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Reuse of PAC and alum sludge (RPAS) process: pretreatment to reduce membrane fouling

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ABSTRACT

In this study, a novel reuse of powdered activated carbon (PAC) and alum sludge (RPAS) process to reduce membrane fouling for drinking water treatment was evaluated by laboratory-scale experiments. As compared to coagulation, PAC combined with coagulation and reuse of alum sludge processes, RPAS process performed better in controlling trans-membrane pressure increase and reducing membrane resistances in both short-term and long-term periodical filtration. Removals of turbidity, DOC, UV_{254}, BDOC and THMFP were 93.8, 37.3, 41.1, 83.0 and 57.9% on average by RPAS pretreatment, respectively. The results of fractionation of organic matters indicated that hydrophobic acids and hydrophilic matters were efficiently removed, while organic matters with the molecular weight (MW) more than 3 k Da was well removed with a removal efficiency of 50.4%, and that of MW less than 1 k Da were reduced by 32.6% by RPAS pretreatment, which could be attributed to the integration of PAC adsorption and the enhanced coagulation of alum sludge.

Keywords: Membrane fouling; Pretreatment; Reuse; Alum sludge; PAC; Organic matters

1. Introduction

One of the critical problems encountered during the membrane process in drinking water treatment is membrane fouling. Membrane fouling usually describes the loss of membrane hydraulic permeability due to the accumulation of aquatic particles and organics on the membrane surface during the filtration process, which results in reduction of the productivity from the membrane process and ultimately increases the cost of operation [1,2]. Therefore, a strategy of membrane fouling control should be preceded for efficient and economical operation of membrane processes.

Pretreatment to lower NOM in the feed has been a useful approach to prevent fouling, such as coagulation, adsorption and ozonation before membranes, which has been used to remove organic matters and to mitigate membrane fouling [3–6]. Coagulation is more widely applied and investigated due to its low cost and easy to use. A few of investigations showed that coagulation could improve flux indeed [7–9]. However, some research work indicated that although coagulation could remove organic matters and decrease membrane filtration resistance, the rate and extent of fouling could not be alleviated by coagulation [10]. Integration between clear size exclusion of UF membranes and adsorption of powdered activated carbon (PAC) could be a retrofit technology in conventional water treatment process [11]. PAC could

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improve membrane productivity by reduction of foulant load toward the applied membranes [12,13]. Meanwhile, some studies reported that PAC addition resulted in more severe flux decline in the integrated PAC-UF membrane [14,15].

Reuse of the alum sludge from water and waste water treatment may not only improve the organic matters removal of a primary sewage treatment but also ease the burden of water treatment works relating to sludge treatment and disposal [16], and the alum sludge could be reused to improve the coagulation of low turbidity water for drinking water treatment [17,18]. When PAC was applied in a conventional process of water treatment plant, the retention time of PAC may be limited to only 10–20 min [19], which may be too short for an adsorption equilibrium to be reached, so reuse of PAC and alum sludge (RPAS) process was investigated by the authors. The removals of turbidity, particulates, DOC and UV_{254} for the raw water were all better than coagulation, reuse of alum sludge and PAC preadsorption process [20]. Particulates and organic matters were well removed by RPAS process, which could possibly have a positive effect on reducing membrane fouling. Therefore, the main objective of this study is to systematically investigate the membrane fouling prevented by RPAS pretreatment, and analyse the inherent mechanism. This work is expected to propose a more effective pretreatment to prevent membrane fouling for drinking water treatment.

2. Materials and methods

2.1. Experimental set-up

Laboratory-scale immersed UF was employed in this study. A schematic illustration of the experimental set-up is shown in Fig. 1. UF membrane modules were made of polyvinyl chloride (PVC) with a nominal pore size of 0.01 μm. The characteristics of UF membrane are shown in Table 1. The effluent was drawn directly from the membrane module by using a peristaltic pump, which was also applied to do backwash with a flux of 60 L/m^2 h. A pressure sensor was set between the membrane module and the suction pump to monitor and record TMP automatically by software. Aeration was provided at the bottom of the membrane tank to reduce fouling with a flux of 45 m^3/m^2 h. UF filtration was operated at a constant flux of 30 L/m^2 h and change of TMP was record to express the fouling. A filtration period of 48 h was applied and a combined membrane cleaning of backwashing and aeration with a cleaning time of 10 min was used.

