



# Membrane fouling control in low pressure membranes: a review on pretreatment techniques for fouling abatement

Samuel Gyebi Arhin<sup>†</sup>, Noble Banadda, Allan John Komakech, Isa Kabenge, Joshua Wanyama

*Department of Agricultural and Bio-Systems Engineering, Makerere University, P.O. Box 7026, Kampala, Uganda*

## Abstract

Conventional treatment techniques cannot meet the stringent modern water quality regulations emanating from the need to provide high quality drinking water. Therefore, a number of studies have suggested low pressure membrane filtration as a worthwhile alternative. However, a major constraint to the extensive use of this technology in low and middle income countries is the high operating and maintenance costs caused by the inherent predisposition to membrane fouling. Notwithstanding, pretreatment of feed water using techniques such as coagulation, adsorption, oxidation and bio-filtration is believed to control fouling. In this review paper, the existing scientific knowledge on membrane fouling and pretreatment techniques for controlling fouling in low pressure membranes is analyzed with the aim of providing new and valuable insights into such techniques, as well as unveiling crucial issues noteworthy for further studies. Among the techniques reviewed, coagulation was observed to be the most cost-effective and will remain the most dominant in the coming years. Although oxidants and magnetic ion exchange resins can also control fouling, the propensity of oxidants to form health treating precursors and the high economic implications of magnetic ion exchange resins will hinder their adoption in developing countries.

**Keywords:** Fouling control, Low pressure membranes, Membrane fouling, Pretreatment techniques, Water quality



This is an Open Access article distributed under the terms of the Creative Commons Attribution Non-Commercial License (<http://creativecommons.org/licenses/by-nc/3.0/>)

which permits unrestricted non-commercial use, distribution, and reproduction in any medium, provided the original work is properly cited.

**Received** February 1, 2016 **Accepted** June 7, 2016

<sup>†</sup> Corresponding Author

E-mail: arhinsamuel32@gmail.com

## 1. Introduction

Aside human survival, clean water is also a fundamental necessity for socio-economic development. However, insufficient supply of safe drinking water is a critical problem facing many countries in the world [1]. According to WHO/UNICEF Joint Monitoring Programme for Water Supply and Sanitation [2], about 663 million people in the world do not get access to improved drinking water supply. Predominantly in developing regions and particularly in sub-Saharan Africa, many people consume untreated water straight from streams, rivers, lakes, inter alia, which are often prone to pathogenic microbes [3]. Consequently, there are frequent outbreaks of waterborne diseases such as cholera, dysentery and diarrhea [4, 5].

Over the years, conventional water treatment techniques have been used to remove waterborne pathogens in order to meet satisfactory drinking water quality. However, such systems cannot effectively meet the ever increasing and more stringent water quality regulations of modern times [6, 7]. Moreover, the presence of residual chlorine that is used for disinfection in conventional water treatment can react with natural organic matter (NOM) to form carcinogenic disinfection by-product precursors (DBPs) such as trihalomethanes (THMs), haloacetic acids (HAAs), and other halogenated organics [8-10]. Direct exposure to DBPs can also lead to miscarriages and nervous system complications [8, 11].

Conscious of these problems, numerous studies have suggested low pressure membrane filtration (microfiltration (MF) and ultrafiltration (UF)) as a sustainable method for treating drinking water due to their compactness, efficacy in pathogens, turbidity, organic matter and DBPs removal, lower energy consumption, environmental friendliness and easy automation [6, 8, 12-16]. Contrasted with other membrane processes like nanofiltration and reverse osmosis, low pressure membrane filtration is a rather economical approach for removing pathogens from drinking water [17-19].

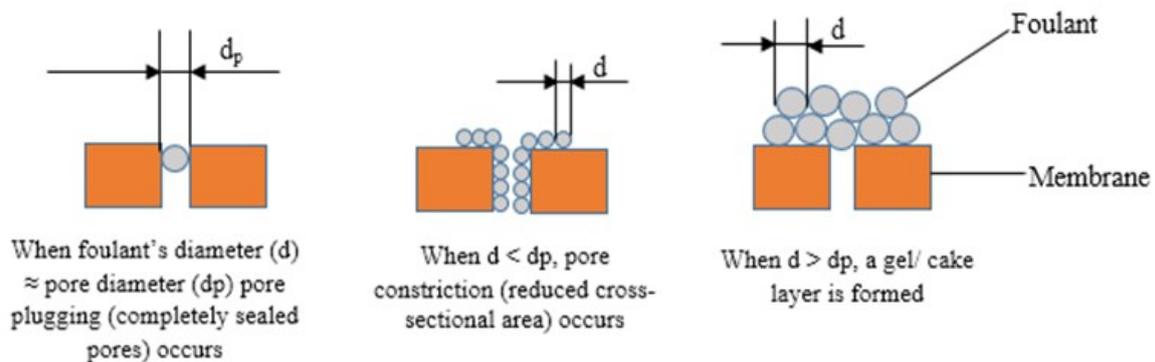
The principle of membrane filtration however implicates sophisticated physical, chemical, and biological reactions between the membrane surface and water contaminants [17]. These reactions usually influence each other, presenting a complicated effect on the filter surface known as membrane fouling. Membrane fouling has been a major hitch to the extensive use of low pressure membranes for drinking water treatment [20-23].

Notwithstanding, pretreatment of feed water has been identified as an efficient way of reducing membrane fouling and increasing permeate quality [19, 24]. In view of that, the aim of this paper is to present a comprehensive review of the recent scientific knowledge on pretreatment techniques used for alleviating membrane fouling. The concept and mechanism of fouling and the influence of techniques such as coagulation, adsorption, oxidation, bio-filtration, and others on membrane fouling are elucidated. In addition, the applicability of the various techniques in low and middle income countries is discussed. It is envisaged that this review paper will provide a comprehensive understanding on fouling reduction abilities of different pretreatment techniques and such knowledge will promote the extensive adoption of the membrane filtration technology for drinking water production in sub-Saharan Africa and other developing regions.

## **2. The concept of Fouling**

Membrane fouling refers to the accrual of impurities on or within the membrane pores. It is a complex phenomenon that describes the blockage of membrane pores during filtration. It is caused by the adsorption or deposition of particulates and compounds on the membrane surface or within the membrane pores [25]. As shown in Fig. 1, when fouling occurs some pores are entirely sealed by dissolved particles while the cross-sectional area of others is reduced. A gel or cake layer may also develop on top of the membrane surface. Membrane

fouling does not only affect the permeate flux, permeate quality and water recovery but also increases the operating cost and shortens the membrane materials' lifespan [12, 26, 27]. Even though Wang et al. [28] reported that fouling layers provide same filtration role as membranes, they also stated that in practical operations, feed water should be pretreated to minimize fouling and improve operational efficiency.



**Fig. 1.** The nature of fouling in low pressure membrane.

Membrane fouling is classified as either based on the origin of foulants or on the fouling reversibility. Depending on the origin of foulants, membrane fouling can be referred to as bio-fouling, organic fouling, inorganic fouling or particulate fouling. Bio-fouling is caused by biofilm formation on membrane surfaces. It originates from colonies formed by aquatic organisms such as algae [21]. Because chemical cleaning routines are often employed in low pressure membrane filtration, bio-foulants may be killed before bio-fouling occurs [17].

Consequently, the specific or possible mechanism of bio-fouling is not well enunciated in literature. Organic fouling on the other hand, has generated a lot of interest in literature with several researchers such as, Zularisam et al. [29], Lee et al. [30], Cui and Choo [31] and Gray et al. [32] reporting on it. NOM from source water is considered to be the cause of organic fouling [17, 33, 34]. Since NOM is ubiquitous in natural waters and at the same time heterogenic in nature, its control is still a major concern. It has been the subject matter of numerous studies yet, the mechanism of NOM fouling is very dissimilar making the findings

of those studies difficult to compare [29, 30]. Aside organic and bio-fouling, inorganic fouling may also occur during membrane filtration. Otherwise known as “scaling”, inorganic fouling is caused by the precipitation of particles (metal hydroxide and metal oxide particles) on an initial layer to form a high resistance cake or gel layer on the membrane surface [35]. Lastly, another type of fouling known as particulate fouling may also result from inert particles and colloids such as silt and clay accumulation inside the membrane pores or on the membrane surface [36].

