



## Membrane fouling control in ultrafiltration technology for drinking water production: A review

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### ARTICLE INFO

#### Article history:

Received 16 September 2010

Received in revised form 12 January 2011

Accepted 15 January 2011

Available online 18 February 2011

#### Keywords:

Ultrafiltration

Membrane fouling

Drinking water production

Pretreatment

### ABSTRACT

Ultrafiltration is a promising process to produce qualified drinking water. The application of ultrafiltration for drinking water production has undergone accelerated development during the past decade. Membrane fouling may be the main obstacle for wider implementation of ultrafiltration, which usually causes higher costs of energy, operation, and maintenance. Fouling is formed due to pore blocking, pore stricture and cake formation. Pretreatments (e.g. coagulation, adsorption, and pre-oxidation) can in various degrees alleviate the fouling by pre-reacting with the foulants in the feed water. However, adverse effects from the pretreatment are also claimed. Moreover, modest operation methods (e.g. running modes, rinsing modes, chemical cleaning, and air scouring) can effectively obtain the fouling reduction. In this report, fouling control in ultrafiltration technology for drinking water production is reviewed in terms of different effective pretreatments and operation methods. Specific mechanisms and future research required are also discussed based on the literature reviewed.

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that dissolved organic matter (DOM) gave rise to more severe fouling than the sum of fouling from each independent DOM. It was demonstrated that a possible adverse interaction existed, though the fouling modes of DOM were not totally independent. Moreover, studies of organic fouling are also concentrated on the fractionation methods to investigate which fraction of NOM is responsible for the fouling. Gel permeation chromatography [20–22], XAD series resins [23,24], different pore size membranes [25], dialysis bag [26], and size exclusion chromatography [27,28], are used to fractionate the NOM according to MW, hydrophobicity or other characteristic of the NOM. However, for the different property of the ultrafiltration membrane, NOM fraction presents different influence. The specific effect of the properties of NOM on fouling of ultrafiltration is not yet elucidated due to the complex or unknown speciation in NOM from the natural waters. Limited details about the behavior of NOM are reviewed by Zularisam et al. [29].

- Bio-fouling stems from aquatic organisms, such as algae [30], which can form colonies and then cause bio-fouling. Due to the lack of data (possibly because of the periodic cleaning with chlorine, which may kill the organisms before the fouling happen) with respect to bio-fouling of ultrafiltration, it is unclear what the specific or possible bio-fouling mechanism is.

Hiroshi et al. [31] classified the membrane fouling as physically reversible fouling which can be totally eliminated by physical cleaning or certain pretreatment and physically irreversible fouling which cannot be entirely counteracted by physical cleaning or certain pretreatment [32,33]. The irreversible fouling can explain the gradual increase of membrane filtration resistance after running a long period, although the physical cleaning and effective pretreatment are routinely implemented.

The study of fouling models is significant for better understanding of the fouling mechanism and better predicting of fouling formation, and would provide a useful tool for practical design and operation. For fouling models of low-pressure membranes, related issues have been extensively reviewed by Kim et al. [34].

### 3. Effect of different pretreatment options

Pretreatment can increase the permeate quality and reduce membrane fouling [35]. Currently, there are several common options: coagulation, adsorption, oxidation, MIEEX, biological treatment, and some integrated pretreatments. The efficiency of pretreatment in removing aquatic contaminants and reducing membrane fouling is strongly associated with the type of the agents (coagulant, adsorbent, oxidant, etc.), dosage, dosing modes (continuous or intermittent), dosing point, mixing ways, temperature, properties (hydrophobicity, charge density, molecular weight, and molecular size) of the aquatic impurities (colloidal or dissolved, organic or inorganic), solution environment (solution pH and ion strength), and characteristics of the membrane (membrane charge, hydrophobicity, and surface morphology).