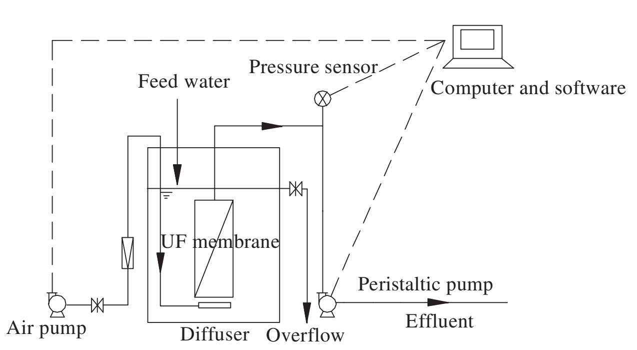

Fig. 1. Schematic diagram of the experimental set-up.

Table 1
The physical characteristics of the UF membranes

Parameters	Characteristics
Material	PVC
Type	Hollow fiber
Filtration mode	External pressure
Weight cutoff (kDa)	100
Contact angle (°)	67.0
Inner diameter (mm)	0.85
Outside diameter (mm)	1.45

2.2. Experiment conditions

Pretreatment experiments were performed with jar tests, and polymer aluminum chloride (Actview Carbon Technology Inc., China) was used as coagulant with an addition of 10 mg/L. The PAC prepared from wood was 200 meshes (Kunshan, China) with a preadsorption time of 10 min and an addition of 10 mg/L. Deionized (DI) water was used to prepare the coagulant and PAC stock solutions. The recycling alum sludge was taken from the jar test of coagulation with coagulant addition only, and the mixed sludge of PAC combined with alum sludge was taken from the jar test with both PAC and coagulant addition. The recycle ratio was chosen for 8% as the optimal value which was determined by tests. Two kinds of characteristics of sludges are shown in Table 2.

Table 2
Characteristics of the two kinds of sludges

Parameters	Alum sludge	Mixed sludge
Solid ratio (w/w%)	0.226 ± 0.021	0.242 ± 0.013
Suspended solid (g/L)	2.276 ± 0.230	2.452 ± 0.212
pH	7.05 ± 0.10	7.11 ± 0.08
DOC (mg/L)	9.83 ± 0.30	9.67 ± 0.19

2.3. Raw water supply

The raw water was taken from the Songhua River in northeast of China, which represented a kind of typical surface water in China. The main water quality characteristics of raw water used in the study are summarized in Table 3.

2.4. Hydrophilic and hydrophobic fractions based on XAD resin separation

XAD resin was used following the procedures of Thurman and Malcolm [21] to determine the hydrophilic and hydrophobic fractions of the organic matter in the UF influent and pollutants. In brief, the XAD-8 and XAD-4 resin (Supelco) sizing from 20 to 50 meshes were cleaned with several solvents and used to fill a glass column. The resin column was thoroughly rinsed with 0.1 M NaOH, 0.1 M HCl and DI water. A water sample that had been prefiltered by a 0.45 μm filter was pumped though the XAD-8 column and then the column was washed by H_3PO_4 of 0.1 mol/L. The organic substances that were reclaimed by H_3PO_4 were assigned as hydrophobic bases (HoB), and the organic substances that stay within the column were hydrophobic neutral fraction (HoN). The water sample that passed though the column was acidified to pH 2.0 with HCl and was pumped through the XAD-8 column for further separation. The organics adsorbed by the XAD-8 resin were the hydrophobic acids (HoA). The remaining water sample was pumped through the XAD-4 column, and the organic substances that passed the column without adsorption (and extraction) were assigned as the hydrophilic matter (HiM). The organics adsorbed by the XAD-4 resin were the weakly hydrophobic acids (WHoA). All hydrophilic and hydrophobic fractions were measured for the TOC concentration.

