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A Comparison of Countercurrent Recycle Membrane Cascades with Some "One-Compressor" Recycle Permeators for Gas Separations in Terms of Ideal Crossflow Stages

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ABSTRACT

A comparison is made of countercurrent recycle membrane cascades (5-stage constant recycle and 6-stage no-mix) with continuous membrane columns, two-unit series, and one-unit recycle membrane modules for the enrichment of O_2 from air, assuming ideal crossflow stages. The crossflow stage equations permit easy comparisons and give insight into the inefficiencies associated with the "one compressor" module designs. For the separation specified (production of O_2 enriched up to $y_P = 0.983$, at a rate $P = 17.6$, from a feed $x_F = 0.21$, rate $F = 100$), the one compressor designs require from about 90 to 16 *times* more total compressor duty, and from about 50 to 10 *times* more membrane area than the no-mix cascade design, depending on module design, to make the same separation. The one unit recycle module is least efficient, followed by the continuous membrane column, and the two unit series module. The basic reasons for the inefficiencies associated with the one compressor modules relative to the recycle cascades are explored.

INTRODUCTION AND BACKGROUND

The literature abounds with papers dealing with the possible efficiency of different membrane module arrangements for gas separations. Most of these deal with module arrangements limited to one, possibly two, compressors, with the implication that *any* membrane-based separation scheme will not be economical if more compressors (and stages) are required. However, none of these studies have made side-by-side comparisons of these "one-compressor" (OC) modules with multistage countercurrent recycle membrane cascades (CRMCs). Such a comparison is the purpose of the present paper which shows that properly designed CRMCs require only a small fraction of the membrane area and compressor duty requirements of the OC modules for some separations. Reasons for the wide difference in the theoretical efficiencies between the OC modules and the cascade configurations are explored.

IDEAL "CROSSFLOW" (NO-MIXING) STAGE

Different flow patterns in membrane stages can result in significantly different stage separation efficiencies, with countercurrent (plug) flow of feed and permeate resulting in the best stage separation (1). However, some modern membranes are manufactured with asymmetric hollow fibers in which a relatively thin active membrane layer is supported on a relatively thick porous support. For such membranes the gas that permeates through the active membrane layer flows perpendicular to, and away from, the membrane layer through the porous support before joining the bulk of the permeate. Under these conditions the permeators behave as a "crossflow" device (2). This flow pattern will be assumed in modeling the different membrane modules and cascades in this study. Not only will this model be realistic for some types of membranes, the resulting equations are very well suited for analyzing the efficiencies of the different modules and cascades.

Consider the "crossflow" ideal membrane stage depicted in Fig. 1. In this stage it is assumed that the gas on the high pressure side of the membrane flows parallel to the membrane with no mixing, and that the mem-

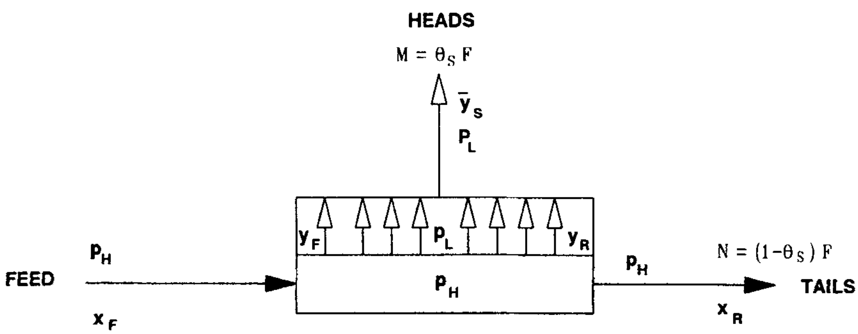


FIG. 1 Schematic diagram of a general crossflow stage.

brane is situated sufficiently far away from the exiting permeate stream so that the permeate flows perpendicular to, and away from, the membrane, again with no mixing taking place near the permeate side of the membrane. With these assumptions, the permeate composition at *any point* near the membrane is determined by the relative rates of permeation of the high pressure gas mixture at that point. The exiting permeate composition from this stage is the pooled average permeate composition.

Saltonstall (3) derived analytically exact stage equations for this ideal crossflow stage for a binary feed mixture. These equations are:

$$y^2 - [1 + r(\Omega^* - 1) + rx]y + \Omega^* rx = 0 \tag{1}$$

$$\theta_s = 1 - \left(\frac{\Omega^* - y_R}{\Omega^* - y_F}\right)\left(\frac{1 - y_R}{1 - y_F}\right)^a \left(\frac{y_R}{y_F}\right)^b \tag{2}$$

$$\bar{y}_s = \frac{x_F - x_R}{\theta} + x_R \tag{3}$$

$$A_s = \frac{\theta_s F_s(\Omega^* - \bar{y}_s)}{p_L R^1 (r - 1)(\Omega^* - 1)} \tag{4}$$

Equation (1) relates *point* compositions on either side of the membrane in terms of pressure ratio and the ideal local point separation factor. Equation (2) relates the *point permeate boundary* compositions for a given stage cut. Equation (3) gives the pooled average permeate composition from material balance around the stage, while the required stage membrane area is given by Eq. (4).

In these equations:

$$a = \frac{1 - \Omega^* r}{r - 1}, \qquad b = \frac{\Omega^* r}{r - 1} - 1, \qquad R^1 = \frac{P^1}{t} \tag{5}$$

$$\alpha^* = \frac{P^1}{P^2}, \qquad \Omega^* = \frac{\alpha^*}{\alpha^* - 1}, \qquad r = \frac{p_H}{p_L}, \qquad \theta_s = \frac{M_s}{F_s} \tag{6}$$

where P^1 and P^2 are the permeability coefficients for the more, and less permeable species, respectively, and p_H and p_L are the pressures on the feed and permeate sides of the membrane. θ_s is usually referred to as the "cut" or "stage cut."

To illustrate the performance of CRMCs, and to contrast the difference between CRMCs and some of the OC membrane modules, it is instructive to use examples for the enrichment of O_2 from air using membrane properties that might be characteristic of some commercially available (polysulfone) membranes. The properties and variables used are:

$$P^{O_2} = \frac{0.00278 \text{ ft}^3(\text{STP})\cdot\text{mil}}{\text{ft}^2\cdot\text{min}\cdot\text{atm}}$$

$$\alpha^* = P^{O_2}/P^{N_2} = 6.0 \tag{7}$$

Calculations were made assuming these properties together with t = 0.5 mil; and assuming the design variables of p_H = 6.80272 (atm) = 100 psia, P_L = 1 (atm); for F = 100 ft^3(STP)/min with x_F = 0.21 for some CRMCs and OC modules. The calculations were conveniently made using a spreadsheet computer program utilizing the crossflow stage equations, incorporating appropriate boundary conditions for the various stages in the cascades and modules. The results of the calculations are presented and discussed below.

COUNTERCURRENT RECYCLE MEMBRANE CASCADES

A schematic flow diagram for a CRMC is shown in Fig. 2. This particular cascade has 5 membrane stages (1 stripping and 4 enriching stages), 3 "recycle" compressors, and 1 combined recycle–feed compressor. Note that the *recycle flow pattern* is characteristic of a general countercurrent recycle cascade (CRC), that is, the tails stream from stage i + 1 and the heads stream from stage i − 1 combined make up the feed to a general stage i. Also note that the term "recycle" compressor is somewhat inconsistent with the concept of recycle in a CRC where *recycle* is associated with the *tails* streams from the next higher numbered stage (i.e., i + 1), which is combined with the heads stream from the next lower numbered stage (i.e., i − 1) to form the feed to stage i. Benedict et al. present an excellent development of CRC theory (4).

