



Attainability and minimum energy of single-stage membrane and membrane/distillation hybrid processes



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ABSTRACT

As an energy-efficient separation method, membrane technology has attracted more and more attentions in many challenging separation processes. The attainability and the energy consumption of a membrane process are the two basic fundamental questions that need to be answered. This report aims to use process simulations to find: (1) at what conditions a single-stage membrane process can meet the separation task that is defined by product purity and recovery ratio and (2) what are the most important parameters that determine the energy consumption. To perform a certain separation task, it was found that both membrane selectivity and pressure ratio exhibit a minimum value that is defined only by product purity and recovery ratio. The membrane/distillation hybrid system was used to study the energy consumption. A shortcut method was developed to calculate the minimum practical separation energy (MPSE) of the membrane process and the distillation process. It was found that the MPSE of the hybrid system is only determined by the membrane selectivity and the applied transmembrane pressure ratio in three stages. At the first stage when selectivity is low, the membrane process is not competitive to the distillation process. Adding a membrane unit to a distillation tower will not help in reducing energy. At the second medium selectivity stage, the membrane/distillation hybrid system can help reduce the energy consumption, and the higher the membrane selectivity, the lower is the energy. The energy conservation is further improved as pressure ratio increases. At the third stage when both selectivity and pressure ratio are high, the hybrid system will change to a single-stage membrane unit and this change will cause significant reduction in energy consumption. The energy at this stage keeps decreasing with selectivity at slow rate, but slightly increases with pressure ratio. Overall, the higher the membrane selectivity, the more the energy is saved. Therefore, the two fundamental questions are answered in a simple and clear manner. These results should be very useful to guide membrane research and their applications.

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1. Introduction

Membrane process is an energy-efficient separation process compared to conventional technologies, particularly to the most widely used thermal distillation process that currently exhausts more than 40% of the total energy in chemical industry [1]. A modern reverse osmosis seawater desalination plant consumes only about 1/3 to 1/4 energy of the conventional thermal distillation processes such as multi-stage flash, multi-effect distillation, etc. Such a huge potential in energy saving has stimulated very intensive research efforts in membrane studies in the last 50 years. As the energy issue becomes more and more serious, using

membrane technology to replace conventional methods becomes more attractive and more urgent in many challenging separation systems such as CO₂ capture, wastewater treatments, etc. Nevertheless, there are still a lot of barriers that have hindered applications of membranes in many potential industrial processes. Some are technically, such as membrane fouling and low membrane performance, but in many other cases it is also due to lack of clear understanding of the membrane process. For example, based on our literature studies we found that even for single-stage membrane process the answers for the following two fundamental questions have not been addressed in a simple and clear manner: (1) at what condition(s) the membrane process can or cannot meet the separation tasks; and (2) at what condition(s) the energy efficiency of a membrane process is competitive to a thermal process and what is(are) the most important parameter(s) that determine the minimum energy?

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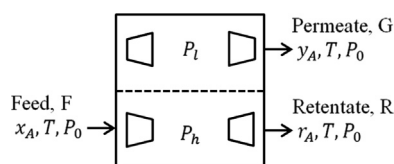


Fig. 1. Separation task performed by a single-stage membrane to a binary mixture. The pressure driving force can be created or recovered by pumps or energy recovery equipment shown inside the membrane box.

In this paper we aim to answer these questions by process simulations. Our analyses are based on the following general membrane separation process that is shown in Fig. 1. A binary mixture (x_A , T , P_0) is fed into a membrane unit and split into a permeate stream as a product and a retentate stream as a by-product. The separation task is that the product should meet certain purity (y_A) and certain recovery ratio (η). The purity requirement determines the concentration of the permeate stream while both requirements (purity and recovery ratio) determine the concentration of the retentate stream (r_A). For general applications, we assume both permeate and retentate streams should leave the system at the same pressure and temperature as the feed stream (P_0 , T). As the pressure within the membrane unit will typically change, so appropriate energy devices will be required to restore the pressure, as illustrated in Fig. 1.