Based on the fouling reversibility, membrane fouling can also be categorized as physically reversible or irreversible [37-39]. Reversible fouling can be eliminated by hydraulic backwashing whereas fouling which cannot be eliminated by backwashing is referred to as physically irreversible. Irreversible fouling accounts for the plodding rise in membrane resistance following a prolonged period of filtration even though hydraulic cleaning is regularly carried out [17, 40].

### 3. Mechanism of Membrane Fouling

Primarily, particles' removal from solution in porous membranes is influenced by the mechanism of straining. Straining occurs when particles larger than the pores are physically retained by the membrane while water and smaller particles flow through [41]. During straining, the fraction of materials removed by the membrane from the permeate stream is known as the rejection and it is given by Eq. (1):

$$R = 1 - \frac{c_p}{c_f} \quad (1)$$

Where  $R$  is the rejection (dimensionless ratio),  $C_p$  is the concentration of particles in the permeate (mg/L), and  $C_f$  is the concentration of particles in the feed solution (mg/L) [42].

The flow of water through low pressure membranes is based on Darcy's law (the fundamental law for flow through porous media) as shown in Eq. (2):

$$v = k_p \frac{h_L}{L} \quad (2)$$

Where  $v$  is superficial fluid velocity (m/s),  $k_p$  is hydraulic permeability coefficient (m/s),  $h_L$  is head loss across porous media (m) and  $L$  is the thickness of porous media (m).

However, the standard equation for membranes flow is given by Eq. (3):

$$J = \frac{TMP}{\mu R_M} \quad (3)$$

Where  $J$  is the permeate flux ( $\text{m}^3/\text{m}^2 \text{ s}$ ),  $TMP$  is the transmembrane pressure (Pa),  $\mu$  is the dynamic fluid viscosity (Pa·s), and  $R_M$  is the hydraulic membrane resistance ( $\text{m}^{-1}$ ) [43-45].

It is assumed that membranes have straight through cylindrical pores [46], hence when no fouling has yet occurred, the membrane flux is proportional to pressure gradient and the medium's permeability. The Hagen-Poiseuille law is used to describe the flux if the flow through the membrane is laminar and it is assumed to be equal to the flow through a capillary tubes with radius ( $r_p$ ). This is illustrated by Eq. (4):

$$J = \frac{\varepsilon r^2 (TMP)}{8\eta\tau\Delta x} \quad (4)$$

Where  $\varepsilon$  is the porosity (dimensionless),  $r$  is the pore radius (m),  $TMP$  is the transmembrane pressure (Pa),  $\eta$  is the dynamic fluid viscosity ( $\text{m}^2/\text{s}$ ),  $\tau$  is the pore tortuosity factor (dimensionless), and  $\Delta x$  is the membrane's thickness (m).

Although this situation is desirable, membrane fouling is inevitable. In point of fact, different fouling mechanisms will occur simultaneously. Consequently, the resistance-in-series model is used to evaluate membranes' performance with regards to membrane fouling [41, 47]. The model presumes that several component influence the hydraulic resistance and

that each component acts independently. Thus if the osmotic pressure is neglected, the revised Darcy's law (Eq. (3)) can be further modified to Eq. (5) and (6):

$$J = \frac{TMP}{\mu(R_m + R_{ir} + R_{hr} + R_{cr})} \quad (5)$$

$$= \frac{TMP}{\mu(R_m + R_c + R_p)} \quad (6)$$

Where  $R_m$  is membrane resistance coefficient ( $m^{-1}$ ),  $R_{ir}$  is irreversible fouling resistance coefficient ( $m^{-1}$ ),  $R_{hr}$  is hydraulically reversible fouling resistance coefficient ( $m^{-1}$ ),  $R_{cr}$  is chemically reversible fouling resistance coefficient ( $m^{-1}$ ),  $R_c$  is cake layer resistance coefficient ( $m^{-1}$ ), and  $R_p$  is pore constriction resistance coefficient ( $m^{-1}$ ). Eq. (5) and (6) are applicable to any number of individual resistances, caused by reversible and irreversible components, fouling mechanisms (pore constriction fouling resistance, cake fouling resistance etc.) or specific foulants [41].

#### 4. Pretreatment Techniques for Controlling Fouling

Several pretreatment options are utilized in membrane filtration. To select the most appropriate method for enhancing membranes' performance, it is important to identify the major membrane foulants [39]. Generally, the efficacy of pretreatment, with regards to membrane fouling abatement, is strongly associated with several crucial factors. These include the pretreatment agent employed (coagulant, adsorbent, oxidant, bio-filter, etc.), dosage used, mode of dosing (continuous or intermittent), mixing method, temperature, NOM properties (charge density, hydrophobicity, molecular size and molecular weight), solution environment (pH and ion strength), and membrane characteristics (hydrophobicity, membrane charge, and surface morphology) [23, 48]. The effectiveness of different pretreatments methods are discussed in the subsequent sections.

#### 4.1. Coagulation

Coagulation as a pretreatment process is used to enhance the rate of particle aggregation. Due to its low cost and comparatively easy operation, it is the most common and effective pretreatment process used for contaminants removal from drinking water [24]. It remains a promising process for abating membrane fouling as well as enhancing turbidity, dissolved organic carbons (DOC) and microorganisms removal [9, 49]. Consequently, coagulation has generated a lot of interest in literature. A number of researchers [9, 16, 40, 49, 50] have examined its effect on filtration performance (NOM, DOC and residual metal removal), independence on operating conditions (pH-values, coagulants type, coagulant dose, type of mixing, mixing intensity, etc.) and the membrane configuration (module design, membrane materials, pore sizes, etc.)

Howe and Clark [50] conducted laboratory experiments to evaluate the effect of coagulation on MF/UF performance. The key variables examined were the source water, type of coagulant, coagulant dose, coagulation application condition, and membrane material. The authors observed that fouling reduced when coagulant doses for enhanced coagulation were used but under-dosed coagulants produced greater fouling than when no coagulant was applied. In a related study, Xiangli et al. [9] tested the effect of ferric chloride ( $\text{FeCl}_3$ ) on the performance of a large-scale UF system in treating high turbidity surface water. Their study showed that by optimizing the dosage of  $\text{FeCl}_3$ , UF installation could operate stably for six months without chemical cleaning and yet produce high quality drinking water. Recently, Lai et al. [16] studied the effects of alum coagulation pretreatment on NOM removal from surface water, and on fouling control. The results of the study indicated that coagulation pretreatment could remove some of the NOM and thereby lessen chances of flux decline associated with

UF membranes. Table 1 summarizes reviewed studies with coagulation followed by low pressure membrane filtration.

Optimizing the coagulation process is very crucial [51]. To begin with, the type of coagulant used can have substantial impact on membranes' performance. As shown in Table 1, various coagulants were used by different authors. Kabsch-Korbutowicz [52] used alum,  $\text{NaAlO}_2$ , and polyaluminum chloride (PACl) in an in-line coagulation-UF and observed that alum and PACl caused an appreciable reduction in membrane fouling by enhancing organic matter removal but  $\text{NaAlO}_2$  had no effect on membrane fouling. Even though Howe and Clark [50] observed no coherent or foreseeable variations in membrane performance based on the coagulant type, they noticed that PACl worked better with a specific water source. PACl has the ability to form more robust flocs, works well under low temperature regimes and produces less sludge compared to alum and  $\text{NaAlO}_2$ . These capabilities of PACl were the reasons why less membrane fouling and better membrane performance was observed when PACl pre-coagulation was used.

