Critical design considerations for harnessing reverse osmosis processes in water/wastewater treatment

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Water Science & Technology: Water Supply **Vol 6 No 6 pp 61–70**

Abstract In this paper, the performance of the full-scale RO process with highly permeable membranes and the governing mechanisms were carefully studied. It was found that the performance of a full-scale RO process could be controlled by two possible mechanisms, namely mass transfer rate and thermodynamic limitations. Under relatively low driving pressure, it was controlled by mass transfer rate (water flux) of the membrane. However, with the highly permeable membrane, it is possible that the performance is limited by the thermodynamic limitation, in which the osmotic pressure becomes equal to the driving pressure inside of the membrane channel. A process controlled by thermodynamic limitation is an extremely case of the hydraulic imbalance problem. When it occurs, it means part of the membranes in the processes do not contribute to permeate production. More complicated are situations in the intermediate pressure range, in which both mechanisms contribute to, but none of them can dominate, the performance of the process.

Some innovative concepts and theories on the performance of the full-scale RO processes were developed. These concepts and theories may provide better qualitative explanations for the behaviors often observed in the full-scale RO processes. A better quantitative simulations or predictions of the performance of the process were developed upon these concepts and theories. Experiments were carried out on a pilot membrane process of 6 m membrane channel to imitate the performance of the full-scale RO under various conditions. The experimental performance data were compared with theoretical simulations and excellent agreement was obtained.

Another focus of this current study was on characterization and modeling of membrane fouling in the full-scale RO process. Colloidal fouling experiments were conducted to study the fouling potential of feed water and a new fouling indicator was proposed. The indicator can be directly used in the mathematical model to simulate fouling development in the full-scale RO processes. Model simulations showed that under certain condition (thermodynamic restriction), the recovery or average permeate flux of a full-scale RO process would maintain a constant value even membrane fouling was taking place. Experimental verification of the simulation results is currently under way. With the new developments and findings in this area, methods or protocols for optimization of full-scale processes of the highly permeable RO membranes were suggested.

Keywords Reverse osmosis; RO; wastewater treatment; water treatment

Introduction

Application of reverse osmosis (RO) processes for water and wastewater treatment has been growing rapidly in recent years and it has become one of the major technologies for producing potable water in many places in the world (Taylor and Jacobs, 1996; Mulder, 1996). However, most of our knowledge about RO membranes still remain on the small scale processes employed in laboratories. A few years ago, my research group attempted to develop a comprehensive model to simulate the full-scale RO system, which was intended for optimization of the system design and operation. One key task of the comprehensive model is to predict the permeate flux from a long RO channel (the length of the RO channel is usually 6 or 7 metres in one stage).

Basic membrane transport equation

Literature review revealed that most published studies on the full-scale RO processes predicted the average permeate flux with the basic membrane transport law (Taniguchi and

doi: 10.2166/ws.2006.950

Kimura, 2000; Taniguchi *et al.*, 2001; Al-Bastaki and Abbas, 1999, 2000):

$$v = l(\Delta p - \Delta\pi) = \frac{(\Delta p - \Delta\pi)}{R_m} \quad (1)$$

where v is permeate flux, Δp is the driving pressure, $\Delta\pi$ is the osmotic pressure, l is permeability of the membrane, and R is the resistance of the membrane.

Although the applicability of Equation (1) to a small piece of membrane is no doubt verified with numerous experiments, the direct use of Equation (1) to the full scale RO processes is not appropriate because the osmotic pressure in the channel is no longer a constant. There is a notable increase in osmotic pressure along the membrane channel as permeate is being produced.

Cascade calculation method

It was also noted from industry practices that the accuracy of permeate flux prediction from a full-scale RO process could be improved by the so-called cascade calculation method, in which the basic membrane transport law (Equation (1)) was applied piecewise to each element in the pressure vessel. Average osmotic pressure of the feed end and the concentrate end of the element, $0.5(\Delta\pi_f + \Delta\pi_c)$, was used in the cascade calculation (Taylor and Jacobs, 1996). The osmotic pressure at the concentrate end of the membrane element was initially guessed and gradually approached to a convergent value with an iteration scheme.

