Long-term studies on hybrid ceramic microfiltration for treatment of surface water containing high dissolved organic matter
A. Abeynayaka, C. Visvanathan, S. Khandaritith, T. Hashimoto, H. Katayama, Y. Matsui and D. R. I. B. Werellagama

ABSTRACT
This long-term pilot-scale study on the performance of ceramic microfiltration (CMF) was conducted at the Bangkhen water treatment plant (BWTP), with the raw water from Chaophraya River, Thailand. Raw water turbidity and dissolved organic carbon (DOC) were varied in the ranges of 20–210 NTU and 3.0–8.5 mg/L respectively. The hybrid pilot-scale CMF (Pilot-CMF) operational parameters were optimized with the aid of jar-tests and laboratory-scale CMF (Lab-CMF) operations. The systems were operated with various polyaluminum chloride dosages and filtration cycle times. Pilot-CMF provided excellent steady turbidity removal compared to the conventional water treatment process. DOC removal percentages of Pilot-CMF and the conventional process at the BWTP were 49% and 30% respectively. With different coagulant dosages, unique patterns in transmembrane pressure (TMP) variations were observed. The daily TMP increment under low turbidity conditions was 0.08 kPa/day. During rainy periods (turbidity over 100 NTU) the TMP increment reached 0.79 kPa/day. However, once the turbidity of raw water reaches normal conditions (30–60 NTU at the BWTP) the Pilot-CMF system recovers the TMP increment due to efficient backwashing.

INTRODUCTION
For decades, rapid sand filtration-based conventional water treatment processes have been used to obtain potable water for city supplies. New challenges, including raw-water quality degradation, quantity limitations, growing attention to consumer safety, such as stringent standards of treated water, and by-products of the conventional treatment process, open the way for new alternative technologies of water treatment. Outbreaks of waterborne diseases such as giardiasis have drawn attention to the reliability of the conventional water treatment process. Moreover, carcinogenic disinfection by-products such as trihalomethanes have drawn more attention to raw water sources with higher dissolved organic matter (DOM). Especially in tropical regions of developing countries where the DOM concentrations in raw water are relatively high, the risk of trihalomethane formation is also high.

Membrane filtration technologies are an alternative option in drinking water treatment. Microfiltration (MF) is effective for the reduction of turbidity and removal of algae, protozoa and bacteria. The combination of a coagulation–flocculation process with MF enhances the removal of viruses (Zhu et al. 2005; Shirasaki et al. 2009) and dissolved substances (Yuasa 1998; Konieczny et al. 2006; Loi-Brügger et al. 2006; Oh et al. 2007; Meyn et al. 2008; Shirasaki et al. 2010) such as DOM. Ceramic microfiltration (CMF) membranes have become noted in the field of potable water treatment (Bottino et al. 2001; Lerch et al. 2005; Matsushita et al. 2005; Kommineni et al. 2010) due to

doi: 10.2166/ws.2013.194
their insensitivity to bacterial action and ability to withstand extreme acidity, alkalinity and higher operating pressures than polymeric membranes. The properties of ceramic membranes allow the use of strong chemical cleaning and application of high pressure for hydraulic backwashing. Apart from that, ceramic membranes have attracted attention due to their long, reliable lifetimes and environmentally safer disposal over polymeric membranes (Finley 2005). Hence, ceramic membranes have become a reliable option for potable water treatment over polymeric membranes. There are a number of pilot- and laboratory-scale studies reported on CMF applications in potable water treatment, with surface water sources (Lerch et al. 2005; Matsushita et al. 2005; Loi-Brügger et al. 2006; Meyn et al. 2008; Shirasaki et al. 2010; Hofs et al. 2011). However, long-term pilot-scale studies on CMF applications for potable water treatment in tropical regions, where the water quality is significantly different from the other regions, are hardly found in the literature. Moreover, Abeynayaka et al. (2012) reported that hybrid CMF (0.1 μm) with coagulation–flocculation processes is capable of removing hydrophilic DOM with higher efficiency than the conventional treatment process. The combined effect with the coagulant and other constituents leads to a higher removal of all the fractions of dissolved organic carbon (DOC). Due to these combined effects, hybrid CMF is capable of providing treated water with lower trihalomethane formation potential (THMFP) under high-DOM tropical surface water conditions.

