Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

Mathematical Modeling of Flux in Ultrafiltration Membrane for Water Treatment

Bao-Guo Wang[a]; Masaaki Ando[b]; Haitao Yin[a]; Bo Hong[a]; Yong Peng[a]
[a] Department of Chemical Engineering, Tsinghua University, Beijing, The People's Republic of China [b] Nitto Denko Co., Kusatsu, Shiga, Japan

To cite this Article Wang, Bao-Guo , Ando, Masaaki , Yin, Haitao , Hong, Bo and Peng, Yong(2006) 'Mathematical Modeling of Flux in Ultrafiltration Membrane for Water Treatment', Separation Science and Technology, 41: 6, 1179 — 1191
To link to this Article: DOI: 10.1080/01496390600641587
URL: http://dx.doi.org/10.1080/01496390600641587

Separation Science and Technology, 41: 1179–1191, 2006

ISSN 0149-6395 print/1520-5754 online
DOI: 10.1080/01496390600641587

Mathematical Modeling of Flux in Ultrafiltration Membrane for Water Treatment

Bao-Guo Wang
Department of Chemical Engineering, Tsinghua University, Beijing, The People's Republic of China

Masaaki Ando
Nitto Denko Co., Kusatsu, Shiga, Japan

Haitao Yin, Bo Hong, and Yong Peng
Department of Chemical Engineering, Tsinghua University, Beijing, The People's Republic of China

Abstract: Calculation of water flux in a spiral ultrafiltration element was conducted by the novel proposed method, in which all necessary parameters were determined solely from membrane sheet test. By comparing water flux of the commercial element with the data from membrane sheet measurement, this study found that the tendency of water flux variation with time in the membrane element was similar to that which occurred in the membrane sheet, including the consideration of scale-up effect due to hydrodynamics influence. Therefore, it is possible to express the variation of water flux in the membrane element based on the results from membrane sheet measurement using a practical water source. Surface water and sea water were separately employed to carry out a pilot test with an 8-inch spiral membrane element, made of polyvinylidene fluoride(PVDF) with a molecular weight cut-off of 150 kDa, and water fluxes under various transmembrane pressures. Calculations were approximate agreement with that of the pilot test, which enables us believe that the proposal method is reliable for designing a practical ultrafiltration system.

Keywords: Spiral element, ultrafiltration, scale-up modeling, water treatment

Received 29 November 2005, Accepted 7 February 2006
Address correspondence to Bao-Guo Wang, Department of Chemical Engineering, Tsinghua University, Beijing 100084, The People's Republic of China. Fax: 86-10-62770304; E-mail: bgwang@tsinghua.edu.cn

INTRODUCTION

The introduction and rapid development of membrane technologies in water treatment during the past decades represent a significant milestone in water industry, including microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) (1–5). Among these techniques, ultrafiltration membrane separation possesses special aspects in drinking water processes due to its capability of providing quality water at a relatively modest capital and operation cost (6). Moreover, development of waste water treatment and recycling technologies has been carried out worldwide, and some successful examples confirm the significance of ultrafiltration separation, in which the UF membrane plays the role of pretreatment for RO process (7, 8). This method is helpful for overcoming the shortage of water as a resource and keeping the environment free from pollution. However, a major obstacle in membrane separation is the permeation flux decline due to concentration polarization and contaminant accumulation on the membrane surface (9, 10). In order to obtain recovery of the water permeation rate, a membrane cleaning process is usually necessary, and then the operation enters the next cycle. Generally, there are three methods to recover membrane flux (11, 12):

i. Chemical cleaning methods including strong acid and basic solution or oxidation agents;
ii. Physical methods such as back flushing and the use of turbulence promoters;
iii. Hydrodynamic methods related to module design.

