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# Removal of natural organic matter (NOM) by the coagulation–ultrafiltration system using an inorganic–organic composite coagulant

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## ABSTRACT

A coagulation–ultrafiltration system to treat drinking water has been widely studied. This study investigated the feasibility and potential of using inorganic-organic composite coagulant in the coagulation-ultrafiltration system for natural organic matter (NOM) removal. The removal efficiency and membrane fouling were studied at different coagulation conditions. And in order to observe how inorganic-organic composite coagulant affects the permeate flux, a resistance analysis was conducted. The results suggest that although the treated water quality in a composite coagulant system was not improved significantly, composite coagulant was more advantageous in terms of improving the permeate flux of the coagulation–ultrafiltration system than inorganic coagulant by reducing the resistance due to cake formation and adsorption. The coagulation conditions of composite coagulant are important for the performance of the coagulation–ultrafiltration system. When dosage is 3 mg $L^{-1}$ and pH is 6, the removal efficiency and the permeate flux are the highest in our study.

*Keywords*: Composite coagulant; Ultrafiltration; Humic acid; Membrane fouling; Resistance

## 1. Introduction

In recent years, natural organic matter (NOM) removal in source drinking water has received increasing attention. The presence of NOM in source water adversely affects water treatments and the quality of produced water. NOM is known to increase disinfectant and coagulant demand, generate potentially harmful disinfection by-products, foul membrane and favor biological re-growth in the distribution network [1]. Therefore, NOM removal is a major challenge in modern drinking water treatment.

Conventional water treatment, using coagulation/flocculation, is the most frequently used process for drinking water treatment targeting at NOM and turbidity removal. However, coagulation only removes a portion of NOM, and removal efficiency depends on water quality and water treatment conditions [2,3], especially the molecular weight of organic compounds [4,5]. As a consequence, drinking water plants tend to improve or develop new technologies for producing water with a low dissolved organic carbon (DOC) content.

In the new technologies, application of the coagulation–ultrafiltration system to the improvement of treated water quality has been widely studied for water production, because coagulation process followed by membrane is easy to be realized for conventional drinking water plants. Fiksdal et al. [6] reported that pre-coagulation in combination with both loose UF and MF membrane filtra-

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tion is an effective hygienic barrier against virus. Chen et al. [7] applied coagulation as a pre-treatment step before the ultrafiltration removed the hydrophobic NOM water fraction, and found that it could increase the flux of water and reduce the fouling effects.

Previous studies also show that although the coagulation could remove NOM and decrease the resistance of membrane filtration, the rate and extent of fouling could not be mitigated by different coagulants, because the characteristics of coagulation chemistry and the coagulated particles have a great impact upon membrane performance [8,9]. Barbot et al. [10] used five organic coagulants (cationic polyelectrolytes) and ferric chloride as coagulants to treat bentonite synthetic water in the coagulation-ultrafiltration system, and find that some coagulants have no influence on permeate flux, some result in a 20% increase in permeate flux whereas other lead to a decrease of 50%. Bergamasco et al. [11] applied chitosan as coagulant in a microfiltration process of nature water, and find that although the coagulation process causes an increase in fouling, the quality of treated water is better compared to the simple microfiltration process. Therefore, proper choice of coagulant is important for the performance of the coagulation–ultrafiltration system.

In recent years, various coagulant categories have been developed including inorganic coagulants, organic coagulants and composite coagulants. Because of their superior efficiency compared with traditional inorganic flocculants, and lower cost compared with organic flocculants, composite flocculants have been extensively studied and applied to water and wastewater treatment systems today [12,13]. Moussas et al. [12] used polyferric sulphate and polyacrylamide to synthesize a new composite coagulant, and find this coagulant exhibits better coagulation performance, when compared with simple PFS, in terms of zeta-potential reduction, turbidity and organic matter removal and residual iron concentration. And the composite coagulant, polyferric-polydimethyldiallylammonium chloride is more efficient and applicable for treating high organic matter and high alkalinity surface water [14]. In addition, the novel composite coagulant, polyaluminum-polydimethyldially lammonium chloride (PAC–PDMDAAC) have better coagulation efficiency and lower residual aluminum compared to traditional inorganic flocculants and organic flocculants [15,16].

Although some studies have addressed the characteristics of composite flocculants, the application of composite flocculants in the coagulation-ultrafiltration system has not been investigated. It is essential to select an optimal coagulant to achieve a high performance in the coagulation–ultrafiltration system. Hence, the aim of this work was to evaluate the feasibility and potential of using inorganic-organic composite coagulant in the coagulation–ultrafiltration system for NOM removal. The fouling mechanism and the corresponding resistances (including cake resistance, resistance due to adsorption, and specific cake resistance) during ultrafiltration were also studied.