2.5. MW fractionation

The MW distribution of the organic matters in the raw water, UF effluent and the fouling substances was determined following the method of UF fractionation [22]. The organic matters washed down by backwash were fractionated to analyse the reversible fouling, and the absorbed organic matters by NaOH after backwash and sponge scrubbing were fractionated to analyse the irreversible fouling in this test. For the raw water and fouling matters sample, they were first filtered through a 0.45 lm membrane. The samples used for the MW distribution were filtered through a number of cellulose membranes in turn, which had nominal MW cut-offs of 30,000, 10,000, 5,000, 3,000 and 1,000 Da (YM30, YM10, YM5, YM3 and YM1, Amicon). For the filtration test, a membrane with a surface area of 63.6 cm^2 was laid in a stirred cell of 300 mL in volume and placed on a magnetic stirrer. The sample liquid filled inside the cell was stirred by a magnetic stir-bar at 120 rpm to prevent the membrane from fouling. Pressure was applied using a nitrogen gas cylinder at 1 bar at room temperature against the liquid enclosed in the stirred cell. The permeated liquid was analysed for TOC concentrations. The concentration of organic matter with different MW ranges was determined by the subtraction method.

Table 3
Characteristics of raw water

Parameters	Results
Turbidity (NTU)	19.5–21.2
pH	7.62–7.72
DOC (mg/L)	7.72–7.83
UV_{254} (cm^{-1})	0.088–0.096
Total hardness ($CaCO_3$) (mg/L)	75.5–81.0
Total alkalinity (mg/L)	62.3–72.0

2.6. Other analytical methods

Water quality analysis was conducted following the standard methods [23], UV_{254} was determined by using the spectrometer (UV754, Cany, China). DOC (prefiltration through 0.45 μm membrane) was measured by the TOC analyser (TOC-VCPH, Shimadzu, Japan). Turbidity was monitored by a turbidimeter (TURBO550, WTW, Germany). THMFP was determined following US EPA Methods 551.1 and 552.2. The extracted sample was analysed for THMs by a GC (Agilent 6890 N, USA) and an electron capture detector (ECD). BDOC was measured with the method of a study [24] by doing some modification. The fouling layer on membrane surface was gold-coated by a sputter and observed under scanning electron microscopy (SEM) (Hitachi S4800 HSD, Japan).

3. Results and discussion

3.1. TMP development of UF membrane in short-term filtration

The evolution of TMP with time in RPAS processes with UF compared with three other different pretreatment processes and raw water filtrated directly by UF during short-term filtration is shown in Fig. 2. The TMP of the raw water filtrated directly by UF membrane increased quickly from 15 to 40.5 kPa after 48 h of run, which increased by 25.5 kPa. TMP of UF

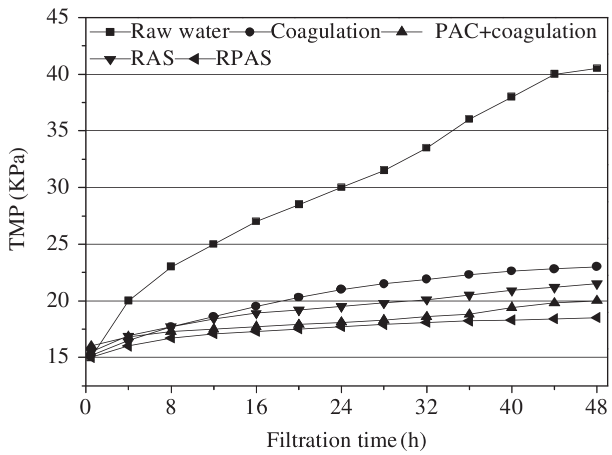

Fig. 2. TMP evolution of RPAS coupled with UF compared with three other different pretreatments (coagulation, PAC + coagulation and reuse of alum sludge).

coupled with RPAS was relatively stable in 48 h of operation with the increase of 3.5 kPa. TMP increased by 7.8, 4.0, and 6.0 kPa, respectively, for coagulation, PAC combined with coagulation and reuse of alum sludge (RAS) only. The results indicated that membrane fouling could be reduced significantly by RPAS pretreatment, which is better than three other different pretreatment processes.