Membranes have two important parameters named selectivity (S) and permeability (β). Permeability is defined as the pressure and thickness normalized flux of the penetrant component [2]. Selectivity is the ratio between the permeabilities of different components in the mixture [3]. We further assume that these parameters depend only on the intrinsic properties of the membrane material and are not a function of the operating conditions [4]. For most gas and liquid separation processes, the driving force of transport is transmembrane pressure drop. Therefore, pressure at the membrane feed side P_h is typically larger than pressure at the permeate side P_l . The transmembrane pressure drop can be created in different ways. The feed stream can be compressed to a higher pressure, which is typically the case for liquid separations such as seawater desalination, and then pressure of the retentate stream should be recovered back to the initial pressure through energy recovery systems; or the permeate side is evacuated by vacuum pump, which is often the case for gas separations, and then the product stream is compressed back to the initial pressure for delivery; or combination of both. The ratio of P_h to P_l is defined as pressure ratio $\gamma = P_h/P_l$. The first important result of this study is to find that pressure ratio is a very important parameter. Once it is combined with membrane selectivity they determine the attainability of a single-stage membrane process. However, the importance of this parameter has been generally overlooked in most of the membrane literature except it recently gained more attentions [5–8].

From the attainability studies it was found that a single-stage membrane process cannot meet the separation task under certain conditions, hence a membrane/distillation hybrid system was used as the benchmark model to evaluate the minimum energy consumption. This approach has the following benefits. First, the membrane/distillation hybrid system can meet the separation task at any membrane conditions. Hence, optimization of the membrane properties becomes possible when minimizing the energy. Second, the hybrid configuration is relatively simple. A superstructure can be found to cover most possible combinations between a membrane unit and a distillation tower. It can also cover a pure membrane unit and a pure distillation unit. Therefore, the true minimum energy consumption can be identified irrespective to specific configurations. Third, the membrane/distillation

hybrid system can retrofit to existing distillation processes. Hence, the results are practically valuable.

Ayotte-Sauve et al. recently proposed a thermodynamic exergy method to find the minimum energy of a membrane/distillation hybrid system [9]. In this method they first used exergy analysis to determine the optimal conditions for the membrane unit, and then the minimum energy of the membrane/distillation system was found at fixed number of trays. However, this method is hard to be used here. First, the membrane properties cannot be changed because they are not involved in the optimization function. Second, at fixed number of trays the energy consumption of the distillation tower may not be the minimum. Third, the exergy analysis is valid when the process is under thermodynamic equilibrium. Practical processes always deviate from the thermodynamic equilibrium state. Hence in this work, the concept of minimum practical energy of separation is adopted for both membrane and distillation unit operations to study their performance as a function of membrane properties. From there the most important membrane parameters are identified.

The separation of propylene/propane was used in this report as a case study. This process is industrially important. The worldwide production rate of propylene was 77 million tonnes in year 2011 and the production rate is anticipated to reach 132 million tonnes by year 2025. The separation is currently achieved by distillation and the process requires more than 230 trays. The energy consumption is one of the highest in petrochemical processes [10]. To reduce energy, many alternative processes, particularly the membrane distillation hybrid system, have been widely discussed in the literature [3,11,12]. Recently, our group developed a ZIF-8 type of metal-organic framework membrane which could efficiently separate propylene from propane with selectivity more than 35 and permeance of propylene more than 3.35×10^{-8} mol/m² s Pa (100 GPU) [13]. Hence, using this system as a case study can help understand the feasibility of the ZIF-8 membrane in practical applications.

The production of propylene is mainly by steam cracking unit (SCU) or fluid catalytic cracking (FCC). These two processes account for about 89% of worldwide propylene production. The concentration of propylene produced from refinery FCC unit or ethylene production process is about 70 mol% while the applications of propylene such as production of polypropylene require high purity. In market, propylene is provided in three grades: polymer grade (> 99.5%), chemical grade (92–96%), and refinery grade (> 60%) [14]. Both propylene and propane have a high market price which mandate high products recovery. Therefore, in this study we used the following conditions for the case study: the feed composition is 50 mol%, and the separation task is to meet the chemical grade purity requirement (96 mol%) and a recovery ratio of 95%.

2. Attainability of single-stage membrane process

For a single-stage membrane process, the simplest model is to assume that both the feed stream and the permeate stream are well mixed, as shown in Fig. 2. The following relationship can be derived from the mass balance [15].

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

$$S = \frac{(\varepsilon - \eta)(\gamma - 1) - (1 - \eta)\varepsilon\gamma x_A + (\varepsilon - \eta)\varepsilon x_A}{(1 - \varepsilon x_A)[(1 - \eta)\gamma - (\varepsilon - \eta)]} \quad (2)$$

where ε is the target component enrichment ($\varepsilon = y_A/x_A$) and η is the target component recovery ratio in the permeate stream ($\eta = Gy_A/Fx_A$). Eq. (2) indicates that both selectivity and pressure

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