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Wan Ramli Wan Daud^a

^a Department of Chemical & Process Engineering, Universiti Kebangsaan Malaysia, Bangi, Malaysia

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Rate-Based Design of Non-fouled Cross-Flow Hollow Fiber Membrane Modules for Ultrafiltration

Wan Ramli Wan Daud*

Department of Chemical & Process Engineering, Universiti Kebangsaan
Malaysia, Bangi, Malaysia

ABSTRACT

Although ultrafiltration has replaced many liquid phase separation equipment, it is still considered as a “non-unit operation” process because the sizing of the equipment could not be calculated using either the equilibrium stage or the rate-based methods. Previous design methods using the dead-end and the complete-mixing models are unsatisfactory because the dead-end model tends to underestimate the membrane area due to the use of the feed concentration in the driving force while the complete-mixing model tends to overestimate the membrane area due to the use of the more concentrated rejection concentration in the driving force. In this paper, a cross-flow model for ultrafiltration is developed by considering mass balance at a differential element of the cross-flow

*Correspondence: Wan Ramli Wan Daud, Department of Chemical & Process Engineering, Universiti Kebangsaan Malaysia, 43600 UKM Bangi, Malaysia; Fax: 603-89252546; E-mail: wramli@vlsi.eng.ukm.my.

module and then integrating the expression over the whole module to get the module length. Since the modeling is rate-based, the length of both modules could be expressed as the product of the height of a transfer unit (HTU) and the number of transfer unit (NTU). The solution of the integral representing the NTU of ultrafiltration is found to be the difference between two exponential integrals ($Ei(x)$). The poles of the solution represent the flux extinction curves of ultrafiltration. The NTU for ultrafiltration is found to depend on three parameters: the rejection R , the recovery S , and the dimensionless gel concentration C_g . For any given C_g and R , the recovery, S is limited by the corresponding flux extinction curve. The NTU for ultrafiltration is found to be generally small and less than unity but increases rapidly to infinity near the poles due to flux extinction. The complete-mixing model is reformulated in terms of the same parameters used in the cross-flow model. The design of membrane modules for ultrafiltration taken from case-studies of previous authors is performed using both the complete-mixing and the cross-flow models developed in this paper and a comparative study of the results is carried out. The length of the hollow fiber module and the membrane area calculated using the cross-flow model for ultrafiltration are found to be always smaller than those given by the complete-mixing model.

Key Words: Ultrafiltration; Hollow fiber module design; Cross-flow model; Number of transfer unit; Height of a transfer unit.

INTRODUCTION

Although membrane processes for liquid separation such as ultrafiltration and hyperfiltration (reverse osmosis) have replaced many other separation equipment in many industries, the process design of membrane modules particularly cross-flow modules such as the cross-flow hollow fiber membrane modules for both is still very empirical and not amenable to the normal unit operation design approach of using equilibrium stages or rates of mass transfer. This difficulty renders membrane processes as “non-unit operation” processes.

Previous design methods are only based on either complete-mixing^[1,2] or dead-end models that mimic the experimental dead-end equipment for ultrafiltration and hyperfiltration systems.^[3–6] These models for ultrafiltration and hyperfiltration suffers from various drawbacks, the most important of which is the increasing solute concentration along the tube, which in the case of ultrafiltration and hyperfiltration decreases the driving force for the solvent flux across the membrane further down the tube. This results in a smaller overall membrane area for the same separation duty compared to that predicted by the complete-mixing model, which uses the flux at the



more concentrated retentate condition. The overall area is also bigger than that predicted by the dead-end model, which uses the flux at the more, dilute feed condition. The corresponding decrease in the rejection flow rate along the tube decreases the superficial velocity as well. The second major drawback is that the pressure drop along the tube due to friction decreases the pressure difference along the tube, which is available to drive the flux resulting in a decrease in the flux across the membrane further down the tube. This again tends to decrease the overall size of the membrane area. In other words, the complete mixing model over estimates the area required while the dead-end model underestimates the membrane area required for a given separation duty.