For the separation of the binary mixture there are six external design variables: F, P, B, x_F, y_P, and x_B. However, these variables must satisfy the material balance relationships:

$$F = P + B$$

$$x_F F = y_P P + x_B B \tag{8}$$

With six variables and two equations which relate the variables, it is possible to specify four of the external variables independently. However, since the cascade design is limited to a discrete number of ideal crossflow stages, exact values of all four "independent" variables in all cases cannot be specified, but rather, are fixed by what the chosen design (e.g., by what the 5-stage cascade with one stripping, four enriching stages, with

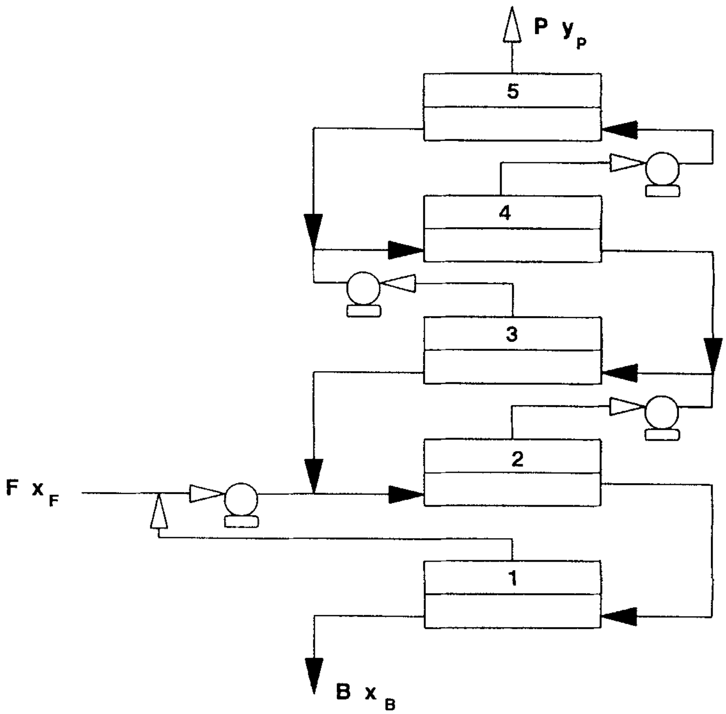


FIG. 2 Schematic diagram of a 5-stage CRMC with one stripping and four enriching stages.

the tails stream from stage $F + 1$ equal to x_F, and with RR specified), will yield.

An iterative procedure is used: for a specified F, x_F, and RR, target values of P and y_P are assumed. These in turn fix B, x_B, and all heads and tails stream rates in the cascade. Calculations are then made starting at the feed stage using the appropriate crossflow stage equations, and "operating lines" (material balance equations around the stripping and enriching sections), to yield calculated values of y_P and x_B. These in turn yield new values of P and B by material balance. Iterations are made until the assumed and calculated values are within a convergence criterion, that is, when all stage "equilibrium" and material and material balance equations are essentially satisfied.

Figure 3 shows a "McCabe–Thiele"-type plot, together with stage α, stage area, and stage RR for a 5-stage CRMC designed for a recycle ratio

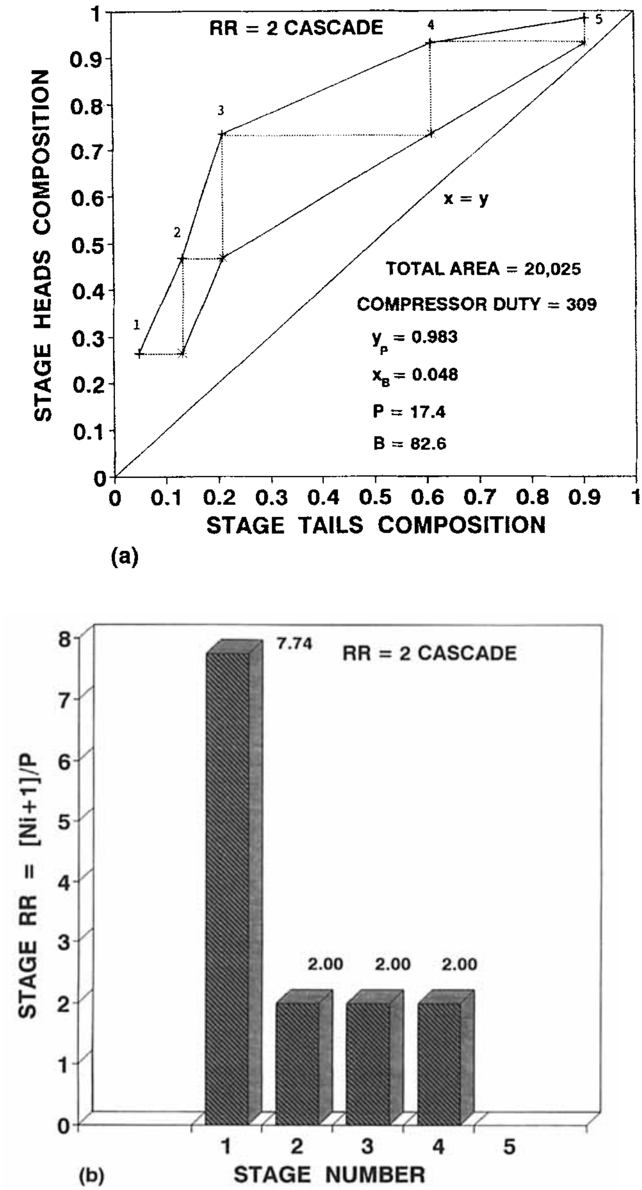


FIG. 3 Five-stage CRMC designed for $RR = 2$. (a) "McCabe–Thiele" plot; (b) stage RR; (c) stage α; (d) stage area, as a function of stage number.

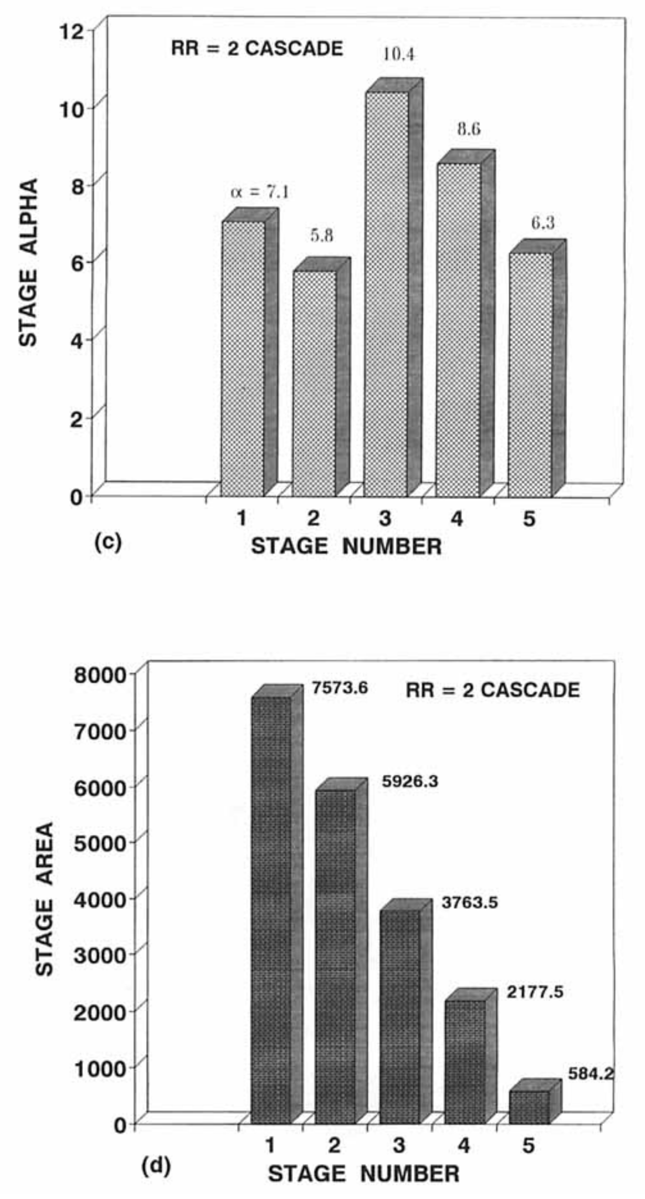


FIG. 3 Continued

$(RR) = N_{i+1}/P = 2$. The stage αs are defined as

$$\alpha_i = \frac{\dfrac{\bar{y}_i}{1 - \bar{y}_i}}{\dfrac{(x_i)_R}{1 - (x_i)_R}} \tag{9}$$

For this design the tails streams from the three enriching stages above the feed stage all have flow rates equal to $2 \times P$, while N from stage 2, the feed stage, is equal to $RR + F$. This design results in the four