The cascade calculation method usually can produce predictions with quite high accuracy for the old generation of membranes, which were operated at driving pressures that was much higher than the osmotic pressure at the concentrate end. For example, when the membrane resistance is 10^{12} Pa.s/m and salt concentration of the feed water is 5,000 mg/l, a driving pressure of about 800 psi is required for a 6 m RO channel to reach about 50% recovery. At this time the salt concentration at the concentrate end of the RO channel is 10,000 mg/l. The osmotic pressure of the concentrate is about 100 psi, which is only one-eighth of the driving pressure. For such situations, permeate flux of the pressure vessel can be excellently predicted with the cascade calculation.

The currently used RO membranes usually have resistance that is more than one order lower in magnitude than the old generations. Consequently, the driving pressure is significantly reduced. For the above example but with a new membrane of resistance of 10^{11} Pa.s/m, a driving pressure of about 120 psi may reach 50% recovery. In this case, the net driving pressure changes from 70 psi at the feed end to 20 psi at the concentrate end. The net driving pressure and the resulted permeate flux change more drastically in the membrane channel with the more permeable RO membranes (Figure 1). When the osmotic pressure is comparable to the driving pressure, the accuracy of the cascade calculation method deteriorates significantly and sometimes it fails to produce convergent results.

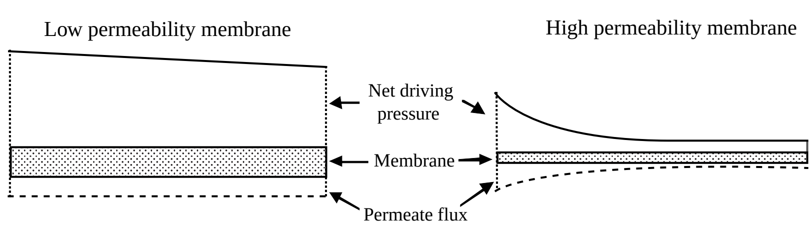

Figure 1 Variations of net driving force and permeate flux along membrane channel with low and high permeable membranes

Finite difference method

To further improve the accuracy of permeate flux calculation in the full-scale RO process, a finite difference method was developed for the membrane processes with highly permeable RO membranes. The whole channel was first divided into many small segments so that the properties in each segment can be treated as constant. In each segment, the permeate flux v, cross flow velocity u, salt concentrate c, and pressure drop Δp were determined with:

$$\text{(Membrane transport law)} \quad v_i = \frac{\Delta p_{i-1} - \Delta\pi(c_{i-1})}{R_m} \tag{2}$$

$$\text{(Mass conservation on water)} \quad u_i = u_{i-1} - v_{i-1}\Delta x/H \tag{3}$$

$$\text{(Mass conservation on salt)} \quad c_i = \frac{u_{i-1}c_{i-1} - (1-r)v_i\Delta x/H}{u_i} \tag{4}$$

$$\text{(Pressure loss due to friction)} \quad \Delta p_i = \Delta p_{i-1} - \frac{12k\eta}{H^2}u_{i-1}\Delta x \tag{5}$$

where Δx is the length of the membrane segment, H is the height of the channel, r is the salt rejection rate, k is the friction coefficient, and η is the viscosity of water.

Although the principle or concept is similar, the advantages of the finite difference method over the cascade method are obvious. First, the membrane transport and conservation principles are applied locally to small segments of membranes. Because the osmotic pressure in the small piece can be treated as a constant, no iteration is needed. For the same reason, there is no overshooting as in the cascade method where iteration is used. Secondly, the finite difference method is more flexible. The total number of the segments used in calculation can be easily adjusted to fit the requirement of more permeable RO membranes. Thirdly, the finite different method is robust and will produce the result for any conditions of operation and never break down.

Therefore, the initial goal of our research was to develop the finite difference method that can produce a more accurate, more stable and robust result for the prediction of the permeate flux in the full-scale RO processes. The result of model development was published in 2002 (Song *et al.*, 2002).