THE COMPLETE-MIXING MODEL

Overall Solute and Solvent Balance of the Complete-Mixing Model

Although the design equations for the complete-mixing model have been developed by many people,^[1-3] it is redeveloped here in terms of dimensionless terms used in this paper so that a better comparison can be made with the cross-flow model developed here. Overall mass balance of the solute for one single hollow fiber module without recycle at steady state using the complete-mixing model as shown in Fig. 1 is given by

$$q_f C_f = q_p C_p + q_r C_r \quad (1)$$

The overall volume balance of the module is given by

$$q_f = q_p + q_r \quad (2)$$

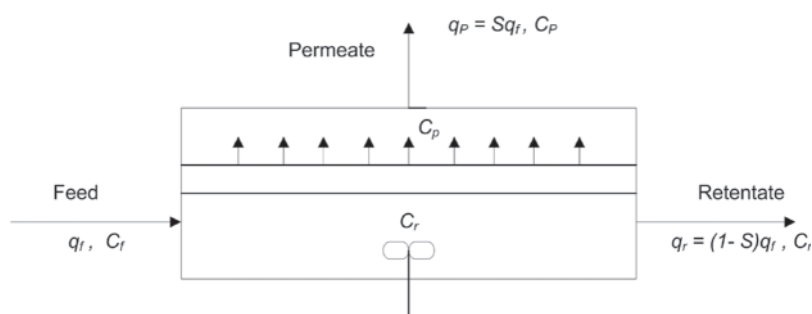


Figure 1. The complete-mixing model of the hollow fiber module without recycle.

where C_f is the feed solute concentration, q_f is the feed volumetric rate, C_p is the permeate solute concentration, q_p is the permeate volumetric flow rate, C_r is the retentate solute concentration, and q_r is the retentate flow rate. Equation (2) can be rewritten as

$$q_r = (1 - S)q_f \quad (3)$$

where S , the recovery is defined by

$$S = \frac{q_p}{q_f} \quad (4)$$

Substituting Eq. (4) into Eq. (1) and solving for the mean solute concentration in the permeate

$$\bar{C}_p = \frac{[C_f - (1 - S)C_r]}{S} \quad (5)$$

The overall rejection in this case, R , is given by

$$R = \frac{(C_r - \bar{C}_p)}{C_r} = 1 - \frac{\bar{C}_p}{C_r} \quad (6)$$

Design Equations for the Complete-Mixing Model of Ultrafiltration Module

Ultrafiltration solvent fluxes of different concentrations of a particular macromolecules or colloids solution increase with applied pressure but taper off to plateaus of maximum fluxes that decrease with increase in solute concentrations at approximately the same pressure. This is caused by the formation of a gel layer at the membrane surface due to the increase in pressure that eventually limits the flux to a plateau where no further increase in flux by increase in pressure is possible. This maximum ultrafiltration solvent flux, J , is often given by

$$J = k \ln \left(\frac{C_g}{C_r} \right) = \frac{q_p}{A} \quad (7)$$

where k is the mass transfer coefficient, A is the membrane area, and C_g is the gel concentration. The membrane module should, therefore, be designed at maximum solvent flux given by Eq. (7). Although the gel layer model in Eq. (7) is problematic because neither does it show the effect of operating parameters such as flow velocity, operating pressure, and feed concentration explicitly on C_g nor does it explain the effect of operating pressure on the



flux.^[7-9] It is still being used extensively to correlate ultrafiltration flux data.^[10] It is because of this reason and because of its simplicity that it is being used in the design method presented in this paper. Following a suggestion by Porter,^[11] the mass transfer coefficient k is calculated using the standard correlation for mass transfer in round pipes in turbulent flow given by

$$\frac{kd}{D} = 0.023\text{Re}^{0.83}\text{Sc}^{1/3} \quad (8)$$

where D is the diffusivity of the solute in the solvent, Re is the Reynolds number given by $\text{Re} = 4\rho q/(\pi\mu d)$, and Sc is the Schmidt number given by $\text{Sc} = \mu/(\rho D)$. Over time the gel layer may precipitate or adsorb a layer of lower permeability material on the membrane surface and fouls the membrane causing the flux to drop. However, the present paper is limited to non-fouled ultrafiltration membranes only and the fouling phenomenon will be taken into account in a future paper.

