Comparison of NF membrane fouling and cleaning by two pretreatment strategies for the advanced treatment of antibiotic production wastewater

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ABSTRACT

The nanofiltration (NF) membrane fouling characteristics and cleaning strategies were investigated and compared for treating membrane bioreactor (MBR) effluent and MBR-granular activated carbon (GAC) effluent of an antibiotic production wastewater by DK membrane. Results showed that the fouling of treating MBR effluent was more severe than that of treating MBR-GAC effluent. After filtering for 216 h, the difference of membrane flux decline was obvious between MBR effluent and MBR-GAC effluent, with 14.9% and 10.3% flux decline, respectively. Further study showed that organic fouling is the main NF membrane fouling in the advanced treatment of antibiotic production wastewater for both of the two different effluents. Soluble microbial by-product like and tyrosine-like substances were the dominant components in the foulants, whereas humic-like substances existing in the effluents had little contribution to the NF membrane fouling. A satisfactory efficiency of NF chemical cleaning could be obtained using combination of acid (HCl, pH 2.0–2.5) and alkali (NaOH + 0.3 wt% NaDS, pH 10.0–10.5). The favorable cleaning strategy is acid-alkali for treating the MBR-GAC effluent, while it is alkali-acid for treating the MBR effluent.

Key words | antibiotic production wastewater, cleaning, membrane fouling, nanofiltration, pretreatment

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INTRODUCTION

Nanofiltration (NF) is a fast growing technology in the advanced treatment of municipal and industrial wastewater for water reuse (Aouni *et al.* 2009; Chon *et al.* 2011; Ellouze *et al.* 2012). Among the various kind of wastewaters, pharmaceutical wastewater discharged globally is not only in large amounts, but also difficult to be treated (Chelilapan *et al.* 2006). In our previous study, NF process was used for the advanced treatment of antibiotic production wastewater. The results showed that the NF permeate could meet China's water quality standard requirements for industrial use (GB21903-2008) (Wang *et al.* 2014b; Wang *et al.* 2015). However, fouling layer formation on the membrane surfaces is still a major obstacle for practical application and spreading of NF process in the advanced treatment of both the antibiotic production wastewater and other types of wastewaters (Wei *et al.* 2010; Chon *et al.* 2013).

According to the literature, water constituents such as particles, colloids, salts, natural organic matters, and soluble microbial products derived from biological wastewater treatment can adsorb and deposit onto membrane surface, which result in membrane fouling (Xu *et al.* 2006). Therefore, the characteristics of the NF membrane fouling are affected by feed water composition. Xu *et al.* (2006) reported that the major components of foulants were polysaccharides, organic sulfonic acids, silicate colloids, Si, Cu, Fe, Zn and Ca when treating the secondary effluent of a local municipal wastewater using NF process in Denver, CO. However, Azais *et al.* (2014) demonstrated that polysaccharides and humic acid were the major compounds adsorbed on the NF membrane of the municipal wastewater treatment plant (WWTP) effluent collected in the south of France. Wei *et al.* (2010) showed that calcium salts and complex organic foulants containing carboxyl acid, amide, and alkyl halide functional groups were deposited

onto the membrane surface in the advanced treatment of pharmaceutical wastewater by NF. Although there were many studies on membrane fouling of NF, the results were quite different due to different feed water qualities. Moreover, many studies focus on simulated wastewater, therefore the studies on membrane foulant identification and cleaning strategy are rather scarce in real wastewater, especially in industrial wastewater.

In addition, many studies showed that employing adequate pretreatment ahead of the membrane process could alleviate membrane fouling (Kumar *et al.* 2006; Al-Amoudi & Lovitt 2007). The pretreatment methods include coagulation (Al-Amoudi & Lovitt 2007), biofiltration (Mosqueda-Jimenez & Huck 2009), H₂O₂/UV (Song *et al.* 2004), ultrafiltration (UF) (Luo *et al.* 2011), activated carbon filtration (Benito-Alcazar *et al.* 2010), and O₃ oxidation (Flyborg *et al.* 2010). Among these pretreated methods, granular activated carbon (GAC) sorption process could achieve an important reduction of the organics in the secondary effluent, which improved the feed water quality for NF process effectively. We have demonstrated that GAC sorption process was a very effective way to pretreat secondary effluent of antibiotic production wastewater in our previous study (Wang *et al.* 2014a). However, the effect of GAC sorption process on NF membrane fouling should be further studied.