If our investigation stopped at this conjecture, we have to confess that the phenomena addressed above were obvious to many people who were knowledgeable of the real-world RO processes. Although the finite difference model has its own merit in more accurate simulation of the full-scale RO processes that is required for optimization of process design and operation, fundamentally it represents only small incremental contributions to the existing knowledge pool of RO processes.

Emergence of thermodynamic restriction

We used the finite difference model extensively to investigate the performance of the long RO channel with different parameters under various operating conditions, including membrane resistance, driving pressure, salt concentration, and channel length. To our big surprise, the simulation results showed us that with the RO membrane of resistance R_m equal to or smaller than 10^{11} Pa.s/m, there were many interesting behaviors of the full-scale RO under common operating conditions, which had not been reported or discussed in the literature.

Thermodynamic restricted flux

From the day one of invention of the RO process, it was known the permeate flux would be zero when the driving pressure was equal to the osmotic pressure, i.e., $(\Delta p - \Delta\pi) = 0$. However, it was not a common knowledge that a full-scale RO could be controlled by the zero net driving force under the common operating conditions employed in the practical RO processes.

Our simulating study with the finite difference model showed that the linear law between flux and driving pressure broke down when concentration polarization was completely not considered (Figure 2). When the channel was sufficiently long, the average permeate flux decreased with the increasing length of the channel (Figure 3). In this case, if the permeate recovery was plotted vs. channel length, it would be a constant invariant with channel length (broken line). These interesting behaviors of the full-scale RO processes prompted us to look for the underlying reason or mechanism for the constant recovery that was not affected by the length of the membrane channel.

The finding of the non-linear dependence of the average permeate flux on the driving pressure was not planned by us. Although it was not known to us before, it must have been known for a long time to many researchers and engineers, especially to these who experienced with module testing and practical process design. It was a little surprise to me that the behavior has not been extensively reported or discussed in the literature. The behavior might be thought too obvious or too trivial to discuss. It also might be because it violated the linear relationship that has been accepted in this field for a long time, or it could not be explained from the known principles, or for other reasons.

We believed that the above behavior of the full-scale RO was significant and hinted another limiting mechanism for the process. It did not take us long to find out that the limitation was actually thermodynamic equilibrium. In a long membrane channel with highly permeable membrane, the equilibrium salt concentration can be reached inside the channel (Figure 4). After the point at which the equilibrium is reached, the rest of the channel does not produce any permeate-dead region. Therefore, the average flux decreases as the dead region increases. This mechanism explains well the behavior of average permeate flux reported in Figure 3.

Calculation of thermodynamic restricted flux

When a process is known at thermodynamic equilibrium, there is no need to use the cascade method or the finite difference method to calculate the permeate flux or recovery.

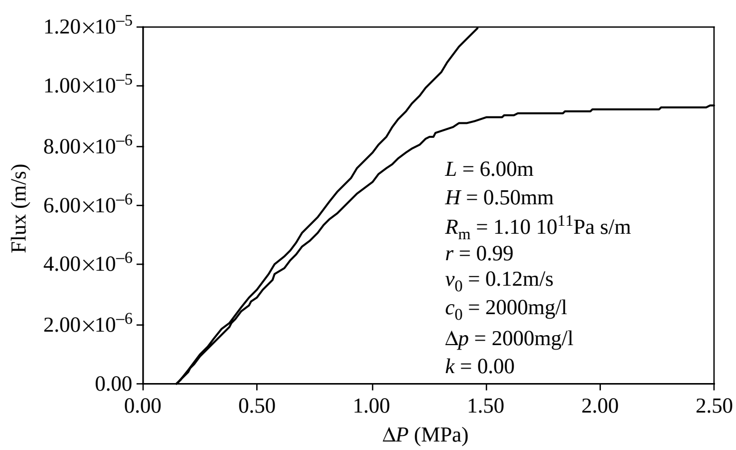

Figure 2 Non-linear relationship of v vs Δp. Parameters for calculation are given in the figure

