ORIGINAL ARTICLE

Construction of a dead-end type micro- to R.O. membrane test cell and performance test with the laboratory-made and commercial membranes

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Abstract

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Construction of a dead-end type micro- to R.O. membrane test cell and performance test with the laboratory-made and commercial membranes

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A dead-end type membrane stirred cell for an RO filtration test has been designed and constructed. Magnetic stirring system is applied to overcome a pressure-induced concentration polarization occurred over a membrane surface in the test cell. A high pressure N_2 tank is used as a pressure source. Feed container is designed for 2.5 ℓ feed solution and a stirred cell volume is 0.5 ℓ. The test cell holds a magnetic stirrer freely moved over the membrane surface. All units are made of stainless steel. A porous SS316L disc is used as a membrane support. The dead-end stirred cell is tested to work properly in an operating pressure ranged 0 - 400 psi. It means that the dead-end cell can be used to test a membrane of different filtration modes, from micro- to Reverse Osmosis filtration. Tests performed at 400 psi for 3 hours are safe but tests at a 500 psi increase leakage possibility.

The cell is used to test the performance of both commercial and laboratory-made membranes. It shows that the salt rejection efficiency of the nano- and RO membranes, NTR759HR and LES90, determined by using the new test cell, is closely similar to those reported from the manufacture. Result of the tests for our own laboratory-made membrane shows a similar performance to the nanofiltration membrane LES90.

Key words : dead-end stirred cell, performance test, nanofiltration, R.O., CA membrane

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Membrane filtration processes can be classified into 4 categories according to the operating pressure. They are (1) microfiltration (MF) for the operating pressure of 1-5 bar; (2) ultrafiltration (UF) for 2-10 bar; (3) nanofiltration (NF) for 10-20 bar; and (4) Reverse Osmosis filtration (RO) for 20-100 bar. Reverse Osmosis (RO) is a general process employed for the separation of substances in fluid mixtures. The substances constituting a solution are allowed to permeate through an appropriate membrane under a high pressure. Due to the principles of RO are derived from a variety of diverse area so that in the present time the applications of RO are widely used in diverse fields like desalination, chemical processing, pollution control, gas separations, food processing, medicine etc. (Winston and Kamalesh, 1992)

In the present time, the authors and coworkers have developed the CA membrane for NF and RO filtration since the first paper of Bhongsuwan (1998) has been presented. A tangential flow filtration test cell or cross flow test cell is designed and constructed by the authors and coworkers at the Membrane Science and Technology Research Center, Faculty of Science, Prince of Songkla University. This cell is needed for a performance test of the membrane developed.

However, by experience with our low-cost cross-flow membrane test system, the disadvantage of the system is that it needs a large volume of feed solution approx. 50 ℓ, which limits the system to study a filtration performance of only inexpensive solutes like NaCl solution. A dead-end membrane test cell is the best solution in this respect as a dead-end cell would need only a few litres of solutes at once. Preparation of different feed solution of expensive or toxic solutes can be possible at all levels. However, for a dead-end membrane test cell a concentration polarization must be taken care of.

Membrane filtration configuration

Membrane filtration configuration can be classified into 2 types. They include (1) Single Pass or Direct Flow Filtration (DFF) or Dead-end Filtration (Figure 1a) and (2) Tangential Flow Filtration (TFF) or Cross-flow Filtration