3.2. *Membrane resistance analysis for RPAS and other pretreatments*

Membrane resistances for UF combined with RPAS and three other different pretreatment processes are shown in Fig. 3. The adsorption resistance (R_a), cake resistance (R_c), concentration polarization resistance (R_{cp}) and plugging resistance (R_g) could be all decreased to a certain degree by RPAS and three other different pretreatment processes. It can be seen from Fig. 3 that the membrane-inherent resistances (R_m) of different tests are not same, the standard specific membrane resistance was used to compare the resistances which is shown in Fig. 4.

The standard specific membrane resistance of adsorption resistance (R_a/R_m), cake resistance (R_c/R_m), concentration polarization resistance (R_{cp}/R_m) and plugging resistance (R_g/R_m) for the raw water filtrated directly by UF was 0.13, 0.90, 0.39 and 0.23, respectively. R_a/R_m was reduced to 0.032, 0.098, 0.065 and 0.065 by RPAS, coagulation, PAC combined with coagulation and reuse of alum sludge processes, respectively. R_c/R_m was reduced to 0.065, 0.20, 0.087 and 0.065 by RPAS, coagulation, PAC combined with coagulation and reuse of alum sludge processes, respectively. R_{cp}/R_m was reduced to 0.065, 0.13, 0.065 and 0.10 by RPAS, coagulation, PAC combined with coagulation and reuse of alum sludge processes, respectively. R_g/R_m was reduced to 0.065, 0.20, 0.11 and 0.21 by RPAS, coagulation, PAC combined with coagulation and reuse of alum sludge processes, respectively.

Results indicate that R_c and R_{cp} could be well reduced by all the four different pretreatments. R_a and R_g were not greatly decreased by coagulation. Although R_a might be reduced to a certain degree by PAC combined with coagulation, R_g was changed very little, which was the same as reuse of alum sludge processes. All the membrane resistances could be well reduced to a great degree by RPAS, and the overall resistance of which was the least in all the four different pretreatments.

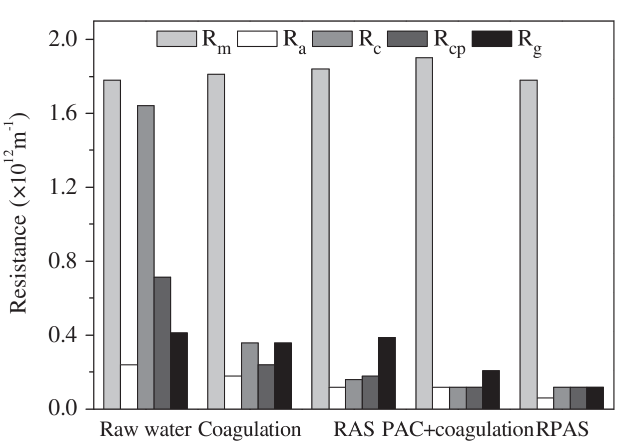

Fig. 3. Effect of RPAS process on the resistance of UF membrane compared to other pretreatments (coagulation, PAC + coagulation and reuse of alum sludge).

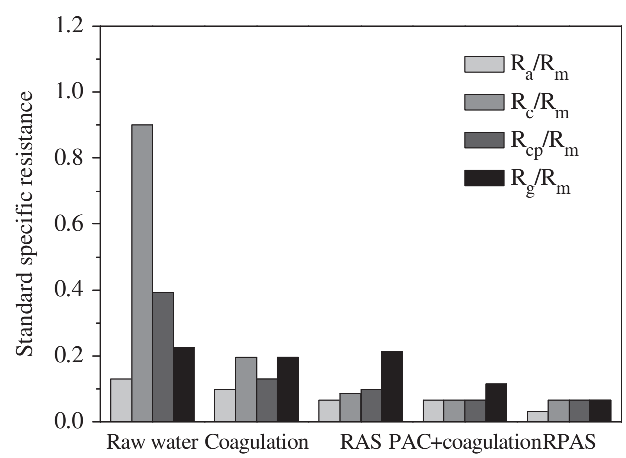

Fig. 4. Standard specific resistance of UF membrane for different pretreatments (coagulation, PAC + coagulation, reuse of alum sludge and reuse of PAC and alum sludge).

3.3. TMP development of UF membrane in periodical filtration

Effects of RPAS process on TMP of UF membrane in periodical filtration compared with other pretreatments were also investigated. Fig. 5 showed the TMP evolution of UF membrane in five continuous filtration periods for RPAS and three other pretreatments.

