Forward and Reverse Osmosis Process for Recovery and Re-use of Water from Polluted Water by Phenol

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ABSTRACT

The research aims to apply the novel forward osmosis (FO) process to recover pure water from contaminated water. Phenol was used as organic substance in the feed solution, while sodium chloride salt was used as draw solution. Membranes used in the FO process is the cellulose triacetate (CTA) and polyamide (thin film composite (TFC)) membrane. Reverse osmosis process was used to treatment the draw solution, the exterior from the forward osmosis process. In the FO process the active layer of the membrane faces the feed solution and the porous support layer faces the draw solution and this will show the effect of dilutive internal concentration polarization and concentrative external concentration polarization.

In the FO process was a run-time for five hours, and the concentration of phenol 100 and 1000 mg/l, and for the NaCl the concentration was 10000 and 30000 mg/l. It was found that recovery percent increases with increasing time, while water flux through membrane decreases with increasing time. Also, it was found that recovery and water flux increases with increasing draw solution concentration, on the contrary, water flux and the percentage of recovery decreases with increasing the concentration of phenol (feed solution). Increase in draw solute (NaCl) concentration has more effect on the water flux in FO process compared with increase in the concentration of phenol. Outlet phenol concentration increases with time, while the outlet salt concentration decreases with increasing the time. The results showed that the cellulose triacetate membrane gave the highest recovery ratio from the thin film composite membrane. The highest recovery was reached in five hours is 51.33%, while using CTA membrane recovery rate increase, by 23% compared with TFC membrane. The value of the resistance to solute diffusion within the membrane porous support layer is 36.83 h/m. Reverse osmosis is perfect method for removal of dissolved salts from water, thus its suitable process for reducing the content of NaCl in draw solution; therefore the sodium chloride rejection percentage was 91.6 – 96 % for polyamide membrane (TFC). Within two hours of work of the reverse osmosis system the recovery percentage of pure water is 58%.

(FO)   .

.

.((TFC)   )   (CTA)

.

.

\   1000   100

.   \   30000   10000   NaCl

.

(NaCl)   .(   )

                    .

%51,33   .

                    .TFC   %23   CTA

                    . \   36،83

                                        .(TFC)   % 96 – 91,6

                                                                    . 58

Keywords: Forward Osmosis; Reverse Osmosis; Recovery of Water; Wastewater; Phenol; Membranes.

INTRODUCTION

The problem of considerable contamination of the aqueous environment with organic pollutants still requires the development of quick and simple methods for the removal, separation and determination of these compounds. The main classes of organic compounds that most of the industries use and discharge into the effluents is phenol, surfactant and dye. All these compounds are troublesome contaminants which pose not only to toxicity and health hazards but also hamper the environmental treatment processes (John et al., 2005). In particular, the removal of phenol is of great interest in wastewater treatment. With a global production of 8 million tons nearly each year, phenol is one of the most important intermediates in chemical industry. Phenol contaminated effluents arise, for example, during the production processes of bisphenol A, phenol formaldehyde resins, and the Hock process (Kujawski et al., 2004). Methods for the recovery of phenols include membrane processes (Ray et al., 1997 and Hoshi et al., 1997), solvent extraction (Krishnakumar and Sharma, 1984 and Shejiao et al., 2001), activated carbon and polymer adsorption (Bercie et al., 1996).

Water scarcity problems in recent years and ground water contamination due to floods have been increasing alarmingly. The aim of wastewater treatment cannot be limited merely on achieving permissible discharge limits; rather its objectives should also focus on possible recycle options within the treatment schemes (Mahesh and Sukumar, 2008). Membrane separation processes are quite useful in concentration, separation and purification. So far, the most widely used membrane processes for water treatment include microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO). All of these are pressure-driven processes, which require energy to pressurize the system. While pressure-driven membrane processes, such as reverse osmosis, have dominated for several decades, new desalination processes are now appear, such as forward osmosis (FO) (Ahmed, 2007 and Hasan 2008).

Reverse osmosis (RO) is a technology that uses pressure to separate the salt (as NaCl) from the water and is capable of reducing water salinity. With this technology, the pressure is increased above the osmotic pressure, which allows the water to pass through semi-permeable membranes, but retains the solid salt particles (George, 1997 and Sourirajan, 1970). The reverse osmosis technology was introduced in the 1970's. After the multi-stage flash distillation (MSF)

technology, it is the most-used technology in all the Gulf countries. Reverse osmosis is considered the best alternative to distillation, due primarily to its low energy consumption, low deposition rate, smaller required space compared to other desalination facilities, and lower costs. This technology is generally very suitable for desalinating brackish water. Most currently available RO membranes fall into two categories: asymmetric membranes containing one polymer, and thin-film composite membranes consisting of two or more polymer layers. There are four main types of membrane modules: plate-and-frame, tubular, spiral wound, and hollow fiber. The most popular module in industry for nanofiltration or reverse osmosis membranes is the spiral wound module (Merten, 1966 and Schutte, 2003). The main advantage of this design is the large surface area of the membrane that is packaged into a relatively small volume of the cylindrical membrane element. The efficiency of water production with this method depends on the salinity level of the feed water and the number of desalination stages (number of membranes). Most reverse osmosis membranes allow less than 1% of the salt content in a single stage. With additional costs, the salinity can be further reduced with second-stage membrane desalination (Ahmed, 2000 and James et al., 2010).

Forward (or direct) osmosis is a process in which osmosis is used to pull water from feed solution (wastewater), through a semi-permeable membrane, into an osmotic agent solution (draw solution or $NaCl – H_2O$ solution in this search). The semi-permeable membrane is permeable to water, but essentially impermeable to other species. Osmotic transfer of water from the feed solution into the draw solution occurs because of the high concentration of dissolved solids maintained in the draw solution (Kravath and Davis, 1975). This gives the draw solution a very high osmotic potential, causing it to pull water through the membrane from the feed. Forward osmosis membranes reject organics, metals and other solids similar to reverse osmosis but resist typical fouling problems (Holloway et al., 2007).

Forward osmosis has several unique benefits of technology. Firstly, FO process overcomes fouling, comparison with other membranes separation processes (such as RO, NF, UF, and MF). Secondly, FO can be treated different feed streams; the water source is dirty or contaminated, which contains high levels of suspended solids (Mi and Elimelech, 2010). Thirdly, in membranes separation processes which using pressure as driving force; all components of a feed are compulsorily forced against the membrane surface. Fourthly, in FO process using different concentration on sides of membrane surface as driving force, this leads to selectively draws molecules through the membrane avoiding membrane fouling and compaction (Yip et al., 2010). And finally, forward osmosis is a process normally occurring in nature, so, requires little or no electricity or external power source (i.e. low cost). The draw solution solute (or osmotic agent) must have very specific characteristics such as highly soluble in water and low molecular weight; from these characteristics obtain a high osmotic pressure which it leads to higher water flux and feed water recovery. Also, osmotic agent solution solute must be nontoxic and probably the solute is edible in some cases. Ideal draw solution does not interact with the membrane or degrade the membrane. It should be noted, the solute in a draw solution non-edible must be separated easily and economically to be used again (McCutcheon et al., 2006).