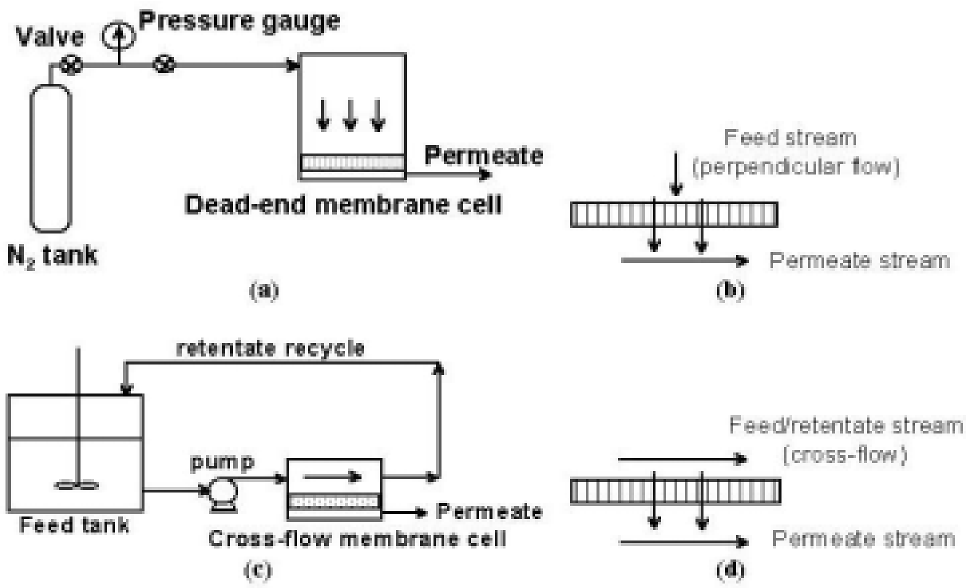

Figure 1. Diagrams showing components of (a) dead-end (c) cross-flow membrane filtration test system, (b) and (d) showing a direction of perpendicular flows (dead-end) and tangential (cross-flow), respectively. (modified from Abatemarco, *et al*, 1999)

(Figure 1c)

The main point of a dead-end filtration is to arrange the flow direction of feed solution perpendicular to the membrane surface (Figure 1b). The permeate solution passes directly through the membrane. The problem encountered in DFF is clogging, which is due to molecules larger than the membrane pore size attached the membrane surface at a high pressure. Clogging will reduce permeate flux and rejection efficiency. A cross-flow filtration arranges the flow direction of feed solution parallel to the membrane surface (Figure 1d) and generates turbulent flow in feed solution. This will prevent large molecules from touching the membrane surface and thereby prohibit clogging.

Concentration polarization is the main source of clogging on membrane surface. When the concentration of solute increases at the membrane surface, it reduces the solubility of solute and then precipitation of solute starts to clog the membrane. Concentration polarization and clogging of the membrane surface can be reduced in a cross-flow system by introducing a sufficient cross-flow velocity of feed solution. This is needed to induce a turbulence flow of feed solution over the membrane surface. For a dead-end system, a magnetically stirred system is needed in order to obtain the turbulence of feed solution over the membrane surface.

This work aims to design and construct a dead-end type RO membrane test cell with stirring system attached and use it to perform the membrane performance tests for commercial MF to RO membranes and also to test our own built CA membrane.

Materials and Methods

There are two parts of work in this study : (1) design and construction of a dead-end stirred cell, and (2) performance tests of the membranes both commercial and laboratory-made CA membranes.

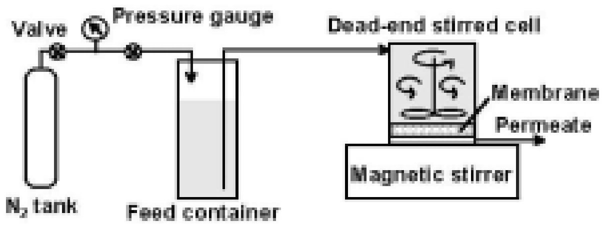

Figure 2. Simple diagram showing a dead-end stirred cell and its components.

