

Enhanced removal of arsenic and fouling mitigation of nanofiltration process via coagulation pretreatment

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ABSTRACT

Arsenic contamination has been considered as one of the serious environmental issues around the world. In this study, the conventional chemical coagulation was applied as pretreatment of the nanofiltration process to enhance the removal efficiency of arsenite (As(III)) and arsenate (As(V)) and mitigate the membrane fouling. Three types of coagulants including aluminum chloride (AlCl₃), polyaluminium chloride and ferric chloride (FeCl₃) were compared. More than 80% of As(V) could be removed by coagulation, nanofiltration, and their combined process, whereas nanofiltration or coagulation using aluminium-based coagulants only achieved approximately 10% of removal efficiency of As(III). FeCl₃ was proven to be the most effective coagulant to remove both As(V) and As(III) with the removal efficiency of 99% and 95%, respectively. The coagulation pretreatment was able to significantly reduce the declining trend of water flux. The formed fouling layer on the membrane surface after the treatment was investigated by a field-emission scanning electron microscopy. Compared to the case of direct filtration of humic acid solution, a more loose and permeable fouling layer could be formed when coagulation was used as a pretreatment process.

Keywords: Arsenate; Arsenite; Coagulation; Nanofiltration; Enhanced removal efficiency; Fouling mitigation

1. Introduction

Arsenic contamination is still a severe environmental problem in such regions as China, Vietnam, Mexico, and the USA. The long-term exposure to arsenic even at a low concentration can lead to adverse effects on human health [1,2]. The maximum permissible contaminant level of arsenic in drinking water is therefore set as 10 µg/L by the World Health Organization (WHO) according to epidemiological evidence of arsenic's carcinogenicity [3,4]. Arsenic can exist in many oxidation states, with As(V) and As(III) as the most common forms in water. Due to the neutral charge of As(III) in natural water, it is generally more difficult to remove As(III)

than As(V). The pre-oxidation process in which As(III) can be effectively oxidized to As(V) is therefore widely applied prior to other techniques when As(III)-containing water is treated.

Numerous treatment technologies have been applied for removing arsenic from water, including adsorption [5], membrane technology [6], and coagulation [7]. Among them, the membrane technology is considered as the most effective solution for the simultaneous removal of multiple contaminants with advantages of no addition of chemical reagents, operation in continuous mode, ease of scale-up and combination with other unit processes [8]. The application of membrane technology in the removal of arsenic

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from water has been reported in the literature [9–14]. For microfiltration or ultrafiltration typical of loose membranes, the rejection efficiency of arsenic is unsatisfied owing to the larger pore size of the membrane than arsenic molecules. In comparison, nanofiltration (NF) can achieve a comparable removal efficiency of As(V) at a far lower energy consumption than reverse osmosis. However, the pre-oxidation process is still needed to increase the rejection of NF membranes towards As(III) (e.g., from 50%–63% to 97%–100% [12]). Conventional chemical coagulation was also reportedly effective to remove arsenic from water and the removal efficiency may vary a lot according to type and dosage of coagulants, a form of arsenic species and water chemistry [15–18]. Meanwhile, coagulation is also proven to be effective for removing colloids and inorganic/organic matters [19,20]. Thus, a combined effect on the enhancement of arsenic removal efficiency and alleviation of membrane fouling can be expected by applying the coagulation process as the pretreatment of nanofiltration. Although application potential of hybrid coagulation–nanofiltration process in water treatment has been heavily studied in recent years [21–24], the performance on the removal of both As(V) and As(III) has been rarely reported.

Herein, the enhanced effect of coagulation pretreatment on the arsenic removal efficiency of the nanofiltration process was systematically studied. Three different types of commonly used coagulants including AlCl_3 , polyaluminium chloride (PAC) and FeCl_3 were compared in terms of arsenic removal efficiency and membrane fouling mitigation. The influence of natural organic matters (NOM) in arsenic removal by coagulation, nanofiltration, and their hybrid process was also investigated.

2. Materials and methods

2.1. Materials

All chemicals used in this study were of reagent grade. The stock solution of As(V) and As(III) with a concentration of 40 mg/L was prepared by dissolving $\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$ and Na_3AsO_3 into deionized water (DI) water, respectively. Humic acid (HA, Sigma-Aldrich, USA) was chosen as the representative of NOM. The HA stock solution was prepared by dissolving 2 g of HA in 1,000 mL of DI water. AlCl_3 , PAC and FeCl_3 were purchased from the Beijing Chemical Company (Beijing, China) and used as coagulants. NF270 membrane was purchased from the Dow FilmTec Company (USA). The molecular weight cut-off (MWCO) of the membrane is approximately 200 g/mol, and pure water flux is 3.8×10^{-8} m/s kPa.