The present study includes two parts, the first stage application novel method (forward osmosis) to recovery of water from wastewater by phenol. Membranes used in the paper are cellulose triacetate (CTA) and thin film composite (TFC) membranes. Draw solution that was used is sodium chloride solution ($NaCl - H_2O$) because it has an ideal specification for draw solution solute. The effect of time, feed solution concentration and draw solution concentration for two types of membranes on recovery percentage, water flux, outlet concentration of phenol and sodium chloride have been determined. The second stage, a technically viable reverse osmosis process has been employed to treat the draw solution outlet from forward osmosis

process, and then recycle draw solution to FO process to be used again.

THEORY AND METHODS

Concentration polarization is a significant problem in pressure-driven membrane desalination processes and has thus been the target of several investigations. Concentration polarization takes place when species that are retained or rejected by the membrane accumulate at the membrane surface. The concentration increases as they approach the membrane surface. The effect of this accumulation depends on the solute concentration (International Atomic Energy Agency, 2004 and Baker, 2004). Below, these two concentration polarization phenomena are quantitatively described.

External Concentration Polarization

Concentrative external concentration polarization occurs in forward osmosis when the feed solution is placed against the active layer of the membrane. To account for this phenomenon, the extent of concentration polarization was calculated from film theory. The Sherwood number, Sh, was first determined by using either the laminar or turbulent flow correlation for a rectangular channel (McCutcheon and Elimelech, 2006):

$$sh = 1.85 \left(Re \, Sc \, \frac{d_h}{L} \right)^{0.33} \text{ (Laminar Flow)} \qquad (1)$$

$$sh = 0.04 \, Re^{0.75} \, Sc^{0.33} \text{ (Turbulent Flow)} \qquad (2)$$

Here, Re is the Reynolds number, Sc is the Schmidt number, d_h is the hydraulic diameter, and L is the length of the channel. The mass transfer coefficient, k, is related to Sh by

$$k = \frac{Sh \, D}{d_h} \qquad (3)$$

Where D is the solute diffusion coefficient. The concentrative external concentration polarization moduli at each permeate flux, J, could be calculated using

$$\frac{\pi_{F,m}}{\pi_{F,b}} = \exp\left(\frac{J}{k} \right) \qquad (4)$$

Where J is the experimental permeate water flux, and $\pi_{F,m}$ and $\pi_{F,b}$ are the osmotic pressures of the feed solution at the membrane surface and in the bulk, respectively. Note that the exponent is positive, indicating that $\pi_{F,m} > \pi_{F,b}$.

The draw solution in contact with the permeate side of the membrane is being diluted at the permeate–membrane interface by the permeating water (Moody and Kessler, 1976). This is called dilutive external CP. Both concentrative and dilutive external CP phenomena reduce the effective osmotic driving force. A dilutive external CP modulus can be defined as above, except that in this case, the membrane surface concentration of the draw solute is less than that of the bulk (i.e. $\pi_{D,b} > \pi_{D,m}$) (Cath et al., 2006):

$$\frac{\pi_{D,m}}{\pi_{D,b}} = \exp\left(-\frac{J}{k} \right) \qquad (5)$$

Where $\pi_{D,m}$ and $\pi_{D,b}$ are the osmotic pressures of the draw solution at the membrane surface and in the bulk, respectively.

To model the flux performance of the forward osmosis process in the presence of external concentration polarization, we start with the flux equation for forward osmosis, given as

$$J = A \left(\pi_{D,b} - \pi_{F,b} \right) \qquad (6)$$

Here, A is the pure water permeability coefficient. We assume that salt does not cross the membrane, the osmotic reflection coefficient (σ), has a value of 1. Equation 6 predict flux as a function of driving force only in the absence of concentrative or dilutive external concentration polarization, which may be valid only if the permeate flux is very low. When flux rates are higher, this equation must be modified to include both the concentrative and dilutive external concentration polarization:

$$J = A \left[\pi_{D,b} \exp\left(-\frac{J}{k} \right) - \pi_{F,b} \exp\left(\frac{J}{k} \right) \right] \qquad (7)$$

Figure 1a show this phenomenon with a dense symmetric membrane (McCutcheon and Elimelech, 2006).

Internal Concentration Polarization

If the porous support layer of an asymmetric membrane faces the feed solution, as in pressure retarded osmosis (PRO), a polarized layer is established along the inside of the dense active layer as water and solute propagate the porous layer (Figure 1b). This is referred to as concentrative internal concentration polarization, this phenomenon is similar to concentrative external concentration polarization, except that it takes place within the porous layer, and therefore, cannot be minimized by cross flow. Lee et al., (1981) derived an expression modeling this phenomenon in pressure retarded osmosis, which Loeb et al., (1997) later described for osmosis. This equation describes internal concentration polarization (ICP) effects and how they relate to water flux (J), salt permeability coefficient (B) and water permeability coefficient (A):

$$K=\left(\frac{1}{J}\right)\ln\frac{B+A\pi_{D,m}-J}{B+A\pi_{F,b}} \tag{8}$$

Where K is the resistance to solute diffusion within the membrane porous support layer, K is defined as

$$K=\frac{t\tau}{D\varepsilon} \tag{9}$$

Where D is the diffusion coefficient of the solute, and t, τ, and ε are the thickness, tortuosity, and porosity of the support layer, respectively. K is a measure of how easily a solute can diffuse into and out of the support layer and thus is a measure of the severity of ICP. We maintain the use of the K term due to convention established in previous studies on internal concentration polarization. Salt permeability coefficient (B) is negligible compared to the other terms in Equation 8. Therefore, we ignore salt flux in the direction of water flux and any passage of salt from the permeate (draw solution) side (Gray et al., 2006). Thus, flux can be solved for implicitly from Equation 8:

$$J=A\left[\pi_{D,m}-\pi_{F,b}\exp(JK)\right] \tag{10}$$

The exponential term in equation 10 is a correction factor that can be considered the concentrative internal concentration polarization modulus, defined as

$$\frac{\pi_{F,i}}{\pi_{F,b}}=\exp(JK) \tag{11}$$

Where $\pi_{F,i}$ is the osmotic pressure of the feed solution on the inside of the active layer within the porous support. The positive exponent indicates that $\pi_{F,i} > \pi_{F,b}$, or that the effect is concentrative. Substitute Equation 5 into 10 to obtain an analytical model for the effect of internal and external concentration polarization on water flux:

$$J=A\left[\pi_{D,b}\exp\left(-\frac{J}{k}\right)-\pi_{F,b}\exp(JK)\right] \tag{12}$$

All the terms in Equation 12 are readily determined through calculations or experiments. From equation we can calculate the water flux through an asymmetric membrane where the feed solution is placed against the support layer and the draw solution against the active layer.