Design and construction of a dead-end stirred cell

Figure 2 shows a simple diagram of a dead-end stirred system, which is constructed in this study. A high pressure N_2 tank is used as a pressure source, which is adjustable between at least 0 - 15 MPa in the range of MF to RO filtration. Feed container is designed for feed solution of up to 2.5 ℓ. Stainless-steel is used in all components to ensure a corrosion-free especially for salt separation study. The stirred cell made of stainless steel is about 0.5 ℓ containing a stirring system, which controls a magnetic stir bar to rotate freely above the membrane surface. A porous SS316L disc allows the permeate solution to pass through and supports the membrane that is not to be destroyed at a high operating pressure. All tubes (1/4" diameter) and tube fitting are made of stainless steel. A magnetic stirrer (Jenway model 1000, England) is used to stir the feed solution in the dead-end stirred cell. An electronic balance (Ohaus, Adventurer, USA) is used to weight the permeate solution continuously throughout the operating time.

Performance tests of the membranes using a newly constructed dead-end stirred cell

The membranes used in performance tests using the newly constructed dead-end stirred cell include the commercial MF and RO membrane, i.e., NTR7450, NTR759HR and LES90 (Somicon, 2002) and our own laboratory-made cellulose acetate (CA) membrane coded R130. Membrane R130 is produced by casting a CA solution on a glass plate to a thickness of ca. 0.150 mm. CA membrane solution is made of a mixture of 22% CA (Eastman, USA), 48 % acetone (Merck, Germany) and 30 % formamide (Merck, Germany). Parameters for the performance tests of the membranes include (1) pure water flux test and (2) permeate flux and salt rejection efficiency of NaCl feed solution. The permeate or pure water flux J_w ($\ell/m^2/hr$) is derived from an equation

$$J_w = \frac{Q}{A\Delta t} \tag{1}$$

where Q is a permeate volume (ℓ) ; Δt is a time interval (hour); and A is an area of the tested membrane (m^2). This test cell has a fixed area of a tested membrane of A = 0.0022911 m^2.

Salt rejection efficiency is present in percent called percent salt rejection (%SR), which is calculated from the equation :

$$\%SR = 1 - \frac{C_p}{C_f} \times 100 \tag{2}$$

where C_P and C_f are NaCl concentrations in permeate and feed solution, respectively.

In this study %SR is determined using conductivity of feed and permeate and is calculated by the equation :

$$\%SR = 1 - \frac{Conductivity_{Permeate}}{Conductivity_{Feed}} \times 100 \tag{3}$$

A conductivity meter (WTW model Inolab Cond Level 2, USA) is used to measure conductivity and TDS of feed and permeate solution in this study.

Results and Analysis

Construction of the high pressure test cell

The system components (Figure 3) includes (1) a high pressure N_2 tank 0 - 1500 psi; (2) SS-316L $^1/_4$" tube for high pressure gas and liquid; (3) 2.5 ℓ feed container and (4) a dead-end stirred cell. A 2.5 ℓ cylinder feed container made of stainless steel is 31.8 cm height and 10.2 cm inner diameter. Feed solution is under a high pressure of N_2 supplied from the tank. This pressure drives feed solution from feed container to the dead-end cell. The dead-end stirred cell is made of stainless steel in cylindrical shape of 10.3 cm height and 5.4 cm internal diameter, 0.5 ℓ capacity. The stirred cell is equipped with a magnetic stir holder, which holds a stir bar to move freely over membrane surface. A porous SS316L disc (5.7 cm ϕ and 0.3 cm thickness) is used as a support for the membrane. Permeate passing through the membrane and support is drained to a small 50 ml beaker placed on an electronic balance.

Pure water flux test

Pure water flux or PWF tests of different membranes are performed using the newly con-