## 2. Materials and methods

### 2.1. Synthetic test water

In this study, humic acid (HA) is chosen to represent NOM. The HA stock solution is prepared as follows: 1 g of HA (Shanghai, China) is dissolved in 1000 ml deionized water which contained 4.2 g of $NaHCO_3$. Under these conditions, the HA was soluble.

Apparent molecular weight (AMW) distribution, which is an important parameter for HA, is determined to use ultra filtration membranes with an Amicon® cell device (Model 8200, Millipore, USA). For ultrafiltration operation, the stock humic acid solution is diluted 200 times with deionized water. Pure nitrogen gas (0.35 MPa) is used as the driving force. The humic acid was divided into four classes: >30, 10–30, 3–10 and <3 kDa. The total organic carbon (TOC) and $UV_{254}$ absorbance of each class are measured. The humic acid mainly consisted of high AMW constituents with fraction greater than 30 kDa accounting for 80.1% of TOC and 87.5% of $UV_{254}$ absorbance.

The synthetic test water is prepared by dissolving the HA stock solution (10 ml) in deionized water (500 ml). The properties of the synthetic test water used were as following: $UV_{254}$ = 0.208–0.234, TOC = 9.28 mg $L^{-1}$, pH = 7.7.

### 2.2. Coagulants

$AlCl_3$, polyaluminum chloride (PAC) and polydimethyldiallylammonium chloride (PDMDAAC) were obtained directly from Bin Zhou Chemical Co., Shandong, China. Stock solutions of $AlCl_3$ and PAC with the total aluminum concentrations of 10% were prepared in distilled water 24 h before being used. The composite coagulant used in this study was synthesized by PAC and PDMDAAC in our laboratory. PDMDAAC (10 g) was injected into the stock solution of PAC (100 ml) with intense stirring at 30°C until the solution became clear. The total aluminum concentration of the composite coagulant (denoted as PAC–PDMDAAC) was 9.10%. All coagulants dosages used in this study are in the unit of mg $L^{-1}$ as Al.

### 2.3. Coagulation–ultrafiltration system experiment

A dead-end batch ultrafiltration unit is used in our study. Ultrafiltration membranes (Mosu, China) with MWCO of 300 kDa are used. The membrane material is modified polyvinylidene fluoride. A fresh piece of membrane is used in every experiment. Prior to the filtration of coagulation suspension, the membrane is "wetted" to its optimal operating condition; whereby deionized water was filtered through the fresh unfolded membrane, at the desired TMP for that experiment, until a steady permeate flux was observed. This value represented the pure water flux of each particular piece of membrane and was recorded in every experiment. The effective membrane area is 48 cm$^2$. Nitrogen gas was used for the application of TMP. Instantaneous mass of cumulative permeate was measured by electronic balances (PB3002-SDR, Mettler Toledo Ltd.) with automated data logging capability. These data were recorded by a connected PC at a predetermined time interval.

Coagulation was conducted in a 1 L cylindrical reactor under 1 min of rapid mixing (120 rpm) and 10 min of flocculation (40 rpm), and then was settled for 10 min. Subsequently, the coagulation suspension was transferred directly from the coagulation reactor into a 0.3 L cylindrical filtration cell and filtered through an ultrafiltration membrane under constant air pressure 30 kPa. A slow stirring velocity was maintained in the reservoir to prevent flocs from settling. The ultrafiltration membrane permeate was analyzed with UV$_{254}$ and TOC for the HA removal efficiency. The coagulation suspension samples were collected from the reactor to measure SC and the particle size of flocs by CAS Charge Analyzing System (Germen) and Malvern Mastersizer2000 (England) respectively. And the fractal dimension was calculated according the particle size data [17].

### 2.4. Resistance in series model

In order to assess the fouling behavior of the investigated systems, resistances due to different fouling mechanisms are determined. Resistances are then calculated following the resistance in series model, as adapted from [18]:

$$J = \frac{\Delta P}{\eta(R_m + R_f + R_c)} \tag{1}$$

where $J$ is permeate flux, $\Delta P$ is trans-membrane pressure (defined as the difference between the applied pressure and the osmotic pressure), $\eta$ is dynamic viscosity, $R_m$ is membrane hydraulic resistance, $R_f$ is resistance due to adsorption and pore blocking, and $R_c$ is resistance due to cake formation.

