

Chemical potential analysis for directing the optimal design of gas membrane separation frameworks



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HIGHLIGHTS

- A thermodynamic model is proposed for mass transfer in gas membrane separation.
- Chemical potential change is used to discern work loss in RMC and CMC frameworks.
- The mix of local permeate differing in composition reduces efficiency seriously.
- Membrane stages with lower efficiency are ascertained in retrofitting frameworks.
- Efficiency is enhanced by partially retrofitting RMC with proper capital cost.

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ABSTRACT

Thermodynamic analysis can discern those energy requirements which might be decreased in the separation system. In this work a theoretical model based on nonequilibrium thermodynamics was established to analyze the free energy change and the key points governing separation efficiency in gas membrane separation. Mass transfer in membranes was treated to be a two-step process in the model: species selectively permeate across membrane to form a series of local permeate gases, then the local permeate gases converge into a bulk permeate stream. Thus the total work required by the membrane process was identified as (i) the work to drive mass permeation, (ii) the work wasted by permeate mixing, and (iii) the minimum separation work. The analysis of two typical systems – the recycle membrane cascade (RMC) and the continuous membrane column (CMC) – revealed that the work wasted by permeate mixing seriously affects the free energy efficiency. This work wastage can be reduced by retrofitting process frameworks. Subsequently, the analysis was used to identify the serious work-losing stages in the RMC. Finally, a partially retrofitted RMC was proposed with tangible reductions in both equipment investment and energy consumption.

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1. Introduction

Gas membrane separation, a promising low energy consuming technology, received extensive researches and wide application in the last 30 years. Its market has kept growing at 15% annually since 1990s (Nunes and Peinemann, 2001). The expanding market has been encouraging great developments in preparation of novel membranes (Zhang et al., 2012; Askari et al., 2012; Du et al., 2010) and design of high-efficiency processes (Baker, 2002; Pathare and Agrawal, 2010).

However, the present commercial membranes can hardly satisfy the need of high selectivity in some industries where highly concentrated products are yielded. What's more, the selectivity of the membrane modules may not be enhanced greatly in the short term

because of the limitation of corresponding materials and formation processes (Baker, 2002; Pathare and Agrawal, 2010; Robeson, 2008). For instance, the industrial polymer membrane modules generally have the value of $\alpha_{\text{CO}_2/\text{N}_2}$ approximately to 12 for CO_2 capture, and $\alpha_{\text{O}_2/\text{N}_2}$ close to 6.0 for air separation; however, membrane module selectivity should be higher than 200 for a single-stage system to capture CO_2 from coal-fired flue gases in power plant with its content up to 90 mol% and its recovery ratio up to 90% (Belaissaoui et al., 2013). Obviously, a single-stage system made of commercial modules cannot efficiently generate high concentration CO_2 , O_2 or N_2 . Furthermore, an inevitable tradeoff exists between recovery ratio and product purity even in the single-stage systems made of highly selective modules (Baker et al., 1998). Therefore, it is critical to develop appropriate membrane process frameworks to obtain products with high yield and purity (Pathare and Agrawal, 2010).

Recycle membrane cascades (RMCs) and continuous membrane columns (CMCs) are two widely known multi-stage membrane systems which can obtain products with both high purity and yield.

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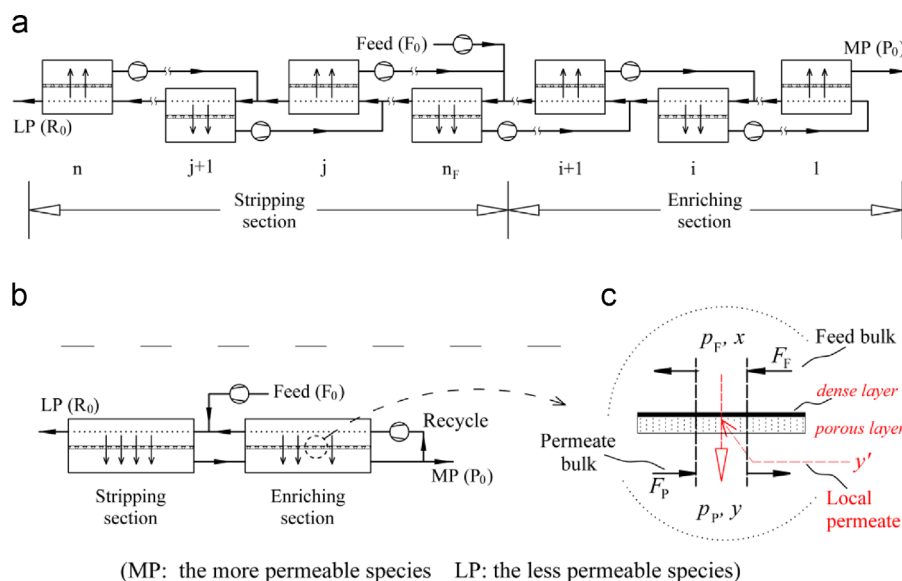


Fig. 1. Schematic diagrams of basic RMC and CMC. (a) Recycle membrane cascade (RMC), (b) continuous membrane column (CMC) and (d) micro membrane section.

