

Performance of a GAC and ultrafiltration hybrid process under high humidity and temperature conditions

Tiejun Qiao, Doris W. T. Au, Xihui Zhang and Jinsong Zhang

ABSTRACT

The performance of a granular activated carbon (GAC) and ultrafiltration (UF) hybrid process and changes in UF membrane properties were studied. This hybrid process reduced total organic carbon by $59 \pm 12\%$ and UV absorbance (UV_{254}) by $88 \pm 31\%$, with GAC accounting for 93 and 98% of total reduction, respectively. Selected pharmaceuticals and personal care products were reduced by 32–92% during the hybrid process. Particles, bacteria and invertebrates were reduced by 98–99, 76–92 and 100%, respectively during the hybrid process. No invertebrates, including rotifers, cyclops and cladocera, were detected in the effluent of the hybrid process. Trans-membrane pressure (TMP) and membrane roughness increased while specific flux (SFLUX) decreased with prolonged operation time. Contact angle increased from $59 \pm 1.2^\circ$ to $70 \pm 2.0^\circ$ after one operation cycle, and then decreased to $66 \pm 1.2^\circ$ after ten cycles. Chemical enhanced backwashing was more effective to reduce TMP, increase SFLUX and control roughness and contact angle than backwashing by water. Membrane fouling not only changed roughness and contact angle, but also affected the removal of organics.

Key words | granular activated carbon, membrane properties, microbial safety, pharmaceuticals and personal care products, ultrafiltration

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INTRODUCTION

Organic matter, both natural and synthetic, has received much attention due to its detrimental effects on human health and water treatment processes (Zularisam *et al.* 2006). Some emerging organics, e.g. pharmaceuticals and personal care products (PPCPs), have also become a key topic worldwide in recent years due to their potential adverse environmental and health impacts (Daughton & Ternes 1999). Conventional treatment processes including coagulation, sedimentation, filtration and disinfection, were generally inefficient in removing these organic substances (Vieno *et al.* 2007; Zularisam *et al.* 2009). However, granular activated carbon (GAC) has been used extensively to remove them since the early 1970s (Yu *et al.* 2008).

Some potential microbial risks have attracted a great deal of attention during GAC application due to the overgrowth and leakage of various microorganisms, e.g.

viruses, bacteria and invertebrates (Qiao & Zhang 2009). Ultrafiltration (UF) is an effective technology to control microbial risk. However, membrane fouling is an important concern during UF application. Some hybrid processes were developed to resolve this problem, involving coagulation pretreatment and activated carbon (Fiksdal & Leiknes 2006; Gur-Reznik *et al.* 2008). The hybrid process using GAC and UF has recently been studied for its efficiency in removing organics and microorganisms (Gur-Reznik *et al.* 2008). Most of this research was conducted in bench-scale or pilot-scale experiments under a specific climate (Gur-Reznik *et al.* 2008). Few studies focused on process performance under high humidity and temperature.

Membrane properties have an important effect on UF performance. Schafer *et al.* (2000) suggested that membrane pore size had a close relationship with UF performance; large membrane pore size resulted in poor performance.

Hu *et al.* (2007) reported that membrane hydrophilicity had an important influence on performance, but could cause high membrane flux. Yoon *et al.* (2007) suggested that UF removed PPCPs mainly due to hydrophobic adsorption. However, UF membrane properties changed during operation. Therefore, other mechanisms may play important roles in removing PPCPs by UF. Also, most of these experiments were conducted in bench-scale experiments or in short-term operation; UF may behave differently due to dynamic raw water quality or in long-term operation.

This paper aims to study the performance of a GAC and UF hybrid process in southern China, where the climate is subtropical, with average humidity and air temperature of 70–80% and 22–24 °C respectively. Changes of membrane properties and their effects on UF performance were explored.

METHODS

Pilot-scale test

A pilot-scale GAC and UF hybrid process, with the capacity of 10 m³ h⁻¹, was investigated. Raw water was from a reservoir, and was pretreated by coagulation and sedimentation. Coagulation conditions were: pH 6.9–7.1, mixing intensity = 500–600 s⁻¹ and mixing time = 1 min. Polyaluminium chloride was used as the coagulant, with dosage of 1.8–2.0 mgAl₂O₃ L⁻¹. The GAC used was coal-based activated carbon (8 × 30 mesh, Xinhua Chemicals Co. Ltd., China). The GAC layer was 1.0 m deep. The filtration velocity was designed to be 8 m h⁻¹. The GAC process was backwashed every 24 h with bed expansion of 15–25%. The UF membrane was made up of PVC alloy. Membrane pore size was 0.01 μm. The operating mode for the membrane module was inside-out. The UF filtration flux was 80 L m² h⁻¹. Chemical enhanced backwashing (CEB) was conducted every 5–7 d during UF operation.