Reported studies of hybrid CMF applications in surface water treatment have mostly been conducted with raw water sources of turbidity less than 10 NTU and DOC concentrations less than 3 mg/L. Water in typical surface-water sources (e.g. rivers) in tropical regions may exceed those limits, with turbidity values over 30 NTU and DOC around 6 mg/L. Reported studies with raw water containing higher DOC (Ericsson & Trägårdh 1997; Hofs et al. 2011) are laboratory-scale batch filtrations. The behavior of CMF in those cases may not simulate the behavior of CMF under actual, complex raw-water conditions and treatment process conditions. Apart from that, the optimization of the treatment part of the hybrid CMF process is an important requirement for the effective implementation of CMF in the potable water treatment industry. Hence, long-term operation under actual field conditions is important. Coagulant dosages in the conventional treatment process are often decided by jar-tests. For the conventional process, big and heavy flocs are favored for efficient settling. However, for hybrid CMF the implementation of standard jar-test observations is uncertain due to differences in post-treatment techniques such as filtration with MF. Supporting these process differences, Lerch et al. (2005) mentioned that the formation of larger flocs in the hybrid CMF process is unnecessary due to the small pore size in CMF and the destruction of larger flocs while entering CMF capillaries. Hence, cross-checking the jar-tests’ optimum values with the hybrid CMF process is important.

When the treatment plant is expected to operate at a regional location with minimum interference from human technicians, performing jar-tests and making frequent adjustments of coagulant dosages is difficult. In a tropical climate and with typical land-use patterns, a sudden rainfall may change the source-water turbidity within hours. Moreover, the treated water quality and membrane fouling in CMF is sensitive to pre-treatment decisions, such as coagulant dosage. Hence, studies on the performance of hybrid CMF systems under actual operating conditions with minimum human interference are important.

**METHODS**

This long-term continuous pilot-scale study on the performance of hybrid CMF was conducted at the Bangkhen water treatment plant (BWTP), Bangkok, Thailand. The experimental framework of the study is illustrated in Figure 1. This study consisted of three phases, namely jar-tests, initial laboratory-scale optimization works and the pilot-scale operation at the BWTP.

**Water source and the BWTP**

Raw water for laboratory-scale CMF (Lab-CMF), pilot-scale CMF (Pilot-CMF) and the actual-scale conventional water treatment plant was drawn from Chaophraya River, Thailand. The BWTP consists of conventional water-treatment units including coagulation,
followed by sedimentation, rapid sand filters and disinfection using chlorine. Operational conditions such as coagulant dosage at the BWTP were decided by the plant operators based on optimization experiments conducted at 4-hour intervals. This conventional water treatment plant (BWTP) has the capacity to produce 3.2 million m$^3$ of water per day.

**Initial jar-tests with raw water**

Selection of optimum coagulant (polyaluminum chloride (PACl)) dosage, velocity gradient ($G_C$) and duration ($t_C$) of the coagulation process, and velocity gradient ($G_F$) and duration ($t_F$) of the flocculation process was made through a series of jar-tests (12 types of jar-test runs with different $G$ and $t$ values). Three different rapid-mixing speeds having corresponding $G$ values of 190, 230, and 270 s$^{-1}$ were applied. Rapid-mixing duration was modified as 50, 100, 150 and 200 s. For each run the coagulant dosages were kept at 0, 0.5, 1.0, 1.5, 1.8 and 2.0 mg Al/L. Slow mixing with a $G$ value of 50 s$^{-1}$ was conducted for another 10 min after the rapid mixings. Samples were collected at the beginning and end of rapid mixing and at the end of every minute of slow mixing. Collected samples were filtered through GF/F (0.7 μm) filter papers with measurements of the turbidity, DOC and UV$_{254}$ (ultraviolet 254 nm wavelength) absorbance of the filtrates. Variation of measured parameters was plotted against coagulant dosages and mixing durations respectively. Floc size and floc formation/breakage were observed. Using these jar-test observations, the optimum ranges of coagulant dosage, $G_C$, $t_C$, $G_F$ and $t_F$ were selected.

