# Retention of natural organic matter by ultrafiltration and the mitigation of membrane fouling through pre-treatment, membrane enhancement, and cleaning - A review

Christian D. Peters[a], Tony Rantissi[b], Vitaly Gitis[b], Nicholas P. Hankins[a]

[a] *Laboratory of Sustainable Water Engineering, Department of Engineering Science, Parks Road, The University of Oxford, OX1 3PJ, United Kingdom*
[b] *Faculty of Engineering Sciences, Ben-Gurion University of the Negev, Beer-Sheva 8410501, Israel*

## Abstract

Natural organic matter (NOM) is a common occurrence in all raw waters (e.g., surface waters, groundwater, and wastewater) that are used for freshwater production. NOM is a leading cause of complications for water treatment systems and is ideally entirely removed early in the process. As an early-stage membrane process, ultrafiltration (UF) can remove the majority of NOM constituents. En revanche, NOM severely fouls UF membranes which can drastically impede the UF process performance. In addition, smaller molecular weight NOM fractions can pass the UF system unhindered and hence, may cause further complications downstream. To address both issues, this paper critically reviews recent publications focusing on enhancing NOM retention and on mitigation of membrane fouling in UF systems. Specifically, the main mitigation routes that are reviewed are UF pre-treatment, in-situ physical and/or chemical cleaning, and membrane enhancement. While several membrane enhancement solutions are promising, the scalability and economic feasibility of these modifications remain questionable. In terms of membrane cleaning, physical solutions are less effective while chemical approaches are more efficient but may cause membrane damage. Of the proposed techniques, multi-process solutions (i.e., combined pre-treatment and UF) are the most promising for NOM retention and prevention of UF membrane fouling. However, optimal operation of multi-process systems is challenging, especially due to the spatio-temporal variability of NOM. Hence, specific focus is given to in-situ, on-line NOM fouling monitoring techniques that can be integrated for dynamic plant operation.

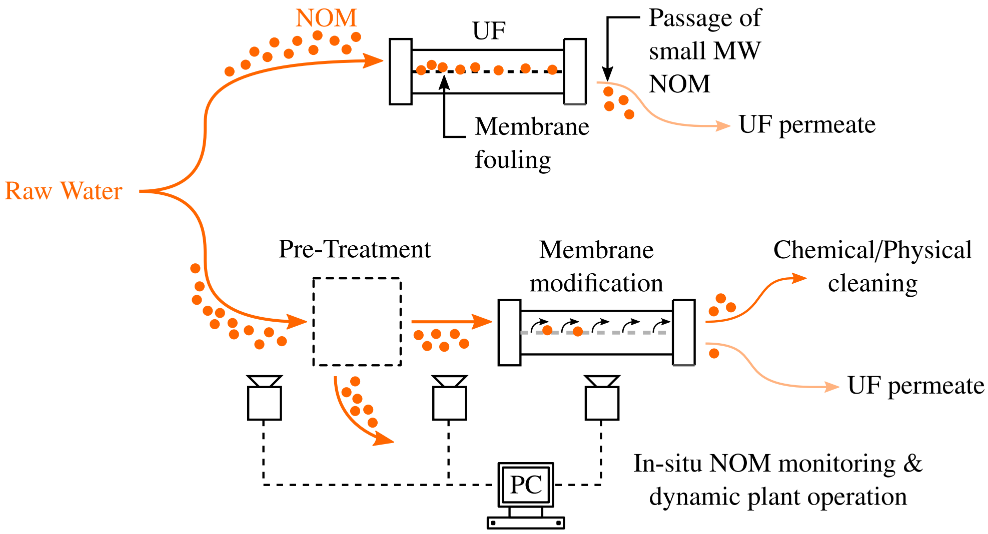
*Keywords:* Ultrafiltration, Natural organic matter, Membrane fouling mitigation, Pre-treatment, Membrane cleaning, Membrane modification

## 1. Introduction

Ultrafiltration (UF) is widely integrated into potable water production, the treatment and reuse of municipal and industrial wastewater, and the desalination of seawater and brackish water [1]. Its popularity can be attributed to its 1) ability to meet stringent water quality regulations by effectively removing pathogens and impurities at low operating pressures of 1-2 bar [2, 3], 2) modularity, compact size and fully automatic operability [4], 3) reliability to produce a high-quality permeate even when subject to fluctuating feedwater quality [5], and its 4) reasonable operational and capital cost when compared to other conventional water treatment processes [6, 7].

Conventional water treatment plants often include a multitude of processes, such as media filtration, coagulation, flocculation, flotation, and adsorption [8, 9]. UF can contribute to the simplification of the water treatment process by replacing several process steps due to its above-mentioned advantages. Furthermore, UF has a wide application range that can vary from treating industrial effluents to concentrating process streams in the food & beverage and in the pharmaceutical industry. All in all, the UF market is rapidly growing and is estimated to hit 2.1 billion USD in 2023 [10].

However, the main drawback of UF technology is its propensity for membrane fouling. Fouling is the narrowing or clogging of membrane pores or forming a cake layer on the membrane surface, all caused by the transport and accumulation of colloids, solutes, or flocs. This phenomenon significantly reduces the membrane permeability and leads to a decline in its performance and lifespan. With its complexity and spatio-temporal variability, natural organic matter (NOM) is one of the oldest,



Graphical abstract

well-known, and still persistent foulants [11, 3].

Figure 2 shows the growing research interest in UF. In 2020 over 1500 publications were published that focus specifically on UF for aqueous applications. Publications on UF membrane fouling contributed to more than 35% while membrane fouling caused specifically by NOM accounted for more than 10% of all publications. The last major reviews focusing on UF membrane fouling and its mitigation date back to 2014 [12, 13] and 2011 [7, 14]. As shown in the figure, the research interest in UF and membrane fouling has significantly grown since then and hence, a critical review and evaluation of the recent progress in this field is needed. This review article specifically surveys recent publications on the causes and mitigation of UF membrane fouling by NOM and ways to improve NOM rejection, as a detailed review is lacking in this research area. Other recent reviews of interest to the reader have focused on 1) NOM removal via other technologies [11, 15, 16], 2) fouling and its mitigation in specific UF applications [17, 18, 19], 3) NOM characterisation [20], and 4) UF fouling mitigation via pre-treatment [2, 21].

NOM is a collective term for naturally occurring organic substances present in surface waters (e.g., seawater, rivers, lakes), groundwater, and wastewater. The NOM is either generated through the biological activity of micro-organisms, algae, and aquatic plants present in the water body, or is derived from decaying plants and animal residues introduced into the water body through drainage from external sources [14, 22].

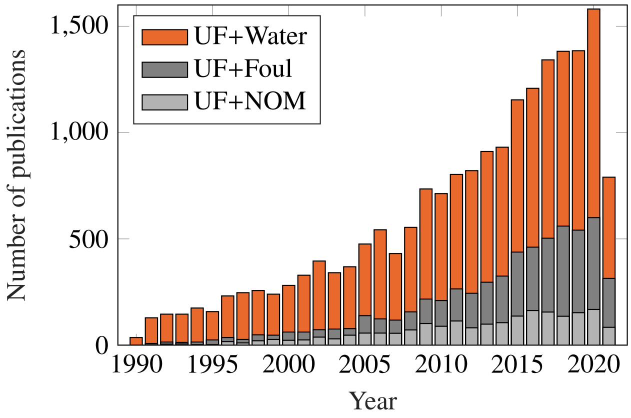


Figure 2: Number of publications per year. Publications in 1) aqueous UF applications ('UF+Water'), 2) Membrane fouling for aqueous UF applications ('UF+Foul'), and 3) NOM membrane fouling for aqueous UF applications ('UF+NOM'). Data is obtained from www.webofscience.com.

NOM is not toxic but its presence in water is undesirable for the following reasons:

- The presence of NOM impairs the organoleptic properties (color, taste and odor) of water [11].
- Various disinfection by-products (DBPs), some of which are carcinogenic, can form when NOM constituents react

| **Abbreviations** | |
|---|---|
| AB | Activated biochar |
| AERs | Anion exchange resins |
| AS | Activated sludge |
| BSA | Bovine serum albumin |
| CA | Cellulose acetate |
| CEB | Chemically enhanced backwash |
| CNCs | Cellulose nanocrystals |
| CNFs | Carbon nanofibers |
| CNTs | Carbon nanotubes |
| DBPs | Disinfection by-products |
| DBP-FP | DBP formation potential |
| DOC | Dissolved organic carbon |
| DOM | Dissolved organic matter |
| EC | Electrocoagulation |
| EEM | Excitation emission matrix |
| EO | Electrocatalytic oxidation |
| FA | Fulvic Acid |
| FH | Ferrihydrite |
| FIX | Fluidized ion exchange |
| GAC | Granular activated carbon |
| GO | Graphene oxide |
| HA | Humic acid |
| HAOPs | Heated aluminium oxide particles |
| HIOPS | Heated iron oxide particles |
| IEP | Isoelectric point |
| IEX | Ion exchange |
| LGC | Lignocellulose |
| MIEX | Magnetic ion-exchange |
| MMMs | Mixed matrix membranes |
| MOFs | Metal-organic frameworks |
| MW | Molecular weight |
| MWCO | Molecular weight cut-off |
| NF | Nanofiltration |
| NOM | Natural organic matter |
| NPs | Nanoparticles |
| OCT | Optical coherence tomography |
| OMCs | Ordered mesoporous carbons |
| PA | Polyamide |
| PAC | Powdered activated carbon |
| PACl | Polyalumimum chloride |
| PAN | Polyacrylonitrile |
| PDMDAAC | Poly dimethyl diallyl ammonium chloride |
| PES | Polyethersulfone |
| PMS | Peroxymonosulfate |
| PNAGA | Poly (N-acryloyl glycinamide) |
| PS | Persulfate |
| PVDF | Polyvinyldifluoride |
| RO | Reverse osmosis |
| SA | Sodium alginate |
| SIX | Suspended ion exchange |
| SPF | Solid-phase fluorescence |
| SUVA | Specific UV absorbance |
| TFC | Thin-film composite |
| UF | Ultrafiltration |
| US | Ultrasound |
| UV | Ultraviolet |
| $UV_{254}$ | Ultraviolet at 254nm |
| UTDR | Ultrasonic time-domain reflectometry |

with disinfectants such as chlorine utilized in water treatment [23, 24, 25].

- NOM enhances the bioavailability, toxicity, and transport of pesticides, radionuclides, and other organic and inorganic pollutants in the source water [26, 27].
- Bioavailability, toxicity, and transport of heavy metals is increased in strong organometallic complexes formed with NOM components such as humic acids (HAs) and fulvic acids (FAs) [28, 29, 30].
- NOM can act as a carbon source for micro-organisms and accelerate their growth in the water distribution network [31, 32]. The removal of NOM from the feed stream is seen as one of the most straightforward and most efficient control measures to minimize biofouling during water distribution [33].

Ultrafiltration is not the only technology that struggles because of the presence of NOM. Depending on the chosen water treatment process, NOM can: 1) increase the demand for chemicals in coagulation [11] and oxidation [34], 2) reduce the regeneration efficiency of ion exchange resins [15] and adsorbents [16], and 3) cause membrane fouling in other membrane processes, such as microfiltration, nanofiltration and reverse osmosis [35, 36, 37, 38, 39].

The retention of low molecular weight (MW) NOM fractions remains difficult with UF membranes, as the majority of these small foulants will pass through the membrane unhindered. A small fraction of the low MW NOM matter can still be adsorbed within the membrane pores, leading to flow obstruction and pore blocking [2]. In contrast, NOM fractions larger than the membrane pores are rejected by UF (e.g., high MW NOM fractions, such as proteins and polysaccharides [14]) and can cause severe membrane fouling.

The paper is structured as shown in figure 3. The first section briefly summarizes the factors affecting UF membrane fouling and some of the novel analytical techniques used to characterize it. Subsequent sections discuss the methods used to enhance NOM retention and control UF membrane fouling through 1) hybrid UF systems that include pre-treatment, 2) UF systems with optimized cleaning and operating procedures, and 3) UF membrane enhancement. The final section discusses current challenges facing UF systems concerning NOM removal, membrane fouling, and its mitigation.

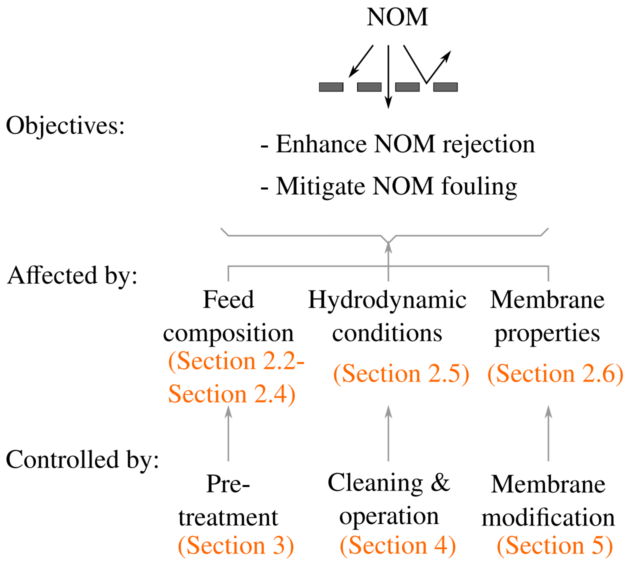


Figure 3: Factors affecting the UF performance and methods to enhance NOM retention, membrane fouling propensity and the UF permeate quality.

## 2. Types, mechanisms, and controlling factors of membrane fouling

### 2.1. *Fouling types and mechanisms*

Size exclusion is the primary mechanism by which UF effectively rejects impurities; this typically includes pathogens, colloids, macromolecules larger than, or close in size to, the membrane pore size. Smaller-sized dissolved substances, such as micropollutants, low molecular weight dissolved organic matter (DOM) and metal ions, are barely retained. Guo et al. [40] reported the UF rejection rates for low MW DOM, such as fulvic acid, citric acid and polysucrose is below 20%. The lowest achievable molecular weight cut-off (MWCO) of tight ultrafiltration membranes typically ranges between 1000 Da to 10000 Da. The size of DOM constituents varies from a few hundred Daltons (low molecular weight acids and neutrals) to over 20000 Da (biopolymers) [41].

While enhancing the UF retention rate improves the product water quality, it may also lead to severe membrane fouling. However, membrane retention and membrane fouling are not necessarily interrelated [41]. Both are dependent on a multitude of factors discussed in section 3 through section 5. It is possible to improve the UF retention rate while maintaining, or even reducing the membrane's fouling propensity.

Fouling is said to occur when the drag force towards the membrane (resulting from the water flux) and the foulant-membrane attraction forces overpower those forces acting away from the membrane (e.g., repulsion forces between membrane and foulant) [42, 43, 44, 45]. Thus, the membrane fouling rate and its reversibility depend principally on the various foulant-membrane and foulant-foulant interactions and may include van der Waals forces, electrostatic interactions, hydration interactions, and hydrogen bonds [46]. Furthermore, as most UF membranes are hydrophobic, it is generally agreed that strong hydrophobic interactions between the membrane and hydrophobic NOM foulants are a principal cause of UF membrane fouling [2, 18].