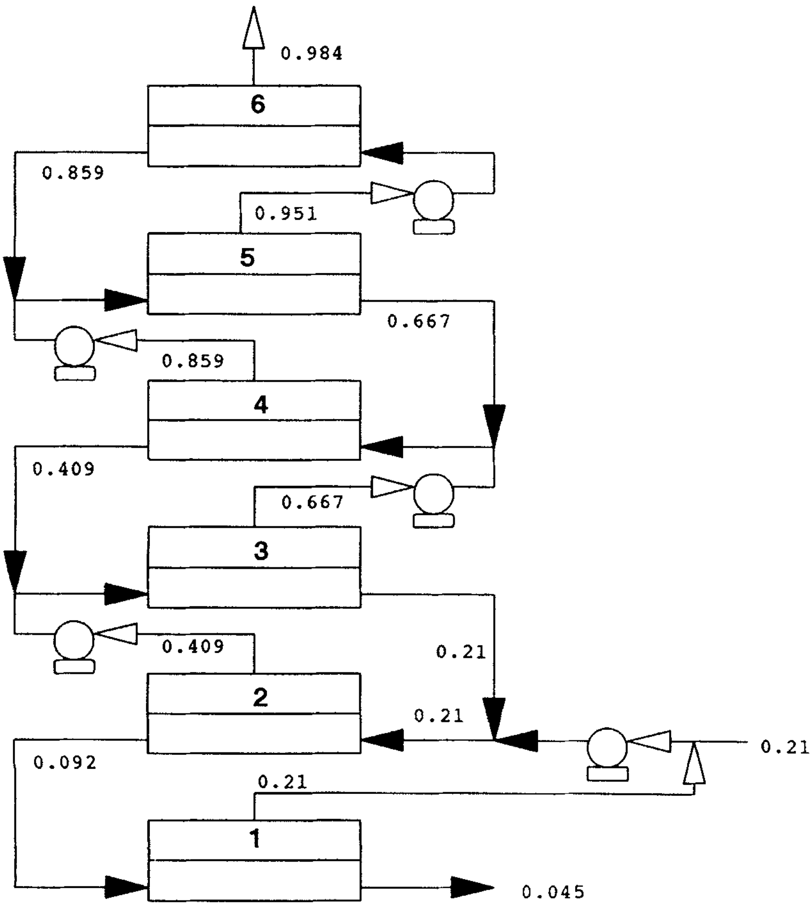


FIG. 4 No-mix CRMC showing the different heads and tails stream compositions.

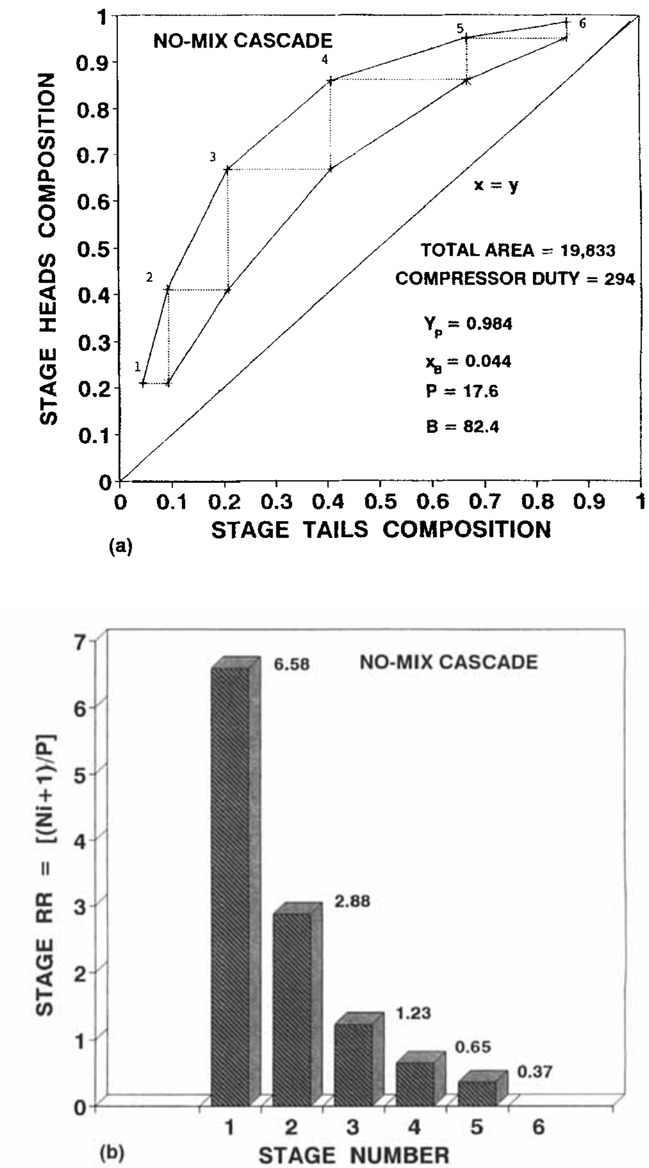


FIG. 5 No-mix CRMC. (a) "McCabe–Thiele" plot; (b) stage *RR*; (c) stage α; (d) stage area, as a function of stage number.

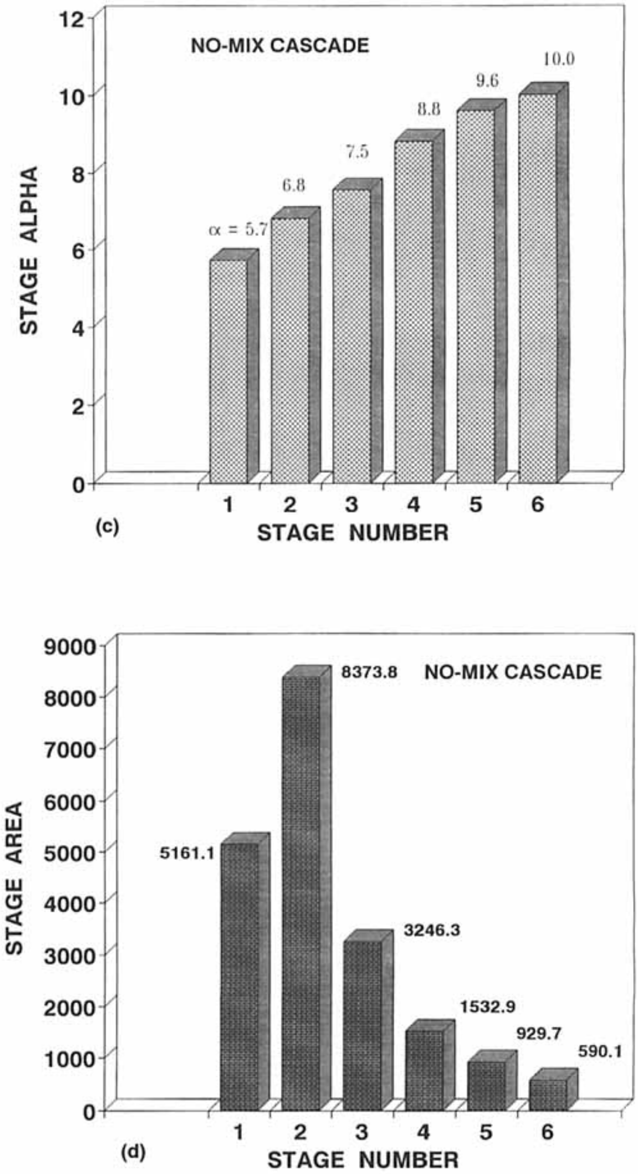


FIG. 5 Continued

"independent" variables being fixed at $y_P \approx 0.983$, $P \approx 17.4$, $x_B \approx 0.048$, and $B \approx 82.6$, with resulting stage αs and required stage areas shown in the figure.

NO-MIX CRMC

A special case of a CRMC is a no-mix cascade, designed such that the various heads and tails streams that combine within the cascade to form the feed to all stages have equal compositions. A schematic diagram of such a cascade is shown in Fig. 4, which also presents all calculated stream compositions for the example discussed below.

An iterative solution similar to that required for the above constant *RR* CRMC is again required to make the spreadsheet calculations.

For this cascade design, 6 stages (5 enriching and 1 stripping) are required to give about the same separation as for the 5-stage, $RR = 2$ design of Fig. 4. Figure 5 presents the "McCabe–Thiele" plot together with stage α, stage area, and stage *RR* as a function of stage number for comparison with the results of the previous example.

Significantly, the no-mix design requires a different *RR* at each stage. As a result, somewhat less total area and compressor duty are required for this design compared with the constant *RR* case, even though one more stage is required to make the same separation.

These two cascade designs will be discussed in more detail below, but here it should be noted that many cascade designs are possible which would result in about the same separation, each design requiring different stagewise *RR* and number of stages, and hence total area and compressor duty. However, a properly designed (IDEAL no-mix) cascade should result in the least total area and compressor duty of all designs (vide infra).