2.2. Coagulation process

All the coagulation experiments were conducted in lab-scale speed controllable agitation tanks. The arsenic concentration ([As]) in solution was 200 $\mu\text{g/L}$, and 5 mM NaHCO_3 was added as background electrolyte. Solution pH was adjusted to 7.5 by adding hydrochloric acid (HCl) and sodium hydroxide (NaOH). To investigate the influence of NOM in arsenic removal, HA was added into the above-mentioned solutions at the HA concentration of 10 mg/L. The mixture solution was then stirred for 24 h to reach equilibrium.

After adding a certain amount of coagulants, the solution was immediately stirred at 250 rpm for 30 s, followed by rapid mixing at 200 rpm for 1 min. The stirring speed was then reduced to 40 rpm for 15 min to allow floc growth to occur. The suspension was left undisturbed for 20 min before taking samples for arsenic concentration measurement. The concentration of HA was determined using a UV spectrophotometer (HACH Corp., USA) at a wavelength of 254 nm. The arsenic concentration was measured using an inductively coupled plasma optical emission spectrometer (ICP-OES, PerkinElmer Optima 3000, USA). The removal efficiency of arsenic can be calculated according to Eq. (1):

$$R = \frac{C_f - C_p}{C_f} \times 100\% \quad (1)$$

where R (%) is the removal efficiency, C_f and C_p (mg/L) are the arsenic concentrations in the feed and permeate, respectively.

2.3. Nanofiltration and hybrid coagulation–nanofiltration processes

Nanofiltration and hybrid coagulation–nanofiltration experiments were carried out by using a dead-end filtration unit consisted of an N_2 gas cylinder, coagulation reactor, membrane filtration cell, electronic balance and a computer as shown in Fig. 1. A piece of NF270 membrane with an effective area of 12.6 cm^2 was soaked in a DI water bath for at least 24 h and then compacted by filtering DI water until a steady-state flux was obtained. After that, arsenic solutions with or without coagulation pretreatment were filtered at the transmembrane pressure (TMP) of 3.5 bar. To measure water flux of the membrane, the weight of permeate water was recorded by electronic balance and computer at different time intervals. After the filtration, the membrane was carefully rinsed by DI water and dried in air. Normalized permeate flux (J_N) was employed to evaluate the membrane fouling as expressed below:

$$J_N = \frac{J}{J_0} \quad (2)$$

where J_N is the normalized permeate flux of membrane, J and J_0 ($\text{L/m}^2 \text{ h bar}$) are the filtrate flux of testing solutions and DI water, respectively.

2.4. Field-emission scanning electron microscopy analysis

The surface morphology of pristine and fouled membranes was examined by field-emission scanning electron microscopy (FESEM) (Hitachi S4500, Japan) at an acceleration voltage of 15 kV. The dried membrane samples were first coated by a thin layer of Pt to obtain electrical conductivity.

2.5. Zeta potential measurement

Zeta potential of pristine and fouled membranes was measured by an electrokinetic analyzer (SurPASS, Anton Paar GmbH, Austria). A nitrogen headspace and a 10 mM NaCl background electrolyte were applied for all measurements.