As organic fouling plays a prominent role in UF membrane fouling, this review focuses on NOM as a principal foulant. In comparison to organic fouling, inorganic fouling is considered less detrimental to UF and only occurs in conjunction with organic fouling [47]. Scale formation is unlikely due to the low retention of scale precursor ions by UF [2]; however, inorganic substances may still contribute to UF membrane fouling. For example, the retention and deposition of organic matter in combination with larger particulate inorganic substances, such as aluminum silicate (a primary component of colloidal clay), manganese dioxide, and ferric hydroxide (oxidation byproducts), on the membrane surface can lead to a substantial flux decline [39, 48]. Furthermore, fouling can be enhanced when metal ions interact with bulk organic matter, which is discussed in greater detail in section 2.3.

Biofouling refers to the growth of deposited microorganisms on the membrane surface. It can be regarded as a biotic form of organic fouling and may cause severe permeability losses during long-term operation [49, 50, 51]. For dense membrane technologies, such as nanofiltration (NF) and reverse osmosis (RO), biofouling is considered to be the most problematic type of fouling to eradicate [33, 52, 53]. However, this is not the case for ultrafiltration processes, as the lack of a transmembrane osmotic pressure gradient, the use of non-spiral-wound membrane modules, and substantially larger membrane pore size, allow for periodical backwashing and surface scouring to help control and mitigate biofouling [50, 41].

As shown in figure 4, UF membrane fouling by organic, inorganic and biological foulants can be categorized into two main mechanisms:

**Pore blockage and adsorption:** Foulants in the size range of the membrane pore, such as high MW biopolymers, may cause complete or partial pore blockage and may result in a severe flux decline [54]. Foulants smaller than the pore size pass through the pores. Adsorption of the foulants to the inner pore surface eventually leads to pore narrowing [55, 56].

**Gel and cake layer formation:** At the following step or as a result of homogeneous flock formation, the foulants of biological, organic, and inorganic origin accumulate on the UF membrane surface, forming a gel/cake layer. The porosity, viscosity, and elasticity of the gel/cake layer affects its flow resistance and ease of removal from the membrane surface [57, 58, 59, 60].

Research findings indicate that both pore narrowing and pore blockage initiate the buildup of a cake/gel layer on the membrane surface [61, 62, 63, 64, 65]. Pore narrowing and pore blockage are considered irreversible and a formation of gel/cake layer primarily reversible [66, 67]. For example, Zhang et al. [68] found that the absorption of low and high MW hydrophilic compounds led to irreversible membrane fouling; due to their hydrophilicity, these fractions are not restricted from entering the membrane pore.

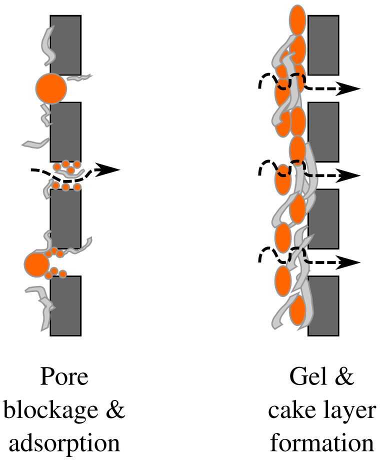


Figure 4: UF fouling mechanisms

## 2.2. *Characteristics of natural organic matter*

NOM is a complex heterogeneous mixture of organic material with molecules that vary in size, molecular weight, hydrophilicity, and charge [2]. A proper characterization might use up to 20 different analytical techniques [69] and characterize just between 1% and 10% of NOM compounds [70]. One of the most straightforward and widely implemented classifications by size divides NOM constituents into particulate and dissolved fractions. The former is filtered out using a 0.45 µm filter [71, 70], whereas the latter passes through practically unattended [61]. A further division of dissolved organic matter into a hydrophobic and hydrophilic fraction is performed with adsorption resins.

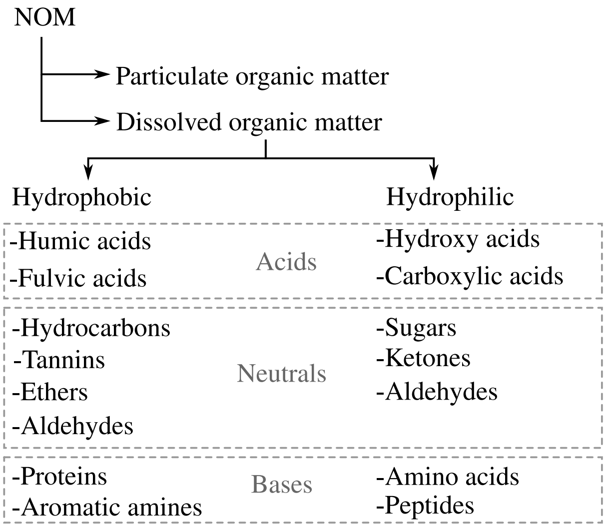


Figure 5: Characterization of natural organic matter [72, 73, 74]. Figure adapted from Levchuk et al. [15] and Bhatnagar and Sillanpää [16].

The considerable variation in NOM components is already evident in the table of typical constituents described in figure 5. For example, simple organic molecules can have molecular weights below 100 Da, whereas biopolymers, such as polysaccharides and proteins, have a molecular weight, between 10000 and 30000 Da [65]. In natural waters, the hydrophobic components are predominant [6, 75], with humic substances generally constituting more than half of the dissolved organic carbon [76]. Humic substances are a collective term for three main groups of organic substances with weak inter-molecular interactions [77, 78, 79]. The 1) fulvic acids with a typical MW between 500 and 2000 Da are water-soluble at any pH, 2) humic acids with MW of roughly 2000-5000 Da are water-soluble at pH>2, and 3) humins that are insoluble at any pH [80, 81]. Humic substances are also considered less biodegradable than biopolymers [82].

According to Li et al. [2], no single NOM fraction has been unanimously identified as the leading cause for UF membrane fouling. Hydrophilic macromolecules (i.e., biopolymers) and organic colloids were identified by Lee et al. [83],Kimura et al. [84], Yamamura et al. [85], Zhang et al. [86], and yu Tian et al. [87] as the major NOM fractions that lead to the loss of permeability. On the other hand, the findings from Yamamura et al. [63], Guo et al. [40], Jacquin et al. [88], Shao et al. [89], and Mu et al. [90] show that an interplay between the various NOM fractions is likely to cause more severe membrane fouling (i.e., a synergistic effect), as fouling caused by one NOM component can initiate or aggravate that of the other. One such example presented by Shao et al. [89] found that the presence of oppositely charged organic foulants, in this case HA and lysozyme, causes the formation of aggregates that are difficult to remove from the membrane surface once deposited. In addition, the findings in Yamamura et al. [63] indicate that hydrophobic, low MW NOM fractions play an essential role in the initiation of membrane fouling, as these are adsorbed within the membrane pores, leading to pore narrowing. Subsequently, larger hydrophilic NOM substances with higher MWs are more likely to be retained by the tightened pores, which inevitably leads to a higher foulant buildup on the membrane surface. Similarly, Mu et al. [90] determined that the deposition of hydrophobic NOM fractions onto the membrane surface alters the membrane's hydrophobicity. Hence, nominally moderate "hydrophobic" fractions tend to agglomerate on the membrane surface and play an essential role in the fouling process. Guo et al. [40] also found that a synergy exists between different dissolved organic substances. An enhanced fouling rate was observed compared to the fouling contribution of the individual compounds.

It can be concluded that the lack of detailed NOM characterization in the source water can affect 1) the membrane's fouling rate and its reversibility, and 2) the product water quality, as for the latter, the types and amounts of DBPs formed during water disinfection strongly depend on the nature of the natural organic matter present in the final product water [23, 91]. Understanding the main NOM characteristics and the associated underlying fouling mechanisms is essential for developing more fouling-resistant filtration approaches.

### 2.3. Presence of metallic ions and inorganic colloids

Membrane fouling and its reversibility depend not only on the NOM fractions present in the UF feedwater; they also depend on the other constituents present, on the membrane properties, and the operational conditions. Although inorganic scaling is unlikely in UF, much research has focused on the synergistic fouling behavior in membrane processes between NOM and metallic ions [92, 93, 94, 95].

Ma et al. [93] found that monovalent ions have no effect on organic membrane fouling, while Gao et al. [96] and Wang and Tang [38] found that an increase in the feed ionic strength (i.e., higher NaCl concentrations) weakened the electrostatic repulsion forces between 1) bovine serum albumin (BSA) and the membrane, and between 2) BSA and itself, which led to a higher membrane fouling tendency. Furthermore, Gao et al. [96] observed that the developed protein fouling layer was eventually dehydrated at higher NaCl concentrations during the long-term process, thus causing a less porous fouling structure with lower permeability. Darowna et al. [97] focused on UF membrane fouling in a photocatalytic membrane reactor using $TiO_2$. Their findings showed that the combination of HA and sodium salts ($NaHCO_3$, $Na_2HPO_4$ and $Na_2SO_4$) in the feed led to an enhanced HA fouling. A high ionic feed strength reduces electrostatic repulsion between HA and the membrane and enhances the fouling. During short-term operations ($\leqslant 10\,h$), both Ding et al. [98] and Gao et al. [96] found that higher $Na^+$ feed concentrations reduced membrane fouling and concluded this was due to an enhanced hydration repulsion force between the membrane and BSA. With an increase in ionic strength, a higher passage of BSA was also observed by Ding et al. [98], which was most likely caused by the reduction of the hydrodynamic radius of BSA due to the compression of the electric double layer [99].

NOM fouling in the presence of divalent ions (such as $Ca^{2+}$ and $Mg^{2+}$) has also been widely studied. Divalent ions can be responsible for 1) forming bridges between various NOM molecules, 2) causing NOM aggregation by charge neutralization, and 3) lowering the electrostatic repulsion between membrane and NOM foulant by decreasing the net negative charge of the NOM molecules [18, 100, 41, 101, 102]. Both points 1) and 2) result in larger, uncharged NOM aggregates [103]. For example, Ding et al. [98] found that larger BSA aggregates were formed in the presence of calcium ions. The membrane easily rejected these larger aggregates. Hence, the predominant fouling mechanism was identified as cake layer formation instead of pore blockage. Similarly, Ma et al. [93] found that HA aggregates were formed in the presence of divalent ions that were well retained by the UF membrane and tended to form a thick cake layer on its surface. A different study by Myat et al. [104] found that BSA and HA could form aggregates on the membrane surface, irrespective of whether calcium ions are present. Still, alginate and HA interactions only occurred in the presence of $Ca^{2+}$.

Other metal ions, such as magnesium ions ($Mg^{2+}$), play a similar role to calcium ions. She et al. [105] demonstrated that $Mg^{2+}$ ions vigorously promote humic acid fouling, whereas calcium ions are responsible for more severe alginate fouling. The observed trend is attributed to the complexation of $Mg^{2+}$ and humic acid, which leads to the reduction of intermolecular repulsion [106]. On the other hand, the intermolecular interaction between $Mg^{2+}$ and alginate is weaker than that of $Ca^{2+}$ and alginate.

Besides metallic ions, inorganic colloids, such as silica and $\alpha$-alumina, were also found to exhibit a synergistic fouling behaviour in conjunction with NOM, causing a less reversible fouling layer and a severe permeability decline [107]. Another study by Ma et al. [108] found that amorphous silica produces a porous fouling layer in combination with BSA due to the strong interaction forces. Still, on the other hand, it forms a compact fouling layer with HA, as the interaction forces are negligible. As hypothesized by Teychene et al. [109] and Li and Elimelech [110], the deposition of particles on the membrane surface may hinder the back diffusion of HA or other NOM fractions away from the membrane surface leading to a synergistic fouling effect between NOM and colloids.

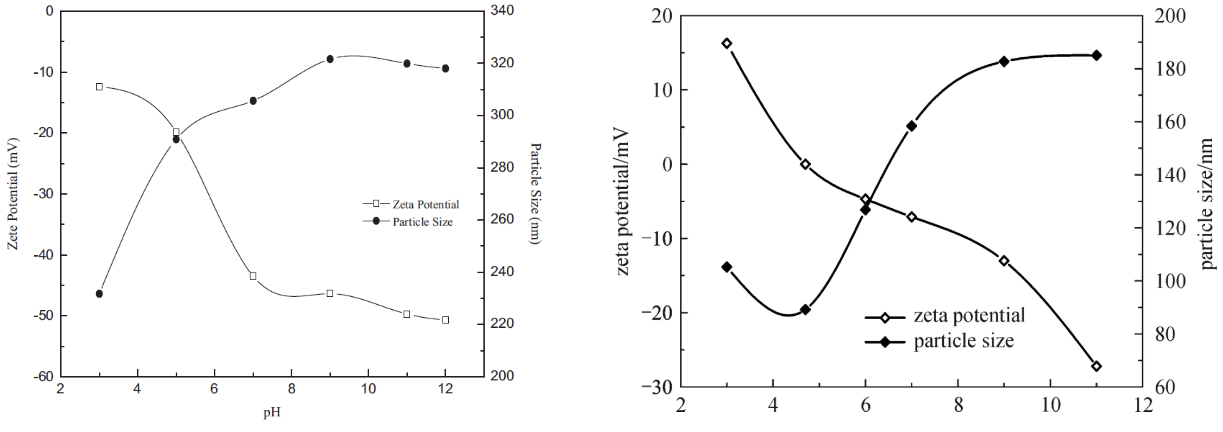
### *2.4. Solution temperature and pH*

The solution temperature is often an uncontrollable spatio-temporal variable that should be considered during UF operation. It directly affects the fluid viscosity and density, affecting the membrane resistance to water transport. Therefore, at higher feed temperatures, a lower fluid viscosity improves the water flux under non-fouling conditions and is thus favorable for filtration performance [111, 112].

Temperature changes between 10C and 30C were found to have a negligible effect on membrane permeability losses caused by the fouling of algae-laden waters using a gravity-driven membrane system [51]. This trend does not necessarily remain at higher feedwater temperatures. Chu et al. [113] found that at higher feed temperatures, membrane fouling is more severe during algae harvesting, as the microalgae excretion rate of extracellular organic matter increases. Furthermore, Ding et al. [98] observed that fouling of a cellulose UF membrane was more severe at higher solution temperatures because 1) the membrane pores expanded at higher temperatures, which reduced their retention and also allowed larger molecules, such as BSA, to be absorbed within the membrane pores, and 2) the BSA molecules unfolded to expose their charged functional groups at higher temperatures leading to a higher adsorption rate of BSA onto the membrane surface. Conversely, van den Brink et al. [114] observed that at low solution temperatures (7C), the particle size of polysaccharides reduces and approaches the membrane pore size. Therefore, lower temperature operation caused fast membrane pore blockage and hence enhanced fouling. Similarly, Alresheedi and Basu [115] found that filtering the same NOM solution with a ceramic membrane at 5C instead of 20C led to an increase in physically and chemically irreversible fouling caused by proteins and polysaccharides; the same components caused only reversible membrane fouling at 20C. However, their findings indicated that fouling caused by humic substances was less influenced by changes in the solution temperature. For dense membranes, Kim et al. [116] found that a higher feed temperature during forward osmosis operation led to a lower fouling rate, as both the solubility of alginate and the organic back-diffusion from the membrane surface increased.