According to previous study [19], $R_m$, $R_f$ and $R_c$ were determined by the following experiments. $R_m$ was determined by filtering deionized water through virgin membrane sheets for at least 2 h under the same operating conditions as mentioned in Section 2.3. Since the other resistances ($R_f$, $R_c$) did not exist at this stage, $R_m$ was calculated using the following equation:

$$R_m = \frac{\Delta P}{\eta J_0} \tag{2}$$

where $J_0$ is the permeate flux of deionized water filtered through the clean membrane. After this step, the same membrane sheet was then immersed in the feed solution until the flux was stable. At this condition, the permeate flux was denoted as $J_1$. The fouled membrane was then gently wiped to remove the cake layer on the surface, and hence only leaving the foulants that adsorbed and blocked the pores. Deionized water was filtered once again through the same membrane sheets for 2 h. This permeate flux was denoted as $J_2$. Since $R_c$ did not exist at this stage, $R_f$ was calculated as follows:

$$R_f = \frac{\Delta P}{\eta J_2} - R_m \tag{3}$$

Finally, $R_c$ was calculated by subtracting all other resistances to the total resistance:

$$R_c = \frac{\Delta P}{\eta J_1} - R_m - R_f \tag{4}$$

## 3. Results and discussion

### 3.1. Effect of different Al(III) based coagulants on the performance of the coagulation–ultrafiltration system

In order to observe whether PAC–PDMDAAC is suitable for the coagulation–ultrafiltration system, the performance of the coagulation–ultrafiltration system was investigated when AlCl$_3$, PAC and PAC–PDMDAAC were used as coagulants. According to previous experiments (not covered in this paper), the dosage of AlCl$_3$, PAC and PAC–PDMDAAC was kept at 7, 6, and 3 mg L$^{-1}$, respectively. And pH of HA water was kept at 6. The permeate flux decline trends of AlCl$_3$, PAC and PAC–PDMDAAC are plotted in Fig. 1, and the HA removal efficiency is shown in Table 1. It can be seen from Fig. 1 that the severest flux decline occurred in the coagulation–ultrafiltration system when AlCl$_3$ was used as coagulant, whereas the slightest decline occurred

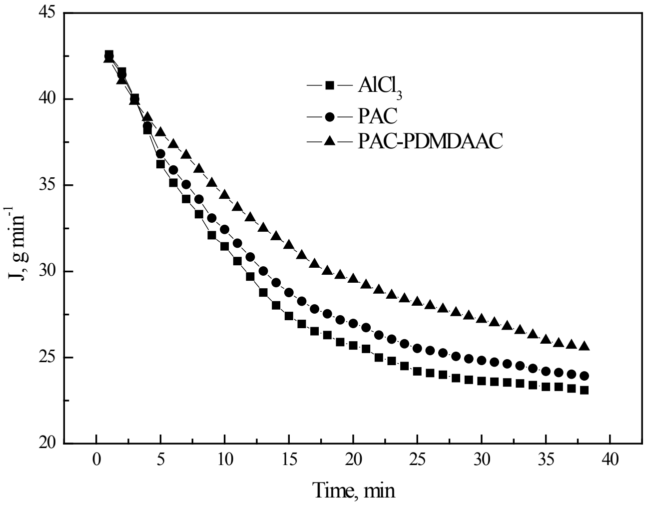

Fig. 1. Variations of the permeate flux at TMP = 30 kPa in the coagulation–ultrafiltration system of coagulated suspensions made by different Al(III) based coagulants.

Table 1
HA removal efficiency of the investigated systems

| Systems | Conditions | UV$_{254}$ | TOC (mg L$^{-1}$) |
|---|---|---|---|
| AlCl$_3$ | 7 mg L$^{-1}$, pH 6 | 0.022 | 0.58 |
| PAC | 6 mg L$^{-1}$, pH 6 | 0.014 | 0.43 |
| PAC–PDMDAAC | 3 mg L$^{-1}$, pH 6 | 0.015 | 0.45 |
| | 0.5 mg L$^{-1}$, pH 6 | 0.033 | 0.79 |
| | 8 mg L$^{-1}$, pH 6 | 0.015 | 0.44 |
| | 3mg L$^{-1}$, pH 4 | 0.024 | 0.63 |
| | 3 mg L$^{-1}$, pH 9 | 0.027 | 0.67 |

