

모델모사를 이용한 막모듈 연결 및 배열이 투과증발 막성능에 끼치는 영향에 관한 연구

염충균[†] · 윤석복 · 박유인*

(주)세프라텍 인천시 남동구 고잔동 719-22

*한국화학연구소, 환경에너지연구센터

(2008년 9월 28일 접수, 2008년 12월 24일 수정, 2008년 12월 24일 채택)

Study on the Effect of Membrane Module Configuration on Pervaporative Performance through Model Simulation

Choong Kyun Yeom[†], Seok-Bok Yoon, and You-In Park*

SepraTek Inc., 719-22 Gojan-dong, Namdong-gu, Incheon 405-821, Korea

*Environment and Energy Research Center, Korea Research Institute of Chemical Technology, Taejon 305-606, Korea

(Received September 28, 2008, Revised December 24, 2008, Accepted December 24, 2008)

요약: 본 연구는 공정모사를 통해서 막모듈 배열 및 연결과 막모듈 내에서 공급 잔류액의 온도 변화가 투과증발 공정에 끼치는 영향을 관찰하였으며 이를 위해서 기본 막모듈이 직렬 혹은 병렬로 연결되어 있는 모듈들의 조합을 통한 투과탈수공정을 예측할 수 있는 모델식들을 확립하였다. 에탄올/물 혼합물을 모델 혼합물로 사용하였고 모델식들에 포함되어 있는 투과 파라메타들을 직접 실험에 의해 구하여 사용함으로써 모사의 현실성을 높였다. 모사를 통해서 모듈의 배열방식, 모듈 단의 갯수와 단사이의 공급액의 재가열등의 중요성을 검토하였다.

Abstract: This study was focused on the investigation of the effects of membrane module configuration and the temperature of feed retentate flowing along with module length on membrane performance through model simulation. A simulation model of pervaporative dehydration through membrane module assemble in which a number of unit modules are connected in parallel or in series has been established. In this study, ethanol/water mixture was used as model mixture. Some of permeation parameters in the model were quantified directly from the real dehydration pervaporation of ethanol through a lab-made membrane. By adopting the coefficients determined empirically the simulation model could be of more practical value. The simulation of pervaporation with two basic module configurations, that is, parallel connection and series connection, could present the importance of process parameters such as feed rate, module connection mode, number of stages, and inter-stage heating.

Keywords: pervaporation, dehydration, simulation, membrane module, plate and frame

1. Introduction

Among membrane processes, pervaporation is a technique that allows the separation of liquid mixtures through polymeric membrane. Now that the membrane process has been proven in the dehydration separation of organic compounds [1-3], coming of age as a dehydration separation process, attention is turning to en-

ergy saving as well as environmental-friendly separation, promising much greater benefits especially in an era of skyrocketing oil price.

The success of the Pervaporation in industry is highly dependent on progress in two fields: 1) the development of membrane with high performance and 2) the provision of engineering tools to optimize pervaporation within the concept of process design, that is, to search a process circumstance which cannot only max-

[†]주저자(e-mail : ckyeom@sepratek.com)

imize membrane performance but also yield its economical value. The development of membrane has been in a constant progress in recent years, but process technology has been widely neglected even though it is very essential to design and build the real system. In process technology, simulation model is a good tool to analyze and optimize process. In previous works [4-8], the simulation models were proposed to predict pervaporative behavior in process and to optimize the process in terms of membrane area and energy requirement in plate-and-frame and hollow fiber modules, respectively.

This study was focused on the investigation of the effects of membrane module configuration and the temperature of feed retentate flowing along with module length on membrane performance through model simulation. A simulation model of pervaporative dehydration through membrane module assemble in which a number of unit modules are connected in parallel or in series has been established. With help of the model, pervaporation processes through membrane module assembles with several configurations were simulated, respectively, for analyzing and optimizing the pervaporation, and ethanol/water mixture with 5 wt% of water content was used as model mixture. Some of permeation parameters in the model were quantified directly from the real dehydration pervaporation of etha-

nol through a lab-made membrane. By adopting the coefficients determined empirically, the simulation model could be of more practical value. The simulation of pervaporation with two basic module configurations, that is, parallel connection and series connection, could present the importance of process parameters such as feed rate, module connection mode, number of stages, and inter-stage heating.

2. Simulation Model

2.1. Mass and Heat Balances Over Differential Element Volume in Unit Membrane Module

The plate-and frame module in this study is characterized by :

- Dimension of each membrane sheet: length l_m and width w_m
- Height of each feed channel
- Number of membrane sheets making up of single unit module

A schematic representation of mass and heat transfers over the differential element volume in feed channel is shown in Fig. 1. When vacuum is applied at the permeate side, a driving force for permeation, i.e. chemical activity gradient can be developed across membrane thickness. As a result, selective permeation