Bench-scale experiments

Membrane properties, in terms of contact angle and roughness, were investigated in a bench-scale syringe apparatus (Rwd252, Rwd Life Science Co. Ltd, China). The GAC

effluent from the pilot process was injected into one membrane fiber to simulate UF. The fiber was backwashed by the apparatus in reverse injection. The membrane flux was set at 80 and 170 L m⁻² h⁻¹ for filtration and backwashing, respectively. Duration of filtration and backwashing were 30 and 1 min, respectively. After several filtration cycles, the test fiber was cut into 5 mm segments. The segments were rinsed in Milli-Q water, and fixed on glass slides, and then dried at 45 °C. Contact angle was analyzed by an optical analyzer (DSA-100, Kruss, Germany). Roughness was measured by atomic force microscopy (Digital Instruments, Veeco Co., USA).

Analytical methods

The ultraviolet absorbance at 254 nm (UV₂₅₄) was measured by a UV spectrophotometer (UV-1700, Shimadzu, Japan). Particle counts were measured by a portable particle count analyzer (WPCS, IBR Inc., USA), with the measurement range of 2–100 μm in diameter. Invertebrates were collected by filtering 500 L water sample at 5 L min⁻¹ with filter screen (pore size of 35 μm). After filtration, the samples were fixed with 4% formaldehyde solution. Invertebrates were classified and identified by optical microscope (BX51, Olympus, Japan). Other water quality parameters were analyzed by standard methods (APHA 1995).

Eight PPCPs were selected on the basis of their occurrence and concentration in the local water sources (Qiao *et al.* 2011). Their properties are shown in Table 1. The

Table 1 | Properties of selected PPCPs analyzed in this study

No	Compound	CAS registry no.	LogKow ^a	LogKoc ^a	pKa
1	Antipyrine	60-80-0	0.38	1.07	1.4
2	Disopyramide	3737-09-5	2.58	2.46	10.4
3	Indomethacin	53-86-1	4.27	2.34	4.5
4	Lincomycin	154-21-2	0.20	0.03	4.6
5	Sulfamethoxazole	723-46-6	0.89	1.54	5.94
6	Sulpiride	15676-16-1	0.57	1.41	9
7	Tiamulin	55297-95-5	4.75	3.21	7.6
8	Trimethoprim	738-70-5	0.91	1.90	7.3

^aThe values were calculated by the EPI suite™.

Kow: octanol-water partition coefficient; Koc: organic carbon partition coefficient; pKa: acidity coefficient.

PPCPs were analyzed by solid phase extraction high performance liquid chromatography (HPLC) (SPD-20A, Shimadzu, Japan) and tandem mass spectrometry (MS/MS) (API 3200, Applied Biosystems, USA) (Qiao *et al.* 2011). The procedure was as follows: water samples were pre-treated with 0.5 g L^{-1} of EDTA and 1 g L^{-1} of ascorbic acid, and then pre-filtered through a glass fiber filter ($1 \mu\text{m}$ pore size, Whatman GF/B, USA). The pre-filtered samples were then filtered at a flow rate of 10 mL min^{-1} (SPC10-C, Sep-Pak concentrator, Japan) through Oasis[®] HLB columns (6 cc 500 mg, Waters, USA). The columns were pre-conditioned with 6 mL of methanol and 6 mL of milli-Q water. The columns were then aspirated and dehydrated, and target PPCPs were eluted with 6 mL of methanol. The eluate was concentrated and evaporated with gentle N_2 , and the residue was redissolved into 1 mL of methanol for HPLC/MS/MS analysis.