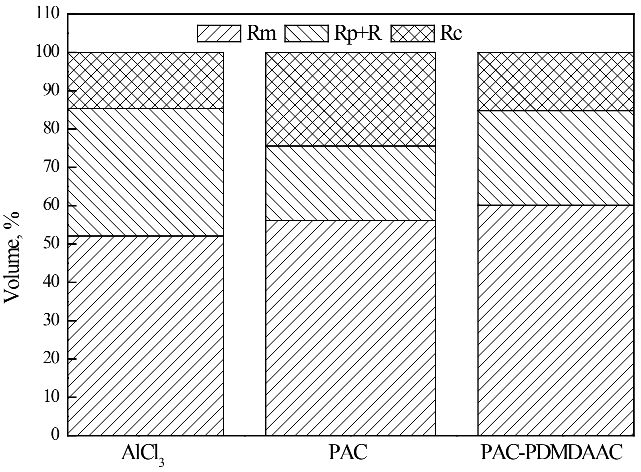

Fig. 2. Resistance analysis of the investigated systems: AlCl$_3$, PAC and PAC–PDMDAAC.

when PAC–PDMDAAC was chosen as coagulant in the system. The HA removal efficiency of PAC–PDMDAAC is similar to that of PAC, which is higher than that of AlCl$_3$ (Table 1). This indicates that although the treated water quality in PAC–PDMDAAC is not improved significantly, PAC–PDMDAAC is more advantageous in terms of improving the permeate flux of the coagulation–ultrafiltration system than AlCl$_3$ and PAC.

In order to observe how PAC-PDMDAAC affects the permeate flux further, a resistance analysis was conducted. The results of this analysis are plotted in Fig. 2 and the values are shown in Table 2. The resistances are divided into three parts in our study: 1) the membrane resistance, $R_m$; 2) the resistance due to cake formation, $R_c$; 3) the resistance due to irreversible adsorptions and pore blockings, $R_f$. All resistances are plotted in percentage relative to the total resistance. As shown in Fig. 2, PAC–PDMDAAC gives the largest $R_m$ relative to the total resistance, whereas PAC gives the largest $R_c$ relative to the total resistance. This shows that the flocs formed by PAC–PDMDAAC are less absorbed or deposited on ultrafiltration compared with AlCl$_3$ and PAC. Therefore, PAC-PDMDAAC could improve the permeate flux by reducing the resistance due to cake formation and adsorption.

Previous studies suggest that the performance of the coagulation–ultrafiltration system would largely depend on the properties of flocs, which is governed by the specific coagulation conditions [20]. Therefore, the properties of flocs in the investigated systems were determined in order to explore the effect of PAC-PDMDAAC on the performance of the coagulation–ultrafiltration system. The results are shown in Table 3.

It can be seen that although the medium diameter ($D_{0.5}$) of flocs formed by AlCl$_3$, PAC and PAC-PDMDAAC are larger than 120 um, the difference in the floc size is observed. In AlCl$_3$ and PAC systems, small flocs could be observed. For example, the concentration below 10 um in AlCl$_3$ and PAC systems are 1.25 and 3.03%, respectively, whereas there is no floc below 10 um in the PAC–PDMDAAC system (Fig. 3). Generally, the small flocs are the major matter for flux decline through internal fouling or cake formation [21]. Thus, the flocs in the PAC–PDMDAAC system reduce the permeate flux less than in AlCl$_3$ and PAC systems due to a larger size. On the other hand, the fractal dimension of flocs formed by AlCl$_3$ is the highest, followed by PAC and PAC–PDMDAAC, indicating that the structure of flocs formed by AlCl$_3$ is much more compact than that formed by PAC and PAC–PDMDAAC. The more compact flocs produce higher resistance, whereas the loose flocs result in lower specific resistance [22]. According to this view,

Table 2
Resistance values summary of the investigated systems

| Systems | Conditions | $R_c$×1010 (m$^{-1}$) | $R_f$×1010 (m$^{-1}$) | $R_m$×1010 (m$^{-1}$) |
|---|---|---|---|---|
| AlCl$_3$ | 7 mg L$^{-1}$, pH 6 | 0.988 | 2.25 | 3.52 |
| PAC | 6 mg L$^{-1}$, pH 6 | 1.53 | 1.22 | 3.47 |
| PAC-PDMDAAC | 3 mg L$^{-1}$, pH 6 | 0.669 | 1.67 | 3.55 |
| | 0.5 mg L$^{-1}$, pH 6 | 3.19 | 1.67 | 3.49 |
| | 8 mg L$^{-1}$, pH 6 | 0.932 | 1.67 | 3.51 |
| | 3 mg L$^{-1}$, pH 4 | 1.20 | 5.15 | 3.52 |
| | 3 mg L$^{-1}$, pH 9 | 2.03 | 1.08 | 3.53 |