Ultrafiltration membranes are often used to deal with various water sources in which the composite solutes and their characteristics (such as size and density of charge, etc.) are not well defined. These water sources may also contain membrane foulant from organic, inorganic, or biological substances. Because of the difficulty in analyzing trace composition in water and confirming the contaminant species, as well as the variation in water sources with season, so far, there is still no completely developed method to calculate water flux for a commercial ultrafiltration membrane element. Water treatment design for membrane separation usually depends on the results obtained by pilot tests which are carried out with commercial elements to filter the actual water, under a defined temperature, pressure, and backwashing conditions. A pilot test usually is time-consuming and requires the construction of equipment and experimental facilities, thus resulting in increased project cost, and decreased efficiency. In order to develop a calculation method for UF membrane-based water treatment, there have been a few studies related to the calculation of permeate concentration in unsteady operation processes, and these have shown useful results (13, 14). However, these models ignored the process of membrane fouling and the effect of backwashing on water flux, and investigation was often achieved by a modeling solution, so that the simulation results usually deviate from the data of the actual water treatment process. The principal objective of this study was to develop a simulation method for designing a practical UF system for water treatment, based on the results from membrane sheet tests, and prove its reliability by comparing the simulation results with that of an element scale pilot test.

THEORY

Unsteady operation is one of the typical characteristics of UF membrane-based water treatment processes, and water flux usually varies with time as a result of membrane fouling. In order to investigate the nature of water permeation behavior, this study compared the flux variation of a membrane sheet for a given wastewater with that of a commercial spiral element between chemical backwashing steps. Both cases showed similar water flux time dependence.

1. Under a constant transmembrane pressure, UF membrane flux is highest initially and then decreases exponentially with time.
2. The initial flux increases with transmembrane pressure, whereas, the flux whose initial value is higher decreases more quickly than that of a low initial flux.

Although the flat membrane sheet has permeation behavior similar to that of the element, the effect of scale-up due to velocity and concentration polarity has to be considered. In particular, the effective transmembrane pressure varies in the axial direction in a spiral membrane element, because of the friction pressure loss and water permeating through the membrane. Additionally, part of the membrane area in a spiral UF element becomes unavailable in dead corners and around the spacer, and membrane fouling also varies in the separation processes. These factors make it difficult to develop an exact mathematical model to describe mass transfer in filtration and estimate water flux time dependence. In order to describe water flux variation with time and eliminate the need for determining complicated model parameters, this study proposes a simple exponential function as follows.

$$J = Flux_0 \cdot time^b \tag{1}$$

$$Flux_0 = c \times \frac{\Delta p^d}{\mu(T)} \tag{2}$$

c: Permeation constant related to water quality and operating conditions;

d: Compressibility factor;

Δp: Transmembrane pressure [kg/cm^2];

Flux$_0$: Initial element flux [m^3/h];

$\mu(T)$: Water viscosity at temperature T. Its value can be calculated with the formula:

$$\mu(T/^\circ C) = x_1 - x_2 T + x_3 T^2 \text{ [mPa.s]}$$

Here, x_1, x_2, x_3 are parameters, used in this case as (15):

$$x_1 = 1.7252; \quad x_2 = -4.767 \times 10^{-2}; \quad x_3 = 5.81 \times 10^{-4}$$

The exponential factor b is relative to transmembrane pressure, temperature, and membrane fouling status where transmembrane pressure is taken as the average across the membrane sheet or element. Because the measurement with a membrane sheet indicated that the tendency of water flux variation was similar to that of the element pilot test, it is reasonable to estimate the model parameters b, c, and d by fitting flat sheet results. Accordingly, we propose a novel method for estimating commercial membrane element performance solely based on the data from membrane sheet measurement in laboratory. The following describes the method of determining model parameters.

1. Measure the flux variation of the membrane sheet at various pressures.
2. Determine the effective filtration area for a commercial membrane element. In order to avoid the influence of membrane fouling, pure water was used to measure the flux of the membrane sheet and the spiral element under the identical transmembrane pressure and temperature. The following formula was used to determine commercial element effective area S_{eff}.

$$S_{eff} = \textit{element specific membrane area} \times \textit{effective area factor } f \tag{3}$$

$$f = \frac{\textit{Element permeating rate}}{\textit{Water flux of the membrane sheet with the equivalent element area}} \tag{4}$$

3. Transform the membrane sheet results into equivalent element values using:

$$J = \frac{\Delta V}{\Delta t} \cdot \frac{S_{eff}}{S_{flat-sheet}} \quad [\text{m}^3/\text{h}] \tag{5}$$

Here ΔV is the accumulated water volume in the membrane sheet test at a defined trans-membrane pressure at various times, and Δt denotes the time interval of sample collection.

