Enhanced antimony(V) removal using synergistic effects of Fe hydrolytic flocs and ultrafiltration membrane with sludge discharge evaluation

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ABSTRACT

The integration of adsorbents with ultrafiltration (UF) membranes is a promising method for alleviating membrane fouling and reducing land use. However, a number of problems have become apparent concerning the granular adsorbents used currently, such as high running cost, high chance of causing membrane surface damage, low in situ chemical cleaning efficiency, etc. Herein, to overcome these disadvantages, loose in situ hydrolyzed flocs were directly injected into the membrane tank, providing strong adsorption ability at low cost. To test the feasibility of this method, the heavy metal pollutant antimony (Sb(V)) in a water plant was chosen as a test case, which is similar to arsenic and difficult to remove. We found that Fe-based flocs integrated with an UF membrane showed a large potential advantage in removing Sb(V), even after running for 110 days. We demonstrated that the observed slow transmembrane pressure development could be ascribed to the loose floc cake layer formed, even though some extracellular polymeric substances were induced during operation. We also found that the floc cake layer was easily removed by washing with feed water or dissolved by in situ chemical cleaning under strongly acidic conditions, and many primary membrane pores were clearly observed. In addition, a relative long sludge discharge interval was feasible for this technology and the effluent quality was good, including the turbidity, chromaticity and iron concentration. Based on the excellent performance, these flocs integrated with UF membranes indeed show potential for application in water treatment.

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1. Introduction

Drinking water treatment technology has been significantly developed in the past few decades, mainly due to developments in materials science (Ajmani et al., 2012). The risks of water quality are of great concern because of the serious water pollution due to increased industrialization, and new methods of guaranteeing water quality are urgently needed. Here, most studies have focused on the application of new materials, aiming to improve the traditional water treatment process with novel technologies. As a result, membrane technology has been widely used in water treatment (Huang et al., 2009; Drioli et al., 2011), and has even been predicted

to become the main water treatment technology in the next few decades (Shannon et al., 2008).

However, researchers have demonstrated that membrane fouling is inevitable as a function of time due to the accumulated pollutants in membrane pores or on the membrane surface (Wang and Tarabara, 2008; Wu et al., 2011). To alleviate membrane fouling, different pretreatment technologies have been investigated to combine coagulation/adsorption and membrane filtration: pre-adsorption, direct-filtration and integrated filtration. Owing to the better membrane performance and smaller land use, more studies have been focused on integrated filtration in recent years (Kim et al., 2010; Cai and Benjamin, 2011; Zhang et al., 2016).

Up to now, various kinds of adsorbents have been used in the integrated filtration process to test their antifouling properties, such as heated iron oxide particles (Zhang et al., 2003), heated aluminum oxide particles (Kim et al., 2008), carbon nanotubes

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(Ajmani et al., 2012), powdered activated carbon (Cai et al., 2013), nanoscale zerovalent iron (Ma et al., 2015), etc. Although these granular adsorbents showed good performance, many problems were found during filtration, including relatively high operation cost, high chance of causing membrane surface damage due to shearing force, and low in situ chemical cleaning efficiency due to the dense cake layer formed by extracellular polymeric substances (EPS). It is known that inexpensive aluminum salts and iron salts are widely used in water treatment, whose flocs exert little shearing force on membranes. Flocs formed in situ, which maintain the adsorption sites as much as possible, were found to have stronger adsorption ability than pre-made adsorbents (Liu et al., 2011; He et al., 2015). To overcome the abovementioned problems, in situ prepared flocs were directly injected into the membrane tank in the current research, which were fully suspended in the membrane tank by aeration from the bottom.

In addition, the sludge discharge interval is one of the main running conditions, which is also crucial to UF membrane fouling (Ozdemir and Yenigun, 2013). It has been reported that sludge discharge could benefit different reversible and irreversible fouling behaviors because of the various compositions of retentate solution in the membrane tank (Bai et al., 2013; Yu et al., 2015). For the water plant, a longer sludge discharge interval not only means a higher water production rate, but also means a smaller amount of sludge produced and lower running cost. However, little is known about the effect of sludge discharge frequency on the integrated membrane process. Furthermore, most research has been focused on the behavior of organic pollutants during membrane filtration (Abdelrasoul et al., 2013; Tabatabai et al., 2014; Meng et al., 2015), while fewer studies have paid attention to the removal of smaller molecular weight (MW) organic matter, especially heavy metals (Barakat and Schmidt, 2010).