RESULTS AND DISCUSSION

Performance in removing organic matters

Total organic carbon and UV_{254}

The GAC and UF hybrid process reduced total organic carbon (TOC) by $59 \pm 12\%$, on average, during operation (Figure 1(a)). After GAC filtration, TOC was reduced by $55 \pm 15\%$ and after UF, by $4 \pm 6\%$, accounting for 93 and 7% of total removal, respectively. Reduction of UV absorbance by the hybrid process is presented in Figure 1(b). The hybrid process reduced UV_{254} effectively, with the

average reduction of $88 \pm 31\%$; UV_{254} was reduced by $86 \pm 21\%$ after GAC, accounting for 98% of total reduction. On average, UF reduced UV_{254} by $4 \pm 21\%$, accounting for 2% of total reduction. Thus, TOC and UV_{254} were mainly removed by GAC rather than UF. Much previous research has reported the effectiveness of GAC in removing organic matter (Gur-Reznik *et al.* 2008). But UF can remove organic substances with large molecular weight, while some small hydrophilic organic molecules were removed poorly (Park & Lee 2005; Gur-Reznik *et al.* 2008). The portion of hydrophilic organic matter can be characterized by the ratio between UV_{254} and TOC. High ratio values mean a low proportion of hydrophilic substances. The ratio decreased greatly after GAC (Figure 1). This indicated that the hydrophobic organic substances were decreased after GAC. Moreover, the increased hydrophilicity could be beneficial in alleviating membrane fouling (Katsoufidou *et al.* 2005).

Pharmaceuticals and personal care products

Figure 2 presents reduction of the selected PPCPs by the GAC and UF hybrid process. The average reduction of antipyrine, disopyramide, sulphiride, tiamulin and trimethoprim was 78 ± 7 , 73 ± 26 , 74 ± 24 , 81 ± 21 and $92 \pm 6\%$, respectively. The reduction efficiencies for lincomycin and indomethacin were lower, with averages of 41 ± 15 and $32 \pm 15\%$, respectively. Sulfamethoxazole was reduced by an average of $59 \pm 19\%$. GAC played a major role in removing antipyrine, disopyramide, sulfamethoxazole, sulphiride and trimethoprim, accounting for 96, 96, 92, 95 and 98% of total reduction, respectively. GAC removed most of the indomethacin, lincomycin and tiamulin present, accounting

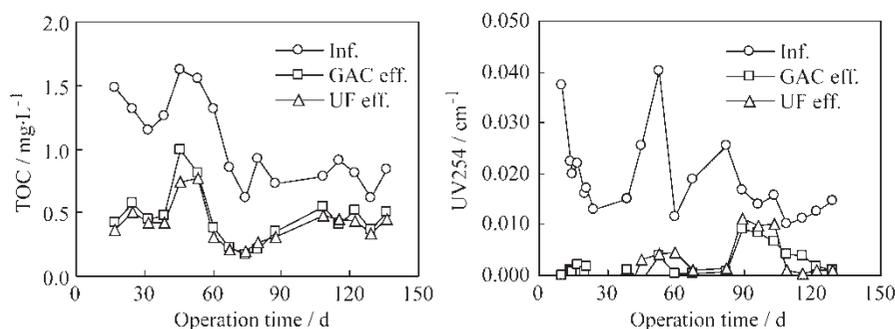


Figure 1 | TOC and UV_{254} reduction by GAC and UF hybrid process during operation. Inf: influent; GAC eff.: GAC effluent; UF eff: UF effluent.

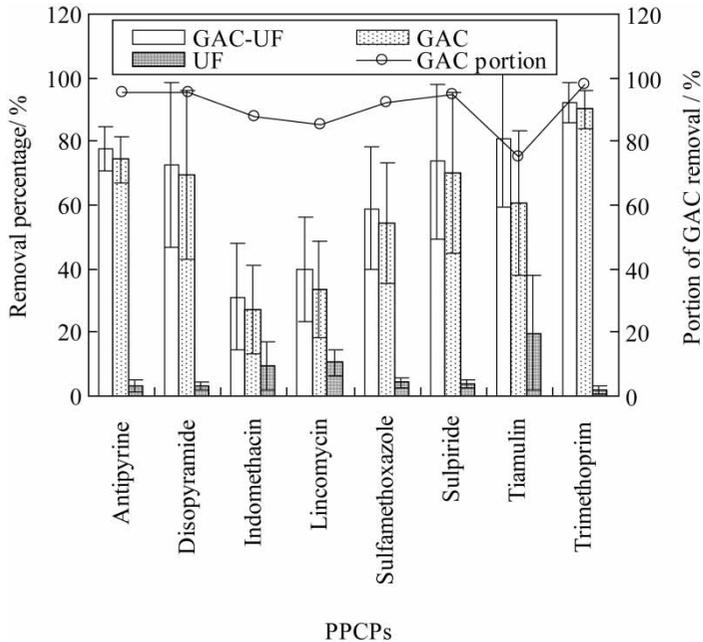


Figure 2 | Reduction of selected PPCPs by the GAC and UF hybrid process during operation.

