Recycling of spent filter backwash water using coagulation-assisted membrane filtration: effects of submicrometre particles on membrane flux

Chihpin Huang, Jr-Lin Lin, C. L. Wu and C. P. Chu

**ABSTRACT**

Membrane separation technology has been widely used for recycling of spent filter backwash water (SFBW) in water treatment plant. Membrane filtration performance is subject to characteristics of the particles in the SFBW. A bench-scale microfiltration (MF) coupled with pre-coagulation was set up to evaluate the recovery efficiency of SFBW. Effect of particle size distribution and zeta potential of the coagulated SFBW on the membrane filtration as well as the coagulation strategies were investigated. Pore clogging was more severe on the membrane with 1.0 \( \mu \text{m} \) pore size than on the membrane with 0.5 \( \mu \text{m} \) pore size due to the fact that submicrometre particles are dominant and their diameters are exactly closed to the pore size of the MF membrane. Pre-settling induced more severe irreversible fouling because only the submicrometre particles in the water become predominant after settling, resulting in the occurrence of more acute pore blocking of membrane. By contrast, pre-coagulation mitigates membrane fouling and improves membrane flux via enlarging particle size on membrane surface. The variations of zeta potential in response to coagulant dosing as well as fractal dimension were also compared with the performance of the subsequent filtration. The result showed that pre-coagulation induced by charge neutralization at the optimum dosage where the zeta potential is around zero leads to the optimal performance of the subsequent membrane filtration for SFBW recycling. At such condition, the fractal dimension of coagulated flocs reached minimum.

**Key words** | coagulation, membrane filtration, MF, spent filter backwash water

**INTRODUCTION**

In the conventional water treatment processes, sand filters are designed to remove pollutants, including organic/inorganic particles, protozoa, bacteria and viruses, from the effluent of the coagulation basin. They are periodical backwashed to recover the capacity and the resultant spent filter backwash water (SFBW) thus contains the pollutants which once accumulated in the sand filter. During filter backwashing, SFBW including colloidal materials, organism (e.g. natural organic matter) and inorganic metals (e.g. aluminum or iron) are generated by dislodging accumulated impurities from the filter (Cornwell & Macphee 2001; Edzwald & Tobias 2002). have reported that the spent filter backwash water is generated with approximately 2 to 10% v/v of the finished water (Adin et al. 2002; Bourgeois et al. 2004). In general, SFBW is normally recycled to the head of WTP for the reclamation of waste streams. However, direct recycling of SFBW may have negative impact on finished water quality and jeopardize the drinking safety because pathogens such as *Giardia* and *Cryptosporidium* or other disinfection by-product precursors (DBPs) could be accumulated in water treatment process (Cornwell & Lee 1994; Arora et al. 2001).

Many approaches for treating SFBW have been reported, among which the membrane filtration can achieve...
the best performance. Studies have indicated that the efficient removal of pathogens, DBP and particulate contaminants in SFBW can be reached by using membrane separation process such as microfiltration (MF) or ultrafiltration (UF). For the recycling of SFBW by membrane filtration, membrane fouling and pore clogging caused by organic matter or colloidal particles will reduce membrane flux and its recovery rate after backwashing (Bourgeois et al. 2004; LeGouellec et al. 2004; Reissmann & Uhl 2006; Walsh et al. 2008).

Recently, a hybrid separation process is usually used to improve membrane flux. Pre-coagulation coupled with membrane separation is generally adopted to form large and permeable floc on membrane surface, which reduces membrane fouling and clogging that can increase the membrane flux (Guigui et al. 2002; Howe et al. 2006; Chen et al. 2007). On the other hand, pre-coagulation conditions such as pH and dosage of Al coagulants also affect the properties of flocs significantly, which influences the subsequent solid-liquid separation, including specific cake resistance as well as dewaterability of coagulated suspension and governs the subsequent filtration efficiency (Lee et al. 2000; Lin et al. 2008a; Walsh et al. 2008). Although investigations have suggested that coagulation-membrane filtration process can facilitate the water quality of filtrated SFBW and the costs for operation and maintenance at such condition are close to that of a conventional water treatment process (Song et al. 2001; Nasser et al. 2002), the effect of submicrometre particles on membrane flux for SFBW recycling by using coagulation-assisted membrane filtration has not been deeply studied.