Table 3
The properties of the flocs in the investigated systems

| System | Conditions | *SC* (mV) | $D_{0.5}$ (um) | Fractal dimension |
|---|---|---|---|---|
| $AlCl_3$ | 7 mg $L^{-1}$, pH 6 | –83 | 120 | 2.42 |
| PAC | 6 mg $L^{-1}$, pH 6 | –81 | 177 | 2.34 |
| PAC–PDMDAAC | 3 mg $L^{-1}$, pH 6 | –42 | 130 | 2.03 |
| | 0.5 mg $L^{-1}$, pH 6 | –65 | 21 | 1.89 |
| | 8 mg $L^{-1}$, pH 6 | 26 | 168 | 2.61 |
| | 3 mg $L^{-1}$, pH 4 | –51 | 106 | 1.82 |
| | 3 mg $L^{-1}$, pH 9 | –39 | 183 | 1.74 |

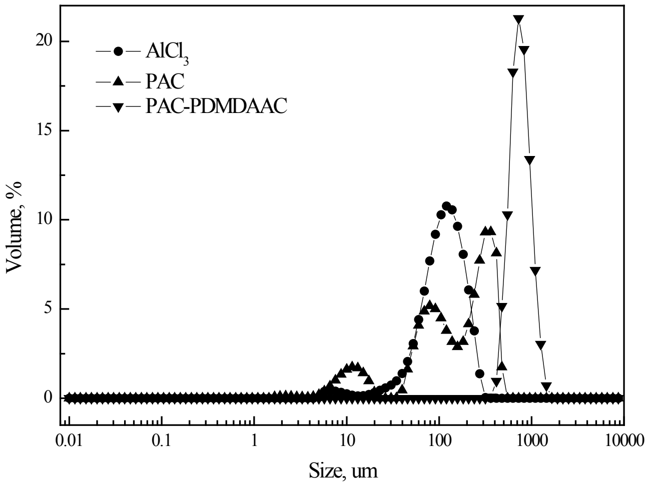

Fig. 3. Particle size distribution of flocs at the investigated systems: $AlCl_3$, PAC and PAC–PDMDAAC.

PAC–PDMDAAC produces the loosest flocs so that the extent of flux decay in the PAC–PDMDAAC system is the smallest in our study. Moreover, SC of flocs in the PAC–PDMDAAC system is the highest, which implies that flocs in PAC–PDMDAAC are formed by the charge neutralization. Lee et al. [20] thought that the cake layer formed through the sweep floc mechanism would certainly be more compressible than that formed through the charge neutralization mechanism. This would also lead to a higher permeate flux for the PAC–PDMDAAC system. According to the above information, PAC–PDMDAAC produces flocs which are more suitable for the coagulation–ultrafiltration system, so PAC–PDMDAAC can improve the permeate flux by reducing the resistance due to cake formation and adsorption.

### 3.2. *Effect of dosage on the performance of the coagulation–ultrafiltration system*

The previous reports suggest that membrane filterability would largely depend on the coagulation conditions [9]. Thus, a series of experiments were conducted with varied dosage at 0.5, 3 and 8 mg $L^{-1}$ to further probe into the effects of PAC–PDMDAAC in the coagulation–ultrafiltration system.

Fig. 4 presents the permeate flux trends of the coagulation–ultrafiltration system with different PAC–PDMDAAC dosages. As seen in this figure, the permeate flux at the same filtering time decreased in the following order: 3 > 0.5 > 8 mg $L^{-1}$. And when the dosage was 8 mg $L^{-1}$, the permeate flux declined rapidly during the first 20 min, and then the flux declined gradually and became less severe. The HA removal efficiency of 3 mg $L^{-1}$ is similar to that of 8 mg $L^{-1}$, which is higher than that of 0.5 mg $L^{-1}$. These results suggest that the system is very sensitive to dosage. Under the optimal dosage, the HA removal efficiency and the permeate flux at the same filtering time increase with increasing the dosage. When the optimal dosage is exceeded, although the HA removal efficiency remained constant, the membrane fouling increased with increasing the dosage.