The TMP of the direct UF for raw water increased quickly from 15 to 45.5 kPa after 5 continuous filtration periods of run, increasing by 30.5 kPa. In the same filtration periods, TMP of UF coupled with RPAS was slightly increased by 6.3 kPa. The TMP increased by 11, 7.0 and 9.5 kPa for coagulation, PAC combined with coagulation and RAS, respectively. After four continuous filtration periods, the TMP of the direct UF to raw water after being backwashed was 22 kPa with an increase of 7.0 kPa, which was mainly caused by irreversible fouling. TMP of UF coupled with RPAS was relatively stable in five continuous filtration periods with an increase of 4.0 kPa, while with an increase of 4.8, 4.3 and 5.0 kPa for coagulation, PAC combined with coagulation and RAS, respectively, which indicted that less membrane fouling happened in RPAS process under the test conditions. It could be inferred from the results that irreversible fouling could be more effectively alleviated by RPAS pretreatment.

3.4. Microscopic observations of the fouling layer on membrane surface

SEM images were taken to determine the morphology of the fouling layer on the membrane surface. In comparison with the flat and smooth surface of clean membrane (Fig. 6(a)), it can be seen that the membrane surface after filtrating raw water (Fig. 6(b)) was covered with a thick fouling layer, while the membrane surface of RPAS (Fig. 6(c)) was relatively clean due to that the particulate and organic matters were well removed by RPAS pretreatment.

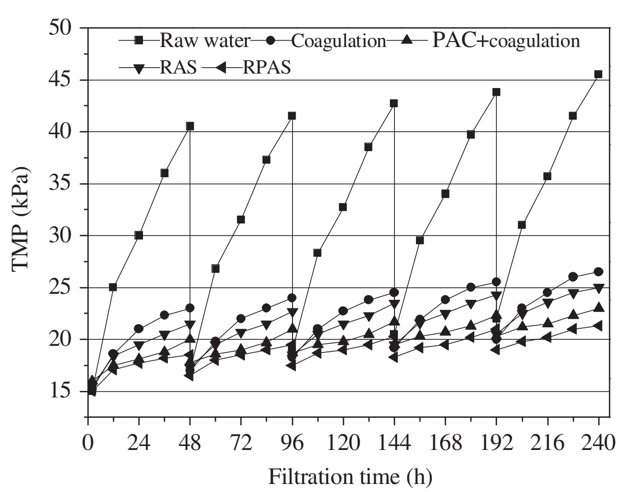

Fig. 5. Comparison of TMP evolution for RASP and other pretreatments during five operating periods (with one operating period of 48 h).