In this study, the fouling characteristics and cleaning strategies of NF membranes by two pretreatment strategies,

membrane bioreactor (MBR) and MBR-GAC, were investigated and compared with the advanced treatment of antibiotic production wastewater. The foulants on the membrane surface were desorbed by different cleaners. Then the cleaning solutions were identified through regular chemical analysis and the three dimensional fluorescence excitation-emission matrix (3D-EEM) spectra. Moreover, scanning electron microscope (SEM), atomic force microscopy (AFM), and attenuated total reflectance – Fourier transform infrared spectroscopy (ATR-FTIR) were employed to analyze the different membrane surfaces and cleaning effects.

MATERIALS AND METHODS

Wastewater characteristics

The raw wastewater was the anaerobic effluent from a wastewater treatment station of a pharmaceutical company producing spiramycin in Wuxi (China). Before NF treatment, the wastewater was pretreated by a MBR and a MBR-GAC, respectively. The GAC used in this study was made from apricot shells with particle size at 10–24 mesh. The characteristics of the MBR-GAC and MBR effluent are shown in Table 1.

Table 1 | Water characteristics of NF feed and permeate in treating the MBR-GAC effluent and MBR effluent

Item	NF feed		NF permeate	
	MBR-GAC	MBR	MBR-GAC	MBR
TOC (mg/L)	29.51 ± 1.76	104.12 ± 3.04	0.00 ± 0.36	0.79 ± 0.33
TDS (mg/L)	3,650 ± 170	3,715 ± 100	2,500 ± 105	2,660 ± 60
UV ₂₅₄ (cm ⁻¹)	0.56 ± 0.03	2.19 ± 0.07	0.03 ± 0.00	0.07 ± 0.00
Turbidity (NTU)	0.09 ± 0.08	0.14 ± 0.13	0.04 ± 0.01	0.04 ± 0.01
Colour (PCU)	122.27 ± 9.73	828.24 ± 33.95	0.00 ± 0.00	1.67 ± 3.83
K ⁺ (mg/L)	161.35 ± 15.97	154.56 ± 9.12	120.92 ± 16.92	122.97 ± 3.29
Ca ²⁺ (mg/L)	276.85 ± 16.09	256.83 ± 7.73	31.92 ± 1.26	33.07 ± 2.18
Na ⁺ (mg/L)	1239.37 ± 51.72	1287.16 ± 79.32	1031.50 ± 57.55	1058.42 ± 28.21
Mg ²⁺ (mg/L)	33.63 ± 2.32	30.96 ± 1.14	1.18 ± 0.12	1.07 ± 0.13
Cl ⁻ (mg/L)	2523.69 ± 141.53	2382.63 ± 154.55	1575.89 ± 159.44	1449.13 ± 118.49
NO ₂ ⁻ (mg/L)	7.32 ± 1.56	5.97 ± 1.87	5.16 ± 0.54	4.80 ± 1.55
NO ₃ ⁻ (mg/L)	1275.63 ± 65.85	1469.09 ± 94.86	909.25 ± 90.41	1062.22 ± 111.68
PO ₄ ³⁻ (mg/L)	13.08 ± 0.76	25.86 ± 1.04	0.50 ± 0.07	0.74 ± 0.09
SO ₄ ²⁻ (mg/L)	157.74 ± 27.80	74.07 ± 5.63	1.61 ± 0.49	0.32 ± 0.29
CO ₃ ²⁻ (mg/L)	2.11	6.15	2.76	5.18

Experimental setup

NF tests were carried out by using a bench-plant (Shanghai Shiyuan Bioengineering Equipment Co., China) equipped with three pressure vessels for 1.8×12 inches spiral-wound membrane modules. DK membrane (DK 1812C-34D), which was provided by GE Osmonics, was employed according to our previous study (Wang *et al.* 2014a). Both permeate and retentate were circulated back to the feed reservoir. NF tests were conducted at pressure of 1.0 MPa and crossflow velocity of 8.0 L/min at 25 °C. Each test was carried out for 216 h.