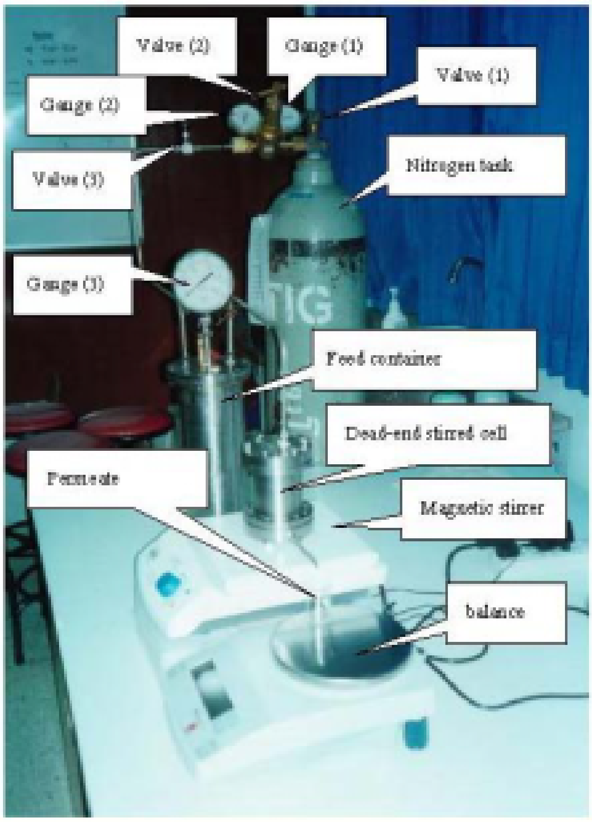

Figure 3. Photograph of the constructed dead-end stirred cell and other components of the system.

structed dead-end stirred cell. Distilled water is used as feed solution. Pure water flux can be used as a reference flux for comparison. Due to purity of distilled water (M.W. = 18) clogging and concentration polarization are not expected to occur on the membrane surface.

PWF result for membrane NTR7450 is shown in Figure 4a. PWFs are 8.36, 18.01, 29.33 and 38.95 ℓ/m^2.hr at a pressure of 25, 50, 75 and 100 psi, respectively. NTR7450 membrane is a nanofiltration membrane, which has a high flux in a pressure of 0 - 100 psi. Membrane NTR759HR shows PWFs between 8 - 39 ℓ/m^2.hr for a range of pressure of 100 - 400 psi (Figure 4b). NTR759HR is a RO membrane, which is normally operated at a high pressure. Flux during 0 - 30 minutes of operation is almost constant. LES90 is a nano-filtration membrane, which has a PWF of 4 - 37 ℓ/m^2.hr when increase a pressure from 100 - 400 psi (Figure 4c). The PWF is almost constant during 0 - 30 minutes at a pressure of 100 - 400 psi. Our own laboratory built CA membrane, R130, shows the PWFs of between 4.66 - 19.23 ℓ/m^2.hr (Figure 4d) for an operating pressure 100- 400 psi and the fluxes are nearly constant during an operating time of 0 - 30 minutes as well.

Imperfect stirring system design and influence of concentration polarization to the membrane performance

To demonstrate the influence of concentration polarization effect due to the dead-end type design geometry of the test cell, we present the result in case of imperfectly constructed stirring system of our dead-end stirred cell. In this experiment a NaCl 3000 ppm solution is used as a feed solution. Operating temperature is the room temperature at 25°C. Performance of the membrane in term of salt rejection is to be determined together with a permeate flux through the membrane. NaCl molecules in the feed solution are large enough to induce a clogging of the membrane when a concentration polarization occurs over the membrane surface under operation. Those behaviors will affect the flux and salt rejection efficiency of the membrane at higher pressure.

The RO membrane NTR749HR gives a permeate flux starting at a threshold pressure of 100 psi. The fluxes at a pressure 100 - 400 psi are between 4.7 - 6.0 ℓ/m^2.hr. Flux-pressure relation is not linear but curved as shown in Figure 5a. Permeate fluxes are generally low for a RO-type membrane, which is needed to be operated at a higher pressure. Decrease of permeate flux during the operating time of 0 - 60 minutes probably indicates the existence of clogging effect at membrane surface due to a gel layer formed at membrane surface when solubility of solute NaCl is reduced. Percent salt rejection at pressures 200, 300 and 400 psi decreases from 84.9 to 61.3 and 41.7, respectively (Figure 5b). Membrane R130 gives a flux of 5.1 - 19.7 ℓ/m^2.hr at a pressure 100 - 400 psi (Figure 5c). Fluxes are nearly constant for an operating time 0 - 60 minutes. The %SR of 81 is observed at 100 psi and 70 at 400 psi (Figure 5d).