**Table 1.** Summary of Reviewed Papers with Pre-Coagulation Followed by MF/UF

Author	Water Source	Coagulant used	Dosage	pH level	Mixing period	MF/UF performance
Fiksdal & Leiknes (2006)	Autoclaved tap water	Alum and PACl	3 and 5 mg Al/L	8.3	1 min fast mixing followed 15 min slow mixing	$\geq 6.7$ log removal of virus
Howe & Clark (2006)	Natural surface water	Alum	0 - 50 mg/L	6.9 - 8.0	Rapid mixing followed 30 min flocculation and 30 min settling	5 - 27% DOC removal
Kabsch-Korbutowicz (2006)	Surface water	Alum, PACl, and NaAlO <sub>2</sub>	1.79 - 3.59 g Al/m <sup>3</sup>	No information	3 min rapid mixing followed by 20 min slow mixing	Enhanced NOM removal
Xiangli et al. (2008)	Surface water (river)	FeCl <sub>3</sub> , FeSO <sub>4</sub> , and PACl	No information	No information	10 min rapid mixing followed by 30 min settling	$\geq 99.8$ bacteria removal
Hatt et al. (2011)	Secondary wastewater	PACl, alum, and Fe <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub>	0.5 - 2 mg/L as Al or Fe	6.7 - 7.2	10 s rapid mixing at followed by 120 s slow mixing	7.5 - 16.5% DOC removal
Matsushita et al. (2013)	Surface water (river)	PACl, FeCl <sub>3</sub> , and alum	0 - 40 $\mu$ mol/L as Al or Fe	6.8 (5.8 and 6.3 for FeCl <sub>3</sub> )	Hydraulic retention time of 1.8s in an in-line coagulation	$\geq 4.3$ log virus removal
Kimura et al. (2014)	Surface water (five different rivers)	PACl	2 mg Al/L	No information	Mixing was done at G = 100 s <sup>-1</sup>	Reduction in membrane fouling
Kim (2015b)	Laboratory prepared NOM solution	pDADMAC	0 - 60 ppm	7.2 $\pm$ 0.1	6 min rapid mixing at G of 400 s <sup>-1</sup>	> 66% TOC removal
Lai et al. (2015)	Surface water	alum (Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub> )	0 - 16 mg/L (as Al)	8.5	3 min rapid mixing at 100 rpm, reduced to 35 rpm for 15min followed 30 min settling	Removal of dissolved organics in 40 - 70 kDa range

Besides the coagulant type, optimizing the coagulant dose is another crucial consideration [53]. As evident from studies presented in Table 1, under-dosed coagulation could be detrimental to membrane performance. With under-dosed coagulants, significant membrane fouling was observed by various authors. However, adequate coagulant dose reduced fouling drastically and enhanced membrane performance momentarily. At optimized coagulation conditions, high microorganisms and other waterborne impurities removal rates were also observed in those studies.

Optimizing operating conditions such as pH of raw water also enhance coagulants' performance which eventually leads to less fouling and improved membrane performance. Even though studies have shown that using PACl coagulant do not require pH adjustments for water sources with pH ranging from 6.7-7.2 [54], other coagulant like alum or ferric chloride ( $\text{FeCl}_3$ ) may require pH adjustments for optimum performance [55].

The mode of coagulation may also affect coagulants' performance. Coagulants may be applied in either in-line or standard mode. In-line coagulation occurs without sedimentation or pre-filtration whereas standard coagulation requires sedimentation. These two configurations have however been a contentious theme in literature with researchers articulating diverse views. Dong et al. [58] observed that during in-line coagulation, floc-cake layers formed on the membrane surface adsorbs hydrophilic neutral fraction of small size but when standard coagulation was used, the membrane rejected much of the hydrophilic neutral fraction with small size which contributed to a slow flux reduction. On the contrary, Kim et al. [59] suggested that for MF membranes with treated water containing few flocs, mechanical mixing with the back mixing-type is more effective in controlling fouling due to residual NOM than pump diffusion mixing with the in-line type.

The specific components of the feed water and properties of NOM could be responsible for these conflicting views on the mode of coagulation. Even though Kim et al. [59] reported that conventional rapid mixing is more effective in controlling fouling caused by humic NOM, they also observed that for non-humic NOM fraction, in-line pump diffusion mixing is more effective [60]. To add to that, the findings of Park et al. [61] indicate that the characteristics of flocs and the ability of the mixing mode to remove dissolved organic matter from the feed water could be responsible for the differences in filterability for standard and in-line coagulation. Therefore for optimum performance, it is important that the mixing method is chosen based on the properties of NOM in the feed water. Nonetheless, in view of ongoing research for simplification of membrane-based waterworks, Kabsch-Korbutowicz [52] proposed that in-line coagulation should be considered for small membrane systems as complex water treatment train may not be advisable for such systems. This view was reiterated by Kimura et al. [40] when they studied the effect of coagulation on different water sources and observed that in-line coagulation could be efficient in controlling fouling.

The impact of coagulation on fouling control may also depend on the kind of membrane material employed. Hydrophobic membranes are easily fouled by hydrophobic NOM fraction in the feed water via strong hydrophobic interactions [62]. Therefore, the ability of the mode of coagulation to remove either hydrophilic or hydrophobic NOM fraction coupled with the hydrophobicity of the membrane material could influence the extent of fouling. Using two different kinds of UF membranes, Lai et al. [16] noted that pre-coagulation had less effect on cellulose acetate (CA) membrane flux than that of polyvinyl chloride (PVC). According to the authors, alum coagulation resulted in higher removal of high molecular weight organics (biopolymers and humic substances) than low molecular weight acids. Because pre-

coagulation was more effective in controlling biopolymers, the effect of coagulation was more significant in the PVC membrane than the CA membrane.

Aside that, the efficacy of pre-coagulation may also be influenced by the flow configuration utilized [63]. Two types of flow configurations known as cross-flow filtration (CFF) and dead-end filtration (DEF) can be used in membrane filtration. During CFF, the shear-induced particle diffusion and inertial lift associated with the circulation of water across the membrane surface prevents particles from plugging the filter pores [25,64]. As a result of this, CFF enhances membrane filterability even for highly turbid water sources following a pre-coagulation process [44]. Nevertheless, CFF requires comprehensive operator training and may not be conducive in conducting rapid-response investigations [65]. Additionally, because surface water has less concentration of solids and most UF/MF plants operate with feed water turbidity less than 100 NTU, the advantages of CFF are less significant in the water industry [41]. Therefore, the single pass DEF with periodic backwashing is mostly used [30]. DEF is however highly dependent on the backwashing efficiency [9, 58, 66] and the mode of operation. In the constant flux mode, membrane and/or cake compression may occur, which could lead to a decrease in backwashing efficiency [13]. Notwithstanding, Lee et al. [30] observed that low constant flux filtration reduces the chances of membrane clogging, though their findings were inconsistent and indistinct to compare with constant pressure filtration in the range of 10-90 kPa.

Considering the low cost of coagulants, the ease of operation, the possibility of using lower doses compared to conventional coagulation [10] and yet, their tremendous ability to control membrane fouling, coagulation pretreatment could be a viable option for low and middle income countries.

## 4.2. Powdered Activated Carbon Adsorption

Adsorption as a pretreatment option refers to the adhesion of foulants to the surface of an adsorbent. Due to their relatively high porosity, adsorbents have a relatively large specific surface area for absorption or accumulation of absorbable impurities [24]. With regards to membrane filtration, the most popular adsorbent is powdered activated carbon (PAC) [35].

Analogous to pre-coagulation, there are two configurations for adsorption coupled with membrane filtration. The system can be in a unified membrane reactor or, a detached reactor, following a PAC reactor. Also there are two ways of dosing – the step input of PAC where the reactor is dosed at a constant rate and the pulse input where all dosing is done at the beginning of the filtration loop. The influence of the dosing method on membrane fouling regulation is however less enunciated in literature.

Several studies have examined the influence of PAC on membrane fouling and membrane performance [35, 67-71]. According to Liu et al. [67], PAC-UF process can significantly reduce membrane fouling of algal-rich source waters. In addition, Gai and Kim [69] studied the effects of PAC on the performance of immersed flat sheet membrane system. The experiments were conducted with PAC dose of 0 g/L and 20 g/L. At the end of the study, they concluded that, PAC coupled with membrane filtration is a viable process for controlling fouling as the TMP of the membrane without PAC rose to 61 kPa after 48 days. But with PAC dosage of 20 g/L, continuous filtration experiments were conducted successfully for 64 days. Yet in another study, Kim et al. [70] also revealed that MF systems combined with high dosage of PAC can be used in treating highly contaminated raw water with organics for advanced water treatment.

