A modified fouling index (MFI40) and fouling predicting approach for ultrafiltration of secondary effluents

B. J. Cai, I. Baudin and H. Y. Ng

ABSTRACT

Fouling indices for evaluating fouling propensity of secondary effluents (SEF) as feed of ultrafiltration (UF) systems are important parameters for the design and operation of the UF process. However, limited fouling indices have been developed and applied for UF feedwater. This study (i) established a modified UF fouling index (MFI40) by raising operating pressure from 30 psi in a traditional MFI test to 40 psi. Standard deviation of MFI40 tests was lower than that of traditional MFI by 68.6%, indicating better stability and repeatability of MFI40. It (ii) investigated the combined effects of UF feedwater characteristics on MFI40. Biopolymers and turbidity played a dominant and secondary positive role in the MFI40, respectively. The effect of conductivity on MFI40 changed from positive to negative with a turbidity increase. It also (iii) validated the MFI40 in both laboratory- and pilot-scale UF membrane units, and UF fouling rates were linearly correlated to the MFI40 of their feeds, and (iv) explored the practical use of the MFI40. It was applied to determine the maximum allowable UF feedwater quality (MFI40max), which could be used to select an appropriate pre-treatment process. A fouling predicting model was established based on the feedwater MFI40 and the operating flux, with an average predicting error of 26.8%.

Key words | fouling index, membrane fouling prediction, MFI40, secondary effluents, ultrafiltration

INTRODUCTION

In order to reuse wastewater, ultrafiltration (UF), as one of the low-pressure membrane processes, is widely used to pre-treat secondary effluent (SEF) to improve the feedwater quality of the subsequent reverse osmosis (RO) membrane process. The UF filtration process is a reliable and cost-effective technology due to its high and guaranteed removal of almost all suspended particles and large dissolved organic matter. Compared with conventional pre-treatment processes for RO, UF membrane technology offers the advantages of a water product with higher quality, a smaller footprint and relatively lower cost (Mulder 1996). However, membrane fouling is a major obstacle for the UF membrane process because it would decrease water productivity, increase operational energy consumption and increase maintenance costs (Huang et al. 2008; Gao et al. 2011). To provide early diagnosis of fouling, fouling indices and fouling prediction models for predicting feedwater fouling propensity and simulating filtration process have been developed (Kim & DiGiano 2009; Peiris et al. 2011). A reliable fouling prediction could help in determining the maximum allowable feedwater quality, selection of pre-treatment process, optimal operational parameters, fouling control strategies and membrane cleaning/replacement cost (Mao et al. 2015; Teychene et al. 2018).

Traditional fouling indices such as silt density index (SDI), modified fouling index (MFI), MFI-UF, MFI-NF and cross-flow sampler modified fouling index ultrafiltration (CFS-MFIUF), have been widely applied to measure water...
with low fouling potential, specifically RO feedwater (Schippers & Verdouw 1980; Schippers et al. 1985; Boerlage 2001, Boerlage et al. 2004; Khirani et al. 2006; Sim et al. 2011; Rachman et al. 2013; Jin et al. 2017). These fouling indices, with standardized methods, monitored the flux variation under constant high pressure (normally 30 psi ≈ 2.069 bar) in a 0.45-μm membrane. However, they failed when measuring UF feedwater samples with high fouling potential, because filter paper experienced heavy clogging by foulants in these water samples (Roorda & van der Graaf 2005). Recently, specific ultrafiltration resistance (SUR), unified fouling index, and total fouling index (TFI)/hydraulically irreversible fouling index (HIFI) were developed for evaluating the fouling potential of UF feedwater (Roorda & van der Graaf 2005; Huang et al. 2008, Janssen et al. 2008). These tests operated under low pressure (0.5 bar ≈ 7.25 psi). Their filter paper, or membrane module, and set-ups have not been standardized. The difference in fabrication of testing apparatus may lead to discrepancies in their results. Besides, their applications in a full-scale UF unit are still limited (Nguyen et al. 2011). Therefore, there is a need to develop a simple, quick, standardized and reliable fouling predicting index to evaluate the fouling propensity of UF feedwater.

In order to establish such a fouling index, modification to traditional fouling tests could be explored. During the traditional fouling indices tests, applied pressure provides the driving force to overcome heavy clogging of foulants. In this case, higher applied pressure could cause higher flux and bring higher fouling load to the filter paper (Alhadidi et al. 2011). Few studies have been conducted to investigate the impact of operating pressure on the sensitivity and repeatability of the fouling index test. Hence, it is interesting to investigate the feasibility of establishing a fouling index for UF feedwater by increasing the applied pressure of traditional fouling indices.

