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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: $S_n=\sqrt{S_1}$, $\gamma_n=\sqrt{\gamma_1}$, 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.

1. Introduction

Membrane processes have played an increasingly important role in many challenging gas separation systems, such as CO_2 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]. 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,

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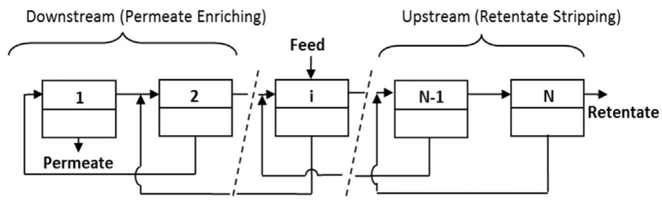

Fig. 1. Schematic of cascade membrane system with recycle streams.

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.

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).

The two most common designs in two-stage systems are shown in Fig. 3a and b, which is often named the two-stage stripping cascade and the two-stage enriching cascade, respectively [13,14] in the literature [15]. This design was found to be the most economically feasible design for a highly diluted target component in the feed mixture [16], while the enriching cascade system is reported as more efficient for the high-purity production of the less permeable component [17,18]. 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.

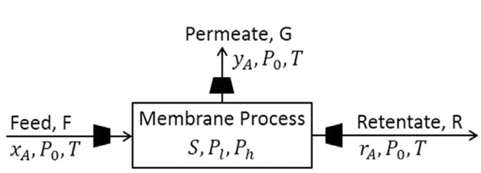

Fig. 2. Black box representation of a membrane process to perform a separation task to a binary mixture. The transmembrane pressure ratio is created by the feed compression. Energy recovery is also considered to minimize the separation energy penalty.

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 (30–70%) [19]. 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.

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:

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

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 [20,21]. 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 [22]. Hence, when τ_E is used as a design variable, the entire cascade can be designed accordingly.