Most studies on membrane filtration of SFBW with pre-coagulation have mainly focused on the filtrate quality. Very little study concerns the effects of SFBW characteristics on membrane filtration performance. In this study, we focused on the effects of particle size distribution in SFBW on recycling by using membrane separation. The strategies of pre-coagulation were discussed as well.

METHODS

SFBW water sample

The SFBW samples were collected from the residual streams discharge ditch of traditional sand filters in Hsinchu Water Treatment Plant (Hsinchu, Taiwan). Hsinchu WTP conventional treatment trains for surface water, with coagulation, flocculation, sedimentation and dual-media filtration. The filters were backwashed by finished water every 24 h for 10 min. Since the composition and turbidity of the SFBW vary tremendously during filter-backwashing, the SFBW samples were withdrawn from the filter at a fixed sampling frequency over one backwash cycle until the desired sample volume was obtained. Characteristics of the SFBW samples are summarized in Table 1.

Coagulants

Commercial polyaluminium chloride (PACl) (Al2O3 = 10%; γ = 1.4) was purchased from Showa Chemicals Inc. and was used as coagulant for this study. Working solutions containing 1,000 mg/L as Al were freshly prepared before each experiment. The Al concentration was analyzed by inductively coupled plasma-atomic emission spectrometry (ICPAES, JY24, Jobin-Yvon Inc., France). The Al speciation in PACl was determined by Ferron method, the Al speciation distribution of PACl used in this study was categorized into three groups, monomeric Al (Ala), polymeric Al (Alp), colloidal Al (Alc), which are 42.3%, 8% and 49.7% (Lin et al. 2009).

Pre-coagulation protocol

The SFBW was pre-coagulated with PACl before the membrane filtration. A pre-determined amount of coagulant was injected into the SFBW sample at a controlled pH

Table 1 | Filtrate quality of filtered SFBW by membrane filtration with pore sizes of 1.0 μm and 0.5 μm

<table>
<thead>
<tr>
<th>Parameters</th>
<th>SFBW</th>
<th>Permeate (1.0 μm)</th>
<th>Permeate (0.5 μm)</th>
<th>Standard</th>
</tr>
</thead>
<tbody>
<tr>
<td>Suspended solid (mg/L)</td>
<td>432</td>
<td>ND</td>
<td>ND</td>
<td>–</td>
</tr>
<tr>
<td>Turbidity (NTU)</td>
<td>685</td>
<td>0.43</td>
<td>0.42</td>
<td>2</td>
</tr>
<tr>
<td>pH</td>
<td>7.9</td>
<td>7.9</td>
<td>7.9</td>
<td>6–9</td>
</tr>
<tr>
<td>DOC (mg/L)</td>
<td>1.31</td>
<td>1.23</td>
<td>1.25</td>
<td>–</td>
</tr>
<tr>
<td>UV254</td>
<td>0.023</td>
<td>0.023</td>
<td>0.023</td>
<td>–</td>
</tr>
<tr>
<td>Total bacteria (cfu/mL)</td>
<td>–</td>
<td>97</td>
<td>46</td>
<td>100</td>
</tr>
<tr>
<td>Total coliform (cfu/100 mL)</td>
<td>–</td>
<td>&lt;1</td>
<td>&lt;1</td>
<td>6</td>
</tr>
</tbody>
</table>
of 7, followed by a rapid mixing at 200 rpm for 1 min, a slow mix of 20 min at 30 rpm and a final 20 min settling. The coagulated sample was used as the feed for the dead-end MF membrane module. The coagulant (Al) dosage is expressed in mg/L as Al in this study.

**Membrane filtration**

PTFE MF (Gore, USA) membranes with polypropylene supporting materials were used in this study. The diameter of the membrane was 5 cm and the effective filtration area was 0.002 m². Two pore sizes, namely, 0.5 and 1.0 μm, were examined.

A schematic diagram of the dead-end MF system used in this study is shown in Figure 1. The membrane module was submerged in the 1.5-L flask. A magnetic stirrer was used to homogenize the test sample and to keep the particles suspended. A constant filtration pressure of 0.63 bar was supplied by a vacuum pump (GAST, USA). The permeate flux was determined by weighing permeates on an electronic balance and recorded via a personal computer equipped with an autoreading program.