In forward osmosis applications for desalination and water treatment, the active layer of the membrane faces the feed solution and the porous support layer faces the draw solution (Kessler and Moody, 1976). As water permeates the active layer, the draw solution within the porous substructure becomes diluted. This is referred to as dilutive internal concentration polarization (Figure 1c). Loeb et al., (1997) similarly described flux behavior in the forward osmosis mode:

$$K=\left(\frac{1}{J}\right)\ln\frac{B+A\pi_{D,b}}{B+J+A\pi_{F,m}} \tag{13}$$

When assuming that B = 0, σ = 0 (i.e., the salt permeability is negligible) and the equation 13 is rearranged, an implicit equation for the permeate water flux is obtained:

$$J=A\left[\pi_{D,b}\exp(-JK)-\pi_{F,m}\right] \tag{14}$$

Here, $\pi_{D,b}$ is now corrected by the dilutive internal concentration polarization modulus, given by

$$\frac{\pi_{D,i}}{\pi_{D,b}}=\exp(-JK) \tag{15}$$

Where $\pi_{D,i}$ is the concentration of the draw solution on the inside of the active layer within the porous support. The negative exponent because the water flux is in the direction away from the membrane active layer surface. In other words, the concentration polarization effect in our case is dilutive, meaning that $\pi_{D,i} < \pi_{D,b}$ by substituting Equation 4 into 14, we get

$$J=A\left[\pi_{D,b}\exp(-JK)-\pi_{F,b}\exp\left(\frac{J}{k}\right)\right] \qquad (16)$$

The terms in Equation 16 are measurable system conditions and membrane parameters. Note that here; dilutive internal concentration polarization is coupled with concentrative external concentration polarization, whereas in the Equation 12, concentrative internal concentration polarization was coupled with dilutive external concentration polarization.

In each of these cases, the external concentration polarization and internal concentration polarization moduli all contribute negatively to the overall osmotic driving force. The negative contribution of each increases with higher flux, which suggests a self limiting flux behavior. This implies that increasing osmotic driving force will provide diminishing increases in flux (Tang et al., 2010).

In this search for ideal state, assuming that the salt permeability coefficient (B) is equal to zero and the small value of the flux (J) compared to osmotic pressure of draw solution (π_{NaCl}), therefore the Equations 8 and 13 it can simplify as follows:

$$K=\frac{1}{J}\left(\ln\frac{\pi_{D,b}}{\pi_{F,b}}\right) \text{ or } J=\frac{1}{K}\left(\ln\frac{\pi_{NaCl}}{\pi_{phenol}}\right) \qquad (17)$$

Osmotic Pressure

Osmotic pressure magnitude is proportional to the amount of dissolved substances in the solution, dissociated ions per molecule and to the temperature of the solution, and is completely independent of the membrane. In 1886, van't Hoff formulated an equation to calculate osmotic pressure (π), based on data for sugar solution and the similarity of dilute solutions to ideal gases (Thain, 1967 and Ahmed, 2007):

$$\pi=\Phi\,i\,R_g\,T\,C \qquad (18)$$

Where C is the concentration of solute, T is the temperature of solution, R_g is the gas constant, i is number of dissociated ions per molecule, and Φ is osmotic coefficient.

Rejection Percentage

The measure of membrane selectivity is solute rejection, the ratio of solute rejected by a membrane to the solute in the feed. It is the most common method of evaluating a membrane's ability to separate solute, because the determination is simple and can be done as accurately in the field as in the laboratory (Hasan 2008 and Yip et al., 2010).

$$R=\left(\frac{C_F-C_P}{C_F}\right)\times100 \qquad (19)$$

Where C_F is the concentration of a specific component in the feed solution to the membrane process and C_P is the concentration of the same specific component in the product stream leaving the membrane system.

Recovery Percentage

The recovery factor measures how much of the feed is recovered as permeate. It is reported as a percentage (Ahmed, 2000). Recovery, or conversion, is defined by:

$$Y=\left(\frac{V_P}{V_F}\right)\times100 \qquad (20)$$

Where V_P is the permeate (or product) volume of water and V_F is the volume of water in feed vessel.
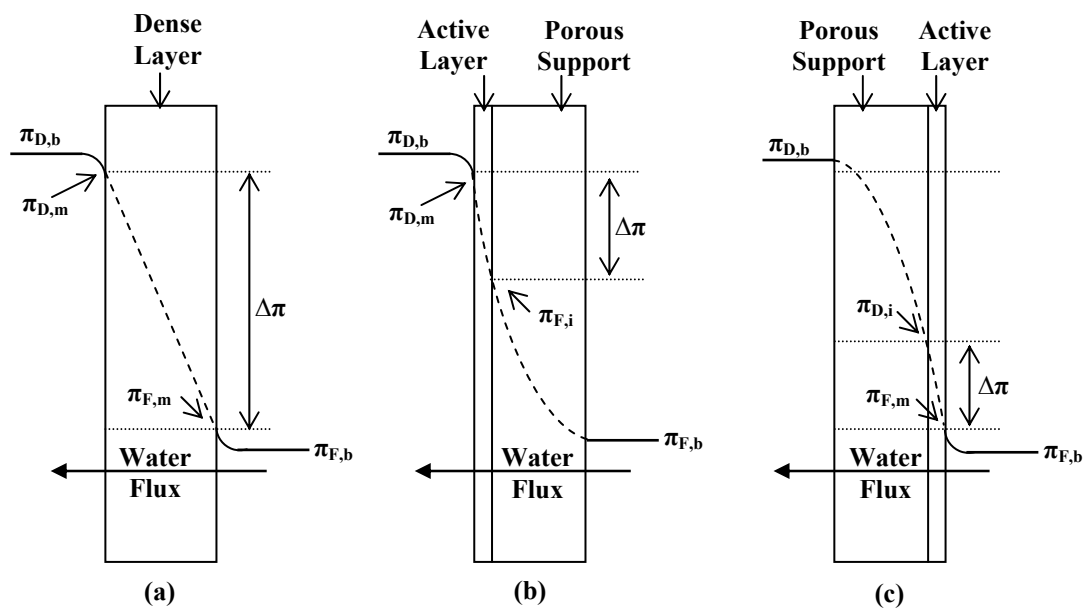
Fig. 1 Illustration of osmotic driving force profiles for osmosis through several membrane types and orientations, incorporating both internal and external concentration polarization. (a) The profile illustrates concentrative and dilutive external CP. (b) PRO mode; the profile illustrates concentrative internal CP and dilutive external CP. (c) FO mode; the profile illustrates dilutive internal CP and concentrative external CP (McCutcheon and Elimelech, 2006).

EXPERIMENTAL

Forward Osmosis

- **Draw and Feed Solutions**

The concentrated draw solution is made by dissolving sodium chloride salt (NaCl) in demineralized water, of 10 – 20 μS/cm conductivity, used for preparing concentrations of 10000 and 30000 mg/l in a QVF glass vessel (25 liter). The high solubility in conjunction with a relatively low molecular weight of the NaCl salt leads to a very high osmotic pressure (i.e. high water flux). As for the preparation of feed solution, it accomplished by dissolving small amounts of phenol in water, to produce concentrations (100 and 1000 mg/l). The chemical analysis of the draw and feed solutions are given in Table 1.

- **The Membranes**

In this study, two types of membranes used. First membrane is cellulose triacetate (CTA) forward osmosis membrane (X-Pack^{TM} supplied by Hydration Technology Inc., Albany, OR) was used for the osmosis experiments. The thickness of the CTA membrane is less than 50 μm and membrane consists of a woven fabric mesh embedded within a continuous polymer layer. The CTA membrane has high sodium chloride rejection 95 – 99%.