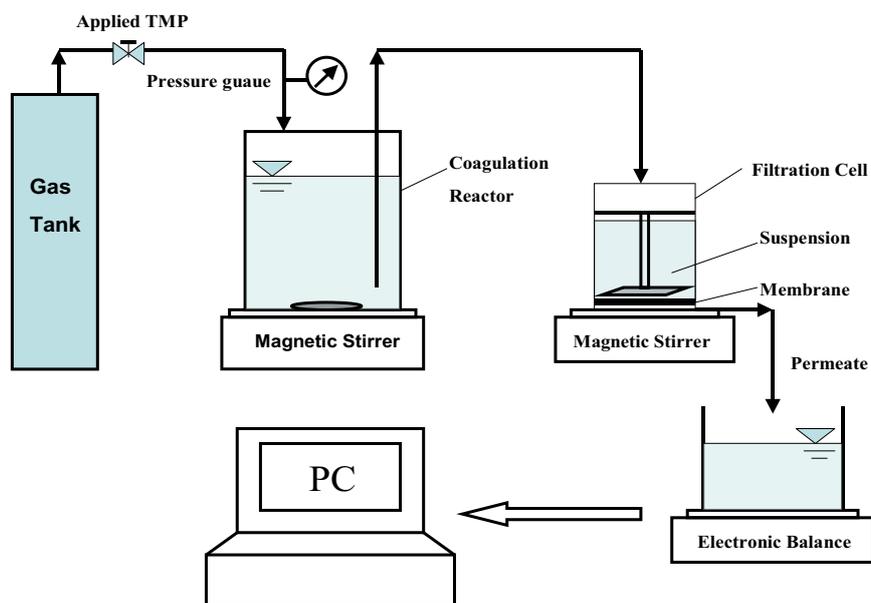


Fig. 1. Schematic diagram of the dead-end filtration unit used in this study.

3. Results and discussion

3.1. Performance of the coagulation process

As shown in Fig. 2, the removal of HA can be significantly enhanced by increasing the dosage of three common coagulants. The optimal dosage of PAC, AlCl_3 , and FeCl_3 at which more than 95% of HA can be removed from the water was determined as 0.40, 0.50 and 0.90 mM, respectively.

The removal efficiency of As(V) and As(III) in the coagulation process was investigated and the results are shown in Fig. 3. More than 80% of removal efficiency of As(V) could be achieved by all the coagulants, while only approximately 10% of As(III) can be removed by Al-based

coagulants. Similar to other studies, better removal of As(III) could be accomplished by using FeCl_3 coagulant [17,25]. The removal mechanism of arsenic in the coagulation process is proposed as adsorption or bridging of arsenic species on active sites which are generated through hydrolysis and polymerization of iron or aluminum salts, followed by separation from the water with settling flocs [26]. Thus, negatively charged As(V) ions would be more easily removed in the coagulation process due to the electrostatic attraction between As(V) ions and positively charged metal hydroxide precipitates. In contrast, As(III) becomes neutrally charged until $\text{pH} > 9$, leading to a weak interaction with metal hydroxide or flocs in water. It is

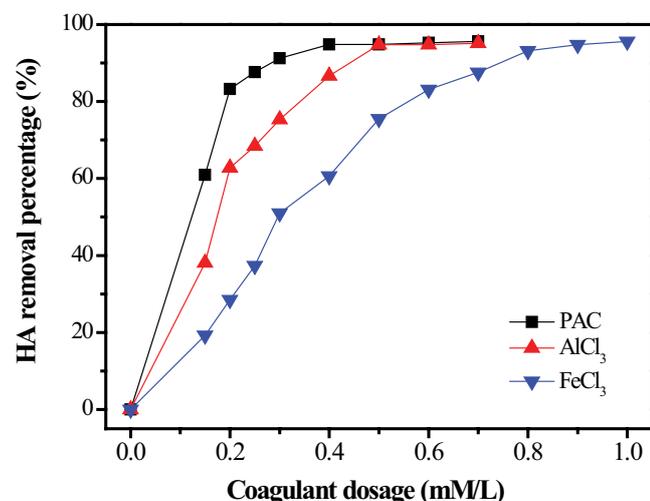


Fig. 2. Removal percentage of HA by adding different dosages of PAC, AlCl_3 and FeCl_3 . Experimental conditions: $[\text{HA}] = 10 \text{ mg/L}$ and $T = 20^\circ\text{C}$.