On the contrary, other reports argue that PAC has minimal influence on fouling control [68, 71, 72]. Even though PAC can remove significant amounts of NOM from water, Li et al. [68]

showed that PAC pretreatment can slightly ameliorate humic acid fouling. Additionally, Kim et al. [71] and Lee et al. [72] reported that PAC preferentially removes non-fouling molecules instead of foulants. Hence, even though they observed high DOC removal, they concluded that PAC could not abate fouling.

These conflicting opinions on PAC's influence on membrane fouling were attributed to the difference in membrane properties and/or NOM characteristics of different water sources [35]. However, Campinas and Rosa [33] reported that irrespective of the NOM characteristics of the feed water, PAC has no effect on the permeate flux as well as on reversible fouling. Hence, the variations in reportage on PAC performance in relation to fouling abatement, could be accredited to variations in membrane properties, types of PAC, dosage and size of PAC particles used in different studies.

Therefore for optimum performance, it is essential to consider the type of PAC. Lee and Walker [73] examined the effect of PAC type on cyanobacterial toxins (microcystin-LR) removal from drinking water and observed that the efficiency of wood-based activated carbon was four times greater than that of coconut-based activated carbon. In a related study, Haberkamp et al. [74] compared the effect of four commercially available PACs on the removal of macromolecular DOC from secondary effluent using flat-sheet polyethersulphone (PES) and polyvinylidene fluoride (PVDF) UF membranes. The authors observed varying results depending on the type of PAC used. Consequently, they concluded that the differences in the adsorption affinities of macromolecular DOC depended on type of PAC applied.

On top of that, it is also crucial to use an adequate dosage of PAC. As proposed by Kim et al. [70], higher total organic carbons (TOC) and ultraviolet absorbance at 254 nm ( $UV_{254}$ ) removal rates were observed at a high dosage of PAC (40 g/L) irrespective of the filtration rate. Additionally, the authors noted that at higher PAC dosage, effluent quality and filtration

efficiency improved. Ying and Ping [75] also asserted that at increasing PAC dosage, cake resistance decreases. Although they also observed that at PAC dosage of 0.75 g/L, irreversible fouling reduced effectively when they studied the effect of PAC dosage of 0, 0.75 and 1.5 g/L on membrane fouling. Torretta et al. [76] however shared a contrasting view that increasing PAC dosage has no effect on membrane fouling. According to the authors, low PAC dosage of 5 mg/L was effective in reducing flux decline by 27% but increasing dosage (to 10 and 20 mg/L) had no significant influence on permeate flux yet, resulted in increased operating costs. These variations in reportage could be due to differences in raw water quality, process configuration (unified or detached reactor) and mode of application (step or pulse input). Thus, a high PAC dosage does not necessarily limit fouling and improve system performance. Therefore preliminary tests should be carried out to determine the optimum PAC dosage before usage.

Besides PAC type and dosage, the PAC size must be also be optimized. Reviewing a number of studies, Stoquart et al. [35] deduced that PAC size greatly affects membrane fouling. Larger PAC sizes provides void spaces for interactions between membrane pores and colloidal matter which intensify PAC cake fouling while very small PAC size can facilitate the adsorption of foulants onto the membrane pores. Thus, PAC particle size of approximately 100 times larger than the pore size is recommended [77].

The specific characteristics of the feed water could also influence PAC's performance. The quality of the feed water can affect PAC's adsorption capacity, its biodegradation kinetics and the rate of microbial development at its surface. Quality parameters such as pH and temperature could influence ammonia oxidation and adsorption of organic matter respectively while the presence of metal ions in the feed water may also influence ion exchange at PAC's surface [35].

Moreover, the mode of operation can influence the efficacy of PAC. Using low operating flux could limit membrane fouling by increasing the contact time between PAC and water [78,79]. Also, when operated in crossflow mode, the abrasion of fouling layers limits the chances of fouling [77]. According to Oh et al. [80] the scouring effect provided by added carbons decreases the deposition of large particles on the membrane. This scouring effect is however dependent on the crossflow velocity (CFV) which is highly influenced by pressure, and the dosage used. At low PAC dosage of 5 mg/L, Campinas and Rosa [33] observed no reduction in fouling after increasing the CFV from 0.5 to 1 m/s. Although operating in the crossflow mode could reduce fouling at a high PAC dose, it could also lead to high operating cost and yet, from a water quality perspective its influence could be minimal [81]. To add to that, depending on the membrane material, type of PAC and configuration used, abrasion could affect the integrity of the membrane. Avoiding contact between membrane surface and PAC by implementing a separation step has therefore been proposed as a way of controlling these challenges associated with CFF [35].

Even though PAC adsorption is cost-competitive [82], in assessing its applicability as a pretreatment option for membrane fouling control in developing countries, it is crucial to ascertain if PAC particles can get into the membrane pore to cause membrane fouling. The possibility that some impurities may not be absorbed by PAC but can easily be adsorbed into the membrane pores could inhibit the extensive use of PAC.

#### **4.3. Pre-oxidation**

Aside pre-coagulation and adsorption, another way to reduce membrane fouling is pretreating the feed water with oxidants. Oxidants regulate membrane fouling by altering the interactions between membrane surface and components of the solution. They suppress microbial growth,

or alter the structure and properties of NOM, and serve as disinfectants. Ozone, permanganate, and chlorine are the main oxidants used in water treatment.

The effect of ozone pretreatment has been extensively reported in literature [83-88]. In a hybrid ozonation–ceramic UF membrane system treating natural water at typical TMPs, Kim et al. [85] examined the effect of ozone dosage and hydrodynamic conditions on permeate flux. The study showed that under suitable operating conditions, the hybrid ozonation – ceramic UF system can significantly reduce membrane fouling. In another study, Zhang et al. [84] observed that with ozone dosages of 2.0-2.5 mg/L, membrane fouling was alleviated and membrane working cycle time doubled under the tested conditions. To add to that, ozone dose of 2.0-5.0 mg/L was also observed to be efficient in controlling membrane fouling and enhancing the removal of multiple contaminants [83].

In spite of these fascinating results, ozone pretreatment seem to be dependent on the quality of the feed water. Even though ozone pretreatment greatly influenced the removal of hydrophobic NOM in both river water and secondary effluent, it could hardly oxidize hydrophilic NOM [87, 88]. Aside that, the concomitant usage of ozone pretreatment and membrane filtration is hindered by low ozone resistance of most polymeric membranes and the predisposition to increase the levels of biodegradable dissolved organic carbon (BDOC) [89]. BDOCs can boost bacterial growth and biofilms formation in the distribution system. To cap it all, the crux of the matter and yet barely pronounced limitation to ozone pretreatment is the possibility of bromate formation [17, 86]. Bromate is carcinogenic and therefore undesirable in drinking water.

Though not extensively, chlorine and permanganate have also been reported as oxidants for fouling control [90-92]. Choo et al. [90] reported on the removal of iron and manganese from water using an in-line pre-chlorination UF system. At chlorine dosage of 3 mg/L as Cl<sub>2</sub>,

they observed over 80% rise in manganese removal efficiency. However, at higher dosage of chlorine (5 mg/L as Cl<sub>2</sub>), no significant increase in metal ions removal was observed and yet more serious membrane fouling occurred. The possibility that chlorine may react with NOM to form carcinogenic DBPs like THMs and HAAs is a major health concern inhibiting the use of chlorine pretreatment [86].

In terms of arsenic removal, permanganate could be a better pretreatment option than chlorine because of its positive seeding effects of in situ formed hydrous MnO<sub>2</sub>(s) on ferric precipitate aggregation [91]. However, high dosage of permanganate can cause pink-colored water. Furthermore, it may form precipitates that cause mudball formations on filters [92]. Such precipitates are difficult to remove and these demerits prevent the extensive use of permanganates as pretreatment for drinking water filtration.

Normally, oxidants are used to degrade precursors in the water sources, but the propensity of forming new DBPs (chlorination), bromate (ozonation), or precipitates (permanganate) may require serious consideration before such systems can be adopted for portable water treatment in developing countries.