The second membrane is thin film composite membrane (TFC) which is commonly used in the process of reverse osmosis, but in this research was used in the two processes (FO and RO). TFC membrane is an aromatic polyamide consisting of three layers: polyester support web (120 μm), micro porous poly sulphone interlayer (40 μm), and ultra thin polyamide barrier layer on the top surface (0.2 μm). The specifications of the TFC membrane are salt rejection (96 – 99 %), maximum operating pressure (6 – 9 Mpa), maximum operating temperature 45 ^{o}C and pH range for continuous operation (2 – 11). The structure of forward osmosis membrane (CTA) is quite different from standard reverse osmosis membranes (TFC).

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Table 1 Chemical Specification of Draw and Feed Solutions

Substance	Properties
Phenol (C_6H_6O) MW = 94.11 Scharlab S.L. Made in Spain Solubility (8.2 g/ 100 ml H_2O)	Phenol, crystallized, reagent grade, ACS Assay 99.5% min. Identity (IR-Spectrum) passes test Chlorides 0.0005% Iron 0.0001% o-cresol 0.05% m-cresol 0.05% p-cresol 0.05% Non- volatile matter 0.01% Water 0.2%
Sodium Chloride (NaCl) MW = 58.44 Fluka chemika Solubility (35.7 g/ 100 ml H_2O)	Assay 99.5% min. Max. limits of impurities (%) Ammonia 0.002 Iron 0.002 Lead 0.0005 Potassium 0.02 Sulphate 0.02

- **The Unit Setup**

Figure 2 describes the forward osmosis apparatus used in laboratory of chemical engineering department – University of Baghdad. The osmosis cell is a plate and frame designed with a rectangular channel on each side of the membrane. The channel has dimensions of 19.7 cm length, 4 cm width, and 10 cm height, providing an effective membrane area of 197 cm^2. The draw solution (NaCl – H_2O) is flowing on the permeate side and the feed solution (phenol) on the feed side. Co-current flow is used to reduce strain on the suspended membrane. Mesh spacers are also inserted for support within both channels, and it serve to increase turbulence and hence mass transport on both sides of the membrane. The feed and draw solutions were pumped by means of a centrifugal pump (11.4 – 54.6 l/min, 3 – 13.7 m. H, 210 Watt, STUART TURNER LTD. HENLEY ON THAMES ENG, England) to pass through channels of osmosis cell. Two submersible electrical coil (220 Volt, 1000 Watt) and thermostat of range

from 0 to 80 °C were used to control on the solutions temperature. The flow rate of draw and feed solutions was regulated by means of globe valve connected at the discharge of the pumps, and measure with a calibrated rotameters with range flow (0 – 100 l/hr). Both the draw and feed solutions were held at the same temperature and flow rate during the FO tests. Concentration of phenol was measured by UV-ultraviolet/visible spectrophotometer (Shimadzu model UV-160 A). While, the concentration of NaCl was measured by digital laboratory conductivity meter (inoLab Cond 720, range (0 – 2 * 10^6 μS/cm), operating temperature (0 – 55 °C), accuracy is ± 0.5% full scale, the electrode material is graphite, and made in Germany (WTW)) and digital total dissolved solid (TDS) meter (Waterproof TDSTestr High+, range (0 – 1 * 10^4 mg/l), operating temperature (0 – 50 °C), accuracy is ± 1% , and Oakton instruments).

- **Experimental Procedure**

In the forward osmosis process, the phenol solution and draw solution flow tangent to the membrane in a cross flow mode. In the typical orientation of forward osmosis process, the draw solution is placed against the support layer and the feed solution is on the active layer. Through osmosis, water transports from the feed solution (low concentration) across the phenol rejecting membrane and into the draw solution (high concentration). The outlet streams of feed and draw solutions, recycled back to the main vessels. For every one hour, the concentrations of phenol and NaCl were measured and the water flux through membrane. The water flux was calculated by dividing the permeate volume by the product of effective membrane area and time. After recording the results, the solution (remaining in feed vessel), was drained by means of a drain valve. The whole system was washed by distilled water.

Reverse Osmosis

To yield pure water, the diluted draw solution exterior from forward osmosis process is sent to a reverse osmosis unit. An experimental rig of reverse osmosis unit was constructed in the laboratory as shown

schematically in Figure 3. The devices used in forward osmosis unit itself was used in reverse osmosis unit, except the selected membrane used a TFC membrane constructed as spiral - wound module (type SSRO50G, length = 12 in., diameter = 2 in., and membrane area = 0.483 m^2). Also, in RO we need to high pressure pump (santoprene and polypropylene materials, maximum pressure = 120 psi, and power = 220 – 240 V)   to overcome on osmotic pressure for salt NaCl in water. The process is carried out in a system where the draw solution flows across the RO membrane and pure water and a very reduced amount of the NaCl will pass through the membrane.

Produce from the reverse osmosis two streams, the first contains pure water can be used, while the second contains the concentrate draw solution, which be recycled to forward osmosis process. For every quarter hour, the conductivities and concentrations of the reject draw solution (concentrate) and product solution were measured by the conductivity and TDS meters, and the flow rate of the product (permeate) solution for each run was recorded.
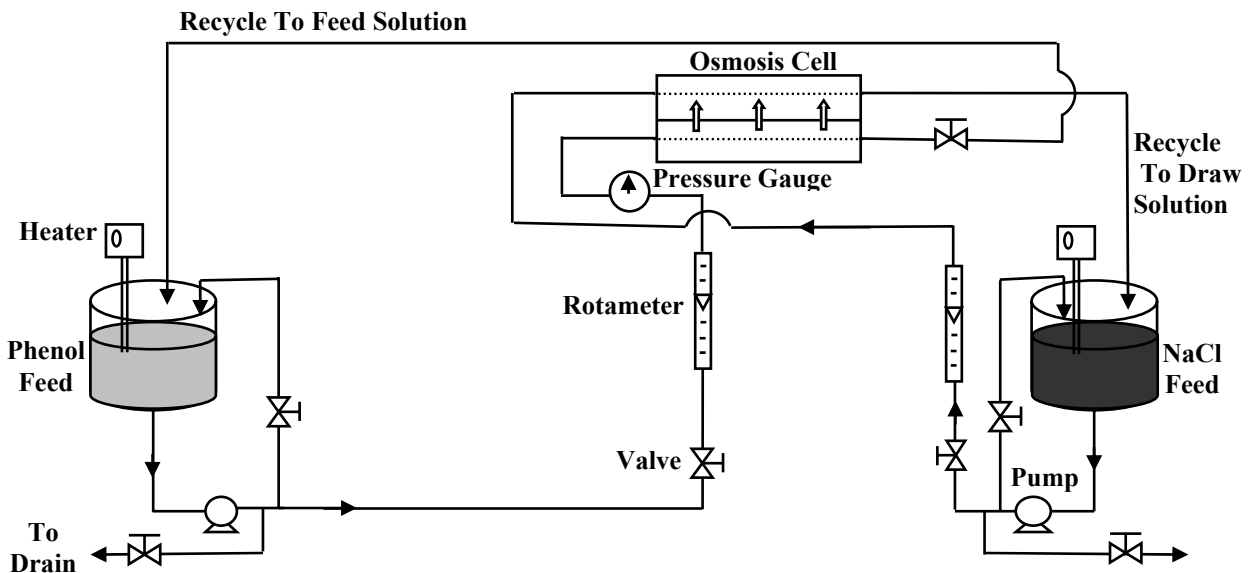
Fig. 2 Schematic Diagram of Forward Osmosis Process
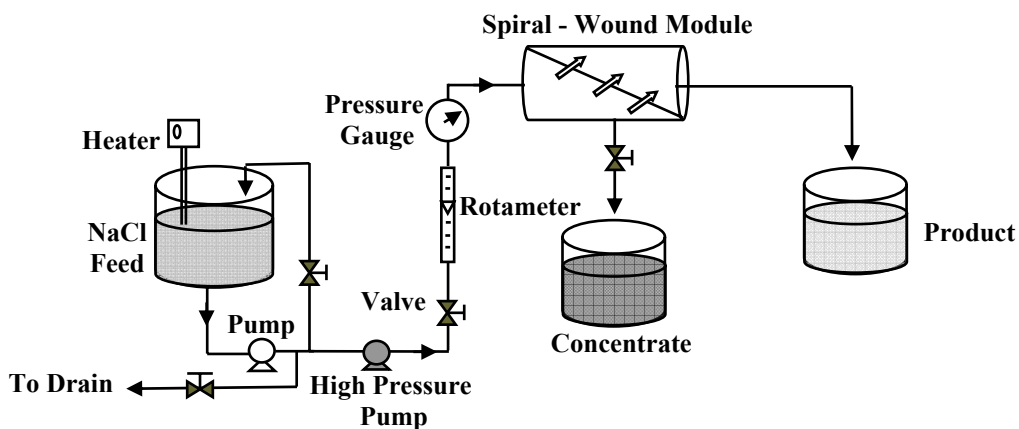
Fig. 3 Schematic Diagram of Reverse Osmosis Process

