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Attainability and Minimum Energy of Multiple-Stage Cascade Membrane Systems

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ABSTRACT

Process design and simulation of multi-stage membrane systems have been widely studied in many gas separation systems. However, general guidelines have not been developed yet for the attainability and the minimum energy consumption of a multi-stage membrane system. Such information is important for conceptual process design and thus it is the topic of this work. Using a well-mixed membrane model, it was determined that the attainability curve of multi-stage systems is defined by the pressure ratio and membrane selectivity. Using the constant recycle ratio scheme, the recycle ratio can shift the attainability behavior between single-stage and multi-stage membrane systems. When the recycle ratio is zero, all of the multi-stage membrane processes will decay to a single-stage membrane process. When the recycle ratio approaches infinity, the required selectivity and pressure ratio reach their absolute minimum values, which have a simple relationship with that of a single-stage membrane process, as follows: $S_n = \sqrt[n]{S_1}$, $\gamma_n = \sqrt[n]{\gamma_1}$, where n is the number of stages. The minimum energy consumption of a multi-stage membrane process is primarily determined by the membrane selectivity and recycle ratio. A low recycle ratio can significantly reduce the required membrane selectivity without substantial energy penalty. The energy envelope curve can provide a guideline from an energy perspective to determine the minimum required membrane selectivity in membrane process designs to compete with conventional separation processes, such as distillation.

Keywords: multi-stage membrane; selectivity; pressure ratio; attainability; minimum energy

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1. Introduction

Membrane processes have played an increasingly important role in many challenging gas separation systems, such as CO₂ capture and natural gas separation.[1] Our previous study on the attainability of a single-stage membrane process revealed that the selectivity S and pressure ratio γ are the only two controlling parameters [2]. Each of these parameters exhibits a minimum value to meet a certain separation task that is defined by the recovery ratio (η) and the enrichment factor (ϵ). This finding is consistent with many other studies in the literature [3, 4]. In real cases, these two parameters are often limited by many factors, including material properties, equipment limitations and economic feasibility. One solution to overcome these limitations is to use multiple membrane separation stages [5-8].

In a multiple stage membrane design, the retentate stream may feed to the next stage to extract more permeate to increase the recovery ratio, while the permeate stream may recycle back to the downstream stages to further purify the product. The stages to further purify the permeate streams are commonly named the enriching section, while the other stages are called the stripping section. The entire structure is called a membrane cascade, which is analogous to the flow cascade in distillation processes. Fig. 1 illustrates the most common membrane cascade design in which the permeate streams recycle one stage back to the previous stage. Typically, the concentration of the recycled streams will not be equal to the streams that they are mixing with, but if their concentrations are designed to be equal, this is called the non-mixing or ideal design. Studies on the separation performance, particularly the possible effect on the membrane area, as well as the power consumption of the ideal and non-ideal cascade designs have been discussed in the literature [9-11]. The process simulation of ideal and non-ideal membrane cascade designs is also well covered in the literature [7, 12-14]. However, the attainability and the minimum energy consumption of multi-stage cascade membrane systems have not been studied.

In this study, we aim to investigate the behavior of the attainability parameters in the most promising two- and three-stage membrane cascade systems. The well-mixed membrane model, which assuming the permeate side and the retentate side in each membrane stage are all well-mixed, is used to obtain simple mathematical models defining the system limiting parameters. This will serve the study objective in producing a conceptual design basis. The separation of propylene/propane, which is important in the petrochemical industry, is used as a bench system

to study the energy consumption, and the results are compared with those obtained using the standard distillation process.

Fig. 1: Schematic of cascade membrane system with recycle streams.

2. Analysis Methodology

All of the analyses performed in this study are based on a general membrane separation process schematically presented in Fig. 2. A binary gas mixture (x_A , T , P_0) is fed into a membrane unit and preferentially split into a permeate stream as a product and into a retentate stream as a by-product. The permeate stream is enriched in the highly permeable component. The separation task requires that the product meet a certain purity (y_A) and a certain recovery ratio (η). To obtain a well-defined energy consumption equation, we assume that the permeate and retentate streams leave the system at the same pressure and temperature as the feed stream (P_0 , T).

Fig. 2: Black box representation of a membrane process to perform a separation task to a binary mixture. The transmembrane pressure ratio is created by the feed compression. Energy recovery is also considered to minimize the separation energy penalty.

The two most common designs in two-stage systems are shown in Figs. 3a and 3b, which is often named the two-stage stripping cascade and the two-stage enriching cascade, respectively [15, 16]. The stripping cascade is also referred to as two strippers in a series permeator system

(TSSP) in the literature [17]. This design was found to be the most economically feasible design for a highly diluted target component in the feed mixture [18], while the enriching cascade system is reported as more efficient for the high-purity production of the less permeable component [19, 20]. In the two-stage stripping cascade, the retentate of the first stage feeds into the second stage, while the permeate of the second stage recycles back to the feed of the first stage to achieve a better recovery ratio. The recycle stream in this case is the permeate stream, whose flow rate is defined as Q and the concentration as y_r . Controlling the amount of Q is equivalent to controlling the stage-cut τ . A higher amount of Q indicates a higher stage-cut as well as recovery ratio. In the two-stage enriching cascade, the permeate of the first stage feeds into the second stage, while the retentate of the second stage recycles back to the feed of the first stage to achieve better product purity. The recycle stream in this case is the retentate stream, and the higher the amount of the recycle stream, the lower the stage-cut but the higher the product purity. The design in Fig. 3b appears to deviate from the cascade design, but by redrawing it as shown in Fig. 3c, one can clearly see that this design is indeed a cascade design, but the feed position shifts to the second stage. However, the recycle streams are different in these two cases. From these discussions, we can make a general definition for recycle streams as follows. For stages in the enriching section, the recycle streams are the retentate streams, while for stages in the stripping stages, the recycle streams are the permeate streams.