4. Determine the model parameters for calculating element flux.

By fitting the curve of the relationship between the equivalent element flux and time with the proposed equation (1), a series of values for b can be obtained corresponding to various transmembrane pressures. In addition, plotting of the initial equivalent element flux against the transmembrane pressure enables parameters c and d to be determined to describe the dependence of viscosity on temperature.

EXPERIMENTAL METHOD

A batch apparatus was used to measure the membrane sheet flux with an actual water sample as feed (Fig. 1). The membrane cell held a poly(vinylidene fluoride) (PVDF) flat membrane sheet, with an effective area of $60\,cm^2$. The cell (C10-T) was connected with 4 valves, and the filtration and backwashing could be switched conveniently using these valves. A centrifugal pump was used to drive water into the cell, and the adjustable valve (V-1) maintained a given transmembrane pressure from $0.4–2.4\,kg/cm^2$, indicated by the pressure meter (PI). Closing the outlet valve (V-5), membrane separation was conducted with a dead-end mode, and the permeated water volume at 5, 10, 15, 20, 25, and 30 minutes was recorded and used to estimate model parameters.

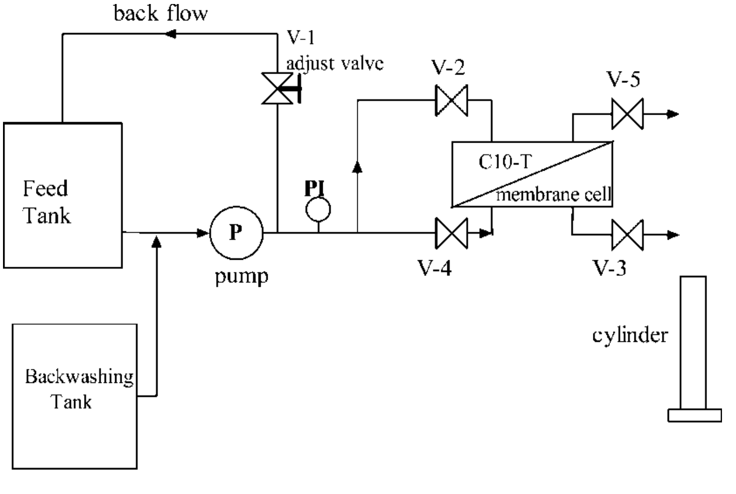

Figure 1. Schematic of flat membrane sheet test apparatus.

A pilot UF system was constructed, equipped with an 8-inch spiral element, made of poly(vinylidene fluoride) (PVDF) with a molecular weight cut-off of 150 kDa, and a specific membrane area of 24 m^2. The pilot apparatus was operated in cross flow mode, and each filtration cycle included three steps:

1. Water permeation through the UF membrane 13 min;
2. Back washing for 90 seconds;
3. Flushing for 30 seconds.

With the help of the control valve system, the operating mode could be changed automatically with the given program. The water flux and cross flow rate were measured by a rotameter. When the flux became very low due to fouling of the membrane surface, 15–20 ppm sodium hypochlorite aqueous solution was employed to remove the foulant, by 30–40 min of immersion and backwashing; thus the flux could be restored.

RESULT AND DISCUSSION

Determination Simulation Parameters

In the ultrafiltration process, both the flat membrane sheet and the element possess the same water flux under the identical membrane fouling status and the same transmembrane pressure. To attain the same amount of foulant on the membrane sheet, filtration with the sample water was performed before the measurement of water flux variation, by repeating membrane permeation and backwashing many times until the flux approached an approximate constant, which was regarded as the steady flux for membrane fouling. After this pretreatment, the variation of the membrane sheet flux over time was measured corresponding to various transmembrane pressures, and these data were transformed into the values equivalent to the element scale performance with the help of equation (5). When the membrane element was used for polluted water filtration, the variation of cross flow velocity had hardly any influence on flux. So the data from the membrane sheet tests obtained in dead-end mode were directly employed to determine model parameters for describing the performance of the membrane element in cross flow filtration.