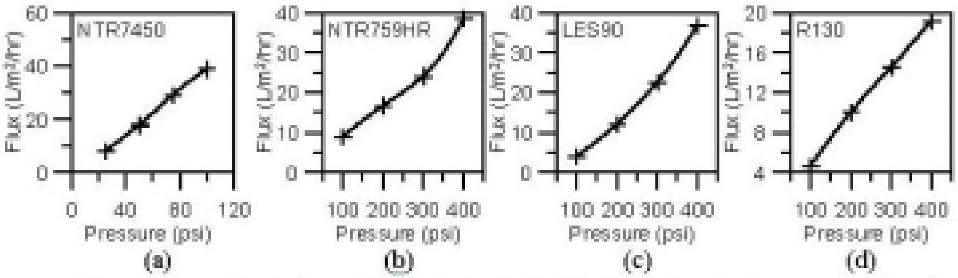

Figure 4. Pure water flux of tested membranes at different operating pressure using the new constructed dead-end stirred cell, (a) NTR7450, (b) NTR759HR, (c) LES90, and (d) R130.

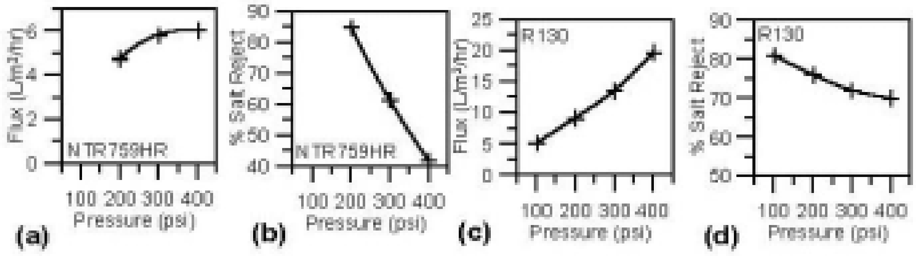

Figure 5. Permeate flux and salt rejection efficiency at different pressures for (a), (b) membrane NTR759HR and (c), (d) membrane R130. The tests are performed at some degrees of concentration polarization in a NaCl 3000 ppm feed solution.

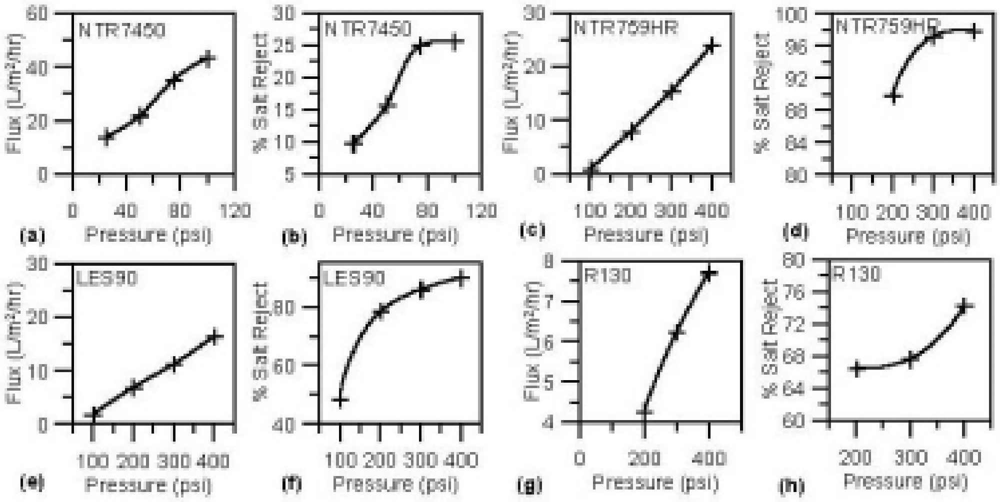

Figure 6. Permeate fluxes and salt rejection efficiencies at different operating pressure using the dead-end stirred cell for (a), (b) membrane NTR 7450; (c), (d) membrane NTR759HR; (e), (f) membrane LES90; and (g), (h) membrane R130. Feed solution is a NaCl 3,000 ppm solution.

