

## Estimation of Concentration Polarization Using the Combined Film theory/ Spiegler-Kedem Model and Empirical Correlation

Taha Yaseen Khalaf

Biomedical Engineering Dept./ Al-Khwarizmi  
Engineering College / University Of Baghdad

Email: Tahayaseen@ymail.com

### Abstract

*At a given operating pressure and in a given apparatus, concentration polarization results in an increase in the effective osmotic pressure of the feed solution on the membrane surface, a consequent decrease in the effective pressure ( $\Delta P$ ) for fluid flow across the membrane, and progressive changes in the mass transfer coefficient, product rate, and solute separation along the length of the membrane in the direction of feed flow. The problem then is to predict the effect of concentration polarization on solute separation and on predict rate, under specified experimental condition. In the present work, the reverse osmosis experiments were conducted in a spiral wound model to get separation data using a NaCl-water system up to 2500 ppm and cellulose acetate membrane.*

*The purpose of this paper is to predict and analyze the concentration polarization using a combined film theory model and empirical correlation. Therefore, the results of the experiments show that the concentration polarization affected when using different models.*

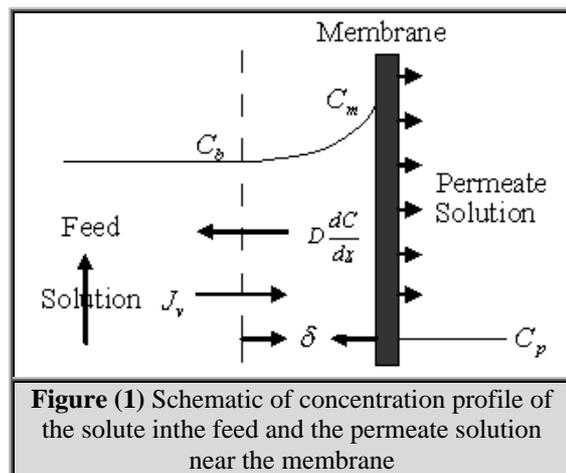
### 1. Introduction

Membrane separation such as reverse osmosis, ultra filtration and others are recently developed separation techniques. As compared with other separation methods, membrane processes possess many advantages such as no phase change, simple equipment, low energy consumption and ease of operation. As a result they have a great tendency to be used in industry as well as in scientific and technological circles.

The main task in designing reverse osmosis devices is selecting the optimum hydraulic parameters to reduce the power consumption of the system and extend the life of the membrane, thus lowering production cost. For the optimization of hydraulic parameters, concentration polarization which seriously affects the performance of the separation system, is one of the important control factors influencing the system design.

It could be reduced to a certain extent by some appropriate measures, but complete elimination of this phenomenon is impossible in any practical operating system [ 1 ].

Figure 1 illustrate the mass transfer system in reverse osmosis for a membrane under steady state conditions [ 2 ]. When the solution flows through the system parallel to the membrane surface at a given rate, both the solute and solvent are forced to pass through the membrane owing to the action of the pressure difference. The solvent can pass through the membrane completely, but most of the solute accumulates at the surface due to the rejection caused by the membrane. Thus a concentration gradient is built between the membrane surface and bulk solution, which makes the solute diffuse back towards the bulk solution. The higher the concentration of solute at the membrane surface, the lower the permeation rate of the solvent. This unfavorable phenomenon is called "concentration polarization" [3-5].



The starting point for our mathematical description of RO/NF separations is the solution-diffusion model. The model assumes that the permeation driving force is the gradient in chemical potential of the solute (Wijmans and Baker, 1995). When the transport equation is expressed in terms of solvent flux ( $J$ ), it is given as

$J_v = A(\Delta P - \sigma \Delta \pi)$	1
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Where A is the solvent permeability through the membrane,  $\Delta P$  is the applied pressure,  $\Delta \pi$  is the osmotic pressure difference between the membrane surfaces and  $\sigma$  is the reflection coefficient. The reflection coefficient represents the intrinsic salt rejection by the membrane, but when intrinsic salt rejection is over a 0.98, which is typical for reverse osmosis separations,  $\sigma$  may be assumed equal to unity (Bhattacharjee et al., 2001). When intrinsic salt rejection is significantly less than 0.98 (i.e., nanofiltration), the reflection coefficient should be used to more accurately predict the resultant trans-membrane osmotic pressure (Murthy and Gupta, 1997)[ 6 ].