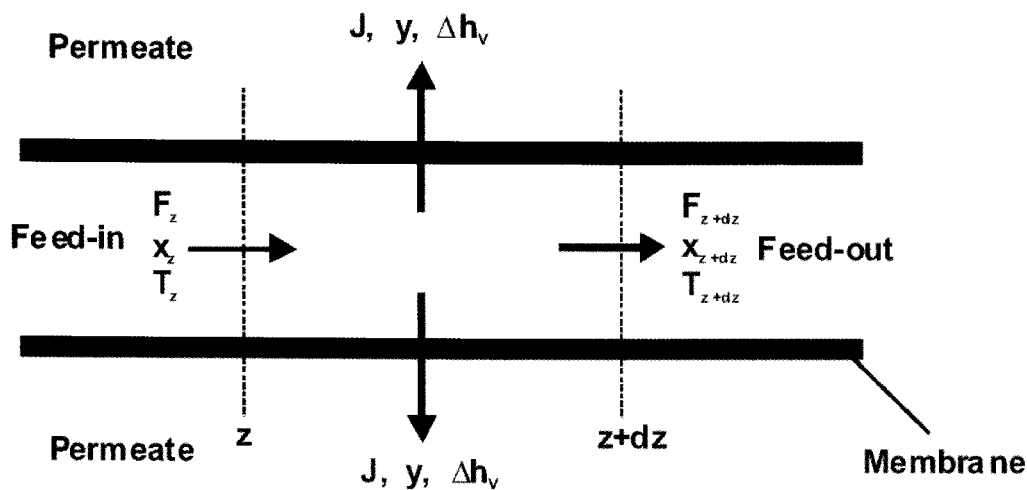


Fig. 1. Mass and heat transfer across membrane during flowing in the differential unit volume of feed channel.

takes place at a certain rate through the membrane, and then feed flow rate as well as feed composition changes over the differential volume as much as the permeation through the membrane. A liquid phase of permeant changes into vapor phase at the permeate side of membrane which is well below saturate vapor pressure, requiring a certain amount of heat or energy corresponding to the vaporization of permeant. The heat of evaporation is supplied from feed flowing in feed channel, so that the feed temperature falls constantly along with feed channel. Therefore three different balances over the differential volume are taken into account as follows;

Mass balance

$$\frac{d}{dz}F = -2Jw_m \tag{1}$$

Concentration balance

$$\frac{d}{dz}(Fx) = -2Jyw_m \tag{2}$$

Heat balance

$$\frac{d}{dz}(Fh_F) = -2J\Delta h_V w_m \tag{3}$$

where F denotes feed flow rate, J total flux, w_m the width of unit membrane sheet, x and y the concentrations of a selectively permeating component in feed and permeate, respectively, h_F the enthalpy of feed flow, and Δh_V the heat of permeant evaporation. Rearranging Eqs. (1)~(3) gives

$$dF = -2Jw_m dz \tag{4}$$

$$dx = \frac{2J}{F}(x-y)w_m dz \tag{5}$$

$$dT = -\frac{2J\Delta h_V}{FC_p}w_m dz \tag{6}$$

where C_p is the heat capacity of feed. The changes in feed flow rate, feed composition, and feed temperature

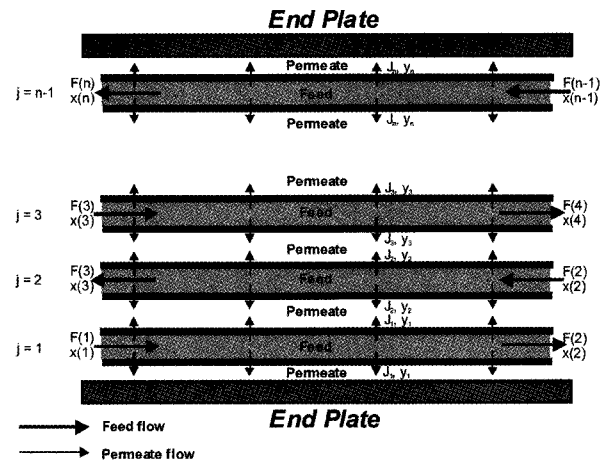


Fig. 2. Representation of the feed and permeate flows in a membrane module.

along with z direction can be determined if flux and permeate composition are expressed as functions of both feed composition and feed temperature, respectively;

$$J = f(x, T) \tag{7}$$

$$y = g(x, T) \tag{8}$$

Hence, flux, feed composition, and permeate composition can also be expressed as a function of location along with z direction. A membrane module unit is structured by stacking a number of membrane sheets with the active layers of a pair of neighboring membranes separated by spacer facing each other for feed channel, as described in Fig. 2. The spaces between sheets are secured for both permeate and feed flows by embedding porous spacers, respectively. The feed channels in the unit membrane module are altogether connected in series.

2.2. Batch Process

In this study, two configurations of membrane modules, series and parallel connections between unit modules are considered in the batch process as shown in Fig. 3. Feed mixture circulates from the feed tank through a configuration of membrane modules. During

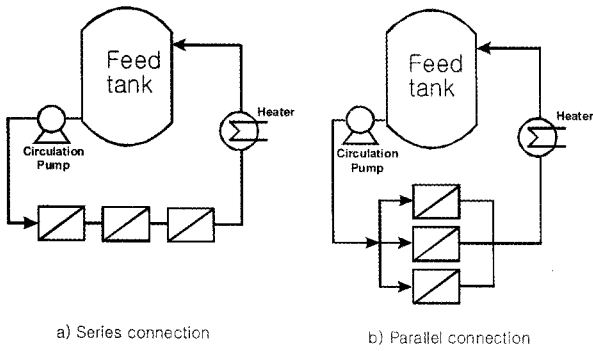


Fig. 3. Module configuration in batch pervaporation process.

the circulation of feed, both the feed amount M_F and the water content of feed in the tank x_F decrease because of water-selective permeation through membrane. Thus total mass change in the feed tank is balanced with the permeation amount for a differential time interval, dt , as follows;

$$dM_F = -J_M A_T dt \tag{9}$$

$$d(M_F x_F) = -J_M y_M A_T dt \tag{10}$$

$$M_F = (M_F)_0 - \int_0^t (A_T J_M) dt \tag{11}$$

where J_M and y_M is average flux and water concentration in permeate through the total area of membranes A_T consisting of a whole assemble of membrane modules, respectively, and $(M_F)_0$ is an initial feed amount in the system and the integration term in Eq. (11) expresses an accumulated permeate amount for a permeating time t .

