



Insight into the combined coagulation-ultrafiltration process: The role of Al species of polyaluminum chlorides



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ABSTRACT

This research focused on the influence of molecular weight (MW) fraction of humic acid (HA) on ultrafiltration membrane (MW cut off of 100 kDa) fouling and further investigated the impact of Al species of polyaluminum chlorides (PACls) on coagulation-ultrafiltration process. Results indicated that MW fraction of 30–50 kDa generated the most severe membrane fouling. On the basis of experiment results in this research and other literatures, we proposed that reversible fouling and irreversible fouling was largely associated with floc properties and residual HA in coagulated water, respectively. This could fully explain the influence of different PACls, PACla (with high monomeric species), PAClb (with high medium polymer species) and PAClc (with slightly higher colloidal or solid species), on membrane fouling. After floc breakage and re-growth, floc size followed the order of PACla > PAClc > PAClb and the sequence of floc fractal dimension was as follows: PAClb > PACla > PAClc. Reversible fouling of PACls was consistent with floc fractal dimension, which was directly correlated with cake layer resistance. In coagulated water, HA with MW more than 50 kDa had similar removal for PACls, whereas HA with MW less than 50 kDa achieved the highest removal efficiency for PAClb, followed by PAClc and PACla.

1. Introduction

Membrane filtration has been one recognized water treatment technology in recent years due to effective improvement on filtered water quality [1–4]. However, a major drawback regarding membrane filtration technology is membrane fouling [5–7]. Therefore, research on how to control membrane fouling receives more attention [8,9]. Numerous studies indicate that natural organic matter (NOM) is the dominant foulant for membrane [10–12]. A small extent of NOM interception by membrane generating significant membrane fouling urge investigators to explore membrane fouling mechanism of NOM. Fan et al. found that high molecular weight (MW) fraction of NOM (> 30 kDa) contributed to the major membrane fouling regardless of the NOM characteristics [13]. Lin et al. also indicated that the largest MW fraction led to the worst flux decline for both hydrophobic and hydrophilic fractions [1]. Thus, it is likely MW of NOM has a profound impact on membrane fouling. Humic acid (HA) is the major component of NOM in the well-protected natural water [14,15]. The influence of MW fraction of HA on ultrafiltration (UF) fouling is still unclear. Thus, it is attractive to ensure the UF membrane fouling of each MW fraction of HA.

To effectively alleviate UF membrane fouling, various approaches are explored, in particular, pretreatments to reduce foulant concentration can enhance the removal of HA and generate large flocs [17,18]. Therefore, coagulation–ultrafiltration (C-UF) hybrid process, i.e., coagulation without sedimentation is used before UF, can significantly mitigate membrane fouling due to lower HA concentration before UF and cake layer induced by flocs [19–21]. Since coagulation parameters play more important role than UF parameters on membrane fouling, more scholars have focused on the effect of coagulation conditions on membrane fouling [22,23].

Aluminum salts, in particular polyaluminum chloride (PACl), have been widely applied in coagulation. Due to the limitations of measurement methods, Al species of aluminum salts is generally divided into three categories: monomeric species (Al_a), medium polymer species (Al_b), and colloidal or solid species (Al_c) [24]. For the combined process of coagulation and membrane filtration, various NOM removal efficiency and floc properties induced by different Al species must result in varying membrane fouling. Thus, some researchers turned their attentions to the influence of Al species on the combined process of coagulation and membrane filtration. Zhao et al. claimed that Al

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species had a profound influence on floc structure and thus led to different membrane flux decline [25]. Xu et al. found that Al_b generated the smallest flocs with the highest density, leading to maximum membrane fouling in all Al species [26].

Considering inevitable floc breakage and re-growth due to prevalent high shear regions in practice, the influence of Al species on floc properties and HA removal efficiency after floc breakage and re-growth may give more indications for actual C-UF hybrid process [27,28]. Feng et al. proposed that after floc breakage and re-growth Al_b was superior to Al_a and Al_c in the decrease of membrane fouling due to higher floc strength [29]. However, only part recovery of membrane fouling by hydraulic cleaning indicates that membrane fouling includes both reversible fouling and irreversible fouling. Humic substances including humic acid and fulvic acid is strong related to irreversible fouling, whereas particulate matter is likely the main foulant to cause reversible fouling [30]. That is, it is reasonable to infer that floc properties should be mainly responsible for reversible fouling instead of total fouling. Therefore, relation between Al species and C-UF process (with floc breakage and re-growth) should be further investigated to access the role of Al species on reversible fouling and irreversible fouling.

The aim of this research was to explore the influence of each MW fraction of HA on UF membrane fouling. Furthermore, with floc breakage and re-growth, the role of Al species in C-UF process was thoroughly investigated to reveal the relation between Al species and membrane fouling by floc properties and HA removal during coagulation.

