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Evaluation of the Performance of Multistage Membrane Separation Cascades

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ABSTRACT

The minimization of the work consumption in a multistage membrane separation cascade operating in isoenthalpic conditions was investigated with regard to the splitting factor of each stage, for the assigned values of the number of stages, the retentate and permeate pressures, and the flow pattern. The mathematical model of the membrane separation cascade is presented, and five possible different flow patterns together with system variance are discussed. Under isoenthalpic conditions the work needs are due to the compression of the permeated stream, and the work losses to the generation of entropy on the mixing of recycled streams. A three-component mixture (NH_3, H_2, N_2) and a polyethylene membrane were used as the standard testing system; the results of the optimization for three, five, seven, or nine stages cascades, using as the initial splitting factor values 0.5, 1.0, and 1.5 for all stages, are discussed. Increasing the number of stages increases the membrane surface area and the total work consumption owing to the increase in recycled stream flow. In the case under examination, both the efficiency index and the specific efficiency decrease for cascades of more than five stages. Therefore membrane staging seems to be unattractive from the point of view of both capital investment and operating costs, at least for more than a certain number of stages, depending on the specific problem.

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INTRODUCTION

Membrane separation processes seem extremely attractive since, for an ideal system, the separation work is equal to the compression work, from the partial pressure of the gases in the mixture to the total pressure of the mixture. Indeed, for real systems the permeating molecules must overcome the frictional resistance within the membrane, and that depends on the operating transport rate, with the result that the pressure of the permeated stream is lower than the partial pressure of the permeated gas in the mixture. Moreover, a real membrane has a certain selectivity of separation. Thus, with a single stage separation it is not possible to isolate a component with a high degree of purity from a mixture. A multistage cascade is needed, but this requires the recycling of streams, thereby increasing the complexity of the system and the work load.

Some companies have claimed (1) the production of high purity gases, mainly nitrogen, with membrane processes, and industry observers in 1990 said that by 1995 up to 30% of the cryogenic nitrogen share of the market could be supplied by membrane separators and pressure swing adsorbers. Actually, high purity nitrogen is produced in industry by the cryogenic process; only technical nitrogen (purity 95–97%) is produced with the membrane process. Nevertheless, there is always somebody who claims the economic feasibility of industrial production of high purity gases with membrane processes.

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In this paper we investigate some aspects of membrane processes. The design of a single-stage permeating apparatus with different flow patterns has been described by many authors (2–6); for a multistage apparatus a lot of work has been done on binary mixtures using shortcut methods (7–10) derived from the McCabe and Thiele procedure, or by the rigorous method using different approaches (11, 12). Only in a few cases has any consideration been given to the optimization of design with respect to operating parameters. Indeed the economics of the membrane separation process is of basic importance in competing with other processes such as distillation, PSA, etc.

For many applications, when only concentration is needed, a single-stage membrane is adequate, but for stream purification more stages are required, the size of the process grows, and the process design becomes complex.

The optimization of a single-stage membrane process is relatively simple and some guidelines can be summarized as follows.

1. Increasing the pressure differential results in a residue purity increase, a permeate purity decrease, and a membrane area requirement decrease.

2. Increasing the membrane area results in a purer residue; decreasing the membrane area results in a purer permeate. In any case, the yield drops as the purity requirements is increased.
3. Both pressure differential and pressure ratio define membrane performance.
4. Since in gas separations there is practically no boundary layer on the membrane, the feed flow rate has little impact on the permeate total flow rate.
5. The countercurrent flow pattern has the best performance; nevertheless, the effect of flow pattern tends to diminish with decreasing permeate/feed pressure ratio.
6. Since the partial pressure of a component in the permeate stream cannot be higher than its partial pressure in the feed ($\pi_{h,m}x'_{i,m} > \pi_{l,m}$ $y'_{i,m}$), the value of $y'_{i,m}$ is limited by the ratio $\pi_{h,m}/\pi_{l,m}$. This means that for a selective membrane the enrichment ratio ($y'_{i,m}/x'_{i,m}$) is limited by the lower value between P_i/P_j and $\pi_{h,m}/\pi_{l,m}$ (5).

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For a membrane process with M stages, operating in isothermal conditions and with known feed streams F_m of NC components, the variance of the system is $3M$. This means that the process is completely described when, for each stage, the values of three variables are assigned, such as the membrane area A_m, the splitting factors θ_m, and the upper and/or the lower pressures, $\pi_{h,m}$ and $\pi_{l,m}$.

In practice, the obtained values of the operating conditions, and/or the outlet stream compositions, may not satisfy the optimum economic conditions or the outlet design specifications. To find the operating conditions an appropriate economic function of the independent variables of the system has to be optimized. The economic function must be a well-balanced estimation of the capital and the operating costs; for instance, the number of stages, the membrane area, and the compression costs of permeated streams (13, 14). Moreover, a relationship between the economic objective function and the independent variables of the system must be found.