If there is concentration polarization, the solute flux in $\text{kg}/\text{m}^2 \text{ sec}$ is given by

$$J_s = \beta C_r(1 - \phi)J \quad (9)$$

where β is the polarization parameter given by $\beta = C_w/C_r$, C_w is the concentration of solute at the membrane surface and ϕ is the fraction of pores rejecting the solute. The values of β and ϕ are difficult to estimate theoretically because of the complex nature of the concentration polarization phenomenon in membranes.^[12] The values of both parameters must be determined empirically for specific types of membrane and used judiciously within the experimental range of operating parameters.

The solute mass balance also gives

$$J_s = C_p J = \frac{C_p q_p}{A} \quad (10)$$

Solving for C_p and then substituting it into Eq. (6) results in

$$R = 1 - \beta(1 - \phi) \quad (11)$$

If there is no concentration polarization, then $\beta = 1.0$ and $R = \phi$. Equation (7) implies that the area of membrane required is given by

$$A = \frac{q_p}{[k \ln(C_g/C_r)]} \quad (12)$$



If both sides of Eq. (5) is divided by C_r and then C_p/C_r in Eq. (6) is substituted into the result, then

$$\frac{C_r}{C_f} = \frac{1}{1 - SR} \quad (13)$$

If Eq. (13) is substituted in Eq. (12), then the total area is given by

$$A = \frac{q_p}{[k \ln(C_g(1 - SR))]} \quad (14)$$

where $C_g = C_g/C_f$. The length of the module is given by

$$L = \frac{q_p}{[n\pi dk \ln(C_g(1 - SR))]} \quad (15)$$

where n is the number of tubes and d is the diameter of each tube.

THE CROSS-FLOW MODEL

Overall Solute and Solvent Balance of the Cross-Flow Model

A solute mass balance at an infinitesimally small section of the membrane module dA of the cross-flow model as shown in Fig. 2 yields

$$qC = (q - dq_p)(C + dC) + dq_p C_p \quad (16)$$

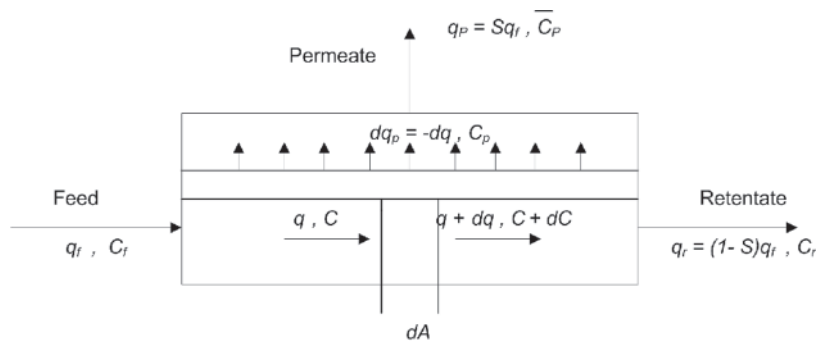


Figure 2. Cross-flow membrane module without recycle.



Non-fouled Cross-Flow Hollow Fiber Membrane Modules

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Rearranging Eq. (16) and ignoring higher order terms gives

$$-\frac{dq_p}{q} = \frac{dC}{(C_p - C)} \quad (17)$$

The rejection R of the solute along the tube is assumed constant and is defined by

$$R = \frac{(C - C_p)}{C} = 1 - \frac{C_p}{C} \quad (18)$$

By applying Eq. (18), the right hand side of Eq. (17) is replaced by

$$\frac{dC}{(C_p - C)} = -\frac{dC}{CR} \quad (19)$$

At any point along the membrane, the relationship between q and q_f is similar to the relationship between q_r and q_f in Eq. (3), that is

$$q = (1 - S)q_f \quad (20)$$

If Eq. (20) is substituted in the left hand side of Eq. (17) and the resulting term dq_p/q_f is replaced by dS by virtue of Eq. (3), then the left hand side of Eq. (17) becomes

