Modeling of Hollow-Fiber Membrane System During Ultrafiltration

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ABSTRACT:

The present study aims to evaluate the performance of hollow fiber membrane module during ultrafiltration of aqueous solutions. The model is represented by a set of differential equations for permeate and residue pressure drop and volumetric flow rates in the axial direction, beside the principle equations of both solvent and solute fluxes through the membrane, while osmotic pressure was neglected in model equations. The shell and tube module type was considered where feed pass in the shell and permeate in the bore side. Tortousily factor of membrane pores in addition to concentration polarization modulus were taken into account in calculations. The model was solved numerically with the help of suitable program in both cocurrent and countercurrent flow pattern and comparison of results were carried out.

Key Words:

Ultrafiltration / Modeling / Hollow-Fiber / Membrane / Counter Current / Cocurrent.

INTRODUCTION:

Ultrafiltration is defined as a pressure deriven membrane process where solvent is the transferred phase and the solute is kept behind, depending on the difference between their molecular size, weight, or shape ^(1,4,13). Osmotic pressure is insignificant factor and the required operating pressure is much lower than that of reverse osmosis processes. For the case study, which is considered in the present work, uranyl nitrate aqueous solution is to be purified.

The purpose of purification is to get a permeate of concentration less than 1 ppm⁽²⁾. Molecular size of the solute as estimated from data available in references ^(1, 3) is arrount the mid-point in the range of ultrafiltration separation ^(2, 4). Previous works ⁽⁵⁻⁹⁾ dealt with separation of different solutions using spiral-wound reverse osmosis system with cellulose acetate membrane, where solution concentration range of 20-40,000 ppm were investigated. The most important characteristic of hollow-fiber membrane module, is the very high surface / volume ratio, in addition to low processing pressure which makes it attractive for comparison with other processes. So, a complementary technique with solvent extraction for separation is introduced. Hybrid separation system can thus be used for different feed concentration conditions. Several studies investigated the performance of both spiral wound and hollowfiber membrane modules on a pilot plant scale in one hand and from the mathematical formulation on the other hand ⁽¹⁰⁻¹²⁾. From the flow pattern point of view, cross-flow accompaning spiral wound module is compared with the parallel flow in hollow fiber one. This in turn may be cocurrent or countercurrent. The main variation of system variables takes place in the axial direction. In the present study, negligible variation in radial

direction was assumed. Since the hollow fibers are nearly uniformly distributed across the bundle. In addition, pores tortousity factor of the value of 1.4 was considered, where steady state operation was the condition basis for model derivation.

MODEL:

Modeling of hollow-fiber module system with different degrees of complexity was developed, depending on the assumptions used in model derivation. Among the assumptions is the concentration profile model in the membrane as found in literature and presented in figure (1). Model (a) is over simplified, while both models (b) and (c) are suitable for separation of large molecular size ultrafiltration. Model (d) is used for reverse osmosis, gas separation and ultrafiltration of low concentration solutions ⁽¹³⁾.

In ultrafiltration, separation is based mainly on a sieving mechanism and not on solution and diffusion. Therefore, friction model was assumed, where the solvent flow through the membrane is mainly laminar convective flow. Hence, the solvent flow can be estimated as Poiseuille flow. The solute flux depends upon diffusion, friction and convective flow ⁽¹³⁾. Also, the flow in bores is laminar, and Poiseuille equation describes the behavior of this stream. On the other hand, shell side fluid is in turbulent flow, hence, Fanning equation was used ⁽¹⁴⁾. The model equations were formulated in generic form, and then, it was solved for the specific case of removal of uranyl nitrate at low concentration. The following equation set describe the system. 1. Water flux, which is independent in osmotic pressure ⁽¹³⁾ is represented by:

$$J_{w} = \frac{A_{w}}{t_{m} \cdot f_{t}} \cdot \Delta P \qquad (1)$$

where:

J $_{\rm w}$: water flux, m/s

A w: water permeability $m^2.s^{-1}.Pa^{-1}$

t $_{\rm m}~$: membrane thickness, m

 f_t : tortousity factor, dimensionless

 Δ P: pressure drop across membrane, Pa.