THEORY

The configuration of the membrane separation process as a multistage cascade (Fig. 1) has been considered (15). A multicomponent stream F_m with NC components may be fed to each stage or, as an alternative, a sidestream, $-F_m$, may be removed.

In node $[m]$ the feed F_m is mixed with the permeate stream P_{m+1} from stage $[m + 1]$ and the retentate stream R_{m-1} from stage $[m - 1]$. The effective feed of each stage Q_m consists of the retentate flow from stage $[m - 1]$, the permeate flow from stage $[m + 1]$, and the entering feed

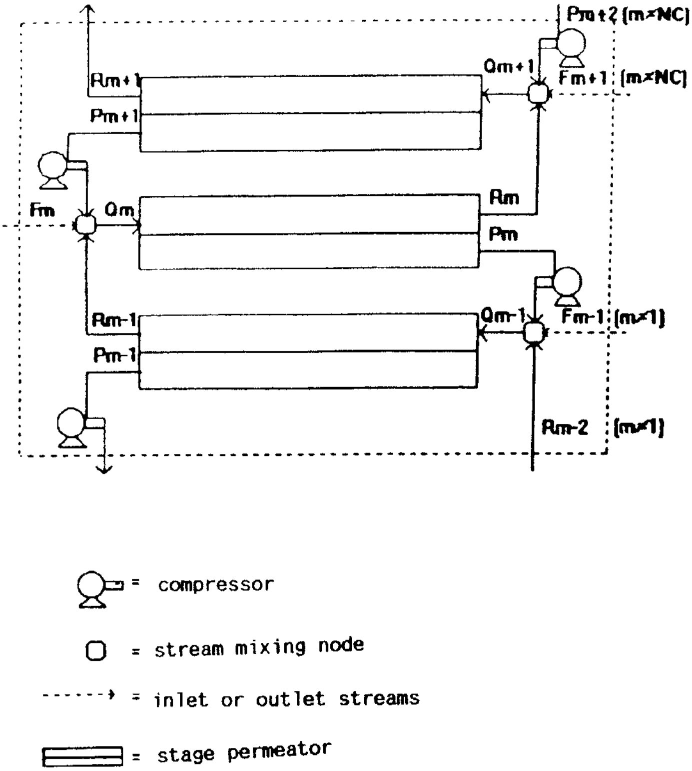

FIG. 1 Configuration of the permeator cascade for the membrane separation process.

stream. No recycling was considered for either end stage: the first stage is fed only with the permeate stream from stage [2] and the last stage with the retentate stream from stage $[M - 1]$.

For an efficient separation process the streams that will mix in each node must have, as far as is possible, similar compositions. The possibility of satisfying this constraint will be discussed with regard to the variance analysis of the system.

With reference to Fig. 1, the following mass balances can be written for each node:

$$Q_m = F_m + P_{m+1} + R_{m-1} \tag{1}$$

and for each stage:

$$Q_m = R_m + P_m \tag{2}$$

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or:

$$F_m + P_{m+1} + \theta_{m-1}P_{m-1} = (\theta_m + 1)P_m \quad (3)$$

where θ_m is the stage splitting factor defined as

$$\theta_m = R_m/P_m \quad (4)$$

The total balance on the M stages is

$$\sum_{m=1}^{M} F_m = R_M + P_1 \quad (5)$$

For assigned values of the F_m feeds and the splitting values θ_m, Eq. (3) forms a tridiagonal system of M linear equations with M unknowns, P_m, that can be solved with a standard procedure.

A node and a stage material balance can be written for each component:

$$Q_m w_{m,j} = F_m z_{m,j} + P_{m+1}y_{m+1,j} + R_{m-1}x_{m-1,j} \quad (6)$$

$$Q_m w_{m,j} = R_m x_{m,j} + P_m y_{m,j} \quad (7)$$

where $z_{m,j}$, is the known molar fraction of the F_m feed stream and $w_{m,j}$, $y_{m+1,j}$, and $x_{m-1,j}$ are the unknown molar fractions of the stream entering stage m, the permeate from stage $m + 1$, and the retentate from stage $m - 1$, respectively.

To find the values of all the unknowns, i.e., to define the problem, the mass balance equations of the permeating flow across the membrane are needed.

On the basis of the working hypothesis: 1) permeation Fick's law; 2) permeability coefficient independent of pressure and composition; 3) null pressure drops in the direction of the flow stream; 4) plug flow, except for mixed flow and crossflow pattern (in any case the concentration gradient is null along the permeation flow); and 5) constant thickness of the membrane; the differential equation of the transport phenomena across the membrane may be written as

$$-dR'_m = dP'_m \quad (8)$$

$$-dR'_m = dA_m \sum_{j=1}^{NC} P_j(\pi_{h,m}x'_{m,j} - \pi_{l,m}y'_{m,j})/\delta \quad (8')$$

$$-d(x'_{m,j}R'_m) = d(y'_{m,j}P'_m) \quad (9)$$

$$-d(x'_{m,j}R'_m) = dA_m P_j(\pi_{h,m}x'_{m,j} - \pi_{l,m}y'_{m,j})/\delta \quad (9')$$

A prime indicates local variables.