for 88, 85 and 75% of total reduction, respectively. The results above demonstrated that the hybrid process can reduce most of the selected PPCPs effectively. Although, UF can reduce selected PPCPs partially, GAC was very effective in removing most of PPCPs (Kim *et al.* 2007; Vieno *et al.* 2007), unlike UF (Yoon *et al.* 2007). However, some hydrophilic PPCPs, such as lincomycin and indomethacin, were not removed effectively, which is consistent with some other reports (Vieno *et al.* 2007), whereas other hydrophilic PPCPs, such as antipyrine, sulfamethoxazole and sulpiride, were removed efficiently. This indicates that hydrophilicity is not the only important factor to affect the removal performance of PPCPs.

Performance in removing microorganisms

Particles

As shown in Figure 3(a), particles were firstly reduced by GAC, and then further removed by UF. Particle counts in the GAC influent were in the range of 260–7,790 counts (CNT)·mL⁻¹. The average particle counts was $2,472 \pm 2,049$ CNT mL⁻¹. The proportion of particles of size range 2–3, 3–5, 5–10 and 10–20 μm were 48, 35, 16 and 1%, respectively. Particle counts in the GAC effluent were in the range 20–600 CNT mL⁻¹. The average particle counts were 41 ± 54 CNT mL⁻¹. The proportion of particles of

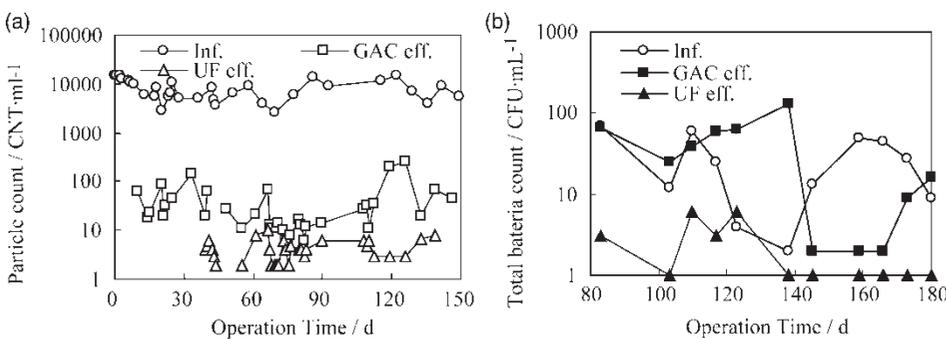


Figure 3 | Particle and bacteria reduction by GAC and UF hybrid process during operation.

size range of 2–3, 3–5, 5–10 and 10–20 μm were 42, 46, 11 and 1%, respectively. Particle counts in the UF effluent were approximately 10 CNT mL^{-1} , and 4.4 ± 2.0 on average. The particle proportion of 2–3, 3–5 and 5–10 μm were 47, 39 and 14%, respectively. Particle counts have a good correlation with microorganisms and are used extensively in water treatment practice (Qiao & Zhang 2009). Particle sizes detected are similar to those of *Cryptosporidium* and *Giardia* (Hatukai et al. 1997). To ensure microbial safety, particle counts should be controlled to below 50 CNT mL^{-1} or even lower. The result above indicated that microbial safety could be enhanced greatly by the hybrid process. Some large particles can be detected in the UF effluent, which indicated that membrane pore sizes were perhaps not uniform, or the shapes of particles were perhaps not spherical. However, some viruses such as noroviruses, hepatitis A viruses and polio viruses cannot detect by the particle count analyzer due to their small size (approximately 20–40 nm in diameters). Therefore, further study could be conducted with a more sensitive analyzer.