#### 3.1. Coagulation

##### 3.1.1. Mechanisms of coagulation in a coupled process

Coagulation, as a physicochemical pretreatment process, is most widely and successfully used due to its low cost and relatively easy operation. Coagulation combined with UF is a promising process with respect to the removal of contaminants [36], the maintenance of a high membrane performance, and the reduction of subsequent formation of disinfection by-product [4]. Coagulants are lingering between organic macromolecules and inorganic salts. This report here only discusses the application of inorganic coagulants, such as aluminum and ferric salts, which are commonly used in drinking water production. Organic coagulants which are not frequently used in pretreatment for ultrafiltration, therefore, are not discussed in this report.

Mechanisms of inorganic coagulation before ultrafiltration should be recognized as follows: neutralizing the charge and sweeping the flocculants to change the size and to stabilize the impurities to the level that they can be rejected by the membrane followed. The speciation is mainly depended on the pH range of the solution.

Chemical reaction may happen between the negatively-charged colloids or the NOM and positively-charged coagulants when the charge neutralization dominates the mechanism. While the coagulant is over-dosed, a metal hydroxide precipitate (such as aluminum hydroxide or ferric hydroxide) can cause a sweep floc effect, provided that there is enough contact time. However, when the excess cationic coagulants encounter a negatively-charged membrane surface (originally charged or with ionizable functional groups), an effect of compressing the electric double layer on the membrane surface may happen, and this may facilitate the adsorption of NOM on the solid membrane surface. Therefore, the specific coagulation conditions and leading mechanisms are used to interpret the different characteristics of the foulants on the membrane surface.

##### 3.1.2. Contaminant removal and contribution to fouling reduction

Pre-coagulation in the UF system is generally categorized into standard coagulation and in-line coagulation in accordance to the presence and absence of the sedimentation.

Standard coagulation can remove destabilized pollutants or their aggregates. Coagulant hydrolytes/precipitates can also partly be eliminated during the sedimentation process. Liang et al. [37] compared coagulation, coagulation–sedimentation, and coagulation–sedimentation–filtration prior to ultrafiltration applied in drinking water production from algae-rich reservoir water, and found that coagulation–sedimentation was the most effective; however, filter presented an adverse effect. Dong et al. [38] presented an opposing view that in-line coagulation could form an initial floc cake layer on the membrane surface for adsorption of the contribution parts of NOM. The phenomenon suggests that there is probably a kind of aggregate benefiting the following ultrafiltration, which can be settled or filtered during the subsequent conventional process. And Kimura et al. [39] studied the efficacy of pre-coagulation/sedimentation on the control of irreversible fouling in a polysulfone ultrafiltration membrane by a pilot scale experiment in a drinking water production plant, and found that pre-coagulation/sedimentation could not remove fractions of organic substances (such as polysaccharides and protein), which accounted for the irreversible fouling in UF in the experiment.

The properties and specific components of the raw water may account for the different results, but the controversial results bring a new insight into the integrated process. It is that during the whole coagulation process, there must be a proper time, from initiating coagulation to ending sedimentation, to form a kind of special flocs to minimize UF fouling. Additionally, the conventional, though classic, coagulation should be improved, and new coagulation way can be established to reduce membrane fouling, Park et al. [40] used a pre-coating method in both standard coagulation and in-line coagulation, demonstrated that removal of DOM could make the membrane more filterable, and found that 13.0 mg/L  $\text{FeCl}_3$  presented a more enhanced filterability than 4.1 mg/L poly-aluminum chloride due to the formation of a liable permeable cake layer.

Coagulation as a pretreatment process focuses on the enhancement of ultrafiltration performance, and ultrafiltration is known to be different from conventional filtration technologies, so some classical coagulation measures may not work well here. For instance, proper dosage, the type of coagulants, and mixing conditions need to be determined by transforming the view from optimized operation of classical coagulation to that of coagulation/UF systems. In terms of water quality, the settling ability of the flocs may not be as considerable as the size for the membrane filtration; there should be a critical size or sustainable size for the flocs to benefit subsequent ultrafiltration.

The type of coagulant has been identified as a significant factor influencing the fouling in ultrafiltration. Kabsch-Korbutowicz [41] used three types of coagulants in a constant pressure in-line coagulation-UF (dead end filtration) system and found that alum and polyaluminum chloride can increase the removal of organic matter and bring a considerable membrane fouling reduction, with no effect for sodium aluminates. However, different reagents may need different optimum conditions, because coagulation may produce different floc with distinct characteristics in different ways due to the various working conditions. Kim et al. [42] compared different mixing conditions for chemical coagulation and found that back-mixing with an in-line coagulation is useful for NOM fouling reduction.