Fig. 3: Different designs of two-stage cascade membrane systems. a) two-stage stripping cascade; b) two-stage enriching cascade; c) a different drawing of the two-stage enriching cascade.

Following the same idea, the three-stage cascade membrane system will have three different designs, as shown in Fig. 4. It was found that the design in Fig. 4b is the most efficient to accomplish high enrichment from a moderate feed molar fraction (30 to 70%) [21]. This is because this design consists of both stripping and enriching stages; hence, it is expected to combine the advantages of the two-stage stripping and enriching cascades.

Fig. 4 Three cascade designs for three-stage membrane systems. (a) feed to the first stage (with only stripping stages); (b) feed to the second stage (with both enriching and stripping stages); (c) feed to the third stage (with only enriching stages).

3. Mathematical Models

Membrane selectivity (S) is defined as the permeability ratio of the most permeable component to other components in the multi-component mixture. The pressure ratio (γ) of each stage is defined as the ratio between its feed and permeate pressures. We also define the recycle ratio (ψ) as the normalized flowrate of a recycle stream to the flowrate of the feed stream of the system. Referring to the cascade membrane design in Fig. 1, the retentate streams flow upward, and the by-product is removed from the last stage, while the permeate streams of the upper stages flow one stage backward to the down stages, except for the last stream, which is removed as the product.

The attainability analysis and the minimum energy consumption calculations are performed under the following three assumptions. First, the well-mixed mode is used to simplify the mathematical derivation of mass transport in a membrane module. Thus, it is assumed that the membrane feed side has a homogenous concentration equivalent to the retentate stream concentration. Similarly, the permeate side of the membrane is considered to have a homogenous concentration equivalent to that of the permeate stream. Combining the mass balance formula for a binary gas mixture with Fick's law for diffusion, the following formula can be obtained to describe the mass transport in each stage of a membrane system.

$$\frac{y_A}{1 - y_A} = S \frac{\gamma r_A - y_A}{\gamma(1 - r_A) - (1 - y_A)} \quad (1)$$

Second, all of the stages are assumed to have the same membrane selectivity and the same pressure ratio. Membrane selectivity is typically an intrinsic property related to membrane

material and microstructure. If the same type of membrane is used in all of the stages, then a constant membrane selectivity can be a good assumption. If the pressure in the retentate does not lose much from the feed pressure, then it is possible to maintain the same pressure ratio in all of the stages with the minimum number of compression units. For the third assumption, two cases are considered in this study. The first case is to assume a constant stage-cut. This assumption has been commonly used in the literature [22, 23]. The second case is to assume a constant recycle ratio that is defined in this study for all of the stages. To the best of our knowledge, this is the first study to make such an assumption in cascade designs; thus, it requires additional explanations as follows. In the constant stage-cut assumption, all of the enriching stages have the same stage-cut τ_E , and all of the stripping stages have the same stage-cut τ_S . The feed stage has the same stage-cut as the stripping stages; therefore, a relationship between τ_E and τ_S can be established from the mass balance. A detailed derivation can be found in the literature [24]. Hence, when τ_E is used as a design variable, the entire cascade can be designed accordingly. However, it can be found easily from Fig. 1 that the flow patterns are different for the following stages: the first stage, the remaining enriching stages, the feed stage, the last stage, and the remaining stripping stages. Use of the constant recycle ratio assumption can improve this situation. In this case, the preceding five stages will have different stage-cuts. Fig. 5 shows the relationship between the stripping and enriching stage-cuts in the constant stage-cut scheme and the relationship between the first and last stage-cut in the constant recycle ratio scheme for a three-stage membrane cascade where the second stage is the feed stage. In both of the schemes, when the enriching stage-cut is increased, the stripping stage-cut is decreased. When the G/R ratio is less than one, there is not much difference between the two schemes. However, when G/R is larger than one, the difference is more significant. Particularly, when the enriching stage-cut is close to 1, the stripping stage-cut reduces to 0 in the constant recycle ratio scheme, but in the constant stage-cut scheme, the stripping stage-cut remains a substantial value. The constant recycle ratio scheme in this case is more reasonable because it indicates that most of the product in the enriching stages goes to the permeate stream; hence, a large permeate stream in the stripping stages is not required. The primary reason that the stripping stage-cut cannot reduce to 0 in the constant stage-cut scheme is because it is limited by the stage-cut of the feed stage, which is close to the G/F value.

Fig. 5: Relationship between the stripping stage-cut and enriching stage-cut at different G/R ratios in a three-stage membrane cascade with enriching and stripping stages. The dotted lines show the relationship in a constant stage-cut scheme, and the solid line shows the relationship in a constant recycle ratio scheme.

As an illustration of the model derivation procedure, the two-stage stripping cascade is presented here while the derivation of other configurations is presented in Appendix A and B. Carrying the mass balance over the first stage and then solving the well-mixed solution-diffusion model gives the following relationship between selectivity S , pressure ratio γ , feed concentration x_A , enrichment factor ε , recovery ratio η , recycle stream concentration y_r and ratio ψ :

$$S = \frac{x_A(\varepsilon\eta x_A - \gamma\psi\varepsilon - \gamma\varepsilon + \gamma\eta + \gamma\varepsilon x_A + \gamma\varepsilon\psi y_r - \gamma\eta\varepsilon x_A + \psi\varepsilon + \varepsilon - \eta - \varepsilon^2\psi x_A - \varepsilon^2 x_A + \eta\varepsilon x_A)}{(\gamma x_A + \gamma\psi y_r - \gamma\eta x_A - \varepsilon\psi x_A - \varepsilon x_A + \eta x_A)(\varepsilon x_A - 1)} \quad (2)$$