Equation (1) serves as the starting point for the design of most modern RO/NF separations. The rejection of ionic species results in an elevated salt concentration near the membrane surface creating a local concentrated layer (CP), this layer quickly reaches a steady state, and the transverse solute flux through the CP layer is constant[ 6,7 ]. The solvent flux (J) may then be determined by the following one-dimensional, steady-state mass balance across the CP layer:

$J_v \cdot C_p = J_v \cdot C - \left[ D \left( \frac{dC}{dx} \right) \right]$	2
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Where,  $J_v$  is the permeate flux through the membrane,  $C_p$  is the permeate solute concentration,  $C$  is the solute concentration in the boundary layer and  $D$  is the solute diffusion coefficient in water[ 8 ].

Integrating the one dimensional (transverse) convection- diffusion mass balance from the membrane surface out to a finite mass boundary (film) layer thickness,  $\delta$ , yields the relationship between concentration polarization and permeate flux. The result is,

$CP = \frac{C_m - C_p}{C_b - C_p} = \exp\left(\frac{J_v \delta}{D}\right)$	4
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or

$CP = \frac{C_m - C_p}{C_b - C_p} = \exp\left(\frac{J_v}{k}\right)$	5
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where  $C_m$  is concentration at the membrane surface, or channel wall, for the rejected salt,  $C_b$  and  $J_v$  are the bulk solute concentration and the permeate water flux through the membrane respectively, and  $(D/\delta) =$  mass transfer coefficient( $k$ )[ 9 ].

## 2. Theory

Several theoretical models have been proposed for estimating concentration polarization. The models are based on the solution of the diffusion-convection differential equations, using appropriate boundary conditions and simplifying assumption. The most commonly accepted CP models are film theory, Spiegler-Kedem and solution-diffusion models(Sablani et al.,2001, Murthy and Gupta, 1997). The alternative approach to calculate CP is to develop correlations for estimating the mass transfer coefficient [10].The development of a generalized mass transfer correlation begins with the assessment of previous work.

## 3. Combined solution-Diffusion / film model

Equation (5) shows that CP is strongly dependant on two parameters,  $J_v$  and  $k$ . Therefore it can be expressed in terms of the membrane rejection fractions(observed rejection( $R_o$ ) and real rejection( $R_r$ ), thus, the concentration polarization expression takes the form[ 11 ]:

$\frac{1 - R_{obs}}{R_{obs}} = \frac{1 - R_r}{R_r} \cdot \exp\left(\frac{J_v}{k}\right)$	6
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The solute transport according to the simple solution-diffusion model is given below[8,12],

$J_s = C_p \cdot J_v = P_s \cdot (C_m - C_p)$	7
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Where  $P_s$  is the overall permeability coefficient, which characterizes solute transport through the membrane. Combining

equations (6) and (7), the unknown parameters  $C_m$  and  $R_r$  can be eliminated, giving:

$$\frac{R_o}{1-R_o} = \left[ \frac{J_v}{P_s} \right] \exp\left(\frac{-J_v}{k}\right) \quad 8$$

#### 4. Combined Spiegler-Kedem/film theory model

A concentration on both sides of membrane causes diffusive transport. When high concentration differences between the reject and the permeate exit, Spiegler and Kedem used the above equation and obtained the following expression of the rejection rate of the solute related to permeation flux:

$$R = \frac{\sigma(1-F)}{(1-\sigma F)} \quad 9$$

$$F = \exp\left(\frac{-J_v(1-F)}{P_s}\right) \quad 10$$

$$\frac{R_o}{1-R_o} = \frac{\sigma}{1-\sigma} \left[ 1 - \exp\left\{-\frac{J_v(1-\sigma)}{P_m}\right\} \right] \exp\left(\frac{-J_v}{k}\right) \quad 11$$

Where  $R$  is the rejection. According to Eq. (9), the rejection increases with increasing the water flux. The Parameters  $\sigma$  and  $P_s$  can be determined from the experimental data of rejection as a function of flux using best-fit method[ 13 ].