Optimized dosage of coagulant in a classic coagulation process may not be the best for UF. Howe et al. [43] found that under-dose of coagulant usually had an adverse effect on the fouling of ultrafiltration, while over-dose was always effective for fouling reduction. Coagulation is the most promising pretreatment for UF, and more researches are needed to optimize and establish a particular coagulation way directly aiming at better ultrafiltration.

### 3.2. Adsorption

#### 3.2.1. Mechanisms of adsorption in a coupled process

Adsorbents often have relatively large specific surface areas, resulted from the relatively high dispersity or porosity. Powdered activated carbon (PAC), with a good removal of certain contaminants and a controversial fouling reduction, is the most popular adsorbent when adsorption and ultrafiltration are coupled because of its thermodynamically unstable surface and widely commercial availability. The mechanism mostly studied and discussed about PAC is to supply enough surface areas or interfaces to absorb or accumulate the absorbable impurities, including “bulk solution transport, diffusion through the hydrodynamic boundary layer of fluid adjacent to the surface of the PAC particle or ‘film diffusion’, internal particle diffusion, and adsorptive reactions between the adsorbate and the adsorbent” [44]. Due to the coexistence of solid phase (such as adsorbent, membrane surface, colloid, etc.) and liquid phase (such as water and other dissolved solute), adsorption is involved into a complicated solid–liquid system rather than aquatic surface chemistry.

The chemical interactions between the solute (such as NOM and other impurities in the water), solvent (usually for water), and the surface (water/membrane interface, solute/water interface, and solute/membrane interface) are significant for understanding adsorption in a coupled process. Given enough contact time and an adequate distance between adsorbent and adsorbate, a charge change may happen in the adsorbent, and this would complicate the interactions between the adsorbent and membrane surface.

#### 3.2.2. Contaminant removal and contribution to fouling reduction

There are two combining modes about the coupled adsorption and filtration process, i.e. in an integrated membrane reactor and a separated reactors following a PAC reactor, and two dosing ways, i.e. step input of PAC (dosed to the reactor at a constant rate) and pulse input (all dosed at the beginning of the filtration loop). The effect of the dosing method on the fouling control is less pronounced. PAC can remove organic or inorganic aquatic contaminants from natural waters by adsorption, usually described as three-step transition of the impurities: from water to carbon, then to the carbon surface, lastly to binding sites.

Much work has been done to study the effect of PAC on the membrane fouling of ultrafiltration. Contact time [45], dosage, and dosing methods are known to account for the contaminant removal in the integrated process. Although a high removal of impurities is discovered, no special phenomena about membrane fouling reduction or increase are found comparing with the UF system alone in the same

conditions [46,47]. At proper impurity removal rate and pH condition, the PAC may be glued to the membrane surface to form a cake layer, which can avoid some foulants approaching the surface, and result in a relatively unchanged membrane surface matrices for ultrafiltration.

Interestingly, other reports stated that PAC had a slight effect on the fouling of the ultrafiltration membrane. It was found [48] that the presence of PAC could not only enhance the removal of organic matter, but also obtain a good control of membrane fouling. However, Lee et al. [49] claimed an opposing view that the combined PAC-UF system can contribute a high removal of synthetic and natural organic matter, but the system displays an adverse influence on the flux rate and more humic acids are detected on the following UF membranes. Zhang et al. [50] also reported an inconsistent result of contaminant removal and fouling reduction. The different types of PAC with various properties and doses may be responsible for the different results of literature. And the crux here is whether the adsorbate adsorbed by PAC can get through or into the membrane pore to impose a pore plugging effect on the membrane, which is reported as a more severe fouling cause than cake layer and pore blocking [6]. The impurities which can or cannot be absorbed by PAC but can affirmably or easily adsorb into the membrane held back the use of PAC as a pretreatment for UF.