Similarly, the mass balance over the second stage yields the following equation:

$$S = \frac{y_r(\gamma\varepsilon - \gamma\eta - \gamma\varepsilon x_A + \gamma\eta\varepsilon x_A - \varepsilon + \eta + \varepsilon y_r - \eta y_r)}{(\gamma\eta\varepsilon x_A + \varepsilon y_r - \eta y_r - \gamma\varepsilon x_A)(y_r - 1)} \quad (3)$$

If the selectivity and pressure ratio are assumed the same for both stages, then Eq. (2) is equal to Eq. (3); therefore, the recycle stream concentration (y_r) can be calculated when the recycle ratio ψ is specified for a given feed and for a defined separation task. Hence, the behavior of selectivity as a function of the applied pressure ratio can be generated. The minimum selectivity ($S_{\min}$) can be obtained at an infinite pressure ratio. Hence, taking the limits of Eq. (2) and (3) in terms of infinity pressure ratio gives the following equation:

$$S_{\min} = \frac{y_r(\varepsilon - \eta - \varepsilon x_A + \eta\varepsilon x_A)}{\varepsilon x_A(1 - \eta)(1 - y_r)} \quad (4)$$

$$S_{\min} = \frac{x_A(\psi\varepsilon + \varepsilon - \eta - \varepsilon x_A - \psi\varepsilon y_r + \varepsilon x_A \eta)}{(\psi y_r + x_A - x_A \eta)(\varepsilon x_A - 1)} \quad (5)$$

Eqs. (4) and (5) indicate that the minimum required selectivity $S_{\min}$ is determined not only by the separation task (x_A , ε , η) but also by the recycle ratio ψ . A higher recycle ratio results in a lower $S_{\min}$. When the recycle ratio goes to infinity, the required selectivity is the absolute minimum below which the two-stage system can never meet the separation task. The absolute minimum selectivity of a two-stage membrane system is given by Eq. (6). The term in the square root is the same to calculate the minimum selectivity of a single-stage membrane system, as derived in our previous report [2]. Hence, the absolute minimum selectivity of two-stage systems is the square root of the single-stage minimum selectivity.

$$S_{\min} = \sqrt{\frac{\varepsilon(1 - x_A)}{(1 - \varepsilon x_A)(1 - \eta)} - \frac{\eta}{1 - \eta}} = \sqrt{S_1} \quad (6)$$

On the other hand, the minimum pressure ratio as a function of the recycle ratio can also be obtained from Eqs. (2) and (3) by taking the limit at infinite selectivity. This resulted in the following relationships:

$$\gamma_{\min} = \frac{y_r(\eta - \varepsilon)}{\varepsilon x_A(\eta - 1)} \quad (7)$$

$$\gamma_{\min} = \frac{x_A(\varepsilon\psi + \varepsilon - \eta)}{x_A + \psi y_r - x_A \eta} \quad (8)$$

The minimum required pressure ratio at a defined recycle ratio can be calculated by solving Eqs. (7) and (8). The absolute minimum pressure ratio is obtained when the recycle ratio goes to infinity to give the following simple form (Eq. 9). Again, the term under the square root is found equal to the minimum pressure ratio of a single-stage membrane system; thus, the absolute minimum pressure ratio of a two-stage membrane system is also the square root of a single-stage minimum pressure ratio.

$$\gamma_{\min} = \sqrt{\frac{\varepsilon - \eta}{1 - \eta}} = \sqrt{\gamma_1} \quad (9)$$

4. Minimum Energy of Multi-Stage Membrane Systems

Comparing the minimum work required by the membrane systems to that consumed by the conventional separation process for a specific application is of great importance to evaluate the technology potential from an energy efficiency perspective. The comparison will also help to

understand the technology limitations. In this discussion, it is assumed that the membrane process is equipped with an energy recovery system, and all of the work equipment (compressors, vacuum pumps and energy recovery system) are 100% efficient. Therefore, the energy consumption by a membrane system is the same regardless of whether the driving force is created by feed compression and/or by permeate vacuum pumping. Hence, the required work for the two-stage system with stripping cascade, two-stage with enriching cascade and three-stage with permeate and retentate recycle is derived in Eqs. (10), (11) and (12), respectively.

$$E_{min}^{2SC} = F \left[\psi + \frac{\eta}{\varepsilon} \right] C_p T \left(\gamma^{\frac{R}{C_p}} - 1 \right) \quad (10)$$

$$E_{min}^{2EC} = F \left[\psi + 2 \frac{\eta}{\varepsilon} \right] C_p T \left(\gamma^{\frac{R}{C_p}} - 1 \right) \quad (11)$$

$$E_{min}^{3S} = 2F \left[\psi + \frac{\eta}{\varepsilon} \right] C_p T \left(\gamma^{\frac{R}{C_p}} - 1 \right) \quad (12)$$

In this study, the membrane technology is to be evaluated against the conventional distillation process for the separation of a propylene/propane mixture. The bench system has a feed mixture at equimolar concentrations and is available at atmospheric pressure and ambient temperature. The separation objective is to produce propylene at a concentration of 96% with a recovery ratio of 95%. The minimum energy of a distillation column is the minimum heat that must be supplied in the reboiler to maintain the minimum vapor at the topmost tray. The minimum vapor flow rate can be calculated from the mass balance as follows:

$$V_{min} = D(1 + RR) \quad (13)$$

Therefore, the minimum energy of a conventional distillation column is as follows:

$$E_{min}^D = \lambda D(1 + RR) \quad (14)$$

The minimum reflux ratio (RR) can be estimated from the underwood formula as follows:

$$RR = \left[\sum_{i=1}^c \frac{\alpha_i x_{D_i}}{\alpha_i - \phi} \right] - 1 \quad (15)$$

where ϕ is the root of the following equation:

$$1 - q = \sum_{i=1}^c \frac{\alpha_i x_{F_i}}{\alpha_i - \phi} \quad (16)$$

A detailed derivation of these equations can be found in our previous report [2] or in the literature [25].