Using the treated surface water as feed, whose composition is listed in Table 1, Fig. 2 shows the variation of water flux with time. A higher transmembrane pressure produces greater initial flux followed by a rapid flux reduction due to fouling. By fitting these data to equation (1), the exponential factor b was determined for various transmembrane pressures. Using the initial flux as the Y axis, the dependence of the initial flux on transmembrane pressure was plotted, as shown in Fig. 3. The parameters c and d in equation

Table 1. Characteristics of surface water composition change by UF membrane separation

Item	Feed water	After UF treatment
pH	7 ~ 7.5	7 ~ 7.5
Suspended solids [mg/L]	19	0
COD_{Cr} [mg/L]	85.6	42.9
Conductivity [μS/cm]	771	768
Color	60	15
Turbidity [NTU]	17.3	0

(2) were obtained from data regression to describe the effect of temperature on viscosity.

Case 1: Surface Water Permeation Behavior

In order to demonstrate the availability of the proposed method for estimating water flux in a large water treatment systems, this study measured the water flux of UF element with surface water from a lake, located at a northern suburban area of Beijing. The pilot test continued over two months, and the feed water composition varied slightly due to the arrival of summer. In addition, surface water represents a typical case in most drinking water processes. Table 1 shows a comparison of the typical items of the product with the feed water.

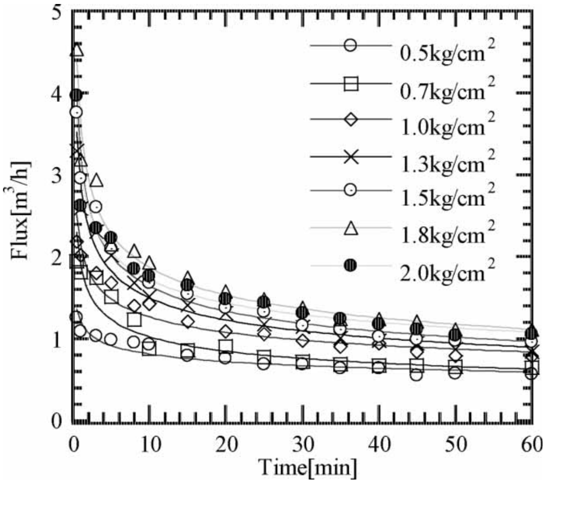

Figure 2. Variation of membrane sheet flux with time under various cross membrane pressure for surface water treatment at ambient temperature.

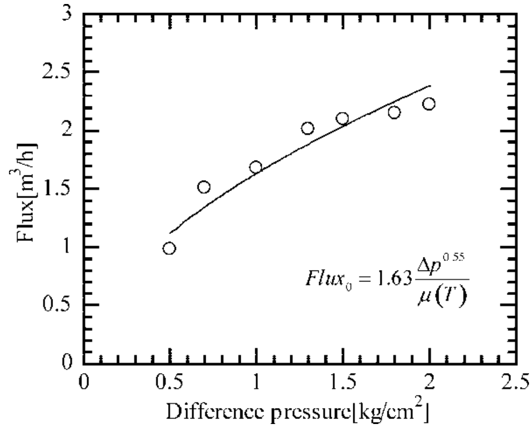

Figure 3. Variation of membrane sheet initial flux with transmembrane pressure for surface water treatment with cross membrane pressure.

Most of the suspended solids and biological material were removed during the UF membrane separation, thus resulting in an obvious decrease in turbidity. As the soluble compound would not have been rejected by the UF membrane, the decrease of COD_{Cr} and color was attributed to the gel layer which was formed on the membrane surface. The gel layer usually leads to water flux decline. Soluble inorganic salt could not be rejected at all, water pH and conductivity almost contained the same values in the separation processes.