#### **4.4. Bio-filtration**

Unconventional pretreatments techniques such as bio-filtration, have also been reported as options for fouling control [93, 94]. Although biological treatment is mostly used for wastewater treatment, Mosqueda-Jimenez et al. [94] investigated the use of bio-filter as a pretreatment for UF of drinking water and observed that membrane fouling was reduced in the bio-filter – UF integrated system than when UF was executed in solitary. Similarly, Hallé et al. [93] evaluated rapid biological filtration as a pretreatment for UF. Based on the findings of the

study, they concluded that bio-filtration as a simple and robust pretreatment may be typically apt for small-scale drinking water treatment.

In view of the rampant upsurge in water pollution in some developing countries like Uganda [95], biological pretreatment, as a chemical-free treatment is seen as a viable option for removing biopolymers and particulate matter from polluted water sources in such countries [96, 97]. However, additional studies are required to evaluate the feasibility of bio-filtration – membrane filtration systems, particularly in large scale water treatment plants.

#### **4.5. Magnetic Ion Exchange Resins**

The use of magnetic ion exchange resins (MIEX<sup>®</sup>) have also been demonstrated by some studies as potential pretreatment for fouling control in low pressure membranes [98-100]. MIEX<sup>®</sup> are three-dimensional structures with polymeric chains. The structures contain magnetized components, and organic matter removal is achieved by means of ion exchange.

According to Zhang et al. [98], MIEX<sup>®</sup> can eliminate a greater part of hydrophilic compounds together with a substantial amount of hydrophobic compounds from biologically treated secondary effluent within a short contact time (20 min). Their study further revealed that at optimal concentration, MIEX<sup>®</sup> could remove over 60% DOC in wastewater and when combined with PAC, over 80% TOC removal can be obtained. Thus, MIEX<sup>®</sup> pretreatment ensures a longer period of membrane filtration with less fouling. In a similar way, the effectiveness of the MIEX<sup>®</sup> in NOM removal was evaluated by Kitis et al. [99]. Their study showed that even at relatively low dose and short contact time, MIEX<sup>®</sup> can effectively removes NOM from raw water. Additionally, Ding et al. [100] revealed that MIEX<sup>®</sup> has the thermodynamic ability to adsorb both NOM and bromide from aqueous solutions.

Other reports however indicate that MIEX<sup>®</sup> preferentially adsorbs non-fouling NOM, but have relatively little influence on the sorption of biopolymers, which are more responsible for irreversible fouling [101-103]. This phenomenon implies that probably there is a variation in the characteristic of NOM reported by different authors. Using three different raw water of fundamentally different NOM characteristics, Mergen et al. [104] showed that an inverse relationship existed between raw water hydrophobicity and NOM removal by MIEX<sup>®</sup>. According to the authors, increasing raw water hydrophobicity resulted in decreasing DOC removal. Consequently, the authors concluded that NOM removal by MIEX<sup>®</sup> was water specific.

Research on MIEX<sup>®</sup> performance on pilot scale or large scale, especially in developing countries, is however lacking due to the high economic implications associated with the technology.

#### **4.6. Integrated Pretreatment Systems**

A handful of authors have capitalized on the theoretical merits associated with specific pretreatment option and have integrated a number of pretreatments into a unified system to supplement each other's shortcomings. For instance Mozia et al. [105] considered an ozonation – adsorption – UF system for treating surface water. Haberkamp et al. [74] also studied the impact of coagulation and/or adsorption pretreatment on DOC removal from secondary effluent. Furthermore, Watson et al. [106] studied the influence of enhanced coagulation (EC) followed by MIEX<sup>®</sup> and EC followed by PAC on DBPs removal while Kim [107] reported on alum under-dose coagulation coupled with MIEX<sup>®</sup> for fouling control.

Usually, integrated system may have high capital costs which could be a challenge for most developing countries. Yet, if the system is effective in controlling fouling and

ameliorating membrane performance, operational costs may decrease. Even if such systems result in high total costs, it may still be the only viable option in situations where the source water has very poor qualities and yet high quality effluent is desired but fouling cannot be effectively controlled using any known single technique [24].

Some integrated system may however exacerbate membrane fouling. As evident from previous studies, such systems were not very efficient and significant flux decline was observed [74, 105]. Yet, other studies reported the contrary. According to Kim [107] combining under-dosed alum and MIEX<sup>®</sup> reduced fouling to a greater extent than when MIEX<sup>®</sup> was used alone. To boot, Watson et al. [106] observed DOC removal rates of  $70 \pm 10\%$  during their EC/PAC experiment, while a DOC removal of  $66 \pm 12\%$  was recorded for EC/MIEX. A possible explanation to the contentious performance of integrated pretreatment systems could be that, combining some pretreatment procedures may induce precipitate formation from the reactions inter se which could have an adverse effect on membrane fouling. Therefore in using integrated systems, it is imperative to ascertain that no adverse effect would ensue. Integrated systems may increase the capital cost of the filtration system. Consequently, it is imperative that modern research is focused on optimizing specific pretreatment options for enhancing membranes' permeability.

## **5. Future Research Outlook**

The influence of pre-coagulation on fouling control has clearly been depicted by a number of studies yet, for optimum performance, the influence of coagulation mode (standard or in-line) needs to be explicitly elucidated. Even though some studies have reported in that regard, the subject still remains controversial. It would be interesting if the influence of those two configurations on membrane fouling could be brought to light for optimum performance of

pre-coagulation – low pressure membrane systems. Aside that, another contentious subject is the influence of PAC on fouling control. Research combining different types of PAC under varying dosage, using different membranes and water sources may provide an insight into the contentious performance report by various studies.

Although some researchers have suggested that bio-filtration prior to membrane filtration is efficient in controlling membrane fouling, research on such system is very minimal. However, such systems may become particularly important in providing high quality drinking water since they are environmentally friendly and have no risk of residual chemicals in the treated water. Hence future studies should focus on optimizing such systems especially on full scale for better assessment of the systems' performance.

## **6. Conclusions**

This paper reviewed the recent scientific knowledge on pretreatment techniques for alleviating membrane fouling. It looked at the concept and mechanism of fouling as well as the effectiveness of techniques such as coagulation, adsorption, oxidation, bio-filtration, and others in controlling membrane fouling. In addition, it also discussed the applicability of the various pretreatment techniques in low and middle income countries. The review showed that different pretreatment technique have distinguished effects on membrane fouling and the permeate flux. The selection of a particular pretreatment technique is therefore dependent of the raw water quality and the purpose of treatment. Pertaining to the evidence gathered from literature, the following are the key conclusions on different pretreatment techniques:

- Coagulation pretreatment limits membrane fouling and improves permeate quality by enhancing the aggregation of waterborne contaminants for rejection via low pressure membrane filtration. It remains the most dominant pretreatment technique in literature

probable due to its low cost, ease of operation and ability to control fouling. The impact of coagulation on fouling control however depends on the type of coagulant (Al and Fe salts, organic and inorganic), dosage used, mode of dosing (standard or in-line), raw water quality, membrane material, flow configuration (CFF or DEF) and the mode of operation (constant pressure or constant flux). For raw water with high humic NOM fraction, standard coagulation may be effective in controlling fouling but for non-humic NOM fraction, in-line coagulation should be considered. Hydrophobic membranes are easily fouled by hydrophobic NOM fraction. Therefore for optimum performance of the combined coagulation – low pressure membrane process, the choice of a particular membrane should be made in consideration with the feed water quality and the ability of the coagulation process to remove either hydrophobic or hydrophilic NOM.

- Even though PAC pretreatment results in high NOM removal, its influence of membrane fouling depends on the type, dosage and size of PAC used, the membrane's properties, the feed water composition and the mode of operation. Usually, increasing the contact time between PAC and water by using low operating flux could limit membrane fouling. Also, using high PAC doses may limit membrane fouling however, depending on the raw water quality, process configuration and the mode of operation. When operated in the crossflow mode, avoiding contact between membrane surface and PAC by implementing a separation step could help limit the loss of membrane integrity via abrasion.
- Oxidants regulate membrane fouling by altering the interactions between membrane surface and components of the solution. Although oxidants can limit membrane fouling, their propensity to form health treating precursors could hinder their use in drinking water treatment. Aside that, ozone could have adverse effect on polymeric membranes.