Before each filtration, the virgin membrane was immersed in ultrapure water for two hours followed by a filtration of 1,000 mL ultrapure water to remove the impurities. The clean-water flux \( J_0 \) was first determined. Then, the same procedure was performed for the SFBW samples, and the measured flux was denoted \( J \). After each filtration, the membrane was backwashed (pulse of air injection in pressure of 0.5 bar) to remove the cake from the membrane surface. Water qualities of the feed water and the corresponding filtrate, including turbidity, pH, dissolved organic carbon (DOC), total number of coliform and bacteria were determined. All analysis followed the standard methods of the EPA of Taiwan (Taiwan EPA 2007).

**Particle size distribution measurement**

Two particle sizers were adopted to determine the size distribution of the SFBW samples before and after coagulation. The particle sizer equipped with small-angle laser light scattering (SALLS) (Mastersizer 2000, Malvern, UK) was applied to measure the particle size in the range from \( 10^{-1} \) to \( 10^3 \) μm. Meanwhile, the fractal dimensions of coagulated flocs were obtained from the data of SALLS, as reported by previous study (Lin et al. 2008b).

The particle size distribution in nano-scale (100–1,000 nm) in the SFBW and filtrate was measured by the Zetasizer nano ZS (Malvern, UK) which also analyzed the zeta potential of the colloids. The majority of micro-size particles in SFBW samples are in the vicinity of 30 μm. The majority of the submicrometre particles of SFBW (5 January 2006) is around 955 nm.

**RESULTS AND DISCUSSION**

**Water qualities of SFBW and membrane filtrate**

Water qualities of the SFBW and filtered SFBW through two dead-end membranes with different pore size were determined and listed in Table 1. Satisfactory pollutants removal was observed for both membranes, especially the total coliform and total bacteria, which comply with the current Drinking Water Quality Standard of Taiwan EPA. However, the DOC was slightly reduced through membrane filtration in which cake or membrane adsorption
could contribute to the reduction of organic matter. The result indicated that organic fouling on PTFE MF membrane is slight. The organic matter in SFBW is rather hydrophilic because the SUVA value (UV254/DOC) of SFBW is lower than 2; as a consequence, the hydrophobic PTFE membrane does not favor organic fouling in this study.

Membrane filtration of SFBW and pre-settled SFBW

For microfiltration, particles in the size range of 0.1 and 1 μm was most likely to influence the membrane flux. Four filtration/backwash cycles of the SFBW was conducted for this study. The relative flux $J/J_0$ was plotted against the accumulated filtrate volume ($V$), as depicted in Figure 2(a) and (b) for membranes of 0.5 and 1.0 μm, respectively. For 0.5 μm membrane filtration, the initial flux maintained about the same level after each backwash. In 1.0 μm membrane filtration, the initial flux declined significantly from 4.12 m³/m²-hr of first cycle to 3.03 m³/m²-hr of the 4th cycle, a 25% decline, indicating a serious irreversible fouling. It can be explained by the submicrometre particles size distribution. Le & Howell (1984) have indicated that severe flux decline occurs when the size of the particle is closed to the pore size of the membrane due to pore blocking. In our experiments, the majority of submicrometre-scale particle (955 nm) in SFBW was larger than membrane pore size of 0.5 μm. As a result, a large number of submicrometre-scale particles can be rejected on membrane surface during filtration. On the contrary, because submicrometre-scale particle was smaller than 1.0 μm of membrane pore size, the membrane fouling was caused due to nano-particle blocking, which resulted in the occurrence of irreversible fouling. Thus, the initial flux cannot be restored by backwashing in 1.0 μm membrane filtration. However, irreversible fouling was not observed for the 0.5 μm membrane because the particles were

Figure 2 | The relative flux $J/J_0$ vs. the accumulated filtrate volume $V$ for SFBW with and without pre-settling for various membrane filtration (a) 0.5 μm (without pre-settling) (b) 1.0 μm (without pre-settling) (c) 0.5 μm (with pre-settling) (d) 1.0 μm (with pre-settling).
mostly twice the pore size of the membrane. The results suggested that submicrometre-scale particle were critical for irreversible membrane fouling.