RESULTS AND DISCUSSION

Forward Osmosis

The effect of operating time (for 5 hour) on recovery percentage (Y%) and water flux (J) is shown in Figure 4. The increase in percentage recovery with increase in time might be due to an increase in the volume of pure water transfer from feed solution (phenol) to the draw solution across the membrane. The water flux decrease with time due to an increase of phenol concentration in feed vessel and decrease of draw solution concentration in feed, subsequently the osmotic power (or the driving force) across the membrane decreased. The water flux calculated by dividing the volume of pure water which transfers from feed to draw solution on time and active area of membrane. The phenol concentrations in these experiments are 100 and 1000 mg/l at the time zero, the recovery and water flux decrease with increasing phenol concentration in feed because the driving force decreased. Figure 5 shows the concentration of phenol in feed vessel and osmotic pressure of phenol versus time. The concentration and osmotic pressure of sodium chloride with time is shown in Figure 6. The solution of phenol loses quantities of pure water and this leads to increased concentration of phenol. The same quantities of pure water transferred across the membrane to the draw solution, as a result, decrease the concentration of sodium chloride. Thus, increases or decreases in concentrations of sodium chloride and phenol are linked to each other.

The Figures 7, 8 and 9 show the effect of NaCl concentration (10000 and 30000 mg/l at t = 0 h) on the recovery percentage, water flux, and outlet concentrations of phenol and sodium chloride respectively. The effect of the change in salt concentration (NaCl) has a greater effect on the water flux through the membrane because the sodium chloride has a high osmotic pressure. The increase in draw solution concentration while the concentration of feed solution (phenol) remains constant this leads to the difference increase in osmotic pressures on sides of membrane ($\pi_{NaCl} - \pi_{phenol}$), subsequently the flux (J) increased

according to the Equation 6. These observations are well agreed with the results of McCutcheon et al., 2006.

The discrimination between cellulose triacetate membrane CTA and thin film composite membrane TFC in forward osmosis process illustrated in Figures 10, 11 and 12. The CTA membrane gives higher water flux than TFC membrane, thus we note that the percentage of recovery for the CTA membrane higher than the TFC membrane and this is evident in Figure 10. The CTA membrane better than TFC membrane because the CTA membrane was originally manufactured for the forward osmosis process, in addition the TFC membrane is composed of several layers and has a supportive layer to withstand the high pressures in the reverse osmosis process. Therefore, the concentration of phenol is more concentrate in the case of the use of CTA membrane, while the concentration of NaCl is high in the case TFC membrane used, and this is due to the high efficiency of the CTA membrane in the forward osmosis process. This is shown in Figures 11 and 12. Practical experiments of the FO process showed that the highest recovery ratio has been reached is 51.33% and this means that it has been recovered 7.7 liter of pure water from contaminated water by phenol, which was the size of 15 liter. Also, by using the CTA membrane increased recovery percentage of water by 23% compared to using the TFC membrane at the same conditions. These results correspond with the results of the researcher Rana, 2011.

The Analysis of Concentration Polarization

In Figures 13, 14, and 15 water flux is presented as a function of the osmotic pressure difference ($\pi_{NaCl} - \pi_{phenol}$) between the bulk feed and draw NaCl solutions. Osmotic pressure was calculated according to the equation 18 where i = 2 for NaCl, i = 1 for phenol and $\Phi = 1$ for an ideal solution. In these figures the membrane was oriented in the forward osmosis mode and also indicates flux under the influence of dilutive internal concentration polarization in the presence of concentrative external concentration

polarization. The relationship between driving force and the rate of transfer of water through the membrane is a linear relationship for the membranes which used in this search. This behavior is agreement with standard equation for forward osmosis process (Equation 6). Therefore, increase in difference between the osmotic pressure of phenol and sodium chloride leads to an increase in the water flux.

Figure 16 show the flux (J) versus $\ln(\pi_D/\pi_F)$, we find the slope of line in figure 16 which represent the inverse of K (Equation 17). The value of resistance to solute diffusion within the membrane porous support layer (K) is 36.83 h/m. This equation considers another way of evaluating the flux behavior and confirming the presence of internal concentration polarization is by normalizing the driving force. These calculations are well agreed with the results of Tang et al., 2010.

Reverse Osmosis

In the second stage of research has been taking the sample of the draw solution to see the possibility of treatment by reverse osmosis process. Also, know specifications of the product water from reverse osmosis process. According to this process the draw solution is separated into two parts, the first part consists of pure water and contain small amounts of salt can be used in several applications, while the second part consists of too salty water and can be retrieved to the forward osmosis process as draw solution.

The recovery percent and water flux from reverse osmosis unit are plotted versus time, as shown in Figure 17. By increasing time, the pure water quantities which transfer across the membrane increased, therefore the recovery percent increase according to Equation 19. While, the flux decreased with increase in operating time. The flux of a reverse osmosis system decrease as fouling occurs, because the foulants on the membrane surface retard the back diffusion of the salt into the bulk solution to cause concentration polarization at the membrane surface. The increase in concentration polarization causes a decrease in the product rate (i.e. water flux), an increase in the concentration of NaCl in concentrate stream and a decrease in the concentration of sodium chloride in permeate

stream. The rejection of salt decreased with increase in operating time. This is shown in Figure 18. The increasing of salt concentration in permeate will decrease the rejection percentage and vice versa. The reason, which was discussed before, for the effect of time on NaCl concentration, can be explain the decreasing of rejection percentage with increase in operating time. These results are similar behavior with the results of the Ahmed, 2000.