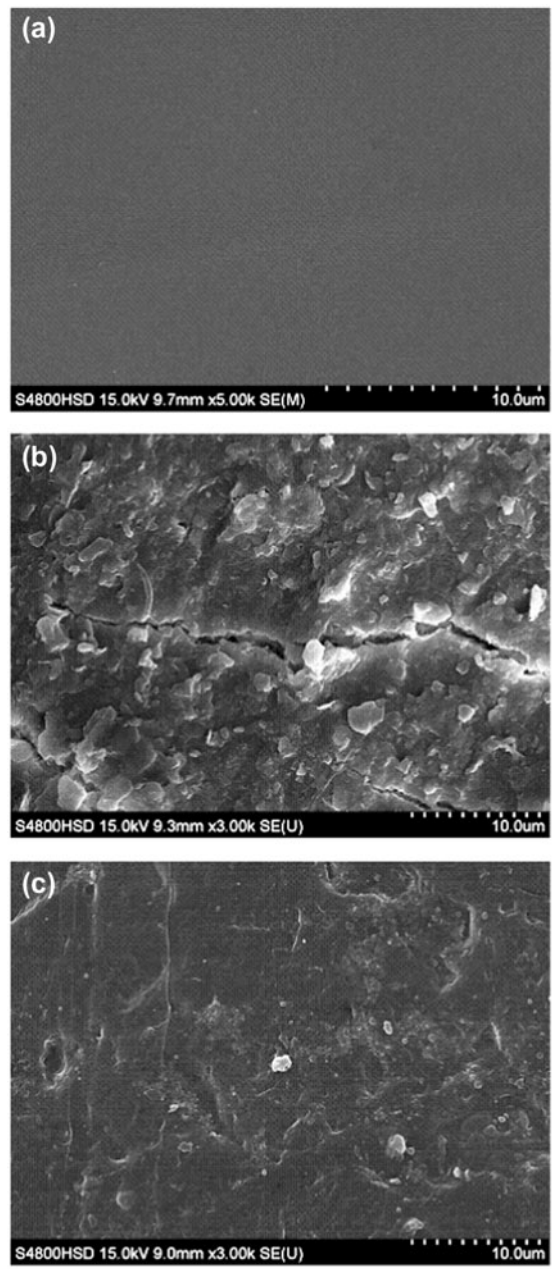

Fig. 6. SEM images of clean membranes surface (a), as well as fouling layer on the membrane surface after filtrating raw water (b) and RPAS effluent (c).

3.5. Mechanism of fouling control for RPAS pretreatment by impurities removal

3.5.1. The overall removal of particulates and organic matters

The removal of turbidity and organic matters by RPAS process are illustrated in Fig. 7. It could be seen that the removal efficiencies of turbidity, DOC, UV254, BDOC and THMFP were 93.8, 37.3, 41.1, 83.0 and 57.9% on average, respectively. The well performance may be attributed to the PAC adsorption for organic matters combined with the enhanced coagulation by reuse of alum sludge.

3.5.2. Chemical fractionation of the organic matters removed by RPAS

Membrane fouling during filtration could be usually caused by organic adsorption. Hydrophobic acids (HoA) are usually considered to induce the reversible fouling, and hydrophilic matters (HiM) to cause the irreversible fouling [2,25,26]. Chemical fractionation of natural organic matters (NOM) in the raw water and in the effluent of RPAS process is shown in Fig. 8. The concentration of HoB, HoN, HoA, WHoA and HiM of the organic matter in raw water was 0.662, 1.32, 2.10, 2.06 and 1.56 mg/L, the removal efficiencies of which could reached up to 35.6, 36.7, 70.6, 3.53 and 36.4%, respectively. Both hydrophobic acids and hydrophilic matters were well reduced by RPAS pretreatment, which might be attributed to the adsorption and sweeping of the particulates and flocs within the recycling sludge.

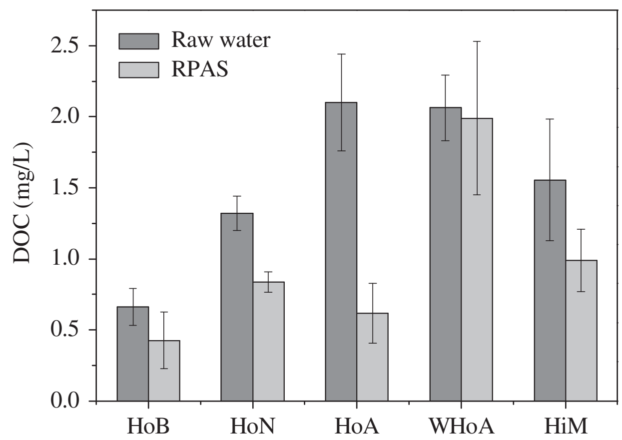

Fig. 8. Fractionation of DOM in raw water and RPAS process effluent.

3.5.3. MW distribution of organic matters removed by RPAS

The MW distribution of organic matters in raw water and RPAS effluent is shown in Fig. 9. It could be seen that the concentration of organic matters with the MW range of <1 k, 1–3 k, 3–5 k, 5–10 k, 10–30 k and >30 k Da in raw water was 4.23, 1.53, 0.185, 0.293, 0.339 and 1.12 mg/L, the removal efficiencies of which could reached up to 32.6, 42.7, 19.5, 46.4, 20.1 and 50.4% on average, respectively. Both reversible and irreversible membrane fouling could be caused by the organic matters with the MW more than 30 k Da, while irreversible membrane fouling were mainly caused by the organic matters with the MW less than 1 k Da [27]. Results indicated that the organic matters