ONE-COMPRESSOR MEMBRANE MODULES

Many "one-compressor" recycle membrane modules can also be designed that will achieve good separation, and many published studies have implied that these devices are the only types of membrane modules that *might be* economically practical to make most separations, because of the cost of multiple compressors (5–7). However, side-by-side comparisons of CRMCs with these OC modules are not reported in the open literature.

THE CONTINUOUS MEMBRANE COLUMN (CMC)

The continuous membrane column (CMC) was introduced by Hwang and Thorman in 1980, who claimed it to be a revolutionary separation

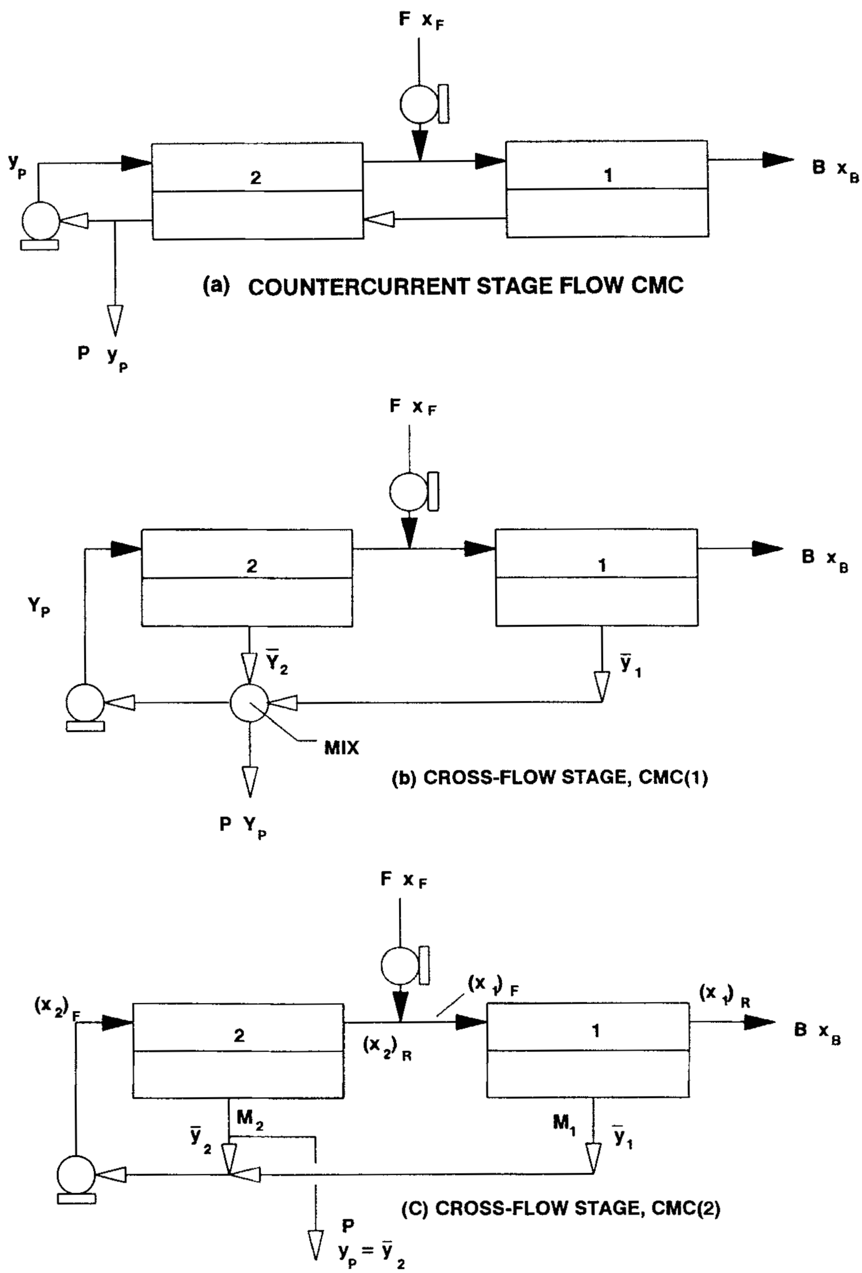


FIG. 6 Three possible "continuous membrane column" flow configurations. (a) Countercurrent stage flow CMC; (b) and (c) crossflow stage CMCs.

technique because it is capable of achieving any desired separation without cascading (5). In a theoretical study it was shown that the CMC design could meet any y_P specification with any given membrane area and with any membrane permselectivity (6).

The original CMC concept envisioned true countercurrent flow throughout the module as shown in Fig. 6(a).

In the no-mix CMC module design, feed is introduced on the high pressure side where $x = x_F$. This arrangement effectively splits the CMC into two stages where the reject stream from stage 1 is the depleted product stream, rate B, with composition x_B. The entire (countercurrent) permeate stream from both stages is enriched to y_P. Part of this stream constitutes the enriched product stream, rate P, while the rest is compressed to p_H and returned as "recycle" as the only feed to stage 2. Stage 2 is referred to as the enriching section, while stage 1 is the stripping section of the CMC.

A consideration of the assumptions of the permeate flow in the *crossflow* stages indicates that the permeate which is sent to the enriching section from the stripping portion of the CMC will have no effect on what happens in the enriching section with respect to the composition of the stage 2 permeate, unlike true countercurrent flow in the two stages. Thus, the CMC flow configuration in terms of the ideal crossflow stage configuration can be viewed in (at least) two ways which are shown Figs. 6(b) and 6(c). Calculations were made for both of these latter flow configurations for comparison with the CRMC designs. For these calculations it is assumed that the feed is introduced on the high pressure side of the CMC exactly where the composition equals the feed composition. This requirement, together with the desired product compositions, fixes all the boundary compositions in the two sections of the CMC, resulting in a very simple solution of the Saltonstall stage equations to model the CMC(1) and CMC(2) configurations. Calculations were made for the same separation as for the previous examples. The results will be discussed in detail below where all the CRMC and OC designs are compared (Table 1), but significantly, the CMC designs require from about 18 to 26 times more membrane area and from 35 to 52 times more compressor duty as compared with the no-mix CRMC to make exactly the same separation!

TWO UNIT SERIES (TUS) AND ONE UNIT RECYCLE (OUR) MEMBRANE MODULES

A 1978 study by Onno et al. (8) compared a two-unit series recycle membrane module with a "conventional" (OUR) module and concluded that the TUS module always enhances the overall separation that can be

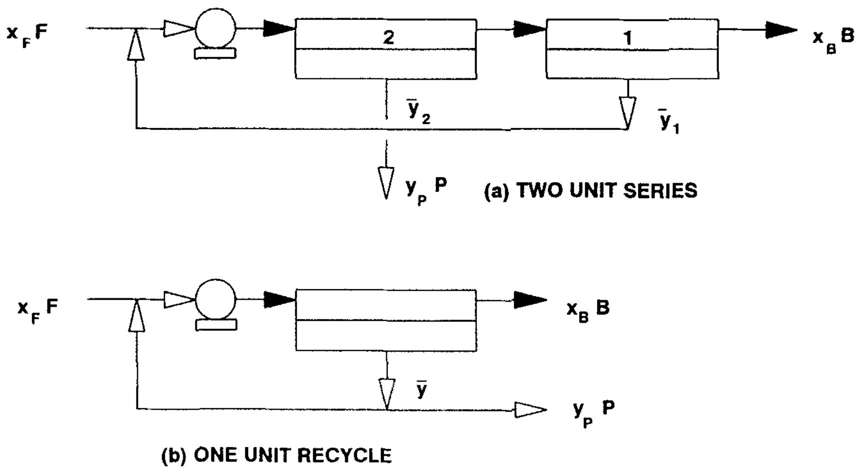


FIG. 7 Schematic diagrams of TUS and OUR membrane modules.

achieved, while requiring less membrane area and compressor duty. The TUS and OUR modules are shown schematically in Fig. 7.

In the "conventional" OUR module, enriched permeate is mixed with the feed stream as "recycle," while the TUS module utilizes a stripping stage in series to produce the permeate "recycle" which is mixed with the feed to stage 2. In the OUR module both the enriched product and recycle are enriched up to composition y_P, while in the TUS arrangement the enriched product is the entire permeate stream from stage 1, with all of the "recycle" coming from the stage 2 permeate.