To analyze the membrane fouling, NF fouling tests were performed by using a cross-flow flat-sheet membrane test unit (SEPA CF II, GE Osmonics). A flat sheet NF membrane of DK was used. The surface area of the NF membrane was 139 cm². Each fouling test was carried out for 45 h in treating the MBR effluent and MBR-GAC effluent as mentioned above. The permeate and retentate were also circulated back to the feed reservoir. The operating parameters were the same as for the NF tests (i.e. TMP 1.0 MPa, 25 °C). After NF fouling test, the fouled flat membrane was divided into six parts. Five of these parts were soaked with deionized (DI) water, acid (HCl, pH 2.0–2.5), alkali (NaOH + 0.3 wt% NaDS, pH 10.0–10.5), acid-alkali (soaking with acid firstly and then with alkali), alkali-acid (soaking with alkali firstly and then with acid) for 24 h, respectively. The NF membranes and cleaning solutions were further analyzed to investigate the fouling characteristic and cleaning strategy.

Analysis

Total organic carbon (TOC) was determined by a TOC-VCPH (Shimadzu, Japan). UV₂₅₄ was measured by an ultra-violet and visible spectrophotometer (Spectrum Laboratory 752sp, Lengquang Tech, China). A color meter (HI 96727, Hanna, Italy) and a turbidity meter (Turb 550, WTW, Germany) were used to measure color and turbidity, respectively. Total dissolved solids (TDS) was measured with a conductivity/TDS meter (HI4321, Hanna, Italy). The contents of Ca²⁺, Na⁺, K⁺, and Mg²⁺ were analyzed by an inductively coupled plasma optical emission spectroscopy (Optima 2100 DV, Perkin Elmer, USA). The anions (Cl⁻, NO₃⁻, NO₂⁻, SO₄²⁻, and PO₄³⁻) were determined using an ion chromatography (ICS-1000, Dionex, USA).

A fluorescence spectrophotometer (F-7000, Hitachi, Japan) was used to investigate major components and the complexity of organic matter that were contained in the NF feed streams and cleaning solutions.

SEM (S-3000N, Hitachi, Japan) analysis was carried out to observe the surfaces of virgin and fouled NF membranes. All samples were freeze-dried for 24 h.

NF membrane surface roughness was determined using an AFM (NanoScope IIIa, Digital Instruments, USA). Imaging was performed in tapping mode on an area of 2 µm × 2 µm. Each sample was measured three times.

ATR-FTIR was performed using a Fourier transform infrared spectrometer (Nicolet 8700, Thermo Electron Corporation, USA) with a resolution of 4 cm⁻¹ in the range 400–4,000 cm⁻¹ to obtain information about the functional groups of the membrane surfaces.

RESULTS AND DISCUSSION

Performance of NF membrane

Water characteristics of NF feed and permeate in treating MBR-GAC effluent and MBR effluent are shown in Table 1. As shown in Table 1, excellent water quality could be obtained using NF process for the treatment of both MBR-GAC effluent and MBR effluent. The TOC of NF permeates were lower than 1.0 mg/L, which illustrated that there was no significant effect on NF permeates of different pretreatment processes. However, the difference of membrane flux decline was obvious between MBR-GAC effluent and MBR effluent, with 10.3% and 14.9% membrane flux decline, respectively, at the end (Figure 1). For the initial permeate flux for treating MBR and MBR-GAC effluents was nearly at the same level, i.e. 69.38 L/h·m² and 72.19 L/h·m². We concluded that the MBR effluent suffered more serious fouling than the MBR-GAC effluent when treated by the NF process. As the main difference of the two feed streams was TOC concentration shown in Table 1 (i.e.