$$-\frac{dq_p}{q} = -\frac{dS}{(1 - S)} \quad (21)$$

Hence Eq. (17) could be rewritten as

$$\frac{dC}{dS} = \frac{CR}{(1 - S)} \quad (22)$$

Mulder^[4] obtained the same equation as Eq. (22) through a different route. The present derivation is more direct and easier to explain in terms of fundamental principles. If this equation is integrated from C_f to C_r , then

$$C_r = (1 - S)^{-R} C_f \quad (23)$$

or

$$N = (1 - S)^{-R} \quad (24)$$

where $N = C_r/C_f$ is defined as the concentration factor. Hence by combining Eq. (23) with Eq. (5), the mean concentration of the permeate is given by

$$\bar{C}_p = \left\{ \frac{[1 - (1 - S)^{1-R}]}{S} \right\} C_f \quad (25)$$



Volumetric Flow Rate and Solute Concentration Relationship

Before attempting to formulate and solve the rate based design equations, the relationship between the volumetric flow rate and the solute concentration in the tube must be established. Rearranging Eqs. (19) and (21) gives

$$\frac{dq}{q} = -\frac{dC}{RC} \quad (26)$$

If Eq. (26) is integrated from C_f to C and q_f to q , then

$$\ln\left(\frac{q}{q_f}\right) = -\left(\frac{1}{R}\right) \ln\left(\frac{C}{C_f}\right) \quad (27)$$

or simply

$$\frac{q}{q_f} = \left(\frac{C}{C_f}\right)^{-1/R} \quad (28)$$

If $C = C_r$ and $q = q_r$ and Eq. (3) is substituted in Eq. (28) above then the latter reduces to Eq. (23). This shows that the derivation is consistent. Although Mulder^[4] derived Eq. (22) using an integral balance, he did not go further to derive Eq. (26) and hence failed to get Eq. (28) that is crucial in solving the final rate-based design equations as shall be shown in the following sections.

Design Equations for the Cross Flow Ultrafiltration Membrane Module

The solvent mass balance across an infinitesimal area of the ultrafiltration membrane gives

$$J\pi Dn dx = \pi Dnk \ln\left(\frac{C_g}{C}\right) dx = dq_p \quad (29)$$

where n is the number of tubes and D is the diameter of each tube. This equation could only be solved if the volumetric flow rate could be expressed in terms of the concentration. According to Eqs. (26) and (28), the right hand side of Eq. (29) is given by

$$dq_p = \frac{q}{RC} dC = \frac{q_f}{RC} \left(\frac{C}{C_f}\right)^{-1/R} dC = \frac{q_f}{RC_f^{-1/R}} C^{-(1+R)/R} dC \quad (30)$$



If Eq. (30) is substituted into Eq. (29), and the resulting equation is integrated from $x = 0$ to $x = L$ and from $C = C_f$ to $C = C_r$, then

$$L = \int_0^L dx = \frac{q_f}{\pi dn k R C_f^{-1/R}} \int_{C_f}^{C_r} \frac{C^{-(1+R)/R}}{\ln(C_g/C)} dC \quad (31)$$

The area of membrane required is given by

$$A = \pi dn \int_0^L dx = \frac{q_f}{k R C_f^{-1/R}} \int_{C_f}^{C_r} \frac{C^{-(1+R)/R}}{\ln(C_g/C)} dC \quad (32)$$

Equation (32) could be rewritten in terms of dimensionless variables.

$$\frac{A}{q_f/k} = \int_1^{C_r/C_f} \frac{(C/C_f)^{-(1+R)/R}}{R \ln[(C_g/C_f)/(C/C_f)]} d(C/C_f) \quad (33)$$

or even more compact as

$$\mathbf{A} = \int_1^{(1-S)^{-R}} \frac{\mathbf{C}^{-(1+R)/R}}{R \ln[\mathbf{C}_g/\mathbf{C}]} d\mathbf{C} \quad (34)$$

where \mathbf{A} is the dimensionless area, \mathbf{C} is the dimensionless solute concentration in the tube, and \mathbf{C}_g is the dimensionless gel concentration given by

$$\mathbf{A} = A/(q_f/k) \quad \mathbf{C} = C/C_f \quad \mathbf{C}_g = C_g/C_f \quad (35)$$