TABLE 1
Comparison of the Different Membrane Configurations Making about the Same Separation

Membrane configuration	y_P	x_B	P (cfm)	B (cfm)	Total area (ft^2)	Compressor duty (cfm)
OUR	0.983	0.0449	17.6	82.4	926,209	27,624
CMC(1)	0.983	0.0449	17.6	82.4	511,767	15,300
CMC(2)	0.983	0.0449	17.6	82.4	356,678	10,390
TUS	0.983	0.0449	17.6	82.4	189,127	4,656
5S-*RR*2	0.9834	0.0470	17.41	82.59	20,025	309
No-mix	0.9839	0.0445	17.62	82.38	19,834	294

These two membrane modules are easily modeled with the crossflow equations by use of the appropriate stage boundary conditions for the specified separation. For this study the same separation of the CRMC and CMC in the previous examples was specified so that a side-by-side comparison could be made. The results of the calculations are presented in Table 1, and show that the OUR module is the least efficient of all membrane configurations studied, while the TUS module has better efficiency than the CMCs but is still much less efficient than the multistage CRMC arrangements for the specified separation.

GENERAL DISCUSSION

The present paper presents for the first time a side-by-side comparison of multistage CRMCs with the OC membrane modules, using membrane properties that might be typical of commercially available membranes for the separation of air. The specified separation for the comparison was rather arbitrary, being that which resulted using the CRMC designed for 5 stages with $RR = 2$. This separation resulted in $y_P \approx 0.983$, $x_B \approx 0.045$, $P \approx 17.6$ ft^3(STP)/min with $F = 100$ ft^3/min. This product rate is about 2140 lb/day of enriched O_2. Some of the results of the calculations are presented in Table 1 for comparison of required membrane area and compressor duty.

The comparison shown in Table 1 is truly striking considering the claims made for the CMC module over the years since it was introduced in 1980. The multistage CRMC design requires significantly less membrane area and power input (compressor duty) than the CMC design and the other two OC modules. As can be seen, the 6-stage no-mix CRMC requires the least area and compressor duty for the specified separation, with the 5-stage $RR = 2$ design only requiring slightly more. The area and compressor duty requirements relative to the no-mix design for the different membrane configurations are shown in Table 2. Here the total compressor duty is the sum of all stream flow rates to the various compressors.

As can be seen, of those considered, the OUR design is the least efficient for the specified separation, requiring 46.7 and 94 times more area and compressor duty, respectively, than the no-mix CRMC design. In comparison, the CMC designs require from 18 to 25.8 times more area and from 35.3 to 52 times more compressor duty. Although the TUS design is more efficient than CMC and OUR designs, it still requires 9.5 times more area and 15.8 times more compressor duty than the no-mix CRMC design. The 5-stage $RR = 2$ CRMC design requires only slightly more area and compressor duty than the no-mix CRMC.

TABLE 2
Relative Area and Compressor Duty Requirements for the Different Configurations

Membrane configuration	Area relative to no-mix	Compressor duty relative to no-mix
OUR	46.7	94.0
CMC(1)	25.8	52.0
CMC(2)	18.0	35.3
TUS	9.5	15.8
5-Stage, RR = 2	1.01	1.05
6-Stage, no-mix	1	1

Because of the large differences in efficiencies, it is interesting (and important) to try to determine the basic reasons for these differences.

All of these designs are capable of producing highly enriched products: CMCs, TUS, and OUR modules designed for higher and higher "recycle" will result in higher and higher enrichments, while in the CRMCs the same overall separation achieved in the OC designs can be accomplished with designs that incorporate the "right" combination of number of stages *and* stage recycle ratios. In the latter case, many designs are possible which will accomplish the desired separation, while only one CMC(2), TUS, and OUR design will result in this separation.

Consider the CMC(2) and TUS modules designed to produce $y_P = 0.983$ with $P = 17.6$ ($x_F = 0.21$, $F = 100$). Each design will require specific values of $(x_1)_F$, $(x_1)_R$, $(x_2)_F$, $(x_2)_R$, M_1, $\bar{y}_1$, M_2, and $\bar{y}_2$ to accomplish the specified separation. Figure 8 shows these quantities for comparison with the various stream flow rates and compositions presented for the constant RR and no-mix CRMCs in Fig. 11.

For the case of CMC(2) the no-mix conditions at the feed point requires that

$$(x_2)_R = x_F = (x_1)_F = 0.21$$

while $(x_1)_R = x_B \approx 0.045$ by overall material balance. The stage calculations then yield the stream flow rates and compositions shown in Fig. 8. For this case the first (stripping) stage is small compared with the second (enriching) stage since it must only reduce the feed side from $(x_1)_F = 0.21$ to $(x_1)_R = x_B$. The total area and compressor duty requirements are dominated by what must occur in stage 2 to result in the desired enrichment for the CMC(2) design. For the discussions that follow, refer to the

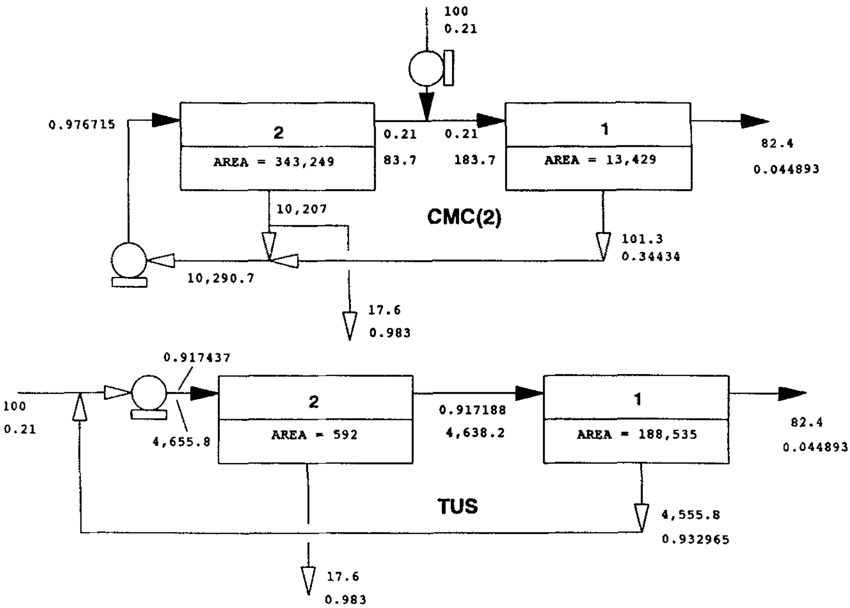
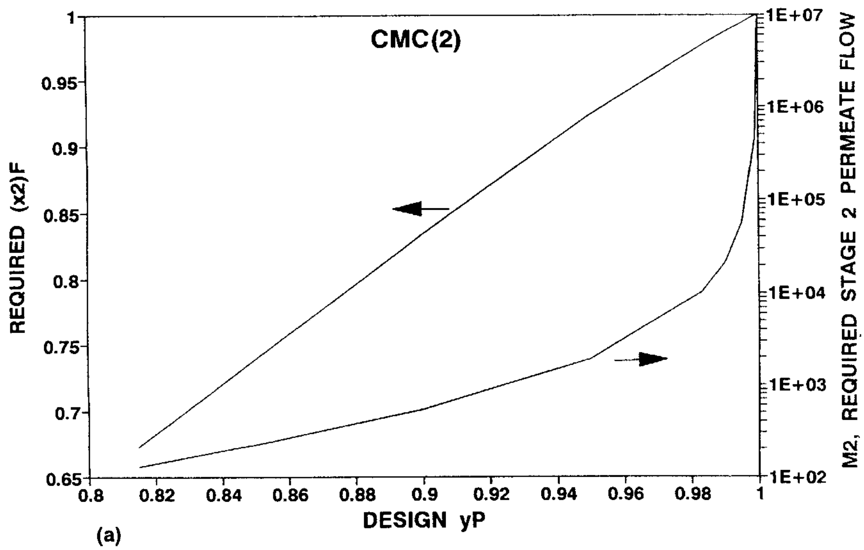


FIG. 8 Area, stream flow rates, and compositions for CMC(2) and TUS designed to produce $y_P = 0.983$, $P = 17.6$.

crossflow stage equations (Eqs. 1 to 4) together with the schematic diagram of CMC(2) to aid in visualizing the effects of recycle on CMC(2) overall enrichment.

Since $(x_2)_R$ is fixed at x_F, the stage 2 contribution to the recycle stream $(M_2 - P)$ must be large enough to result in a value of $(x_2)_F$ which will yield a stage 2 permeate with composition $\bar{y}_2 = y_P$. For this case $(x_2)_F$ must equal 0.976715, which requires a stage cut $\theta_2 = 0.991869$ and a value of $M_2 = 10{,}207$ ft^3/min ($\bar{y}_2 = 0.983$) from stage 2 to increase the composition of the combined recycle streams from stages 1 and 2 up to the required value of $(x_2)_F$. Thus, the mechanism for increasing the enrichment with increasing "recycle" in the CMC(2) design is simply a matter of increasing $(x_2)_F$ with enriched permeate to result in the desired value of $\bar{y}_2 = y_P$. A very large amount of "recycle" is required to accomplish this, especially at higher enrichments. Figure 9 presents the required stage 2 permeation rate and the required $(x_2)_F$ as a function of the (design) y_P for a series of CMCs, all designed to produce the indicated y_P at a rate of $P = 17.6$.