**Laboratory-scale CMF (Lab-CMF)**

Lab-CMFs were conducted to check the applicability of the optimum ranges selected through jar-tests. Three different rapid-mixing speeds with corresponding $G_C$ values of 210, 230 and 250 s$^{-1}$ were applied (jar-test optimum was 230 s$^{-1}$). Rapid-mixing duration was modified as 90, 100 and 110 s (jar-test optimum was 100 s). For each run the coagulant dosages were kept at 0, 1.0, 1.2, 1.5, 1.8 and 2.0 mg Al/L.

Properties of the Lab-CMF are given in Table 1 and Figure 2(a). The feed-water sample (coagulated and rapid-mixed) was added to a 2 L pressure tank. Constant transmembrane pressure (TMP) for filtration was employed using the pressurized nitrogen gas with a pressure regulating system.

**Table 1 | Properties of CMF systems**

<table>
<thead>
<tr>
<th>Description</th>
<th>Property</th>
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</thead>
<tbody>
<tr>
<td>Membrane area of Pilot-CMF</td>
<td>0.18 m$^2$</td>
</tr>
<tr>
<td>Membrane area of Lab-CMF</td>
<td>0.04 m$^2$</td>
</tr>
<tr>
<td>Operated membrane water flux of Pilot-CMF</td>
<td>60 L/m$^2$.h</td>
</tr>
<tr>
<td>Nominal pore size</td>
<td>0.1 μm</td>
</tr>
<tr>
<td>Operated filtration pressure range</td>
<td>10–45 kPa</td>
</tr>
<tr>
<td>Backwash pressure</td>
<td>400 kPa</td>
</tr>
<tr>
<td>Blowdown pressure</td>
<td>200 kPa</td>
</tr>
<tr>
<td>Backwash water volume per unit area of membrane</td>
<td>3.3 L/m$^2$</td>
</tr>
<tr>
<td>Material</td>
<td>Ceramic</td>
</tr>
<tr>
<td>Channel diameter</td>
<td>2.5 mm</td>
</tr>
<tr>
<td>Number of channels</td>
<td>55</td>
</tr>
<tr>
<td>Supplier</td>
<td>NGK insulators</td>
</tr>
</tbody>
</table>
Pressurized feed passed through the spiral static flocculator \((G_F = 50 \text{ s}^{-1}, t_F = 120 \text{ s})\) before reaching the CMF. Constant-pressure \((30 \pm 1 \text{kPa})\) batch filtrations were performed and the filtrate reduction rate was measured. Between every run, the membranes were chemically cleaned as described in Lerch et al. (2005).

Pilot-scale CMF (Pilot-CMF)

Pilot-CMF, illustrated in Figure 2(b) and having properties indicated in Table 1, was operated for a period of 21 months at the BWTP. Water was pumped from the intake of the BWTP to the coagulation tank, where PACl was added and mixed \((G_C = 230, t_C = 100 \text{ s})\). Then the water was pumped through the static flocculator \((G_F = 50 \text{ s}^{-1}, t_F = 120 \text{ s})\) before reaching CMF. Constant filtrate flux was maintained using a flowmeter-coupled control system. TMP was measured using pressure gauges and recorded continuously in a data logger. The overall system was programmed and automatically operated. At the initial stage, the plant was operated under different coagulant dosages of 0, 1.0, 1.5, 2.0 and 3.0 mg Al/L with a backwash interval of 2 h (backwash was conducted automatically with stored filtrate in a backwash tank. There was no chemical addition for the automated backwashing). Then the backwash interval was altered as 2, 2.5, 3 and 3.5 h. During this period the coagulant dosages were kept at the optimum values obtained from the initial steps of the study. At the final stage the system was operated under the optimized backwash interval (3 h) and constant coagulant dosage 1.5 mg Al/L.