The concentration of NaCl in concentrate stream (or reject stream) versus time is shown in Figure 19. The decrease in salt concentration and the osmotic pressure in the reject stream with time due to a decrease in the transfer of pure water across the membrane with time. After two hours of work of the reverse osmosis system we get a recovery ratio of 58%, means it has been obtained 11.6 liters from feed vessel which it has volume is 20 liters.
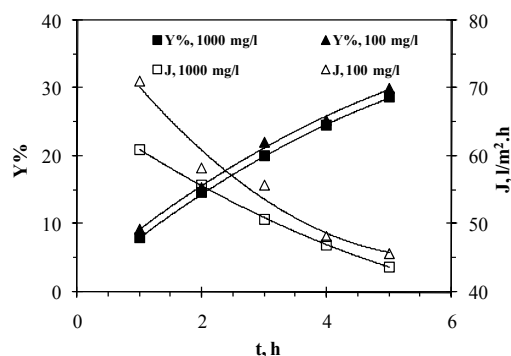

Fig. 4 Effect of Time on Recovery Percent and Flux for different Phenol Concentration (CTA Membrane, C_{NaCl} = 10000 mg/l, Q_{NaCl} = 50 l/h, Q_{phenol} = 50 l/h, P = 0.3 bar and T = 25 °C)

Membrane, C_{phenol} = 1000 mg/l, Q_{NaCl} = 50 l/h, Q_{phenol} = 50 l/h, P = 0.3 bar and T = 25 °C)

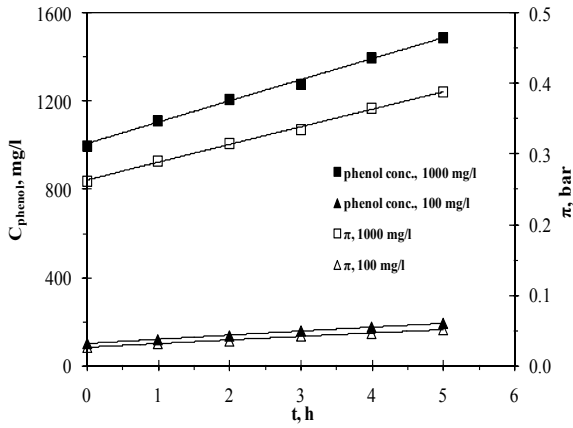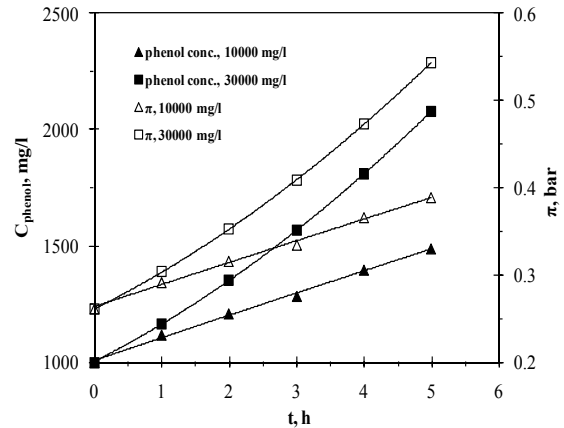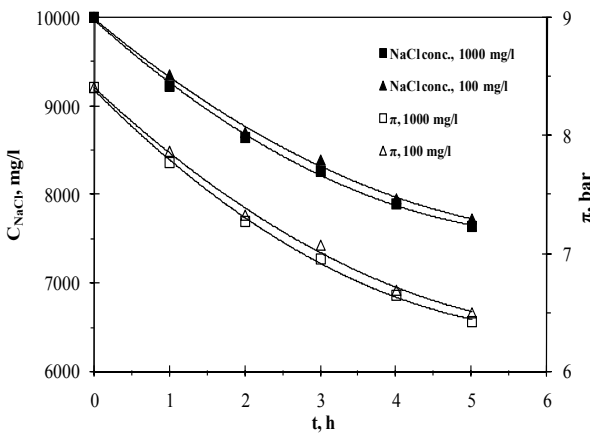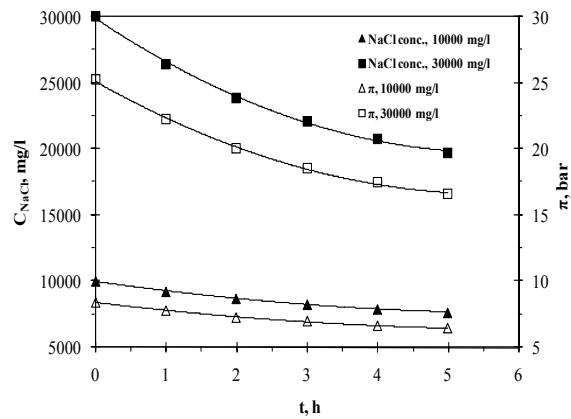Fig. 5 Effect of Time on Phenol Conc. and Osmotic Pressure of Phenol for different Phenol Concentration (CTA Membrane, C_{NaCl} = 10000 mg/l, Q_{NaCl} = 50 l/h, Q_{phenol} = 50 l/h, P = 0.3 bar and T = 25 °C)

Fig. 8 Effect of Time on Phenol Conc. and Osmotic Pressure of Phenol for different NaCl Concentration (CTA Membrane, C_{phenol} = 1000 mg/l, Q_{NaCl} = 50 l/h, Q_{phenol} = 50 l/h, P = 0.3 bar and T = 25 °C)

Fig. 6 Effect of Time on NaCl Conc. and Osmotic Pressure of NaCl for different Phenol Concentration (CTA Membrane, C_{NaCl} = 10000 mg/l, Q_{NaCl} = 50 l/h, Q_{phenol} = 50 l/h, P = 0.3 bar and T = 25 °C)

Fig. 9 Effect of Time on NaCl Conc. and Osmotic Pressure of NaCl for different NaCl Concentration (CTA Membrane, C_{phenol} = 1000 mg/l, Q_{NaCl} = 50 l/h, Q_{phenol} = 50 l/h, P = 0.3 bar and T = 25 °C)
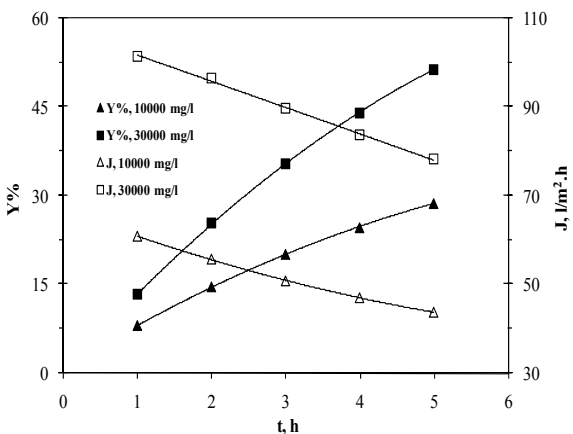
Fig. 7 Effect of Time on Recovery Percent and Flux for different NaCl Concentration (CTA