This behavior clearly indicates the concentration polarization effect, which is still dominant at solution-membrane interface due to imperfect former design of stirring system. Concentration polarization is caused by the accumulation of solute (NaCl) on the membrane or in the boundary layer adjacent to the membrane surface. This NaCl concentration will be highest at the membrane surface, and decreases exponentially toward the feed solution. Imperfect former design of the stirring system in this study cannot properly induce a well-mixed solution and then the concentration polarization is still dominant. A modification of stirring system must be made.

Membrane performance tests using a dead-end with an improved stirring system

This test is performed after a modification of stirring system is complete. Permeate fluxes and salt rejection efficiencies are the parameters of different membranes to be tested. Feed solution is NaCl 3000 ppm solution. Operating temperature is the room temperature at 25°C.

Result shows that at 25 - 100 psi operating pressure, membrane NTR7450 has a flux increasing from 14 to 43 ℓ/m^2.hr (Figure 6a). Flux is almost constant during operating time 0 - 30 minutes. It is noted that a pure water flux (38.95 ℓ/m^2.hr) is comparable to salt water flux (43.41 ℓ/m^2.hr) at the same pressure of 100 psi. This is probably due to a problem in uniformity of the membrane surface cut at different positions of a sheet. The salt rejection (%SR) at pressure 25, 50, 75 and 100 psi is 9.72, 15.73, 24.94 and 25.60, respectively (Figure 6b). It shows a higher %SR at higher pressure. This probably indicates that stirring system can (almost) completely overcome the concentration polarization effect. However, %SR determined using the newly constructed dead-end stirred cell shows the %SR less than that reported by the manufacture (40%SR) (Somicon, 2002).

RO-type membrane NTR759HR is tested for operation pressure 100 - 400 psi. Fluxes increase linearly with pressure and range between 0.89 - 24.04 ℓ/m^2.hr (Figure 6c). Permeate fluxes are constant for an operating time 0 - 30 minutes. Pure water flux at a 400 psi (38.70 ℓ/m^2.hr) is higher than a flux for NaCl feed solution (24.04 ℓ/m^2.hr) at the same pressure. This indicates an influence of clogging and concentration polarization for NaCl feed solution. The %SRs at 200, 300 and 400 psi are 89.89, 97.26 and 97.93, respectively (Figure 6d). They are comparable to the certified values (99 - 99.5%) reported by the manufacture (Somicon, 2002). It is observed that salt rejection increases as a function of pressure. This also indicates that the stirring system is improved and can overcome the concentration polarization at membrane surface.

Membrane LES90 is tested at pressures 100 - 400 psi. Permeate fluxes increase linearly with pressure and range between 1.8 and 16.43 ℓ/m^2.hr (Figure 6e). Pure water flux at 400 psi (36.95 ℓ/m^2.hr) is higher than the flux for NaCl feed solution (16.43 ℓ/m^2.hr) at the same pressure. The %SRs change from 48.66 - 89.97 for the pressures of 100 - 400 psi (Figure 6f). These values are comparable to those reported by the manufacturer (90-95 %SR).

Membrane R130, our own laboratory-made membrane, is tested at pressures 100 - 400 psi. Permeate fluxes increase linearly with pressure and are between 4.27 and 7.72 ℓ/m^2.hr (Figure 6g). Fluxes measured during an operating time 0 - 110 minutes are nearly constant. Pure water flux at 400 psi (19.23 ℓ/m^2.hr) is almost three times higher than the flux for NaCl feed solution (7.72 ℓ/m^2.hr) at the same pressure. The %SRs vary from 66.49 to 74.29 for the pressures of 100 - 400 psi (Figure 6h).