If each membrane sheet in the membrane module is not big enough for permeation through it to significantly change feed composition or feed temperature, each feed channel confined by a pair of membrane sheets would be taken as an element volume. Thus, $w_m dz$ in Eqs. (4)-(6) can approximate to the area of a membrane sheet A_m .

Finite different schemes are employed to get a numerical solution for the model equations. This method involves dividing permeating time and membrane

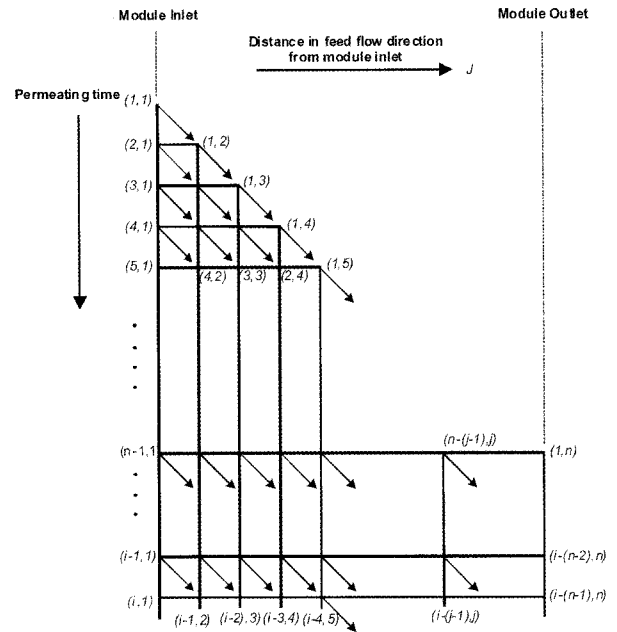


Fig. 4. Finite difference grid used in the numerical solution.

length into finite elements, respectively. In continuous process, feed mixture flows through a sequence of finite element volumes to such an extent that the feed in an element volume positioned closer to the outlet of the module has a longer residence time at a given permeating time. Two-dimensional finite difference grid for single membrane module is applied to obtain a numerical solution as shown in Fig. 4. Horizontal directional parameter j denotes the j^{th} element or j^{th} membrane sheet from the entrance of the module while vertical directional parameter i refers to feed introduced into the module inlet at the i^{th} time interval.

2.2.1. Series Connection

When the module is the k^{th} among the multi-modules in series connection, three-dimensional coordinate is used to describe the grid structure. Thus, a coordinate (i, j, k) in the grid indicates a feed that is located at the j^{th} element volume from the inlet of the k^{th} module and introduced into the I^{st} module inlet at the i^{th} time interval. The differential forms of Eqs. (4)-(6) can be transformed into the difference form of them, respectively, as follows;

$$F(i, j + 1, k) = F(i, j, k) - 2J(i, j, k) A_m \tag{12}$$

$$T(i, j + 1, k) = T(i, j, k) - 2J(i, j, k) \Delta h_v / \tag{13}$$

$$F(i, j, k) C_p A_m$$

$$x(i, j + 1, k) = x(i, j, k) + 2J(i, j, k) / \tag{14}$$

$$F(i, j, k) (x(i, j, k) - y(i, j, k))$$

The initial and boundary condition for each parameter can be given as follows, respectively,

Initial condition for each parameter;

$x(1, 1, 1) = x_0$: initial water concentration in feed

$T(1, 1, 1) = T_0$: initial feed temperature in the tank

$F(1, 1, 1) = F_0$: initial feed pumping speed

Boundary condition at the i^{th} time interval over unit module;

• For $k = 1$,

$x(i, 1, 1) = x_F(i)$: water concentration of feed to enter the 1st membrane module

$T(i, 1, 1) = T_0$: temperature of feed to enter the 1st membrane module

$F(i, 1, 1) = F_0$: flow rate of feed to enter the 1st membrane module

• For $k > 1$,

$x(i, 1, k) = x(i, n, k-1)$: water concentration of feed to enter the k^{th} membrane module

$T(i, 1, k) = T(i, n, k-1)$: temperature of feed to enter the k^{th} membrane module

$F(i, 1, k) = F(i, n, k-1)$: flow rate of feed to enter the k^{th} membrane module

Now the parameters in Eq. (11), that is, the total permeation rate through a whole assemble of modules and remnant feed amount M_F in feed tank at time t can be calculated by using the finite difference scheme;