Here, to fully investigate this integrated membrane process, antimony (Sb) was chosen, which is similar to arsenic and is also difficult to remove (He et al., 2015). It has caused great concern due to its potential toxicity and carcinogenicity towards humans after long-time exposure (Wilson et al., 2010). The maximum contaminant level (MCL) in drinking water for Sb is 5 µg/L in the USA and China (Filella et al., 2002a, 2002b; Xu et al., 2011). Sb(III) and Sb(V) are the two main species, of which Sb(III) is dominant under anoxic conditions with the main species of Sb(OH)₃, while Sb(V) is dominant in oxic surface waters with the main species of Sb(OH)₆⁻ over a wide pH range (Filella and May 2005; He et al., 2015).

The current test was carried out in a drinking water plant in the province of Hunan, located in central China (N: 27.7°; E: 111.2°). The concentration of Sb is relatively high in the surface water of the local area owing to mining, and even in the effluent of the water plant. Fe flocs hydrolyzed in situ were used in this study due to the strong adsorption affinity toward Sb of iron salts, especially under acidic conditions (Leuz et al., 2006; He et al., 2015). To understand the membrane behavior in detail, the effect of sludge discharge frequency was evaluated. Some factors responsible for the membrane performance were also investigated, such as the running time, the in situ chemical cleaning efficiency, etc.

2. Materials and methods

2.1. Characteristics of feed water

Table 1 presents the specific characteristics of the feed water. The main species of antimony in the effluent was Sb(V) because of the dissolved oxygen in the surface water. Owing to the low removal efficiency of traditional water treatment, the concentration of Sb(V) in effluent was higher than the MCL before upgrading the plant. To enhance the removal efficiency toward Sb(V), the final

Table 1

The characteristics of feed water.

Parameters	Feed water
Water temperature (°C)	8.9–28.5
pH	7.3–7.9
Concentration of Sb (V, µg/L)	4.1–13.7
Turbidity (NTU)	0.2–2.1
Chromaticity	1.0–8.0
Residual chlorine (mg/L)	0.3–0.9
Total organic matter (TOC, mg/L)	0.5–3.8
UV ₂₅₄ (cm ⁻¹)	0.01–0.16

solution pH in the membrane tank was maintained at 6 with 1 M HCl by using a peristaltic pump, and no additional disinfection was used during the experiment.

2.2. Preparation of flocs

All chemical reagents were analytical grade except when specified, and FeCl₃·6H₂O (Sinopharm chemical reagent Beijing Co., Ltd, China) was used. The coagulants were dissolved in 400 mL deionized (DI) water each time and the corresponding flocs were prepared by adjusting the solution pH to 6 with 1 M NaOH. To prevent a high concentration of iron in effluent, the prepared flocs were washed by DI water three times before using. The concentration of Fe-based flocs was almost equal to the concentration of Fe-based coagulants, because Fe hydrolytic flocs are the dominant iron species around neutral pH conditions (Eilbeck and Mattack, 1987).

2.3. Adsorption kinetics of Sb(V) on flocs and EPS

Deionized (DI) water and potassium antimony tetrahydrate (KSbO₃·3H₂O) were used for all the adsorption kinetic experiments. In order to obtain a proper dose of flocs during the test afterward, 20 µg/L Sb(V) was chosen according to the highest concentration observed (18.9 ± 0.9 µg/L) in the effluent. The prepared Sb(V) solution was added into a 1 L beaker with magnetic agitation (100 rpm) in the presence of 0.01 M NaCl to provide background ionic strength at pH 6. The prepared flocs were dosed with the concentrations of 1, 2, 5, 10, 15 mg/L as Fe.

To fully understand the adsorption kinetics of Sb(V) on EPS, bovine serum albumin (BSA) and polysaccharide (SA) were used to simulate EPS (Coseza et al., 2013). The adsorption kinetic experiment procedures were similar to those used for the adsorption kinetics of Sb(V) on flocs. The BSA, SA or their 1:1 mixture were dosed with the concentrations of 0.5, 1, 5, 10, 20, 50, 100 mg/L. All adsorption kinetic experiments were conducted at room temperature.