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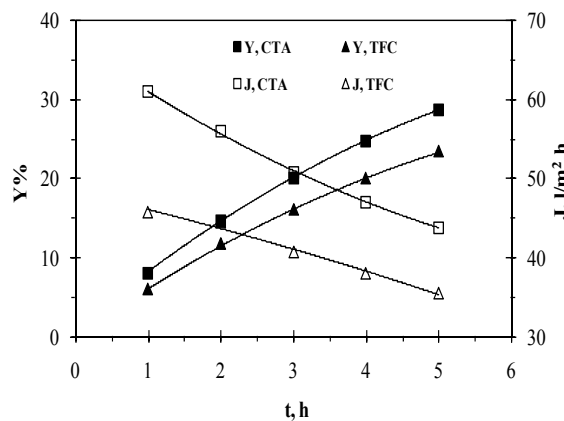
Fig. 10 Effect of Time on Recovery Percent and Flux for CTA and TFC Membranes (C_{NaCl} = 10000 mg/l, C_{phenol} = 1000 mg/l, Q_{NaCl} = 50 l/h, Q_{phenol} = 50 l/h, P = 0.3 bar and T = 25 °C)
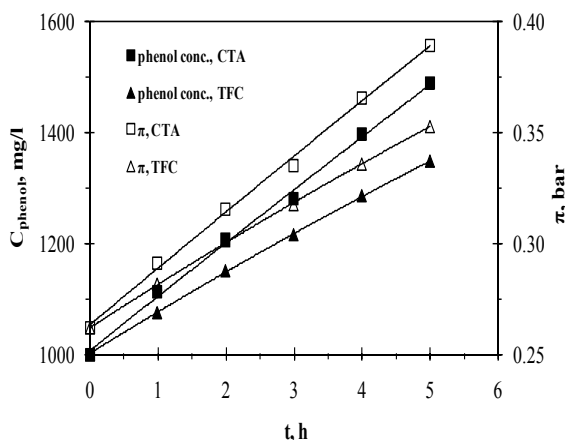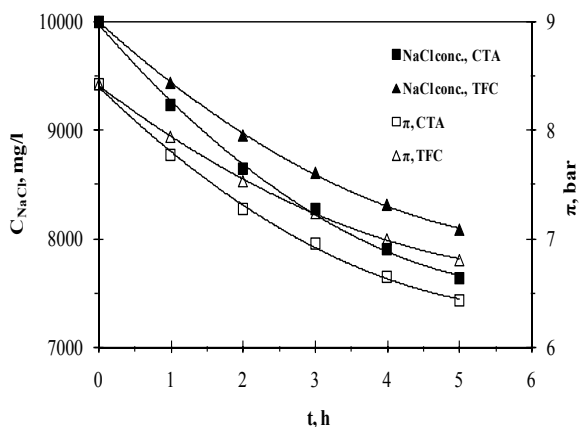

Fig. 11 Effect of Time on Phenol Conc. and Osmotic Pressure of Phenol for CTA and TFC Membranes (C_{NaCl} = 10000 mg/l, C_{phenol} = 1000 mg/l, Q_{NaCl} = 50 l/h, Q_{phenol} = 50 l/h, P = 0.3 bar and T = 25 °C)

Fig. 12 Effect of Time on NaCl Conc. and Osmotic Pressure of NaCl for CTA and TFC Membranes (C_{NaCl} = 10000 mg/l, C_{phenol} = 1000 mg/l, Q_{NaCl} = 50 l/h, Q_{phenol} = 50 l/h, P = 0.3 bar and T = 25 °C)
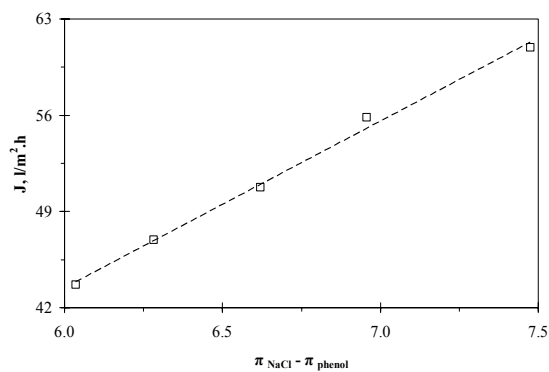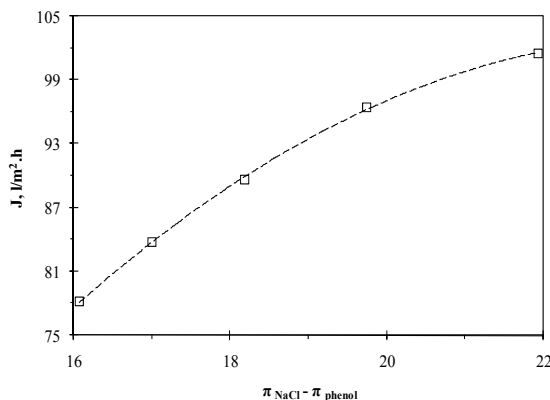

Fig. 13 Effect of Driving Force on Water Flux for CTA Membrane (C_{NaCl} = 10000 mg/l, C_{phenol} = 1000 mg/l, Q_{NaCl} = 50 l/h, Q_{phenol} = 50 l/h, P = 0.3 bar and T = 25 °C)

Fig. 14 Effect of Driving Force on Water Flux for CTA Membrane (C_{NaCl} = 30000 mg/l, C_{phenol} = 1000 mg/l, Q_{NaCl} = 50 l/h, Q_{phenol} = 50 l/h, P = 0.3 bar and T = 25 °C)
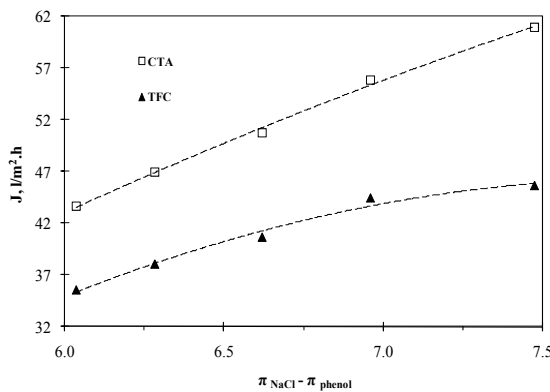

Fig. 15 Effect of Driving Force on Water Flux for CTA and TFC Membranes (C_{NaCl} = 10000 mg/l, C_{phenol} = 1000 mg/l, Q_{NaCl} = 50 l/h, Q_{phenol} = 50 l/h, P = 0.3 bar and T = 25 °C)
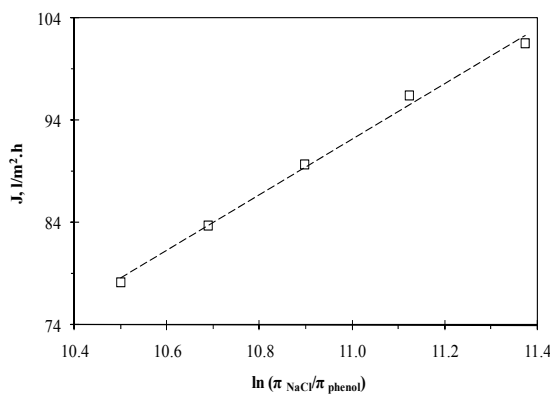