Besides the feed composition and its temperature, the solution pH also affects UF operation and its fouling propensity. As shown in figure 6, the particle size and the zeta potential of HA and BSA, both model organic foulants, are affected by the solution pH. Under neutral and alkaline conditions, the functional groups of humic acid are deprotonated, increasing their electronegativity and hence, stretching their molecular conformation [117, 118]. On the other hand, humic substances tend to curl (i.e., reduce in size) when the solution pH is close to their isoelectric point (IEP), due to the reduction of intra-molecular repulsion [119]. A deviation from IEP that for BSA is at pH=4.7 [38] leads to stronger intra-molecular repulsion and increase in BSA size.

When considering the interaction between the membrane and the foulants, their respective IEPs become essential. For example, Almécija et al. [121] found that under extreme acidic (pH=3) or alkaline (pH=10) conditions, the membrane and the investigated proteins had the same positive or negative charge, respectively. Thus, if the solution pH exceeds both the IEPs of the UF membrane ($pH > IEP_{Mem}$) and the NOM func-



(a)

(b)

Figure 6: The zeta potential and the particle size of (a) HA [117] and (b) BSA [120] with respect to the solution pH. Adapted with permission of Elsevier Science Ltd.

tional groups ($pH > IEP_{NOM}$) (or is below both, such that: $pH < IEP_{Mem}$ and $pH < IEP_{NOM}$), charge repulsion between the membrane and the foulant counteracts foulant adhesion to the membrane. Therefore, operation at neutral or alkaline pH where, in general, $pH > IEP_{Mem}$ and $pH > IEP_{NOM}$ is generally preferred due to the higher retention of the larger sized NOM constituents and the stronger charge repulsion between the negatively charged membrane and NOM components [18].

Various researchers have found that membrane fouling is more severe when the solution pH is below or close to the IEP of the foulant ($pH \leqslant IEP_{NOM}$) but remains above that of the membrane ($pH > IEP_{Mem}$). Zhou et al. [122] and Zhang et al. [123] hypothesized that the membrane and the fouling species are oppositely charged under these conditions, which encouraged the strong adsorption in this case of algal cells onto the membrane surface during the filtration of algal-laden waters. Similar findings were reported by Ding et al. [98] and Wang et al. [120] for the filtration of BSA solutions.

### *2.5. Hydrodynamic conditions*

The existing hydrodynamic conditions within the membrane module can be optimized to mitigate membrane fouling continuously. It is generally agreed that operation 1) below the critical flux [124, 125, 126], 2) at low operating pressures [127, 38, 128] and 3) under high shear-stress conditions [129, 130, 131], minimize fouling.

During the filtration process, foulants are carried towards the membrane surface via hydrodynamic drag forces by the water permeating through the membrane. In contrast, lift forces reduce NOM deposition by continuously removing detached foulants from the membrane surface [132, 133]. High cross-flow velocities, vibration, or gas sparging can be utilized to induce turbulent flow regimes, and high shear stresses in the membrane system to combat membrane fouling. According to Du et al. [134], the shear stresses achieved via cross-flow in low-pressure membrane filtration systems (i.e., microfiltration and UF) are in the range of 0-6 Pa for low viscosity solutions, which are relatively low in comparison to those achievable via rotation of stirred cells or gas sparging. Furthermore, studies by Chan et al. [135] and Gaucher et al. [136] have highlighted that shear stress fluctuations created by gas sparging, for example, can be beneficial for fouling control.

There are, however, instances in which higher shear stresses in the membrane system can cause more severe membrane fouling. For example, at high cross-flow velocities or high aeration rates, microbial cells can be damaged and release extracellular polymeric substances or larger amounts of sludge. Microalgal flocs can breakup into smaller colloids increasing membrane fouling and causing a more significant flux decline [137, 138, 139]. A different issue arising from high shear stresses is that high shear stresses more readily remove larger particles, whereas smaller particles are less affected. This is because the lift force acting on the foulant is proportional to its size cubed [140, 141]. Therefore, this process of size fractionation, with the deposition of smaller foulants on the membrane surface and the removal of larger ones, can lead to the formation of denser and less porous cake layers [142, 143, 144]. Therefore, the optimal cross-flow velocity, or aeration rate, depends strongly on the composition of the feed stream and should be chosen so that membrane fouling is alleviated rather than enhanced.

Among the most critical parameters affecting membrane fouling is the operational water flux. When operating at a constant transmembrane pressure, the achieved water flux is at its highest during plant start-up, as no fouling layer has yet formed on the membrane surface. As Voutchkov [145] said, more pollutants are transported towards the membrane at a higher water flux, thus fouling its surface. This fouling layer increases the membrane resistance and causes a rapid decline in water flux (for a plant operated at fixed feed pressure). The lower water flux reduces the fouling rate, and thus the initial rapid decrease in water flux is followed by a more gradual decline. After a certain operational period, few additional foulants accumulate on the membrane surface due to the lower water flux, and thus a steady state is reached in many cases [146].

To prevent this rapid descent in water flux at plant start-up, operation at constant flux, rather than at constant transmembrane pressure, is a preferred and widely investigated operational choice in experimental and industrial MF and UF applications [147, 148, 139, 149]. Bacchin et al. [125]said that UF operation below the critical flux is necessary to minimise irreversible fouling. In addition, many researchers state that it is also essential to operate at low hydraulic pressures and low permeate drag forces (i.e., low water fluxes) to prevent the compaction of any formed gel/cake fouling layers, which may reduce membrane permeability and make the removal of the fouling layer more complex [149, 150, 151, 128]. In case of pore blocking, operation at high water flux pushes foulants deeper into the membrane pores and hence, enhances pore narrowing [152].

### *2.6. Membrane properties*

Commercially available UF membranes are made of various polymers, such as cellulose acetate (CA), polyamide (PA), polysulfone (PS), polyethersulfone (PES), polyacrylonitrile (PAN), and polyvinyldifluoride (PVDF) [13, 18, 10]. An additional class of membranes is made of ceramics, such as alumina, titania, zirconia, or hafnia. With excellent mechanical, thermal, and chemical stability [153], the ceramic membranes outperform polymeric ones in terms of selectivity [154, 155] and fouling propensity. Both features are due to a narrower pore size distribution of ceramic membranes. Lower organic and microbial fouling propensity [156] made the ceramic membranes the preferable choice for treating complex streams.

Due to their extended stability, ceramic membranes can be cleaned under extreme conditions. However, the range of application of ceramic membranes is more limited due to their substantial cost, brittleness, and significant resistance to water transport [13]. While the development of new, low-cost, highly stable ceramic membranes remains an active research field [157, 158], polymeric membranes remain the market leader for UF applications.

Although the extent of NOM membrane fouling differs between ceramic and polymeric membranes, similar fouling trends were observed by Lee and Kim [159]. Independent of whether polymeric or ceramic membranes are utilized, it is generally agreed that UF membrane fouling is affected by the membrane's: 1) morphology (e.g., pore size, porosity, and surface roughness), 2) surface charge, and 3) its hydrophilicity/hydrophobicity [19, 13].

As mentioned in section 2.1, the relative difference in size between the foulant and the membrane pore dictates whether pore blockage, which is linked to irreversible membrane fouling or the formation of a gel/cake layer, which may be more reversible, is more likely. Complete pore blockage causes a rapid decline in water flux and is most likely when feed particles are in size range of the membrane's pores [160, 161]. On the other hand, smaller particles can be absorbed within the membrane pore, leading to partial pore blockage, and larger foulants tend to accumulate and agglomerate on the membrane surface. Therefore, tight MF and UF membranes with pores smaller than the feed foulants are preferred to ensure foulant retention and prevent irreversible membrane fouling [162, 163, 164, 165].

Hydrophilic membranes are less foulable by hydrophobic NOM fractions and are therefore preferred over hydrophobic membranes [19, 120, 163, 166]. Foulant adsorption can be further prevented via electrostatic repulsion. The electronegativity of both the membrane and the different NOM functional groups are highly pH-dependent, as discussed in section 2.4. At higher pH (pH >7), most polymeric membranes have a negative surface charge due to adsorption of charged entities from a solution [167]. Similarly, most proteins and humic substances are more electronegative (negative zeta potential) at alkaline pH due to the deprotonation of carboxylic and amino groups. Electrostatic repulsion between the negatively charged NOM groups and the membrane surface decreases the fouling tendency [168]. On the other hand, electrostatic attraction between oppositely charged NOM groups and membranes within a specific pH range can increase the membrane's fouling potential [59].

### 2.7. *Novel characterization techniques for membrane fouling*

As detailed in section 2.2, the heterogeneity of NOM complicates its proper characterization. The statement is twice as accurate in the characterization of membrane fouling by NOM. The interplay between different NOM fractions and between NOM and a membrane requires many analytical methods for proper definition. This subsection reviews some novel in-situ, non-destructive, and real-time analytical methods implemented in recent publications to observe membrane fouling. These techniques offer new insights that can help to 1) elucidate various NOM fouling mechanisms, 2) identify the NOM fractions most active at different fouling stages, 3) describe the effect fouling has on the membrane's properties, and 4) evaluate the efficacy of various fouling mitigation techniques. For a more detailed review on various analytical techniques used for the characterization of fouled and pristine membranes, the reader is advised to consult other articles on the matter, including Johnson et al. [169], Rudolph et al. [170], and Chen et al. [171].

One novel technique worthy of mention is optical coherence tomography (OCT), a non-invasive imagining characterization technique (figure 7). Studies involving OCT have offered valuable real-time insights into the progression of biofouling [172, 173, 174, 175, 176] scaling [177], the compressibility of biofilms and its effect on flow properties and fouling reversibility [178, 179, 57]. In low-pressure membrane systems, OCT helped to track the formation of fouling cake layers in low-pressure membrane systems [180, 181, 182, 183] and during membrane distillation [184]. What makes OCT attractive is that it is versatile compared to other characterization methods, such as SEM and MRI, applicable to opaque media layers and requires no dyes or chemicals to analyze the fouled membrane surface [178]. Recently, Kerdi et al. [178] utilized OCT to monitor the evolution of the biofilm structure from initiation to maturation. It was also used to record how the biofilm thickness and elasticity are affected by compression and decompression cycles. OCT was also utilized by Fortunato et al. [185] for analyzing the biofilm formation in a spacer-filled UF feed channel. The results indicated that the feed spacer itself was the cause of the initial biomass accumulation and offered new growth sites which caused a progressive increase in feed channel pressure drop.

As depicted in figure 7, OCT is 1) limited to fouling investigations on flat membrane surfaces, and 2) requires a transparent window, limiting the maximum operational pressure [174, 183]. Furthermore, the scanning depth is limited to approximately 1mm due to light scattering within the sample [186]. Therefore, this method is not ideal for in-situ fouling investigations of existing membrane modules; instead, it is used to investigate fouling phenomena and mechanisms. OCT is also utilized to investigate surface deposition and cake/gel fouling due to its relatively coarse spatial resolution of $\approx 10\,\mu m$ [176, 183]. Thus, the analysis of fouling caused by dissolved organic matter and the related phenomenon of UF membrane pore blockage and pore narrowing (which is often associated with irreversible fouling) requires a more satisfactory resolution currently not possible using optical-based approaches, such as OCT.

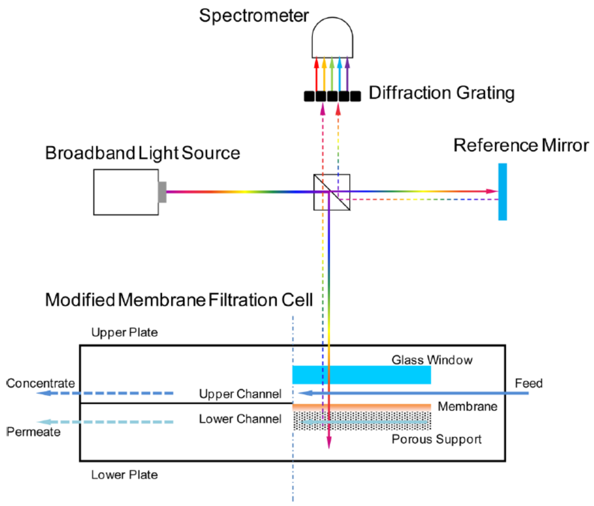


Figure 7: Schematic of optical coherence tomography for real-time tracking of membrane fouling. Adapted from Li et al. [182] with permission of Elsevier Science Ltd.

Yamamura et al. [187] and Yu et al. [188] have adopted the technique of liquid-phase fluorescence excitation-emission matrix (LPF-EEM) analysis, which is commonly used to characterize DOM in solution, to characterize NOM membrane fouling. Fluorescence EEM spectroscopy analysis has been widely used to analyze foulant characteristics because it can detect the presence of tiny amounts of fluorescent NOM without any special pre-treatment [189]. While Yu et al. [188] analyzed a dried membrane sample with front-face EEM, Yamamura et al. [187] managed to analyze membrane fouling in-situ and in real-time using solid-phase fluorescence EEM (SPF-EEM). Valuable observations were made using SPF-EEM; an important one was that transphilic proteinaceous substances caused both physically reversible and irreversible fouling. Hydrophilic proteinaceous gels were mainly responsible for irreversible membrane fouling. SPF-EEM offered a better quantification of foulants than LPF-EEM and has proven its ability to identify DOM fractions responsible for irreversible membrane fouling.

In-situ Raman spectroscopy is another promising method for qualitative and quantitative monitoring of membrane fouling [169, 190]. Raman spectroscopy is a spectroscopic technique typically used to determine vibrational modes of molecules, although other low-frequency modes of systems may also be observed. Tang et al. [191] investigated the synergistic effect between foulant and coagulant during UF and found that in-situ Raman spectroscopy could predict the onset of fouling before any indications of flux reduction. Virtanen et al. [192] used vanillin as a model organic foulant and found that higher vanillin concentrations led to higher membrane adsorption rates, visible by stronger foulant peaks on the IR spectra. Although this method is capable of predicting the type of foulant and the quantity which is deposited onto the membrane, in-situ Raman spectroscopy is still a relatively new technique for membrane fouling observation and faces several issues, such as fluorescence interference [193].