In terms of resistance, an increase of dosage from 0.5 to 3 mg $L^{-1}$ results in a slight decrease of $R_c$ relative to the

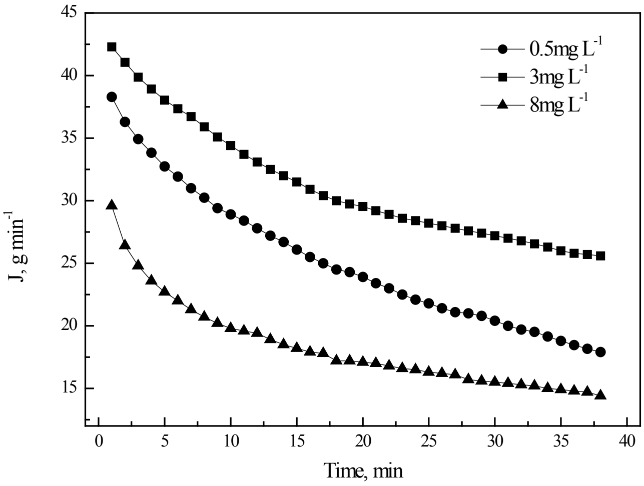

Fig. 4. Variations of the permeate flux at TMP = 30 kPa in the PAC–PDMDAAC system of coagulated suspensions made at different dosages.

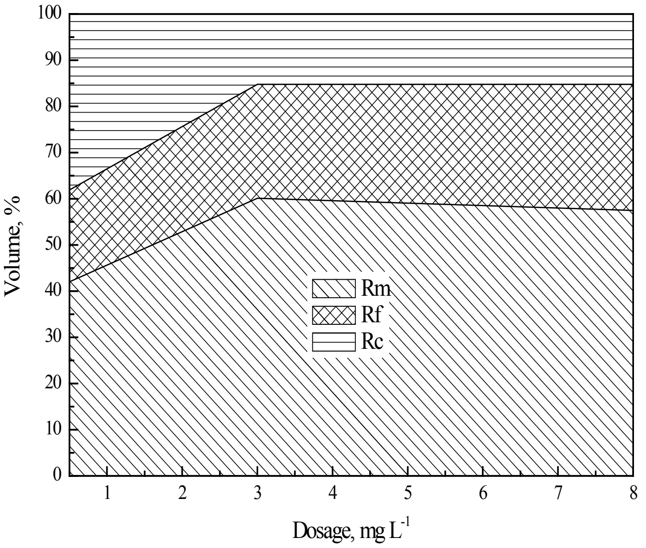

Fig. 5. Resistance analysis of the investigated systems at different dosages.

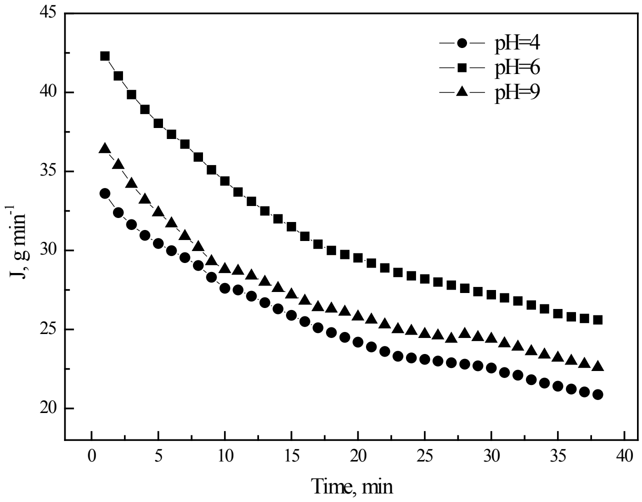

Fig. 6. Variations of permeate flux at TMP = 30 kPa in the PAC–PDMDAAC system of coagulated suspensions made at different pHs.

total resistance, and a slight increase of $R_f$ relative to the total resistance (Fig. 5). When the dosage is higher than 3 mg $L^{-1}$, there is a slight change for $R_m$, $R_f$ and $R_c$ relative to the total resistance. According to Table 2, when the dosage was 3 mg $L^{-1}$, $R_c$ was the lowest in our study. This indicates that at the optimal dosage, the permeate flux at the same filter time is the highest by reducing the cake formation.

According to the properties of flocs at different dosages (Table 3), it can be seen that *SC*, $D_{0.5}$ and fractal dimension of flocs increase when the dosage is increased. When the dosage is 0.5 mg $L^{-1}$, *SC*, $D_{0.5}$ and fractal dimension of flocs are the lowest compared to 3 and 8 g $L^{-1}$. This implies that although there is not enough dosage to produce the loosest flocs, the size of the flocs is the smallest because of insufficient charge neutralization. This results in the highest $R_c$ and hence the lower permeate flux. While for 8 mg $L^{-1}$, although excessive coagulant could produce the largest flocs, the structure of flocs becomes too compact. This is the reason why the excessive dosage gives the higher $R_c$, which results in the lowest permeate flux.