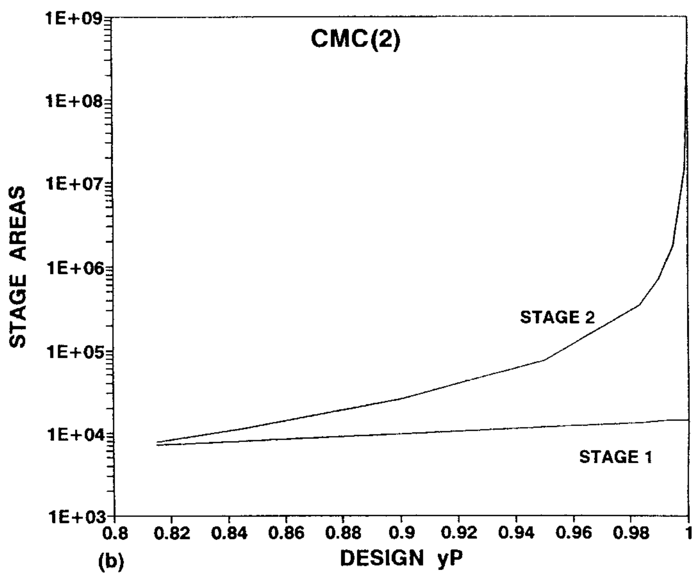


FIG. 9 Series of CMC(2)s designed to produce various y_P with $P = 17.6$. (a) required $(x_2)_F$ and M_2 for the indicated y_P; (b) stage areas required for the design y_P.

As can be seen from this figure, there is a very rapid increase in the stage 2 permeate, and the resulting area requirement for y_P is greater than about 0.95. For CMC(2) all of the permeate from stage 2 (P + required recycle) must be enriched up to y_P.

A similar analysis of CMC(1) would show that more stage 2 permeate/area is required than in CMC(2) because *all* of the combined stage 1 and stage 2, plus the product stream, must have composition y_P, which requires a higher value of $(x_2)_F$ because $\bar{y}_2$ must be larger than y_P to accomplish this increase in $(x_2)_F$.

The TUS [sometimes referred as a two stripper-in-series module (6)] design is more efficient than the CMC designs for this separation, but still considerable less efficient than the CRMC designs. As shown in Fig. 8, for this design only P must be enriched up to y_P, which results in a low area requirement for stage 2, while all of the recycle is produced in stage 1. However, for this case the permeate (recycle) from stage 1 must be at a high enough rate, and at a high enough concentration, so that, when mixed with the fresh feed, $(x_2)_F$ is high enough to result in $\bar{y}_2 = y_P$. For this example, a very large recycle is required with corresponding large area, although it is less than 1/2 that required for CMC(2). Also, the fact that the high pressure stream in stage 2 must be reduced to x_B from a relatively high $(x_1)_F$ contributes to this large area requirement.

The OUR design is even less efficient than the two CMC designs because in the *single* stage the reject stream concentration must be reduced to x_B [$(x_1)_R = x_B$], which results in a much greater stage cut and area requirements. In addition, the recycle rate must be large enough so that the combined (recycle + feed) stream composition, $(x_1)_F$, is sufficient to produce $\bar{y}_1 = y_P$ with $(x_1)_R = x_B$. This is in contrast to the CMC designs in which the feed is introduced where $(x_1)_R = x_F = (x_2)_F = 0.21$, but, in this case, the stage 1 permeate is mixed with permeate from stage 2 which has a higher concentration than x_F.

The concentration gradient that develops on the feed (high pressure) side of the crossflow stage is necessary for the recycle of enriched permeate to increase the enrichment in the stage or module. Simple calculations using the perfect-mix equations of Stern and Walawender (1) show that recycle will not increase the separation over the no-recycle perfect-mix design for the OUR. Although, with recycle, the feed composition to the stage will be greater, the two product compositions must remain constant (for a specified cut) in order to satisfy both overall material balance and "equilibrium" requirements. For the recycle case the area must be larger to accommodate the higher permeation rate.

This perfect-mix stage behavior means that a CMC module with perfect-mix conditions on both sides of the membrane will give an overall *separa-*

tion equal to a single perfect-mix OUR regardless of the amount of recycle returned to stage 2.

However, it can easily be shown that a CRMC composed of perfect-mix stages can be designed to make any desired separation.

COUNTERCURRENT RECYCLE MEMBRANE CASCADES

Figures 3 and 5 present "McCabe–Thiele"-type plots, together with stage α, stage *RR* (N_{i+1}/P), and stage area as a function of stage number for the two CRMC designs under discussion. As can be seen, stage αs are not constant for these designs but vary with stage number. These stage αs are not part of the design specifications, but rather are fixed by the required cascade interstage flow rates and or interstage compositions as calculated from Eqs. (1)–(7). This is different than the "normal" McCabe–Thiele plot (as in distillation, for example), in which an "equilibrium" curve is specified which relates stage heads and tails stream compositions over the entire composition range. Another difference is that the "top" stage in the CRMCs has no recycle returned to it, although a CRMC could be designed which incorporates a recycle to the product stage.

The lower, "operating" lines are analogous to those in McCabe–Thiele plots, and represent material balance requirements for the stripping and enriching sections of the cascades as reflected in the locus of $(x_{i+1})_R$, $\bar{y}_i$ pairs which are the compositions of the heads and tails streams "passing" one another between stages.

The two cascade designs result in different interstage flow and area requirements, with the stage *RR* being decreased from the feed stage to the *P* stage in the enriching section, while it is constant in the constant *RR* design.

Significantly, the no-mix design requires less area and compressor duty requirements than for the constant *RR* design even though one more stage is required to make the same separation.

As previously mentioned, the term "recycle" has different meanings (and separation consequences) when referring to this phenomena for the OC modules and the CRMCs. In the former case it refers to permeate (heads) streams that are recycled back to the feed of the stage that produces it, or to another stage in the module with the purpose of 1) increasing the possible enrichment achievable in the module, and/or 2) increasing the recovery of the desired product(s). From the previous discussion for these one-compressor cases, the main mechanism for increased enrichment is increasing the composition of the feed to the stage that *produces the enriched product.*

The overall purpose of the recycle is the same in the CRMCs, but this is accomplished by *recycle* of *tails* streams from the next higher numbered stage (i.e., $i + 1$) to become part of the feed to stage i. Separation will *continue* toward the separation goal from stage-to-stage if this recycle stream rate is large enough. Thus, CRMCs can designed to achieve any desired separation, provided an adequate number of stages and sufficient recycle are used. However, for a fixed number of stages, separation is limited no matter how much recycle is used, unlike the CMC, TUS, and OUR designs as shown by the following example.

TWO-STAGE CRMC

Consider the two-stage CRMC shown in Fig. 10. As can be seen, the only physical difference between this design and the TUS design discussed above is that the feed is introduced between the stages. For the present discussion, only the no-mix case will be considered, that is, $(x_2)_R = x_F = (x_1)_F$. Also, all of the recycle for this design is produced in stage 1 with $(x_2)_F = \bar{y}_1$. Because $(x_1)_F = x_F$ and $(x_1)_R = x_B$, the magnitude of $\bar{y}_1$ is limited, hence y_P can only be in a limited range, unlike the CMC and TUS designs.