Therefore, work to modify PAC has been done by Hong et al. [51]. They implemented a process combining surfactant-modified powdered activated carbon (SM-PAC) with ultrafiltration to treat water containing anionic contaminants, and obtained a good performance at different removal efficiency of various anionic impurities. But little information about the membrane fouling was presented. For the uncertain impact of PAC on the membrane fouling, modifying PAC or replacing novel adsorbent to guarantee the contaminant removal and the fouling reduction may be an effective measure for the amphibolous view.

And although PAC is so widely studied, a new adsorbent should be developed for membrane fouling control, e.g. carbon black, which is modified by the manufacture, is demonstrated to be a better selection in river water ultrafiltration than PAC [51].

### 3.3. Preoxidation

Oxidants used in a conventional water treatment plant are usually ozone, permanganate, and chlorine. Oxidants can suppress the growth of microorganisms or change the structure and properties of the NOM and provide a disinfecting environment for the water.

#### 3.3.1. Ozone

In the review of oxidants' compatibility with low-pressure membranes by Farahbakhsh et al. [52], ozone was reported to be not compatible with the most commercially available polymeric UF membranes. Therefore, studies [53–55] were carried out by using the ceramic membranes while combining ozone pre-oxidation and UF. Kim et al. [53] investigated the effect of ozone dosage and hydrodynamic conditions on the performance of the ozonation/ultrafiltration process in natural water treatment. The system demonstrated that ozone dosage, which could bring a permeate flux increase over various operational conditions, presented less fouling when a higher cross-flow velocities and a lower TMP were applied, and with increased ozone dosage, TOC removal increased. Ozone can alter the size and characteristics of the impurities in the water sources, thus influencing the following membrane process. A high removal efficiency of TOC by pre-oxidation was also observed by Schlichter et al. [55] and Mozia et al. [54]. And Schlichter et al. stated that a concentration of 0.05 mg/L could maintain no reduction of membrane flux without any use of membrane backwashing. It is worth mentioning that ozone can increase the bio-degradability [56,57], which is significant while combining MBR for micro-polluted water treatment. However, a significant aspect of ozone rarely announced



while combining with UF is the formation of bromate. With the stricter standards all over the world, the subsequent removal of bromate, if produced, by UF and its effect on the UF membrane permeability are critical.

### 3.3.2. Permanganate and chlorine

Permanganate and chlorine are widely used in developing countries, such as China, for pre-oxidation in a conventional water treatment plant. Despite of their preferential advantage in developing world, research is limited on the combination of permanganate, chlorine, or permanganate and chlorine with ultrafiltration. Chlorine was reported to be able to oxidize inorganic ions, such as manganese [58], and account for a little fouling of UF because of its ability to reduce the size of particulate matter [59]. Liang et al. [60] examined the algae fouling reduction by permanganate and chlorine while treating algae laden reservoir water in a coagulation/UF process, and found that 1 mg/L of chlorine and 0.5 mg/L of permanganate could much effectively reduce the fouling of the UF membrane used. However, not much literature pronounced the specific mechanism of fouling reduction by permanganate and chlorine.

Pre-oxidants are always used to degrade the precursors in the water sources, but with respect to existing and new disinfect by-products (DBP), such as N-nitrosodimethylamine (NDMA) [61], more consideration must be taken into account in the following filtration ability of the DBP, and in investigating how the products impact the membrane filtering and membrane fouling.

### 3.4. Other pretreatment

MIEX, biological treatment, and some integrated pretreatment processes were also studied prior to ultrafiltration as pretreatment options to enhance the membrane performance and reduce fouling.

#### 3.4.1. MIEX

MIEX resins, a three-dimensional structure with polymeric chains, which have electrically charged functional groups located on the chains and different charged ions (counter-ions) attached to the functional groups through electrostatic forces, could remove the ions in the raw water by ion exchange [62], and can exchange weak organic acid ions from a pH around 7.0 due to its strong base functionality [63–65]. Kabsch-Korbutowicz et al. [27] compared the efficacy of coagulation and MIEX resin adsorption as a pretreatment of ultrafiltration, and reported that both of the pretreatment methods could increase the permeate quality and decrease the membrane fouling. Importantly, MIEX resin could remove organic matter more effectively than coagulation due to its ability for the removal of low-molecular fractions, which is claimed by Lee et al. [17] as a main foulant to the UF membrane. However, there is a lack of data about the pilot-scale or large-scale of MIEX as a pretreatment option for ultrafiltration of various source waters, so more studies are needed to better understand the mechanism of MIEX on fouling of ultrafiltration. Besides, due to the higher cost, the research and application of MIEX are more popular in developed countries than developing countries.