Furthermore, assessment and prediction of UF fouling performance by fouling index is difficult due to the complexity of feedwater (SEFs) in both quantity and quality (Mosset et al. 2008; Ayache et al. 2013; Teychene et al. 2018). UF membrane fouling has been reported to be related to the amount and characteristics of potential foulants as well as the interactions between these foulants (Park et al. 2006; Haberkamp et al. 2008; Zheng et al. 2009; Chen et al. 2014; Wang et al. 2017). In previous studies, the effect of foulants on the fouling index has been studied individually. Park et al. (2006) found that MFI significantly increased with increasing particle concentration in a linear relationship, and Chuang et al. (2009) reported that MFI of larger molecular organics was higher than that of smaller ones. Sim et al. (2010) reported that CFIS-MFIUF increased with increasing humic acid concentration, and Ayache et al. (2013) found that TFI were largely impacted by biopolymers and humic substances. Besides, it was found that pH and salt concentration influenced the fouling indices (Kabsch-Korbutowicz et al. 1999; Mousa & Al-Hitmi 2007). These studies have given some insights into the effect of critical water quality parameters on fouling indices; however, the quantitative information of this effect is still limited. Additionally, the combined effect of these parameters on the fouling index needs to be investigated because the combined effect significantly influences the fouling index value (Jermann et al. 2008; Teychene et al. 2018). However, this has been seldom reported and thus is poorly understood. Therefore, there is a need to quantify the individual and combined effect of major foulants on the fouling index.

Based on the research needs mentioned above, the objectives of this study are to: (i) raise the operating pressures of existing MFI tests to improve their stability and establish a modified fouling index; (ii) investigate the effects of UF feedwater characteristics on the established fouling index; (iii) validate the built fouling index in laboratory- and pilot-scale UF membrane units; and (iv) explore the practical use of the index. This study may provide insights into defining a systematic approach to predict fouling propensity of SEFs as UF feedwater.

MATERIAL AND METHODS

Fouling index test

The fouling index method was developed to estimate the SEF fouling propensity, based on the MFI method (Schippers & Verdouw 1980) operated at a constant pressure of 30 psi (MFI30). A water sample was fed to a 0.45-μm pore-sized membrane filter with a diameter of 47 mm (GE Osmonics, USA) at a constant pressure of 40 psi, and the MFI40 was obtained. Permeate was collected and weighed over
the test every 30 seconds. Based on the cake filtration theory, a curve of Filtration time as a function of Permeate volume was plotted and the MFI value was calculated according to Equation (1):

\[ \frac{t}{V} = \frac{\eta R_m}{\Delta P A} + \frac{\eta I}{2 \Delta P A^2} V \]

with

\[ MFI = \frac{\eta I}{2 \Delta P A^2}, \quad I = \alpha W \] (1)

where \( V \) = permeate volume produced (m\(^3\)); \( A \) = membrane area (m\(^2\)); \( \Delta P \) = trans-membrane pressure (Pa); \( R_m \) = the membrane resistance (m\(^{-1}\)); \( R_c \) = cake resistance (m\(^{-1}\)); \( \eta \) = dynamic viscosity of the water (N s m\(^{-1}\)); \( I \) = cake resistivity (m\(^{-2}\)); \( \alpha \) = specific cake resistance (m/kg); \( W \) = concentration of particles (kg/m\(^3\)).

For the compressible cake layer, \( \alpha \) changes with pressure as follows (Almy & Lewis 1912):

\[ \alpha = \alpha_0 \Delta P^\omega \] (2)

where \( \omega \) is the compressibility factor of the cake and \( \alpha_0 \) is a constant. For incompressible cakes, \( \omega \) is zero and \( \alpha \) is a constant. The larger the \( \omega \), the more compressible the cake layer is.

**Feedwater**

Various SEF samples were collected at different times from three domestic wastewater treatment plants (WWTPs) in Singapore and one industrial WWTP in Shanghai. These WWTPs employed a conventional activated sludge process for secondary treatment. Their SEFs were sequentially fed to a UF membrane system, which is described below under ‘UF membrane units’.

Meanwhile, makeup SEFs were used as the feedwater for a few UF filtration runs in order to control the foulants concentration at a pre-determined level. Makeup SEFs were prepared according to the following protocol. SEFs collected from WWTPs were firstly filtered by a microfiltration membrane (pore size of 0.45 \( \mu \)m, GE Osmonics, USA). The rejected solution was then stocked as the particle concentrates, while the permeate was sent to a cation exchange resin (Dowex Monosphere 650C UPW(H), DOW, USA) column to remove the cations and was further concentrated by RO membrane (SWC 1-4040, Hydranautics & Nitto Denko Pte. Ltd, USA). Various test solutions were made up by mixing the particle concentrates, RO concentrates, and CaCl\(_2\) solution in different ratios.