The following equations hold true for the molar fractions:

$$\sum_{j=1}^{NC} x'_{m,j} = 1; \qquad \sum_{j=1}^{NC} y'_{m,j} = 1 \tag{10}$$

By developing Eq. (9′) and considering Eq. (8′), it is possible to write

$$dx'_{m,j} = dA_m[P_j(\pi_{h,m}x'_{m,j} - \pi_{l,m}y'_{m,j}) - x'_{m,j}\sum_{j=1}^{NC} P_j(\pi_{h,m}x'_{m,j} - \pi_{l,m}y'_{m,j})]/(\delta R'_m) \tag{11}$$

The boundary conditions for the integration of the previous equations depend on the flow pattern of the permeate and retentate streams. In general, the following flows can be considered:

1. Cocurrent flow
2. Countercurrent flow
3. Crossflow
4. One side mixing
5. Perfect mixing

Some authors have considered three or more components when studying the various flow patterns in a permeating stage (16–19).

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OPTIMIZATION OF CASCADE PERFORMANCE

With reference to the general scheme of Fig. 1 for a system with M stages, there are $(M - 2)$ possible external feed streams and $(3M - 4)$ interstreams (the two exit streams are also included). Therefore the total number of independent variables for a system with M stages and NC components is

$$4NC(M - 1) + 3M \tag{12}$$

where the last term considers the upper $\pi_{h,m}$ and lower $\pi_{l,m}$ pressures and the membrane area A_m of each stage.

The total number of relations among the variables is

$$\text{Stages equations} = 2MNC$$

$$\text{Node equations} = (M - 2)NC$$

The variance V of the system is

$$V = NC(M - 2) + 3M \tag{13}$$

Since all feed F_m are assigned, it has to be assumed that $(M - 2)NC$ feed

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composition streams are known and therefore the variance is

$$V = 3M \tag{14}$$

For instance, if the splitting factors (θ_m) and the upper and lower pressures ($\pi_{h,m}$, $\pi_{l,m}$) are assigned for all stages, the problem is defined, and it is possible to calculate the amount and composition of each stream and the membrane area needed for each stage.

Obviously, for any given arbitrary value of independent variables the calculated cascade is probably not the best in terms of either capital cost (number of stages and total membrane area) or operating costs (work of compression of recycled streams).

Since from the application point of view the problem is not only to calculate the cascade but also to find the best design for the goals in mind, an optimization procedure leading to the best values for the independent variables must be set up. Such optimization calls for criteria that reduce the generation of entropy due to the mixing of the streams in each node. Since from the application point of view it is not practical for each stage to have different $\pi_{h,m}$ and $\pi_{l,m}$ values, the only operating variables that can be used as independent variables for optimization are the splitting factors θ_m.

The total entropy of mixing S_{mix} is

$$\frac{S_{mix}}{R} = \sum_{m=2}^{M-1} \left[R_m \sum_{j=1}^{M} (x_{m,j} \ln x_{m,j} - w_{m,j} \ln w_{m,j}) + P_m \sum_{j=1}^{M} (y_{m,j} \ln y_{m,j} - w_{m,j} \ln w_{m,j}) + F_m \sum_{j=1}^{M} (z_{m,j} \ln z_{m,j} - w_{m,j} \ln z_{m,j}) \right] \tag{15}$$

The minimization of this function in terms of the splitting factors θ_m is not easy since it always gives the banal solution of zero value for all the recycled streams: the retentate streams under the feed stage and the permeated streams over the feed stage. If one tries to eliminate this solution by introducing some bootstrap, the solution found is not satisfactory.

An equivalent approach is to constrain the compositions of the streams at the node to be equal:

$$z_{m,j} = x_{m,j} = y_{m,j} \tag{16}$$

This implies the introduction of $2(NC - 1)(M - 2)$ constraints. Since for assigned feed compositions and stage pressures $\pi_{h,m}$ and $\pi_{l,m}$ the variance is M on introducing the previous constraints, the residual variance becomes

$$V = M - 3(NC - 1)(M - 2) \tag{17}$$

where $M > 2$ and $NC > 1$. It can be easily verified that for $M = 3$ and $NC = 2$, the variance is zero.

The impossibility of satisfying the constraints (16) indicates that a multistage membrane separation cascade has an intrinsically irreversible configuration owing to the entropy generation in the mixing nodes, and it is not possible to develop a theoretically reversible cascade. Therefore, a suitable objective function is needed for the optimization and a nonlinear regression procedure can be used to determine the best values of the unknown variables.