In comparison, the techniques of electrochemical impedance spectroscopy, fluid dynamic gauging, and ultrasonic time-domain reflectometry (UTDR) are functional on an industrial scale [170]. Of particular interest is UTDR, as it is an acoustic technique that is not constrained by the module type. It can monitor membrane fouling in hollow fiber [194, 195] and spiral wound [196, 197] membrane modules. UTDR is a non-destructive technique where the fouling layer can be observed through the opaque pressure vessel housing, as shown in figure 8. The transmission and reflection of ultrasonic waves provide information on the medium (i.e., membrane housing, membrane, fouling layer, and fluid) through which the waves travel. One major issue faced by this technique is that the fouling layer's density must differ from that of the membrane to be identifiable [198]. Furthermore, UTDR cannot offer any qualitative information on the fouling species and generally only provides information on the density and thickness of the fouling layer [170]. Similarly, electrochemical impedance spectroscopy can be applied to any module type, but in contrast to UTDR, it can also offer limited information on the fouling layer composition [199, 200, 201].

The authors of this article believe that these real-time and in-situ membrane fouling monitoring techniques have the potential to exceed their current use as principal membrane fouling investigation techniques. These methods can predict the onset of membrane fouling and offer qualitative and quantitative information on the foulants responsible for membrane fouling, which is precious information in the context of both operational water treatment plants and desalination plants to mitigate fouling. However, these sensors produce vast amounts of data that can be overwhelming in full-scale operational plants. With the development of advanced control systems involving machine learning and artificial intelligence, the potential exists to process this data faster, and operational decisions can be made in real-time to enhance NOM removal and mitigate UF membrane fouling. The interest in machine learning and artificial intelligence in the water treatment and desalination sector is rising. Bagheri et al. [202] and Fan et al. [203] review the use of these intelligent data processing approaches for anti-fouling and improving the product water quality, respectively.

As discussed in section 2, UF membrane fouling depends on various spatio-temporal factors, which complicate the choice of a universal UF fouling mitigation approach. Therefore, the development of in-situ, real-time, and qualitative membrane fouling measurement techniques and their integration into smart control systems for water treatment and desalination plants is required to locally and rapidly optimize their operation.

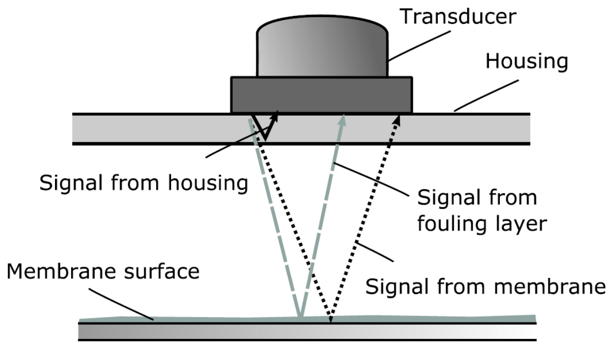


Figure 8: Schematic of ultrasonic time-domain reflectometry (UTDR) for real-time tracking of membrane fouling through the membrane module housing. Adapted from Rudolph et al. [170] with permission of Elsevier Science Ltd.

## 3. Pre-treatment techniques for controlling UF membrane fouling

This section reviews the current scientific knowledge on pre-treatment methods for controlling fouling in UF membranes. Table 1 below details the efficiency of each pre-treatment technique in handling NOM fouling and maintaining membrane performance. A closer review of the relevant literature reveals that coagulation remains the most common pre-treatment and cost-effective approach. A detailed discussion and recent research for each pre-treatment method are presented in subsequent subsections.

### 3.1. Coagulation pre-treatment

Coagulation plays an essential role in controlling the fouling of UF membranes. In general, coagulants neutralize the charge of dissolved and suspended particles and bind them together to form larger flocs. Coagulation efficiency depends on operational conditions such as dosage, pH level, mixing rate and temperature [226, 227, 228]. NOM properties such as particle size, charge and hydrophobicity also play an important role in the effectiveness of the coagulation process [228]. Recently, researchers and industry practitioners have doubled their efforts in finding the optimal doses of coagulant and coagulant aid to minimize fouling [229, 230, 231]. After studying the effect of coagulant for two different samples of surface water, Peleato et al. [232] found that low doses ($<1$ mg/L) of coagulant significantly reduced irreversible fouling [210]. In contrast, an increased dosage of coagulant aid may cause severe membrane fouling [233, 234]. An increase in the concentration of NOM may also challenge its effectiveness as a strategy in controlling fouling, further stressing the need to optimize coagulant type and dosages, the need to combine it with other pre-treatments, and the need for more efficient coagulants.

Coagulant chemicals used to treat water are categorized as either organic or inorganic. Organic polymers can generally be classified as natural or synthetic. Natural polymers are environmentally friendly and non-toxic, while synthetic polymers can sometimes be toxic to humans. However, due to the properties of natural polymers (e.g., Chitosan), they are generally less effective in treating wastewater [235]. Furthermore, inorganic coagulants are much cheaper and therefore widely used in the treatment of high turbidity water. Synthetic organic polymers are more expensive and are sometimes added to the inorganic coagulants. The selection of a suitable coagulant is critical and depends on the water chemistry, the hydrodynamics and operating conditions of the processing system [236, 237]. However, their application includes the generation of potentially toxic residuals, which requires careful administration to avoid health risks [238]. Treatment goals are also important. Operators may choose to treat the water to minimize membrane fouling, remove NOM, or to do both.

Aluminum (Al) salts, such as aluminum chloride, aluminum sulfate, and sodium aluminate, and iron salts (Fe), such as ferrous sulfate, ferric sulfate, and ferric chloride are commonly used inorganic coagulants for the primary treatment [235]. Until recently, Al-based salts have been more popular as coagulants in the pre-treatment of drinking water due to their low cost and their efficiency in removing turbidity and color. Furthermore, Al-based salts are active in the typical pH range between 6 and 8. Iron species are active at pH between 4 and 6; therefore, sufficient charge neutralization requires a significant drop in pH values. Polyaluminium chloride (PACl) is a multicomponent coagulant that contains different species of low, medium, and high molecular weight aluminium species. $Al_a$, the mononuclear species of PACl, presented a better removal efficiency than $Al_b$ (the medium polymer species) or $Al_c$ (the higher polymers species) [239]. The most commonly used chemical coagulant is aluminum sulfate, known commercially as alum. Its disadvantage to other coagulants is the limited pH range over which it is most effective, which range lies between 6.5 and 7.5 [240].

The use of Al-based salts removes 34%-53% of NOM [204]. However, Al-based salts have been criticized for health risks associated with residual aluminum and Alzheimer's disease [241]. As a result, Fe-based salts have gained popularity over Al-based salts due to the absence of known health effects of its residuals [242]. Fe-based salts can remove 28%-40% of NOM [205]. When comparing the Fe(II) activated peroxymonosul-

Table 1: NOM removal efficiency and degree of fouling control using various pre-treatment methods. Extended after Cui and Choo [6].

| Technological option (chemical or additive) | Dosage | NOM removal efficiency (%) | Degree of fouling control [a] | Reference |
|---|---|---|---|---|
| **Chemical coagulation** | | | | |
| Aluminum salt | 3.5 mg/L as Al | 34–53 | 3.2–4.0 | [204] |
| Iron salt | 5–20 mg/L as Fe | 28–40 | 3.4–3.8 | [205] |
| Polyaluminum Chloride | 2.2–5.3 mg/L as Al | 33–38 | 1.15–2.05 | [206] |
| Titanium salt | 10-80 mg/L | 52-89 | 0.42-1.0 | [207, 208] |
| **Activated carbon adsorption** | | | | |
| Powdered activated carbon | 10–500 mg/L | 10–98 | 0.8–1.8 | [209, 210] |
| Granular activated carbon [b] | 1.5 g/L | 11–46 | 3.0–5.5 | [211] |
| Biochar | 20-40 mg/L | 85-97 | 2.55-2.96 | [212] |
| **Mineral adsorption** | | | | |
| Ferrihydrite | 100 mg/L | 22.4–22.9 | 1.69–1.84 | [213, 214] |
| Heated iron oxide particles | 10–100 mg/L as Fe | 21–74 | >2.7 | [215] |
| Heated aluminum oxide particles | 0.7–50 mg/L as Al | 15–46 | About 6.0 | [216, 217] |
| **Chemical oxidation** | | | | |
| Ozone | 0.5–3.0 mg/L as $O_3$ | <1–10 | 1.0–1.7 | [218] |
| Chlorine | 2–11 mg/L as $Cl_2$ | 38–62 | 0.88–1.0 | [219] |
| Chlorine dioxide | 2-15 mg/L | 40-97 | 0.83-1.33 | [220] |
| Permanganate | 0.1-0.5 mg/L | 13-14 | 1.11-1.26 | [221] |
| **Ion exchange** | | | | |
| Magnetic ion exchange | 2.5–15 mL/L | 57–86 | About 1.0 | [222, 223] |
| Fluidised ion exchange | 0.7 L [c] | 60-90 | 0.65-1.0 | [224] |
| Suspended ion exchange | 4-20 mL/L | 55-85 | Not reported | [225] |

[a] The degree of fouling control is defined as the ratio of membrane permeability with each fouling control option to that without it.

[b] The TMP data at operation times of 10 and 70 days were used.

[c] The amount of resin used led to a fixed bed height ∼ 0.7 m. Contact times 1-2 min, and a set flow of 20 L/h for anion FIX and 40 L/h for cation FIX.

fate process as a pre-treatment for membrane fouling control during sludge water treatment, Fan et al. [243] found that after complexing with thiosulfate, adding Fe (II) best mitigated fouling in UF-based systems caused by sludge water. An emerging alternative is Fe(II)/UV/chlorine as a pre-treatment strategy. A higher dosage of Fe(II) and chlorine reduces the effect of fouling [244]. Ma et al. [245] studied the thickness of the cake layer and found that it decreased with the application of Fe in a rotating membrane tank. The faster the rotation speed, the thinner the cake layer was [245].

Polyaluminium chloride is less commonly used as a pre-coagulant agent. Nonetheless, its main advantage over other aluminum and iron salts is its ability to form denser flocs more rapidly and allow for faster settling [246]. The application of PACl improves the removal of nutrients, lowers ultraviolet 254 nm ($UV_{254}$) absorbance, and lowers dissolved organic carbon (DOC) by over 65% [247]. When compared to using PACl alone, Shen et al. [248] found that combining PACl with Poly dimethyl diallyl ammonium chloride (PDMDAAC) with optimal doses of 1 mg/L and 0.1 mg/L, respectively, substantially improves NOM removal. Shen et al. [249] found that PACl-PDMDAAC resulted in higher flux recovery and increased membrane resistance to irreversible fouling in HAs-sodium alginate and HAs-BSA-sodium alginate mixtures than in HAs-BSA mixtures. In general, the addition of synthetic polymers to mitigate fouling of polymeric membranes is not recommended due to potential linkage between the polymers and the membrane and the enhanced irreversible fouling.

Titanium salts have also been used to remove NOM and decrease membrane fouling. Their ability to recover highly valued titanium dioxide from the settled flocs, their ability to form larger and denser flocs compared to other conventional coagulants, and the fact that residual titanium in treated water does not cause any health risks have all gained it much attention as an effective coagulant [250, 251]. Huang et al. [207] compared the floc characteristics of titanium sulfate $Ti(SO_4)_2$ to aluminum sulfate $Al_2(SO_4)_3$ and ferric sulfate $Fe_2(SO_4)_3$. They found that $Ti(SO_4)_2$ forms flocs with a more compact structure, higher strength and larger size. Wu et al. [252] reported similar observations and found that $Ti(SO_4)_2$ flocs were three times greater than aluminum flocs.

Compared to Al- and Fe-based ferric salts, the coagulation efficiency of titanium salt showed comparable or higher performance. It has been reported that the $UV_{254}$ and DOC removal efficiencies of $Ti(SO_4)_2$ were 89.2% and 52.3%, respectively [207]. Furthermore, the final normalized permeate flux for $Ti(SO_4)_2$ was about 0.416, which was slightly higher than those for $Al_2(SO_4)_3$ and $Fe_2(SO_4)_3$ [207]. Okour et al. [253] experimented with titanium tetrachloride ($TiCl_4$) and reported greater reductions in turbidity, $UV_{254}$ absorbance, and DOC, as compared with the treatment using ferric chloride and aluminum sulfate ($Al_2(SO_4)_3$). Zhao et al. [208] compared the performance of $TiCl_4$ coagulation with $Al_2(SO_4)_3$, PACl, iron chloride $FeCl_3$, and polyferric sulfate and found that $TiCl_4$ achieved higher removal of $UV_{254}$ (98%), DOC (84%), and turbidity (93%). In a different study, Wan et al. [254] reported that $TiCl_4$ showed better coagulation performance than $FeCl_3$ with the following $UV_{254}$ removal efficiency $TiCl_4$ (72.4%) > $FeCl_3$ (62.4%) > $AlCl_3$ (59.4%) and the following DOC removal rates $FeCl_3$ (53.4%) > $TiCl_4$ (52.9%) > $AlCl_3$ (42.7%). Another study found the following order for $UV_{254}$ removal $TiCl_4$ (54.9%) > $FeCl_3$ (47.6%) > $Al_2(SO_4)_3$ (41.2%) and following order for DOC removal $FeCl_3$ (57.9%) > $TiCl_4$ (55.1%) > $Al_2(SO_4)_3$ (43.2%) [208]. The study also found that $TiCl_4$ had the largest floc size but the weakest floc strength and the worst re-growth ability compared to coagulants [208].

In recent years electrocoagulation (EC) has offered an alternative to the use of chemical coagulants to remove NOM [255, 256, 257]. This process involves a release of aluminum or iron ions from an immersed anode connected to an external power source [258]. The efficiency of this process depends highly on the electrode and on the chemical properties of contaminants and the pH of the solution [259]. Nonetheless, EC has gained interest in the water and wastewater treatment sector due to its cost-effectiveness, easy procedural approach, and its ability to remove complex organics such as emulsified oil, total petroleum hydrocarbons, and refractory organics.

### *3.2. Adsorption pre-treatment*

Adsorption is yet another pre-treatment process employed in water and wastewater treatment. In this process, compounds (the adsorbate) bind to solid material's external and internal surface (the adsorbent). This process has the potential for NOM removal; however, its effect on membrane fouling is debatable. In some studies, it was shown to reduce fouling [260, 261, 262] while in other studies exacerbates fouling [263, 264], and in some studies, it neither reduced nor exacerbated reversible fouling [3]. Nonetheless, its advantages over other methods lie in its simple operation, reliability, and low energy cost. Such benefits have spurned the search for cheap adsorbents with optimal binding capacity.

Employed in either powder, granular, or impregnated form, activated carbon is the most frequently used adsorbent. Other adsorbents include natural or synthetic zeolites such as Clinoptilolite and mineral adsorbents such as clay minerals, silica gel, and activated aluminum [265]. Activated carbon is used to remove pollutants with intermediate (500-4000 Da) and low molecular weights in wastewater streams and water sources. In comparison, high molecular weight (>10,000 Da) fractions of NOM are typically removed by coagulation [81].