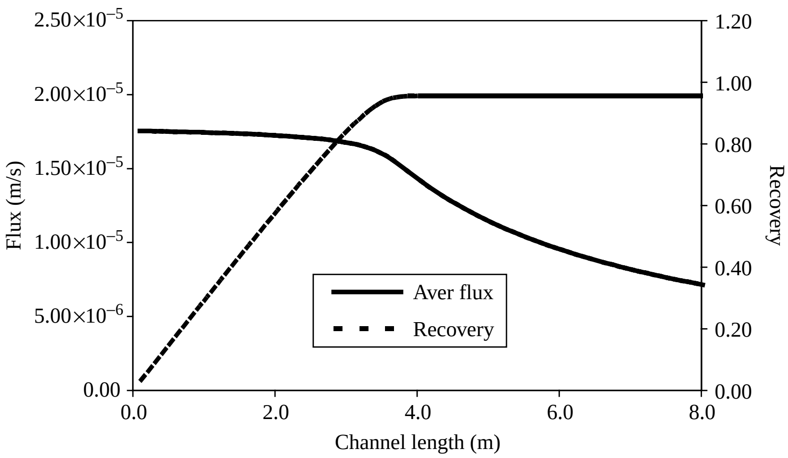

Figure 3 Average flux decreases with channel length. Parameters used in simulation are the same as in Figure 2

The recovery can be determined easily from thermodynamic principle, i.e.,

$$R = 1 - \frac{\Delta\pi_0}{\Delta p} \tag{6}$$

where R is the recovery and $\Delta\pi_0$ is the osmotic pressure of the feed water. With the conversion coefficient (respect ratio) Hv_0/L, the average permeate flux is determined as:

$$\bar{v} = \left(\frac{Hv_0}{L}\right) \cdot R = \frac{Hv_0}{L}\left(1 - \frac{\Delta\pi_0}{\Delta p}\right) \tag{7}$$

Equations (6) and (7) are important because they give us the upper limit of the performance of full-scale RO under the given salt concentration and driving pressure. The most direct use of these two equations is to evaluate the full-scale RO processes. If a process has recovery or average flux much smaller than the values calculated with these two equations, the process can be improved by prolonging the membrane channel and increasing mass transfer coefficient. If the recovery or average flux is near to the predicted value of Equations (6) or (7), the process efficiency cannot be further increased by the above measures.

Condition for thermodynamic equilibrium

It is demonstrated that there are two possible controlling mechanisms for the performance of the full-scale RO processes, namely mass transfer (Equation 1) and thermodynamic equilibrium (Equation 6 or Equation 7). Mass transfer would be the controlling mechanism under low pressure because the low production of permeate should not cause significant change in salt concentration in the flow. The thermodynamic restriction would take over under sufficiently high pressure due to the substantially elevated salt concentration.

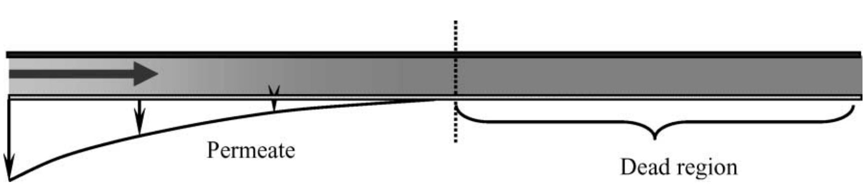

Figure 4 Substantial variation of the permeate flux along the channel and dead region

There was a transition pressure, ΔP_T, for each full-scale RO process (Figure 5). The average flux increases linearly with the driving pressure when it is smaller than the transition pressure, while increases marginally when the driving pressure is higher than the transition pressure. With the knowledge of transition pressure, the appropriate equation can be chosen for the calculation of permeate flux. Further study resulted in an analytical formula of the transition pressure, which can be expressed with the basic parameters of the full-scale RO processes (Song *et al.*, 2003):

$$\Delta P_T = \frac{u_0 H R_m}{L} = \frac{Q}{S} \cdot R_m \quad (8)$$

where Q is the feed flow rate to a pressure vessel and S is the total membrane area. The transition pressure can be expressed as the product of the feed flow rate per unit membrane area and the membrane resistance. The linear dependence of the transition pressure on the membrane resistance indicates the enormous effect of the improvement of RO membrane permeability on the reduction of the transition pressure. To our surprise, the transition pressure is not related to the salt concentration in the feed.