Fig. 16 Water Flux Data Plotted Against the Logarithm of the Ratio of Feed and Draw Solution Osmotic Pressures for Calculate K,

Equation 20 (CTA Membrane, C_{NaCl} = 30000 mg/l, C_{phenol} = 1000 mg/l, Q_{NaCl} = 50 l/h, Q_{phenol} = 50 l/h, P = 0.3 bar and T = 25 °C)
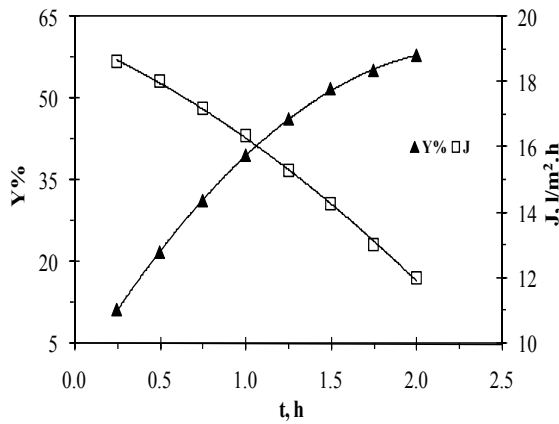

Fig. 17 Effect of Time on Recovery Percent and Flux in RO Process (C_{NaCl} = 7500 mg/l, Q_{Feed} = 20 l/h, pH = 6.5, P = 8.5 bar, V_F = 25 l and T = 25 °C)
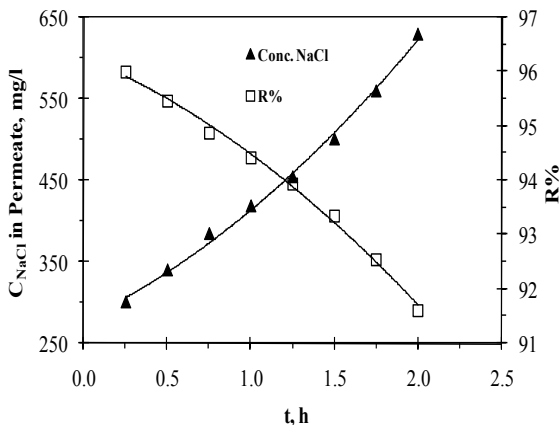

Fig. 18 Effect of Time on Product NaCl Concentration and Rejection Percent in RO Process (C_{NaCl} = 7500 mg/l, Q_{Feed} = 20 l/h, pH = 6.5, P = 8.5 bar, V_F = 25 l and T = 25 °C)
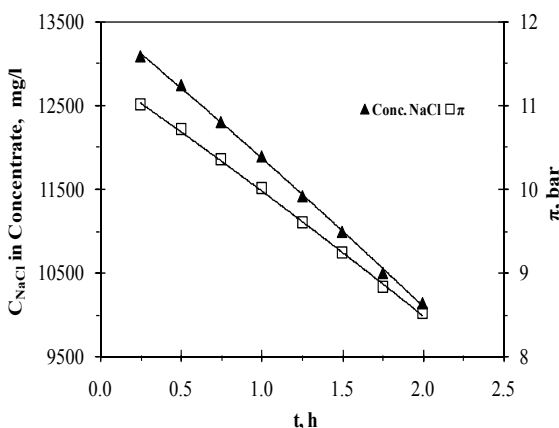

Fig. 19 Effect of Time on Rejected NaCl Concentration and Osmotic Pressure in RO Process (C_{NaCl} = 7500 mg/l, Q_{Feed} = 20 l/h, pH = 6.5, P = 8.5 bar, V_F = 25 l and T = 25 °C)

CONCLUSION

- Forward osmosis process is a convenient method for recovery of water from wastewater by phenol.

- Cellulose triacetate membrane (CTA) gave better results than the thin film composite membrane (TFC). Therefore, membrane CTA prefers more in the forward osmosis process.

- The recovery percentage for water through membrane increases with increasing operating time. Water flux decreases with time due to the decrease in driving force.

- The recovery percentage and water flux increases with increasing concentration of draw solution (i.e. osmotic pressure). While, the recovery and flux decreases with increasing concentration of feed solution.

- Outlet concentration of each of salt and phenol is associated with the amount of pure water transfer through the membrane, where the increase in water flux cause increase in concentration of phenol and decrease the salt concentration.

- The highest recovery ratio is 51.33%. Using CTA membrane recovery rate increase by 23% compared to using TFC membrane.

- The influence of dilutive internal concentration polarization and concentrative external concentration polarization appeared in forward osmosis process. Numerical value was calculated for the resistance to solute diffusion within the membrane porous support layer (K) is 36.83 h/m. .

- Reverse osmosis process is a good method to treatment of draw solution to

be used again. Recovery percentage from pure water is 58% after the operating time of two hours.

- In the RO process, the TFC membrane has high salt (sodium chloride) rejection 91.6 – 96 %. This leads to improve the performance of the forward and reverse osmosis processes together to recover the pure water from wastewater.

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NOMENCLATURE

Symbol	Definition	Units
A	Pure Water Permeability Coefficient	$l/m^2.h.bar$
B	Salt Permeability Coefficient	m/s
C	Concentration of Solute	mg/l
C_F	Feed Concentration	mg/l
C_P	Product Concentration	mg/l
CP	Concentration Polarization	
D	Solute Diffusion Coefficient	m^2/s

d_h	Hydraulic Diameter	m
J	Water Flux	$l/m^2.h$
k	Mass Transfer Coefficient	m/s
K	Resistance to Solute Diffusion	m/h
P	Pressure	bar
Q	Flow Rate	l/h
L	Length	m
R	Rejection Percentage	
Re	Reynolds Number	
R_g	Gas Constant	bar. l/gmol. k
Sc	Schmidt Number	
Sh	Sherwood Number	
t	Time	h
T	Temperature	°C
V_F	Volume of Water in Feed Vessel	l
V_P	Product Volume of Water	l
Y	Recovery Percentage	

reek Symbols

Symbol	Definition	Units
π	Osmotic Pressure	bar
$\pi_{D,b}$	Osmotic Pressure of the Draw Solution in the Bulk	bar
$\pi_{D,i}$	Osmotic Pressure of the Draw Solution on the Inside of the Active Layer	bar
$\pi_{D,m}$	Osmotic Pressure of the Draw Solution at the Membrane Surface	bar
$\pi_{F,b}$	Osmotic Pressure of the Feed Solution in the Bulk	bar
$\pi_{F,i}$	Osmotic Pressure of the Feed Solution on the Inside of the Active Layer	bar
$\pi_{F,m}$	Osmotic Pressure of the Feed Solution at the Membrane Surface	bar
σ	Osmotic Reflection Coefficient	
Φ	Osmotic Coefficient	