#### 3.4.2. Biological treatment

Biological processes are often welcomed in wastewater treatment rather than drinking water production due to the lack of culture in source water for growth of microorganisms. However, with the gradually polluted source water, the concept and application of biological treatment are again valued for drinking water production. Mosqueda-Jimenez et al. [66] examined the effect of bio-filter, as a pretreatment option, on the performance of ultrafiltration applying to the treatment of humic acid rich water, and found that combination of bio-filter presented lower fouling rate than ultrafiltration alone. Cynthiahalle et al. [67] claimed that bio-filter with a longer contact

time can reduce both the hydraulically reversible and irreversible fouling. Except for the separated biological pretreatment, some media are introduced into the membrane tank to integrate the process as an incorporate technology. Tian et al. [9,10] introduced coagulant and adsorbent directly into the submerged ultrafiltration reactor for treating simulated source water contaminated by sewage. Both the membrane coagulation bioreactor (MBCR) and membrane adsorption bioreactor (MABR) showed high removal efficiency of TOC, COD<sub>Mn</sub>, DOC and UV<sub>254</sub>, as well as disinfection by-product formation potential. However, no corresponding information of the membrane fouling was discussed.

Additional research with respect to the mechanism of biological treatment on the UF membrane fouling in the drinking water production is necessary to better evaluate the feasibility of biological-pretreatment coupled with ultrafiltration, especially on a large-scale plant.

#### 3.4.3. Integrated pretreatment

Integrated pretreatment means utilizing the advantages of each pretreatment to theoretically or practically enhance the membrane performance and control the membrane fouling, to supplement each other's disadvantages. But in practice, integrated pretreatment could present an adverse effect on the membrane fouling. Mozia et al. [56] studied an ozonation-adsorption-ultrafiltration system by treating a mixture containing humic acids and phenol, and found that the addition of PAC increased the fouling of the regenerated cellulose UF membrane comparing with UF alone. Therefore, when an integrated pretreatment is introduced, it must guarantee no adverse influence between the different pretreatment options being used. For the removal of impurities, there is probably some common ground among the various pretreatment options, designers have to avoid wasting and to optimize the increasing part and decreasing part of the cost due to the excessive pretreatment and better membrane permeability, respectively.

## 4. Fouling control through operational conditions

In addition to the pretreatments (e.g. coagulation, adsorption, and pre-oxidation), it is also important that the different operation conditions can reduce membrane fouling to some extent. As previously reported, fouling is formed because of the impurities rejected by or adsorbed on the membrane surface, and some complicated interactions then may happen between the impurities and the membrane surface. Therefore, theoretically, there should be a reasonable way to apply some proper operation or cleaning strategies to inhibit the complicated reaction before it happens, and their combined benefits could be an ideal way to control or reduce the membrane fouling. The strategy must be based on a better understanding of the mechanism of membrane fouling when ultrafiltration is applied for drinking water production. And the control of different fouling mechanism or foulants in the ultrafiltration process may need different running and cleaning strategies.

### 4.1. Running modes

Running modes, which can influence the rate and extent of membrane fouling, here discussed, involve different permeate fluxes [66,68], backwashing frequency [69], and constant pressure or constant flux [17,19]. Change of running modes may be very practical for the membrane fouling control in the pilot-scale or large-scale application. A relatively low flux is bound to bring a lower rate of fouling, but more membrane required would increase the building and operating cost. A proper backwashing frequency is significant for the better operation of ultrafiltration. According to a more-than-two-years operating experience in Germany, Lipp et al. [69] suggested that it was better to manage backwashing in terms of filtered volume than



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