2.4. Filtration process

Fig. S1 shows the specific schematic illustration of the integrated membrane process. Feed water was continuously fed into the membrane tank, in which a hollow-fiber membrane module (polyvinylidene fluoride, Litree, China) was immersed. The membrane module was composed of 30 membranes, each one being 53 cm long. The inner diameter of the membrane tank was 64 mm, and the height was 800 mm. The effluent from the membrane was pumped by a peristaltic pump (1 L/h), which was operated in a cycle of 30 min filtration and 1 min backwashing (2 L/h). A ceramic aerator (diameter: 40 mm) was placed in the bottom, allowing flocs to be suspended in the membrane tank. The flocs were freshly prepared before injecting to maintain their activity (Chen et al., 2015).

During filtration, the flocs were injected into the membrane tank with the peristaltic pump. The transmembrane pressure (TMP) was monitored by pressure sensors. The hydraulic retention time was kept at 2.2 h and the accumulated sludge was released once every 5 days or 10 days. Before sludge discharge, the aeration was ceased for 30 min, and then half of the sediment was discharged.

At day 100, the membrane module with sludge discharge frequency of 5 days (water production rate 99.5%) was taken out carefully and washed with feed water, aiming to understand the contribution of cake layer resistance. For the membrane module with sludge discharge frequency of 10 days (water production rate: 99.7%), the solution pH in the membrane tank was adjusted to 2 by HCl and maintained at that pH for 1 h at day 100, aiming to investigate the efficiency of in situ chemical cleaning of the cake layer. Before measuring the morphology of the layered membrane surface with scanning electron microscopy (SEM, JSM-7401F, JEOL Ltd., Japan), 2 cm of membrane was carefully cut off from the middle of the membrane module at day 100, while the ends were tied in knots to ensure normal operation in the filtration afterward.

2.5. Characteristics of flocs in the membrane tank

To minimize the impurities and characterize the flocs accurately, floc samples were taken from below the water surface in membrane tank at day 1. A laser diffraction instrument (Mastersizer 2000, Malvern, UK) was used to measure the dynamic floc size and fractal dimension (D_f) of flocs. Floc size was taken every 30 s and recorded by a computer, while D_f was calculated based on the small-angle light scattering, which has been described extensively by other studies (Jarvis et al., 2005; Yu et al., 2012). In addition, the surface charge of flocs was measured by a nano-particle sizing & zeta potential analyzer (BECKMAN COULTER Ltd., USA), and the specific surface area of flocs was measured by the Brunauer-Emmett-Teller method (BET, ASAP2020HD88). The corresponding characteristics of the Fe-based flocs were shown in Table S1.

2.6. Measurement of EPS in the sludge

The sludge was analyzed by the methods described below to characterize the membrane fouling. To prevent EPS being released from the bacterial cells, a heating and extraction method was modified to extract the EPS from the sludge (Morgan et al., 1990). The sludge suspension was first dewatered by centrifugation (Model 5417C, Eppendorf, Germany) in a 50 mL tube at 3000 rpm (5 min). The sludge pellet in the tube was re-suspended in 20 mL phosphate buffer saline solution, sheared by a vortex mixer (Vortex-Genie 2, Mo Bio Laboratories, Inc, USA) for 15 min, ultrasonically treated for 2 min, and heated to 80 °C for 30 min. The mixture was centrifuged at 10,000 rpm for another 15 min, and then the supernatant solution was collected for EPS analysis.

Following a modified Bradford method (Peterson, 1977), BSA was used as the standard to quantify the absolute concentrations of proteins. The phenol-sulfuric acid method was used to quantify the absolute concentration of SA in EPS with glucose as the standard (Dubios et al., 1956).

2.7. Other analytical measurements

Solution pH was measured by a pH meter (Orion benchtop) and water temperature was obtained using a thermometer. The UV absorbance of the waters was measured at a wavelength of 254 nm (with 0.45 µm sample pre-filtering) by using an ultraviolet/visible spectrophotometer (U-3010, Hitachi High Technologies Co., Japan). TOC was measured by a total organic matter analyzer (TOC-Vcph, SHIMADZU, Japan). The concentrations of Sb(V) and iron in the

effluent were measured by inductively coupled plasma mass spectrometry (ICP-MS, 7500a, Agilent Technology, USA).