Many different objective functions have been tested with the aim of reducing the operating costs, including the simple relationship

$$\mathrm{FUN0} = \sum_{m}^{M} \sum_{j}^{NC} [\mathrm{abs}(1 - x_{\mathrm{m-1,j}}/w_{\mathrm{m,j}}) + \mathrm{abs}(1 - y_{\mathrm{m+1,j}}/w_{\mathrm{m,j}})] \tag{18}$$

which has the disadvantage of not taking into account the separation yield of the components. Moreover, the exit streams, the interstreams, and their composition depend strongly on the initially assumed values of the splitting factors θ_{m}.

A term that takes into account the yield of separation of two components, arbitrarily chosen as the key components i and k, has been introduced into the final objective function:

$$\mathrm{FUN} = M\left[\mathrm{abs}\left(1 - P_1 y_{1,\mathrm{i}} \Big/ \left(\sum_{m=1}^{M} z_{\mathrm{m,i}} F_{\mathrm{m}}\right)\right) + \mathrm{abs}\left(1 - R_{\mathrm{M}} y_{\mathrm{M,k}} \Big/ \left(\sum_{m=1}^{M} z_{\mathrm{m,i}} F_{\mathrm{m}}\right)\right)\right] \tag{19}$$

$$+ \sum_{m}^{M} \sum_{j}^{NC} [\mathrm{abs}(1 - x_{\mathrm{m-1,j}}/w_{\mathrm{m,j}}) + \mathrm{abs}(1 - y_{\mathrm{m+1,j}}/w_{\mathrm{m,j}})]$$

The first two terms of the second member should maximize the yield of the two key components i and k, while the last term should equalize the streams recycled at the nodes.

SOLUTION PROCEDURE

The solution of a rather complex optimization problem, like that of the multicomponent multistage permeation cascade, is not an easy task since in many cases the objective function may have many local minima or a

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rather large flat region of minima. This occurs when some correlation exists among the variables. In all these cases the problem is not well defined by a single solution: many solutions with similar objective function values can be found, depending on the assumed initial value of the independent variables.

In the case of optimization problems, the usual procedure, to verify the stability of the solution or the presence of a multiplicity of solutions, is to perform an optimization procedure using different initial values for the independent variables.

In fact, as was shown in Eq. (14), the permeation cascade can be optimized through $3M$ independent variables. Nevertheless, in order to avoid an exotic approach, we have assumed that all stages operate with assigned values of $\pi_{h,m}$, $\pi_{l,m}$.

Since it is not possible to exclude some correlation among the independent θ_m and the calculated A_m variables, the optimization was performed for the following initial values of the stage splitting factor: $\theta_m = 0.5, 1.0, 1.5$. Except for the perfect mixing flow pattern case, where the material balance is defined by a set of algebraic equations solved by an iterative procedure, the stage material balances are defined by a set of differential equations. The solution of these equations with an iterative procedure requires a good estimation of the initial values of the streams for each stage. The optimized solution of the perfectly mixed stage was used as the initial estimation for all other flow patterns.

The first step in the solution procedure is the estimation of the cascade streams through the tridiagonal system of Eq. (3) for given values of the operating variables $\pi_{h,m}$, $\pi_{l,m}$, θ_m, and known values of the feed streams and membrane transport properties. The following step is the evaluation of θ_m values that optimize the objective function. To reduce the computing time, a preliminary optimization is made assuming perfect mixing for each stage. The results are then used as the initial values in the second part that considers the effective flow pattern: all data reported were calculated according to the crossflow pattern.

The differential equations were solved by Runge–Kutta–Merson with an optimized integration step with a minimum value of 10^{-6}.

The computing program uses the computer library to solve nonlinear equations, to optimize functions, and to evaluate the numerical integration of differential equations of the Jacobian and the inverse matrix.

RESULTS AND DISCUSSION

A typical literature problem was analyzed: ammonia–hydrogen separation in the presence of nitrogen as proposed by Shindo (15). Ammonia,

TABLE 1
Parameters Used for the Calculations of the Columns (15)

Membrane: Polyethylene at 373 K; thickness = 7.5 μm
Feed: 1 mole of mixture

Feed composition (mole fraction)		Permeability data [mole/(s·m·Pa)]	
Ammonia	0.45	36.9×10^{-15}	key-one component
Hydrogen	0.25	11.7×10^{-15}	key-two component
Nitrogen	0.30	2.41×10^{-15}	

Pressure (Pa): $\pi_h = 5.006 \times 10^5$; $\pi_l = 0.659 \times 10^5$

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the most permeable component, was assumed as the key-one component and hydrogen, which has an intermediate permeability, as the key-two component. All design variables are reported in Table 1.