Analytical parameters

DOC and UV\textsubscript{254} were measured according to standard methods (APHA 2005) using filtrates of GF/F (0.7 μm). Specific ultraviolet absorbance (SUVA) was calculated using UV\textsubscript{254} and DOC (UV\textsubscript{254}/DOC). The turbidity, pH and energy consumption of the system were measured using a turbidity meter (HACH 2100N), pH meter (Oakton®) and power meter (Energy logger® 4000) respectively.

RESULTS AND DISCUSSION

Jar-test and Lab-CMF observations

The optimum \(G\) and \(t\) values of the coagulation and flocculation process were found to be \(G_C = 230 \text{ s}^{-1}, t_C = 100 \text{ s}, G_F = 50 \text{ s}^{-1}\), and \(t_F = 120 \text{ s}\). Hence, the Pilot-CMF \(G\) and \(t\) values were fixed at those values. Lab-CMF operating under those
$G$ and $t$ values with raw water (turbidity = 35 NTU and DOC = 6 mg/L) and optimized PACl dose (1.5 mg Al/L) gave filtrate of turbidity <0.02 mg/L and DOC = 2.7 mg/L. Moreover, an offset of coagulant dosage by ±0.5 mg Al/L from the optimum range doubled the flux decline percentage (Table 2). This could have been due to poor floc formation leading to membrane fouling (Lerch et al. 2005). Raw water pH (water from the Chaophraya River) was around 7.5 (during the long-term operation period it was 7.46 ± 0.24), and during jar-tests the pH dropped by amounts ranging from 0.0 to 0.18 based on the raw water quality and amount of coagulant added. However, these slight pH changes have not shown a notable effect on the coagulation process. Hence, it was decided to conduct the Pilot-CMF system operations without pH adjustment.

### Table 2

<table>
<thead>
<tr>
<th>Coagulant dosage (mg Al/L)</th>
<th>Flux decline (%)</th>
<th>DOC removal (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>48.4 ± 2.7</td>
<td>9.5 ± 1.6</td>
</tr>
<tr>
<td>1.0</td>
<td>10.4 ± 1.8</td>
<td>27.3 ± 4.1</td>
</tr>
<tr>
<td>1.2</td>
<td>5.3 ± 1.2</td>
<td>38.2 ± 3.9</td>
</tr>
<tr>
<td>1.5</td>
<td>4.8 ± 1.2</td>
<td>40.9 ± 5.2</td>
</tr>
<tr>
<td>1.8</td>
<td>4.8 ± 1.1</td>
<td>38.0 ± 2.2</td>
</tr>
<tr>
<td>2.0</td>
<td>9.7 ± 1.2</td>
<td>36.7 ± 1.3</td>
</tr>
</tbody>
</table>

Pilot-CMF provided better removal than the conventional water treatment process. The filtrate turbidity from CMF and the conventional treatment process at the BWTP was <0.02 and 0.05–2.0 NTU respectively (Figure 3(a)). Raw water turbidity varied over a wide range, indicating values greater

**Figure 3**

Turbidity and DOC variation of raw and treated waters: (a) turbidity variation; (b) DOC variation.
than 100 NTU during rainy periods. Turbidity of filtrate from the conventional process varied with time, especially during rainy sessions (reaching 2 NTU), while the CMF filtrate remained at low turbidity and steady. Higher treated-water turbidity reduces the efficiency of post-disinfection processes (LeChevallier et al. 1984). Therefore, irrespective of raw water turbidity, the CMF system could provide treated water with favorable conditions for post-disinfection.