All water samples were fractionalized into particles, dissolved organic matters and mineral contents. The dissolved organic matters were further separated into biopolymers, humics and low molecular weight (LMW) organics (sum of building blocks and LMW neutrals), and quantified by liquid chromatography–organic carbon detection (LC–OCD) (DOC-LABOR Dr. Huber, Germany). Turbidity was measured by a turbidity meter (Hach 2100N, USA). Dissolved organic carbon (DOC) was measured by a total organic carbon (TOC) analyzer (Model 1010, O.I. Analytical, Shimadzu TOC VCSH, Japan) and water samples were filtered through a membrane with a pore size of 0.45 \( \mu \)m ( Pall, USA) prior to TOC analysis. Ionic concentration was measured by ion chromatography (Diomex LC20 Chromatography, Dionex Corporation, USA) and conductivity was measured by a conductivity meter (HACH, USA). The characteristics of the tested domestic and industrial SEFs are summarized in Table 1.

**UF membrane units**

**Laboratory-scale UF unit**

SEFs were fed to a hollow fiber UF membrane system (Figure 1) in dead-end filtration mode at a flux of either 35

**Table 1 | Characteristics of domestic and industrial effluents**

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Unit</th>
<th>Domestic SEFs</th>
<th>Industrial SEFs</th>
</tr>
</thead>
<tbody>
<tr>
<td>Turbidity</td>
<td>NTU</td>
<td>2.5 ± 1.9</td>
<td>11.8 ± 9.8</td>
</tr>
<tr>
<td>DOC</td>
<td>mg/L</td>
<td>7.3 ± 1.7</td>
<td>31.2 ± 4.9</td>
</tr>
<tr>
<td>Biopolymers (measured as DOC)</td>
<td>mg/L</td>
<td>0.91 ± 0.5</td>
<td>1.95 ± 0.9</td>
</tr>
<tr>
<td>Humics (measured as DOC)</td>
<td>mg/L</td>
<td>1.78 ± 0.5</td>
<td>3.26 ± 1.2</td>
</tr>
<tr>
<td>LMW organics (measured as DOC)</td>
<td>mg/L</td>
<td>3.11 ± 1.1</td>
<td>9.9 ± 2.6</td>
</tr>
<tr>
<td>Ca(^{2+})</td>
<td>mg/L</td>
<td>21 ± 12.2</td>
<td>39 ± 2.7</td>
</tr>
<tr>
<td>Mg(^{2+})</td>
<td>mg/L</td>
<td>1.4 ± 0.3</td>
<td>14 ± 3.5</td>
</tr>
<tr>
<td>Conductivity</td>
<td>ms/m</td>
<td>51.4 ± 7.8</td>
<td>1487.9 ± 407.8</td>
</tr>
</tbody>
</table>
or 40 LMH. The UF system was operated by filtration cycles of 10 min on, 30 s off, 30 s backwash and another 30 s off. Meanwhile, aérations for scouring the membrane surface were performed at the initial and final 30 s of filtration period, as well as during 30 s of backwash. The hollow fiber UF membrane module used was the laboratory-scale GE ZeeWeed-1 (GE Water & Process Technologies, Europe), and its membrane characteristics are summarized in Table 2. During each filtration run, the trans-membrane pressure (TMP) values were measured and recorded every 30 s by a pressure transducer (Shanghai Ke Qi Automation Co., Ltd, Shanghai, China) and a data logger, respectively.

![Schematic diagram of the submerged UF membrane setup.](image)

Table 2 | Properties of ZeeWeed-1 membrane module

<table>
<thead>
<tr>
<th>ZeeWeed – 1 module properties</th>
<th>Polyvinylidene fluoride (PVDF)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Membrane material</td>
<td>0.036</td>
</tr>
<tr>
<td>Membrane pore size (μm)</td>
<td>0.036</td>
</tr>
<tr>
<td>Module type</td>
<td>Outside/in hollow fiber</td>
</tr>
<tr>
<td>Membrane surface area (m²)</td>
<td>0.046</td>
</tr>
<tr>
<td>Outer/inner diameter (mm)</td>
<td>1.9/0.8</td>
</tr>
<tr>
<td>Maximum operating pressure (kPa)</td>
<td>62</td>
</tr>
<tr>
<td>Operating pH range</td>
<td>5–9</td>
</tr>
</tbody>
</table>

**Pilot-scale UF unit**

A UF pilot unit with hollow-fibre membrane modules (ZeeWeed-500d, GE Water & Process Technologies, USA) was used to filter industrial SEFs in dead-end mode at a flux of either 35 or 40 LMH. The filtration cycle and membrane material were identical to that of the laboratory-scale ZeeWeed-1 unit.

The fouling performance of these two systems was assessed by fouling rate, which was calculated as the slope of TMP increase with time over the filtration process. As suggested by Doyen et al. (1998), the filtration and fouling phenomena that occur during this short time interval would influence the long-term fouling. The fouling rate was then correlated to feed the fouling index measured at the same time.