As shown in Fig. 4 and Fig. 5, the water flux decreases over time due to the increase in fouling on the membrane surface. A higher transmembrane

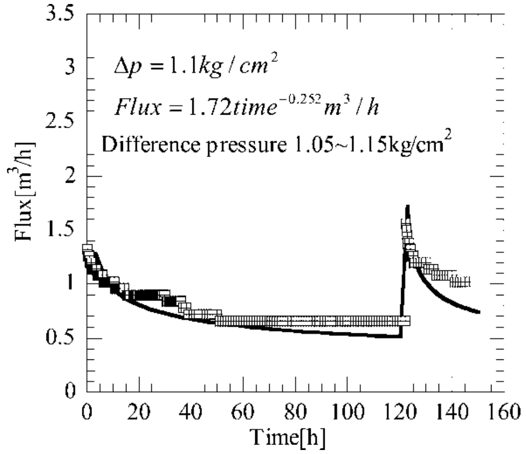

Figure 4. Comparison of water flux calculated in this study with element measurement for surface water treatment.

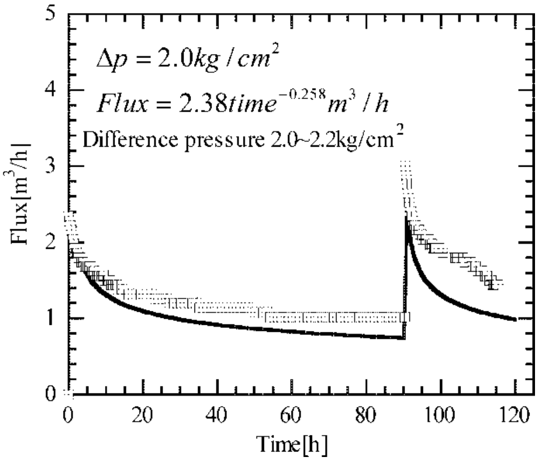

Figure 5. Comparison of water flux calculated in this study with element measurement for surface water treatment.

pressure led to an increase in initial water flux but the water flux curve showed a more rapid decline. Thus more frequent chemical washing had to be performed to restore the water flux. The average error between the calculated and measured water flux was less than 15%. These results have encouraged us to employ this method in the design of a practical water treatment system.

In addition, the composition of the surface water usually varies with time, leading to water flux of UF element changes seasonally (16), because a great

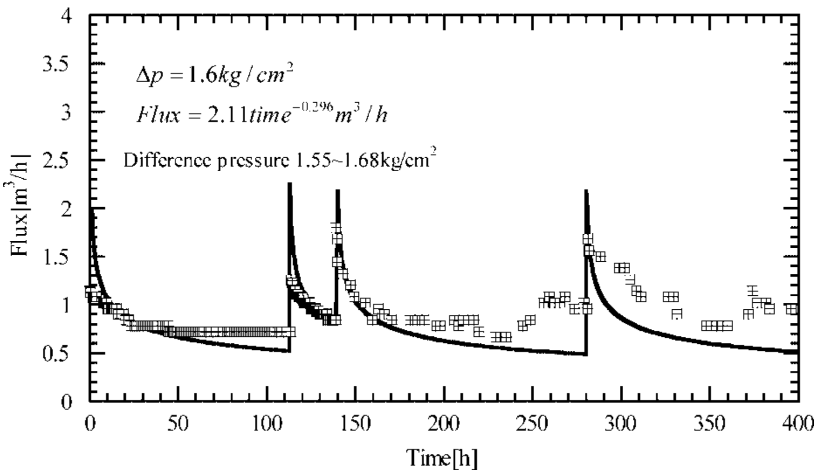

Figure 6. Comparison of water flux calculated in this study with element measurement for surface water treatment.

deal of plant and plankton grows in the water in the summer. For understanding the influence of water composition upon filtration properties, this study observed membrane flux change during 400 hours test, which was carried out continuously, keeping transmembrane pressure around $1.6\,kg/cm^2$ and cleaning membrane element with sodium hypochlorite every week. Figure 6 compares the simulation results with the measurement, and shows a good agreement for the most time. With the arrival of summer, rain diluted the pollutant concentration in the lake and water temperature rose somewhat, thus reducing foulant accumulation on the membrane and leading to an increase in water flux. Whereas, the calculation still used the original parameters, calculation deviated the element data slightly.