**DOC removal**

Average DOC removal by CMF was 49% while the conventional treatment process achieved only 30% (Figure 3(b)). Unlike the steady turbidity removal by CMF, DOC removal was unsteady during the operation. However, the DOC variation for CMF filtrate shows a similar pattern of variation to the raw water DOC. The main mechanism of DOM removal by hybrid CMF is aggregation and floc formation followed by filtration. However, this process is sensitive to the nature of the DOM, the coagulation–floculation process and water quality conditions such as pH, alkalinity and temperature (Yan et al. 2009). In Pilot-CMF system operation, the treated water pH was observed to be around 7 (7.1 ± 0.18). Hence, there was no negative effect of the operation pH on the coagulation process. During the long-term operation of Pilot-CMF (with 1.5 mg Al/L dosage), the DOC concentration of raw water had varied (Figure 3(b)). Moreover, the SUVA had varied from 2.2 to 7.3 indicating the nature of the DOM had varied during the operation. Hence, the observations of varying DOM removal efficiencies are understandable. Apart from that, DOM could be adsorbed to the ceramic membrane surface causing irreversible membrane fouling. Raw water with higher DOM concentrations often causes higher levels of irreversible membrane fouling. Moreover, the remaining DOM may generate harmful disinfection by-products at later stages of treatment if chlorination is the disinfection method. However, with 1.67 times more DOM removal compared to the conventional treatment, CMF could provide filtrate with less THMFP. Abeynayaka et al. (2012) reported that a Pilot-CMF system operated under the same conditions as in this study was capable of reducing THMFP by 1.54 times more than the conventional water treatment process.

**TMP variation during a cycle (interval between two backwashes) of Pilot-CMF**

Figures 4(a) and 4(b) indicate TMP variations during two consecutive cycles under different coagulant dosages and different cycle times. The figures show that the TMP increases as the cycle time increases, indicating the need for more frequent backwashing to prevent membrane fouling.
filtration cycle times respectively. Coagulant dosage has a direct influence on TMP increment. Absence of coagulation or deviations of dosage from the optimum value induces a higher TMP increment during a cycle. Yonekawa et al. (2004), who researched with a similar membrane module to the one in this study, described the floc formation and hydraulic flow pattern in a ceramic membrane module as unique. Furthermore, during a filtration cycle a number of processes occur such as particle aggregation, floc lifting, and cake formation. Yonekawa et al. (2004) also mentioned that flocs larger than 1 μm tend to lift from the membrane surface. However, oversized flocs could be destroyed while entering membrane channels (Lerch et al. 2005). In this study, the rapid TMP increment (4 kPa during a cycle) without coagulation could be due to no floc lifting due to minimum aggregation. Coagulation-aided floc formation could minimize the TMP increment due to floc-lifting, during the cycle operation. If the preceding coagulation–flocculation process is conducted under optimized conditions (corresponding to 1.5 mg Al/L dosage), there is little retention on the membrane, which leads to a smaller TMP increment. When the dosages deviate from the optimum value, the floc properties change (from jar-test observations under different dosages). Smaller, strong flocs may accumulate on the membrane surface causing a relatively larger TMP increment (likely to occur with 1.8 and 2.0 mg Al/L dosages). Perfect description of these unique TMP variation patterns is difficult without physical investigation (which is difficult with ceramic tubular membranes during operation) or computer-aided modeling. However, these experimental observations can be effectively utilized for CMF process optimization in full-scale water treatment plants. One possible concern is optimizing energy consumption for filtration. TMP is the major governing parameter of the energy requirement for filtration. With optimized coagulant dosage, average TMP during a cycle could be reduced by 1–3 kPa. If the filtration process takes place at a low pressure range such as 10–30 kPa, this variation equals to 5%–10% of the filtration energy. Hence, for low-pressure (such as MF) filtration applications, the use of optimum coagulant dosages could reduce energy consumption by a considerable amount.