Figure 1 | Changes of NF membrane flux decline for treating the MBR-GAC effluent and MBR effluent.

29.51 ± 1.76 mg/L and 104.12 ± 3.04 mg/L, respectively), we can infer that organics is the primary cause of this phenomenon. In addition, Figure 1 shows that the rapid decline of flux occurred at the initial stage of NF treatment, which was unlike Ang's study (Ang *et al.* 2011). This may be also caused by the influence of the organics in the feed streams.

NF membrane morphological analysis

The samples of virgin and fouled NF membranes through NF fouling tests were tested with SEM and AFM, respectively (Figures 2 and 3). As shown in Figure 2, the surface of virgin NF membrane was rather clean and smooth. In contrast, it was obvious that the NF membrane surface was seriously fouled for treating these two effluents after fouling tests. A cake layer was formed by the foulants attached to the NF membrane surface.

The mean roughnesses of the virgin and fouled NF membrane changed apparently in Figure 3, i.e., the mean roughnesses of the virgin NF membrane, fouled NF membrane treating the MBR-GAC effluent, and fouled NF membrane treating the MBR effluent were 11.186 nm, 23.710 nm, and 34.638 nm, respectively. As the surface of DK membrane was rather smooth, the foulants built up on the membrane surfaces and resulted in an increase in surface roughness. This result was consistent with Xu's study (Xu *et al.* 2006). The changes of roughness also showed that it

suffered worse fouling from treating the MBR effluent compared with the MBR-GAC effluent.

Identification of membrane foulant and cleaning strategies

Characterization of the foulant desorbed by different cleaning strategies

In order to identify the foulant composition of the fouling layer, the cleaning solutions by different cleaners was analyzed through regular chemical analysis and 3D-EEM. The extent of organics desorbed from the fouled membrane was determined by TOC measurements using the different cleaning strategies described above (Figure 4). Higher concentrations of TOC were detected desorbing by alkali cleaning, acid-alkali cleaning and alkali-acid cleaning. However, none of the inorganic ions (including Ca^{2+} , Mg^{2+} , SO_4^{2-}) were found in each cleaning solution. This demonstrated that organics were the dominant factor of membrane fouling in the advanced treatment of antibiotic production wastewater by NF process. Furthermore, on the basis of TOC concentrations in the desorption solutions, the favorable cleaning strategy is acid-alkali when treating the MBR-GAC effluent, while it is alkali-acid when treating the MBR effluent. This might be due to the different composition of the two effluents, which resulted in different composition of membrane foulants. Generally, acid cleaning

Figure 2 | Comparison of SEM photos of different NF membranes: (a) virgin membrane, (b) fouled membrane treating MBR-GAC effluent, and (c) fouled membrane treating MBR effluent.

Figure 3 | Comparison of AFM photos of different NF membranes: (a) virgin membrane, (b) fouled membrane treating MBR-GAC effluent, and (c) fouled membrane treating MBR effluent.

Figure 4 | TOC concentrations using the cleaning solutions for treating MBR-GAC effluent and MBR effluent.

is suitable for the removal of precipitated salts, while alkali cleaning is used to remove adsorbed organics. Xiao *et al.* (2012) also demonstrated that HCl solution could remove the substance of aromatic protein, while NaOH solution could remove both aromatic protein and other organics, such as soluble microbial by-product (SMP)-like substance. The different effectivity between alkali-acid and acid-alkali might due to the

different composition of membrane foulants. As shown in Table 1, the inorganic ions concentrations of the two effluents were at the same level, whereas the organic concentrations were obviously different. This led to different inorganic/organic ratio of the two effluents. Kappel *et al.* (2014) showed that change of Ca^{2+} /TOC ratio could induce different membrane fouling characteristics. Therefore, the different

inorganic/organic ratio of the two effluents might lead to different membrane fouling characteristics, which influenced the cleaning strategies and the difference between alkali-acid and acid-alkali in treating the two effluents.