Figure 11(a) presents y_P, x_B, P, B, and $(x_2)_F = \bar{y}_1$ for a series of two-stage CRMCs designed for different $RR = N_2/P$ ($x_F = 0.21$, $F = 100$),

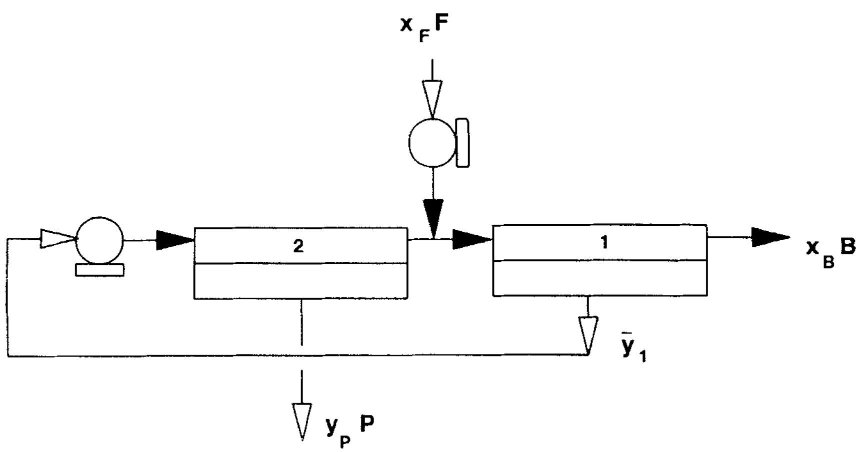
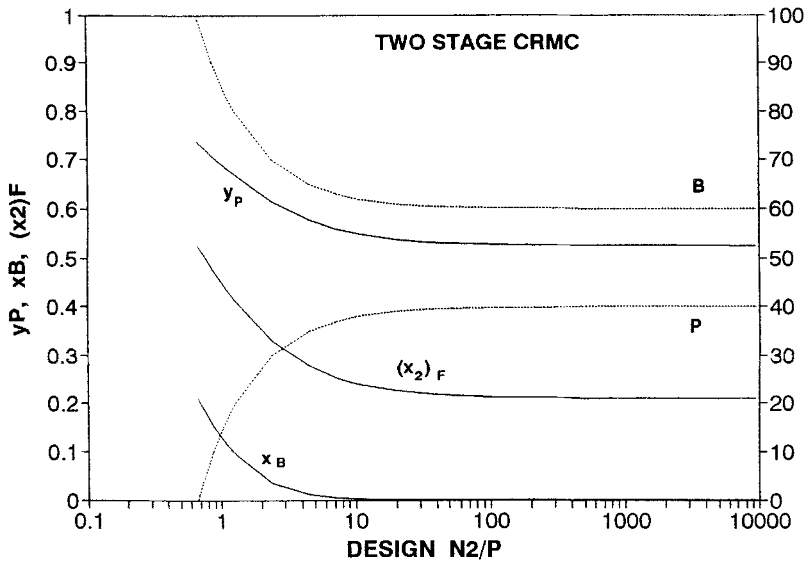
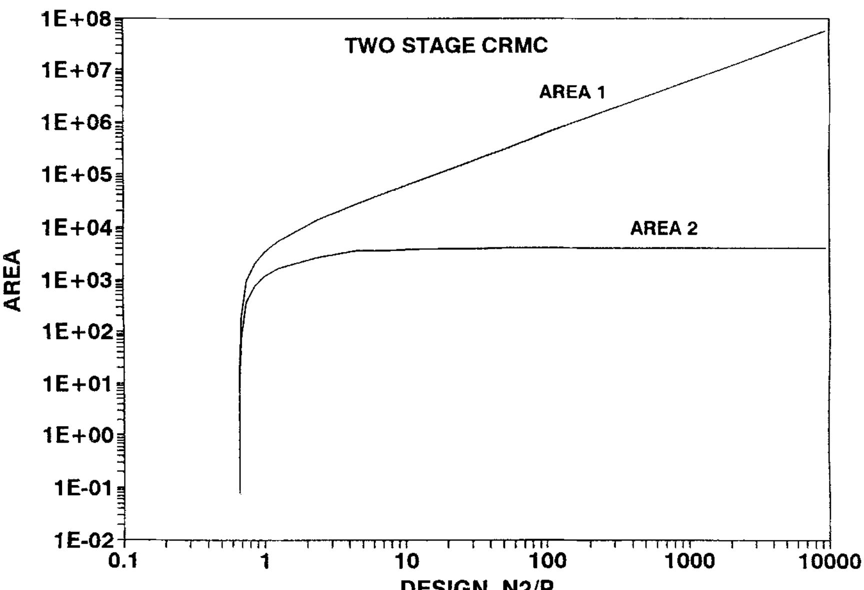


FIG. 10 Two-stage CRMC.



(a)



(b)

FIG. 11 y_P, x_B, P, B, $(x_2)_F$, and stage areas for a two-stage CRMC designed for various RR.

TABLE 3
Possible Compositions and Product Rates for Two-Stage (no-mix) CRMCs Designed for Different *RR*

$RR = N_2/P$	y_P	P	x_B	B
$\rightarrow 0$	$\rightarrow 0.736$	$\rightarrow 0$	$\rightarrow 0.21$	$\rightarrow 100$
2.14	0.679	17.6	0.11	82.4
10.5	0.549	38	0.002	62
∞	0.525	40	0	60

while Fig. 11(b) presents the required areas for the two stages for each *RR*.

For this design, $(x_2)_F$ can vary from about 0.525 at $RR = 0.6666$ (where $x_B \rightarrow 0.21$) to 0.21 as *RR* is made very large. The resulting y_Ps can vary from about 0.736 (for low *RR*) to 0.525 as the cascade is designed for large *RR* as shown in Table 3. A two-stage CRMC designed for $RR = 2.14$ would produce $P = 17.6$, $B = 82.4$, as in the previous comparison examples, but under these conditions $y_P = 0.679$, $x_B = 0.11$.

In the limiting case of zero recycle there would be no separation although $y_P \rightarrow 0.736$ as zero recycle is approached from finite values. In the other limiting case (infinite recycle), as $RR \rightarrow \infty$, $x_B \rightarrow 0$, and $\bar{y}_1 = (x_2)_F \rightarrow 0.21$, which requires that $y_P \rightarrow 0.525$ from Eq. (3). For these compositions, overall material balances require that $B \rightarrow 60$ and $P \rightarrow 40$.

Significantly, $x_B \rightarrow 0$ for the higher *RR*, being very close to zero at fairly low *RR*. For example $x_B \approx 0.002$ with $RR = 10.5$. However, the total required area for this design would be quite high, due mainly to the requirements for stage 1. Figure 11(b) presents area requirements for the two stages for the different *RR* designs. As can be seen, stage 1 area requirements become very large even for moderate *RR*. The basic reason for the large area requirements for stage 1 is that all of the recycle must be produced in that stage, and at higher *RR*, θ_1 must approach 1.

Thus, for the two-stage CRMC the overall separation is limited even as $RR \rightarrow \infty$, unlike the CMC and TUS designs. However, design experience shows that any desired separation can be made using multistage cascades, provided enough stages are used *and* a high enough $RR = N_{i+1}/P$ is used for each stage in the cascade.

MULTISTAGE CRMC

The stream flows and stream compositions for the 5-stage $RR = 2$ and the 6-stage no-mix CRMCs presented in Figs. 12(a) and 12(b) *clearly show*