• For $i < n$,

$$J_M(i) A_T = \sum_{j=1}^j J(i-(i-1), j, 1) A_m \tag{15}$$

$$M_F(i) = (M_F)_o - \sum_{i=1}^i \sum_{j=1}^j \tag{16}$$

$$J(i-(j-1), j, 1) A_m \Delta t$$

• For $i > n$ and $< nN$,

$$J_M(i) A_T = \sum_{k=1}^k \sum_{j=1}^j \tag{17}$$

$$J(i-(i-1), j, k) A_m$$

$$M_F(i) = (M_F)_o - \sum_{i=1}^i \sum_{k=1}^k \sum_{j=1}^n \tag{18}$$

$$J(i-(i-1), j, k) A_m \Delta t$$

• For $I \geq nN$,

$$J_M(i) A_T = \sum_{k=1}^N \sum_{j=1}^n \tag{19}$$

$$J(i-(i-1), j, k) A_m$$

$$M_F(i) = (M_F)_o - \sum_{i=1}^i \sum_{k=1}^N \sum_{j=1}^n \tag{20}$$

$$J(i-(i-1), j, k) A_m \Delta t$$

where n is the number of element volume in an unit module and N the number of unit modules in the system. Eqs. (9) and (10) gives

$$x_F(i+1) = x_F(i) + (x_F(i) - y_M(i)) J_M(i) A_T / \tag{21}$$

$$M_F(i) \Delta t$$

The initial condition for each parameter can be given;

$x_F(1) = x_0$: initial water concentration in feed

$M_F(1) = (M_F)_o$

$J_M(1) = f(x(1), T_F)$

$y_M(1) = g(x(1), T_F)$

2.2.2. Parallel Connection

Model equations for the permeation through parallel-connected modules are simpler in shape than the series-connected modules. Two dimensional coordinate (i, j) is valid for the parallel connection mode which is

equivalent to the case of $k=1$ in three dimensional coordinate (i,j,k) in the series connection mode. In the module configuration, since a certain number (N) of unit modules are connected in parallel, the whole assembly of modules can be considered as single module having a membrane area of N times larger than the unit module. As a result, the total membrane area A_m in the unit membrane module in Eqs. (12)-(14) is replaced with $N A_m$ to constitute model equations for the parallel connection.

3. Results and Discussion

3.1. Determination of Permeation Parameters

The permeation parameters which had been presented in the previous work (4) were utilized for the simulation in this work. The permeation data, such as flux and permeate concentration were directly obtained the pervaporation of ethanol/water mixture through a hydrophilic composite membrane at different feed compositions and temperatures. The composite membrane had a thin active layer of modified poly(vinyl alcohol) coated on a porous substrate of poly(acrylonitrile). Curve fitting of the obtained permeation data was performed to get the permeation parameters as functions of feed composition and temperature as follows, respectively

$$J = A_0 \exp(-E_p / T) \quad (20)$$

$$A_0 = 475 \exp(2.84x),$$

$$E_p = 3300 + 839.6x$$

$$y = [(ax)^{-2} + 98.3^{-2}]^{-0.5} \quad (21)$$

$$a = 440.9 - 112700/T$$

where x and y are water concentrations (wt%) in feed and permeate, respectively, J a flux ($\text{kg}/(\text{m}^2 \cdot \text{h})$), A_0 a pre-exponential factor ($\text{kg}/(\text{m}^2 \cdot \text{h})$) and E_p a permeation activation energy (cal/mol). The pre-exponential factor and permeation activation energy were given as func-

tion of feed composition. The determined permeation functions were in good agreement with experimental data within $\pm 4.5\%$ in the given range of operation condition. These determined permeation parameters were used in the simulation of the pervaporation dehydration process.

3.2. Simulation of Pervaporation Process

With help of the simulation model established in this study, the dehydrations of ethanol/water mixtures through the flat sheet membrane were simulated in a commercial scale of batch processes in which a number of unit modules are connected in sequence and parallel, respectively, as described in Fig. 3. In the batch process, the final product can be taken from feed retentate in the tank after circulating of the feed through respective configuration of unit modules. Therefore, each process may have its own features because of its different module configuration and feed flow regime. The simulation of these two pervaporation processes can help to understand the difference between them and provide guidelines for the design of optimum pervaporation process.

3.2.1. Series and Parallel Connections of Unit Modules

As mentioned earlier, pervaporation is characterized by the evaporation of permeate through membrane, requiring an equivalent heat for the evaporation. The heat is supplied from feed source, so that a temperature gradient in direction of the feed flow can be developed, constantly decreasing feed temperature and then decreasing flux correspondingly. In a real industrial scale where a large area of membrane is employed, feed temperature falls significantly along with membrane length due to the evaporation of permeate and thus falling of feed temperature has to be taken into consideration in process design. In order to figure out how seriously the feed temperature drop occurs with the number of unit modules in series connection at different feed flow rate, simulations were run for

Table 1. Base Parameters for Simulation of Batch Pervaporation Process

Process parameters	Unit	Value
Initial temperature	°C	90
Module inlet temperature	°C	90
Permeate pressure	torr	10
Flow velocity per each channel	Kg/h	25 ~ 1000
Initial water concentration in feed	wt%	5
Final water concentration in feed	wt%	0.5
Initial water amount	kg	1000
Dimension of unit membrane sheet	m W×L:	0.5×0.8
Membrane area of unit membrane module	m ²	28 (70 ea)
Feed channel height	mm	2.8

different sets of pervaporation. Table 1 summarizes base parameters for the simulations of pervaporative dehydration.