The classical membrane arrangements are illustrated in Fig. 1. For a long time, the CMC has been claimed to be one of the most compact membrane processes which are able to break the tradeoff between product purity and yield (Hwang and Thorman, 1980). In the CMC, any two-component mixture can be separated into two highly concentrated products, provided that the flow rate of the recycle stream is large enough. However, the demand of both high compression power and large membrane area would lead to an expensive operation (McCandless, 1994). As a result, the CMC remains impractical so far. In contrast, the RMC can achieve a much higher energy efficiency and process capacity (McCandless, 1994). In RMC, the permeate side is specially designed as a series of independent channels. The permeate streams from each channel are separately pressurized and recycled to the high-pressure side at appropriate positions. The separation degree is improved by increasing the number of membrane stages. By virtue of high efficiency and process capacity, the derivative processes of the RMC have been recommended in many studies to yield products with both high recovery ratio and purity (McCandless, 1994; Agrawal and Xu, 1996; Agrawal, 1997; Avgidou et al., 2004; Gassner and Maréchal, 2010).

It is widely recognized that the power consumption of a multi-stage system can be reduced by increasing the number of recycle loops (Agrawal, 1997). In the light of this, membrane stages of specific substructures have been introduced into the RMC to further improve its energy efficiency (Agrawal and Xu, 1996; Agrawal, 1997). A series of systematic methods also have been proposed to design membrane cascade frameworks (Agrawal, 1997; Kookos, 2002; Marriott and Sørensen, 2003; Chang and Hou, 2006). Nevertheless, an inevitable defect of RMC and its derivatives is the multiple recycle compressors and auxiliary equipments, which may result in high capital cost and restrict the application seriously. Hence, only the schemes using fewer compressors and recycle loops have been applied in practice (Zhao et al., 2012).

In order to make a balance between the addition in capital cost and the increase in efficiency, it is necessary to optimize process framework by modifying the most inefficient membrane stages and eliminating those unwanted recycle compressors. The intrinsic factors resulting in the large difference in separation efficiency between the CMC and the RMC are very important in directing the conceptual design of membrane processes for gas separation.

Some researchers attempted to explain why the CMC and other one-compressor schemes need much larger compression duty and membrane area than the RMC does for the same separation

(McCandless, 1994; Xu and Agrawal, 1996a). Besides, some intensive simulations were made for various applications of CMC to improve its efficiency (Mercea and Hwang, 1994; Purnomo and Alpay, 2000). Our analysis around these results shows that the convergence of the local permeates with different compositions contributes largely to the poor efficiency of CMC. For example, in the case of air separation with CMC (McCandless, 1994), the local permeate gases with a molar fraction of O_2 ranging from 98.8% to 3.6% are mixed directly to yield a bulk permeate stream with an O_2 molar fraction of 95.0%. This back-mixing lowers the separation efficiency seriously. In contrast, independent channels are used in RMC to separately collect the local permeate gases by stages, so that the serious back-mixing is impaired. Thus the separation efficiency is enhanced.

To optimize membrane frameworks, qualitative studies of these potential factors are not enough. It is necessary to understand the framework characteristics, find the key factors and quantify their effects on separation efficiency. On that basis, it is desired to develop practical heuristic methods to guide the retrofit of inefficient membrane stages and the removal of unnecessary compressors.

The minimum separation work and the chemical potential loss determine the energy consumed by compressing the feeds and the recycle streams in multistage membrane systems. The minimum work cannot be decreased. Thus the only approach to improve energy efficiency is reducing the loss in chemical potential. A theoretical model is established to analyze chemical potential change in gas membrane separation systems in this paper. A comparison between the CMC and the RMC is then conducted to quantify the intrinsic factors which affect separation efficiency in the change of frameworks. The chemical potential loss induced by the non-ideal behavior of mass transfer is associated with framework characteristics. Finally, chemical potential analysis is applied to direct the conceptual design of membrane systems to achieve a balance between capital cost and energy efficiency. This is illustrated by the retrofit of the RMC for CO_2 separation.

2. Theory and modeling

The separation system studied here is a binary gas mixture. The more and the less permeable species are labeled as MP and LP, respectively. The basic element for process development is the asymmetric high-flux hollow fiber membrane module. The schematic membrane microstructure is shown in Fig. 1c. The porous supporting

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