Bacteria

As can be seen from Figure 3(b), total bacteria counts (TBC) in GAC influent were on average, 29 ± 23 colony-forming units (CFU) $\cdot\text{mL}^{-1}$ (range 2–70 CFU mL^{-1}). The GAC effluent's TBC fluctuated greatly in the range

2–130 CFU mL^{-1} , with an average of 37 ± 39 CFU mL^{-1} . Almost no bacteria were detected in the UF effluent. These results showed that the hybrid process reduced TBC very effectively, especially UF. The change of TBC in the influent and effluent of GAC was not consistent (Figure 3). Sometimes TBC in the influent was higher than that in the effluent, showing that bacteria were removed by GAC. However, at other times, TBC in the GAC effluent was higher than that in the influent, indicating that the overgrowth of bacteria had occurred in the GAC. TBC are an essential parameter to evaluate microbial safety. A high TBC may mean the microbial risk is too high. The experimental results suggested that GAC could increase or decrease microbial risk. However, this risk could be reduced greatly by UF.

Invertebrates

Figure 4 shows that invertebrates in the GAC influent were abundant during days 65–80, with counts of 150–3,120 organisms per 100 L. The predominant species were rotifers in the GAC influent, accounting for 99%. In the GAC effluent, invertebrate counts were 0–40 organisms per 100 L. The predominant species were rotifers and cladocera, accounting for 50–100% of the total count. Other invertebrates included cyclops and nauplius.

From day 81 to 210, invertebrate counts were 30–720 organisms per 100 L in the GAC influent. Rotifers, as

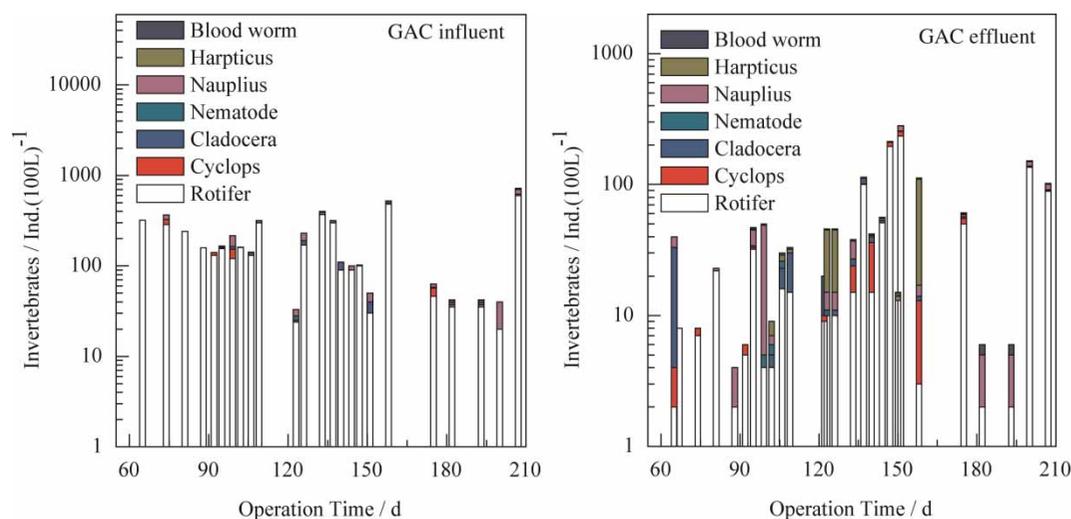


Figure 4 | Invertebrates in the influent and effluent of GAC process during operation. Note the different scale for invertebrate counts in the two diagrams.

predominant species, accounted for 50–100% of total invertebrates. Other invertebrates included cladocera, nauplius, cyclops, nematodes, harpacticus and blood worms. Counts were 4–50 organisms per 100 L in the GAC effluent. The predominant species were rotifer and nauplius, accounting for 22–91 and 8–87%, respectively. Others included cladocera, harpacticus, nematodes, cyclops and blood worms.