**Establishment of fouling predicting approach by MFI40 and operating flux**

To establish a fouling predicting model based on the feedwater MFI40 and flux, a model-building dataset of 13 filtration runs fed by various makeup feedwaters at different fluxes was designed according to the Box–Behnken design. This design, which is a modified fractional factorial design,
has been widely used to screen the maximum number of factors with the least number of experiments (Box et al. 1978). In model establishment, feedwater MFI40 and operating flux of filtration runs were measured and then correlated to the fouling rate acceleration \( \frac{\Delta \text{Fouling Rate}}{\Delta T} \) by utilizing the method of quadratic response surface regression. Furthermore, five additional filtration runs fed by SEFs samples collected from WWTPs were conducted and their results were used to verify the established statistical correlation.

RESULTS AND DISCUSSION

The feasibility of increasing applied pressure of MFI40 test

The determination of MFI40 value

The plots of \( t/V \) versus \( V \) of MFI tests on one SEF collected from a domestic WWTP in Singapore under 30 and 40 psi are shown in Figure 2. Similar trends of \( t/V \) increasing with \( V \) were observed for MFI tests carried out under these two different operating pressures. During the first 5 min, \( t/V \) increased gently with \( V \). Subsequently, until the end of the test (15 min), \( t/V \) increased linearly with \( V \) at a much more rapid and stable rate. The MFI value was then defined as the slope of this linear curve of \( t/V \) versus \( V \) after the initial 5 min, on the basis of the cake filtration theory. According to Boerlage et al. (2005), the filtration process of the MFI test is initially the occurrence of pore blocking (the first 5 min), followed by cake filtration with compression of the cake layer (the next 10 min). Similar filtration curves observed at 30 and 40 psi suggested the occurrence of similar filtration mechanisms at these two pressures.

Subsequently, 10 MFI tests for another one SEF collected from domestic WWTP in Singapore were repeated under both operating pressures of 30 and 40 psi. Figure 3 shows that the MFI30 values at 30 psi ranged from 9,747 to 20,568 s/L², with an average value of 14,769 s/L²; while MFI40 values at 40 psi ranged from 10,019 to 13,121 s/L², with an average value of 11,716 s/L².

The average MFI40 value was lower than MFI30. A low fouling index at higher operating pressure was also observed by Sim et al. (2011), where the explanation of cake compression effect was provided. By combining Equations (1) and (2), the MFI was reformatted and represented as follows:

\[
\text{MFI} = \frac{\eta \alpha W}{2A^2} \Delta P^{n-1}
\]

Equation (3) shows that the MFI values would decrease with a \( \Delta P \) increase when the compressibility \( \omega \) is between 0 and 1, depending on how much the cake layer could be compressed. The compressibility of the cake layer is influenced by the characteristics of the components in the water sample. The relevant components in SEFs mainly included particles and colloids with multi-sizes that can be easily deformed under high pressure. Medium compressibility with a \( \omega \) value of 0.6 was proposed for the foulants in SEFs by Roorda & van der Graaf (2005). Thus, MFI values obtained at different pressures need to be corrected to the same pressure for comparison purposes. In this study, the average MFI40 (11,761 s/L²) was corrected to be 13,145 s/L² at 30 psi with \( \omega = 0.6 \). This value was closer to the tested MFI30 (14,769 s/L²) with a slight difference of 10%. The above results indicated that the MFI40 and
MFI\textsubscript{30} values were rather similar when corrected to the same pressure.

**Improvement of repeatability of fouling index test**

Figure 3 also shows that the standard deviation of MFI\textsubscript{40} (1,156 s/L\textsuperscript{2}) was lower than that of the MFI\textsubscript{30} (3,676 s/L\textsuperscript{2}) by 68.6\% over the 10 repeating tests, which indicates that MFI\textsubscript{40} were more reproducible and stable. Alhadidi et al. (2011) suggested that during the fouling index test, dominated by the cake formation process, a more stable filtration process may occur due to a more stable cake layer. Under higher pressure, a more stable structure of cake was formed when more foulants were brought to the surface of the filter paper by a larger driving force, leading to the formation of a denser cake layer and an increase in the specific cake resistance (Sim et al. 2011). By combining Equations (1) and (2), the cake resistivity could also be expressed as Equation (4):

\[ I = a_0 \Delta P^\omega W \]  

(4)

As previously discussed, \( \omega \) is between 0 and 1 for SEFs in this study. Thus, it could be deduced that \( I \) would increase with operating pressure according to Equation (4).

**Influence of feed water characteristics on MFI\textsubscript{40}**

In order to investigate the effect of feed water characteristics on MFI\textsubscript{40}, eight water quality parameters (turbidity, DOC, biopolymers, humics, LMW organics, calcium, magnesium and conductivity) and MFI\textsubscript{40} values of 42 SEFs samples (including SEFs collected from WWTPs and makeup SEFs) were measured.