Working pressure

The knowledge of the characteristic pressure can help to determine the working pressure of membrane process, which was not able to be done before. Figure 6 is a schematic description of the guideline. For any RO process, the recovery-pressure (or average flux-pressure) curve can be developed with the method presented in this paper. The typical curve has a turning point (at pressure P_1 in Figure 6) that divides the pressure domain into two regions. In the region of lower pressure, recovery (or average flux) basically increases with pressure linearly. In the region of higher pressure, the recovery (or average flux) only increases marginally with pressure. It is obviously that P_1 is the optimal working pressure in the sense of recovery (or flux) increment per unit pressure. It can be seen that when pressure increase significantly from P_1 to P_2, the recovery increment $R_2 - R_1$ is rather small.

It is worth to point out that the turning point is obvious for low salt concentration. As the salt concentration increases, the turning point may become less obvious and the recovery (or flux) shows a strong nonlinear dependence over the entire pressure domain and the rate of flux increase gradually decreases with pressure. However, the determination of the working pressure for this case is less critical.

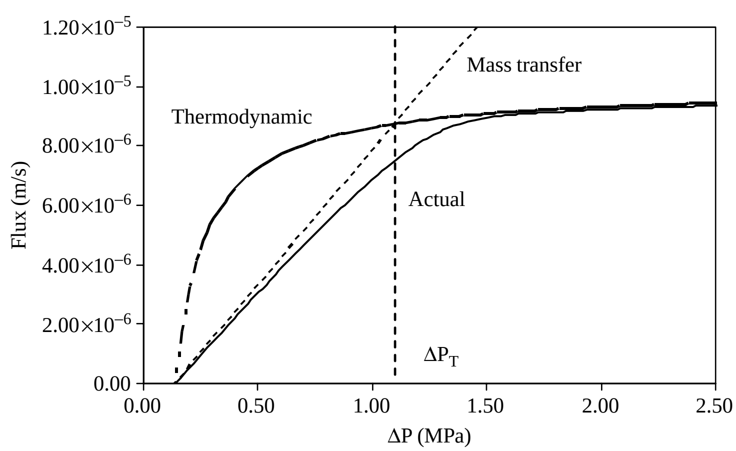

Figure 5 Transition pressure and two distinguished regimes

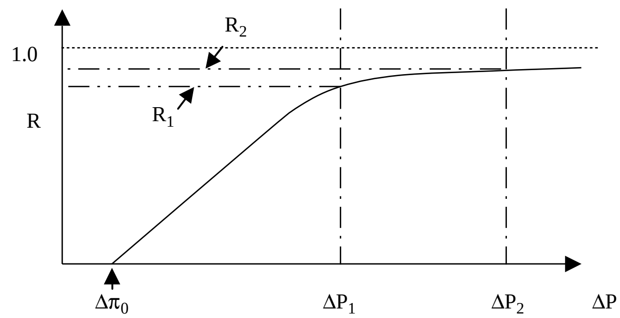

Figure 6 Schematic for determination of working pressure for a RO membrane process

Evidences of thermodynamic restriction in literature

There were plenty of indirect evidences of the thermodynamic restriction in the literature. The first evidence was the significant reduction of the driving pressure in the practical RO processes in recent years. RO membrane used to be operated at very high driving pressure ($>$500 psi) but has been reduced gradually to around 100 psi for brackish water. One obvious reason to operate RO at much lower pressure is that the increment of permeate flux is only marginal when the RO process hits the wall of thermodynamic restriction. Another piece of evidence was the reports of the imbalanced permeate production rate along the membrane channel (Wilf, 1997; Nemeth, 1998). It was reported that the flux decreased rapidly along the membrane channel and the last few membrane elements in a long pressure vessel do not contribute much to the permeate production.

Effective fouling characterization

It demonstrated that when a full-scale RO was operated under thermodynamic restriction, the average permeate flux was *not* affected by change in membrane resistance. A new method was proposed for a better fouling characterization that is valid for membrane fouling under any conditions.