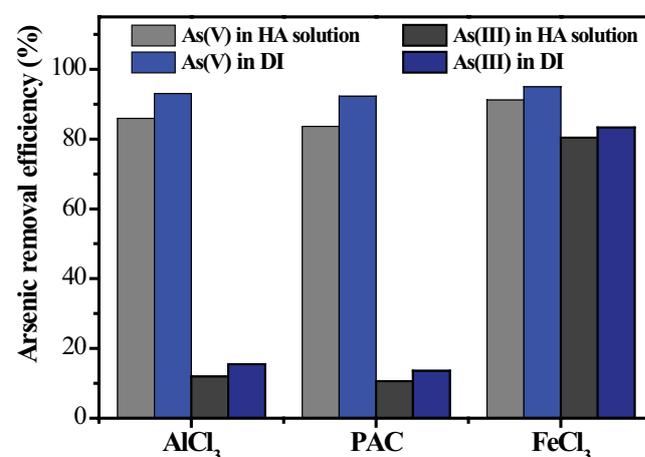


Fig. 3. Removal efficiency of As(V) and As(III) in coagulation process. Experimental conditions: $[\text{PAC}]$ dosage = 0.40 mM, $[\text{AlCl}_3]$ dosage = 0.50 mM, $[\text{FeCl}_3]$ dosage = 0.90 mM, $[\text{HA}] = 10 \text{ mg/L}$, and $[\text{As}] = 200 \text{ }\mu\text{g/L}$.

worthwhile to note that the removal of arsenic is decreased to some extent at the presence of HA in solution. This is mainly due to the competitive adsorption between HA and arsenic on metal-based precipitates [27–29]. In addition, owing to the presence of carboxylic and phenolic groups in HA, the complex between HA molecules and metal ions (e.g., Al^{3+} and Fe^{3+}) can be rapidly formed by electrostatic interaction in which the hydrolysis process of coagulants would be disturbed [30].

3.2. Performance of the nanofiltration process

The performance of the nanofiltration process on the removal of As(V) and As(III) was investigated by using a dead-end filtration module at the operating pressure of 3.5 bar. The arsenic concentrations at three different permeate volumes were determined. As shown in Fig. 4, approximately 80% of As(V) can be effectively rejected by the NF270 membrane, while the As(III) rejection efficiency was only around 10%. The rejection mechanism of the nanofiltration membrane is generally considered as size exclusion and electrostatic interaction between negatively charged membrane surface and pollutants. At pH 7.5, the dominant form of As(V) and As(III) is HAsO_4^{2-} and H_3AsO_3 , respectively [31]. The molecular weight of HAsO_4^{2-} and H_3AsO_3 are 143 and 126 g/mol, lower than the MWCO of the NF270 membrane. Therefore, electrostatic repulsion between NF270 membrane surface and arsenic species should play a more important role in the rejection of As(V) and As(III).

The removal efficiency of arsenic is slightly increased at the presence of HA in solution. The NOM fouling layer can affect the membrane rejection performance by changing the membrane surface charge, pore distribution and concentration polarization phenomenon [32]. Membrane surface pore blocking and cake formation caused by the attachment of HA molecules may enhance the arsenic rejection. The salt rejection was also reportedly enhanced by HA deposition on NF membrane surfaces (NF90 and NF270) through the plugging of the least resistant paths (“hot spots”, having high local flux and low salt rejection) [33]. Moreover,

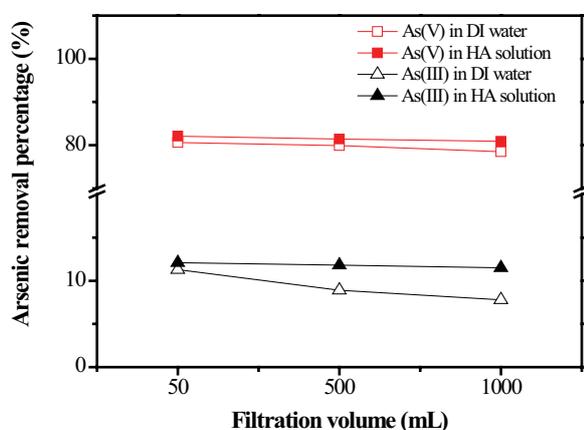


Fig. 4. Removal percentage of As(V) and As(III) in nanofiltration process. Experimental conditions: [HA] = 10 mg/L, [As] = 200 $\mu\text{g/L}$, and TMP = 3.5 bar.

according to the zeta potential of pristine, HA-fouled and floc-fouled NF270 membranes as presented in Fig. 5, the pristine NF270 membrane has a strongly negative surface charge of -34.6 mV, and the surface charge is further decreased to -40.9 mV after filtering the HA solution. The lower zeta potential of the HA-fouled membrane surface can cause stronger electrostatic repulsion between As(V) ions and NF270 membrane. Notably, the change of nanofiltration membrane surface charge after treatment seems to depend on the membrane type. As reported by Bellona et al. [34], the surface charge of TFC-S (Koch membrane systems NF membrane) and NF270 membranes could be decreased after treating secondary treated wastewater effluent, while it was found to be increased for the NF90 membrane.