**Selection of most important water quality parameters for MFI\textsubscript{40}**

**Independence of fouling-relevant water quality parameters.**

In order to avoid the co-correlation of selected water quality parameters and select the independent parameters, the dependency between these parameters was firstly investigated by calculating the correlation coefficient \( \rho \) for every
pair of parameters, with equation of $\rho = \frac{\text{Cov}(y_1, y_2)}{\sigma_1 \sigma_2}$ (Box et al. 1978). Table 3 shows the result of the correlation analysis for the eight tested parameters. According to statistical explanation for the matrix, two parameters are significantly correlated when the correlation coefficient is larger than 0.6. The larger the coefficient is, the more significantly these two parameters interact. A positive value of the coefficient indicates a positive correlation between two parameters, while a negative value indicates a negative correlation.

For the tested organic parameters, it can be observed from Table 3 that the turbidity and biopolymers correlation coefficient was 0.19, indicating their independence from each other. Thus, turbidity and biopolymers were selected as independent parameters to represent particles and organic foulants. Turbidity/DOC, DOC/humics, and DOC/LMW organics were found to have a high correlation coefficient (>0.6) and correlated with each other. Teychene et al. (2018) also found that a high Pearson’s coefficient $r$, over 0.9, was found for both fluorophores humic- and fulvic-like substances against total organic carbon (TOC) parameters. For the inorganic parameters, correlation coefficients for calcium/turbidity, calcium/biopolymers, magnesium/turbidity, magnesium/biopolymers, conductivity/turbidity and conductivity/biopolymers were found to be lower than 0.6. This indicates that calcium, magnesium, and conductivity were relatively independent on turbidity and biopolymers. Thus, turbidity, biopolymers, calcium, magnesium and conductivity were relatively independent of each other, and were selected to be studied.

### Table 3 | Correlation matrix of the water quality parameters

<table>
<thead>
<tr>
<th></th>
<th>Turbidity</th>
<th>DOC</th>
<th>Bio-polymers</th>
<th>Humics</th>
<th>LMW organsics</th>
<th>Ca$^{2+}$</th>
<th>Mg$^{2+}$</th>
<th>Conductivity</th>
</tr>
</thead>
<tbody>
<tr>
<td>Turbidity</td>
<td>1</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>DOC</td>
<td>0.68</td>
<td>1</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Biopolymers</td>
<td>0.19</td>
<td>0.53</td>
<td>1</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Humics</td>
<td>0.59</td>
<td>0.75</td>
<td>0.03</td>
<td>1</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>LMW organics</td>
<td>0.72</td>
<td>0.98</td>
<td>0.50</td>
<td>0.79</td>
<td>1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ca$^{2+}$</td>
<td>0.58</td>
<td>0.80</td>
<td>-0.05</td>
<td>0.59</td>
<td>0.49</td>
<td>1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Mg$^{2+}$</td>
<td>0.21</td>
<td>0.72</td>
<td>-0.29</td>
<td>0.90</td>
<td>0.80</td>
<td>0.51</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>Conductivity</td>
<td>0.22</td>
<td>0.37</td>
<td>0.15</td>
<td>-0.04</td>
<td>0.25</td>
<td>0.20</td>
<td>-0.133</td>
<td>1</td>
</tr>
</tbody>
</table>

**Importance of the independent water quality parameters.** The significance of these five independent parameters was compared to determine the most important ones relevant to MFI$_{40}$. This was performed by utilizing a multi-level factorial analysis, which was used to analyze the impact of multi-level factors on response by Ng & Ng (2010). The importance of these five independent water quality parameters is shown in Figure 4. The parameter with larger importance is supposed to have a more dominant impact on the response (MFI$_{40}$).

The obvious effects of particles and organics on the fouling index have been widely reported by previous studies (Park et al. 2006; Huang et al. 2009; Sim et al. 2010; Ayache et al. 2015). MFI$_{40}$ was observed to increase with increasing particle and organics concentrations. Clearly, the increasing effect of particles on MFI$_{40}$ could be explained by the fact that a cake layer was formed on the filter paper when all particles were retained (Bourgeois et al. 2003). The significant effect of biopolymers observed on MFI$_{40}$ indicated that the biopolymers, rather than total organics, would play a principal role in MFI$_{40}$. It was because biopolymers, rather than overall organic matter, could be largely retained either by the filter paper or the formed cake layer in the fouling index test (Zheng et al. 2010, 2017; Ayache et al. 2013; Wang et al. 2017). In this case, increased particles and biopolymers concentrations would lead to an increased foulants loading, thus a higher fouling index.