- Due to the escalating levels of water pollution in many developing countries, bio-filtration prior to MF/UF is seen as a suitable option for small scale water treatment plants. Although study on bio-filtration had previously focused on wastewater treatment, recent evidence indicates that such system could be used for portable water treatment as well. Yet, further studies are required in assessing their performance
- Although some studies reported that MIEX<sup>®</sup> have substantial impact on fouling abatement, others researchers reported contradictory results. The contention on MIEX<sup>®</sup> performance could be due to the variation in NOM properties reported by different researches. It appears MIEX<sup>®</sup> works better with feed water having high hydrophilic NOM fraction and may not perform well with feed water containing high hydrophobic NOM. Aside that, the main limitation to that usage of MIEX<sup>®</sup> in developing region, especially sub-Saharan Africa is the high economic implication associated with them.
- The efficacy of integrated pretreatment systems vary based on the pretreatment options combined. Hence a proper understanding of the possible reactions between different pretreatments techniques is required before such systems are implemented

The findings of this review is of particular importance to sub-Saharan Africa and other developing regions. Even though the initial cost of low pressure membranes have decreased over time, the high operational and maintenance costs caused by fouling has hampered the adoption of this technology for drinking water production in these regions. However, with the appropriate pretreatment technique, prolonged membrane filtration can be done at minimized operational and maintenance costs, within the financial capabilities of developing regions. This review will therefore help alleviating drinking water crisis in developing regions by ensuring the extensive adoption of the low pressure membrane technology for high quality, sustainable, effective, low cost and socially acceptable drinking water supply.

## Acknowledgments

The study was supported by the Regional Universities Forum for Capacity Building in Agriculture (REFORUM) under the grant: RU 2015 FAPA 063. The authors also wish to express their gratitude to the European Union for the Mobility to Enhance Training of Engineering Graduates in Africa (METEGA) project. The support of Emmanuel Mensah is duly acknowledged.

## References

1. Prüss-Ustün A, Bartram J, Clasen T, Colford JM, Cumming O, Curtis V, et al. Burden of disease from inadequate water, sanitation and hygiene in low- and middle-income settings: A retrospective analysis of data from 145 countries. *Trop. Med. Int. Heal.* 2014;19:894-905.
2. WHO/UNICEF Joint Monitoring Programme for Water Supply and Sanitation. Progress on Sanitation and Drinking-Water: 2015 Update and MDG Assessment. Geneva, Switzerland; 2015. p. 90.
3. Arnal JM, Garcia-Fayos B, Verdu G, Lora J. Ultrafiltration as an alternative membrane technology to obtain safe drinking water from surface water: 10 years of experience on the scope of the AQUAPOT project. *Desalination* 2009;248:34-41.
4. Bhavnani D, Goldstick JE, Cevallos W, Trueba G, Eisenberg JNS. Impact of rainfall on diarrheal disease risk associated with unimproved water and sanitation. *Am. J. Trop. Med. Hyg.* 2014;90:705-11.
5. UNICEF/WHO. Diarrhoea: Why children are still dying and what can be done? Geneva, Switzerland: WHO Press; 2009. p. 68.

6. Li M, Wu G, Guan Y, Zhang X. Treatment of river water by a hybrid coagulation and ceramic membrane process. *Desalination* 2011;280:114-119.
7. Choi YH, Nason JA, Kweon JH. Effects of aluminum hydrolysis products and natural organic matter on nanofiltration fouling with PACl coagulation pretreatment. *Sep. Purif. Technol.* 2013;120:78-85.
8. Zularisam AW, Ismail AF, Salim R. Behaviours of natural organic matter in membrane filtration for surface water treatment - a review. *Desalination* 2006;194:211-231.
9. Xiangli Q, Zhenjia Z, Nongcun W, Wee V, Low M, Loh CS, et al. Coagulation pretreatment for a large-scale ultrafiltration process treating water from the Taihu River. *Desalination* 2008;230:305-313.
10. Zularisam AW, Ismail AF, Salim MR, Sakinah M, Matsuura T. Application of coagulation - ultrafiltration hybrid process for drinking water treatment: Optimization of operating conditions using experimental design. *Sep. Purif. Technol.* 2009;65:193-210.
11. Jeong CH, Postigo C, Richardson SD, Simmons JE, Kimura SY, Marin BJ, et al. Occurrence and Comparative Toxicity of Haloacetaldehyde Disinfection Byproducts in Drinking Water. *Environ. Sci. Technol.* 2015;49:13749-1359.
12. Chon K, Kim SJ, Moon J, Cho J. Combined coagulation-disk filtration process as a pretreatment of ultrafiltration and reverse osmosis membrane for wastewater reclamation: An autopsy study of a pilot plant. *Water Res.* 2012;46:1803-1816.
13. Gamage NP, Chellam S. Aluminum electrocoagulation pretreatment reduces fouling during surface water microfiltration. *J. Memb. Sci.* 2011;379:97-105.
14. Hill VR, Kahler AM, Jothikumar N, Johnson TB, Hahn D, Cromeans TL. Multistate evaluation of an ultrafiltration-based procedure for simultaneous recovery of enteric microbes in 100-liter tap water samples. *Appl. Environ. Microbiol.* 2007;73:4218-4225.

15. Kearns EA, Magaña S, Lim D V. Automated concentration and recovery of micro-organisms from drinking water using dead-end ultrafiltration. *J. Appl. Microbiol.* 2008;105:432-442.
16. Lai C, Chou Y, Yeh H. Assessing the interaction effects of coagulation pretreatment and membrane material on UF fouling control using HPSEC combined with peak-fitting. *J. Memb. Sci.* 2015;474:207-214.
17. Gao W, Liang H, Ma J, Han M, Chen Z, Han Z, et al. Membrane fouling control in ultrafiltration technology for drinking water production: A review. *Desalination* 2011;272:1-8.
18. Hill VR, Polaczyk AL, Hahn D, Narayanan J, Cromeans TL, Roberts JM, et al. Development of a rapid method for simultaneous recovery of diverse microbes in drinking water by ultrafiltration with sodium polyphosphate and surfactants. *Appl. Environ. Microbiol.* 2005;71:6878-6884.
19. Xu H, Chen W, Xiao H, Hu X. Stability of an ultrafiltration system for drinking water treatment, using chlorine for fouling control. *Desalination* 2014;336:187-195.
20. Her N, Amy G, Plottu-Pecheux A, Yoon Y. Identification of nanofiltration membrane foulants. *Water Res.* 2007;41:3936-3947.
21. Rosas I, Collado S, Gutiérrez A, Díaz M. Fouling mechanisms of *Pseudomonas putida* on PES microfiltration membranes. *J. Memb. Sci.* 2014;465:27-33.
22. Peiris RH, Hallé C, Budman H, Moresoli C, Peldszus S, Huck PM, et al. Identifying fouling events in a membrane-based drinking water treatment process using principal component analysis of fluorescence excitation-emission matrices. *Water Res.* 2010; 44:185-194.
23. Pezeshk N, Narbaitz RM. More fouling resistant modified PVDF ultrafiltration membranes for water treatment. *Desalination* 2012;287:247-254.