Equations (30) and (31) could be rewritten in the NTU form by

$$L = \left(\frac{q_f}{n \pi d k} \right) \int_1^{(1-S)^{-R}} \frac{\mathbf{C}^{-(1+R)/R}}{R \ln[\mathbf{C}_g/\mathbf{C}]} d\mathbf{C} = H_T N_T \quad (36)$$

where H_T is the height of a transfer unit based on the tube side given by

$$H_T = \left[\frac{q_f}{(n \pi d k)} \right] \quad (37)$$

and N_T is the number of transfer unit based on the tube side given by

$$N_T = \int_1^{(1-S)^{-R}} \frac{\mathbf{C}^{-(1+R)/R}}{R \ln[\mathbf{C}_g/\mathbf{C}]} d\mathbf{C} \quad (38)$$



RESULTS AND DISCUSSION

Analytical Solution of Design Equation

Equations (38) for the NTU of the ultrafiltration membrane module can be integrated by substituting $z = (\ln C - \ln C_g)/R$ such that

$$N_T = \frac{C_g^{-1/R}}{R} \int_{\ln[C_g/(1-S)^{-R}]/R}^{\ln C_g/R} \frac{e^z}{z} dz \quad (39)$$

Equation (39) can then be expressed in terms of standard exponential integrals as

$$N_T = \frac{C_g^{-1/R}}{R} \left\{ \text{Ei} \left(\frac{\ln C_g}{R} \right) - \text{Ei} \left(\frac{\ln[C_g/(1-S)^{-R}]}{R} \right) \right\} \quad (40)$$

where

$$\text{Ei}(x) = \int_{-\infty}^x \left(\frac{e^t}{t} \right) dt \quad (41)$$

If the series expansion for this integral given by^[13]

$$\text{Ei}(x) = \gamma + \ln x + \sum_{n=1}^{\infty} \frac{x^n}{nn!} \quad (42)$$

where γ is the Euler constant, is substituted in Eq. (40) then

$$N_T = \frac{C_g^{-1/R}}{R} \left\{ \ln \left(\frac{\ln C_g}{\ln[C_g/(1-S)^{-R}]} \right) - \sum_{n=1}^{\infty} \frac{\{\ln[C_g/(1-S)^{-R}]/R\}^n - (\ln C_g/R)^n}{nn!} \right\} \quad (43)$$

Equation (43) has a pole at

$$S = 1 - C_g^{-1/R} \quad (44)$$

or

$$R = -\frac{\ln C_g}{\ln(1-S)} \quad (45)$$

corresponding to zero flux and $N_T = \infty$. Design charts of NTU for ultrafiltration hollow fiber module are shown in Figs. 3–5.



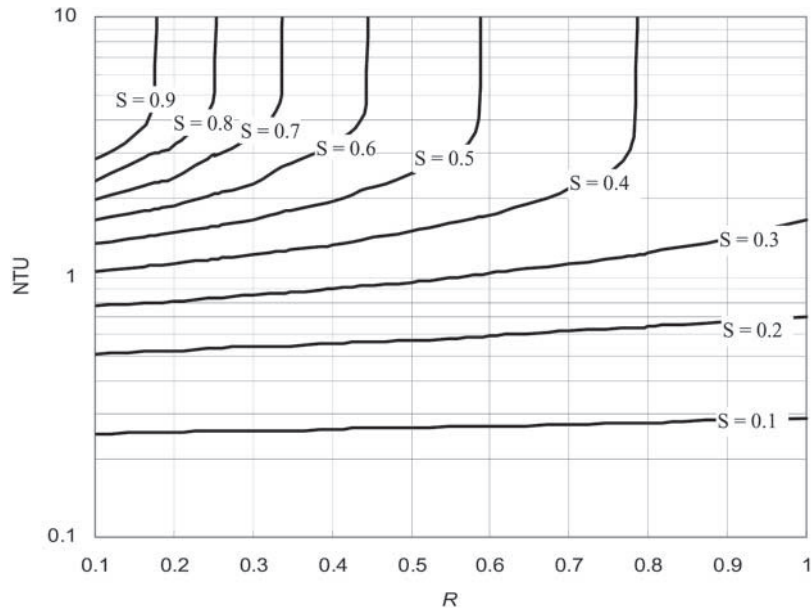


Figure 3. Design chart for NTU of ultrafiltration hollow fiber module for $C_g = 1.5$.