Assume that each batch system includes a different number of unit modules in series connection and no heat loss happens into its surroundings. The pervaporative dehydrations of ethanol/water mixture were simulated at 90°C of feed temperature and different feed flow rates. When the feed mixture in the tank was dehydrated with permeating time from the initial water content of 5 wt% to the final water content of 1 wt% in the simulation, the temperature of feed retentate was investigated with permeating time at the outlet of the module assemble composed of a certain number of unit membrane modules in the system. Fig. 5 exhibits the result of the simulation. The term "stage" in Fig. 5 means "the number of unit membrane modules" in the module assemble. At the beginning stage of permeation, the temperature of feed retentate at the outlet of the module assemble is well lower than that at the inlet of module assemble, depending on the number of unit module constituents in the system. As the permeation keeps on going, the feed temperature at the outlet increases with time and then approaches to that at the inlet of module assemble. It is because water content in feed is the highest at the beginning of permeation among a whole permeation period, so permeation through the membrane would occur to the greatest extent, require a amount of heat taken from the feed flow corresponding to the evaporation of the

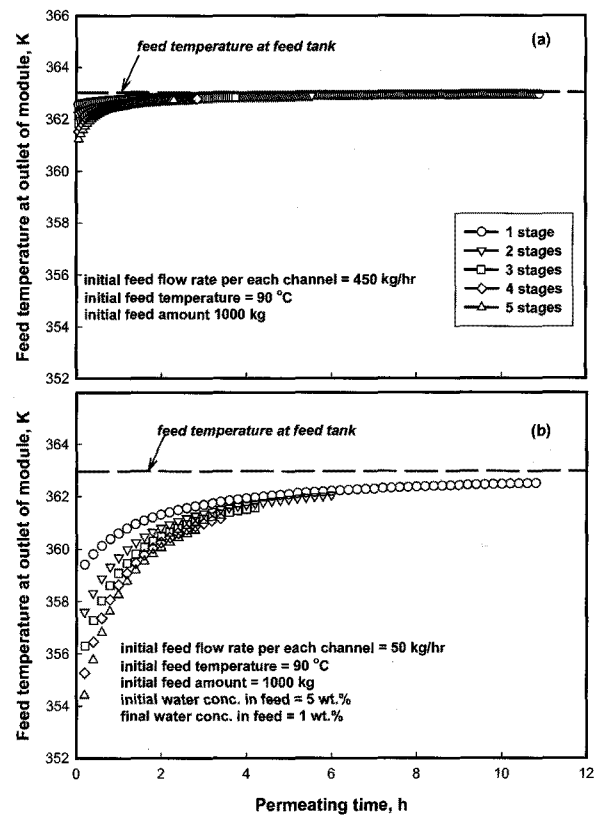


Fig. 5. Feed temperature at the outlet of module assemble having different number (stages) of unit modules in series connection with permeating time.

resulting permeate, and in turn chill down the feed, lowering its temperature. As the dehydration of feed by water-selective permeation progresses, water content in feed reduces further and then flux decreases with permeating time as much, resulting in less energy taken from the feed flow. Thus, the chill feed could be recovered toward its inlet temperature with permeating time. The more the unit modules are connected in the module assemble, the lower the feed retentate temperature at the outlet is at the beginning of permeation due to the permeation through larger membrane area for longer residence time of feed in the module assemble in its single circulation at a given flow rate, requiring more heat removal from the feed. However, as water content in the feed reduces with permeating time, flux goes down and the heat for evaporation decreases to such an extent that the temperature curves can get asymptotical into a single line approaching the inlet

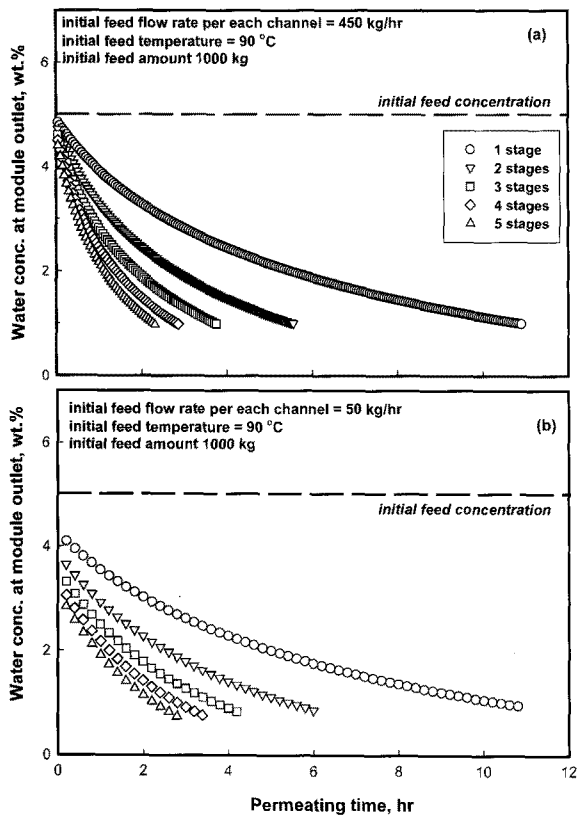


Fig. 6. Feed composition at the outlet of module assemble having different number (stages) of unit modules in series connection with permeating time.

temperature regardless of the number of unit modules. The effect of the number of unit modules on the feed retentate temperature at the outlet is observed more remarkable at lower feed flow rate as can seen in Fig. 5(b). Slower feed flow brings a longer residence time of feed in the membrane module. The residence time of feed in the module would be identical with permeating time in each circulation of feed. As a result, slower feed flow will yields a longer permeating time resulting in more permeation and then more heat for evaporation.