The observed significant effect of conductivity on MFI$_{40}$ values has occasionally been reported. Boerlage et al. (2003) found that the MFI value increased with salinity, but it started to drop at NaCl concentrations of 0.1 M. Similarly,
Sim et al. (2010) reported their fouling index value increased as the NaCl concentration was increased, but it also started to decrease when the NaCl concentration was 0.2 M. When the NaCl concentration exceeded 0.43 M, any increase in NaCl concentration had no further effect on the fouling index. This phenomenon could be explained by the combined effects of electrostatic double layer compression and particle aggregation (Nikkola et al. 2014; Abdelrasoul et al. 2017). On the one hand, the compression of the electrostatic double layer that occurs at high conductivity would result in tighter bonding between the particles and cake layer, which consequently increases the fouling index value. On the other hand, particle aggregation occurring at high conductivity would lead to a more permeable cake layer formed by larger particles and, consequently, a decrease in the fouling index value. In this study, the importance of conductivity on MFI_{40} was found to be even more significant than turbidity and biopolymers, which may be explained by the wide range of conductivity tested (from 49 to 2,782 ms/m). The high conductivity of industrial SEFs is around 20 times higher than domestic SEFs in most studies. This wide range would result in a significant change in the status of the electrostatic double layer and particle size, and hence a drastic difference in the MFI_{40} value. In this special case, the effect of conductivity on MFI_{40} could overwhelm the effects of turbidity and organics.

The influences of calcium and magnesium on the MFI_{40} are observed to be minor in this study. The effects of these ions have been seldom reported by previous studies, possibly due to the weak interaction between divalent ions and the membrane/cake layer compared with the strong drag force on calcium. Another explanation is that the effects of divalent ions on MFI_{40} did not manifest due to the narrow testing ranges of calcium and magnesium concentration, specifically from 12.5 to 99.7 mg/L and from 0.7 to 14.0 mg/L, respectively. These ranges were chosen based on the quality of SEFs collected from practical treatment processes; hence, the obtained results were likely to represent their effects on the MFI_{40} in practical application.

**Combined effects of most important water quality parameters on MFI_{40}**

The combined effects of the three most important parameters, namely turbidity, biopolymers and conductivity, on MFI_{40} values were investigated using the method of response quadratic surface regression. This regression method was used because it could deal with the two-order main effects and two-way interaction effects of the variables (Hill & Lewicki 2007). Based on the 42 datasets of various SEFs collected, three quadratic response surface regression equations illustrated in Equations (5)–(7) were generated.
to interpret the combined effects of turbidity/DOC, turbidity/chloride and DOC/chloride on MFI$_{40}$, respectively. R$^2$ values of these regressions were close to 0.6, which indicated the reliability of these regressions. They were further visualized by three-dimensional surface figures as shown in Figures 5–7.

$$MFI_{40} = 8.0(T)^2 - 17816.6(B)^2 + 255.4(T)(B) - 642.9(T) + 45232.6(B) - 4676.2$$  \hspace{1cm} (5)

\[n = 18, \quad R^2 = 0.54\]

$$MFI_{40} = -72.5(T)^2 - 0.01(C)^2 + 0.4(T)(C) - 210.8(T) + 32.8(C) + 11285.2$$  \hspace{1cm} (6)

\[n = 23, \quad R^2 = 0.53\]

$$MFI_{40} = -17276.1(B)^2 - 0.003(C)^2 + 8.3(B)(C) + 38613.0(B) + 6.6(C) - 4881.4$$  \hspace{1cm} (7)

\[n = 23, \quad R^2 = 0.59\]

where $T$, $B$, and $C$ stand for turbidity, biopolymers and conductivity, respectively.

Figure 5 visualizes Equation (5) and illustrates both the single and mutual effects of turbidity and biopolymers on the MFI$_{40}$. It was also found that MFI$_{40}$ significantly increased with the increase of biopolymers and slightly increased with increasing turbidity when the biopolymers concentration was high. It suggests that biopolymers had a predominant effect in MFI$_{40}$ increase, while turbidity had a secondary increasing effect on the MFI$_{40}$. It also gave further support to the results of the variable importance analysis (see above under ‘Importance of the independent water quality parameters’), which reveals that biopolymers affected MFI$_{40}$ more significantly than turbidity. Moreover, it also suggests that the effects of biopolymers and turbidity on MFI$_{40}$ increase were synergized when the concentrations of biopolymers and turbidity were higher, which could be explained by the interaction between particles and biopolymers. When particles and biopolymers increased, a heterogeneously denser fouling layer was formed by the particles and biopolymers.
Figure 6 | Relationship between turbidity, conductivity and MFI_{40}.

Figure 7 | Relationship between biopolymers, conductivity and MFI_{40}.
particles and biopolymers. Such a dense cake layer could reject extra particles and organics that could not be rejected by the filter paper alone, which leads to an increase in the fouling index. Such a mutual effect was also reported by Jermann et al. (2008), who observed that particle fouling was aggravated by the presence of organics. Sun et al. (2016) used humic acid (HA), bovine serum albumin (BSA), and kaolin clay to synthesize natural water, and found that BSA-kaolinite exhibits more substantial irreversible fouling than that of HA-kaolinite and HA-BSA-kaolinite.