Powdered activated carbon (PAC) is most commonly used in combination with UF membrane systems and the most widely studied form in integrated processes. PAC was found to adsorb low molecular weight and non-fouling molecules yet was found to be ineffective in removing highly hydrophilic extracellular organic matter [210, 266, 267]. In some studies, PAC was even found to exacerbate fouling by forming denser NOM carbon layers on the filtration membrane. On the other hand, granular activated carbon (GAC) appeared to remove intermediate molecular weight fractions and reduce irreversible fouling of UF membranes [211, 261]. Marais et al. [268] found that as indicated by $UV_{254}$, a GAC filtration column preceded by a sand filter column removed 86% of NOM, UF removed 73% of NOM, and conventional water treatment only removed 61% of NOM. At any rate, the structural properties of PAC were found to affect fouling in UF applications. Decreasing the particle size of PAC, for instance, improved the adsorption of NOM [269].

In contrast, NOM portions greater than two kDa failed to diffuse into the PAC particles and instead bonded around the external surface [269, 270, 271]. Shao et al. [272] found that PAC resulted in a synergistic effect with humic acid to form a fouling cake layer on the membrane surface. Fouling increased with increasing PAC dosage and decreased with PAC size. Furthermore, the presence of calcium resulted in the formation of an irreversible dense cake layer on the membrane surface [272]. On the other hand, Cheng et al. [273] found that combining PAC and peroxymonosulfate (PMS) had a synergistic effect for reducing both reversible and irreversible fouling by NOM, with the DOC reduction rates of 53.6%, 24.3%, 27.1%, and 31.4% for HA, BSA, sodium alginate (SA) and their mixture HA-BSA-SA, respectively. Furthermore, the modification of adsorbents such as PAC was found to have a higher potential for removing NOM [274]. However, modified adsorbents could find their way back to the environment and thus cause a threat to aquatic organisms. Although no information was revealed on fouling, combining surfactant-modified PAC with UF resulted in different removal efficiencies of various anionic impurities [274].

When compared to PAC, ferrihydrite (FH) as a pre-treatment adsorbent was found to be more effective in removing high and low to medium molecular weight HA and SA molecules via ligand exchange between the phenolic and the carboxylic groups of NOM and the hydroxyl groups of FH, thus reducing membrane fouling in a hybrid adsorption-UF membrane system [210, 213, 214, 275, 276]. Zhang et al. [277] found that the adsorption capacity of FH followed the order of HA>BSA>SA, while PAC followed the order of BSA>HA>SA. When combined with UF, nanoscale carbon black was also found to remove NOM better than PAC [16]. It was reported that in the absence of coagulation, carbon black removed over 60% of NOM [278]. Higher efficiencies of NOM removal by carbon black was recorded for lower pH levels and in combination with low dosages of coagulants, while over- or insufficient- stirring decreased NOM removal efficiency [279].

In contrast to carbon adsorbents, mineral oxide adsorbents such as iron oxide and aluminum oxide seemed to capture high MW NOM components contributing to cake formation [280]. It was reported that heated aluminum particles (HAOPs) and iron oxide particles (HIOPs) remove more NOM, and NOM with higher $SUA_{254}$ than alum or ferric chloride adsorbents [281, 282]. Studies have found that HAOPs removed 15-46% of NOM while HIOPs removed 21-74% of NOM [215, 216, 217]. A more recent study showed that HAOPs removed 88%, 90%, and 84% of HA, BSA, and SA, respectively [283].

Newer types of adsorbents such as biochars have also been studied to improve the efficiency in an integrated process with a UF membrane [284, 285, 286, 287, 288]. Compared to activated carbon, biochar's hydrophobic characteristics showed a good adsorption performance of organic compounds [289]. Shankar et al. [212] demonstrated that a 6% decrease in total fouling resistance was experienced when using biochar as an adsorbent. Chu et al. [290] compared the adsorption capacity for HA molecules of oxygen and nitrogen-based activated biochar (AB) with PAC and found that nitrogen-based AB showed a higher adsorption capacity than oxygen-based AB and PAC.

Nonetheless, to ensure maximum removal of NOM, adsorption is generally combined with other technologies. Bu et al. [291] found that dosing the adsorbent in a slow-mixing unit after adding a coagulant in a rapid mixing unit better removed NOM. It also resulted in the accumulation of fewer particles on the membrane and the reduction of transmembrane pressure by 4.9 kPa. However, its effectiveness depended on the concentration of the pollutant substance and increased with lower temperature and pH values [291, 292, 293, 16]. A different study employed alum as a coagulant and magnetic chitosan nanoparticles as an adsorbent to increase floc size and improve its settlement performance [294]. Another study showed that synthesized magnetic nano adsorbents combined with alum removed 98.7% of DOC [295]. This adsorption-conjugate process removed hydrophobic fractions of NOM through the interaction of acidic functional groups with hydroxylated oxide surface sites. An additional study employed biochar as an adsorbent and polyaluminum chloride as a coagulant to improve HA and tannic acid removal [287]. Zhou et al. [296] found that MgO is highly effective in removing NOM and served as both a coagulant and an adsorbent. More specifically, in the same study, $Mg^{2+}$ from dissolved $Mg(OH)_2$ removed around 92% of NOM through coagulation while $Mg(OH)_2$ removed the remaining NOM through adsorption, all the while minimizing the generation of solid waste in the process [296].

### *3.3. Oxidation pre-treatment*

Pre-oxidation has also attracted significant interest as a pre-treatment process in abating membrane fouling [2]. The pre-oxidants commonly used include ozone, chlorine, chlorine dioxide, and permanganate. Other pre-oxidants used are peroxy acetic acid and pure oxygen, ferrate (Fe(VI)), or a combination of oxidants [297]. Each pre-oxidant has its advantages and disadvantages. In general, pre-oxidation changes the chemical structure of organic matter, such as the molecular weight and size, functional groups, charge, and hydrophobicity, and promotes the bond formation and mineralization of organic compounds and inhibits microbial growth. All these alternations affect its interaction with the membrane [298].

Pre-oxidation may also have side effects, including the production of disinfection by-products (DBPs). One study demonstrated that pre-oxidation decomposed high MW components but could not effectively remove lower MW components that contribute to the blockage or constriction of the membrane [299]. Other studies showed that the resultant decrease of macromolecules reduced the cake layer and abated flux decline [300, 301, 302].

Ozone is presently the most common oxidant for UF membranes. More than 70% of studies have proven that ozone reduces fouling by more than 30% [2]. Through this pre-treatment method, ozone degrades the colloidal NOM responsible for membrane fouling [303]. One study found that for raw water with an initial DOC level of 38 mg/L, 9 mg/L of ozone can remove up to 35.5% of biopolymers. At the same time, the building block of acids and humic increased by 7.25%, and low molecular weight neutrals increased by 14.62% [304]. However, residual ozone may damage polymeric membrane performance by changing its physical and chemical characteristics such as surface charge, surface hydrophilicity, and pore size, affecting its permeability [305]. Despite this, more than two-thirds of studies have used polymeric membranes; this is possibly due to their low cost and their availability for research [8]. Other disadvantages of ozone include its low mineralization rate and its ability to decompose mainly high MW [306, 307]. Furthermore, ozone damages algae cells and releases intracellular organic matter, increasing fouling [308]. In the presence of ozone, bromide concentrations higher than 0.1 mg/L increase bromate formation, which is not rejected by low-pressure membranes [309]. Other DBPs include low molecular weight aldehydes, ketones, and organic acids [310].

Chlorine is also a commonly used oxidant. Using an in-line pre-chlorination UF system, chlorine was reported to reduce iron, manganese, and sulfite from water [311, 312]. Chlorine was found to remove 38% to 68% of NOM [219]. However, it was found that chlorine may generate a wide range of toxic DBPs such as trihalomethanes and haloacetric acids and was found ineffective in removing micropollutants [313]. Recently, chlorine dioxide has gained popularity as an alternative pre-oxidant to chlorine and was found to remove iron and manganese while minimizing the significant levels of toxic DBPs produced [314]. One study showed impressive results in a hybrid chlorine dioxide UF membrane [220]. In this study, two mg/L of chlorine dioxide removed 97% of the 4.07 mg/L total organic carbon and increased permeate flux to about 80% from the initial permeate flux while maintaining trihalomethanes levels below the United States Environmental Protection Agency guidelines [220]. However, the challenges of chloride dioxide are in the formation of inorganic DBPs such as chlorite and chlorate and undesirable taste and odor.

Permanganate is also used to remove NOM, dissolved iron, and manganese, as well as taste and odor [315]. One study showed that, compared to the results obtained from a direct UF process, using an optimal dose of 0.3 mg/L of potassium permanganate ($KMnO_4$) in a UF hybrid process reduces average effluent concentrations as measured by $UV_{254}$ and total organic content by 14.29% and 4.83%, respectively. It also resulted in lowered total membrane resistance [221]. However, an overdose of potassium permanganate might result in an increase of manganese in permeate water [316].

Other novel oxidation processes of increasing interest include electrocatalytic oxidation, ultrasound sonication, and ultraviolet irradiation. It is now recognized that electrocatalytic oxidation (EO) produces hydroxyl radicals with strong oxidation ability [317]. Xu et al. [318] found that EO reduced HA by 53% and sodium alginate (SA) by 55% and decomposed high molecular weight organics into relatively smaller fractions. Ultrasound sonication can contribute to both the physical disruption and inactivation of microbial structures and the chemical degradation of humic matter [319]. The combined treatment with other oxidants such as ozone was observed to improve NOM oxidation and mineralization. Higher intensity of ultrasound sonication improved the oxidation rate [320]. The combined oxidation by sulfate radicals and coagulation by Fe(II), ferrous-activated peroxymonosulfate (Fe(II)/PMS) was found to reduce membrane fouling caused by NOM significantly [61]. Improved mitigation potential was experienced with increased Fe(II) or PMS dosage [61]. Fe(II)-PS was also reported to effectively alleviate fouling in algae-laden water [321]. However, using Fe(II)-PMS or Fe(II)-PS as pre-treatment processes may increase the amount of sludge due to the precipitation of ferric flocs.

Employing ultraviolet (UV) irradiation and persulfate (PS) as a pre-treatment retarded irreversible fouling of the UF membrane by around 75%. It also improved recorded fouling performance with increased PS doses and extended UV irradiation times [322]. UV-PS at a lower dosage (10 mg/L) aggravated BSA fouling due to the formation of aggregates, which contributed to cake formation [323]. Cheng et al. [324] found that UV-Fe(II)-PMS showed the best performance for fouling reduction, while Fe(II)-PMS significantly mitigated both reversible and irreversible membrane fouling and UV-PMS only reduced reversible fouling. A different study showed that at 4 mg/L of chlorine, a hybrid process of UV-chlorine as a pre-oxidation method and a UF membrane achieved a 49% reduction in membrane fouling and a 59% reduction in reversible membrane fouling [325].

### *3.4. Ion exchange pre-treatment*

Ion exchange (IEX) has been claimed to be more effective than activated carbon, carbonaceous resins, or metal oxides for NOM adsorption [326]. In this process, IEX resins, which may be a crystalline lattice or a gel, carry ions that are exchanged for the ions of other species with a similar charge. Therefore, IEX can only remove charged NOM species and does not affect uncharged NOM species. Most commonly used are synthetic resins, which are durable and designed to suit the specific application required. Ion exchangers can be classified as strong or weak acid cation exchange resins, or strong or weak base anion exchange resins. Cation IEX resins exchange positive ions, while anionic IEX resins exchange negative ions.

In some studies, the use of anion-exchange resins (AER) for NOM removal proved more effective than activated carbon treatment or coagulation. The AER efficiency was influenced by water quality, the nature of the organic compounds, and the type of the resins used [327, 328]. It has been reported that AERs can effectively remove the majority of NOM and prevent the formation of DBPs [329]. Residual NOM, between 10% and 40%, correlates with the uncharged fractions of NOM [330]. AERs also selectively target low and medium-MW organic compounds, while coagulation selectively removes high MW compounds to provide higher combined removal of NOM [331]. Furthermore, compared to coagulation alone, the combination of AERs with coagulation was found superior for reducing DBP formation potential (DBP-FP) [332]. This combined process also showed an increase in floc strength and a reduction in coagulant dose [333]. After testing for AER pre-treatment at water treatment plants in Belgium, the United Kingdom, and the Netherlands, Caltran et al. [334] found that AERs removed 40-60% of total NOM fractions, lowered DPD-FP, and reduced the coagulant doses.

Resins are regenerated after they are exhausted. However, the regeneration and reuse of IEX resins decrease its efficiency in removing DOC and DBP-FP. Finkbeiner and Moore Pereira Jefferson [335] found that virgin IEX resins removed 86-89% of DOC while pre-used IEX resins achieved 67-79% DOC removal. The researchers further found that virgin resins decreased DBP-FP by 31-63%, while pre-used resins decreased DBP-FP by 2-43% [335]. This is due to a partial surface coverage of resin pores by high MW organic molecules [331].

Magnetic ion exchange (MIEX) particle is a robust base anion exchange regenerable resin that contains magnetic iron oxide in its polymer matrix [336, 337]. The MIEX particles are often applied with UF membranes when the membrane plays the role of physical barrier needed to separate the particles from the permeate. Despite its novelty, the reports on the efficiency of MIEX particles are controversial. Positive [338, 339] and no [340, 341] effects on the retention of DOC and minimization of organic fouling are intermixed. The researchers try to relate the general trends to specific definitions of water chemistry and DOC sub-groups.

Mergen et al. [331] found that 56%, 33%, and 25% DOC were removed from water containing 21%, 50%, and 75% hydrophobic NOM, respectively, suggesting that MIEX removes hydrophilic compounds of low MW. The MIEX dose is less important than membrane properties [339]. Kitis et al. [342] found that with a contact time of 10-20 minutes and MIEX dose of 5-

10 mL, DOC was <1.5 mg/L, specific UV absorbance (SUVA) was <2 mg/L and the need for alum coagulant decreased by 0-30 mg/L. The anionic strength of raw water and level of SUVA was found to lower DOC removal and specific UV absorbance efficiency [343]. However, a dose of 8 mL/L MIEX, however, removed more than 75% of DOC after 30 minutes contact time. Adding PAC after MIEX decreased the residual DOC from 1.4 to 1.2 mg/L [344]. 7.97 mL/L MIEX comprehended by 8 mg/L ozone removed 59.77% of DOC [345]. A maximum retention was found at a pH of 9.98, far into the alkaline pH values range [345].