The Number of Transfer Units as Functions of S and R

Figures 3–5 show that the NTU for constant recovery S at first increases slowly as the rejection R is increased and then increases sharply as the pole or the dimensionless gel concentration is reached due to the zero flux or flux extinction there. The NTU value is generally less than 1.0 but increases sharply to infinity at the flux extinction point. At a lower value of the dimensionless gel concentration C_g , the flux extinction is reached much more quickly than at a larger C_g .

As C_g is increased, the flux extinction is reached less quickly at the outlet and which for smaller S tends to be outside the domain of R . The NTU also increases as S is increased for all cases. The effect of the poles is to increase the NTU sharply near it. In other words, the length of the hollow fiber ultrafiltration module and therefore the area at the poles is very large. It implies that for very high rejection R , the operating point should be at a low S so that the area is not so large.

Flux Extinction Curves

Not all possible pairs of values of rejection R , and recovery S , however, have NTU values due to the flux extinction. This effect can be seen clearly on

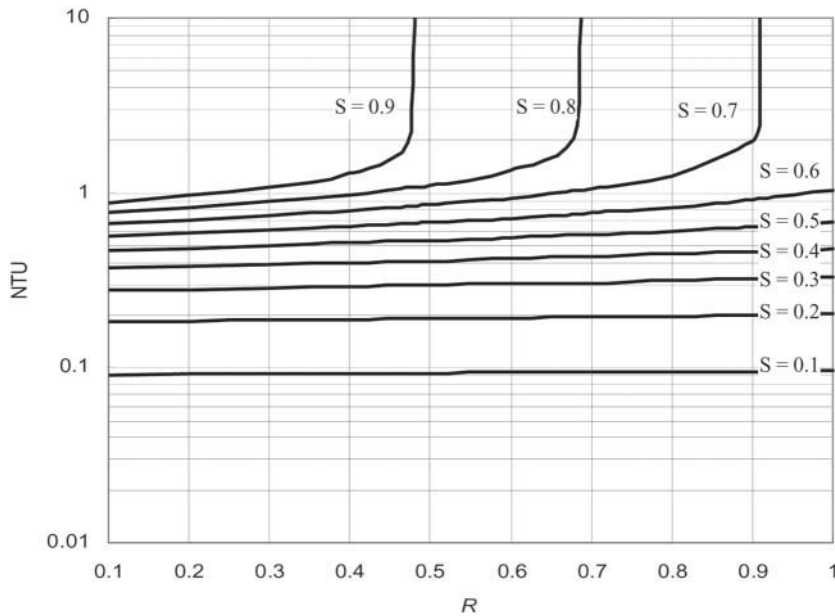


Figure 4. Design chart for NTU of ultrafiltration hollow fiber module for $C_g = 3$.

the flux extinction curves given by Eq. (44), which are plotted in Fig. 6. The flux extinction curves represent the maximum attainable value of recovery, S for any given C_g and R . It means that for any given C_g and R the recovery, S , is limited by the corresponding flux extinction curve. The flux extinction curves also show that the feasible value of recovery, S , is lower at lower values of C_g compared to that at higher C_g . Conversely, solutions with higher C_g can achieve higher rejection, R , and higher recovery, S , compared with those with a lower value of C_g .

Sample Calculations of Membrane Areas

The required membrane areas for several cases of ultrafiltration taken from Geankoplis,^[2] and Wankat^[3] calculated using the complete-mixing and the cross-flow models are compared in Table 1. For the purpose of this calculation, the values of β and ϕ are taken from Geankoplis^[2] and Wankat.^[3] The design procedure is given in Fig. 7. Once the length of the hollow fiber is determined by using Eq. (36), then the area of the membrane in the module is given by