5. Results and Discussion

5.1 Attainability of multi-stage membrane systems

Our previous studies showed that for a single-stage membrane system, there is a boundary defined by the membrane selectivity S and pressure ratio γ where below this boundary, as shown in the shaded area in Fig. 6a, the separation task cannot be achieved. Accordingly, the curve defining the boundary is called the S - γ attainability curve. For multi-stage cascade membrane systems, this relationship between S and γ is also found to exist. For example, for a two-stage stripping cascade, an equation between S and γ can be obtained by combining Eqs. (2) and (3). The equation parameters include separation task parameters (x_A , ε , η) and the recycle ratio ψ . In the constant recycle ratio scheme, the recycle ratio ψ is assumed the same for all of the stages, while in the constant stage-cut scheme, the recycle ratio of each stage can be calculated from the enriching stage-cut value τ_E . A detailed derivation is shown in Appendix B.

Figs. 6b-f show the S - γ attainability curves under the constant recycle ratio assumption for the bench separation system by the two-stage stripping cascade, two-stage enriching cascade, and three-stage cascades with feed to the first, second and third stages, respectively. In all of these diagrams, the attainability curves move towards a lower selectivity and lower pressure ratio when the recycle ratio ψ increases, indicating that the required selectivity and pressure ratio decrease when the recycle ratio increases. It is interesting to observe in each diagram that the attainability curves are located within two boundaries. The high-end boundary is when the recycle ratio is 0. This boundary was identical in all of the diagrams and was identical to the attainability curve of the single-stage membrane system. This result indicates that when the recycle ratio decreases to 0, all of the multi-stage membrane systems decay to a single-stage membrane system. The low-end boundary is when the recycle ratio is infinity. In Fig. 6, the recycle ratio is studied only up to 100, while from the trend of different recycle ratios, the curve at an infinite recycle ratio should be very close. This boundary is identical for the same number of stages, regardless of the feed position. Because this is the low-end boundary, it can be defined as the absolute minimum selectivity and pressure ratio attainability curve. The diagrams in Fig. 6 show that the absolute minimum selectivity and pressure ratio for a single-stage are 461 and

19.4, for two-stages are 21.5 and 4.4, and for three-stages are 8.5 and 2.9, respectively. Hence, a square root relationship is realized between the absolute minimum selectivity and pressure ratio of the two-stage systems to the single-stage system, and a cubic root relationship is realized between the three-stage systems to the single-stage system. The square root and cubic root relationships are also derived mathematically in Eqs. (6) and (9) and in Appendix B. This relationship can be further extended to any multi-stage membrane systems as follows:

$$S_n = \sqrt[n]{S_1} \quad (17)$$

$$\gamma_n = \sqrt[n]{\gamma_1} \quad (18)$$

where S_n , γ_n , S_1 and γ_1 are the absolute minimum selectivity and the absolute minimum pressure ratio of n-stage and single-stage membrane systems, respectively.

Fig. 6: Attainability of multi-stage cascade membrane systems under the constant recycle ratio scheme for the bench separation system. (a) single-stage; (b) two-stage stripping cascade; (c) two-stage enriching cascade; (d) three-stage with feed to the first stage; (e) three-stage with feed to the second stage; and (f) three-stage with feed to the third stage.

Fig. 7 shows the attainability curves in a constant stage-cut scheme for two- and three-stage membrane systems. The separation task is the same as that in Fig. 6. For comparison purposes,

Fig. 7a shows again the attainability curve of the single-stage membrane system. The two-stage system in Fig. 7b and the three-stage system in Fig. 7d have no enriching section. In this case, the stripping stages will have a fixed stage-cut value; therefore, there is only one curve shown in each diagram. Fig. 7c shows the attainability curves of the two-stage enriching cascade at different enriching stage-cut values. When the enriching stage-cut decreases, the attainability curve moves towards the direction of a lower selectivity and lower pressure ratio. The enriching stage-cut value of 1 and 0 define the two boundaries of these curves. When the enriching stage-cut is 1, the attainability curve is identical to the single-stage membrane system in Fig. 7a. This is because in this case, the entire feed in the enriching stage will go to the permeate; therefore, there is no flow in the retentate stream. Because this can be achieved simply by a membrane stage with zero membrane area, it is equivalent to the case in which the enriching stage has disappeared, and only the stripping stage remains in the system. This explanation can also be used to understand the diagrams in Fig. 7e and 7f for three-stage membrane systems with feed to the second and third stages, respectively. In Fig. 7e, when the enriching stage-cut is 1, the attainability curve is identical to the two-stage stripping cascade shown in Fig. 7b, while in Fig. 7f, the attainability curve is identical to the single-stage membrane system. The reason is because the enriching stages in this case have disappeared; thus, two stages in Fig. 7e and one stage in Fig. 7f remain in the stripping section. When the enriching stage-cut becomes 0, the attainability curve in Fig. 7c is identical to the absolute minimum attainability curves for the two-stage systems in Figs. 6b and 6c, while the attainability curves in Figs. 7e and 7f are identical to the absolute minimum attainability curves for the three-stage systems in Figs. 6e and 6f. This can be explained by considering the flows in the first enriching stage in which the product is extracted from the permeate stream. To reach a stage-cut of 0, the feed flow must be infinity, along with the retentate flow and the recycle ratio. It can be easily derived that the recycle ratio in the stripping stages will also be infinity. Hence, in this case, the attainability behaviors of Fig. 7c is equivalent to that of Figs. 6b and 6c, and the attainability behaviors of Figs. 7e and 7f are equivalent those of Figs. 6e and 6f. From these discussions, a generalized conclusion can be drawn as follows for all multi-stage membrane systems in a constant stage-cut scheme. When the enriching stage-cut is 1, the attainability behaviors of multi-stage membrane systems will decay to only stripping stages, while when the enriching stage-cut is 0, the attainability behavior reaches the absolute minimum attainability curve of the same number of stages.