Figures 2 shows the molar fraction of the key-two component, hydrogen, in the permeate exit stream, and Fig. 3 shows the composition of the retentate as a function of the number of stages for the three initial values

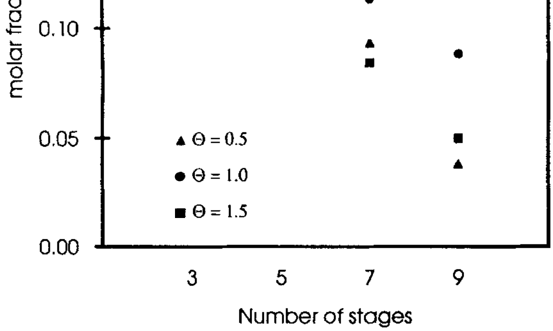

FIG. 2 Molar fraction of key-two component (hydrogen) in the permeate exit stream.

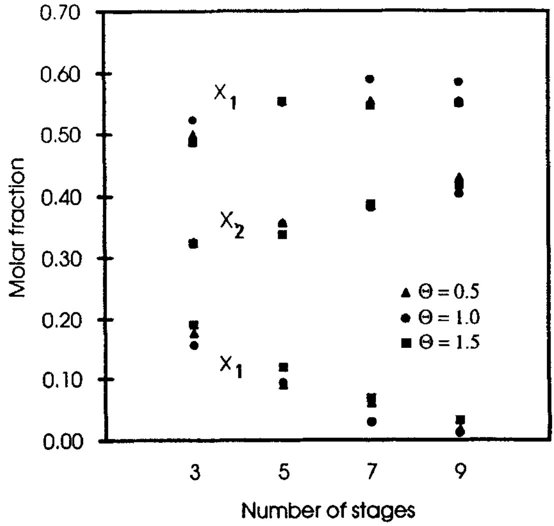

FIG. 3 Molar fraction of the components in the retentate exit stream.

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of the splitting factors θ_m. The lowest permeable component, nitrogen, is practically absent in the permeate stream: its molar fraction is lower than 0.01. As expected, by increasing the number of stages, the separation between the two key components increases. In any case, the initial values of θ_m have some influence on the permeate composition and only a minor influence on retentate exit stream.

The permeate and retentate exit flow rates are shown in Fig. 4 as a function of the cascade stage number. Even if the results are rather scattered for the various initial values of θ_m, the general trend is a decrease of retentate and an increase of permeate flow rate with cascade stages.

Figures 5 and 6 show, respectively, the yield $(P_i y_{i,m}/\Sigma F_m z_{m,1}) \times 100$ of the key-one component in the permeate and key-two component in the retentate as a function of the number of stages for the initial values of θ_m. In both cases the yield increases by increasing the number of stages but with a different trend: the component one points markedly at high values of the yield while the yield of the component two increases smoothly.

Figure 7 shows the total permeating areas of each membrane cascade for different initial values of θ_m. The general trend is an increase of membrane area with the number of stages. An anomalously high value of the total area has been obtained for the case of a nine-stage cascade and θ_m

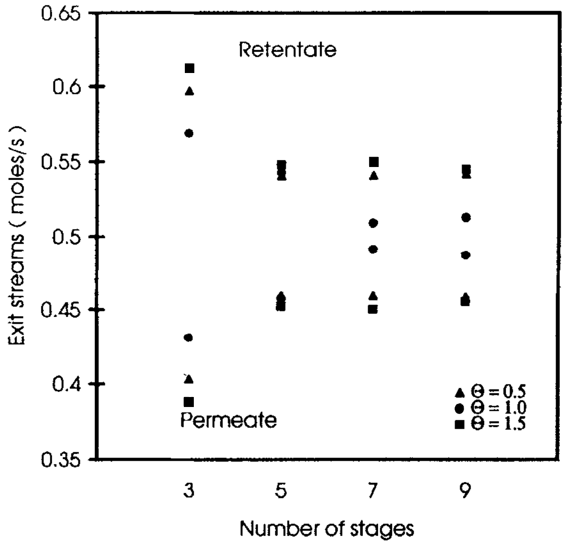

FIG. 4 Permeate and retentate exit flow rates.

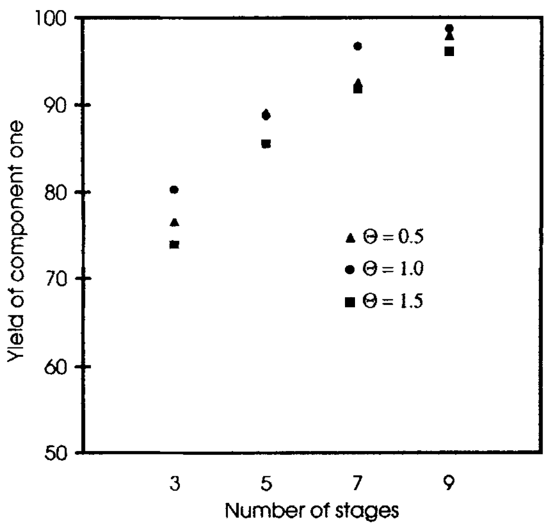

FIG. 5 Yield of component one (ammonia).