#### 5. Empirical correlation of mass transfer coefficient

Generalized correlations of mass transfer for developed flow (in both laminar and turbulent regions) suggest that the Sherwood number (Sh) is related to the Reynolds number (Re), Schmidt number (Sc) and the flow geometry in the form (Cussler,1997)[ 8 ]:

$$Sh = a(\text{Re})^\alpha (\text{Sc})^\beta \left(\frac{d_h}{L}\right)^\gamma \quad 12$$

$$Sh = \frac{kd_h}{D} = a \left(\frac{d_h u}{\nu}\right)^\alpha \left(\frac{\nu}{D}\right)^\beta \left(\frac{d_h}{L}\right)^\gamma \quad 13$$

Where  $d_h$  is the hydraulic diameter,  $u$  is the flow velocity,  $\nu$  is the kinematic viscosity,  $D$  is the solute diffusivity in water,  $L$  is the length of the tube channel and  $a$  is a numerical constant characterizing the flow channel geometry. The values of constants  $a$  and  $\alpha, \beta, \gamma$  are parameters, experimentally.

For laminar flow in a thin rectangular channel, the mass transfer coefficient (k) may be related to the Sherwood number (Sh) through the following equation[4 ]:

$$Sh = \frac{kd_h}{D} = 1.62 \left(\frac{d_h u}{\nu}\right)^{\frac{1}{3}} \left(\frac{\nu}{D}\right)^{\frac{1}{3}} \left(\frac{d_h}{L}\right)^{\frac{1}{3}} \quad 14$$

For turbulent flow, the mass transfer coefficient (k) may be related to the Sherwood number (Sh) through the following equation (Dittus and Boelter relation ) [12 ]:

$$Sh = \frac{kd_h}{D} = 0.2487 \left(\frac{d_h u}{\nu}\right)^{0.7604} \left(\frac{\nu}{D}\right)^{0.392} \quad 15$$

The diffusivity and kinematic viscosity data for the NaCl-water system are taken from the reference [15].

#### 6. Experimental Work

Pilot scale experiments were performed using a custom made pilot scale membrane tester (Berkefeld Filter commercial ) which holds a spiral wound membrane module with 20.1 cm (8 inch) nominal diameter and 101.6 cm (40 inch) effective length. The experiment was performed using commercially available pilot scale RO membrane ROGA-HR manufactured by Koch Membrane Systems, Inc., (USA). ROGA-HR is the type high rejection Cellulose Acetate (CA) RO membrane element for brackish water desalination.

The trans-membrane pressure and volumetric flow rate were adjusted using the concentration (reject) outlet valve. The pressure was varied between 15 bar and 35 bar. The experiments were carried out with a NaCl-water solution, the feed temperature is constant and feed concentration varied from 725 ppm – 2500 ppm.

#### 7. Results and Discussion

Experimental data were obtained at various feed concentration and transmembrane pressure. Under a fixed set of conditions,  $J_v$  and  $R_o$  were measured. These data were curve fitting using Levenberg-Marguardt [14] nonlinear regression technique to obtain the values of the parameters for the Eqs. (8) and (11). Table 1 shows that the membrane parameters and mass transfer coefficient for the three models (Combined solution-Diffusion / film, Combined Spiegler-Kedem/film theory and Empirical correlation), which shows that the parameters for Eq.(8) are relatively constant and the  $k$  values for the same equation differ markedly. The most important observation is that the  $k$  values estimated from Eq.(8) shows a negligible variation with feed flow rate, as observed early by a simple graphical method as shown clearly in figures (1) and (2), thus the true concentration polarization values cannot be predicted from this model.

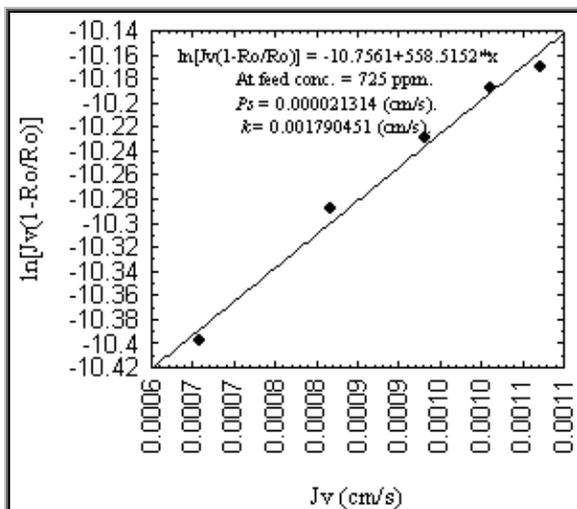


Figure (2) Computation of parameter and mass transfer coefficient for the (CFSD) model.