Fig. 7: Attainability of multi-stage cascade membrane systems under the constant stage-cut scheme for the bench separation system. (a) single-stage; (b) two-stage stripping cascade; (c) two-stage enriching cascade; (d) three-stage with feed to the first stage; (e) three-stage with feed to the second stage; and (f) three-stage with feed to the third stage.

5.2 Energy consumption of membrane systems

The energy consumption in multi-stage cascade systems is mainly used to drive the inter-stage pumps to provide the required pressure ratio in each stage. The energy consumption is a function of the selectivity and pressure ratio. However, the energy consumption is an optimal function over the pressure ratio. Eqs. (10)-(12) are derived to calculate the minimum energy at the optimal pressure ratio for single-stage, two-stage and three-stage membrane systems, respectively. The separation of propylene/propane is used as a bench system in which 96% purity of propylene in the permeate stream and 95% recovery ratio should be achieved from an equimolar feed. For comparison purposes, the energy consumption by a conventional distillation process to achieve the same separation task is calculated from Eq. (14). The physical properties used in the calculation are the following: $\alpha = 1.3$ (relative volatility of propylene to propane), $C_p = 56$ J/mol K (mixture specific heat capacity) and $\lambda = 18.8$ kJ/mol (propane latent heat of

vaporization). The minimum distillation energy is reached at the reflux ratio of approximately 7.0, where the energy consumption is approximately 78 kJ/mol.

Figs. 8a-8c show the energy consumption of the bench system under optimal pressure ratios in single-stage, two-stage enriching cascade and three-stage cascade with feed entering into the second stage, respectively. For single-stage membrane systems, the energy increases sharply when the selectivity is less than a certain value, and this value is the minimum selectivity at the optimal pressure ratio. This result indicates that a single-stage membrane system can achieve a much lower energy consumption, but the required selectivity is very high. For multi-stage membrane systems, when the recycle ratio is zero, the energy curves are equal to the single-stage membrane system. However, slightly increasing the recycle ratio, for example, a recycle ratio of 0.1 in all of the three multi-stage membrane systems, leads to slight reduction in energy consumption but a significant reduction in the required membrane selectivity. Three-stage membrane systems require much lower selectivity than that of the two-stage membrane systems, indicating that a higher number of stages results in more reduction in the required membrane selectivity. However, further increasing the recycle ratio will increase the energy consumption. When the recycle ratio is 100, the energy consumption in multi-stage membrane systems is too high to compete with the existing distillation process because too much energy will be required to drive inter-stage streams.

Fig. 8: Energy consumption by multi-stage cascade membrane systems for the bench separation system (a) single-stage; (b) two-stage enriching cascade; and (c) three-stage with feed to the second stage. The red dotted line shows the energy consumption of the distillation process for the bench separation system.

To demonstrate the effect of the recycle ratio, we can further use the mathematical technique known as “envelope of family of curves in plane” [26] to plot the tangent line to the lowest energy points at various recycling ratios, as illustrated in Fig. 9 for three-stage cascade with feed entering into the second stage. Each point on this curve reveals the minimum energy that can be achieved by the three-stage cascade at that membrane selectivity. For example, the envelope curve meets the distillation energy curve at a selectivity of approximately 10, indicating that when a membrane with a selectivity of 10 is used in each stage, the minimum energy that can be achieved by a three-stage cascade will be equal to the distillation process. Thus, for a three-stage membrane cascade to compete with the distillation process, the minimum required membrane selectivity should be approximately 10. Therefore, this curve will provide a guideline for selectivity in multi-stage membrane cascade designs.

Fig. 9: Illustration of the minimum energy consumption at different membrane selectivities in the three-stage system with feed to the second stage cascade system for the bench separation system. The blue line is the energy envelope curve generated by the theory of “envelope of family of curves in plane”. The red dotted line shows the energy consumption of the distillation process for the bench separation system.

6. Conclusions

Studies on the attainability curves and the minimum energy consumption of multi-stage membrane cascade systems use the simplest well-mixed membrane model under two schemes: constant recycle ratio and constant stage-cut. The following useful guidelines are obtained in both of the schemes. First, to meet a certain separation task that is defined by feed concentration x_A , product purity enrichment ϵ and recover ratio η , the attainability curves of multi-stage membrane cascade systems are defined by membrane selectivity S and pressure ratio γ . The

recycle ratio or enriching stage-cut will shift the attainability curve. When increasing the recycle ratio from 0 to infinity or when decreasing the enriching stage-cut from 1 to 0, the attainability curve will move towards a lower selectivity and lower pressure ratio. In the constant recycle ratio scheme, when the recycle ratio is 0, the attainability behavior of any multi-stage membrane cascade system will decay to that of a single-stage membrane process. When the recycle ratio approaches infinity, the selectivity and pressure ratio will reach the absolute minimum selectivity and absolute minimum pressure ratio, which have the following simple relationships with the minimum selectivity and pressure ratio of a single-stage membrane system: $S_n = \sqrt[n]{S_1}$, $\gamma_n = \sqrt[n]{\gamma_1}$. In the constant stage-cut scheme, when the enriching stage-cut is 1, the attainability behavior of multi-stage membrane cascade systems will decay to the membrane system that contains only the stripping stages. When the enriching stage-cut approaches 0, then it is equivalent to the infinity recycle ratio scheme, and the selectivity and pressure ratio will reach the absolute minimum selectivity and absolute minimum pressure ratio.