Invertebrates have also received much attention due to not only aesthetic problems, but also because of concerns about pathogen survival (Scholz & Martin 1997; Castaldelli *et al.* 2005). The results demonstrated that GAC provided circumstances for their growth and removed them to some extent. The average removal of total invertebrates was approximately 85% by GAC. This is different from some previous studies (Schreiber *et al.* 1997; Li *et al.* 2010). The difference could be due to backwashing frequency. Backwashing of GAC was performed every 24 h in this study, and the growth of invertebrates could be controlled well. In addition, GAC media layer was 0.8 m, much less than previous studies (Li *et al.* 2010). However, UF can remove invertebrates very effectively. No invertebrates were detected in UF effluent. The results showed that invertebrates were reduced effectively by the hybrid process, especially by UF. Predominant species were not consistent with other studies. Some reported that rotifers were predominant, then nematodes, copepods and cladocera, etc (Schreiber *et al.* 1997; Li *et al.* 2010). But in this study, rotifer and nauplius were dominant. The difference could result from raw water quality, climate characteristic and operation parameters. Invertebrate species were related to raw water quality, but affected by operation parameters. Therefore, measurements should be taken under specific conditions to control their overgrowth.

Membrane specific flux (SFLUX) and trans-membrane pressure (TMP)

As shown in Figure 5, TMP increased and SFLUX decreased with operating time. TMP was generally in the range 13–30 kPa. The increase in TMP was 0.1–1.25 kPa d⁻¹, on average. SFLUX was generally in the range 2.5–5.5 L m⁻² h⁻¹ kPa⁻¹. The decrease in SFLUX was 0.005–0.07 L m⁻² h⁻¹ kPa⁻¹ d⁻¹ on average. TMP decreased greatly after chemical rinsing with 0.5% NaOH and 0.2% HCl, on day 80. Both

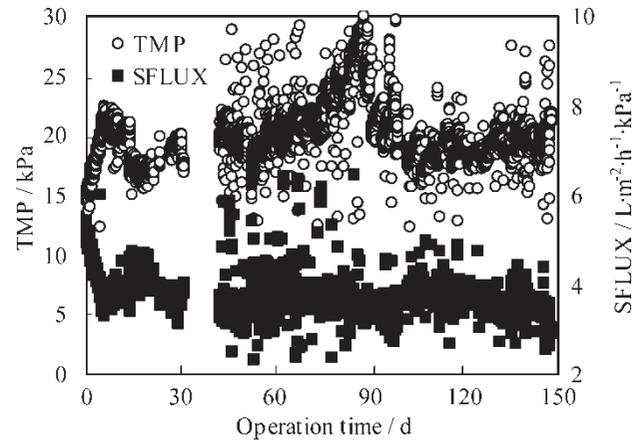


Figure 5 | Trans-membrane pressure (TMP) and membrane specific flux (SFLUX) during UF operation.

parameters are important in describing UF performance. Variations of TMP and SFLUX were related to UF operation condition, such as backwashing, CEB and chemical rinsing. Increased operation time caused the increased TMP and decreased SFLUX, while CEB and chemical rinse could reduce TMP and improve SFLUX. This is consistent with previous studies (Kim & DiGiano 2006).

Change of membrane properties during operation

Roughness

As shown in Figure 6(a), roughness increased from 6.9 ± 1.1 to 8.0 ± 2.1 nm after one operation cycle, and decreased to 7.2 ± 1.2 nm after backwashing. After five cycles, roughness increased to 11.3 ± 2.1 nm and decreased to 11.0 ± 1.2 nm after backwashing. After ten cycles, roughness increased to 20.8 ± 10.6 nm and then decreased to 14.7 ± 2.0 nm after backwashing. The roughness was then reduced to 13.1 ± 5.1 nm after CEB. High roughness means high membrane fouling. High membrane fouling can cause increased TMP and decreased SFLUX. The change of roughness could have a close relationship with cake formation. Roughness increased with prolonged operation time, as indicated by Figure 6(b). Backwashing can reduce the increasing rate of roughness, but not effectively. CEB was effective in controlling roughness, but cannot reduce it to that of a clean membrane. This was also consistent with variations of