3. Results and discussion

3.1. Kinetics of Sb(V) adsorption on Fe-based flocs

Sb(V) adsorption on Fe-based flocs was investigated at pH 6 (Fig. 1). It was found that the concentration of Sb(V) in the effluent was higher than 5 µg/L after 300 min when the concentration of flocs was lower than 2 mg/L. However, when the floc concentration increased to 5, 10, 15 mg/L, the concentration of Sb(V) was lower than 5 µg/L after 120, 20, 5 min, respectively. The higher removal efficiency was ascribed to the strong electrostatic interaction between the positively charged Fe-based flocs (Table S1) and the negatively charged Sb(V) ($\text{Sb}(\text{OH})_6^-$), which was in accordance with the previous studies (Leuz et al., 2006; He et al., 2015).

As seen from Fig. 1, the Fe-based flocs at 2 mg/L did not have enough adsorption ability to reach the critical concentration for Sb(V) (5 µg/L), but the dosage of 5 mg/L Fe-based flocs provided optimum reactive surface for removing Sb(V). The equilibrium time decreased with increasing concentration of Fe-based flocs due to the abundant surface sites for adsorption. Although the concentration of Sb(V) was lower than 5 µg/L in the presence of 5 mg/L Fe-based flocs, previous studies have demonstrated that common co-existing anions showed different effects on the adsorption of Sb (Wu et al., 2010; Xu et al., 2011), especially phosphate (He et al., 2015). Owing to the similar structures of phosphorus and antimony anions, they would compete for the same adsorption sites (Ghosh et al., 2006; Kolbe et al., 2011). In addition, the concentration of Sb(V) in the effluent was 4.01 ± 0.58 µg/L, which was close to the MCL. Thus, 10 mg/L Fe-based flocs was chosen as the optimum concentration for further experiments.

3.2. Variation of effluent quality

To investigate the effluent quality achieved with the integrated membrane process, the concentration of Sb(V) was first analyzed. As seen from Fig. 2, the variation of Sb(V) concentration in the influent was large due to variations in the local water sources, varying between 4.1 µg/L and 13.7 µg/L during the test. However, the removal efficiency of Sb(V) was high and the corresponding concentration in the effluent was lower than 1 µg/L, even after

Fig. 1. Kinetics of Sb(V) adsorption on Fe-based flocs. Reaction conditions: initial Sb(V): 20 µg/L, 0.01 M NaCl, react for 5 h at pH 6.

Fig. 2. Variation of Sb(V) as a function of time before and after filtration with different sludge discharge frequencies.

running for 110 days.

Although Sb(V) could be adsorbed/rejected by the UF membrane or EPS in the membrane tank, the adsorption capacity was almost negligible. Fig. 2 shows that the removal efficiency of Sb(V) by the UF membrane alone was low (about 7.4%), due to the large average membrane pore diameter (30 nm, provided by manufacturer). Regarding the adsorption ability of EPS (produced by microorganisms because of non-disinfection, to be discussed later) toward Sb(V), Fig. S2 shows that little adsorption of Sb(V) occurred on protein or polysaccharide. As a result, the high Sb(V) removal efficiency was due to the strong adsorption ability of Fe-based flocs toward Sb(V), which was in accordance with the results shown in Fig. 1. In addition, due to the strong adsorption ability of Fe-based flocs, sludge discharge frequency had little influence on the Sb(V) removal efficiency, and almost no variation was observed in the concentration of Sb(V) before and after sludge discharge.

Based on this strong adsorption ability, the removal efficiency of Sb(V) was also high after running for 100 days, with periodic cleaning by washing with feed water (sludge discharge frequency of 5 days) or immersion in strongly acidic conditions (pH 2) for 1 h (sludge discharge frequency of 10 days), respectively. The concentration of Sb(V) in the effluent was 0.32 µg/L and 0.41 µg/L at day 100 and 101 with sludge discharge frequency of 5 days, while the concentrations were 0.26 µg/L and 0.28 µg/L with sludge discharge frequency of 10 days. Furthermore, Fig. S3a and S3b show that little variation in turbidity and chromaticity occurred before and after filtration with different sludge discharge frequencies. Table S1 shows that the average floc diameter was 113.9 ± 22.7 µm (considerably larger than the pore diameter of the filter membrane), resulting in low iron concentration in the effluent (Fig. S3c).