Case 2: Seawater Filtration for RO Feed Pretreatment

In recent years, ultrafiltration is expected to be a useful technique for RO feed pretreatment in seawater desalination, for its high quality produced water and modest capital. With the same simulation method, filtration measurement was conducted with seawater from Bo-Hai gulf in northern China, and the obtained data was used to evaluate the calculation accuracy. In the pilot measurement, seawater was directly introduced into the UF spiral element. Parts of representative terms are listed in the Table 2.

As can be see in Fig. 7 and Fig. 8, an increase in transmembrane pressure leads to the relative high initial flux and rapid flux deduction, similar to the behaviors that occurred in the surface water treatment. Using the same water source, membrane sheet test was carried out to obtain the necessary parameters for calculating water flux in element measurement. Generally, simulation results can describe flux variation of the element scale measurement, even though no direct pilot data was employed in determining modeling parameters. Calculation shows a negative deviation in Fig. 8, and this needs further investigating the characteristic of membrane fouling for transmembrane pressure below $1.0\,kg/cm^2$ and improving the calculation accuracy in the future study.

Table 2. Typical composition of the treated seawater

Item	Feed source
Temperature [°C]	22 ~ 26
pH	6.8 ~ 7.2
Suspended solids[mg/L]	25 ~ 30
COD_{Mn} [mg/L]	25
Conductivity[μS/cm]	61,000 ~ 65,000
Total iron ion [mg/L]	~0.3
Turbidity[NTU]	32 ~ 40
Total soluble solids [mg/L]	37858

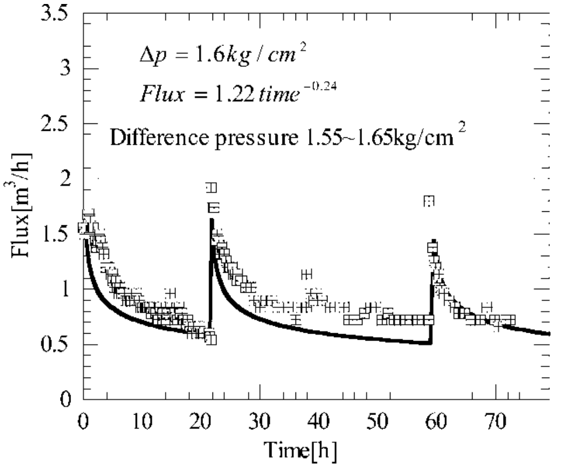

Figure 7. Comparison of water flux calculated in this study with element measurement for seawater treatment.

CONCLUSION

This study proposed a method for calculating the flux in a commercial spiral UF element for water treatment. All of the necessary parameters can be determined solely from membrane sheet tests, including the measurement of the dependence of water flux on transmembrane pressure and the effect of scale-up. The element scale pilot test was carried out using surface water and seawater

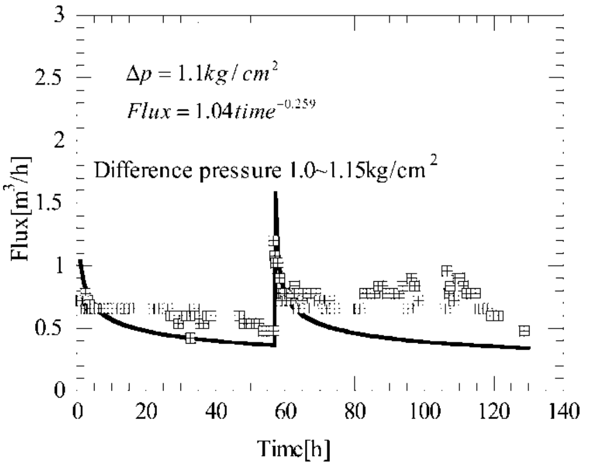

Figure 8. Comparison of water flux calculated in this study with element measurement for seawater treatment.

separately, both cases of UF membrane showed an excellent rejection rate for removing suspended solids and decreasing water turbidity. Because the parameters obtained in this way reflect the membrane fouling, and the model's development was based on unsteady permeation, the calculated results provide a reasonable description of the flux variation in the element scale pilot tests for various transmembrane pressures. The developed method should be useful for designing UF membrane water treatment processes.