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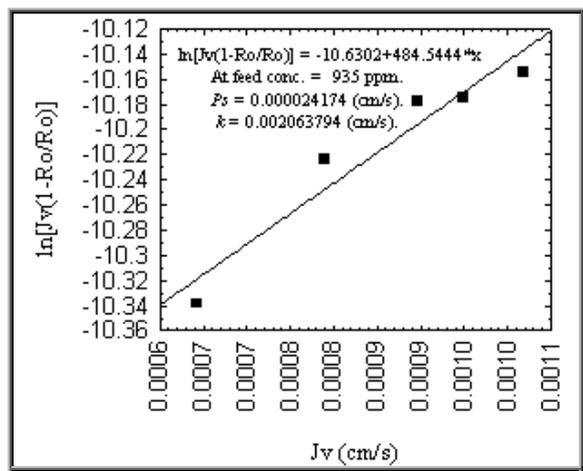


Figure (3) Computation of parameter and mass transfer coefficient for the (CFSK) model.

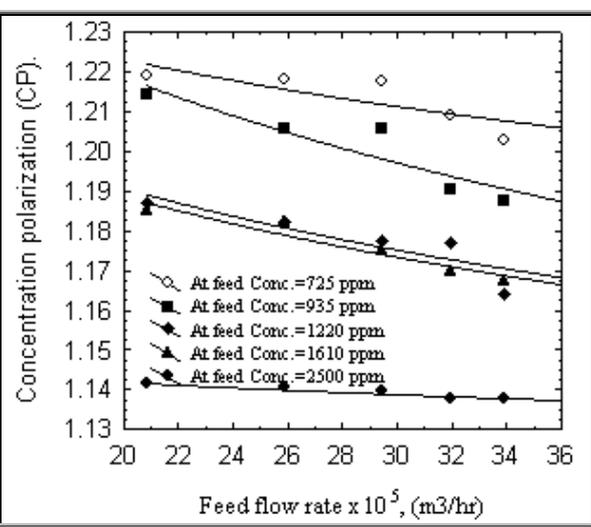


Figure (4) Effect of feed flow rate on concentration polarization using CFSK

Table (1) Parameters and mass transfer coefficients estimated from the combined film theory/solution diffusion (CFSD) model, the combined film theory/ Spiegler-Kedem (CFSK) model and the empirical correlation by a nonlinear parameter estimation program for the NaCl-water system.

S.no.	Feed rate, (m <sup>3</sup> /hr)	Feed conc., ppm	CFSD model $P_s \times 10^5$ , cm/s	$k \times 10^4$ , cm/s	CFSK model $\sigma$	$P_m \times 10^5$ , cm/s	$k \times 10^4$ , cm/s	Empir. Correl. $k \times 10^4$ , cm/s
1	20.833	725	2.145	18.06701	0.9874	2.0037	33.298	32.431
2	25.833	725	2.148	18.18968	0.9856	2.0918	41.466	41.849
3	29.444	725	2.148	18.18969	0.9842	2.0820	47.285	47.053
4	31.944	725	2.148	18.19046	0.9830	2.0739	53.267	53.562
5	33.888	725	2.113	20.0787	0.9822	2.0681	58.088	58.759
6	20.833	925	2.293	19.06832	0.9917	2.5096	33.072	34.198
7	25.833	925	2.317	19.20387	0.9897	2.5007	42.265	42.731
8	29.444	925	2.317	19.20395	0.9883	2.4935	47.908	48.045

9	31.944	925	2.317	19.20786	0.9869	2.4860	54.472	54.690
10	33.888	925	2.594	20.00036	0.9860	2.4808	59.283	59.997
11	20.833	1220	2.362	19.65126	0.9984	2.7787	36.387	36.120
12	25.833	1220	2.363	19.69739	0.9965	2.7727	44.052	44.100
13	29.444	1220	2.363	19.69742	0.9951	2.7677	48.929	49.584
14	31.944	1220	2.363	19.69841	0.9947	2.7807	52.770	56.442
15	33.888	1220	2.344	20.66709	0.9946	2.8774	62.893	61.919
16	20.833	1600	2.511	20.88620	0.9979	2.9525	36.337	36.390
17	25.833	1600	2.551	21.47867	0.9973	2.9507	44.111	44.100
18	29.444	1600	2.551	21.47895	0.9958	2.9453	49.578	49.583
19	31.944	1600	2.510	21.47896	0.9946	2.9407	54.875	56.442
20	33.888	1600	2.551	21.49437	0.9945	3.0063	61.881	61.193
21	20.833	2500	2.801	24.22169	0.9986	3.2908	45.901	45.620
22	25.833	2500	2.816	24.72346	0.9967	3.2842	52.595	52.405
23	29.444	2500	2.816	24.72366	0.9954	3.2789	57.809	58.922
24	31.944	2500	2.816	24.72366	0.9943	3.2753	64.220	67.072
25	33.888	2500	2.816	24.73257	0.9942	3.3233	72.966	77.581