The 3D-EEMs is a collection of a series of emission spectra over a range of excitation wavelengths, which can be used to identify the fluorescent compounds present in complex mixtures (Wang *et al.* 2009). Chen *et al.* (2003) divided the EEM spectra into five regions, including humic acid-like, fulvic acid-like, tyrosine-like, tryptophan-like, and soluble microbial by-product-like organics, respectively (Table 2). In this study, 3D-EEMs were employed to identify the organics in feed streams and membrane foulants, and the results are illustrated in Figure 5 and Table 2.

As shown in Figure 5 and Table 2, strong humic-like fluorescence of Peak A was found both in the

MBR-GAC effluent and MBR effluent. The concentration of humic-like substances in the MBR effluent was much higher than in MBR-GAC effluent, with intensities of 15,496 nm and 3,242 nm, respectively. However, cleaning solutions showed two other maximum peaks (Peak B and Peak C) which corresponded to SMP-like and tyrosine-like substances for treating these two effluents. This demonstrated SMP-like and tyrosine-like substances were the dominant components in the foulants, whereas humic-like substances had little contribution to the NF membrane fouling, which was consistent with other literature (Zazouli *et al.* 2010). Moreover, the concentrations of SMP-like and tyrosine-like substances in cleaning solution treating the MBR effluent were much higher than that of treating the MBR-GAC effluent as shown in Table 2, which further demonstrated that fouling of MBR effluent was more severe.

Table 2 | EEM peaks and intensities in the different solutions

EEM region	Rang (Ex/Em)	MBR-GAC effluent		MBR effluent		Cleaning solution treating MBR-GAC effluent		Cleaning solution treating MBR effluent	
		Rang (Ex/Em)	Inten. max.	Rang (Ex/Em)	Inten. max.	Rang (Ex/Em)	Inten. max.	Rang (Ex/Em)	Inten. max.
Tryptophan-like	<250/280–330	–	–	–	–	–	–	–	–
Tyrosine-like	<250/330–380	–	–	–	–	230/330	2,682	225/335	4,102
SMP-like	>250/280–380	–	–	–	–	280/330	8,774	285/335	9,360
Humic acid-like	>250/ > 380	325/400	3,242	355/405	15,496	–	–	–	–
Fulvic acid-like	<250/ > 380	–	–	–	–	–	–	–	–

Figure 5 | 3D-EEMs in the different solutions: (a) MBR-GAC effluent, (b) cleaning solution treating MBR-GAC effluent, (c) MBR effluent, and (d) cleaning solution treating MBR effluent.

Figure 6 | ATR-FTIR spectra of different NF membranes for treating MBR-GAC effluent and MBR effluent.

ATR-FTIR analysis of different NF membranes

The virgin, fouled and cleaned NF membranes had been investigated by ATR-FTIR spectral analysis as presented in Figure 6. As shown in Figure 6, the spectra of the virgin DK membrane were similar to the specific ATR-FTIR absorbance peaks typical for polyamide membranes (Mo et al. 2010). After fouled for 45 h, the peaks of virgin membranes decreased in both the absorbance intensity and peak quantity, suggesting that the membranes were severely fouled. It is shown that there was poor cleaning effect by DI water or acid separately. However, cleaning by alkali could obtain a perfect result, which also demonstrated that the main foulants of the fouling layer were organics. After cleaning by acid and alkali, there were significant similarities in the spectra obtained for the cleaned membrane with the virgin membrane, which proved that it could get satisfactory cleaning performance through combining acid and alkali cleaning.

CONCLUSIONS

Organic fouling is the main membrane fouling in the advanced treatment of antibiotic production wastewater by NF process. The membrane fouling of treating the MBR effluent was worse than that of treating the MBR-GAC effluent. SMP-like and tyrosine-like substances were the dominant components in the foulants treating these two effluents, whereas humic-like substances existing in the effluents had little contribution to the NF membrane fouling. The favorable cleaning strategies is acid (HCl, pH 2.0–2.5)-alkali (NaOH + 0.3 wt% NaDS, pH 10.0–10.5) for treating the MBR-GAC effluent, while it is alkali-acid for treating the MBR effluent.

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