### *3.3. Effect of pH on the performance of the coagulation–ultrafiltration system*

In order to look further into the performance of the coagulation–ultrafiltration system when PAC–PDMDAAC is chosen as coagulant in the system, a series of fouling experiments at different pHs were conducted. The normalized permeate flux trends as a function of filtered time at different pH are shown in Fig. 6. The resistance analysis is shown in Fig. 7 and Table 2. It can be seen that a significant variation of HA removal efficiency occurred when pH was changed, and the permeate flux varied with pH. The permeate flux at the same filtering time decreased in the following order: pH 6 > pH 9 > pH 4. The results are different from the HA removal efficiency in composite coagulation treatment. Wei et al. [13] used different composite flocculants to treat HA solution, and found that the HA removal efficiency decreased when pH increased.

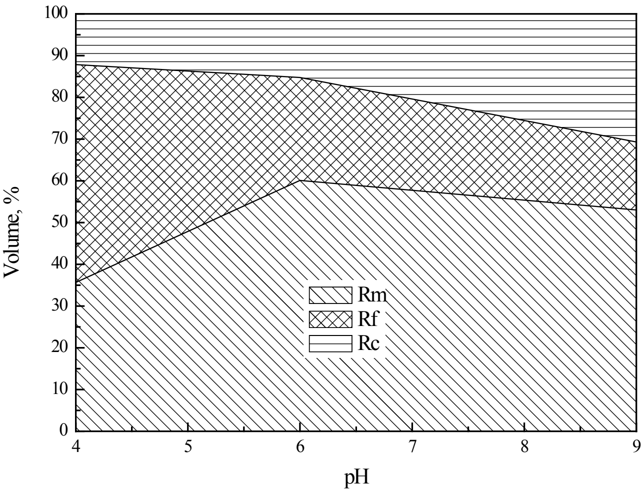

Fig. 7. Resistance analysis of the investigated systems at different pHs.

According to the resistance analysis, it can be seen that the resistance is very sensitive to pH. $R_c$ relative to the total resistance increased and $R_f$ relative to the total resistance decreased with increasing pH (Fig. 7). At pH 6, $R_c$ was the smallest compared with pH 4 and pH 9 (Table 2). This shows that the mechanism of membrane fouling is different when pH changes. At low pH, the reason for membrane fouling is mainly irreversible adsorption and pore blocking. At high pH, reversible cake formation becomes the major factor for membrane fouling.

These results could be explained by the properties of flocs at different pH conditions (Table 3). Although the floc size increases as pH increases, the fractal dimension at pH 4 and pH 9 is too small ($< 2$) which implies that the flocs are too loose to be broken. For pH 4, the looser flocs with the smallest size lead to the highest $R_f$ and hence the lowest permeate flux. For pH 9, the loosest flocs with the largest size result in the highest $R_c$ and hence the lower permeate flux.

## 4. Conclusion

1. Compared with $AlCl_3$ and PAC, the flocs formed by composite coagulant, PAC–PDMDAAC, are less absorbed or deposited at ultrafiltration. Therefore, a composite coagulant could reduce the membrane fouling by reducing the resistance due to cake formation and adsorption, although it does not contribute to the improvement of the quality of treated water.
2. The coagulation dosage of a composite coagulant is important for the quality of the treated water and membrane fouling. Under the optimal dosage, the HA removal efficiency and the permeate flux at the same filtering time increase with increasing the dosage due to cake formation reduction. Above the optimal dosage, although the HA removal efficiency remains constant, the permeate flux at the same filtering time decreases when the dosage increases.
3. The quality of the treated water depends on pH. At low pH, membrane is fouled mainly because of irreversible adsorption and pore blocking. At high pH, reversible cake formation becomes a major factor to membrane fouling.

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## References

[1] V. Jegatheesan, P.R. Lamsal, C. Visvanathan, H.H. Ngo and L. Shu, Effect of natural organic compounds on the removal of organic carbon in coagulation and flocculation processes, Wat. Sci. Technol.: Wat. Supply, 2 (2002) 473–479.

[2] J.K. Edzwald, Coagulation in drinking water treatment: particles, organics and coagulants, Wat. Sci. Technol., 27 (1993) 21–35.

