as feed solutions. The present study evaluated various physical and chemical cleaning protocols for organic fouled membrane and the efficiency of cleaning methods for the removal of organic fouling in the forward osmosis process. The study made recommendations on future cleaning technologies such as Ultraviolet and Ultrasound. Generally, a combination of physical and chemical cleaning is the best for restoring the water flux in the FO process.

Keywords: Forward Osmosis ; Organic Fouling ; Wastewater

1. Cleaning Protocols for Fouled FO Membranes

During the FO process, the membrane surface gets fouled, which causes a decline in the permeability and selectivity of the membrane. Several cleaning methods were suggested and developed by researchers to restore the membrane water flux. However, the cleaning protocols should not damage the membrane and must be practical and less time consuming with an ease of installation. Moreover, the cleaning solutions and cleaning cycles are one of the critical factors to decide the membrane life.

1.1. Physical Cleaning

The initial step in the physical cleaning involves the unwinding and redissolution of the foulant layer into the feed stream. Direct flushing at high cross-flow velocity is applied to remove firmly attached foulants. Loosely bounded foulants can be easily removed by flushing the membrane module with water. For plate and flat sheet modules, backflushing can be applied to reduce the fouling within pores and to eliminate the cake layer or gel layer formation on the active site of the membrane. In contrast, the non-homogeneous flow through the pores restricts its applicability. Recently, researchers reported the energy cost and an ultra-sonication cleaning to remove foulants, but the main challenges are scaling up. Air scouring is utilized as a cleaning strategy to control the membrane fouling effect and to prevent any complicated reaction before it occurred [1].

The cake layer formed in the FO and RO processes is very different. Compressible foulants in RO formed a dense cake layer, which can reduce flux and increase the energy due to increasing the hydraulic resistance along the membrane. On the other hand, the flux decline in the FO was due to cake formation on the membrane surface because of the enhanced osmotic pressure [2]. The fouling in FO was reversible and not compressed when compared to the RO mode, which involved the hydraulic pressure, and may affect the morphology of the alginate gel. As a result, cleaning the FO membrane was easier than the RO mode due to lower hydraulic compaction of the foulant layer [3]. Many factors can affect the efficiency of membrane cleanings such as type of feed water, membrane type, duration frequency, and intensity [4]. Surface flushing is an effective method for cleaning the reversible foulant on the FO membrane surface. The membrane can simply obtain cleaning efficiency by around 98% with increased cross-flow velocity and cleaning duration. Additionally, air bubbles can be added to enhance the cleaning efficiency due to increasing shear force on the membrane and the turbulence, and reducing the time of cleaning. Hence, this may achieve 100% efficiency without any chemical reagents [5]. However, this method is insufficient for the RO membrane due to the porous factor of the support layer, which can decline the shear force and reduce the cleaning efficiency [6]. Osmotic backwashing is another physical cleaning method that can be used for the FO membrane. The osmotic backwash is implemented by reducing the salinity of the draw solution and increasing the feed solution salinity or swapping the feed with the draw solution inlet to break down the foulant on the support layer [7]. Some studies recommended osmotic backwash as an effective cleaning method, especially with membrane fouled by complex water such as anaerobic digester concentrate, MBR feed water, industrial wastewater, and municipal wastewater [8][9][10][11][12].

In contrast, other studies reported low flux recovery obtained when using osmotic backwashing for membrane cleaning [6][13]. Some foulants may form gypsum in the support layer and required additional hydraulic backwash to obtain 100% flux recovery, especially in the RO mode due to a compressed foulant. In case of using the cellulose acetate (CA) membrane for both FO and RO, apply 4 M NaCl Draw solution for FO and, under 28 bars, hydraulic pressure for RO for 20 h until the flux notably reduced. The cleaning process by rinsing the foulant membrane with 50 mM NaCl solution for 15 min found that the flux on the FO membrane recovered by around 100%, while, for RO, was around 70% [3]. Therefore, osmotic backwashing can be used for the FO mode due to the lower compaction of foulant on the membrane while, in case of the RO, the suitable method for physical cleaning is hydraulic backwashing in case of cleaning without chemical reagents.