A slight reduction in the rejection efficiency with the filtration volume can be observed in Fig. 4. This might be due to the occurrence of concentration polarization on the membrane surface with the filtration time.

3.3. Performance of hybrid coagulation-nanofiltration process

The removal efficiency of arsenic and HA by hybrid coagulation-nanofiltration process is shown in Fig. 6. Benefiting from the excellent removal capability of coagulation and nanofiltration processes, more than 98% of HA can be effectively removed in all experiments. As shown in Fig. 6a, over 92% of As(V) can be removed by the hybrid coagulation-nanofiltration process. Especially, the removal efficiency of As(V) can reach higher than 98% when FeCl_3 was used as a coagulant. Similar to the nanofiltration process, a decrease in the arsenic rejection with the filtration time is observed. The fouling layer formed by the deposition of coagulation flocs should be looser and more permeable (will be discussed later). The pore-blocking on the membrane surface is therefore negligible. As well known, the zeta potential of coagulation flocs is generally around the isoelectric point. As seen from Fig. 5, the zeta potential of the NF270 membrane turns to be more positive after filtering the coagulation solution (-30.8 mV), which could weaken the electrostatic repulsion between membrane and As(V) ions.

As shown in Fig. 6b, the removal efficiency of As(III) can vary from 16.5%–26.2% for AlCl_3 and 12.9%–15.8% for PAC, respectively. However, when FeCl_3 was used as a coagulant,

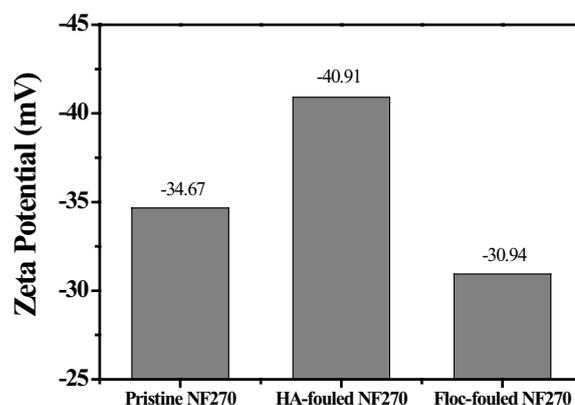


Fig. 5. Zeta potential of membranes before and after filtration.

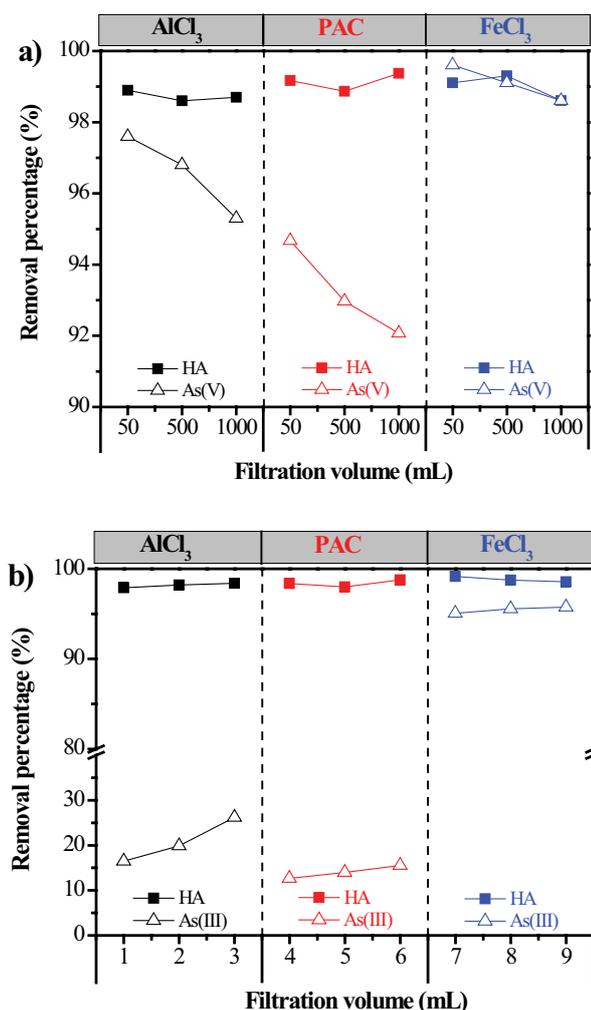


Fig. 6. Removal percentage of arsenic and HA in hybrid coagulation-nanofiltration process: (a) As(V) and (b) As(III). Experimental conditions: [HA] = 10 mg/L, [As] = 200 µg/L, and TMP = 3.5 bar.