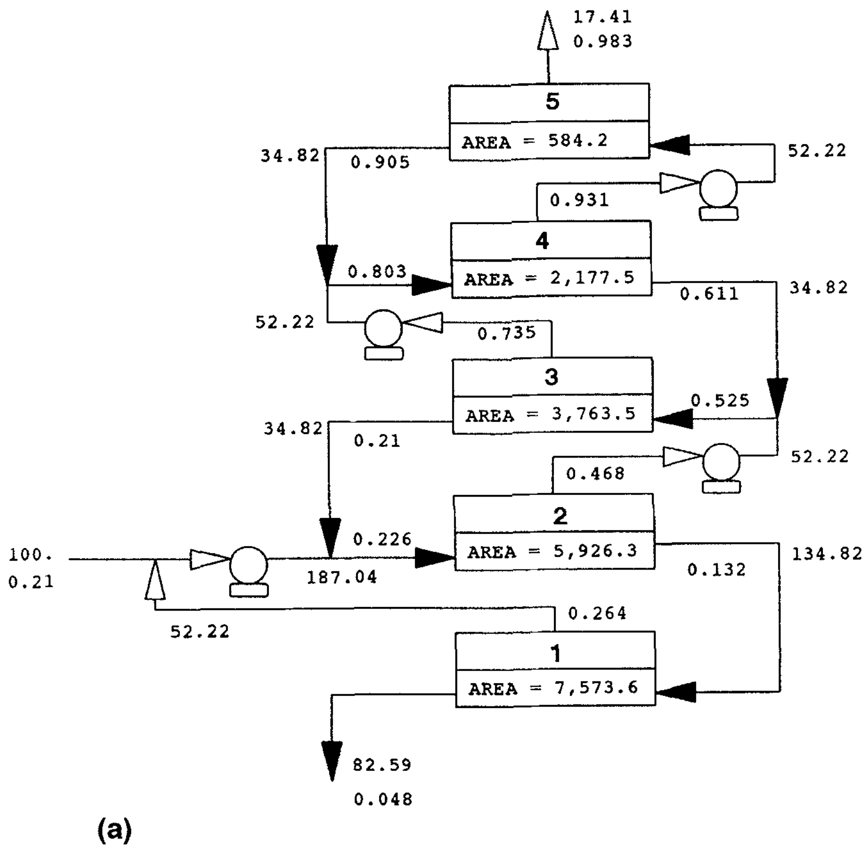
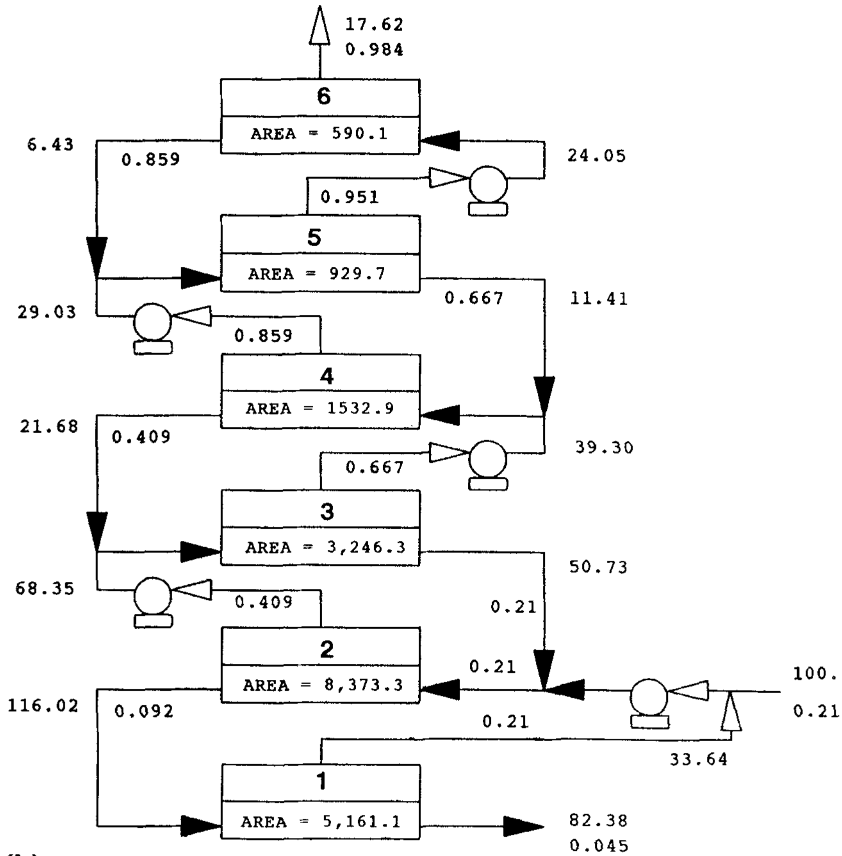


FIG. 12 Stream rates and compositions for the 5-stage $RR = 2$ CRMC and the 6-stage no-mix CRMC designed to produce $y_P \approx 0.983$, $P \approx 17.6$.

that CRMCs can be designed to accomplish a good separation utilizing small interstage flow rates compared with the OC designs.

Better separation could be achieved with CRMC designs that utilize more stages and/or higher stage *RR*. A detailed economic study would be required to determine the optimum design.

It is not known whether or not enriched O_2 produced in a CRMC plant would be competitive with O_2 produced by other processes (e.g., by PSA or cryogenic distillation), but the potential for a CRMC plant is probably better than for one based on one of the OC designs considering the fact that the CRMC design requires much less membrane area and compressor



(b)

FIG. 12 Continued

duty. A detailed plant design and economic analysis would be required before any firm conclusions can be made. Such a study was beyond the scope of the present study.

It is believed that CRMCs can be analyzed in ways very similar to CRCs (9), as discussed by Benedict (10). It is shown in CRC theory that separation will proceed from stage to stage in a multistage CRC provided that the recycle rate to all stages is higher than the respective stagewise minimum values. These minimum stage recycle values depend on stage αs, stage heads or tails compositions, and on the required (design) values of y_P, P, x_B, and B. As a result, it appears that a CRMC can be designed to achieve any desired separation of a binary mixture provided that *enough* stages are used *and* all interstage flow rates are above the respective minimum values. A great number of stages may be required if all stage αs are small, or if minimum flow rates are approached at any point in the cascade. In this study, however, it is shown that stage αs are relatively high, and that only a few stages and moderate interstage flow rates are required to obtain a relatively good separation for the membrane/feed system assumed. This study has shown that the required total interstage flows are much less in "properly designed" CRMCs than in OC designs, something that probably could not have been predicted prior to making the calculations. Thus, design experience is required before conclusions can be made regarding the efficiency of alternate membrane separation schemes. Some of the statements made in the above discussion assume that CRC theory directly applies to the membrane cascades.

In this regard, in "conventional" CRC theory (7), an *ideal cascade* is one in which:

1. The composition of the heads and tails streams forming the feed to a stage have the same compositions

$$\bar{y}_{i-1} = (x_i)_F = x_{i+1}$$

2. The heads separation factor $\beta_i = [\bar{y}_i/(1 - \bar{y}_i)]/[(x_i)_F/(1 - (x_i)_F)]$ is a constant.

When these conditions are met, it is easily shown that β_i is equal to the square root of the (constant) stage separation factor, α_i. It is possible for Condition 1 to be met but not Condition 2 (11). As discussed by Benedict et al. (4), the ideal cascade assures minimum total interstage flow of any CRC design. It is not known if this theory applies directly to the present CRMC designs where there is a rather wide variation in the magnitude of

TABLE 4
Stage α, β, Square root of α, as a Function of Stage Number for the No-Mix CRMC

Stage no.	Stage α	Stage β	$\sqrt{\alpha_i}$	$\beta_i / \sqrt{\alpha_i}$
1	5.71	2.61	2.39	1.09
2	6.80	2.61	2.61	1.00
3	7.54	2.89	2.75	1.05
4	8.81	3.05	2.97	1.03
5	9.60	3.15	3.10	1.02
6	10.00	3.18	3.16	1.01

the α_i; however, the no-mix CRMC discussed above nearly meets Condition 2 as shown in Table 4.

From this it appears that the present no-mix design is "close" to being an ideal cascade in which the total membrane area and compressor duty are near minimum values for the specified separation.

It is not known whether or not an ideal cascade can be designed for this system, and, to our knowledge, no one has presented a comprehensive theory for CRMCs composed of nonporous polymeric membrane stages for gas separations. As a result, some of the statements made above need be qualified to reflect that void. Such a study may be the subject of a future paper.

NOMENCLATURE

a	exponent in Eq. (2), defined in Eq. (6)
A	membrane area
b	exponent in Eq. (2), defined in Eq. (6)
B	bottoms (reject) rate from membrane module or cascade
CFM	gas rate, cubic feet per minute (STP)
CMC	continuous membrane column
CRMC	countercurrent recycle membrane cascade
F	feed rate to cascade, module or stage
M	heads rate from a membrane stage
N	tails rate from a membrane stage
OC	"one-compressor" membrane module, CMC, TUS, or OUR
OUR	one unit recycle membrane module
p	pressure
P	product rate
r	ratio of pure gas permeabilities in membrane
R	ratio of permeability coefficient to membrane thickness
RR	recycle ratio in a CRMC
t	membrane thickness
TUS	two unit series membrane module
x	composition of high pressure (feed) stream, mole fraction
y	composition of low preaaure (permeate) stream, mole fraction
$\bar{y}$	pooled average permeate composition leaving a stage
α	stage separation factor = $[(\bar{y}/(1 - \bar{y})]/[x_R/(1 - x_R)]$
α^*	ratio of pure gas permeabilities, Eq. (5)
β	heads separation factor
Ω^*	$= \alpha^*/(\alpha^* - 1)$
θ	cut or stage cut

Subscripts

i	general number in a stage
$i + 1$	stage which sends recycle to stage i in a CRMC
B	bottoms composition
F	feed composition to a module, cascade, or stage
H	high pressure
L	low pressure
P	product (tops) composition
R	reject composition from a stage
s	stage or stage number

Superscripts

*	indicates ratio of pure gas parameters
1,2	compound 1,2 in a binary gas mixture
O_2	permeability of O_2
N_2	permeability of N_2

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