Membrane performance tests using a cross-flow test cell

A cross-flow test cell is used to perform the membrane performance tests in order to compare with the results from a new constructed dead-end stirred cell. Feed solution used in the cross-flow tests is a NaCl 3000 ppm solution, similar to what we have used in dead-end tests. Operating temperature is the room temperature at 25°C.

Membrane NTR7450 gives a flux between 40.33 and 362.36 ℓ/m^2.hr in a range of operating pressure 50 - 400 psi (Figure 7a). The %SR ranges of 25.98 - 10.20 at the same range of operating pressure (Figure 7b). The %SRs are comparable for a pressure 100 -200 psi, but decrease at higher pressure 300, 400 psi. Decrease of %SR probably indicates a concentration polarization when the tangential flow velocity over the membrane surface is so low, especially at high pressure above 200 psi.

Membrane NTR759HR gives a flux increasing from 3.8 and 31.56 ℓ/m^2.hr when operating pressure increases from 100 - 400 psi (Figure 7c). The %SRs of 91.24 - 95.10 are obtained at the same range of operating pressure (Figure 7d), but are nearly constant for a range of 200 to 400 psi. Membrane LES90 shows a progressive increase of flux from 6.01 and 17.69 ℓ/m^2.hr when operating pressure increases from 200-400 psi (Figure 7e). The %SRs of 86.84 - 87.13 are obtained at the same operating pressures (Figure 7f).

A laboratory-made membrane R130 gives an increasing flux from 4.5 to 15.23 ℓ/m^2.hr in a

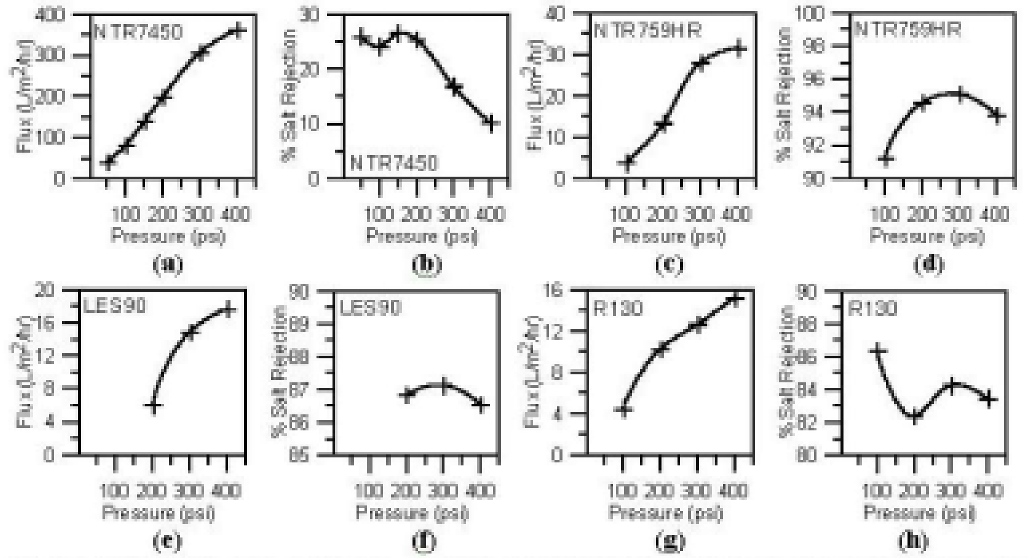

Figure 7. Permeate flux and %SR versus pressures resulted from membrane performance tests using a cross-flow test cell. (a) (b) membrane NTR7450; (c) (d) membrane NTR759HR; (e) (f) membrane LES90; and (g) (h) membrane R130. (Feed is a NaCl 3000 ppm solution)

range of operating pressure 100 - 400 psi (Figure 7g). The %SRs range from 82.37 to 86.37 at the same range of operating pressure (Figure 7h), and are comparable for a pressure 200 - 400 psi.