Halogens (such as chlorine, iodine, or bromide) used to disinfect water react with NOM to produce unwanted brominated DBPs [346]. MIEX had a higher affinity towards removing DOC than for bromide ($Br^-$) [347]. In a hybrid MIEX-UF system, Gibert et al. [348] found that 1 mL/L of virgin MIEX, in the form of MIEX-Cl, removed 32% of DOC and 16% of $Br^-$, and an increase of MIEX dosage to 3 mL/L removed 55% of DOC and 37% of $Br^-$. The hybrid MIEX-UF membrane system removed 32-46% more DOC than UF alone [348]. The commercially available Br-selective IX resins, Purolite-Br Plus/9218, proved superior to other resins in the removal of $Br^-$. Purolite-Br resin showed $Br^-$ removal efficiencies that reached 93.5 ± 4.5% out of 0.25 mg/L $Br^-$ in the feed and decreased the generation of brominated DBPs, haloacetic acids, and haloacetonitriles, as well as total organic halogenated compounds by ∼90% [349]. A different study showed that by using MIEX, 64% of the organic micropollutant, carbamazepine, could be removed from water with a turbidity of 20 ± 1.1 NTU and 74% turbidity of 60 ± 1.0 NTU [350]. The addition of MIEX reduces the coagulant dose.

Fluidized ion exchange (FIX) emerged as a different configuration used to introduce AERs into the feed water for NOM removal. This process is based on the sedimentation rate of suspended solids in feed water, which is slower than the sedimentation rate of AERs. The sedimentation rate depends on the diameter and density of the resins as well as the water temperature [224]. Pumping water at a certain speed into an up-flow reactor configuration fluidizes the resins and removes suspended solids from the feed water. When tested, the feasibility of this method before UF proved efficient in removing 80-90% of NOM after five days of using fresh resins and decreased to 60% after that [224]. A different process for removing DOC from surface water is the suspended ion exchange (SIX) process. In this process, the feed water and resins flow through plug flow reactors [351]. The resins are then collected in a lamella settler for regeneration 10-30 minutes after they are introduced into the feed water. The SIX process removes suspended solids using commercially available resins (4-20 mL/L) appropriate for NOM concentration of the surface water [352]. One study found that SIX removes 55-85% of NOM for resin concentrations in the range 10-20 mL/L and contact times between 20 and 40 minutes [225]. Other studies showed 52% DOC removal, 80-85% removal of humic fractions, and a low trans membrane pressure increase of 0.01 KPa/day (68 LMH and 30 minutes filtration time) at one site [353, 354]; and 54-60% DOC removal, 60-80% humic fractions removal and a high trans membrane pressure increase of about 6.5 KPa/day at a different site [355]. After investigating three different surface waters, Metcalfe et al. [356] found that when compared to conventional treatment SIX, followed by in-line coagulation, reduced DOC by an additional 50% and $Br^-$DBP by an extra 47%. This process was chosen for the Andijk Water Treatment Plant in the Netherlands and was found feasible for NOM removal using 13-15 mL of strong basis acrylic gel anion resin with contact time that ranged between 25 and 30 minutes. The selection of the resin was based on adsorption-desorption kinetics, capital cost, and sedimentation properties [352].

Recently, nanotechnology has also been proposed for NOM removal. Where AERs provide a low surface area for ion exchanges, nanomaterials provide a larger adsorption surface area. To exploit the large surface area of nanomaterials, Johnson et al. [357] employed "NanoResins", which are strong base type I anion exchange resins synthesized on the surface of single-walled carbon nanotubes using a robust polymerization mechanism. In this study, NanoResins have a specific capacity of 50 mg-C/g. When tested for NOM removal, NanoResins yielded a specific capacity of 37 mg-C/g, while MIEX yielded a specific capacity of 6.2 mg-C/g after 10 seconds of incubation. Furthermore, when subjected to the same conditions, 0.16 mg of the NanoResins removed 8.31 $\times 10^{-3}$ mg-C of low MW fluorescein, while 0.35 mg of MIEX removed only 0.14 $\times 10^{-3}$ mg-C of low MW fluorescein [357]. Despite this, additional research should be conducted to test and modify the use of nanotechnology for the IEX removal of NOM and attempt to mitigate the environmental risks from the use of such materials.

## 4. Mitigation of NOM fouling through membrane cleaning

It is crucial to maintain membrane performance and sustain membrane permeability to reduce operational costs associated with membrane service life and energy use. The different methods used to remove fouling and clean membranes can be categorized as physical and chemical cleaning methods. Adapted from Kan et al. [358], the effectiveness of each cleaning method is presented in table 2. A detailed discussion of each method's effectiveness and other novel and integrated cleaning methods will be presented in succeeding sub-sections.

In general, chemical cleaning provides a higher membrane flux recovery than a physical one. The drawbacks of chemical cleaning are potential damage to the membranes and the generation of secondary pollutants. Prolonged cleaning periods reduce the permeate flow, generate secondary pollutants and require the filtration unit to be out of operation for the cleaning period [359, 360]. Physical cleaning has documented advantages over chemical cleaning. Physical cleaning is easily integrated as part of the water treatment system, does not require heavy maintenance, does not require plant shut-down for the cleaning period, and does not generate chemical waste. However, this method fails to remove irreversible foulants from the surface or the inner pores of membrane structures.

### *4.1. Physical membrane cleaning*

In general, physical cleaning restores reversible membrane fouling. Standard physical-cleaning methods for membranes include air sparging, sponge ball cleaning, backwashing, backpulsing, and electric field application. Air sparging and sponge ball wiping are simple cleaning methods. Air bubbling presents several possible scenarios for both fouling prevention and membrane cleaning. Small micro and nanobubbles may form a dense layer near the membrane surface that prevents direct contact between the foulants and the membrane surface. Large $>10$ mm air bubbles and slugs form a turbulent movement with wake zones after the front that can remove foulants, typically from the cake layer [371, 372]. Backwashing, or backflushing, involves pumping back water from the permeate side to loosen the fouling cake layer from the membrane surface [359]. Frequent backwashing controls reversible fouling and reduces the need for chemical cleaning [359]. This method, however, is ineffective in widening pore blocking, and reducing irreversible fouling.

Backwashing effectiveness depends on variables, such as membrane and feed properties, and the time and frequency of the applied backwashing [373, 374, 375, 376]. In terms of membrane type, ceramic membranes have higher hydraulic fouling reversibility than polymeric membranes. Alresheedi et al. [155] found that ceramic UF membranes had a 1.5–2 times higher backwash efficiency than polymeric UF mem-

Table 2: Membrane cleaning strategies and efficiencies. Adapted from [358].

| Membrane | Foulant | Membrane cleaning strategy | Flux recovery (%) | Reference |
|---|---|---|---|---|
| **Physical cleaning** | | | | |
| PES, MWCO 70kDa | Ottawa river water, rich in NOM | 7-hour filtration at 100 LMH, 1-2 minute cyclic backwash | 85 | [361] |
| PSF, MWCO 25kDa | Algae-laden reservoir water from South China | Backwash followed by forward flushing, 30 min | 84 | [362] |
| PSF, MWCO 25kDa | Algae-laden reservoir water from South China | Backwash, 30 min | 68 | [362] |
| PSF, MWCO 25kDa | Algae-laden reservoir water from South China | Forward flushing, 30 min | 59 | [362] |
| 0.1$\mu$m PSF | Wastewater | Ultrasonic water bath, 35kHz | 57 | [363] |
| 0.45$\mu$m PVDF | Protein solution | Ultrasonic water bath, 20W; 60min | 43 | [364] |
| 0.60$\mu$m PTFE | Humic acid | Ultrasonic probe, 15W; 25min | 45 | [358] |
| 0.10$\mu$m PVDF | Activated sludge | 15min backwash with DI water | 25 | [365] |
| **Chemical cleaning** | | | | |
| PES, MWCO 100kDa | Water treatment plant, Brabant water, Netherlands | RO permeate backwash, 30s; 250 LMH, and daily CEB, 20min | 93 | [366] |
| 0.01$\mu$m PVDF | Water treatment plant | Steam 90s, and 1M HCl; 30min | 83 | [367] |
| 0.1$\mu$m tubular ceramic | Whey protein | 1.0wt% NaOH; 30min | 80 | [368] |
| 0.10$\mu$m PVDF | Activated sludge | 1M NaOH, 1M $HNO_3$; 12h | 77 | [365] |
| Sulfonated PES UF | Surface and groundwater NOM | 0.1M NaOH; 24h | 71 | [369] |
| 0.1$\mu$m PVDF | Industrial wastewater | 0.5wt% $H_2O_2$; 2h | 53 | [370] |
| CA | Humic acid | NaOH, pH 9.5; 15min | 45 | [64] |

branes. The configuration of UF units also affected the propensity of fouling [377, 378]. To shed light on the effect of filtration mode on total fouling behavior, Chang et al. [66] found that the total fouling of an HA and SA solution under constant pressure filtration compared to, or was slightly higher than under constant flux filtration. Ye et al. [377] found that increasing the duration of filtration from 20 minutes to 60 minutes did not increase the percentage of reversible fouling that could be removed via backwashing; increasing the filtration period from 60 minutes to 90 minutes further exacerbated irreversible fouling; while expanding the backwash duration from 10 to 30 seconds, decreased the initial transmembrane pressure at the beginning of the following filtration cycle and fouling rate by over 50% [377].

Applying short (typically less than one second) and frequent reverse flow, known more commonly as back pulsing, effectively removes surface and pore foulants [379]. In back pulsing, the cake layer is removed by the dynamic and tangential recirculation flow created [380]. It has been reported that back pulsing improved the steady-state net flux by 17% for ceramic membranes with a nominal pore size of 50 $\mu$m [381]. However, back pulsing is less effective for feed water with a high concentration of foulants [8]. Back pulsing can be induced either as a fluid or as compressed gas in pulses to the permeate side. At the same operating parameters, gas back pulsing was found less effective than fluid back pulsing and was also found to embrittle specific membranes [8]. When compared to standard filtration, one study reported a 3.7-fold improvement for water back pulsing and a 3.2-fold improvement for gas back pulsing at the end of a one hour filtration cycle [382].

Back pulsing efficiency depends on several parameters, including the amplitude of the transmembrane pressure in each back pulse, the duration of each pulse, and the frequency of the pulse applied. Gao et al. [383] report that higher amplitude and longer time or higher frequency of back pulsing improves the recovery ratio while back pulsing with low amplitude, short duration, or low frequency is not practical to remove membrane fouling. Mores et al. [384] attempted to study the optimal conditions for back pulsing efficiency and found that backpulse duration is significantly more critical than filtration time. They also found that the longer the filtration cycle, the longer the back pulse duration that was needed. The model also predicted that under optimal conditions, a 6-fold improvement in net flux could be achieved.

Other parameters that influence the efficiency of back pulsing include feed concentration and properties as well as membrane characteristics. In general, the effect of back pulsing decreases as the concentration of foulants in feed water increases [380, 385]. It was observed that a 2-5 fold improvement in permeate flux could be achieved for feed water that mainly contains inorganic compounds [386], and a 10% improvement could be achieved for biological wastewater [387]. Ma et al. [388] observed an 80% flux recovery with backwashing for bentonite suspensions and a 30% recovery for oil emulsions, confirming the difficulty of removing adhesive substances.

In terms of membrane material, polymeric membranes are more sensitive than ceramic membranes to back pulsing pressure differentials [389]. It was further found that backpulsing improved the permeate flux in membranes with higher hydrophilicity [390]. A smaller membrane pore size restricts macromolecules; therefore, a lower back pulsing amplitude is required to remove the external fouling cake layer formed [391]. The surface charge of a membrane also affects back pulsing efficiency. Due to its affinity to water, membranes with higher hydrophilicity produce higher water fluxes. One study showed a 6.33-fold increase in permeate volume with back pulsing for a polyvinyl chloride membrane modified to increase the surface hydrophilicity and a 4.16-fold increase for an unmodified membrane [390]. A different study observed that for carboxylate modified latex particles foulant, an increase in permeate volume with back pulsing was achieved by modifying a polypropylene membrane to be hydrophilic and negatively charged. This modification was performed using acrylic acid. On the other hand, a lower permeate volume with back pulsing was observed by modifying the membrane to be hydrophilic and positively charged. This was achieved using dimethyl aminoethyl methacrylate, which mitigated the electrostatic repulsive forces [382].

### *4.2. The use of ultrasound for membrane cleaning*

Ultrasound has also been used to control membrane fouling and to overcome membrane flux decline. For homogeneous nucleation, high-frequency ultrasound (US) waves generate cavitation bubbles that collapse causing localized pressures as high as 1000 atm as well as possibly form free radicals that decompose foulants in the fouling layers [392, 393, 394]. Heterogeneous nucleation requires lower frequency and results in the formation of bubbles that do not collapse after the sonication stops but act to disintegrate the fouling cake layer. This method does not require a chemical cleaning reagent, therefore avoiding the interruption of the filtration process during cleaning [395]. Continued application of US may, however, damage the membrane surface [396].

Factors that influence the effectiveness of this method include the sonication intensity and frequency. Sonication intensity increases the amplitude of the US wave and hydrodynamic turbulence and thus improves the efficiency of membrane cleaning [394]. Intermittent ultrasonic power was found to increase membrane flux and consume less energy [397]. Higher sonication frequencies generate smaller bubbles that can collapse commonly and less violently, resulting in lower localized temperatures and pressures at the cavitation site. Borea et al. [363] assessed the efficiency of ultrasound cleaning on the degree of UF fouling by the rate of development of TMP needed to maintain a constant flux along a filter cycle. The highest development rate was consistently observed in experiments with no sonication. Low frequency 35 kHz sonication reduced the fouling rate from 27.7 to 11.8 kPa/h for a membrane flux of 150 LMH and from 18.2 to 11.5 kPa/h for a membrane flux of 75 LMH. Moderate reductions to 20.0 and 15.4 were observed when the sonication frequency was increased to 130 kHz. The experiments were performed in a cross-flow mode, so a successful reduction of fouling at higher flux can partially be explained by the shearing force exerted on a cake by the flow [363]. Sonication at 20 kHz and 16W increases the recovery ratio in flat ceramic membrane with 0.2 $\mu$m pores, from 0.21 to 0.70 [395]. The ultrasonic control of membrane fouling caused by hydrophilic NOM was most effective with lower calcium concentrations and moderate pH values. These results are consistent with the work of others [398, 399, 400].

Other influencing factors include viscosity and pressure, feed properties, and membrane properties. Increased temperature reduces cavitational collapse, likely due to a decrease in viscosity and vapor saturation pressure. Optimal cavitation was observed at around 60–70C [401]. Application of sonication to control membrane fouling during filter runs [402] showed an increase in filtration pressure during cross-flow filtration exerted a more substantial drag on the particles on the membrane surface. As a result, the relative permeate flux improvement with ultrasound declined. Furthermore, high feed particle concentrations and fluid viscosity decrease the effect of the sound waves [394]. Sonication is more efficient in reducing fouling cakes than in the release of pore plugging and narrowing. Higher recovery ratio through 0.2 than through 0.8 $\mu$m ceramic membrane was explained by plugging the pores of the latter and a little effect of ultrasonic waves at removing the plugged particles from the pores [403]. The existence of membrane housing will decrease the cleaning effect and will require additional power input. Chen et al. [402] also found that increasing the distance between the ultrasound source and the membrane from 1.7 cm to 2.6 cm and then 3.5 cm decreased permeate flux from 97% to 75%, and then to 60%, respectively.