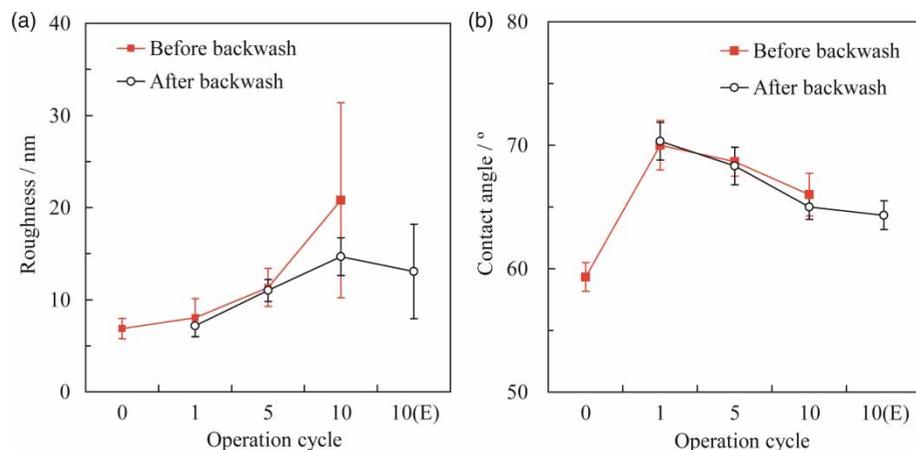


Figure 6 | Change of membrane properties during operation.

TMP and SFLUX (Figure 5). Therefore, roughness had important effects on membrane fouling and UF performance.

Contact angle

Figure 6(b) shows that contact angle increased from $59 \pm 1.2^\circ$ to $70 \pm 2.0^\circ$ after one operation cycle due to membrane fouling. However, after several operation cycles, membrane hydrophilicity increased, as indicated by decreased contact angle, which could be due to the formation of a cake layer by hydrophilic compounds. The hydrophilicity did not change obviously after backwashing. Therefore, membrane foulants could not be removed by only backwashing. In addition, CEB was effective to reduce TMP or increase SFLUX, while it only changed membrane contact angle from $66 \pm 1.7^\circ$ to $65 \pm 1.2^\circ$. These results showed that membrane hydrophilicity did not change obviously during operation, or that even the UF membrane became more hydrophilic with increasing filtration cycles. High hydrophilicity improved SFLUX generally (Kim & DiGiano 2006). However, SFLUX was always decreasing during operation (Figure 5). Therefore, contact angle had little effect on the SFLUX in this study.

Effect of membrane fouling on UF performance

Membrane fouling occurred during UF operation (Figure 5), resulting in increased TMP and decreased SFLUX. Membrane fouling could be attributed to two mechanisms: pore blocking and cake formation (Wang & Tarabara 2008). Pore blocking could play an important role in the earlier

membrane fouling stage and cake formation in the later stage. The cake layer was thin in this study, approximately in tens of nm, which could be estimated on the basis of membrane flux, filtration time, organic matter removal and porosity. This can be confirmed by the results in Figure 6 (a). Membrane fouling changed not only roughness, but also contact angle. Contact angle was dependent on the portion of hydrophilic organic matter, which increased after GAC, as suggested in Figure 1. This is consistent with the variance of contact angle, shown in Figure 6(b).

Membrane fouling could have another important effect on the removal of organic matters. UF was usually not efficient in removing organic matter, especially for small molecular organics (Park & Lee 2005; Gur-Reznik *et al.* 2008). However, some organic matter was, nevertheless, removed by UF in this study. Selected PPCPs were removed partially by UF although the opposite result had been reported previously (Yoon *et al.* 2007). The phenomenon could be due to the function of the cake layer. It was also reported recently that cake layer formation enhanced the removal of organic matter by UF (Choi *et al.* 2005; Xu *et al.* 2006). Biodegradation may also occur due to some microorganisms on the membrane surface (Choi *et al.* 2005; Park & Lee 2005).

CONCLUSIONS

The GAC and UF hybrid process reduced TOC and UV_{254} by 59 ± 12 and $88 \pm 31\%$, respectively. Selected PPCPs

were reduced by 32–92%, with GAC accounting for 75–90%. Particles, bacteria and invertebrates were reduced by approximately 98–99, 76–92 and 100%, respectively during the hybrid process. No invertebrates were detected in the effluent.

TMP increased while SFLUX decreased along with operation time. TMP and SFLUX were 13–30 kPa and 2.5–5.5 L m⁻² h⁻¹ kPa⁻¹, respectively. The change had a close relationship with variation of membrane properties. Roughness also increased with prolonged operation time. Contact angle increased from 59 ± 1.2° to 70 ± 2.0° after one operation cycle, and decreased to 66 ± 1.2° after ten cycles. CEB was effective to reduce TMP, increase SFLUX and control membrane roughness and contact angle, while backwashing by water had little effect on them.

Membrane fouling not only changed membrane roughness and contact angle, but also could have an important effect on the organics removal performance.

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