ACKNOWLEDGEMENTS

The authors gratefully acknowledge the financial support from the National Natural Science Foundation of Science (20276033, 20276014) and the Major State Basic Research Development Program of China (973 Program) (2003CB615701).

REFERENCES

1. Klomfas, G. and Konieczny, K. (2004) Fouling phenomena in unit and hybrid processes for potable water treatment. *Desalination*, 163 (1–3): 311–322.
2. Arora, M., Maheshwari, R.C., Jain, S.K., and Gupta, A. (2004) Use of membrane technology for potable water production. *Desalination*, 170 (2): 105–112.
3. Shaalan, H.F. (2003) Development of fouling control strategies pertinent to nanofiltration membranes. *Desalination*, 153 (1–3): 125–131.
4. Kiso, Y., Sugiura, Y., Kitao, T., and Nishimura, K. (2001) Effects of hydrophobicity and molecular size on rejection of aromatic pesticides with nanofiltration membranes. *J Membr. Sci.*, 192 (1–2): 1–10.
5. Enegess, D., Togna, A.P., and Sutton, P.M. (2003) Membrane separation applications to biosystems for wastewater treatment. *Filtration & Separation*, 40 (1): 14–17.
6. Choi, H., Kim, H.S., Yeom, I.T., and Dionysiou, D.D. (2005) Pilot plant study of an ultrafiltration membrane system for drinking water treatment operated in the feed-and-bleed mode. *Desalination*, 172 (3): 281–291.
7. Qin, J.J., Oo, M.H., Lee, H., and Kolkman, R. (2004) Dead-end ultrafiltration for pretreatment of RO in reclamation of municipal wastewater effluent. *J Membr. Sci.*, 243 (1–2): 107–113.
8. Bonnelye, W., Sanz, M.A., Durand, J.P., Plasse, L., Gueguen, F., and Mazounie, P. (2004) Reverse osmosis on open intake seawater: Pre-treatment strategy. *Desalination*, 167 (1–3): 191–200.
9. Sivik, E. and matthiasson, B.C. (1980) Concentration polarization and fouling. *Desalination*, 35: 59.
10. Hanemaaijer, J.H., Robbertson, T., van den Boomgaard, T., and Gunnink, J.W. (1989) Fouling of ultrafiltration membrane: The role of protein adsorption and salt precipitation. *J. Membr. Sci.*, 40: 199–217.
11. Srijaroonrat, P., Julien, E., and Aurelle, Y. (1999) Unstable secondary oil-water emulsion treatment using ultrafiltration: flouing control by backflushing. *J. Membr. Sci.*, 159: 11–20.

12. Laitinen, N., Luonsi, A., Levanen, E., and Nystrom, M. (2001) Effect of backflushing conditions on ultrafiltration of board industry wastewaters with ceramic membranes. *Separation & Purification Technology*, 25: 323–331.
13. Muller, A., Daufin, G., and Chaufer, B. (1999) Ultrafiltration modes of operation for the separation of α-lactalbumin from acid casein whey. *J. Membr. Sci.*, 153: 9–21.
14. Darnon, E., Morin, E., Belleville, M.P., and Rios, G.M. (2003) Ultrafiltration within downstream processing: Some process design considerations. *Chemical Eng. & Processing*, 42: 299–309.
15. Jiang, W.J., Dai, Y.Y., and Gu, H.J. (1992) *Principle of Chemical Engineering*. Tsinghua University Press: Beijing, PRC.
16. Zhang, B. and Yamamoto, K. (1996) Seasonal change of microbial population and activities in a building wastewater reuse system using a membrane separation activated sludge process. *Water Sci. & Technology*, 34 (5–6): 295–302.