Water fouling potential

The fouling strength of the feed water is usually quantified in numerical values with some parameters, such as SDI, MFI. It is desirable the parameter used to define water fouling potential can be used to predict the development of fouling in full-scale RO processes. For this purpose, water fouling potential, k, was defined by Equation (9):

$$k = \frac{R_t - R_0}{\int_0^t v(\tau)\mathrm{d}\tau} \tag{9}$$

where t is the filtration time, R_t and R_0 are the nominal membrane resistance at time t and 0 respectively, and τ is the dummy integration variable. The water fouling potential can be determined with a small piece of membrane in a lab filtration device in a few hours.

Modeling of fouling development in full-scale RO processes

When the feed water fouling potential is known, the membrane resistance in the whole membrane channel at any time can be calculated with Equation (10):

$$R(x,t) = R_0 + k\int_0^t v(x,\tau)\mathrm{d}\tau \tag{10}$$

Combined with Equations (2)–(5), the permeate flux in the membrane channel at any time can be fully determined. Therefore, the fouling development in a full-scale RO process can be well modeled. A sample calculation is shown in Figure 7.

Membrane fouling characterization

Fouling will inevitably result in increase in membrane resistance under any time and therefore change in membrane resistance can be a fault-free indicator for membrane fouling. That means if the membrane resistance keeps constant over operating period, it is safe to say there is no fouling. However, because membrane resistance is a distributive parameter that has different value at different location due to uneven membrane fouling, the direct use of the localized membrane resistance to indicate membrane fouling is not convenient and it is unpractical to find out membrane resistance at every point in the membrane channel. To overcome this difficulty, a lumped (collective) parameter, *filtration coefficient*, was defined

$$F = \frac{1}{u_0 H} \int_o^L \frac{dx}{R_m(x)} \tag{11}$$

where F is the filtration coefficient, u_0 is the feed flow velocity, H is the channel height, and $R_m(x)$ is the localized membrane resistance. For a given RO process, F is a function of membrane resistance solely. Any change of membrane resistance at any point of the membrane channel will be reflected in F.

With the filtration coefficient, the recovery can be linked to the net driving pressure with the following equation:

$$R = F(\Delta p - \Delta \pi) \tag{12}$$

Equation (12) showed that the filtration coefficient would be the slope when the recovery is plotted versus the driving pressure. Because the information of the localized membrane resistance is unavailable, the filtration coefficient usually cannot be calculated directly

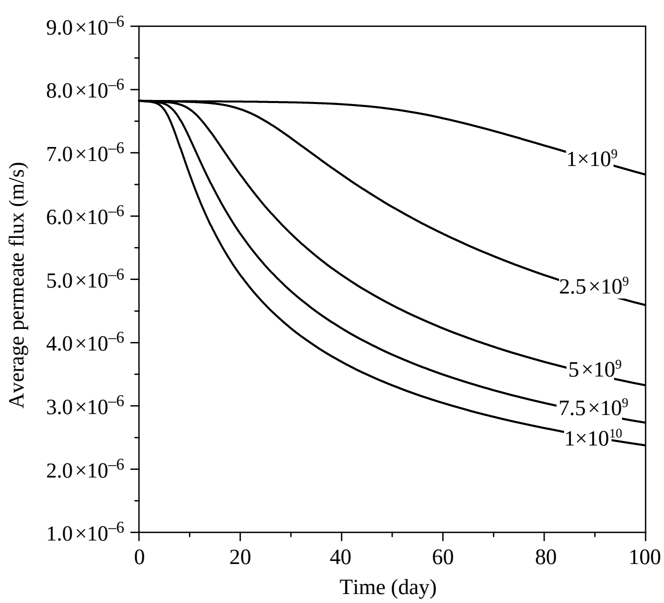

Figure 7 Change of average permeate flux in a 6-m membrane channel for different k-values