1.2. Chemical Cleaning

Since complete removal of foulants is not possible, using physical cleaning, various chemicals are used to break down the chemical structure and bonding of foulants with the membrane. Chemicals used in cleaning reacts with foulants to enhance their solubility by the degradation of foulants in a more soluble form. Surfactants and wetting agents are also used as cleaning chemicals since they can easily remove the foulants inside the pore. The sanitization operation is performed to avoid the growth of microorganisms on the membrane surface. In some studies, hypochlorite is applied as a disinfectant agent. Mass transfer of foulants and chemicals to and from the membrane decides the efficiency of cleaning. Circulation time and flow rate at various pH of cleaning with chemicals is a critical factor in obtaining highly efficient cleaning. In general acids, bases, steam, gas sterilization, and sequestrants are used as cleaning agents. Acid solution mainly reduces efficiently inorganic fouling while the whole alkali solution is responsible for removing organic fouling [14].

Information about the main components of the water source and fouling composition helps to obtain better cleaning in a short amount of time while considering safety, cost, and the impact of the cleaning agent to the environment and membrane [15]. Gao, Liang et al., 2011, reported that sometimes two agents need to be mixed to reduce foulant from the surface of one membrane [1]. Tragardh (1989) stated that chemical cleaning should follow some steps to avoid damaging the system and sustainable working such as keeping the fouling loose and solute to avert forming new fouling and keep the membrane clean and safe with all other wetted surfaces sterilized. Additionally, the efficiency of the chemical cleaning depends on the flow rate of the cleaning, concentration, and temperature of cleaning material and the percentage of the foulant [16]. The positive effect of the interaction between the foulant and the chemical agent can be presented in three ways: (i) the foulant can be removed, (ii) the morphology of the foulant can be changed such as swelling or compacting, and (iii) the hydrophobic or charged is modified because the surface chemistry of the deposit may be altered [17]. This cleaning agent may react chemically with foulant so they can break down the cohesion force between the foulant ions and reduce the adhesion force between the foulants and the membrane surface.

In the case of the FO membrane, thin-film composite (TFC) required chemical cleaning due to strong adhesion with alginate when compared to the cellulose triacetate (CTA) membrane [3]. Sodium hydroxide (NaOH), sodium ethylenediaminetetraacetic acid (EDTA), and sodium hypochlorite (NaOCl) with a concentration range between 0.5–1.0%. Several studies reported that chemical cleaning could achieve highly efficient membrane cleaning from organic foulant, which may have a strong interaction to the membrane surface [8][18][19][20][21]. Although chemical cleaning has a high cleaning efficiency, hardly removed foulant from membrane pores [8], can be harmful to the FO membrane, may reduce the membrane life [13], and might cause an environmental problem due to the effluent stream during the cleaning process [3][22]. For an example of RO mode, cleaning reagents chosen for an organic foulant similar to FO as NaOH as an alkaline solution, certified grade disodium ethylenediaminetetraacetate (Na2-EDTA) as a metal chelating reagent, certified grade sodium dodecyl sulphate (SDS) as an anionic surfactant, and NaCl as a salt cleaning solution at pH 11 [23]. Thin-film composite LFC-1 as an RO membrane was used, and the foulants within the irreversible fouling layer on the membrane surface, which may have a strong intermolecular adhesion force between particles from wastewater effluent [24]. The cleaning process starts by adding the chemical reagent as a feed solution for a suitable duration time, which may also affect the cleaning efficiency. Some flounts required one chemical cleaning reagent to obtain a high cleaning efficiency, while others need to combine more than one at the same time. Generally, Ang, Yip et al. (2011) stated that adding NaOH as a cleaning reagent (alone or combined with another reagent) can improve the cleaning performance due to its capability to loosen the fouling layer.

1.3. Physio-Chemical Cleaning

Some foulants cannot be effectively removed by physical cleaning only, so are required to combine a chemical cleaning process to recover membrane permeability [25]. This method utilizes physical cleaning methods gathering some chemical agents to improve the capability of the cleaning process. Some researchers use the backflush water method by adding a small amount of the chemical agent to the water. This procedure can enhance the cleaning efficiency [26]. This method keeps improving, and currently, they use a developed method for a mechanical-chemical cleaning process with ultra-sonication-enhanced chemical cleaning [27]. Adding low frequencies of the ultrasound to a chemical cleaning agent like ethylene diamine tetra-acetic acid (EDTA) will enhance the cleaning process more than using the chemical or ultrasound separately [28].