24. Huang H, Schwab K. Critical Review Pretreatment for Low Pressure Membranes in Water Treatment : A Review. *Environ. Sci. Technol.* 2009;43:3011-3019.
25. Li NN, Fane AG, Ho WSW, Matsuura T. Advanced Membrane Technology and Applications. John Wiley & Sons, Hoboken, New Jersey; 2008. p. 989.
26. Munla L, Peldszus S, Huck PM. Reversible and irreversible fouling of ultrafiltration ceramic membranes by model solutions. *J. Am. Water Works Assoc.* 2012;104.
27. Ahmadiannamini P, Bruening ML, Tarabara V V. Sacrificial polyelectrolyte multilayer coatings as an approach to membrane fouling control: Disassembly and regeneration mechanisms. *J. Memb. Sci.* 2015;491:149-158.
28. Wang L, Wang X, Fukushi K. Effects of operational conditions on ultrafiltration membrane fouling. *Desalination.* 2008;229:181-191.
29. Zularisam AW, Ismail AF, Salim MR, Sakinah M, Ozaki H. The effects of natural organic matter (NOM) fractions on fouling characteristics and flux recovery of ultrafiltration membranes. *Desalination.* 2007;212:191-208.
30. Lee EK, Chen V, Fane a. G. Natural organic matter (NOM) fouling in low pressure membrane filtration - effect of membranes and operation modes. *Desalination* 2008;218:257-70.
31. Cui X, Choo K. Natural Organic Matter Removal and Fouling Control in Low-Pressure Membrane Filtration for Water Treatment. *Environ. Eng. Res.* 2014;19:1-8.
32. Gray SR, Ritchie CB, Tran T, Bolto BA. Effect of NOM characteristics and membrane type on microfiltration performance. *Water Res.* 2007;41:3833-3841.
33. Campinas M, Rosa MJ. Assessing PAC contribution to the NOM fouling control in PAC/UF systems. *Water Res.* 2010;44:1636-1644.
34. Zhao Y, Song L, Ong SL. Fouling behavior and foulant characteristics of reverse

- osmosis membranes for treated secondary effluent reclamation. *J. Memb. Sci.* 2010;349:65-74.
35. Stoquart C, Servais P, Bérubé PR, Barbeau B. Hybrid Membrane Processes using activated carbon treatment for drinking water: A review. *J. Memb. Sci.* 2012;411-412:1-12.
36. Koo CH, Mohammad AW, Suja' F, Meor Talib MZ. Review of the effect of selected physicochemical factors on membrane fouling propensity based on fouling indices. *Desalination* 2012;287:167-177.
37. Chen F, Peldszus S, Peiris RH, Ruhl AS, Mehrez R, Jekel M, et al. Pilot-scale investigation of drinking water ultrafiltration membrane fouling rates using advanced data analysis techniques. *Water Res.* 2014;48:508-518.
38. Yamamura H, Kimura K, Watanabe Y. Mechanism involved in the evolution of physically irreversible fouling in microfiltration and ultrafiltration membranes used for drinking water treatment. *Environ Sci. Technol.* 2007;41:6789-6794.
39. Tian JY, Ernst M, Cui F, Jekel M. Correlations of relevant membrane foulants with UF membrane fouling in different waters. *Water Res.* 2013;47:1218-1228.
40. Kimura K, Tanaka K, Watanabe Y. Microfiltration of different surface waters with/without coagulation: Clear correlations between membrane fouling and hydrophilic biopolymers. *Water Res.* 2014;49:434-443.
41. Crittenden JC, Trussell RR, Hand DW, Howe KJ, Tchobanoglous G. *MWH's Water Treatment Principles and Design*. Third Edit. New Jersey: John Wiley & Sons, Inc; 2012.
42. Kaya Y, Barlas H, Arayici S. Evaluation of fouling mechanisms in the nanofiltration of solutions with high anionic and nonionic surfactant contents using a resistance-in-series model. *J. Memb. Sci.* 2011;367:45-54.
43. Meng F, Zhang H, Yang F, Li Y, Xiao J, Zhang X. Effect of filamentous bacteria on membrane fouling in submerged membrane bioreactor. *J. Memb. Sci.* 2006;272:161-168.

44. Bergamasco R, Konradt-Moraes LC, Vieira MF, Fagundes-Klen MR, Vieira AMS. Performance of a coagulation-ultrafiltration hybrid process for water supply treatment. *Chem. Eng. J.* 2011;166:483-489.
45. Rai P, Rai C, Majumdar GC, Dasgupta S, De S. Resistance in series model for ultrafiltration of mosambi (*Citrus sinensis* (L.) Osbeck) juice in a stirred continuous mode. *J. Memb. Sci.* 2006;283:116-122.
46. Duclos-Orsello C, Li W, Ho CC. A three mechanism model to describe fouling of microfiltration membranes. *J. Memb. Sci.* 2006;280:856-866.
47. Fabris R, Lee EK, Chow CWK, Chen V, Drikas M. Pre-treatments to reduce fouling of low pressure micro-filtration (MF) membranes. *J. Memb. Sci.* 2007;289:231-240.
48. Kabsch-Korbutowicz M. Ultrafiltration as a method of separation of natural organic matter from water. *Mater. Sci.* 2008;26.
49. Fiksdal L, Leiknes T. The effect of coagulation with MF/UF membrane filtration for the removal of virus in drinking water. *J. Memb. Sci.* 2006;279:364-371.
50. Howe KJ, Clark MM. Effect of coagulation pretreatment on membrane filtration performance. *J. Am. Water Work Assoc.* 2006;98:133-146.
51. Jung J, Kim Y, Park Y, Lee S, Kim D. Optimization of coagulation conditions for pretreatment of microfiltration process using response surface methodology. *Environ. Eng. Res.* 2015;20:223-229.
52. Kabsch-Korbutowicz M. Removal of natural organic matter from water by in-line coagulation/ultrafiltration process. *Desalination.* 2006;200:421-423.
53. Lee BB, Choo KH, Chang D, Choi SJ. Optimizing the coagulant dose to control membrane fouling in combined coagulation/ultrafiltration systems for textile wastewater

reclamation. *Chem. Eng. J.* 2009;155:101-107.

54. Hatt JW, Germain E, Judd SJ. Precoagulation-microfiltration for wastewater reuse. *Water Res.* 2011;45:6471-6478.

55. Matsushita T, Shirasaki N, Tatsuki Y, Matsui Y. Investigating norovirus removal by microfiltration, ultrafiltration, and precoagulation-microfiltration processes using recombinant norovirus virus-like particles and real-time immuno-PCR. *Water Res.* 2013;47:5819-5827.

56. Kim H-C. Microfiltration of humic-rich water coagulated with cationic polymer: The effects of particle characteristics on the membrane performance. *J. Memb. Sci.* 2015;475:349-356.

57. Wan M-W, Yang H-L, Chang C-H, Reguyal F, Kan C-C. Fouling Elimination of PTFE Membrane under Precoagulation Process Combined with Ultrasound Irradiation. *J. Environ. Eng.* 2012;138:337-343.

58. Dong BZ, Chen Y, Gao NY, Fan JC. Effect of coagulation pretreatment on the fouling of ultrafiltration membrane. *J. Environ. Sci.* 2007;19:278-283.

59. Kim H-C, Hong JH, Lee S. Fouling of microfiltration membranes by natural organic matter after coagulation treatment: A comparison of different initial mixing conditions. *J. Memb. Sci.* 2006;283:266-272.

60. Kim H-C, Lee S-J, Byun S., Y M. Application of improved rapid mixing for enhanced removal of dissolved organic matter and DBPFP (disinfection by-product formation potential) control. *Water Sci. Technol. Water Supply* 2006;6:49-57.

61. Park P, Lee C, Choib S, Choo K, Kimd S, Yoone C. Effect of the removal of DOMs on the performance of a coagulation-UF membrane system for drinking water production. *Desalination* 2002;145:237-245.

62. Miller DJ, Kasemset S, Wang L, Paul DR, Freeman BD. Constant flux cross flow

filtration evaluation of surface-modified fouling-resistant membranes. *J. Memb. Sci.* 2014;452:171-183.

63. Guigui C, Roucha JC, Bonnelyeb V, Aptel P. Impact of coagulation conditions on the in-line coagulation-KF process for drinking water production. *Desalination* 2002;147:95-100.

64. Liu P, Hill VR, Hahn D, Johnson TB, Pan Y, Jothikumar N, et al. Hollow-fiber ultrafiltration for simultaneous recovery of viruses, bacteria and parasites from reclaimed water. *J. Microbiol Methods* 2012;88:155-161.

65. Smith CM, Hill VR. Dead-end hollow-fiber ultrafiltration for recovery of diverse microbes from water. *Appl. Environ. Microbiol.* 2009;75:5284-5289.

66. Liang H, Gong W, Chen J, Li G. Cleaning of fouled ultrafiltration (UF) membrane by algae during reservoir water treatment. *Desalination* 2008;220:267-272.

67. Liu Y, Li X, Yang Y, Liang S. Fouling control of PAC/UF process for treating algal-rich water. *Desalination* 2015;355:75-82.