2. Solute flux equation

$$J_{s} = M \cdot C_{r} (1-R) \cdot J_{w}$$
 (2)

where:

 J_s : solute flux, kg . m⁻². s⁻¹

M : concentration polarization modulus = -----

 C_{b}

 C_{w}

 C_b : bulk concentration, kg . m⁻³

3. Permeate volumetric flux, J_v :

$$J_v = J_w + J_s / \rho_s \tag{3}$$

where:

- ρ_s : solute density , kg . $m^{\text{-}3}$
- 4. Area of transfer, A:

$$A = \pi d_o \cdot L.N \tag{4}$$

- d_o : hollow-fiber outer diameter.
- L : length of hollow-fibers bundle , m.
- 5. Steady state mass balance for the solute in differential element of membrane thickness dr :

$$dc
 J_w \cdot c - D \cdot - = 0
 dr
 dr
 (5)$$

which by integration gives:

$$C_{w} \qquad J_{w} / k$$

$$---= exp \qquad (6)$$

$$C_{b}$$

where :

 $C_{\rm w}$: concentration polarization at membrane wall, kg . $m^{\text{-}3}$

k : mass transfer coefficient, m . s^{-1}

6. Pressure drop in bores is assumed to follow Poiseuilli equation:

$$\frac{d P_p}{dz} = \frac{128 \cdot \mu_p \cdot Q_p}{\pi \cdot d_i^4}$$
(7)

where :

- P_p : permeate pressure, Pa,
- μ_p : permeate viscosity, centipoise,
- Q_p : permeate volumetric flow rate, $m^3.s^{-1}$,
- z : axial coordinate,
- d_i : inner bore diameter.

7. Permeate volumetric flow rate gradient:

$$\frac{dQ_{p}}{dz} = \pi d_{o} \cdot J_{v}$$
(8)

8. Overall volumetric balance:

$$Q_{\rm f} = Q_{\rm p} + Q_{\rm b} \tag{9}$$

9. Component mass balance:

$$Q_{f} \cdot C_{f} = Q_{p} \cdot C_{p} + Q_{b} C_{b}$$
 (10)

 Q_f . C_f : feed volumetric flow rate and concentration respectively.

10. Bulk pressure drop was assumed to follow Darcy' equation, since it is turbulent flow ⁽¹⁴⁾

$$dP_{b} - 32 f \cdot Q_{b}$$

$$- \frac{2}{dz} = - \frac{1}{\pi^{2} \cdot g \cdot D^{5}}$$
(11)

where:

- f : friction factor of bulk flow, Pa. m^{-1}
- D : bundle diameter, m
- 11. Pressure gradient pressure gradient inside membrane pores is assumed to follow Poiseuille equation, where the flow is laminar:

Pressure drop inside the pores depends partially on friction factors between the different species. These are: solvent-solute, solventmembrane and solute-membrane. As mentioned by Wankat⁽¹³⁾ these factors are difficult to be predicted. Therefore, they can be collected in one factor and included in the equation of pressure drop, which yields

$$d P_{m} = -128 \cdot \mu_{p} \cdot Q_{p}$$

$$dr = \pi \cdot d_{m}^{4}$$

$$(12)$$

where :

 P_m : pressure inside the pore ,

- d_m : pore diameter,
- r : pore length coordinate,
- Q_c : correction factor of frictions inside the pores.
- 12. Finally, permeate concentration can be expressed in terms of fluxes as follows:

$$C_{p} = ----$$
(13)
$$J_{v}$$

The above equations were solved with boundary conditions:

when the flow of both streams are countercurrent. In case of cocurrent, the boundary conditions are the same except the pressure of permeate equals 1 atm at the end of the module length, i.e.

$$P_p = 1$$
 at $z = L$

Figure (2) shows both cocurrent and countercurrent systems schematically with boundary conditions.

Equations (7), (8), (11) and (12) can be rearranged to yield:

$$\frac{d^2 P}{dz^2} = \pi d_o C_1 (P_b - P_p)$$
(14)
$$\frac{dz^2}{dz^2}$$

$$\frac{d P_{b}}{d z} = C_{2} \cdot (Q_{f} - \frac{1}{C_{1}} - \frac{d P_{p}}{d z})^{2}$$
(15)
$$\frac{d z}{C_{1}} = C_{2} \cdot (Q_{f} - \frac{1}{C_{1}} - \frac{1}{C_{2}})^{2}$$
(15)

and

$$\frac{d P_m}{d r} = C_3 \cdot \frac{Q_c}{C_1} \cdot \frac{d P_p}{d z}$$
(16)

Equation (14) to (16) show the linear relation between permeate pressure drop and pores pressure drop, and a quadratic relation between bulk pressure drop and that of permeate. Constants C_1 , C_2 and C_3 are as follow:

$$C_1 = 128 \ \mu_p / \pi \ di^4$$
, $C_2 = 32 \ f / \pi \ g$
 $C_3 = 128 \ \mu_p / \pi \ d^4_m$.