3.3. Variation of TMP

The TMP development was further investigated in the absence/presence of flocs (Fig. 3). The results showed that the TMP dramatically increased to 67.9 kPa at day 10 in the absence of flocs, while decreasing to 18.4 kPa after washing the membrane with the feed water, indicating that cake layer formation was the main fouling mechanism. The possible reason was the dense cake layer formed by EPS because of the high water temperature (Fig. S4). Fig. S5 shows that the average particle size in the feed water was smaller than 10 nm, leading to fewer possibilities of cake layer formation.

Fig. 3. TMP development as a function of time with different sludge discharge frequencies.

With continuous floc injection, the development of TMP became extremely slow. The corresponding TMP only increased to 36.1 kPa and 33.7 kPa after running for 100 days with sludge discharge frequencies of 5 and 10 days, respectively. Owing to the thicker and looser floc cake layer formed with sludge discharge frequency of 10 days (Fig. S6), more pollutants were rejected/trapped. Thus, the TMP development with sludge discharge frequency of 10 days was lower than that of 5 days.

A previous study reported that the membrane pore size became smaller and the cake layer became denser with decreasing water temperature (Sharma et al., 2003). As a result, the TMP increased relatively rapidly when the water temperature was lower. As seen from the red dotted area in Fig. 3, the water temperature was 16.6 °C at day 83, and the corresponding TMP were 22.1 and 18.3 kPa with sludge discharge every 5 and 10 days. The water temperature rapidly decreased to 12.7 °C at day 85, and the corresponding TMP significantly increased to 33.7 and 21.3 kPa, respectively. In fact, the TMP varied even during the same day due to the difference in water temperature between AM and PM (Fig. S7).

At day 100, the membrane module with sludge discharge frequency of 5 days was washed with feed water, and the TMP decreased from 36.1 kPa (day 100) to 16.9 kPa (day 101). Owing to the existence of flocs, the solution in the membrane tank was adjusted to pH 2 with HCl and maintained at that pH for 1 h for the sludge discharge frequency of 10 days. The corresponding TMP decreased from 33.7 kPa (day 100) to 12.3 kPa (day 101), indicating that the cake layer dominated by flocs was broken. Fig. 3 shows that cake layer formation was the main fouling mechanism after physical cleaning and chemical cleaning, and the in situ chemical cleaning also demonstrated the feasibility of the integrated membrane process.

3.4. Variation of EPS in the sludge

Owing to the lack of disinfection during the whole filtration, microorganisms easily proliferated in the membrane tank, especially at the appropriate water temperature (Arora and Kazmi, 2015). Fig. 4 shows that the water temperatures were 24.1, 20.1, 18.7, 15.3 and 11.9 °C at day 20, 40, 60, 80 and 100, respectively. Although the water temperature gradually decreased as a function of time, the concentrations of both protein and polysaccharide in the sludge increased first and then decreased. A potential reason was that the proliferation rate of microorganisms at day 60 (18.7 °C) was still strong, while it gradually became weak due to further

Fig. 4. Variation of protein (a) and polysaccharide (b) in the sludge as a function of time with different sludge discharge frequencies.

Fig. 5. SEM of membrane surface at day 100: sludge discharge frequency of 5 days before (a) and after (b) washing by feed water; sludge discharge frequency of 10 days before (c) and after (d) in situ chemical cleaning, and (e) statistical details of pores for membranes cleaned by physical or chemical method.

reduction in the water temperature.

As seen from Fig. 4, the concentrations of protein and polysaccharide were a little higher with the longer sludge discharge interval (10 days). A potential reason was that the microorganism community was broken up by frequent sludge discharge (Massé et al., 2006; Wang et al., 2014). In comparison with protein, polysaccharide was more easily utilized by microorganisms due to the molecular structure (Wardle, 1992). As a result, the average concentration of protein was higher than the concentration of polysaccharide, especially at day 60. The concentrations of protein were 51.23 ± 1.51 and 63.19 ± 2.65 $\mu\text{g/g}$ with 5 and 10 days sludge discharge interval at day 60, while the concentrations of polysaccharide were 8.86 ± 0.41 and 8.98 ± 0.56 $\mu\text{g/g}$, respectively.

Additionally, although the concentration of EPS was a little higher with the sludge discharge interval of 10 days, the membrane fouling was less serious due to the strong adsorption/rejection of flocs, reducing the possibilities getting to the membrane surface (Figs. 3 and Fig. 4). Therefore, the TMP development was relatively slow with the sludge discharge frequency of 10 days. However, the membrane fouling will be more serious if the sludge discharge interval is continuously prolonged.

