The Effects of Soluble Organic Matters on Membrane Fouling

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Abstract

This study investigated the effects of soluble organic matters on membrane fouling characteristics, using silt density index (SDI) and modified fouling index (MFI) to evaluate the fouling potential. Experimental results demonstrated that humic acid had significant effects on membrane fouling indexes. When its concentration was in the range of 0.01-0.05 mg/L, the SDI$_{15}$ and MFI were 2.9-3.9 and 5.4-13.8 s/L$^2$, respectively. According to the linear equations of MFI measurements, the fouling potential was in the order of humic acid $>$ nucleic acid $\approx$ protein $>$ glucose. Moreover, the molecular weight of dextran played an important role in membrane fouling indexes. Furthermore, a mathematical analysis of filtration experiments based on saturation curve was developed in this study. The maximum accumulated filtrate ($V_{\text{max}}$) and the constant of filtration ($k_f$) could be obtained to improve the precision of membrane fouling prediction.

Keywords: Fouling, Membrane, Modified Fouling Index, Silt Density Index, Soluble Organic Matters.

1. INTRODUCTION

The reclamation and reuse of water and wastewater is becoming an essential and urgent issue in water shortage areas, especially in the developing countries, which require a great amount of high-grade water for industrial cleaning process. Membrane units predominantly act as the core to produce high-grade water for industries. However, membrane fouling is always a serious problem when the secondary effluent of wastewater treatment plants was employed for reclamation and reuse (Jarutthirak et al., 2002; Sadr Ghayeni et al., 1998; Shon et al., 2004).
There are many complex soluble organic compounds in secondary effluent. Both the characteristics of wastewater and the operations of biological processes will bring about dissimilar compositions of soluble organic compounds in the effluent, such as polysaccharide, protein, nucleic acid and humic acid etc. (Barker and Stuckey, 1999; Boero et al., 1996; Liang et al., 2007). These soluble organic matters will possibly bring organic fouling and reduce the life span of membrane.

Since 1980, researchers focus on the development of index of fouling in order to predict, prevent and reduce the membrane fouling problem. The silt density index (SDI), modified fouling index (MFI) and mini plugging factor index (MPFI) are proposed sequentially (ASTM, 1995; Kremen and Tanner, 1998; Schippers and Verdouw, 1980). These indexes are useful for evaluating the suspended solid and colloid fouling for the operations of membranes (Vrouwenvelder et al., 2003; Khirani et al., 2006; Yiantsios and Karabelas, 2002). However, studies dealing with the effects of soluble organic on membrane fouling are limited in the literature (Jucker and Clark, 1994; Park et al., 2006). The characteristics of fouling potential of soluble organic compounds need more comprehensive studies. Therefore, various soluble organic compounds with different chemical structure are selected for comparing their effects on membrane fouling potential in this study. Moreover, a mathematical analysis of filtration experiments based on saturation curve is developed for improving the precision of membrane fouling prediction.

2. MATERIALS AND METHODS

2.1 Silt Density Index measurement
The measurement of the Silt Density Index (SDI) conformed to the ASTM D4189 standards with a time interval of 15 and 30 min (ASTM, 1995). The main equipment included an air compressor, pressure tank, membrane filter holder, valves, and graduated cylinder. The pressure was automatically controlled at 30 psi (207 kPa) in the pressure tank to minimize experimental error. A filter with 47 mm diameter and average pore size of 0.45-μm provided by CRITICAL was used for the test. The filtrate was collected and recorded continuously over a period of 30 minutes for the calculations of SDI_{15}, SDI_{30}, and MFI (ASTM, 1995; Schippers and Verdouw, 1980). Experiments were repeated to ensure the reproducibility.

2.2 Soluble organic matters

Various soluble organic compounds with different chemical structure were selected for comparing their effects on membrane fouling potential in this study. These soluble organic compounds can present different component of pollutants in wastewater. The target soluble organic matters included glucose provided by TEDIA, humic acid provided by Fluka, nucleic acid provided by ACROS, protein provided by Protose Be, and dextran with varying molecular weight provided by Sigma. The glucose used in this study is for comparing with other different chemical structure pollutants. The solutions were freshly prepared to adequate concentration with deionized water. Moreover, the concentration range of various soluble organic matters is depended on the SDI experimental limitation and its characteristics.