$$A = n\pi dL \quad (46)$$



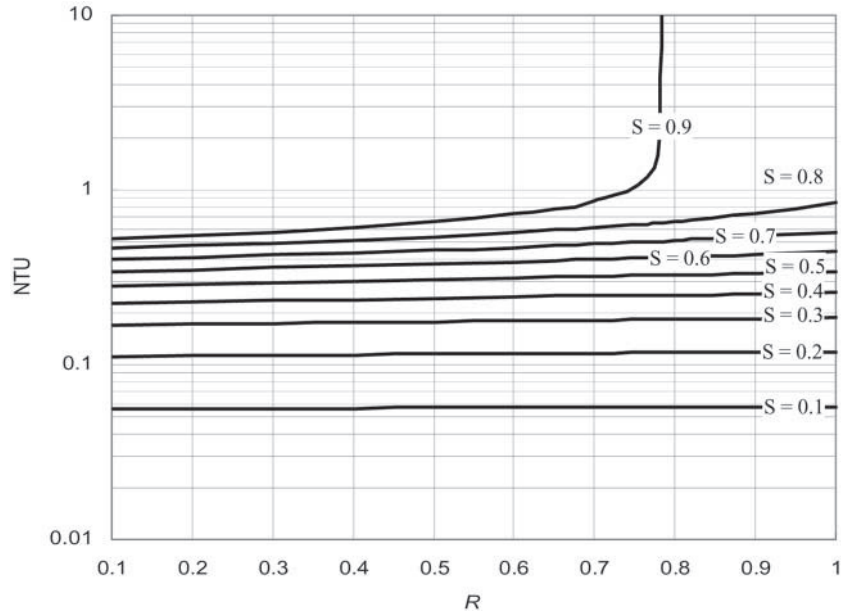


Figure 5. Design chart for NTU of ultrafiltration hollow fiber module for $C_g = 6$.

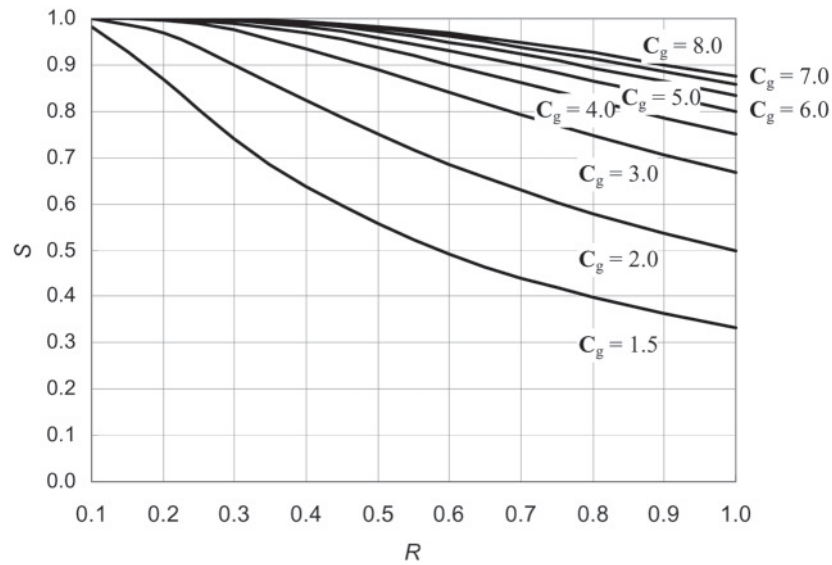


Figure 6. The flux extinction curves of ultrafiltration.



Table 1. Comparison of design of hollow fiber ultrafiltration membrane module using complete-mixing and cross-flow models $q_f = 0.01 \text{ m}^3/\text{sec}$, $D = 0.0005 \text{ m}$, $n = 1 \times 10^5$.