Figure 6 depicts Equation (6) and illustrates the combined effects of turbidity and conductivity on the MFI40. Increasing turbidity obviously increased the MFI40; however, the conductivity effect on the MFI40 was impacted by the turbidity. MFI40 increased with increasing conductivity when the turbidity was low and it decreased with increasing conductivity when the turbidity was high. These observations reinforced the effect of turbidity on MFI40 increase, which agrees with the result of Figure 5. The opposite effects of conductivity on MFI40 could be explained by the combination effects of the compression of the electrostatic double layer aggravating fouling (She et al. 2009) and the aggregation of particles mitigating fouling (Shon et al. 2004; Abdelrasoul et al. 2017). When the turbidity was low, the particle amount was so small that its size effect on the MFI40 was insignificant; thus, the interaction between the foulants and membrane filter dominated the fouling process. In this case, the effect of compressing the electrostatic double layer might overwhelm the effect of aggregating particles, which led to a positive response of MFI40 to conductivity. In contrast, the negative response of MFI40 to conductivity could be caused by the predominant effect of particle aggregation when the turbidity was high. In this case, size screening dominated the filtration process, and therefore the particle and colloidal status influenced the MFI40 more significantly than interactions between the foulants and the filter paper. It leads to the effect of particle aggregation subduing the effect of electrostatic double layer compression. Abdelrasoul et al. (2017) also found that aggregation of particles mitigated membrane fouling due to reduction of pore blocking and irreversible fouling.

The single and mutual effects of biopolymers and conductivity on the MFI40 illustrated by Equation (7) are depicted in Figure 7. It is observed that the MFI40 generally increased with increasing biopolymers concentration and conductivity. However, when the biopolymers concentration was low, the MFI40 remained constant despite the variation of conductivity. This could be as a result of the weakened influence of conductivity on filtration and fouling behaviour because the particles and organics were scattered and the electrostatic force was weaker in the solution at low biopolymers concentration. However, when the biopolymers concentration was increased, the effect of electrostatic force on the MFI40 started to manifest. Higher conductivity would lead to tighter bonding between foulants and the membrane filter, resulting in higher MFI40. This explanation is partly supported by Jermann et al. (2008), who pointed out that the interaction between organics and membrane largely influenced the fouling propensity of the solution when the particle effect was excluded.

**Correlation of MFI40 to UF fouling rate**

Overall, fouling rates caused by 50 various SEFs samples (including SEFs collected from WWTPs and makeup SEFs) at two fluxes (i.e. 35 and 40 LMH) in both laboratory-scale and pilot-scale UF units were correlated to their MFI40 as shown in Figures 8 and 9.

It can be seen that feedwater with higher MFI40 caused a faster fouling rate in an obvious linear relationship with an average R2 value of 0.75. Besides, the gradient of fouling rate versus MFI40 curve in the laboratory-scale system was steeper than that in the pilot-scale system, suggesting that the impact of MFI40 variation on fouling rate was less significant in the pilot-scale system due to the scale-up effect, which was also found by Ayache et al. (2013). It indicates that patterns of correlation between the fouling rate and the MFI40 were the same for systems with different scales, whereas the coefficients values in the correlation changed with system scale. Similar noticeable correlations between flux decline rate and fouling indices were reported by Choi et al. (2009) and Yu et al. (2010). However, these studies were not conducted under typical conditions that were used in full-scale membrane systems. It may undermine the reliability of the established correlation to assess membrane fouling behaviour. In this study, the system using a commercialized hollow fiber UF module was operated under typical...
operating conditions of a full-scale unit, making the obtained correlation more practically applicable.

It is also interesting to note that the gradient of the fouling rate versus the MFI40 curve was steeper at higher flux, indicating that a rise in MFI40 caused a more obvious increase in fouling rate. It reveals that the positive response of MFI40 to fouling rate was more significant under higher flux. This phenomenon was explained by the increased resistance of the compressed fouling layer and subsequently amplified TMP increase caused by the same feedwater (Roorda & van der Graaf 2005).

**Practical use of MFI40**

**Determining maximum allowable UF feedwater quality**

One possible application of the MFI40 is to determine the maximum allowable UF feedwater quality by calculating MFI40max. In order to calculate MFI40max, the following information was collected first: (i) highest TMP limit for the membrane (TMPfoul) recommended by membrane supplier; (ii) initial TMP of UF membrane process (TMPinitial); (iii) designed filtration duration between two cleanings (T); and (iv) the correlation equation of fouling rate versus feedwater MFI40 obtained from filtration runs, which could be written as Fouling rate = a × MFI40 + b, as shown in Figures 8 and 9. MFI40max value was then calculated by substituting TMPfoul, TMPinitial, and T into the correlation equation as follows:

\[
MFI_{40}\text{max} = \frac{(\text{TMP}_{\text{foul}} - \text{TMP}_{\text{initial}})}{a \times T} - \frac{b}{a}
\]  

where a and b are the coefficients in the linear correlation between the fouling rate and MFI40max.