As pervaporative dehydration is exerted on the system, the water contents of both the feed retentate at the outlet of the module assemble and the feed in the tank decrease with permeating time as shown in Fig. 6 and Fig. 7. As discussed above, the module assemble which includes more unit modules connected in series has a larger membrane area, more permeation and lon-

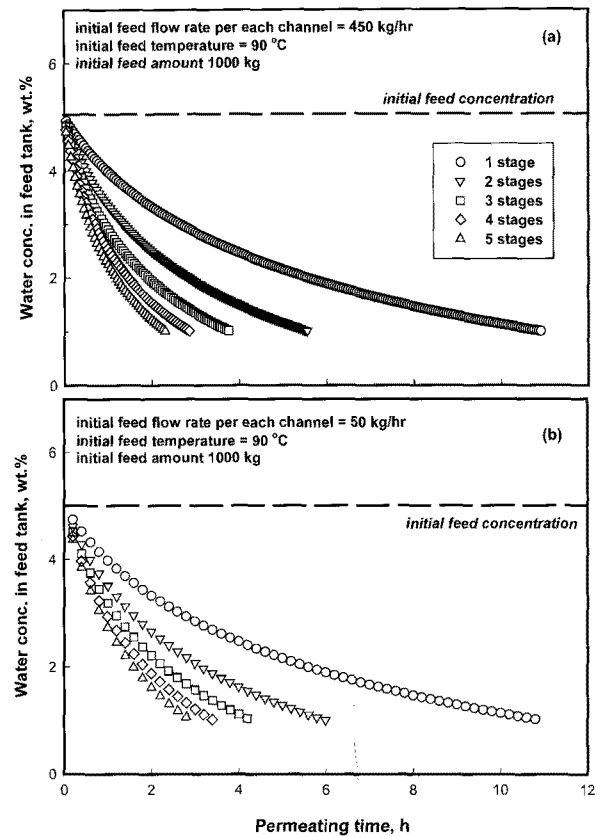


Fig. 7. Feed composition in feed tank with permeating time in pervaporative dehydration of ethanol through a module assemble having different number (stages) of unit modules in series connection.

ger residence time of feed during single circulation. So it is quite normal for the system with larger membrane area to require shorter permeating to dehydrate the ethanol/water mixture into the target composition. At an incipient permeation, the water content of feed retentate at the outlet of module assemble is lower than that at the inlet of module assemble, depending on the number of unit modules in the system. The water content at the outlet is even lower when feed flows slower or/and the number of unit modules is larger in the membrane assemble. It is because water-selective permeation through more membrane area with longer permeating time dehydrates the retentate feed more at the beginning state of permeation, as already explained in the previous section. However, permeation decreases the temperature of retentate feed as much as the heat used for the evaporation of permeate which is taken

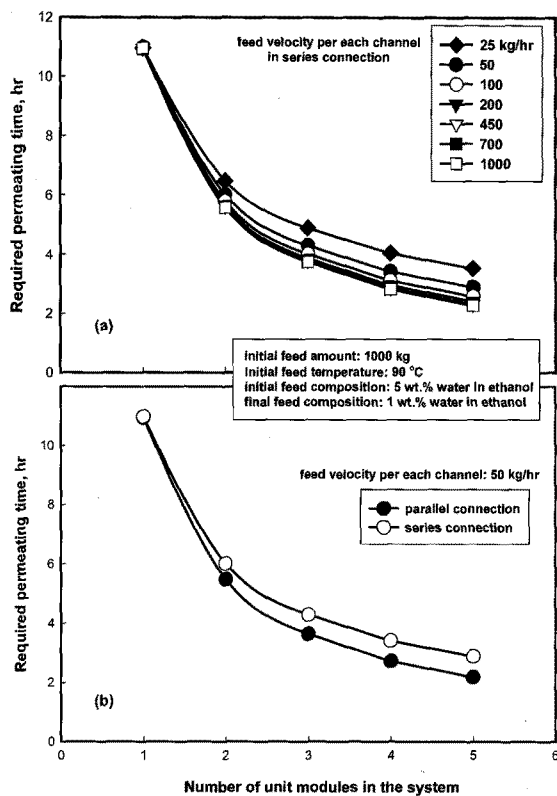


Fig. 8. Required permeating time to get the target composition of feed with number of unit modules (number of stages) composing of the membrane assemble at different feed flow rates.

from the retentate feed, resulting in lowering average retentate temperature in module assemble. Therefore, when the number of unit module is larger or/and feed flow is slower in module assemble, it takes longer to get a target composition of retentate because of lower temperature of retentate feed in the module assemble.