Conclusions

It is already known that water flux depends on an area of membrane (A), pressure difference at both sides of the membrane (ΔP) and osmotic pressure difference (Δπ) between feed and permeate solutions. They follow the equation :

$$F_w = A(\Delta P - \Delta \pi) \quad (4)$$

This shows that flux increases linearly when ΔP - Δπ increases. Results of flux tests in this work follow this relation quite well. However, at high ΔP, flux tends to be flattened or saturated, as seen in Figures 5a, 7c, which is probably due to mechanical deformation of the membrane at high pressure. This effect results in closing of several pores of the membrane.

In the case of salt rejection, an ideal membrane should allow only pure water to pass through it but it is not true for actual membrane. This means that permeate solution must always contain some impurities. In the case of NaCl feed solution, salt rejection should be higher by increasing pressure. This is because increase of pure water flux must be higher than salt flux at a given pressure due to the size of molecules. In our case, dead-end filtration test cell, if stirring system cannot overcome the polarization, salt rejection will be lower at higher pressure as seen in Figures 5b, 5d.

Results show that pure water flux increases when operating pressure increases. The newly constructed dead-end stirred cell can be operated properly at a pressure between 0 - 400 psi for at least 3 hours continuously. It means that the test cell can be used to test a full range of filtration membrane from MF to RO. Operation performed at 500 psi increases the possibility of leakage.

An increase of operating pressure in membrane filtration does not affect the solute diffusion through a membrane but increases the permeate flux, which results in an increase of the salt rejection efficiency. However, increase of operating pressure may cause a compression of membrane that lowers its permeate flux apart from a linear increase at high pressure as shown in Figures 5 and 6 and found in both cases of imperfect and perfect stirring systems dead-end test cell. The salt rejection derived from cross-flow tests is comparable at a pressure of 100 - 200 psi, but reduced with pressure for 300 - 400 psi. This clearly indicates an effect of cross-flow or tangential-flow velocity of feed solution over a membrane in operation. At pressures of 300 and 400 psi, the cross-flow velocity (85.7 and 7.9 ℓ/hr, respectively) is probably so low and that the concentration polarization survives. Stirring system in the newly constructed dead-end test cell acts as a high cross-flow velocity in a cross-flow test cell in order to reduce the concentration polarization over the membrane surface.

Salt rejection efficiency (% SR) is lower at a higher pressure for an imperfect stirring system operation, which is affected by a concentration polarization near the membrane surface.

In a perfect stirring operation the %SR increases when the pressure increases. Flux and %SR of the studied membranes tested with a perfect stirring dead-end operation are comparable with those determined from the cross-flow tests and certified values reported by the manufacture (Table 1).

Table 1. comparison of the %SR of the dead-end stirred and cross-flow tests with certified values.

Membrane	Certified value (%SR)	Result of this study	
		Dead-end (%SR)	Cross-flow (%SR)
NTR7450	40 - 50	9.7 - 25.6	10.2 - 26.6
NTR759HR	99 - 99.5	89.9 - 97.9	91.2 - 95.1
LES90	90 - 95	48.7 - 90.0	86.5 - 87.1
R130	-	66.5 - 74.3	82.4 - 86.4

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References

Abatemarco, T., Stickel, J., Belfort, J. and Frank, B.P. 1999. Fractionation of multiwalled carbon nanotubes by cascade membrane microfiltration. J. Phys. Chem. : B, 103 : 3534-3538.

Bhongsuwan, D. 1998. Production of reverse osmosis membrane from cellulose acetate. Songklanakarin J. Sci. Technol., 20(4) : 477-484.

Somicon, AG. 2002. Nitto Denko Membrane. Http://www.somicon.com/products/ Products_Nitto_files/Nitto_Specs/

Winston, W. S. and Kamalesh, K. S. 1992. Membrane Handbook. Van Nostrand Reinhold, p. 266.