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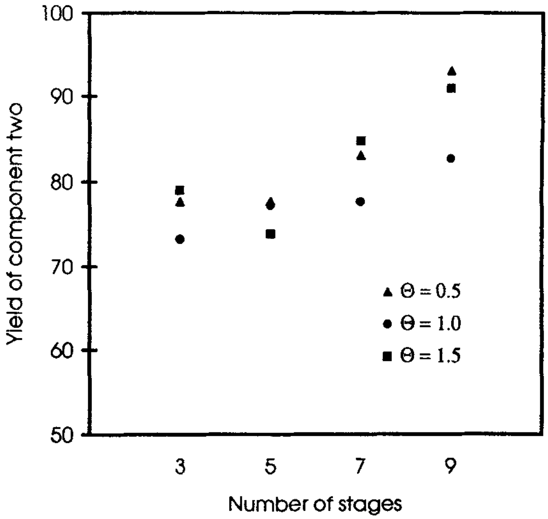

FIG. 6 Yield of component two (hydrogen).

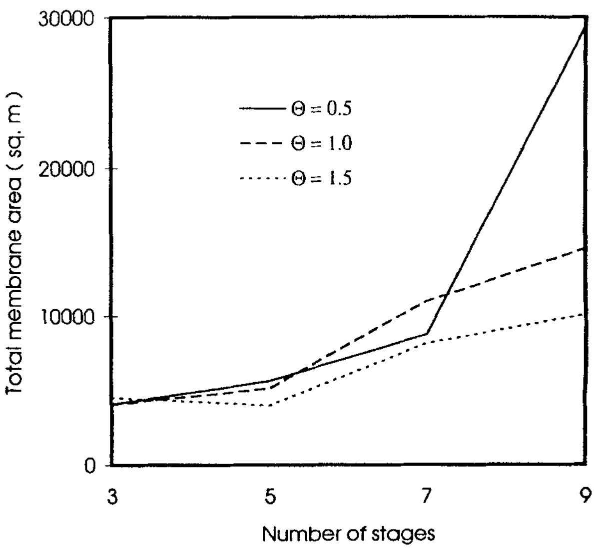

FIG. 7 Total membrane area.

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= 0.5 as the initial value. By comparing Figs. 2 and 7, a correlation between the cascade membrane area and the molar fraction of key-two component in the permeate is suggested: in all cases a high value of the membrane area corresponds to a low molar fraction of the key-two component in the permeate.

The data on the stage area, not reported for the sake of brevity, point out that the stage below the feed-stage has the greater membrane area and all stages over the feed-stage have low values of the membrane area. As a general conclusion, it is suggested that the number of effective working stages is no more than five.

At the end of the calculations one has to determine which figures have merit for the cascades in order to allow a choice among equipment that differs in component yield, operating conditions, membrane surface area, and number of stages. The first step is to evaluate the efficiency of each cascade. For this, we introduce an efficiency index, defined as

$$\text{e.i.} = \frac{\text{thermodynamic work of isothermal separation}}{\text{isothermal work of all permeate stream compression}} \times 100 \tag{20}$$

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The thermodynamic work of isothermal separation W_S is defined according to the Gibbs function. By assuming an isoenthalpic separation process, i.e., exit stream enthalpy equal to feed enthalpy (under isothermal conditions this assumption implies ignoring the excess mixing enthalpy, a quite reasonable assumption for gases not at elevated pressure), the work is determined only by the entropy variation of the process:

$$W_S = \Delta G = -T\Delta S \tag{21}$$

where ΔS may be evaluated from the stream entropy variations at the exit and the entrance. It is possible to show that this work equals the ideal work of compression of all components from their partial pressure in the exit stream to their partial pressure in the feed stream.

The isothermal work of compression W_c of all permeate streams is the ideal work of compression from the low pressure value $\pi_{l,m}$ to the feed pressure $\pi_{h,m}$ under isothermal conditions. This work is the sum of the following needs:

1. Thermodynamic energy needs for the separation.
2. Kinetic work to overcome the membrane resistance or, from another point of view, the work of the forces driving the process.
3. Configurational losses of work due to the presence of mixing irreversibilities in the nodes.

Obviously, no retentate pressure drop occurs in this ideal model since there is no friction.

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Figure 8 shows the efficiency index of the various cascades as a function of the number of stages. The first and most important consideration is the very low value of the efficiency index for all the cascades: the values range from 1 to 5, i.e., under ideal conditions the work needs range from 20 to 100 times the thermodynamic separation work.