Feed	Casein solution ^[3]	Flexible polymer solution ^[3]	Albumin solution ^[3]	Albumin solution ^[3]	Colloidal solution ^[2]
C_f (kg/m ³)	20.41	26.0	41.67	250	50
C_g (kg/m ³)	162.79	80.0	818.18	818.18	300
C_g	7.98	3.08	19.64	3.27	6.0
k (m ³ /m ² sec)	1.62×10^{-5}	2.31×10^{-5}	2.43×10^{-5}	2.43×10^{-5}	2.00×10^{-5}
β	1.4	1.0	0.996	0.996	1.0
ϕ	0.964	0.996	0.996	0.996	1.0
S	0.4	0.5	0.5	0.5	0.5
R	0.95	0.996	0.992	0.992	1.00
Complete-mixing model					
A (m ²)	386.1	993.6	179.5	822.1	455.0
L (m)	2.46	6.33	1.14	5.24	2.90
Cross-flow model					
NTU	0.2134	0.6440	0.1854	0.5934	0.5940
H_T (m)	3.93	2.75	2.62	2.62	3.18
L (m)	0.84	1.77	0.49	1.55	1.89
A (m ²)	131	278	76.3	244	297

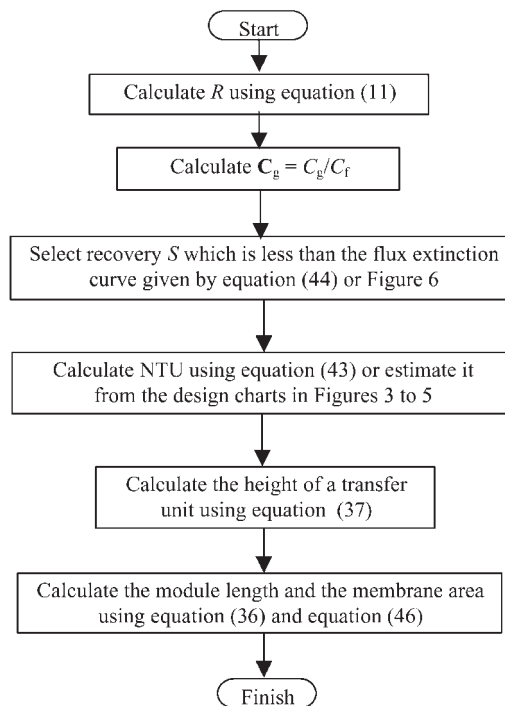


Figure 7. Cross-flow design flowchart for the hollow fiber ultrafiltration membrane module.

The length of the hollow fiber module and therefore the membrane area calculated using the cross-flow model is always lower than those given by the complete-mixing model. This is because of the use of a decreasing solvent flux driving force along the hollow fiber membrane due to increasing concentration of solutes along the hollow fiber in the cross-flow model. The NTU values are generally less than 1.0.

CONCLUSIONS

It can be concluded that the length and membrane area of hollow fiber modules for ultrafiltration could be determined using the cross-flow model whose solution could be expressed in terms of the NTU and HTU. The solution of the integral representing the NTU of ultrafiltration is found to be the difference between two exponential integrals (Ei). The poles of the



solution represent the flux extinction curves of ultrafiltration. The NTU for ultrafiltration is found to depend on three parameters: the rejection R , the recovery S , and the dimensionless gel concentration C_g . For any given C_g and R , the recovery, S is limited by the corresponding flux extinction curve. The NTU for ultrafiltration is found to be generally small and less than unity but increases rapidly to infinity near the poles due to flux extinction. The length of the hollow fiber module and the membrane area calculated using the cross-flow model for ultrafiltration are found to be always smaller than that given by the complete-mixing model. This is because of the decreasing solvent flux driving force along the hollow fiber membrane due to increasing concentration of solutes in the cross-flow model.

NOMENCLATURE

A	membrane area
A	dimensionless membrane area
C	dimensionless solute concentration
C_f	feed solute concentration
C_g	dimensionless gel concentration
C_g	gel concentration
C_p	permeate solute concentration
C_r	retentate solute concentration
d	diameter of tube
D	diffusivity of the solute in the solvent
H_T	height of a transfer unit
J	solvent flux
J_s	solute flux
k	mass transfer coefficient
L	module length
n	number of tubes
N	concentration factor
N_T	number of transfer units
q	volumetric flow rate
q_f	feed volumetric flow rate
q_p	permeate volumetric flow rate
q_r	retentate volumetric flow rate
R	rejection
Re	Reynolds number
S	recovery
Sc	Schmidt number
z	dummy variable



Greek Letters

β	polarization parameter
γ	Euler constant
ϕ	fraction of pores rejecting the solute
μ	viscosity of solution
ρ	density of solution

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