2.3 Saturation curve analysis method

A mathematical analysis based on the saturation curve was established to interpret the data of the SDI filtration experiment. Equation [1] shows the pattern of the saturation curve used to identify the fouling behavior in this study:
Equation [2] shows the linear form of the aforementioned saturation curve. The slope and intersection of the inverse filtrate \((1/V)\) versus inverse time \((1/t)\) curve were used to calculate the maximum accumulated filtrate \((V_{\text{max}})\) and the constant of filtration \((k_f)\) for comparing the degree of fouling potential of soluble organic matters. According to the saturated curve model, the higher values of \(V_{\text{max}}\) and \(k_f\) exhibit lower fouling potential of soluble organic matters.

\[
\frac{1}{V} = \frac{1}{V_{\text{max}}} + \frac{k_f}{V_{\text{max}}} \left( \frac{1}{t} \right)
\]  

[2]

3. RESULTS AND DISCUSSION

3.1 Effects of soluble organic matters on SDI and MFI

The SDI experiments of glucose, nucleic acid, protein, and humic acid were carried out repeatedly under various feed concentrations for comparing their fouling potential. SDI experimental data of glucose showed that the SDI value increased linearly with the concentration. The SDI\(_{15}\) varied from 1.03 to 2.66 when the glucose concentration was in the range of 50 - 250 mg/L. Moreover, the values of SDI\(_{15}\) were always higher than that of SDI\(_{30}\). Obviously, extending the time interval of SDI measurement would reduce the variation and significance of the index on fouling potential determination. The SDI data of humic acid demonstrated another pattern. The SDI\(_{15}\) values increased sharply in the lower concentration regions. Then, the increasing trend was slow down. Finally, the SDI\(_{15}\) values became flat in the higher concentration regions. Other research about humic acid fouling also demonstrated the same behavior (Park et al., 2006). According to the suggested criteria on fouling control, the SDI\(_{15}\)
should be lower than 2.0 for the water solution prior to conventional RO membrane
treatment (Eaux, 1996). Consequently, the experimental results revealed that the feed
congentration of glucose and humic acid should be controlled below approximately 180
and 0.005 mg/L, respectively, to reduce the fouling potential. The humic acid showed
evidently higher fouling potential than glucose.

MFI experimental data illustrated that a positive linear relationship between the
MFI values and the concentration of soluble organic matters was established with an
excellent correlation. The slope of linear correlations could be applicably used to
quantify the fouling potential of soluble organic matters. Table 1 summarizes the MFI
measurements and linear correlations of different soluble organic matters. According to
the linear correlations of the MFI values and organic concentration, the fouling
potentials of soluble organic matters were in the order of humic acid ≒ nucleic acid ≒
protein ≒ glucose.

3.2 Effects of molecular weight on SDI and MFI

The SDI experimental data of dextran demonstrated that the molecular weight of
dextran was an essential factor on SDI measurement. A higher molecular weight would
bring about a stronger fouling potential on the membrane. According to the criteria of
RO operation (Eaux, 1996), the concentration of dextran with molecular weight of 60-
90 kDa should be lower than 10 mg/L for avoiding membrane fouling. On the other side,
only a tiny amount of about 0.01 mg/L of dextran with molecular weight of 5-40 MDa
would cause serious fouling.

Table 2 summarizes the MFI values and its correspondent linear correlations of
dextran with different molecular weight. Based on the slope of linear equations of the
dextran concentration and its MFI values, the dextran with molecular weight of 60-90
kDa and 100-200 kDa had the same degree of fouling potential. The dextran with molecular weight of 4-50 MDa obviously showed higher fouling potential.

3.3 Fouling potential analysis based on saturation curve model

Table 3 shows the comparisons of SDI\textsubscript{15} values and the constants of the saturation curve model for various soluble organic matters under different levels of concentration. The glucose of 50 mg/L demonstrated slight effects on the SDI\textsubscript{15} value; the maximum accumulated filtrate ($V_{\text{max}}$) and filtration constant ($k_f$) approximated to the value of deionized water. The SDI\textsubscript{15} value of nucleic acid was very close to that of protein when the concentration was 1.0 mg/L. The SDI values could not clearly identify the difference of fouling potential between nucleic acid and protein in such conditions. However, based on the analysis of saturation curve model, the $V_{\text{max}}$ of nucleic acid and protein was 161.3 L and 103.1 L, respectively; and the correspondent $k_f$ was 94.1 and 78.9 minutes. The difference of accumulated filtrate is significant. Thus, the saturation curve model could provide a more evident distinction to identify fouling potential when the SDI values were very close.

The SDI\textsubscript{15} values of humic acid and dextran were close when the concentration was 0.01 mg/L. Comparing the constants of the saturation curve model, dextran with molecular weight of 5-40 MDa exhibited apparently lower values of $V_{\text{max}}$ and $k_f$ than humic acid. Therefore, the $V_{\text{max}}$ and $k_f$ obtained by the saturation curve model could be used to improve the precision of membrane fouling prediction caused by soluble organic matters.