the hybrid coagulation-nanofiltration process can achieve higher than 95% removal efficiency of As(III) in which the permeate concentration of arsenic is below 10 µg/L, meeting the drinking water standard set by the WHO. As stated above, electrostatic repulsion between membrane and As(III) can be ignored. Meanwhile, the size exclusion effect of the membrane is hardly influenced by the floc layer formed on the membrane surface. The increase in the rejection efficiency of As(III) with the filtration time is likely caused by the adsorption of As(III) in the floc layer that can work as a second functional membrane.

3.4. Water flux and membrane fouling

To investigate the effect of coagulation pretreatment on membrane fouling mitigation, the normalized permeate fluxes of membranes when filtering 10 mg/L HA solution and coagulant effluents were measured at the operating pressure of 3.5 bar. As shown in Fig. 7, the most significant

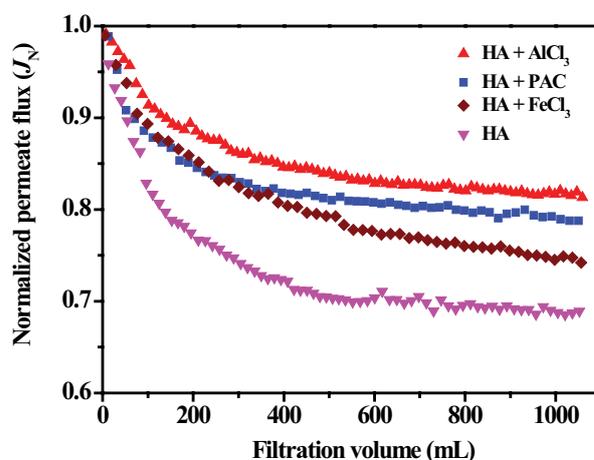


Fig. 7. Normalized permeate flux of hybrid coagulation-nanofiltration process. Experimental conditions: [HA] = 10 mg/L, [PAC] dosage = 0.40 mM, [AlCl₃] dosage = 0.50 mM, [FeCl₃] dosage = 0.90 mM, and TMP = 3.5 bar.

permeate flux decline occurs when the HA solution is directly filtered. The normalized permeate flux decreases by about 32% at the filtration volume of 1,000 mL. In contrast, the flux reduction trend can be slowed down when the coagulation process is applied as pretreatment. The improvement in flux decline by three types of coagulants follows the order: AlCl₃ > PAC > FeCl₃. The hybrid coagulation-nanofiltration process suffers about 19%, 22%, and 25% of flux reduction for AlCl₃, PAC and FeCl₃, respectively. Since the removal efficiency of HA is the same for three coagulants at chosen dosages, the flux decline trend could be determined by properties of the floc layer on the membrane surface.

As shown in Fig. 8 and Table 1, the floc size follows the order of FeCl₃ > AlCl₃ > PAC, while the fractal dimension (D_f) of floc is in the order of FeCl₃ > PAC > AlCl₃. A higher D_f is reportedly corresponding to a more compact floc structure [35]. Thus, the flocs formed by FeCl₃ coagulant is larger

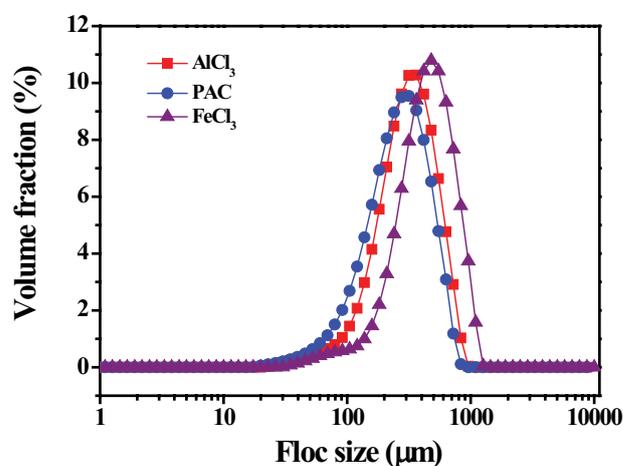


Fig. 8. The volume fraction of different floc sizes formed by AlCl₃, PAC and FeCl₃.