A module assemble composed of certain number of constituent modules connected each other in parallel has the same length of path way as single unit module. Thus, the residence time of feed in the module assemble with parallel connection is identical with that in single unit module of the module assemble with the series connection in each circulation. It was found from the simulation that pervaporation performance through the modules connected in parallel is almost independent of feed flow rate for a given range of feed flows in this study, and is very similar to one in series with fast feed flow regime because of smaller

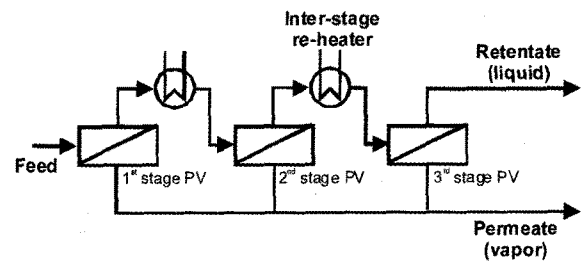


Fig. 9. Schematic representation of continuous pervaporation process with inter-stage re-heater.

residence time of feed in the module assemble with parallel connection in each circulation. Thus simulation curves of parallel connection could not be plotted in Fig. 8 because of overlapping them with ones of series connection with high feed flow rate. Fig. 8(a) shows the plots of required permeating time with the number of unit module employed in membrane assemble with series connection at various feed flow rates. There is no big distinction between curves with flow rate above 200 kg/h. It is because the residence time of feed is too short to yield significant permeation in each circulation. When feed flow rate is smaller, the feed dwells longer in the module during single circulation, more permeation takes place and then more heat is taken from feed correspondingly. Therefore the resulting average temperature of feed in the module lowers, in turn, depressing the permeation through membrane and requiring longer permeating time to reach the target composition in feed. Fig. 8(b) presents a comparison of required permeating times between parallel and series connection modes at 50 kg/h of feed flow. Parallel connection which has shorter path way in module assemble or smaller residence time of feed favors the permeation more, resulting in shorter permeating time required for the dehydration.

3.2.2. Supplementary Series Connection Mode

The term "feed flow rate" used so far refers to "feed velocity through respective feed channel" in module. At a given feed flow rate, parallel connection of modules shows some advantage over series connection in terms of membrane performance because of short residence time of feed. However, feed mixture should be

Table 2. Base Parameters for Simulation of Continuous Pervaporation Process

Process parameters	Unit	Value
Initial temperature	°C	80, 90
Module inlet temperature	°C	80, 90
Maximum temperature drop	°C	10
Permeate pressure	torr	10
Flow velocity per each channel	kg/h	100
Initial water concentration in feed	wt%	5
Final water concentration in feed	wt%	0.5
Dimension of unit membrane sheet	m	W×L: 0.5×0.8
Feed channel height	mm	2.8

Table 3. Total Membrane Area and Membrane Area in Each Stage Module Calculated in Dehydration of Ethanol at Different Initial Temperature

Stage number	Required membrane area, m ²	
	Initial feed temp = 80°C	90°C
1	7.2	4.4
2	15.2	10.4
3	18.8	12.4
Total membrane area	41.2	27.2

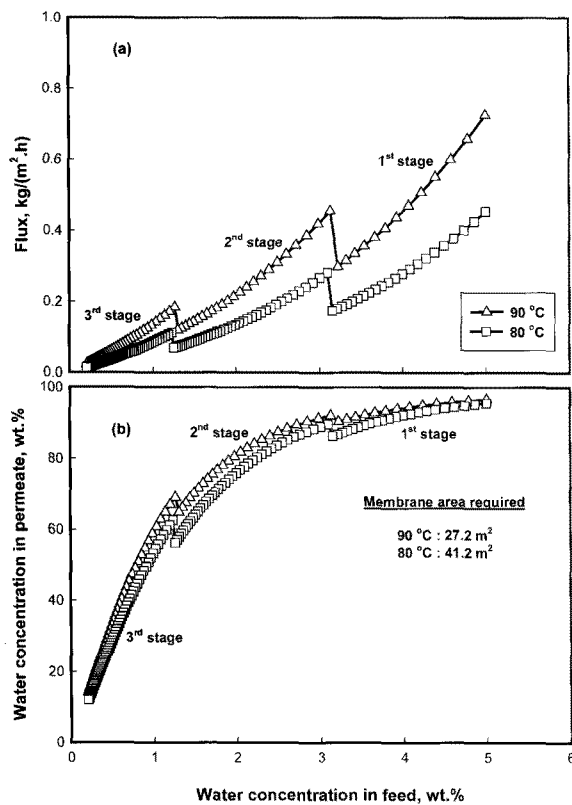


Fig. 10. Simulated flux and permeate concentration with feed composition in the continuous pervaporation of ethanol/water mixture at different feed temperature: initial feed flow rate = 100 kg/h.

simultaneously supplied into all of channels in the module assemble with parallel connection. Thus parallel connection needs a bigger capacity of circulation pump and consumption of more electricity than series connection, requiring larger installation and operation costs. Therefore, from the economical point of view, it

might be worthy of taking into consideration of a hybrid system of parallel and series connections, compromising the system in terms of membrane performance, energy consumption and installation cost. Alternatively, supplementary series connection mode could be employed; the feed is re-heated when the feed temperature fell below an acceptable module entrance temperature and thus the temperature falling could be compensated by installing re-heaters between different unit modules to reheat the feed to a specified module entrance temperature (Fig. 9). For a given set of dehydration, the number of re-heater or the number of stage is determined by two parameters, 1) the initial feed temperature (or module entrance temperature) and 2) the allowable maximum temperature drop along the module unit. A typical maximum temperature drop is 10°C. Thus, in this study, the maximum temperature drop was fixed to be 10°C. In this case, each stage of modules would have different number of membrane sheets. The simulations were run for different sets of pervaporation described in Table 2. Fig. 10 shows the influence of different initial feed temperature on the membrane area and stage number required. Looking at Fig. 10(a), each curve segment between points depicts a change in respective permeation performance with feed flowing over respective membrane sheet. The flux change is observed to be more significant at higher feed temperature or/and higher water content in feed, resulting in more temperature drop over a membrane sheet and less number of membrane sheets involved in an unit module. Since the dependency of flux on temperature is an Arrhenius-type equation as described in Eq. (20), the influence of temperature on membrane

performance is found to be significant to achieve a predefined separation. As feed temperature increases, the membrane area required decreases and number of membrane sheets in each stage also decreases as much (Table 3). It is observed from simulation that the depressed permeation due to the temperature falling can be recovered to some extent by reheating the feed of which temperature fell as a result of permeation. The permeate concentration increases slightly with feed temperature but decreases more with decreasing water concentration in feed (Fig. 10(b)). It reflects that more separation permeation could be achieved at higher temperature. Hence, one of the basic design rules to apply to pervaporation is maintain the feed temperature as high as possible.