The values of concentration polarization as a function of feed flow rate at a constant temperature are presented in Figs. (4) and (5). This figures showed that CP decreased with increase in flow rate. The increase in feed flow rate reduces CP values due to increase in turbulence near the membrane. At a constant temperature, the properties of the fluid are not change. Since remaining the effect of feed flow rate on concentration polarization is only.

Permeation flux is an important parameter in the design. Flux is affected by several factors such as feed pressure, operating temperature, feed velocity and/or composition. As shown in Fig. 6, flux decreased with increasing recovery rate. While recovery was increasing, feed stream concentration increased. If the membrane plant was operated at a recovery of 50%, the feed stream concentration was doubled. At high concentration factors, the negative effect of concentration polarization on membrane

polarization using empirical correlation

performance can be so serious that the flux is decreased. Deposition of solute on the surface of membranes can change the separation characteristics and the high concentration of solute at the membrane interface increases the risks of change in composition of the membrane material due to chemical attack.

As recovery rate rises, the salt concentration in the feed stream increases, which causes an increase in the driving force for salt flow or salt passage. Higher salt concentration levels in the feed stream increase the osmotic pressure, which reduce the net pressure driving force and therefore permeate flow. As shown in Fig. 7, conductivity rejection was decreased with increasing recovery rate.

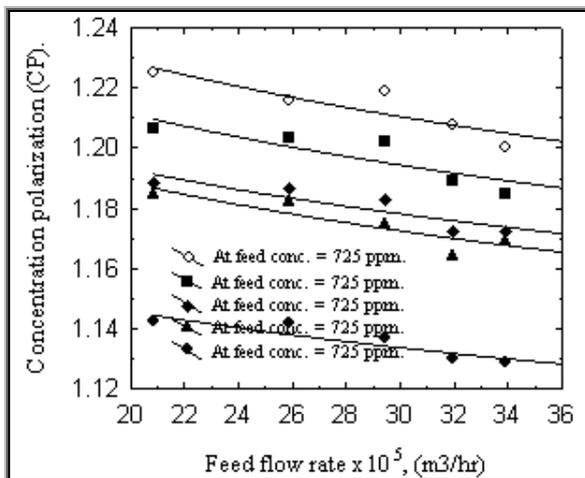


Figure (5) Effect of feed flue rate on concentration

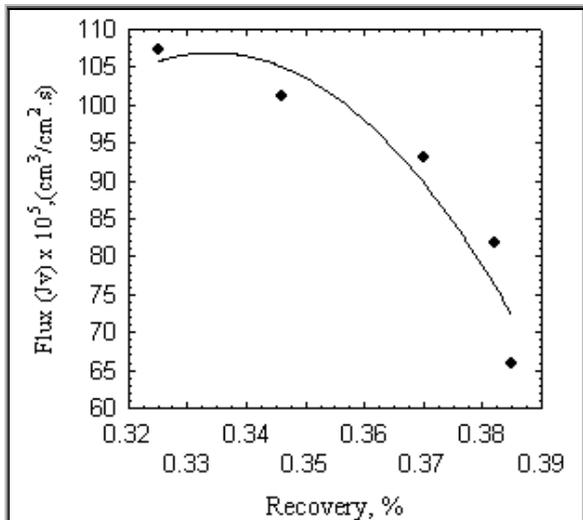
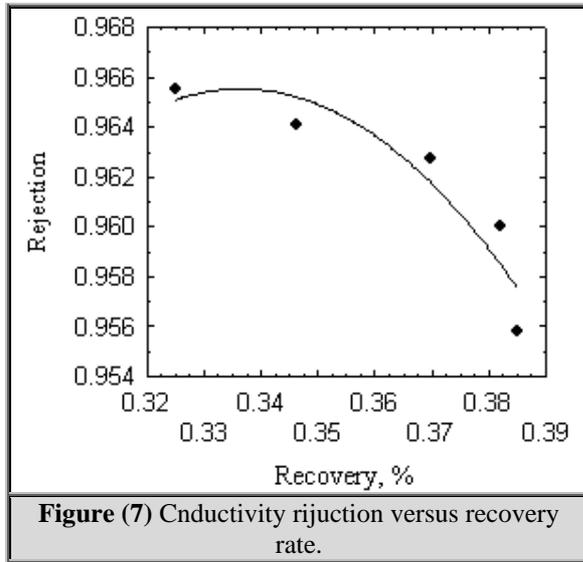