Second, the energy consumption of multi-stage membrane systems is primarily determined by membrane selectivity. In general, a higher membrane selectivity results in a lower energy consumption. Using a small recycle ratio will not substantially change the energy consumption but can significantly reduce the required membrane selectivity. This indicates the benefit of using recycle streams in the membrane cascade designs. However, using a large recycle ratio will cause a large energy penalty, although it may further reduce the required membrane selectivity and pressure ratio, and this benefit will become increasingly marginal at larger recycle ratios. The minimum energy consumption at different membrane selectivities can be found by the energy envelope. This envelope curve can provide a simple method to determine the minimum required membrane selectivity for multi-stage membrane designs to compete with conventional separation techniques from an energy perspective.

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Nomenclature

Roman letters

C_p	gas specific heat J/(mol·K)
D	distillate flow (mol/s)
E	separation energy (J/mol)
F	feed flow rate (mol/s)
G	permeate flow rate (mol/s)
P_0	feed initial pressure (Pa)
P_h	feed side pressure (Pa)
P_l	permeate side pressure (Pa)

R	retentate flow rate (mol/s) or ideal gas constant
r	molar fraction of retentate stream
$R_{\min}$	minimum reflux ratio
S_j	selectivity, defined as the permeability ratio of the most permeable component relative to component j
T	feed temperature (K)
T_{in}	compressor inlet temperature (K)
V	vapor flow rate, mol/s
x	molar fraction of feed stream
y	molar fraction of permeate stream

Greek Letters

α_i	relative volatility of component i
β	permeability, defined as the flux normalized with transmembrane pressure drop and membrane thickness (mol/m·s·Pa)
β_m	permeability of the most permeable component (mol/m·s·Pa)
γ	feed to permeate pressure ratio (P_h/P_l)
ε	target component enrichment (y_A/x_A)
τ	stage-cut defined as the flow rate ratio of permeate stream to the stage feed
λ	latent heat/enthalpy of vaporization (J/mol)
θ	underwood polynomial root
η	recovery ratio of the target component (Gy_A/Fx_A)
ψ	ratio between recycle stream flow rate to the raw feed flow rate (Q/F)

Subscripts

A	relating to component A
D	relating to distillate stream
h	relating to membrane high pressure side
l	relating to membrane low pressure
min	relating to the minimum
P	relating to permeate stream
R	relating to retentate stream or recovery stream
r	relating to recycle stream
n	relating to the number of stages

Superscripts

2SC	relating to a two-stage enriching cascade
2SC	relating to a two-stage stripping cascade
3S	relating to a three-stage design

Appendix A: Dimensionless Parameters

$$\text{Recovery Ratio } (\eta) = \frac{\text{TC permeate flow rate}}{\text{TC feed flow rate}} = \frac{G y_A}{F x_A}$$

$$\text{Enrichment } (\varepsilon) = \frac{\text{TC permeate concentration}}{\text{TC feed concentration}} = \frac{y_A}{x_A}$$

$$\text{Recycle Ratio } (\psi) = \frac{\text{Recycle stream flow rate}}{\text{feed flow rate}} = \frac{Q}{F}$$

$$\text{Pressure Ratio } (\gamma) = \frac{\text{Feed side pressure}}{\text{Permeate side pressure}} = \frac{P_h}{P_l}$$

Appendix B: Model Derivation

(1) Constant recycle ratio

Fig. B1 Schematic of N-stage membrane cascade with constant recycle ratio

By conducting an overall mass balance over the cascade system shown in Fig. B1, the following relationships can be obtained regardless of the number of stages.

$$G = G_1 = \frac{\eta}{\varepsilon} F \quad (\text{B1})$$

$$y_A = y_{A1} = \varepsilon x_A \quad (\text{B2})$$

$$R = R_N = \left[1 - \frac{\eta}{\varepsilon}\right] F \quad (\text{B3})$$

$$r_A = r_{AN} = \frac{\varepsilon x_A (1 - \eta)}{(\varepsilon - \eta)} \quad (\text{B4})$$

By definition of the constant recycle ratio, any retentate stream in the enriching stage has a constant flow rate of Q, and the ratio of Q to the feed flow rate is fixed and equal to ψ . Similarly, the recycle stream in a stripping stage is the permeate stream and has a flow rate and ratio to feed of Q and ψ , respectively. Therefore, the mass balance is dependent on the stage location and can be described for stage i in a cascade of N-stages as follows:

Enriching section

$$G_i = Q + G \quad (B5)$$

$$y_i = \frac{Q r_{i-1} + (Q + G)y_{i+1} - Q r_i}{Q + G} \quad (B6)$$

$$R_i = Q \quad (B7)$$

$$r_i = \frac{Q r_{i-1} + (Q + G)y_{i+1} - (Q + G)y_i}{Q} \quad (B8)$$

Feed stage

$$G_i = Q + G \quad (B9)$$

$$y_i = \frac{Q r_{i-1} + Fx_A + (Q)y_{i+1} - (Q + R)r_i}{Q + G} \quad (B10)$$

$$R_i = Q + R \quad (B11)$$

$$r_i = \frac{Q r_{i-1} + Fx_A + (Q + G) y_{i+1} - (Q + G) y_i}{Q + R} \quad (B12)$$