1.4. Biological/Biochemical Cleaning

A biological foulant can attach reversibly or irreversibly to the membrane surface due to the interaction between the biomass and the membrane surface. The interaction between colloidal particles and bacteria cells blocks the membrane pores. Some chemical compounds such as hydrogen peroxide (H_2O_2) can harm the membrane and may cause fouling with a negative effect on the microbial community. If there is any bio-fouling formed on the membrane, using one of the biological cleaning methods with different chemical agents such as biocide solution can minimize the effect of the bio-fouling [29] or using mixtures of cleaning contains bioactive agents like enzymes or single molecules to promote the foulants extract [30]. Using an environmentally-friendly and mild cleaning agent like purified enzymes and surfactants to avoid damage or corrosion to the membrane surface during the biological cleaning process is advised [31]. For example, some researchers used a new enzymatic protocol for biological cleaning to UF fouled membrane used for abattoir effluent and obtain a 100% flux recovery [32]. Although two more strategies can be used to control osmotically-driven membrane processes (ODMPs), membrane fouling such as energy uncoupling and quorum quenching [33], enzymatic cleaning is the most common biological method for FO and RO membrane cleaning [26].

1.5. Factors Influencing Cleaning Efficiency

Many factors impact the cleaning efficiency of the fouled membrane, such as the sequence of the cleaning steps. Practically, physical cleaning can remove loose particles on the membrane surface (reversible fouling). Generally, physical cleaning cannot retrieve membrane permeability effectively, and a combination of chemical and physical cleanings are usually used for the removal of such fouling [25][34]. In addition, the sequence in the chemical cleaning may affect the membrane permeability degree. Some studies reported that an alkaline cleaning substance could be more efficient when followed by an acid cleaning substance better than when cleaning the membrane from dominant organic matter foulants [35][36]. Other studies found that precipitation of an inorganic scaling or metal hydroxide on the membrane surface prefers acid than the alkali substance sequence for the cleaning process [14][37].

Temperature is considered another factor that may take effect on the membrane cleaning strategy. Increasing temperature is substantial for cleaning the fouling membrane by increasing solubility due to reactivity of functional groups at high temperatures of the organic matters and increasing mass transfer dispersive with mechanical destabilization of biofilm layers on the membrane surface [38]. In contrast, high-temperature biofilm tends to precipitate on the membrane surface in the form of calcium carbonate, ferric hydroxide, and silicates, which makes the process more difficult to separate [39].

Increasing the pH has a direct proportion with membrane cleaning efficiency in case all carboxylic functional groups of EDTA are deprotonated [40]. For instance, increasing pH from 4.9–11.0 will affect the cleaning percentage from 25%–44% and, at pH 11, are very easy to break down the gel layer on the membrane surface when compared to the lower pH. While Ang, Lee et al. (2006) found that, in the case of sodium dodecyl sulphate (SDS), the effect of the pH is very low on the cleaning efficiency because the chemical reaction between the foulants and the SDS effect slightly by changing pH [40]. Table 1 shows the cleaning protocols for fouled membranes. Furthermore, Al-Amoudi, A. concluded that permeable efficiency relies on the membrane cleaning step within the membrane process [36].

Table 3. Cleaning protocols for fouled forward osmosis (FO) membranes.

Cleaning Method	Membrane Type	Feed Solution	Draw Solution	Factors/Cleaning Condition	Cleaning Efficiency %	Flux %	Ref.
Mechanical	Vertical hollow fibres	Bentonite	-	8 mm amplitude + 8 Hz frequency vibration compared to no vibration	90	-	[41]
	γ-Al_2O_3 ceramic	Natural water include colloidal silica	-	3.5 cm 2.6 cm 1.7 cm Distance between the ultrasonic source probe and the membrane surface for 1.56 μm particles	100	60 75 97	[42]
Chemical	Hydrophobic Polyvinylidene fluoride (PVDF) membrane	Surface water	-	Oxidants and bases 400 ppm NaClO 0.1 M NaOH	-	150 60 10	[43]
	Cellulose acetate (CA) FO	200 mg/L alginate, 50 mM NaCl, and 0.5 mM Ca^{2+}	(DI) water with 28 bars (400 psi) 4 M NaCl	21 cm/s, 50 mM NaCl solution for 15 min	-	RO = 70 FO = 100	[3]
Combined chemicals	Polyethersul-fone (PES)	Synthetic wastewater	-	NaClO + citric acid recover membrane permeability higher than single-agent cleaning	80%	70–78	[44]