4. Conclusions

The simulation model for pervaporative dehydration through membrane module assemble in which a number of unit modules are connected in parallel or in series has been established. With help of the model, pervaporation processes through the membrane module assembles were simulated, respectively, for analyzing and optimizing the pervaporation process.

In series connection, when feed flows slower or/and the number of unit modules is larger in the membrane assemble, the water content at the outlet of module assemble is lower because water-selective permeation through more membrane area with longer permeating time dehydrates the retentate feed more at the beginning state of permeation. In the other hand, permeation decreases the temperature of retentate feed as much as the heat for the evaporation of permeate which is taken from the retentate feed, resulting in lowering average retentate temperature in module assemble. Thus, it takes longer to get a target composition of retentate because of lower temperature of retentate feed in the module assemble when the number of unit module is larger or/and feed flow is slower in module assemble.

Pervaporation performance through the modules connected in parallel is almost independent of feed flow

rate for a given range of feed flows in this study, and is very similar to one in series with fast feed flow regime because of smaller residence time of feed in the module assemble with parallel connection in each circulation. There is no distinction between curves with flow rate above 200 kg/h because the residence time of feed is too short to yield significant permeation in each circulation. When feed flow rate is smaller, the feed dwells longer in the module during single circulation, more permeation takes place and then more heat is taken from feed correspondingly. Therefore the resulting average temperature of feed in the module lowers, in turn, depressing the permeation through membrane and requiring longer permeating time to reach the target composition in feed. Parallel connection which has shorter path way in module assemble or smaller residence time of feed favors the permeation more, resulting in shorter permeating time required for the dehydration.

In supplementary series connection mode, re-heaters between different unit modules are installed to compensate the decline of feed temperature through reheating the feed to a specified module entrance temperature. It is observed from simulation that the depressed permeation due to the temperature falling is recovered to some extent by reheating the feed of which temperature fell as a result of permeation.

Acknowledgements

Financial support on this work from Korean Energy Management Corporation is gratefully acknowledged.

Notation

- A_m : effective membrane area per unit membrane sheet in a module (m^2)
- C_p : heat capacity of feed (kcal/(kg·°C))
- dz : differential length of channel in z direction (m)
- E_p : permeation activation energy (kcal/mol)
- F : Feed flow rate per individual feed channel (kg/h)

- h_F : enthalpy of feed (kcal/mole)
- Δh_v : heat of the evaporation of permeate (kcal/mole)
- J : flux (kg/(m².h))
- l_m : effective length of unit membrane sheet (m)
- M_F : feed mass in feed tank at time t (kg)
- $(M_F)_0$: initial feed mass in feed tank (kg)
- n : number of membrane sheets in an unit module
- N : number of unit modules in system
- t : permeating time (h)
- T : feed temperature (K)
- x : concentration of a selectively permeating component in feed (fraction)
- y : concentration of a selectively permeating component in permeate (fraction)

References

1. G. W. Meindersma and M. Kuczynski, "Implementing membrane technology in the process industry", *J. Membr. Sci.*, **113**, 285 (1996).
2. F. W. Greenlaw, W. D. Price, R. A. Shelden, and E. V. Thompson, "Dependence of diffusive permeation rates on upstream and downstream pressure: II. Two component permeant", *J. Membr. Sci.*, **2**, 141 (1977).
3. R. Rautenbach, C. Herion, M. Franke, A. A. Asfour, A. Bemquerer-Costa, and E. Bo, "Investigation of mass transport in asymmetric pervaporation membranes", *J. Membr. Sci.*, **36**, 445 (1988).
4. J. M. Won, C. K. Yeom, S. Yoon, J. W. Rhim, S. R. Bae, and B. H. Ha "Modeling of pervaporation process: prediction of feed temperature distribution in a frame and plate type of membrane module," *Membrane Journal*, **6(1)**, 44, (1996).
5. C. K. Yeom, M. Kazi, and F. U. Baig, "Simulation and process design of pervaporation plate-and-frame modules for dehydration of organic solvents", *Membrane Journal*, **12(4)**, 226 (2002).
6. C. K. Yeom and F. U. Baig, "Simulation and pervaporation process through hollow fiber module for treatment of reactive waste stream from a phenolic resin manufacturing process", *Membrane Journal*, **13(4)**, 257 (2003).
7. C. K. Yeom and F. U. Baig, "A characterization of pervaporation-facilitated esterification reaction with non-perfect separation", *Membrane Journal*, **13(4)**, 268 (2003).
8. S. H. Choi, Y. I. Park, S. S. Chang, and C. K. Yeom, "A parametric study of pervaporation-facilitated esterification", *Membrane Journal*, **17(2)**, 146 (2007).