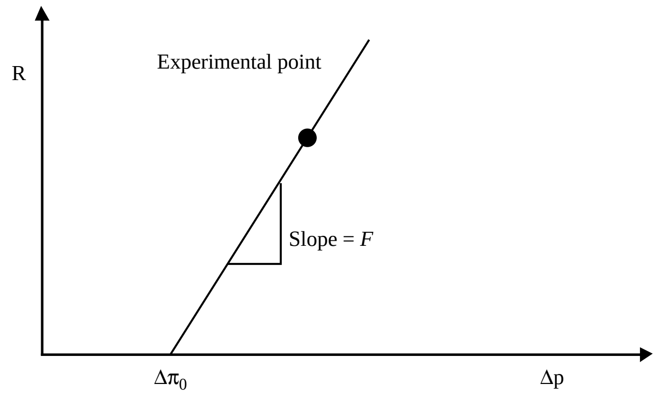

Figure 8 Filtration coefficient is the slope of the line of R vs Δp

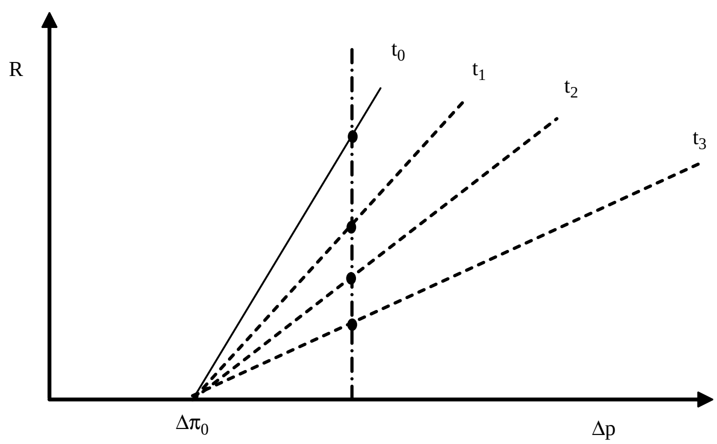

Figure 9 Fouling can be indicated by the straight lines with different slopes

from Equation (11). Instead, the filtration coefficient can be easily determined from Equation (12) with a simple pilot test, by finding the slope in the R vs. $(\Delta p - \Delta \pi)$ plot (Figure 8). (The testing has to be done in a low pressure such that the full-scale RO is operated in the mass transfer controlled regime.)

From Equation (9), it can be seen that the filtration coefficient F decreases as the overall membrane resistance increases. Therefore, membrane fouling in a full-scale RO process can be described by a series of straight lines with different slopes at different times (Figure 9). For a given driving pressure (the vertical line in the figure), recovery of the full-scale RO process (the intersection point of the recovery line with the vertical pressure line) decreases with time as membrane fouling is going on.

To our best knowledge, it is the first time that this complex but rather important fouling phenomenon in the full-scale RO processes is explained with the shifting between controlling mechanisms of the process. It can provide profound insights rather than phenomenological descriptions of the processes. For example, the use of filtration coefficient for the evaluation of cleaning efficiency can provide more accurate information than the use of average permeate flux.

Concluding remarks

One conventional practice in both membrane module and process designs is to facilitate the use of large membrane area. The spiral-wound RO modules squash as large as possible membrane area into a limited space. In the full-scale RO processes, long pressure vessels holding 6–7 modules are standard design and 2 or 3 stages of the long pressure vessels are connected in series. These measures were necessary for the old generations of RO membranes that had permeability orders smaller in magnitude than the membranes we are using currently. It can be proven that even with these measures to increase membrane areas for a given feed water supply, the processes with old generations of membranes are still controlled by mass transfer. This result justifies the design strategy as described above for the old generations of membranes.

With the emergence of the thermodynamic restriction with the new generation of highly permeable RO membranes, a new strategy of module and process design is required.

The emergence of thermodynamic restriction itself reveals there is more than needed membrane area in the processes. It is an indicator that the capacity of RO membranes has not been fully utilized because there is not sufficient feed water supply to the entire membrane. This is evidenced by the hydraulic imbalanced problem as reported in the literature. It was reported with the highly permeable RO membranes that the last few modules do not have water to produce because the net driving force has been significantly reduced. This problem can be improved by increasing the channel height and reducing the total membrane area in the modules.

It seems that the module and process design is a bottleneck for further improvement of membrane permeability. If the design cannot even utilize the current capacity of the RO membranes, there is no need to further improve the membrane permeability. A new design strategy must be developed to fully use up the capacity of the current RO membranes and leave some room for further improvement.

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