**Cycle time and water recovery**

Increments of filtration cycle time allow accumulation of larger amounts of foulants at the membrane, leading to a larger TMP increment. Furthermore, increments of higher contact time for organics with the ceramic membrane may increase irreversible fouling. However, frequent backwashing reduces water recovery due to loss of treated water. Looking at the long-term average energy consumption (energy consumption of the 2, 2.5, 3 and 3.5 h filtration cycles was 0.35, 0.32, 0.30 and 0.31 kWh/m² respectively) and overall TMP patterns, 3 h was selected as the cycle time for long-term operation (for the turbidity range 20–50 NTU). Yet for different feed-water conditions the optimum cycle time may vary from this. The relative energy consumption decreases from 2 to 3 h cycles, due to reduced backwash frequency. However, further increase of cycle time increased the TMP increment within the cycle, causing a slight increase in energy consumption. Apart from energy consumption, the water recovery of the CMF system also varied with the cycle time. Pilot-CMF with operation cycle times 2, 2.5, 3 and 3.5 h provided water recovery of 96.8%, 97.5%, 98.0% and 98.2% respectively. Hence, increments of operation cycle time provide higher water recovery.

Average water recovery at the BWTP during the 21-month period was 95.2% (water recovery by the CMF system is 96.8 to 98.2% under tested conditions). Monolith CMF, having efficient backwash, which consumes a low water volume compared to the conventional process, could provide higher water recovery than the conventional process. Moreover, the issues associated with treating larger volumes of sludge can be overcome with the CMF system, which produced 33% to 62% less sludge volume. Presently the sludge treatment facility to treat sludge produced from the conventional water treatment process of the BWTP occupies more than half of the treatment-plant land area.

**Effect of turbidity on TMP**

Long-term TMP variation under constant coagulation dosage (1.5 mg Al/L) and filtration cycle time (3 h) with varying raw water turbidities is given in Figure 5. In the initial period (zone A), the turbidity varied between 20 and 30 NTU. During this period, TMP gradually increased at a rate of 0.08 kPa/day. However, the sudden increase in turbidity (zone B) caused a rapid TMP increment at a rate of 0.79 kPa/day. Due to prolonged high turbidity, the Pilot-CMF system was unable to recover the TMP increment.
through backwashing with filtrate water. In this study period, no chemical enhanced backwash (CEB) was conducted. However, for similar membrane modules, CEB (with acid added to the filtrate) has shown effective TMP recovery (Lerch et al. 2005). After this period (on the 156th day), the membrane was replaced with an identical new one. Then the dismantled membrane was chemically treated and the initial flux was restored. Hence, in actual operations, instead of membrane replacement, CEB could be conducted to recover the TMP. After replacing the membrane in this study a slight increment of TMP (similar to that in the initial operation) under low turbidity conditions was observed. However, the sudden peak of turbidity (zone D) caused a sudden increase in TMP. Yet, after the turbidity reached the lower range (zone E) the original TMP increment was almost recovered. This is due to progressive removal of foulants by the backwashing process and the reduction of fouling due to the optimum coagulant dosage matching the reduced turbidity. A similar relation between turbidity and TMP can be seen during the next operation period. This proves that rapid TMP increments due to high turbidity are reversible in a long-term operation. However on some occasions, if the high turbidity is prolonged, due to rapid increments of TMP, the membrane may have to be chemically cleaned to remove the membrane fouling, as backwashing is not enough for recovery.

CONCLUSIONS

Considering this long-term continuous data, the CMF system is shown capable of treating high-DOM surface water to provide better quality drinking water, compared to the conventional water treatment process, under the operated conditions. The hybrid CMF process gives higher water recovery and a lower volume of sludge. The sensitivity of TMP variation to the coagulant dosage within a filtration cycle is quantified. Further studies on energy consumption variation with coagulant dosage and filtration cycle time should be conducted on CMF systems for minimizing energy consumption. The rapid TMP increase due to short-term high turbidity is reversible during long-term low turbidity operation with the operated raw water source. For these types of raw water which are common in many large city supplies, to provide safer water with sustainable operation, irreversible membrane fouling and DOM removal efficiency are identified as important factors for further study.

ACKNOWLEDGEMENTS

The authors would like to acknowledge Mr Manit Pongchalemporn from the Metropolitan Water Works Authority, Bangkok, Thailand and the rest of staff of Bangkhen water treatment plant for their support and provision of facilities to conduct the study.

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First received 30 April 2013; accepted in revised form 21 August 2013. Available online 13 September 2013