To enhance the filtration performance, the SFBW was pretreated by two methods: settling and coagulation/flocculation. Large particles in SFBW can be removed preferentially through pre-settling, and then the solids of supernatants became less, which is favorable for the flux improvement in membrane filtration. First, the SFBW was settled at various durations before the filtration. The supernatant after 15 min-settling was used as the pre-settled SFBW for microfiltration. The flux profiles were presented in Figure 2(c) and (d). Contrast to the flux profiles of the unsettled SFBW, as seen in Figures 2(a) and (b), the initial flux of both 0.5 and 1.0 μm membrane filtration decreased dramatically with filtration cycle. The irreversible fouling for both 0.5 and 1.0 μm membranes were much more severe when the SFBW was pre-settled. The relative flux of the fourth cycle dropped 64% for the 0.5 μm membrane filtration. For 1.0 μm membrane, more severe irreversible fouling was observed with an 80% decline. Since the pre-settling removes large particles, leaving only the submicrometre particles in the supernatant, the initial flux was severely reduced by the submicrometre particles which were smaller than the membrane pore. With respect to the filtration of raw SFBW, the cake layer of large particles removes a significant portion of submicrometre-scale particles before they reach the primary membrane surface.

Membrane filtration of pre-coagulated SFBW

Jar tests were performed to determine the optimum coagulant dosage for the SFBW. The PACI applied here destabilized the particles of SFBW predominately by enmeshment at pH 7 since the large amount of aluminium hydroxide formed (Lin et al. 2008c). The variations of residual turbidity and zeta potential (measured after 1-min rapid mixing) indicated an optimum dosage of 2.5 mg/L. The filtration profiles of the coagulated SFBW samples were depicted in Figure 3. Pre-coagulation has clearly slowed down the decline in membrane flux, and the degree of effect is related to the efficiency of coagulation. The relationship is discussed in terms of particle characteristics including particle size, zeta potential and fractal dimension in the following sections.

The effects of coagulation on particle size distributions are analyzed in micrometre and nano scales. The results are shown in Figures 4(a) and (b), separately. The mode of the particle size distribution shifted toward large sizes in both scales. The largest transition occurred at optimum dosage and then gradually decreased with the increasing coagulant dosage.

The zeta potential of the coagulated SFBW was compared with the corresponding average flux, which is depicted in Figure 5(a). The zeta potential increased continuously with the addition of coagulant. It reached zero around the optimum dosage (2.5 mg/L), indicating that charge neutralization was the main coagulation mechanism. Lee et al. (2000) has indicated that cake layer is related to
the mechanism of pre-coagulation in dead-end MF. The specific cake resistance of those from charge neutralization is smaller than that from sweep coagulation. The result indicated that charge neutralization plays an important role in pre-coagulation coupled with membrane filtration for the recycling of SFBW.

Moreover, the fractal dimension of the pre-coagulated SFBW under various coagulant dosages was determined and compared with the average flux, as shown in Figure 5(b). The floc with lowest fractal dimension was also found at the optimum coagulant dosage where the highest average flux of membrane occurred, which could be explained by the flocs structure. Many previous studies have suggested that low cake resistance were associated with large floc or low fractal dimension (Veerapaneni & Wiesner 1996; Lee et al. 2003). In this study, the floc formed by Pre-coagulation with charge neutralization is largest and has the lowest fractal dimension. The smaller the fractal dimension, the loose the flocs and, therefore, the higher the porosity of the cake as well as the lower specific cake resistance (Cho et al. 2005). As a result, the flow can quickly pass through the pore of cakes formed by pre-coagulation at the optimum dosage.

CONCLUSIONS

Particle size distribution determines the mechanism of pore plugging. The presence of submicrometre particles in SFBW increases the irreversible fouling in MF membrane filtration. Irreversible fouling occurs when the size of the submicrometre particles is smaller than the pore size of the MF due to pore plugging. For SFBW recycling, pre-settling of SFBW before membrane filtration will worsen the performance of membrane separation. The coagulation pretreatment enhances the performance of dead-end MF operation via the increase in flux and decrease in the degree of pore blockage. It was found that pre-coagulation induced by charge neutralization at the optimum dosage, where the zeta potential is around zero and the fractal dimension of floc is the lowest, leads to the optimal
performance of the subsequent membrane filtration for SFBW recycling. The performance of MF membrane with pore size of 0.5 μm is superior to that of 1.0 μm due to the presence of submicrometre-scale particles. Overall, MF membrane filtration with pre-coagulation for SFBW recycling is technically feasible.

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REFERENCES


Taiwan EPA 2007 Standard Method for the Examination of Water Quality in Environmental Analysis Laboratory, EPA, Taipei, Taiwan.