Stripping section

$$G_i = Q \quad (B13)$$

$$y_i = \frac{(Q + R) r_{i-1} + Q y_{i+1} - (Q + R) r_i}{Q} \quad (B14)$$

$$R_i = Q + R \quad (B15)$$

$$r_i = \frac{(Q + R) r_{i-1} + Q y_{i+1} - Q y_i}{Q + R} \quad (B16)$$

Substituting these equations in the well mixed model will result in N equations with $N+2$ unknowns (selectivity, pressure ratio, recycle ratio and $N-1$ concentrations). Therefore, the attainability behavior is obtained by fixing the recycle ratio and solving the model equations over the pressure ratio range. The obtained formulas for the two-stage enriching cascade are as follows:

$$S = \frac{(r_1 \psi + x_A \eta)}{\left(\frac{\varepsilon \psi + \eta}{\varepsilon}\right) \left(1 - \frac{\varepsilon(r_1 \psi + x_A \eta)}{\varepsilon \psi + \eta}\right)} \frac{\left[\gamma \left(1 - \frac{\varepsilon x_A (1 - \eta)}{\varepsilon - \eta}\right) + \frac{\varepsilon(r_1 \psi + x_A \eta)}{(\varepsilon \psi + \eta)} - 1\right]}{\left[\gamma \left(\frac{\varepsilon x_A (1 - \eta)}{\varepsilon - \eta}\right) - \frac{\varepsilon(r_1 \psi + x_A \eta)}{(\varepsilon \psi + \eta)}\right]} \quad (B17)$$

$$S = \frac{\varepsilon x_A}{1 - \varepsilon x_A} \frac{[\gamma(1 - r_1) + \varepsilon x_A - 1]}{[\gamma r_1 - \varepsilon x_A]} \quad (B18)$$

As recognized in our recent study about single-stage attainability, the minimum selectivity (S_{min}) to accomplish a certain separation task at a given recycle ratio exists at an infinite pressure ratio. Hence, taking the limits of Eqs. (B17) and (B18) gives the following equation:

$$S_{min} = \frac{(\eta x_A + \psi r_1)(\varepsilon(1 + \eta x_A - x_A) - \eta)}{x_A(\varepsilon(\eta x_A + \psi(r_1 - 1)) - \eta)(\eta - 1)} \quad (B19)$$

where,

$$r_1 = \frac{\varepsilon x_A}{S - S\varepsilon x_A + \varepsilon x_A} \quad (\text{B20})$$

Substituting Eq. (B20) into Eq. (B19) and then taking the limit as the recycle ratio goes to infinity, the absolute minimum selectivity to accomplish the separation task is as follows:

$$S_{min} = \sqrt[2]{\frac{\varepsilon(1 - x_A)}{(1 - \varepsilon x_A)(1 - \eta)} - \frac{\eta}{1 - \eta}} \quad (\text{B21})$$

To obtain the minimum pressure ratio at a given selectivity and recycle ratio, Eqs. (B2) and (B3) are solved for the pressure ratio, and the limit is taken as the selectivity goes to infinity. This gives the following relationships:

$$\gamma_{min} = \frac{\eta^2 x_A - \eta x_A \varepsilon + \psi \eta r_1 - \psi \varepsilon r_1}{x_A(\eta^2 + \eta \psi \varepsilon - \eta - \psi \varepsilon x_A)} \quad (\text{B22})$$

where,

$$r_1 = \frac{x_A \varepsilon}{\gamma} \quad (\text{B23})$$

Substituting Eq. (B23) into Eq. (B22) and then taking the limit as the recycle ratio goes to infinity, the absolute minimum pressure ratio to accomplish the separation task by this membrane configuration is as follows:

$$\gamma_{min} = \sqrt[2]{\frac{\varepsilon - \eta}{1 - \eta}} \quad (\text{B24})$$

To illustrate the systematic method followed to derive the relationships in a three-stage cascade, we select the system consisting of two stripping stages and a single enriching stage shown in Fig. 3b. The well-mixed model can be written for the three stages as follows:

$$S = \frac{\varepsilon x_A}{1 - \varepsilon x_A} \frac{[\gamma(1 - r_1) + \varepsilon x_A - 1]}{[\gamma r_1 - \varepsilon x_A]} \quad (\text{B25})$$

$$S = \frac{(r_1 \psi + x_A \eta)}{(\varepsilon \psi + \eta) - (r_1 \psi + x_A \eta)} \frac{\left[\gamma \left(1 - \frac{\varepsilon [x_A(1 - \eta) + \psi y_3]}{\varepsilon(\psi + 1) - \eta} \right) + \frac{\varepsilon(r_1 \psi + x_A \eta)}{(\varepsilon \psi + \eta)} - 1 \right]}{\left[\gamma \left(\frac{\varepsilon [x_A(1 - \eta) + \psi y_3]}{\varepsilon(\psi + 1) - \eta} \right) - \frac{\varepsilon(r_1 \psi + x_A \eta)}{(\varepsilon \psi + \eta)} \right]} \quad (\text{B26})$$

$$S = \frac{y_3}{1 - y_3} \frac{\left[\gamma \left(1 - \frac{\varepsilon x_A(1 - \eta)}{(\varepsilon - \eta)} \right) + y_3 - 1 \right]}{\left[\gamma \frac{\varepsilon x_A(1 - \eta)}{(\varepsilon - \eta)} - y_3 \right]} \quad (\text{B27})$$

Hence, the system is described by three nonlinear equations with five unknowns, which are selectivity, pressure ratio, recycle ratio, stage-1 retentate stream concentration (r_1) and stage-3 permeate stream concentration (y_3). Therefore, the system can be solved by fixing the applied pressure ratio and recycle ratio.