2. Material and methods

2.1. Suspensions

Humic acid solution was prepared by using a similar way to our previous work [12]. 10 g of HA (Shanghai, China) was dissolved in 0.1 mol/L NaOH and stirred for 24 h. Then HA solution was filtered using 0.45 μ m fiber filter membrane and adjusted pH to 7.5 by addition of 0.01 mol/L HCl or NaOH, followed by diluted to 1 L in a flask. The original HA solution (denoted as HA_0) was prepared by diluting HA solution mentioned above in tap water (Harbin, China), to give a dissolved organic carbon (DOC) concentration of 2.25 mg/L and UV_{254} absorbance of 0.200 cm^{-1} . To determine MW distribution of HA_0 , UF membranes (Mosu, China) with MW cut-offs of 50, 30 and 10 kDa were applied to fractionate HA_0 solution sequentially since HA_0 mainly contained high molecular weight components [13,31]. The total dissolved carbon (TOC) of effluent of each membrane was measured. Subsequently, on the basis of HA_0 solution, four synthetic HA solutions were prepared by adding different volume of each MW component (obtained by UF fractionation). The DOC concentration of five HA solutions were fixed at 2.25 mg/L. The final UV_{254} absorbance of five HA solutions were around 0.200 cm^{-1} . Four synthetic HA solution was denoted as HA_1 , HA_2 , HA_3 and HA_4 , respectively. The MW distribution characteristics of HA solutions are summarized in Table 1.

Kaolin suspension was prepared by dispersing 100 g of kaolin clay in 1 L ultrapure water with high-speed magnetic mixing for 24 h, followed by 1-h sedimentation. Then the top 800 mL was decanted as stock suspension of kaolin clay. This suspension was diluted to give the

Table 1
Molecular weight distributions of HA solutions.

Sample	< 10 k (%)	10–30 k (%)	30–50 k (%)	> 50 k (%)
HA_0	34.87	11.13	13.80	40.20
HA_1	17.97	11.13	30.70	40.20
HA_2	17.97	28.03	13.80	40.20
HA_3	34.87	11.13	30.70	23.30
HA_4	34.87	28.03	13.80	23.30

final kaolin concentration of 95 mg/L.

For the flocculation tests, HA_0 solution and kaolin solution was added into the tap water (Harbin, China) to achieve the raw water. The raw water was put aside for long hours to obtain adsorption balance [32]. The final suspension had a kaolin clay concentration of 95 mg/L, humic acid concentration of 5 mg/L and pH around 7.2.

2.2. Coagulants

PACls with various basicity and production methods were used as coagulants in this work. The conventional $AlCl_3$ (Tianjin, China), which was directly dissolved in deionized water, was considered as PACla. PAClb was prepared by the alkali titration method in laboratory at room temperature [33]. A predetermined amount of 0.2 mol/L $AlCl_3$ solution was transferred into a 300-mL glass reactor. With rapid stirring, 0.5 mol/L NaOH was titrated into $AlCl_3$ solution at 0.1 mL/min by a peristaltic pump (BT100-2J, Longer CO., China). The amount of NaOH varied with the target B value (OH/Al ratio), i.e., 2 in this work. PAClc (Tianjin, China) was a commercial PACl product containing 28% of Al_2O_3 . The sample of each PACl was measured by Ferron assay after aging for one week. The results of Al species distribution of PACls are shown in Table 2. All the reagents used in this study were of analytical grade.

2.3. Apparatus

The C-UF combined process includes coagulation process and UF process. The schematic diagram of C-UF process is shown in Fig. 1. For coagulation process, the reactor was a modified version of jar-test apparatus, which was a rectangular stirred tank (5 mm thick plexiglas) with the length of bottom $D=150$ mm and the height of liquid $H=D=150$ mm, and filled with 3.4 L of testing water sample. Moreover, a R1342-type impeller (IKA, Germany) with a diameter $d=50$ mm was applied to provide stirring condition and the center of the impeller was positioned at $C=H/3$ from the tank bottom. This mixing system was successfully used in our previous researches [34,35].

To calculate floc properties, a digital charged coupled device (CCD) camera (SVS-VISTEK GmbH, Germany) was used to capture digital images of aggregates. Then a process control and image processing (PCIP) software package (FMans 10, China) was applied to measure geometrical parameters of flocs. The in-situ recognition system including digital CCD camera and PCIP software package has been widely used by our lab and other researchers since there is no input and output of samples [12,36,37].

For UF process, a dead-end UF setup with a 400 mL cylindrical beaker (Amicon 8400, Millipore, US), was used in this research. Commercial polyethersulfone (PES) UF membranes (Mosu, China) with effective membrane area of 45.34 cm^2 and MW cut-offs of 100, 50, 30 and 10 kDa were applied during UF process. The UF membrane with MW cut-off of 100 kDa was used to determine the membrane fouling during UF and C-UF process. While UF membranes with MW cut-offs of 50, 30 and 10 kDa were applied to determine the removal of each MW fraction of HA during coagulation. Before each UF test, the fresh membrane has been “wetted” to optimal operating condition. In addition, nitrogen gas was applied to supply constant pressure at 100 kPa. Cumulative permeate mass was determined via electronic

Table 2
Characteristics of PACls by Ferron method.

Coagulant	pH	Ala (%)	Alb (%)	Alc (%)
PACla	2.71	92.4	7.6	0
PAClb	4.03	12.3	79.5	8.2
PAClc	4.50	26.7	32.4	40.9

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