As a second consideration, the efficiency index for a cascade with a given number of stages changes randomly with the assumed initial values of the splitting factors θ_m. Nevertheless, it appears that by increasing the number of stages the efficiency index decreases. For the problem under consideration, this decrease begins after five stages.

The efficiency index may be correlated to the operating costs of the separation plant. To combine the operating costs and the capital costs we have introduced the specific efficiency, defined as

$$\text{s.e.} = \frac{\text{e.i.}}{A_{dl}} \tag{22}$$

where A_{dl} is the dimensionless total membrane area, defined as

$$A_{dl} = (\Sigma A_m) \frac{P_j \pi_h}{\Sigma F_m \delta} \tag{23}$$

where ΣA_m is the total surface area of a cascade, ΣF_m is the total feed flow rate, and P_j is the permeability of a reference component, usually the most permeable one.

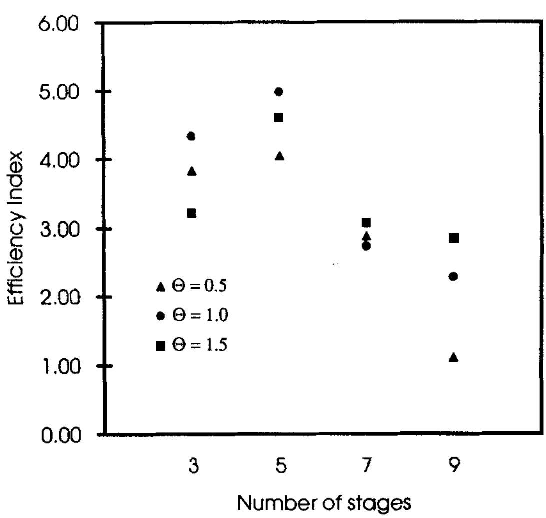

FIG. 8 Efficiency index of the various columns.

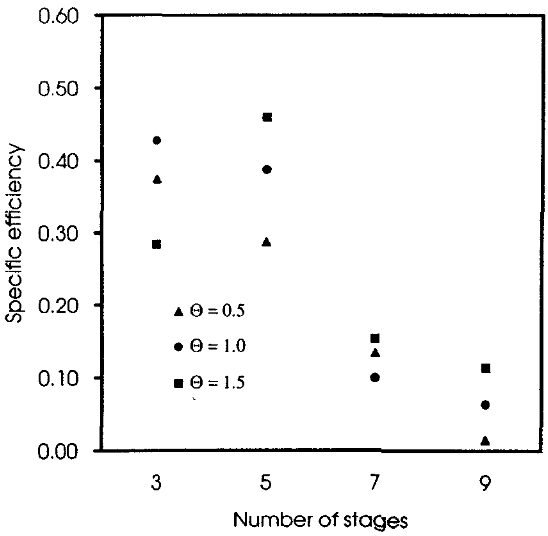

FIG. 9 Specific efficiency of the various columns.

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Figure 9 illustrates the values of the specific efficiency. The trend of the data clearly shows that after five stages there is a marked decrease in specific efficiency. This can be explained by considering that the increase in the number of stages raises the separation yield, but this is obtained at the expense of membrane surface area investments and recycling stream work. All this suggests that in permeating cascades there is a stage threshold beyond which any increase in the number of stages is hardly of economic interest.

CONCLUSIONS

The studied cases indicate that increasing the number of stages in a separation cascade also increases the separation yields, but the efficiency index and the specific efficiency decrease markedly.

One of the factors that lowers the efficiency of a cascade is the pressure drop across the membrane: a 5-fold decrease in the pressure drop across a membrane decreases the efficiency of separation by one order.

In accordance with other authors (13, 17, 19), we, too, conclude that membrane staging appears to be unattractive with regard to both capital investment and operating costs, at least beyond a certain number of stages that depends on specific problems.

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It is important to point out that a single permeation stage may be considered ideal when the membrane is semipermeable, i.e., permeable to only one component. Moreover, this stage has a potentially reversible configuration since by increasing the membrane surface area the driving forces may be decreased to zero for an infinite membrane surface area. With real membranes that have a certain value of selectivity and an internal resistance to transport, a single permeation stage is intrinsically irreversible. Moreover, when these stages are interconnected in a cascade, the resulting configuration is also intrinsically irreversible due to the mixing entropy of the recycled streams.

A clear disadvantage of a permeation cascade is the impossibility of recovering energy put into the process as compression work, since this energy in the form of heat must be withdrawn from the system in order to maintain isothermal conditions. Thus the operating costs of the membrane process increase with respect to processes based on equilibrium phenomena, such as distillation processes. In this case there are two streams, not in equilibrium, entering a stage; this nonequilibrium creates the driving force of separation, producing two streams, in equilibrium, that feed two other stages. The flow of streams not in equilibrium allows the reuse of the energy, as heat flow, from stage to stage.