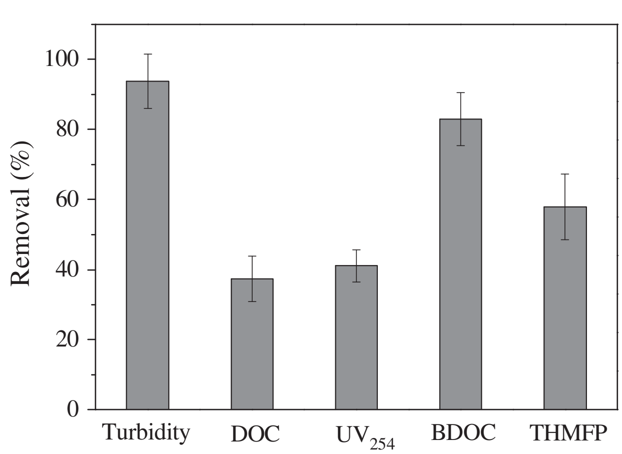

Fig. 7. Effect of RPAS on turbidity and organic matters removal in raw water.

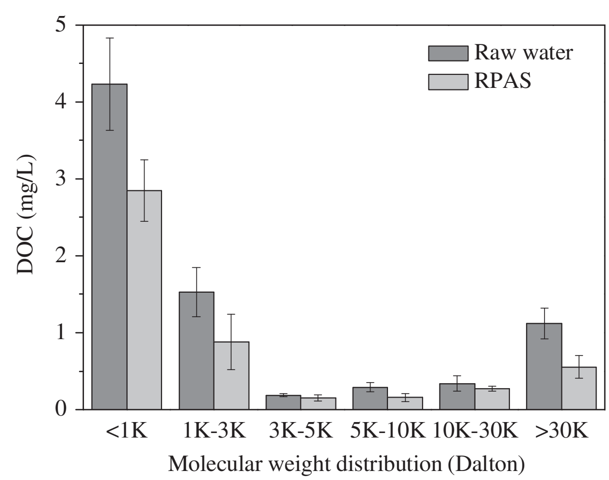

Fig. 9. MW distribution of DOM in raw water and RPAS process effluent.

with the MW more than 30 k Da was well removed by RPAS pretreatment with a removal efficiency of 50.4%, and that of MW less than 1 k Da were reduced by 32.6%, which could partly explain the very slightly increasing TMP evolution during the period of UF for RPAS effluent.

3.5.4. Mechanism analysis of reducing fouling for RPAS

The results indicate that membrane fouling could be well alleviated by RPAS pretreatment due to the efficient removal of particulates and organic matters of raw water. Cake resistance could be mainly caused by the suspended particles and macromolecules organics, while adsorption resistance and concentration polarization resistance are mainly caused by micromolecules and macromolecules organics, respectively [28,29]. Suspended particles and macromolecules organics could be partly removed by coagulation, so cake resistance and concentration polarization resistance could be reduced a lot by coagulation. PAC has a good ability to adsorb low molecular organic compounds [4], and adsorption resistance may be reduced to a certain degree, by which irreversible fouling could be alleviated. Reuse of the alum sludge from water treatment may not only improve the removal efficiency of the organic matters but also enhance the coagulation of low turbidity water for drinking water treatment [16,17]. Both alum sludge and PAC existed in the mixed sludge, and suspended particles and macromolecules organics could be better removed by the enhanced coagulation of alum sludge, while low molecular organic matters could be significantly removed by PAC adsorption coupled with adsorption and sweeping by flocs within the mixed sludge. Addition of PAC could also improve membrane permeability [30]. Integration of PAC adsorption and enhanced coagulation of alum sludge have a better removal for membrane fouling substances in raw water, and RPAS process could be chosen as an effective pretreatment for membrane filtration in polluted raw water treatment.

4. Conclusion

Results of this investigation indicated that membrane fouling could be reduced significantly by RPAS pretreatment due to the well removal of particles and organic matters in raw water. TMP development of UF membrane for RPAS pretreatment in both short-term and periodical filtration grew slower than three other pretreatments, and membrane resistances could be well reduced to a great degree by RPAS process. PAC adsorption combined with enhanced coagulation of alum sludge have a better removal for particles and organic matters in raw water, so RPAS process could be chosen as an effective pretreatment to reduce membrane fouling in drinking water treatment for polluted raw water.

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