Combining the US with chemical and/or physical methods could amplify the cleaning efficiency. Popovic et al. [404] found that combining a 30 min sonication at 35 kHz with a mixture of chemical cleaning solution recovered 87 ± 3% of the flux through a pristine 200 nm ceramic membrane. The second round of sonication improved the recovery to 96.3 ± 0.4%. Jin et al. [405] found that combining the US with a 2 g/L citric acid aqueous solution recovered 81% of the initial flux through a PVDF hollow fiber UF membrane. Soaking for seven hours without sonication recovered 73.3% of the flux. Sonication without soaking recovered 56.2% of the initial flux. A similar observation was reached in experiments on polyethersulfone spiral-wound membrane fouled by a 1% solution of skimmed milk and cleaned by a combination of 3 mM of ethylenediaminetetraacetic acid and sonication [406].

Li et al. [407] investigated the cleaning of nylon membranes fouled by effluent from the paper industry using ultrasound with forward flushing. They found that this cleaning process increased the permeate flux of the membrane from 350 LMH to 738 LMH. Lim and Bai [408] found that a combination of complex cleaning cycles that contain NaOH, backwash with deionized water, $HNO_3$, and sonication at 42 kHz recovered 95.7% of the initial flux.

### 4.3. Chemical membrane cleaning

Physical cleaning restores reversible membrane fouling while chemical cleaning effectively removes irreversible membrane fouling [75, 409]. In a chemical cleaning, membranes are soaked in a cleaning solution and then flushed to remove chemicals and foulants [13]. This process could be either performed in-situ without removing a membrane from a rack, ex-situ performed in a separate tank, by chemical washing when chemicals are added during the filtration cycle, and by chemical enhanced backwash when chemicals are added during physical cleaning [410]. Chemical cleaning agents could be classified as alkaline or acidic solutions, metal chelating agents, enzymes, and oxidizing agents. A combination of several cleaning agents is also commonly adopted. Alkaline solutions are used to remove organic foulants through hydrolysis; metal chelating agents withdraw metals from the fouling layer; and oxidizing agents oxidize organics forming oxygen-containing hydrophilic functional groups [411, 412, 413, 414]. An optimal cleaning sequence depends on the feed composition and fouling type and intensity, and its efficiency varies with cleaning frequency, temperature, and pH [415]. Sodium hypocholorite (NaOCl), for example, removes organic matter from the membrane surface and pores and operates better at a higher pH value than hydrogen peroxide [416].

A chemically enhanced backwash (CEB) could be used as a membrane cleaning method to reinforce the physical cleaning process and to avoid periodic shutdown of the membrane unit for the chemical cleaning procedure. Compared to in-situ or ex-situ cleaning, CEB can be fully automated and requires relatively lower concentrations of chemicals and shorter contact times with the membrane [417].This method prevents the need for frequent aggressive cleaning sequences with high chemical concentrations, thus prolonging the life of the membrane. A survey of 83 UF plants in Germany showed that without CEB cleaning-in-place is required at least 1 to 4 times a year, while routine CEB significantly reduced that need to a maximum of once every one to 5 years [418]. Given these advantages, CEB methods gained popularity amongst practitioners and operators and is therefore given more attention in this section.

Oxidizing agents such as NaOCl and hydrogen peroxide

$H_2O_2$ are often used for chemically enhanced backwash. As a backwash reagent, NaOCl causes a reaction with hydrophilic organic matter in the fouling layer and produced chemical by-products [419]. However, no potential health risk was recorded with 4 minutes of application of NaOCl reagent, a 120 minutes backwash interval, a 20 LMH backwash flux, and 25 mg/L of reagent concentration [419]. Gibert et al. [420] found that high transmembrane pressures, and lower frequency and prolonged durations of chemical backwashing effectively removed foulants.

Hydrogen peroxide $H_2O_2$ was added intermittently during chemical washing to the feed tank of synthetic feed treated by coagulation and UF filtration with PVDF membrane. After a 60-day operation, this procedure significantly reduced the growth of bacteria in the membrane tank and reduced by more than 50% the adherence of extracellular polymeric substances, including proteins and polysaccharides, to the membrane surface [421]. The study also found that the low oxidation strength of $H_2O_2$ did not adversely affect the membrane's integrity [421].

Chang et al. [66] reported a reduction of irreversible fouling when the backwash was performed with ultrapure water instead of UF permeate. A similar degree of fouling was found under constant flux and constant pressure filtration modes. Shao et al. [50] found that using chlorinated water for backwashing UF membranes decreased the soluble organic matter level by 12% and the fouling resistance by 8.6%, while pure water backwashing was found more effective at lowering the fouling resistance. Pure water weakens the interaction between organic foulants, which leads to a looser and more porous fouling layer, decreases soluble organic matter level by 29.2% and decreases the fouling resistance by 33.4%. Other researchers have found that using UF permeate for backwashing is ineffective [375, 422, 423] and that using deionized water was useless for cleaning HA-fouled membranes [424]. In addition, researchers showed the effectiveness of using pure NaCl solutions instead of deionized water for backwashing UF membranes fouled by hydrophilic organic matter [425].

The composition of backwash water impacts the control efficiency of the cleaning process. The presence of divalent ($Ca^{2+}$) and monovalent ($Na^{+}$) cations were reported to influence backwash efficiency negatively [375, 118]. $Ca^{2+}$ cations bind negatively charged NOM compounds to negatively charged UF membranes, thus reducing the backwash efficiency [118]. Researchers have further found that increasing the $Na^{+}$ concentration in backwash water decreases the hydraulic cleaning effect [375, 426]. For SA-fouled UF membranes, Chang et al. [427] found that $Ca^{2+}$ in backwash water decreased backwash efficiency by $<$50.3% while using a salt concentration $\geqslant$0.1 mmol/L had a backwash efficiency of $>$91%. Similar results were reported in a different study using NaCl solution as backwash water to achieve around 97% hydraulic cleaning efficiency of HA and SA fouled UF membranes [428]. A comparative study investigated the effects of various backwash waters on the hydraulically irreversible fouling index, total surface tension, and residual HA. It was found that a Milli-Q water backwash cleaned HA-fouled membranes better than a backwash with UF permeate, while backwash water containing $Na^{+}$ or HA outperformed Milli-Q water in alleviating HA fouling [429].

A promising experimental procedure applied a carbon dioxide saturated solution during backwashing to recover membrane permeability after fouling [430]. Reducing the temperature and increasing the pressure ensured a maximum $CO_2$ saturation level in the solution. The $CO_2$ dissolved in the solution then acted as both a physical and a chemical cleaning process. When dissolved in water, $CO_2$ creates a carbonic acid that acts as a chemical cleaning agent. With pressure drops during the cleaning process, the saturated solution becomes supersaturated, thus creating gas bubbles that remove foulants from the surface of membranes [431]. Other gases such as nitrogen and oxygen form stagnated bubbles and reduce the ability to scour the membrane [432]. The study showed that this process could potentially recover 80-100% of the initial flux even under severe irreversible fouling conditions and reduce the use of Milli-Q water to 44% [430]. This is a promising alternative to other methods described above as it prolongs the life span of the membrane and lowers the operational costs of treatment plants.

Applying a constant electric field is another eco-friendly solution to minimize the fouling of the membrane surface. This process coats the membrane surface in electric charges to prevent the adhesion of negatively charged NOM colloids and bacteria to the membrane surface [433]. Cornelissen et al. [434] found that the application of 15 kV/m DC field strength significantly increased UF membrane flux during emulsion filtration but did not prevent irreversible fouling. At the same time, other studies have shown a reduction in transmembrane pressure by 33.7% with the application of 3V and by 51.1% with 5 V after 100 minutes of treatment [435], and a 3.5-fold increase in water flux with 6 V/cm DC electric field in a membrane bioreactor [436]. This process, however, is not without its drawbacks. The heat produced during its application may weaken the membrane properties or generate DBPs with chlorine-containing compounds' decomposition. To mitigate these drawbacks, researchers have proposed studying the use of alternating current instead of direct current [437, 438, 439].

## 5. Membrane modification for NOM fouling mitigation

This section presents studies on promising UF membrane modification, focusing on NOM fouling mitigation and retention. The discussion centers around enhancing polymeric UF membranes, as these are the most widely used in industry. A performance comparison of all the investigated studies is given in table 3 and table 4.

For a detailed discussion about the modification techniques available for polymeric UF membranes, the reader is advised to consult Zhao et al. [440] and Nady et al. [441]. In addition, 3D printing has gained significant research interest as a novel membrane fabrication and modification technique [442, 443, 444]. Although 3D printing allows for the rapid development of new membranes, this technique faces several challenges, such as its 1) limited resolution and printing accuracy, which is especially disadvantageous for fabricating membranes directly, 2) poor scalability, which prevents its use for membrane manufacture on a commercial scale, and 3) high cost in comparison to more conventional membrane manufacturing techniques [443].

### *5.1. Carbon modifications of UF membranes*

Among the various UF membrane enhancements listed in table 3 and table 4, the modification of polymeric UF membranes with different carbon nanomaterials is one of the most promising. Studies show that it is indeed possible to enhance NOM retention while lowering the membranes fouling propensity. Cheng et al. [445] prepared PES UF membranes with either ordered mesoporous carbons (OMCs), namely CMK-3 and CMK-8, or with powdered activated carbon pre-deposited onto the membrane surface via surface coating. Cheng et al. [445] show that both the CMK-3 and the CMK-8 coating layers could counterbalance the flux reduction caused by HA and BSA. In contrast, the PAC layer had little impact on the fouling resistance and could worsen NOM fouling due to the formation of a denser and more complex fouling layer with NOM. CMK-3 showed a higher adsorption rate of HA and BSA of the two OMC coatings and thus achieved a higher DOC removal rate and a lower irreversible fouling resistance. However, neither CMK-3 nor CMK-8 could adsorb the hydrophilic SA molecules. SA could penetrate the OMC layer unhindered and severely foul the underlying PES membrane. Furthermore, the retention of SA only slightly increased with the addition of the OMC layers due to the low hydrophobic interactions between the OMC and SA.

Similar research findings were obtained by Cheng et al. [446], that investigated PES UF membranes coated with carbon nanotubes (CNTs) or with carbon nanofibers (CNFs) layers. While the carbon coating layers well adsorbed HA and BSA, SA was not adsorbed. SA is a high molecular weight compound and caused a severe flux decline by forming a cake/gel layer on the PES membrane surface. However, this fouling layer was easily removed after backwash. Especially the CNTs coating layer showed a remarkable antifouling improvement, even when filtrating natural surface waters [446]. Farid et al. [447] reported fouling prevention by CNT-enhanced UF membranes in natural seawater. In addition to the improved fouling reversibility and the lower flux decline offered by CNT coatings, the CNT surface was also found to significantly inhibit the cell growth of E.coli, which may mitigate biofouling in the UF system and any subsequent membrane process.

UF membranes modified by graphene oxide (GO) showed similar antibacterial and antifouling properties as CNTs [448, 449, 450]. High rejections of BSA and significant fouling reversibility were achieved by Hu et al. [448], even at deficient GO concentrations between 0.004 wt% and 0.016 wt%. The aggregation of GO was prevented by using mixed matrix membranes (MMMs), which enhanced the membrane's rejection, antibacterial and antifouling properties, and increased the membrane's porosity size and it's break strength. Similarly, Yang et al. [451] successfully prevented the aggregation of GO using metal-organic frameworks (MOFs). The incorporation of porous MOFs and hydrophilic GO in the membrane structure also enhanced the water transport through the membrane while not compromising the membrane's rejection capabilities. On the other hand, Algamdi et al. [449] found that at high GO concentrations of 5wt%, the rejection of the enhanced membrane reduced below that of the virgin membrane. Furthermore, the 5 wt% GO membrane had a higher HA adsorption rate, possibly due to an increase in surface roughness. Algamdi et al. [449] found that UF membranes enhanced with low amounts of GO achieved a higher HA rejection and had a lower fouling tendency. In addition, Zhao et al. [450] observed that with more GO layers, the PVDF membrane's initial water flux decreased significantly. However, this was caused by a reduction in pore size, which in conjunction with higher surface hydrophilicity and electronegativity improved the membrane's rejection and its irreversible fouling tendencies.

### 5.2. *Modification of UF membranes using inorganic nanoparticles*

Another promising membrane modification pathway is introducing inorganic nanoparticles (NPs) within the UF membrane structure to mitigate irreversible membrane fouling and maintain, or even improve, the membrane's rejection capabilities. Fouling mitigation using UF membrane modification was proven for singular and synthetic fouling solutions and activated sludge (AS) of concentrations of up to $14000\,mg/L$ [452]. Similar to the findings presented in the other studies, the lower fouling propensity of the enhanced UF membrane was attributed to the membrane's improved hydrophilicity, which ensured the formation of a hydration layer on its surface to prevent the adhesion of hydrophobic fouling constituents.

The chosen nanoparticle may offer other additional benefits. For example, the incorporation of $SiO_2$ can prevent high-pressure membrane compaction, decreasing the membrane pore size and water flux. Furthermore, the incorporation of silver [453] and copper [454] NPs can give the membrane a long-term bacterial resistance. Potential health risks associated with potential leaching of the NPs were proven insignificant as the amounts of leached silver and copper are far below the thresholds set by the World Health Organisation for drinking water safety.

One issue faced in all studies involving the use of inorganic NPs is that beyond a certain threshold, the addition of more NPs lead to inconsistencies in the membrane fabrication, such as the increase in surface roughness, the reduction in hydrophilicity, and to the accumulation of the NPs. Therefore, it is essential to determine the optimal weight percentage of NPs to achieve the desired membrane properties.

### *5.3. Polymeric modifications of UF membranes*

Blending additional polymers can lead to the improvement of conventional UF polymeric membranes. Bai et al. [455] coated cellulose nanocrystals (CNCs) and cellulose nanofibrils onto the membrane surface of PES UF membranes to enhance their hydrophilicity and electronegativity, which reduced the amount of both reversible and irreversible fouling caused by HA and BSA. In a different study led by Cheng et al. [446] on carbon nanotubes and carbon nanofibers, the nanocrystals showed a better dispersibility than the fibers, and thus the CNC coated membranes showed a better antifouling performance, as these had a reduced membrane pore size, a higher electronegativity, and hydrophilicity, and a lower surface roughness than the CNF coated membranes. Based on these findings, Bai et al. [456] focused on developing these CNC coated membranes and showed that irreversible fouling caused by SA could also be mitigated. At the same time, the retention of HA, BSA and SA can be further increased. In this study, the membrane's HA retention increased although the CNC coated membrane had larger membrane pores than the unmodified PES membrane, which can be attributed to the higher hydrophilicity and greater negative surface charge of the modified membrane. A similar finding by Yong et al. [457] showed that at higher lignin weight fractions, the membrane pore size increased. However, the rejection of HA and the oil emulsion still improved due to the improved hydrophilicity and higher surface charge. This proves that surface charge and hydrophilicity are also important membrane characteristics that affect foulant rejection besides the membrane pore size.