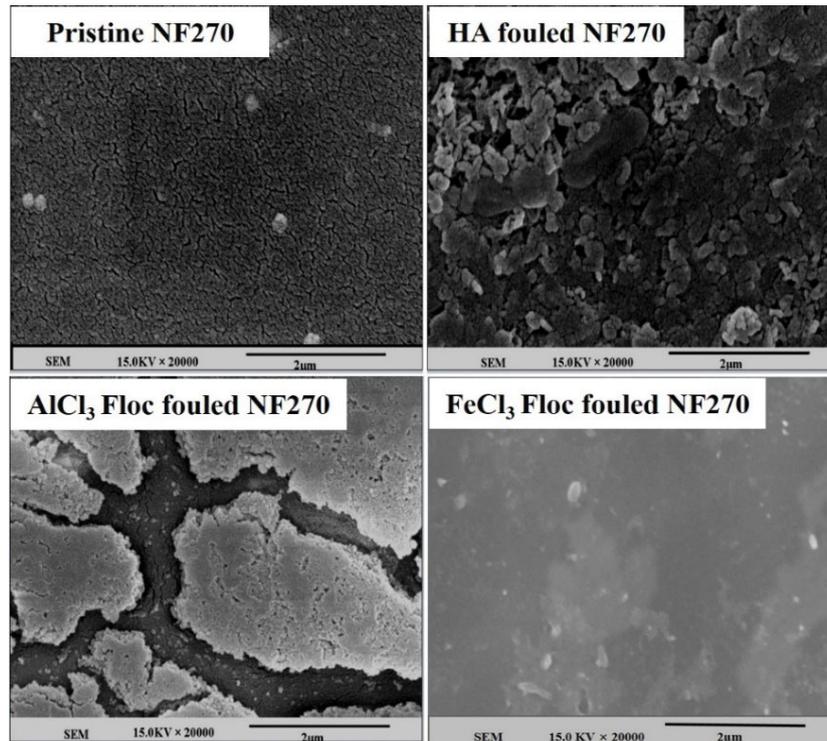


Fig. 9. FESEM images of pristine, HA-fouled, AlCl_3 floc-fouled and FeCl_3 floc-fouled NF270 membranes.

Table 1
Mean floc size $d(50)$ and fractal dimension (D_f) of different coagulants

Floc parameter	Coagulant		
	AlCl_3	PAC	FeCl_3
$d(50)$, μm	335.7	287.5	473.2
D_f	2.47	2.56	2.83

but more compact, leading to the formation of a tight and high-resistance fouling layer on the membrane surface. In contrast, AlCl_3 flocs have the smallest D_f value and a relatively larger floc size than PAC. This might address the issue of why a better improvement in the permeate flux of NF270 membrane can be achieved by AlCl_3 coagulant.

The surface morphology of membranes after filtering HA solution and coagulant effluents were observed by FESEM. As shown in Fig. 9, the pristine NF270 membrane has a porous surface with a pore size of several hundred nanometers. A thick and dense fouling layer is observed on the membrane surface after directly filtering the HA solution. The fouling layer formed by AlCl_3 flocs becomes more permeable and loosely compacted. Additionally, the presence of cracks on the fouling layer which are formed during the drying procedure also indicates that the water content in the floc layer should be higher than the HA fouling layer. Water molecules can, therefore, more easily pass through the floc layer during the filtration process. In comparison, the fouling layer formed by FeCl_3 flocs is much tighter than AlCl_3 .

4. Conclusions

A hybrid coagulation-nanofiltration process was applied to effectively remove both As(III) and As(V) and mitigate membrane fouling. Compared to As(III), As(V) can be more easily removed by coagulation, nanofiltration, and the hybrid process. Only about 10% of As(III) can be removed by coagulation or nanofiltration when aluminium-based coagulants were used. In contrast, FeCl_3 coagulant was proven to be more effective for the arsenic removal by the hybrid coagulation–nanofiltration process in which over 98% of As(V) and 95% of As(III) can be successfully removed from the water at the initial arsenic concentration of 200 $\mu\text{g/L}$. The water flux decline of membranes can be retarded by the coagulation pretreatment. A more permeable and looser fouling layer was found to be formed by coagulation effluent than the HA solution. The findings in this study show the hybrid coagulation–nanofiltration process has a promising application potential on the treatment of arsenic-containing water.

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