To obtain a formula that describes the absolute minimum selectivity existing at an infinite recycle ratio and pressure ratio because the selectivity in all of the stages is equal; hence, the selectivity also equals the cubic root of the product of the right hand sides of Eqs. (B25), (B26) and (B27).

$$S = \left[\frac{\varepsilon x_A}{1 - \varepsilon x_A} \frac{[\gamma(1 - r_1) + \varepsilon x_A - 1]}{[\gamma r_1 - \varepsilon x_A]} \frac{y_3}{1 - y_3} \frac{\left[\gamma \left(1 - \frac{\varepsilon x_A(1 - \eta)}{(\varepsilon - \eta)} \right) + y_3 - 1 \right]}{\left[\gamma \frac{\varepsilon x_A(1 - \eta)}{(\varepsilon - \eta)} - y_3 \right]} \right. \\ \left. \frac{(r_1 \psi + x_A \eta)}{(\varepsilon \psi + \eta) - (r_1 \psi + x_A \eta)} \frac{\left[\gamma \left(1 - \frac{\varepsilon [x_A(1 - \eta) + \psi y_3]}{\varepsilon(\psi + 1) - \eta} \right) + \frac{\varepsilon(r_1 \psi + x_A \eta)}{(\varepsilon \psi + \eta)} - 1 \right]}{\left[\gamma \left(\frac{\varepsilon [x_A(1 - \eta) + \psi y_3]}{\varepsilon(\psi + 1) - \eta} \right) - \frac{\varepsilon(r_1 \psi + x_A \eta)}{(\varepsilon \psi + \eta)} \right]} \right]^{1/3} \quad (\text{B28})$$

By taking the limits of formula (B28) at an infinite pressure ratio and recycle ratio, formula (B29) is obtained for the absolute minimum selectivity in the three-stage membrane system.

$$S_{min} = \sqrt[3]{\frac{\varepsilon(1 - x_A)}{(1 - \varepsilon x_A)(1 - \eta)} - \frac{\eta}{1 - \eta}} \quad (\text{B29})$$

Solving Eqs. (B25), (B26) and (B27) for the pressure ratio and then following the same solution methodology performed for the selectivity, Eq. (B30) is obtained for the absolute minimum pressure ratio (exists at infinite selectivity and recycle ratio).

$$\gamma_{min} = \sqrt[3]{\frac{\varepsilon - \eta}{1 - \eta}} \quad (\text{B30})$$

(2) Constant stage-cut

Fig. B2: Schematic of N-stage cascade system with a constant stage-cut

The overall mass balance will also give the same formulas described earlier in Eqs. B1 to B4. To derive the general formulas describing the stage permeate and retentate concentration and flow rate based only on the stage-cut (τ), it is been assumed that the feed can enter into any stage

in the cascade system with a flow of F_i . Therefore, conducting mass balance over stage i in an n -stage cascade results in the following relationships:

$$G_i = \tau_i [R_{i-1} + G_{i+1} + F_i] \quad (\text{B31})$$

$$y_i = \frac{R_{i-1} r_{i-1} + G_{i+1} y_{i+1} + F_i x_A - R_i r_i}{G_i} \quad (\text{B32})$$

$$R_i = (1 - \tau_i) [R_{i-1} + G_{i+1} + F_i] \quad (\text{B33})$$

$$r_i = \frac{R_{i-1} r_{i-1} + G_{i+1} y_{i+1} + F_i x_A - G_i y_i}{R_i} \quad (\text{B34})$$

where $F_i = 0$ for all of the stages, except for the first stripping stage. In addition, $\tau_i = \tau_E$ for an enriching stage and τ_S for a stripping stage. Substituting these equations in the well-mixed model will result in n equations with $N+2$ unknowns (selectivity, pressure ratio, enriching stage-cut and $N-1$ concentrations). Therefore, the attainability behavior is obtained by fixing the enriching stage-cut and solving the model equations over the pressure ratio range.

(3) Minimum energy of cascade systems

The assumption of equal pressure ratio throughout all of the cascade stages simplifies the system energy calculations. The minimum energy required to create the pressure ratio (γ) in the cascade system is equal to the sum of all of the permeate streams multiplied by the minimum compression specific energy.

Under the constant recycle ratio assumption, the permeate streams of the feed stage and enriching stages have identical flow rates and are equal to $Q+G$, while the stripping section permeate streams have an identical flow rate of Q . Therefore, the sum of permeates flow rate for the cascade system illustrated in Fig. B1 with N total number of stage and feed entering into stage Z can be described by the following general formula:

$$\sum \text{Permeates Flow Rate} = ZF \left(\frac{\eta}{\varepsilon} + \psi \right) + (N - Z)F\psi \quad (\text{B35})$$

The minimum compression specific energy assuming isentropic work is given as follows:

$$E_{min}^S = C_p T \left(\gamma^{\frac{R}{C_p}} - 1 \right) \quad (\text{B36})$$

Hence, the minimum energy of separation for a cascade system of equal recycle ratio is as follows:

$$E_{min} = \left(ZF \left(\frac{\eta}{\varepsilon} + \psi \right) + (N - Z)F\psi \right) C_p T \left(\gamma^{\frac{R}{C_p}} - 1 \right) \quad (\text{B37})$$

Graphical Abstract

Attainability of multi-stage membrane cascade systems are defined by selectivity and pressure ratio and can be shifted by recycle ratio from multi-stage to single-stage. The minimum selectivity of a multi-stage membrane system to have the same energy consumption as conventional separation processes can be found by the energy envelope curve.

Highlights

- (1) Attainability of multi-stage membrane systems is defined by selectivity and pressure ratio.
- (2) Recycle ratio shifts the attainability between single-stage and multi-stage systems.
- (3) A simple root relationship is found between multi-stage and single stage systems.
- (4) The energy envelope curve can be used to find the minimum required selectivity.
- (5) Recycle ratio in small value is beneficial in multi-stage systems.