Cellulose, or more specifically lignocellulose (LGC), was also used by Lavanya and Geetha Balakrishna [458], as a hydrophilic additive, combined with caramel as a pore-forming agent to enhance a PSF UF membrane. While the rejection of lower MW proteins, such as trypsin, was reduced the rejection of BSA remained constant. On the other hand, the modified membrane had a significantly higher rejection of Methylene Blue and Congo Red, which can be attributed to the stronger charge repulsion between the modified membrane and Congo Red and the higher adsorption rate of Methylene Blue.

One very promising study by Fu et al. [459] showed that with the addition of poly(N-acryloyl glycinamide) (PNAGA) hydrogel the contact angle of the modified PVDF could be reduced to zero degrees within 11 seconds. Furthermore, the weight and morphology of the modified membrane remained almost constant after a 30 min ultrasonic treatment and a seven day operation period showing the stability of the hydrogel-modified membrane. In comparison to other studies [460, 461, 462, 463], this membrane modification could ensure an almost complete flux recovery after four cycles when fouled by BSA at pH 7. Above 90% of the initial flux was recovered when the membrane was fouled by BSA at pH 4.7. This high recovery ratio was attributed to the superior hydrophilicity of the modified membranes, which minimizes protein adhesion to the membrane surface.

In summary, the studies reviewed show that polymeric modification can substantially improve the membrane's anti-fouling properties and NOM retention. The enhancement aims at the higher membrane's hydrophilicity, increased surface charge, reduced surface roughness, and/or smaller pore size. While these studies have highlighted the various possibilities to improve the UF performance, the scalability and economic feasibility of the different membrane modifications have not been discussed. Therefore, it is recommended that researchers focus on new modifications and manufacturing methods that can produce enhanced membranes at a large scale for application in industry.

Table 3: Recent studies on UF membrane modifications for antifouling and enhanced NOM removal. The following abbreviations are used in the table: V= Virgin membrane, SRNOM= Suwannee River NOM, AS= Activated sludge, A= Permeability, CA= Contact angle, ZP= Zeta potential (electronegativity at pH=7), P∅= Membrane pore size, SR= Surface roughness, COD= Chemical oxygen demand, TN= Total nitrogen, TP= Total phosphorous. Modification abbreviations: OMC= Ordered mesoporous carbons, CNT= Carbon nanotubes, CNF= carbon nanofibers, GO= Graphene oxide, MMM= Mixed matrix membranes, MOF= Metal-organic frameworks, NP= Nanoparticles, CNC= Coated cellulose nanocrystals, LGC= Lignocellulose, PNAGA= Poly(N-acryloyl glycinamide).

| Modification | Membrane material | Fouling solution | Membrane characterisation | $J/J_0$ | Irreversible fouling resistance | NOM rejection | Reference |
|---|---|---|---|---|---|---|---|
| OMC (CMK-3) | PES | HA, BSA & SA ($3\,mg/L$), pH=7.0 | A: V > CMK-3 | HA: V= 45%, CMK-3= 61%<br>BSA: V= 42%, CMK-3= 93%<br>SA: V= 3.1%, CMK-3= 3.1% | HA: V > CMK-3<br>BSA: V > CMK-3<br>SA: V ≈ CMK-3 | HA: V= 46.4%, CMK-3= 84%<br>BSA: V= 15.3%, CMK-3=83.5%<br>SA: V= 70%, CMK-3=70-80% | [445] |
| CNT & CNF | PES | HA, BSA & SA ($3\,mg/L$), pH=7.0 | A: V > CNT | HA: V= 45%, CNT= 56%<br>BSA: V= 42%, CNT= 90%<br>SA: V= 3%, CNT= 3% | HA: V > CNT<br>BSA: V > CNT<br>SA: V > CNT | HA: V= 46.4 %, CNT= 90.4 %<br>BSA: V = 15.3%, CNT= 90.1%<br>SA: V= 70.5%, CNT= 70-80% | [446] |
| CNT | PVDF | HA ($10\,mg/L$)<br>& SW<br>pH=7.0 | CA: V= 71.1°, CNT= 19.9°<br>ZP: V= -30mV, CNT= -40 mV<br>P∅: V > CNT | HA: V= 9%, CNT= 60.4%<br>SW: V< 10%, CNT> 20% | HA: V > CNT | HA: V= 80.4%, CNT= 94.6% | [447] |
| GO MMMs | PES | BSA ($1000\,mg/L$)<br>pH=7.4 | A: V < GO<br>CA: V= 65.2°, GO= 43.1°<br>ZP: V= -51.8mV, CNT= -68.6mV<br>P∅: V < GO | BSA: V= 27.4%, GO= 31.3% | BSA: V > GO | BSA: V= 99.2 %, GO= 99.6% | [448] |
| MOF@GO | CA | BSA ($1000\,mg/L$) | A: V < MOF@GO<br>CA: V= 73.2°, MOF@GO= 49.5°<br>SR: V > MOF@GO | N/A | BSA: V > MOF@GO | BSA: V= 91.4%,<br>MOF@GO= 95.4% | [451] |
| GO | PES | HA ($50\,mg/L$)<br>pH=7.0 | A: V < GO<br>CA: V= 80.6°, GO= 56.0° | N/A | HA: V > GO | HA: V= 96.5%, GO= 94.5% | [449] |
| GO | PVDF | SRNOM ($5\,mg/L$)<br>pH=8.0 | A: V > GO<br>CA: V= 73.0°, GO= 56.0°<br>ZP: V= -23.4mV, CNT= -46.8mV<br>P∅: V > GO | NOM: V= 40%, GO= 75% | NOM: V > GO | HA: V= 28.3%, GO= 51.4%<br>FA: V= 30.6%, GO= 59.4%<br>BSA: V= 18.3%, GO= 44.1% | [450] |
| Ag NPs | PEI | HA, BSA, Oil ($1000\,mg/L$) | A: V < Ag<br>CA: V= 90.5°, Ag= 62.1°<br>SR: V < Ag | N/A | HA: V > Ag<br>BSA: V > Ag<br>Oil: V > Ag | HA: V= 82.5%, Ag= 97.0%<br>BSA: V= 85%, Ag= 97.5%<br>Oil: V= 87.5%, Ag= 97.5% | [453] |
| CuO NPs | PES | BSA ($500\,mg/L$) | A: V < CuO<br>CA: V= 69.8°, CuO= 61.9°<br>P∅: V ≈ GO | N/A | BSA: V > CuO | BSA: V> 97%, CuO> 97% | [454] |
| PC-A NPs | PES | AS<br>($1000\,mg/L$)<br>pH= 7-8 | A: V < PC-A<br>CA: V= 62.6°, PC-A= 51.6°<br>P∅: V < PC-A | AS: PC-A= 67%<br>@ $14000\,mg/L$ | AS: V > PC-A | COD: PC-A= 94.2%<br>TN: PC-A= 38%<br>TP: PC-A= 34.2% | [452] |
| $SiO_2$ NPs | PVDF | HA ($100\,mg/L$)<br>BSA ($1000\,mg/L$)<br>Oil ($100\,mg/L$) | A: V < $SiO_2$<br>CA: V= 68.7°, PC-A= 50.7° | N/A | HA: V > $SiO_2$<br>BSA: V > $SiO_2$<br>Oil: V > $SiO_2$ | HA: V= 90%, $SiO_2$= 90%<br>BSA: V= 71%, $SiO_2$= 91%<br>Oil: V= 69%, $SiO_2$= 89% | [464] |

Table 4: Recent studies on UF membrane modifications for antifouling and enhanced NOM removal. The following abbreviations are used in the table: V=Virgin membrane, SRNOM= Suwannee River NOM, AS= Activated sludge, A= Permeability, CA= Contact angle, ZP= Zeta potential (electronegativity at pH=7), P∅= Membrane pore size, SR= Surface roughness, COD= Chemical oxygen demand, TN= Total nitrogen, TP= Total phosphorous. Modification abbreviations: OMC= Ordered mesoporous carbons, CNT= Carbon nanotubes, CNF= carbon nanofibers, GO= Graphene oxide, MMM= Mixed matrix membranes, MOF= Metal-organic frameworks, NP= Nanoparticles, CNC= Coated cellulose nanocrystals, LGC= Lignocellulose, PNAGA= Poly(N-acryloyl glycinamide).

| Modification | Membrane material | Fouling solution | Membrane characterisation | $J/J_0$ | Irreversible fouling resistance | NOM rejection | Reference |
|---|---|---|---|---|---|---|---|
| CNC & CNF | PES | HA & BSA<br>(1000 $mg/L$)<br>pH= 7.0 | A: V $>$ CNC<br>CA: V $>$ CNC<br>$\|ZP\|$: V $<$ CNC<br>P∅: V $>$ CNC<br>SR: V $<$ CNC | HA: V= 11%, CNC= 42%<br>BSA: V= 13%, CNC= 30% | HA: V $>$ CNC<br>BSA: V $>$ CNC | N/A | [455] |
| CNC | PES | HA, BSA & SA<br>(10 $mg/L$), pH= 7.0 | A: V $<$ CNC<br>CA: V= 63.0°, CNC= 48.2°<br>ZP: V=-3mV, CNC=-7mV<br>SR: V $<$ CNC | N/A | HA: V $>$ CNC<br>BSA: V $>$ CNC<br>SA: V $>$ CNC | HA: V= 37.6%, CNC= 71.9%<br>BSA: V= 70.5%, CNC= 82.7%<br>SA: V= 82.5%, CNC= 85% | [456] |
| Lignin | PVC | HA (100 $mg/L$)<br>Oil (1000 $mg/L$) | A: V $<$ Lig<br>CA: V=106.7°, Lig= 41.5°<br>P∅: V $<$ Lig | | HA: V $>$ Lig<br>Oil: V $>$ Lig | HA: V= 73.0%, Lig= 95.3%<br>Oil: V= 65.5%, Lig= 98.4% | [457] |
| LGC-C | PSF | BSA (200 $mg/L$)<br>Dyes<br>pH= 7.0 | A: V $<$ LGC-C<br>CA: V= 73.6°, LGC-C= 51.1°<br>ZP: V=-15.5mV, LGC-C=-25.7mV | N/A | BSA: V $>$ LGC-C | BSA: V$>$ 90%, LGC-C$>$ 90%<br>Dyes: V$<$ 60%, LGC-C$>$ 95% | [458] |
| PNAGA Hydrogel | PVDF | BSA<br>(1000 $mg/L$)<br>pH=4.7 & 7.0 | A: V $>$ PNAGA<br>CA: V=106.2°, PNAGA= 0°<br>ZP: V=-60mV, PNAGA=-40mV<br>P∅: V $>$ PNAGA<br>SR: V $<$ PNAGA | N/A | BSA: V $>$ PNAGA | N/A | [459] |

## 6. Conclusions and future research

The complex, heterogeneous composition and spatio-temporal variability of NOM in source waters remains challenging for water treatment systems. Due to its high variability, it is generally difficult to combat NOM using a single method. Hence, so far no single, universal and economically viable solution has been developed that allows for the complete removal of NOM from source waters using membrane systems without the associated drawback of membrane fouling.

In this comprehensive review, recent studies on hybrid UF treatment systems, UF membrane modifications and cleaning protocols have been discussed to portray these alternative approaches and the progress made to combat UF membrane fouling and to enhance NOM removal rates. The following aspects and conclusions drawn from this review highlight the remaining challenges and also touch on potential research foci:

- Each pre-treatment method reviewed has its advantages and disadvantages. The efficiency of all methods strongly correlate with the NOM and feedwater composition and the operating conditions. Novel methods, such as electro-coagulation, have been proposed as an alternative pre-treatment method for NOM removal. Nonetheless, coagulation remains the most common pre-treatment method reviewed, and it also remains the most cost-effective. In general, integrating different pre-treatment methods capitalizes on the strengths of each pre-treatment process for controlling membrane fouling and maintaining membrane performance.

- Pre-treatment in combination with UF offers the best solution to remove the majority of NOM (also low MW substances) to prevent UF membrane fouling while simultaneously ensuring a higher permeate quality. However, with the inclusion of several processes in series, it is more challenging to operate the larger system more dynamically to cope with temporal changes, such as variation of NOM species in the feedwater.

- It is crucial to develop different NOM characterization techniques, specifically online and real-time monitoring methods to ensure that the plant operation can be dynamically optimized with cutting-edge control systems. The development of such practices and their implementation into large operational plants remains a technical challenge. Therefore, methods and tools for rapid sensor data processing and data transformation into functional choices have gained significant research interest.

- The characterization methods described in the paper, except for UTDR, will be challenging to turn into fully online tools for large-scale water treatment systems, as they are complicated and generate a lot of data. The online tools that will be needed will be based on detecting simple parameters, such as total oxygen, organic carbon, pH, temperature, etc, to generate a data set that can be rapidly analyzed to determine a possible NOM composition to choose the best pre-treatment option. Pre-treatment choices and adjustments should be limited to those already installed on-site, as retrofitting others, such as ion exchange or oxidation devices, to existing plants is cumbersome and expensive. Among easily applicable methods or adjustments are changing the coagulation dose or adding PAC. All others are doubtful, unless already installed previously.

- In terms of the UF cleaning methods reviewed, chemical cleaning provided the highest flux recoveries. However, this process is likely to damage the membrane, generate secondary pollutants, and typically require filtration unit downstream. On the other hand, solely using physical cleaning does not generate chemical waste but fails to remove irreversible foulants from the membrane surface or the inner pores of the membrane structure. One promising physical cleaning method is ultrasound, but its continued use may damage the membrane surface. Another promising cleaning procedure is the application of a saturated carbon dioxide solution during backwashing. Integrated cleaning methods could achieve better flux recovery than any single method reviewed.

- The review of recent studies on UF membrane enhancement shows that the features of conventional polymeric UF membranes can be enhanced to improve both the membrane's antifouling properties and NOM retention. The standard approach of all studies is to improve the membrane's antifouling properties by either enhancing the membrane's hydrophilicity, increasing its surface charge, reducing the surface roughness, and/or reducing the membrane pore size. While these studies have highlighted the various possibilities to increase the UF performance, the scalability and economic feasibility of the various membrane modifications have not been discussed. A major limiting factor preventing the roll-out of these modified membranes is that their increase in performance is not worth the additional production complexity and cost incurred. Hence, it is suggested that researchers focus on new modifications and manufacturing methods that can produce these enhanced membranes at a large scale and at an economically viable cost for actual application in industry.

- Although challenges remain, the recent developments made in terms of fouling mitigation and NOM removal highlight the capabilities and potential of UF as an early-stage purification process.

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