Figure (6) The reLation between recovery rate and permeate flux.



## 8. Conclusions

For determination of scaling of the propensity of the fouling species in water, it is essential to evaluate the concentration polarization level near membrane surface under the given experimental condition such as flow rate, pressure and feed concentration. The proposed mass transfer and the CP obtained from the combined film theory/ Spiegler-Kedem (CFSK) model gave good agreement with the empirical correlation against the combined film theory/solution diffusion (CFSD) model. The results show that even though the parameters of the Spiegler-Kedem model and the solution-diffusion model are constant over the operating conditions for our experimental data, the  $k$  values differ considerably. The reason for this may be the presence of a reflection coefficient in the Spiegler-Kedem model. Finally, at a constant temperature the effect of flow shows much influence on permeate concentration and the accumulate solute (CP) at the membrane shows some effect on the recovery and rejection.

## 9. Nomenclature

- A Solvent permeability  
 $C$  Solute concentration in the boundary layer  
 $C_b$  Feed concentration  
 $C_m$  Membrane surface concentration  
 $C_p$  Permeate concentration  
 $D$  Solute diffusion coefficient in water  
 $J_s$  Solute flux through membrane

- $J_v$  Permeate water flux  
 $k$  mass transfer coefficient  
 $\Delta P$  Trans-membrane pressure difference  
 $P_m$  Solute permeability coefficient for SK model  
 $P_s$  Overall permeability coefficient  
 $R_o$  Observed rejection ( $1 - C_p / C_b$ ) (dimensionless)  
 $R_r$  Real rejection ( $1 - C_m / C_p$ ) (dimensionless)  
Sh Sherwood number  
Re Reynolds number  
Sc Schmidt number  
 $d_H$  Hydraulic diameter  
 $u$  Flow velocity  
 $\nu$  Kinematics viscosity  
 $D$  Solute diffusivity  
 $L$  Length of the tube channel  
 $\alpha, \beta, \gamma$  Parameter experimentally  
 $\delta$  Film thickness  
 $\pi$  Osmotic pressure  
 $\sigma$  Reflection coefficient, 0 for no rejection, 1 for total rejection

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### الخلاصة:-

عندما يكون لدينا منظومة تعمل بالتنافذ العكسي تحت ضغط معين فان ظاهرة الاستقطاب (concentration polarization) تكون النتيجة الحتمية بتواجدها على سطح غشاء المنظومة بسبب زيادة الضغط الاوزموزي هناك مما يؤدي إلى ضعف في أداء المنظومة وهبوط بالضغط تكون نتيجته نقصان في كمية المياه الناتجة ونقصان في معامل انتقال الكتلة وكذلك النقصان في نسبة الفصل.

إن المشكلة الحقيقية هي في معرفة مدى دقة تأثير ظاهرة الاستقطاب (concentration polarization) على نسبة الفصل وعلى كمية المنتج. لذا فانه في هذا البحث سوف نستخدم غشاء من نوع (Spiral wound) والمصنع من مادة السليلوز اسيتيت مع استخدام المحلول القياسي (NaCl-water) لغرض الاختبار بتركيز تصل إلى 2000 ppm.

إن الهدف من هذا البحث هو معرفة وتحليل ظاهرة الاستقطاب (concentration polarization) باستخدام نوعين من الموديلات الرياضية وهي (combine film theory model and empirical correlation) وقد اظهرت التجارب النهائية إن هذه الظاهرة تتأثر بنوع الموديل الرياضي المستخدم.

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