Using the above approach, MFI40max values for laboratory-scale (ZW1) and pilot-scale (ZW500d) systems were calculated and are shown in Table 4. It was found that MFI40max values for the both systems at lower flux...
were higher than those at higher flux, indicating that the UF system operated at lower flux could accept a feedwater with a higher fouling potential. In addition, MFI\textsubscript{40}\text{max} value for the pilot-scale system were close to that for the laboratory-scale system at similar flux, with a slight difference of 3–5%. This suggests that MFI\textsubscript{40}\text{max} value obtained from the small-scale system could be used to predict that for the large-scale system under similar operating conditions.

**Predicting and monitoring fouling development by MFI\textsubscript{40} and flux**

The predicting model involving the fouling rate acceleration, MFI\textsubscript{40} and flux was built on the dataset of 13 different makeup SEFs filtration runs at different fluxes. The established correlation is shown in the following equation:

\[
\frac{\Delta \text{ Fouling Rate}}{\Delta T} = 1.42 \left( \frac{\text{MFI}_{40}}{10000} \right)^2 + 1.14 \left( \frac{\text{flux}}{10} \right)^2 \\
+ (3.72E - 5) \times (\text{MFI}_{40} \times \text{flux}) - 0.002 \times (\text{MFI}_{40}) \\
- 1.56 \times (\text{flux}) + 51.72
\]  

\( n = 13, R^2 = 0.95, F = 37.6, F_{\text{significant}} = 2.44 \times 10^{-5} \)

Analysis of variance (ANOVA) for this regression reveals that \( R^2 \) value was as high as 0.95 and the \( F \) value
of 37.6 was larger than $F_{\text{significance}}$ value of $2.44 \times 10^{-5}$, which implies that this regression was reliable. It was also found that the $P$ value for flux (equal to 0.003) was smaller than that for the MFI$_{40}$ (equal to 0.014), suggesting that the flux effect on fouling development was more significant than the MFI$_{40}$.

Furthermore, five additional filtration runs fed by makeup SEF samples with different characteristics to the above model, establishing 13 makeup SEFs, were conducted and their results were used to create a validation dataset to verify the established statistical correlation. The predicting capability of this model was investigated by comparing the predicted and measured fouling behaviour of the validation dataset. A plot of measured versus predicted fouling rate acceleration is demonstrated in Figure 10.

It can be seen that the average error between the measured and predicted one for these five filtration runs was 26.8%, suggesting that the established correlation can be used to approximately predict fouling rate evolution by flux and feedwater MFI$_{40}$. This predicting methodology differs from those mathematically complex and computationally expensive semi-empirical models that required a very detailed knowledge of the fouling layer and were applied to ideal operating conditions defined by model assumption, which were not readily obtainable for a large-scale UF system (Al-Zoubi et al. 2007; Kim & DiGiano 2009). In short, the established predicting model offers the advantage of easy adoption and a small prediction error.

Therefore, it can be a reliable tool to predict fouling development in UF membrane systems.

**CONCLUSIONS**

In this study, the MFI$_{40}$ of SEFs with high fouling potential was measured by raising the operating pressure of existing fouling index tests from 30 to 40 psi. The standard deviation of 10 repeating MFI$_{40}$ was lower than MFI$_{30}$ by 68.6%, indicating better stability and repeatability of the MFI$_{40}$. It is explained that a denser and more stable cake layer was formed at a higher operating pressure, with higher cake resistivity.

Turbidity, biopolymers and conductivity have been found to significantly co-affect the MFI$_{40}$ value. Biopolymers concentration played a dominantly positive role in the MFI$_{40}$ and turbidity had a secondarily positive impact on the MFI$_{40}$. Their positive effects were amplified when biopolymers concentration and turbidity were high due to the formation of a heterogeneous denser fouling layer. The effect of conductivity on MFI$_{40}$ changed from positive to negative with turbidity increase, probably because the dominant influencing process in the MFI$_{40}$ test transferred from electrostatic double layer compression to particle aggregation. Additionally, the positive feedback of MFI$_{40}$ to conductivity manifested only when biopolymers concentrations were high, which can be explained by the effect of reducing electrostatic force on fouling development, excluding particle influence, that started to display and become dominant when the biopolymers concentration was increased.

Validation of the MFI$_{40}$ was performed by correlating the feedwater MFI$_{40}$ and fouling rate of laboratory-scale and pilot-scale UF membrane systems using a commercialized UF module operated at typical conditions of a full-scale system. A linear correlation between the MFI$_{40}$ and the overall fouling rate was found, indicating the reliability of using the MFI$_{40}$ for interpreting the SEFs fouling potential.

A possible application of the MFI$_{40}$ was to determine the maximum allowable UF feedwater quality (MFI$_{40,\text{max}}$), which could be used to evaluate UF pre-treatment efficiency and to select an appropriate pre-treatment process. Additionally, a regression fouling predicting model was...
established based on the feedwater MFI$_{40}$ and the operating flux, with an average predicting error of 26.8%. Such an easily adopted model could be a promising fouling predicting approach for practical design and operation.

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