3.5. Variation of membrane surface

SEM images were also obtained to investigate the characteristics of the cake layer (Fig. 5), which showed that the presence of EPS-like substances in the cake layer with sludge discharge frequency of 10 days was more visible compared to the cake layer with sludge discharge frequency of 5 days (Fig. 5a and c). Thus, the SEM images provided information that was consistent with the physical results discussed previously, which showed that the concentration of EPS in the sludge with sludge discharge frequency of 10 days was higher than that with sludge discharge frequency of 5 days.

After the cake layer was washed with feed water (sludge discharge frequency of 5 days) or immersed under acidic conditions for 1 h (pH 2, sludge discharge frequency of 10 days), membrane pores were clearly observed. Because the cake layer was formed by loose Fe hydrolyzed flocs, many primary pores were visible on the surface of both membranes (Fig. 5b and d). On one hand, the floc cake layer was thicker with sludge discharge frequency of 10 days, resulting in more EPS being trapped with less possibility of getting to the membrane surface. On the other hand, even the blocked membrane pores could be cleaned under acidic conditions. Therefore, more membrane pores were observed after in situ chemical cleaning. This was consistent with the statistical number of open pores of the two fouled membranes as quantified by imageJ software. As seen from Fig. 5e, a higher percentage with a wide size range was present with sludge discharge frequency of 10 days. The average membrane pore size after washing with feed water was 10.16 ± 0.41 nm, while that increased to 14.42 ± 1.19 nm after in situ chemical cleaning. As a result, the TMP decreased more dramatically with sludge discharge frequency of 10 days after chemical cleaning compared to that of 5 days after physical cleaning (Fig. 3, day 100 and 101).

3.6. Variation of MW distribution

Due to the existence of TOC in the influent (Table 1), the corresponding removal efficiency and the variation of MW distribution were analyzed with different sludge discharge frequencies at day 100 (Fig. 6). The results showed that both larger (>10 kDa) and smaller (<10 kDa) MW organic molecules were removed, and the removal efficiency was a little higher with sludge discharge frequency of 10 days. For sludge discharge frequency of 5 days, the specific removal efficiency of larger MW organic molecules was

Fig. 6. Variation of MW distribution as a function of time with different sludge discharge frequencies at day 100.

32.8%, while the removal efficiency of smaller MW organic molecules was 20.7%. For sludge discharge frequency of 10 days, the specific removal efficiency of larger MW organic molecules was 41.1%, while the removal efficiency of smaller MW organic molecules was 26.2%. The results were in accordance with the thickness of the cake layer, and more pollutants were rejected by the thicker cake layer (sludge discharge frequency of 10 days).

As seen from Fig. 6, with the removal efficiency of different MW pollutants, the peak value of the MW distribution also varied, especially for the sludge discharge frequency of 10 days. The peak value of the larger MW distribution was 10551.3 Da in the influent, while it decreased to 10317.6 Da with the sludge discharge frequency of 10 days. However, owing to the lower removal efficiency of smaller MW pollutants, the peak value (7819.4 Da) was almost the same before and after filtration. Additionally, no variation was observed in the removal efficiency of TOC before and after sludge discharge (data not shown).

4. Conclusions

The integrated UF membrane process performed better and less severe membrane fouling occurred, compared to the granular adsorbents used. Based on their strong adsorption ability and loose structure, the in situ Fe-based flocs were directly injected into the membrane tank, and many problems encountered with other adsorbents were solved, such as higher operation cost, greater chance of causing membrane surface damage, lower in situ chemical cleaning efficiency, etc.

For the flocs-integrated membrane process, we found that the removal efficiency toward Sb(V) was high and stable. The effluent quality was excellent, even under weakly acidic conditions. Although microorganisms gradually proliferated and the main membrane fouling mechanism was ascribed to cake layer formation, most EPS were wrapped by flocs, and the cake layer was relatively loose. As a result, the cake layer was easily washed off by water or dissolved under strongly acidic conditions with in situ chemical cleaning.

Sludge discharge also played an important role in water treatment. However, it had little influence on effluent quality, including the turbidity, chromaticity, iron concentration, etc. This indicates that longer sludge discharge frequency is feasible during actual operations, which also leads to a higher water production rate. Based on the excellent performance, the in situ flocs-integrated UF membrane process indeed shows promising potential for

application during water treatment.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.watres.2017.05.025>.

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