Implicity of the last three equation tends the solution to a numerical one, beside the arise of the two-points boundary value problem.

CASE STUDY:

Table (1) presents the value of constants and parameters used in model solving as a case study.

Table ((1)): I	Numerical	values	of	parameters	used	in	model	solv	ing

Parameter	Numerical value				
Membrane characteristics *:					
hollow-fiber inside diameter	300 micron				
hollow-fiber outside diameter	500 micron				
cartirage diameter	25.4 cm				
Number of hollow fibers in cartirage	1300				
Length of hollow fiber	1 m				
Operating conditions:					
Feed temperature	25 °C				
Feed concentration	$0.2 \text{ kg} \cdot \text{m}^{-1}$				
Feed volumetric flow rate	$8.33 \times 10^{-5} \text{ m}^3.\text{s}^{-1}$				
Feed pressure	5 x 10 ⁵ Pa				
Permeate pressure	1 x 10 ⁵ Pa				

* Data obtained from reference (13).

As mentioned in literature $^{(4, 13)}$ ultrafiltration may interfere with reverse osmosis from the point of view of molecular size and weight of the solute. It is referred to that with molecular weight greater than 300 or molecular size in the range of 10-200 Å, the membrane process is considered to lay in ultrafiltration zone. For the case of uranyl nitrate which has the molecular formula UO₂(NO₃)₂.6H₂O, the molecular weight is 502, and the molecular size is 89.3 Å as calculated depending on data given in reference (3).

An important relation between pore size of the membrane and solute molecular size must be satisfied. That is the smaller particle size must be greater than the largest pore size. If the largest particle size is only one half the smaller pore size; zero percent retention is obtained. This condition can be fulfilled by proper manufacturing technique for membrane, in case of hollow fiber membrane, the skin layer is the inner layer of the fiber. Also, complexing agent can be used to increase molecular size of solute ⁽²⁾.

The results of solution of model equations are presented. Figure (3) shows the variation of pressure of bulk and permeate streams for both cocurrent and countercurrent flow pattern. For the case of cocurrent, a numerical problem exist, that is the boundary condition of pressure is known at z= o for the feed (bulk) stream and at z= L for the permeate. So two points boundary value problem arises in model solving. For the case of countercurrent flow, one point boundary is the case, and no solution difficulty was found. In figure (1) variation of pressure for the different streams is shown where feed of $5x10^5$ Pa was used. In figure (4) concentrations of bulk and permeate streams are presented. Sensible difference at z= o and z= L were observed with greater rate of variation in case of cocurrent. The balance of volumetric flow rates are presented in figure (5). Unsensible changes in bulk variables for both flow patterns, which is attributed to low concentration of the feed.

CONCLUSION:

The present study introduce ultrafiltration as reverse osmosis alternative membrane process for low concentration solutions. The process pressure is much lower than that for reverse osmosis. Countercurrent flow pattern yields permeate concentration of about one third that if cocurrent system is used. The study may help in estimation of the friction factors insides the pores, and thus high lights the application of friction model in future.

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نمذجة منظومة الألياف المفرغة أثناء الترشيح الدقيق

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تهدف الدراسة الحالية إلى تقييم أداء نموذج أغشية الألياف المفرغة أثناء الترشيح الدقيق للمحاليل المائية. ويمثل النموذج الرياضى مجموعة من المعادلات الرياضية التفاضلية لكل من التيار الراشح والتيار المتبقى معبرة عن الانخفاض فى الضغط ومعدل السريان الحجمى فى الاتجاه المحورى بالاضافة إلى المعادلات الاساسية الخاصة بالفيض لكل من المذيب والذاب خلال الغشاء وقد تم اهمال الضغط الاسموزى فى معادلات النموذج الرياضى. وأخذ فى الاعتبار نموذج الطبقة والانابيب المشابهة للمبادلات الحرارية حيث يمر تيار التغذية حول الانابيب الشعرية ويمر الراشح بداخلها. وكذلك اخذ عامل الاعوجاج فى المسارات المسامية الغشاء و عامل تركيز الاستقطاب. وتم حل النموذج فى كل من حالتى المتوافق والمتعاكس وقورنت النتائج فى الحالتين.





Fig.(1): Schematic concentration profile in membrane system (a) without concentration polarization (b) with polarization (c) Gel-polarization model (d) Friction model at low concentration feed.





Fig.(2): Flow pattern inside the module

a: cocurrent

b: countercurrent



Fig.(3): Variation of bulk and permeate pressure for both cocurrent and counter current systems.



Fig.(4): Variation of bulk and permeate concentrations for cocurrent and counter current systems.