68. Li K, Qu F, Liang H, Shao S, Han ZS, Chang H, et al. Performance of mesoporous adsorbent resin and powdered activated carbon in mitigating ultrafiltration membrane fouling caused by algal extracellular organic matter. *Desalination* 2014;336:129-137.

69. Gai X-J, Kim H-S. The role of powdered activated carbon in enhancing the performance of membrane systems for water treatment. *Desalination* 2008;225:288-300.

70. Kim H-S, Takizawa S, Ohgaki S. Application of microfiltration systems coupled with powdered activated carbon to river water treatment. *Desalination* 2007;202:271-277.

71. Kim J, Cai Z, Benjamin MM. Effects of adsorbents on membrane fouling by natural organic matter. *J. Memb. Sci.* 2008;310:356-364.

72. Lee JW, Chun JI, Jung HJ, Kwak DH. Comparative Studies on Coagulation and Adsorption as a Pretreatment Method for the Performance Improvement of Submerged MF

Membrane for Secondary Domestic Wastewater Treatment. *Sep. Sci. Technol.* 2005;40:2613-2632.

73. Lee J, Walker HW. Effect of process variables and natural organic matter on removal of microcystin-LR by PAC-UF. *Environ. Sci. Technol.* 2006;40:7336-7342.

74. Haberkamp J, Ruhl AS, Ernst M, Jekel M. Impact of coagulation and adsorption on DOC fractions of secondary effluent and resulting fouling behaviour in ultrafiltration. *Water Res.* 2007;41:3794-3802.

75. Ying Z, Ping G. Effect of powdered activated carbon dosage on retarding membrane fouling in MBR. *Sep. Purif. Technol.* 2006;52:154-160.

76. Torretta V, Urbini G, Raboni M, Copelli S, Viotti P, Luciano A, et al. Effect of powdered activated carbon to reduce fouling in membrane bioreactors: A sustainable solution. Case study. *Sustain.* 2013;5:1501-1509.

77. Saravia F, Naab P, Frimmel FH. Influence of particle size and particle size distribution on membrane-adsorption hybrid systems. *Desalination* 2006;200:446-8.

78. Jia Y, Wang R, Fane AG. Hybrid PAC-submerged membrane system for trace organics removal II: System simulation and application study. *Chem. Eng. J.* 2009;149:42-49.

79. Vigneswaran S, Guo WS, Smith P, Ngo HH. Submerged membrane adsorption hybrid system (SMAHS): process control and optimization of operating parameters. *Desalination* 2007;202:392-399.

80. Oh H, Yu M, Takizawa S, Ohgaki S. Evaluation of PAC behavior and fouling formation in an integrated PAC - UF membrane for surface water treatment. *Desalination* 2006;192:54-62.

81. M YM, Yuasa A, Ariga K. Removal of a synthetic organic chemical by PAC-UF Systems-I: Theory and modeling. *Water Res.* 2001;35:455-463.

82. Khan MMT, Takizawa S, Lewandowski Z, Jones WL, Camper AK, Katayama H, et al. Membrane fouling due to dynamic particle size changes in the aerated hybrid PAC-MF system. *J. Memb. Sci.* 2011;371:99-107.
83. Fan X, Tao Y, Wang L, Zhang X, Lei Y, Wang Z, et al. Performance of an integrated process combining ozonation with ceramic membrane ultra-filtration for advanced treatment of drinking water. *Desalination* 2014;335:47-54.
84. Zhang X, Guo J, Wang L, Hu J, Zhu J. In situ ozonation to control ceramic membrane fouling in drinking water treatment. *Desalination* 2013;328:1-7.
85. Kim J, Davies SHR, Baumann MJ, Tarabara V V, Masten SJ. Effect of ozone dosage and hydrodynamic conditions on the permeate flux in a hybrid ozonation-ceramic ultrafiltration system treating natural waters. *J. Memb. Sci.* 2008;311:165-172.
86. Van Geluwe S, Braeken L, Van der Bruggen B. Ozone oxidation for the alleviation of membrane fouling by natural organic matter: A review. *Water Res.* 2011;45:3551-3570.
87. Song Y, Dong B, Gao N, Xia S. Huangpu River water treatment by micro filtration with ozone pretreatment. *Desalination* 2010;250:71-75.
88. Wang X, Wang L, Liu Y, Duan W. Ozonation pretreatment for ultrafiltration of the secondary effluent. *J. Memb. Sci.* 2007;287:187-191.
89. Williams MD, Pirbazari M. Membrane bioreactor process for removing biodegradable organic matter from water. *Water Res.* 2007;41:3880-3893.
90. Choo KH, Lee H, Choi SJ. Iron and manganese removal and membrane fouling during UF in conjunction with prechlorination for drinking water treatment. *J. Memb. Sci.* 2005;267:18-26.
91. Lihua S, Ruiping L, Shengji X, Yanling Y, Guibai L. Enhanced As (III) removal with permanganate oxidation, ferric chloride precipitation and sand filtration as pretreatment of

ultrafiltration. *Desalination* 2009;243:122-131.

92. Chaturvedi S, Dave PN. Removal of iron for safe drinking water. *Desalination* 2012;303:1-11.

93. Hallé C, Huck PM, Peldszus S, Haberkamp J, Jekel M. Assessing the performance of biological filtration as pretreatment to low pressure membranes for drinking water. *Environ. Sci. Technol.* 2009;43:3878-3884.

94. Mosqueda-Jimenez DB, Huck PM, Basu OD. Fouling characteristics of an ultrafiltration membrane used in drinking water treatment. *Desalination* 2008;230:79-91.

95. Banadda EN, Kansiime F, Kigobe M, Kizza M, Nhapi I. Landuse-based nonpoint source pollution: A threat to water quality in Murchison Bay, Uganda. *Water Policy* 2009;11:94-105.

96. Beukes LS, Schmidt S. Microbiological Assessment of the Biofilter Matrix Within a Biofiltration system Treating Borehole water in KwaZulu-Natal (South Africa). *Int. J. Environ. Res.* 2015;9:263-272.

97. Maeng M, Choi E, Dockko S. International Biodeterioration & Biodegradation Reduction of organic matter in drinking water using a hybrid system combined with a rock bio filter and membrane in developing countries. *Int. Biodeterior Biodegradation* 2015;102:223-230.

98. Zhang R, Vigneswaran S, Ngo HH, Nguyen H. Magnetic ion exchange (MIEX®) resin as a pre-treatment to a submerged membrane system in the treatment of biologically treated wastewater. *Desalination* 2006;192:296-302.

99. Kitis M, Ilker Harman B, Yigit NO, Beyhan M, Nguyen H, Adams B. The removal of natural organic matter from selected Turkish source waters using magnetic ion exchange resin (MIEX®). *React Funct. Polym.* 2007;67:1495-1504.

100. Ding L, Deng H, Wu C, Han X. Affecting factors, equilibrium, kinetics and thermodynamics of bromide removal from aqueous solutions by MIEX resin. *Chem. Eng. J.* 2012;181-182:360-370.
101. Croue J, Humbert H. Combination of coagulation and ion exchange for the reduction of UF fouling properties of a high DOC content surface water. *Water Res.* 2007;41:3803-3811.
102. Croue J, Humbert H. Performance of selected anion exchange resins for the treatment of a high DOC content surface water. *Water Res.* 2005;39:1699-1708.
103. Filloux E, Gallard H, Croue JP. Identification of effluent organic matter fractions responsible for low-pressure membrane fouling. *Water Res.* 2012;46:5531-5540.
104. Mergen MRD, Jefferson B, Parsons SA, Å PJ. Magnetic ion-exchange resin treatment: Impact of water type and resin use. *Water Res.* 2008;42:1977-1988.
105. Mozia S, Tomaszewska M, Morawski AW. Application of an ozonation-adsorption-ultrafiltration system for surface water treatment. *Desalination* 2006;190:308-314.
106. Watson K, Farré MJ, Knight N. Enhanced coagulation with powdered activated carbon or MIEX® secondary treatment: A comparison of disinfection by-product formation and precursor removal. *Water Res.* 2015;68:454-466.
107. Kim HC. High-rate MIEX filtration for simultaneous removal of phosphorus and membrane foulants from secondary effluent. *Water Res.* 2015;69:40-50.