SYMBOLS

A_m	membrane surface area of stage *m*
A_{dl}	dimensionless total membrane area
δ	thickness of the membrane
F_m	feed flow rate at the stage *m*
M	total number of stages
NC	total number of component the feed mixture
P_m	permeate flow rate from stage *m*
P_i	permeability of the component *i*
$\pi_{h,m}$	high permeating pressure at the stage *m*
$\pi_{l,m}$	low permeating pressure at the stage *m*
Q_m	stream rate to stage *m*
R_m	retentate flow rate from stage *m*
$\Delta S_{mix}/R$	adimensional mixing entropy
θ_m	splitting factor (R_m/P_m) of stage *m*
V	variance
$x_{m,i}$	molar fraction of the component *i* in the retentate stream from stage *m*
$y_{m,i}$	molar fraction of the component *i* in the permeate stream from stage *m*

$w_{m,i}$ molar fraction of the component i in the stream to stage m
$z_{m,i}$ molar fraction of the component i in the feed to stage m
$'$ local variable

REFERENCES

1. Anonymous, *Chem. Eng.*, p. 37 (April 1990).
2. S. Weller and W. A. Steiner, "Engineering Aspects of Separation of Gases: Fractional Permeation through Membranes," *Chem. Eng. Prog.*, *46*, 585 (1950).
3. R. W. Naylor and P. O. Backer, "Enrichment Calculation in Gaseous Diffusion: Large Separation Factor," *AIChE J.*, *1*, 95 (1955).
4. W. P. Walawender and S. A. Stern, "Analysis of Membrane Separation Parameters. II. Countercurrent and Cocurrent Flow in a Single Permeation Process," *Sep. Sci.*, *7*, 553 (1972).
5. C. Y. Pan and H. W. Habgood, "An Analysis of a Single-Stage Gaseous Permeation Process," *Ind. Eng. Chem. Fundam.*, *13*, 323 (1974).
6. S. A. Stern and S. C. Wang, "Countercurrent and Cocurrent Gas Separation in a Permeation Stage. Comparison of Computing Methods," *J. Membr. Sci.*, *4*, 141 (1978).
7. G. Schulz, H. Michele, and U. Werner, "Membrane Rectification Columns for Gas Separation and Determination of the Operating Lines Using the McCabe–Thiele Diagram," *Ibid.*, *12*, 183 (1982).
8. N. Boucif, S. Majumdar, and K. K. Sirkar, "Series Solution for Gas Permeators with Countercurrent and Cocurrent Flow," *Ind. Eng. Chem. Fundam.*, *23*, 470 (1984).
9. R. Rautenbach and W. Dahm, "Simplified Calculation of Gas Permeation Hollow-Fiber Modules for the Separation of Binary Mixtures," *J. Membr. Sci.*, *28*, 319 (1986).
10. K. R. Krovvidi, A. S. Kovvali, S. Vemury, and A. A. Khan, "Approximate Solution for Gas Permeators Separating Binary Mixtures," *Ibid.*, *66*, 103 (1992).
11. K. Li, D. R. Acharya, and R. Hughes, "Mathematical Modelling of Multi-Component Membrane Permeators," *Ibid.*, *52*, 205 (1990).
12. J. K. F. Keurentjes, L. J. M. Linders, W. A. Beverloo, and K. van't Riet, "Membrane Cascades for the Separation of Binary Mixtures," *Chem. Eng. Sci.*, *47*, 1561 (1992).
13. S. L. Matson, J. Lopez, and J. A. Quinn, "Separation of Gases with Synthetic Membranes," *Ibid.*, *38*, 503 (1983).
14. R. W. Spillman, "Economics of Gas Separation Membranes," *Chem. Eng. Prog.*, p. 41 (January 1989).
15. Y. Shindo, T. Hakuta, H. Yoshitome, and H. Inoue, "Calculation Method for Multicomponent Gas Separation by Permeation," *Sep. Sci. Technol.*, *20*(5&6) 445 (1985).
16. D. W. Brubaker and K. Kammermeyer, "Separation of Gases by Plastic Membranes—Permeation Rates and Extent of Separation," *Ind. Eng. Chem.*, *46*, 733 (1952).
17. C. Y. Pan and H. W. Habgood, "Gas Separation by Permeation. Part I. Calculation Methods and Parametric Analysis," *Can. J. Chem. Eng.*, *56*, 197 (1978).
18. S. A. Stern, T. F. Sinclair, P. J. Gareis, N. P. Vahldieck, and P. H. Mohr, "Helium Recovery by Permeation," *Ind. Eng. Chem.*, *57*, 49 (1965).
19. S. A. Stern, in *Membrane Separation Process* (P. A. Meares, Ed.), Elsevier, Amsterdam, 1976.

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