[3] B. Eikebrokk, Coagulation–direct filtration of soft, low alkalinity humic waters, Wat. Sci. Technol., 40 (1999) 55–62.

[4] S.W. Krasner and G. Amy, Jar tests evaluations of enhanced coagulations, J. AWWA, 87 (1995) 93–107.

[5] B. Bolto, D. Dixon, R. Eldridge and S. King, Removal of THM precursors by coagulation or ion exchange, Wat. Res., 36 (2002) 5066–5073.

[6] L. Fiksdal and T. Leiknes, The effect of coagulation with MF/UF membrane filtration for the removal of virus in drinking water, J. Membr. Sci., 279 (2006) 364–371.

[7] Y. Chen, B.Z. Dong, N.Y. Gao and J.C. Fan, Effect of coagulation pretreatment on fouling of an ultrafiltration membrane, Desalination, 204 (2007) 181–188.

[8] K.Y. Choi and B.A. Dempsey, In-line coagulation with low-pressure membrane filtration, Wat. Res., 38 (2004) 4271–4281.

[9] C. Guigui, J.C. Rouch, L. Durand-Bourlier, V. Bonnelye and P. Aptel, Impact of coagulation conditions on the in-line coagulation UF process for drinking water production, Desalination, 147 (2002) 95–100.

[10] E. Barbot, S. Moustier, J.Y. Bottero and P. Moulin, Coagulation and ultrafiltration: Understanding of the key parameters of the hybrid process, J. Membr. Sci., 325 (2008) 520–527.

[11] R. Bergamasco, C. Bouchard, F.V. Silva, M.H.M. Reis and M.R. Fagundes-Klen, An application of chitosan as a coagulant/flocculant in a microfiltration process of natural water, Desalination, 245 (2009) 205–213.

[12] P.A. Moussas and A.I. Zouboulis, A new inorganic–organic composite coagulant, consisting of polyferric sulphate (PFS) and polyacrylamide (PAA), Wat. Res., 43 (2009) 3511–3524.

[13] J. Wei, B. Gao, Q. Yue, Y. Wang, W. Li and X. Zhu, Comparison of coagulation behavior and floc structure characteristic of different polyferric-cationic polymer dual-coagulants in humic acid solution, Wat. Res., 43 (2009) 724–732.

[14] J.C. Wei, B.Y. Gao, Q.Y. Yue, Y. Wang and L. Lu, performance and mechanism of polyferric-quaternary ammonium salt composite flocculants in treating high organic matter and high alkalinity surface water, J. Hazard. Mater., 165 (2009) 789–795.

[15] B.Y. Gao, Y. Wang and Q.Y. Yue, The chemical species distribution of aluminum in composite flocculants prepared from polyaluminum chloride (PAC) and polydimethyldiallylammonium chloride (PDADMAC), Acta Hydrochim. Hydrobiol., 33 (2005) 365–371.

[16] Y. Wang, B. Gao and Q. Yue, Study on residual aluminum and mechanism of coalgulation of composite flocculants prepared by polyaluminum chloride and ploydimethyldiallylammonium chloride, Environ. Chem., 22 (2003) 474–477.

[17] J. Guan, T.D. Waite and R. Amal, Rapid structure characterization of bacterial aggregates, Environ. Sci. Tech., 32 (1998) 3735–3742.

[18] A.I. Schaefer, A.G. Fane and T.D. Waite, Nanofiltration: Principles and Applications, 1st ed., Elsevier Advanced Technology, Oxford, 2005.

[19] K. Listiarini, W. Chun, D.D. Sun and J.O. Leckie, Fouling mechanism and resistance analyses of systems containing sodium alginate, calcium, alum and their combination in dead-end fouling of nanofiltration membranes, J. Membr. Sci., 344 (2009) 244–251.

[20] J. Lee, S. Lee, M. Jo, P. Park and J. Kwak, Fouling mechanism and resistance analyses of systems containing sodium alginate, calcium, alum and their combination in dead-end fouling of nanofiltration membranes, Environ. Sci. Tech., 34 (2000) 3780–3788.

[21] T. Carroll, S. King, S.R. Gray, B.A. Bolto and N.A. Booker, The fouling of microfiltration membranes by NOM after coagulation treatment, Wat. Res., 34 (2000) 2861–2868.

[22] J. Wang, J. Guan, S.R. Santiwong and T.D. Waite, Characterization of floc size and structure under different monomer and polymer coagulants on microfiltration membrane fouling, J. Membr. Sci., 321 (2008) 132–138.