Cleaning Method	Membrane Type	Feed Solution	Draw Solution	Factors/Cleaning Condition	Cleaning Efficiency %	Flux %	Ref.
Mechanical-Chemical	**RO/LFC-1**	**Suwannee River natural organic matter**	**-**	**−ethylene diamine tetraacetic acid (EDTA) 0.5 mM + pH 5.7**	**25**	**-**	[45]
				−EDTA 0.5 mM + pH 11 for 15 min T20 °C	**43**		
				−EDTA 2 mM + pH 11 for 15 min	**100**		
				−EDTA 0.5 mM + pH 11 for 60 min	**85**		
				−EDTA 0.5 mM + pH 11 for 15 min T40 °C	**97**		
				−sodium dodecyl sulfate (SDS) 2 mM + pH 11	**15**	**-**	[45]
				−SDS 2 mM + pH 11 for 15 min	**18**		
				−SDS 2 mM + pH 11 for 60 min	**25**		
				−SDS 10 mM + pH 11 for 15 min	**70**		
				−SDS 10 mM + pH 11 for 60 min	**95**		
	Hollow fibre	**Seeding sludge + synthetic municipal wastewater**	**-**	**0.5 ppm NaClO solution + Backflush 15 min**	**-**	**77**	[46]

Cleaning Method	Membrane Type	Feed Solution	Draw Solution	Factors/Cleaning Condition	Cleaning Efficiency %	Flux %	Ref.
	Polyethersul-fone (PES) flat sheet UF	Extract of Radix astragalus (RA)	-	Ultrasound 20 kHz and 120 W + 0.1 M NaOH	-	80	[27]
	FO cellulose acetate (CA)	Sodium alginate (10 g/L)	-	−NaCl solution at 8.5 cm/s for 24 h	-	96	[3]
				−NaCl solution at 21 cm/s after 15 min of rinsing		98	
Biological	Ultrafiltration (UF) membrane	wastewater treatment plant	-	New enzymatic cleaning protocol (25–30 °C)	100	-	[32]
	Bulgarian UF 60 Polyacrylonitrile (PAN) spiral-wound	Water from the Kamchia dam after its chlorination		Preliminary treatment of the water-chlorination or UV irradiation by low-pressure mercury- vapor lamps at a wavelength of 253.7 nm	80	-	[47]
	Flat-sheet polysulfone (PSf)	Abattoir process	-	Pseudomonas lipase in conjunction with the identified proteases	-	−100	[48]

2. Future Outlook and Conclusions

Water and wastewater treatment using the FO process holds a significant promise for addressing the global water crisis. In general, the biggest challenge for FO commercialization originates from the financial feasibility. However, to maintain the FO process at commercial scale for wastewater treatment, there are critical challenges to overcome, such as fouling problems. An ideal FO membrane must exhibit high pure water flux with a low structural parameter value. One of the most complicated membrane fouling is organic fouling since it affects membrane performance and could result in irreversible fouling. Real-time monitoring of complex formation between organic foulants and membrane materials can help develop strategies to alleviate organic fouling. Several chemical and physical cleaning methods were developed to remove membrane fouling. However, a combination of chemical and physical cleaning methods is more efficient for membrane cleaning. New and innovative cleaning methods such as UV and ultrasound have not been thoroughly covered in the literature, and more experimental work should be done in future studies to investigate the efficiency and cost-effectiveness of non-conventional cleaning methods. Moreover, the research outcomes from various studies involving advanced membrane materials, draw solutions, pre-treatment, and post-treatment should be tested for real-time wastewater applications to facilitate the commercialisation of FO at industrial scale. Pilot-scale FO experiments need to be performed for a long-term operation to understand the formation of irreversible fouling in the FO process.

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