4. CONCLUSIONS
The focus of this study was to investigate various soluble organic matters on membrane fouling index. The SDI and MFI were employed as indicators to interpret the fouling potential. Based on the results of this research, conclusions were made as follows:

1. The humic acid showed significant effects on membrane fouling indexes. The SDI<sub>15</sub> and MFI were 2.9-3.9 and 5.4-13.8 s/L<sup>2</sup>, respectively, when its concentration was in the range of 0.01-0.05 mg/L. The feed concentration of humic acid should be controlled below approximately 0.005 mg/L in order to reduce RO membrane fouling.

2. The SDI<sub>15</sub> and MFI illustrated similar behavior regarding fouling potential prediction of soluble organic matters.

3. According to linear correlations of organic concentration and the MFI, the fouling potentials were quantified in the order of humic acid > nucleic acid ≒ protein > glucose. Moreover, the molecular weight of dextran also played an important role in fouling indexes.

4. The maximum accumulated filtrate (\(V_{\text{max}}\)) and the constant of filtration (\(k_f\)) obtained from the saturation curve model were valuable to improve the precision of membrane fouling prediction caused by soluble organic matters.

References


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2003. Tools for fouling diagnosis of NF and RO membranes and assessment of the
on RO membrane colloidal fouling experiments with iron oxide particles.
Desalination 151, 229-238.
### Table 1. The MFI and linear correlations of various soluble organic matters

<table>
<thead>
<tr>
<th>Materials</th>
<th>Range of concentration (mg/L)</th>
<th>Range of MFI (s/L^2)</th>
<th>Linear correlations</th>
<th>R²</th>
</tr>
</thead>
<tbody>
<tr>
<td>glucose</td>
<td>50-250</td>
<td>1.2-2.6</td>
<td>MFI = 0.01(C)^3 + 0.96</td>
<td>0.98</td>
</tr>
<tr>
<td>humic acid</td>
<td>0.01-0.05</td>
<td>5.4-13.8</td>
<td>MFI = 207.4(C) + 3.74</td>
<td>0.98</td>
</tr>
<tr>
<td>nucleic acid</td>
<td>1.0-2.0</td>
<td>1.1-2.6</td>
<td>MFI = 1.38(C) - 0.33</td>
<td>0.95</td>
</tr>
<tr>
<td>protein</td>
<td>1.0-5.0</td>
<td>1.8-4.9</td>
<td>MFI = 0.76(C) + 0.91</td>
<td>0.98</td>
</tr>
</tbody>
</table>

* (C) represents the concentration of soluble organic matters.
Table 2. The MFI and linear correlations of dextran with different molecular weight

<table>
<thead>
<tr>
<th>Molecular weight</th>
<th>Range of concentration (mg/L)</th>
<th>Range of MFI (s/L²)</th>
<th>Linear correlations</th>
<th>R²</th>
</tr>
</thead>
<tbody>
<tr>
<td>60-90 kDa</td>
<td>10-50</td>
<td>1.3-3.4</td>
<td>MFI=0.05(C)+0.50</td>
<td>0.81</td>
</tr>
<tr>
<td>100-200 kDa</td>
<td>100-500</td>
<td>0.8-7.8</td>
<td>MFI=0.02(C)-1.98</td>
<td>0.94</td>
</tr>
<tr>
<td>5-40 MDa</td>
<td>0.01-0.1</td>
<td>1.1-8.9</td>
<td>MFI=69.6(C)+0.29</td>
<td>0.81</td>
</tr>
</tbody>
</table>

* (C) represents the concentration of dextran.
Table 3. The comparisons of SDI and the constants of saturation curve model

<table>
<thead>
<tr>
<th>Soluble organic matters</th>
<th>Concentration (mg/L)</th>
<th>SDI$_{15}$</th>
<th>$V_{max}$ (L)</th>
<th>$k_f$ (min)</th>
</tr>
</thead>
<tbody>
<tr>
<td>glucose</td>
<td>50</td>
<td>1.5</td>
<td>192.3</td>
<td>124.3</td>
</tr>
<tr>
<td>nucleic acid</td>
<td>1.0</td>
<td>1.9</td>
<td>161.3</td>
<td>94.2</td>
</tr>
<tr>
<td>protein</td>
<td>1.0</td>
<td>2.0</td>
<td>103.1</td>
<td>78.9</td>
</tr>
<tr>
<td>humic acid</td>
<td>0.01</td>
<td>2.9</td>
<td>86.2</td>
<td>53.1</td>
</tr>
<tr>
<td>dextran (5-40 MDa)</td>
<td>0.01</td>
<td>3.2</td>
<td>59.9</td>
<td>27.2</td>
</tr>
</tbody>
</table>