



Pressure ratio and its impact on membrane gas separation processes



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ABSTRACT

The feed and permeate pressures achievable in most gas separations are limited by economics. As a result, commercial membrane separation processes usually operate with a feed-to-permeate pressure ratio (θ) in the range of 5–15. This limited pressure ratio means that the optimum membrane may not be the one with the highest selectivity; rather, a membrane with lower selectivity, but tailored to the pressure ratio, may result in a more economical process. In other words, when a process is pressure ratio limited, the useful selectivity is also limited. The balance between pressure ratio and selectivity is often ignored by membrane developers. The importance of this balance is illustrated with two applications of current interest: CO₂ capture from power plant flue gas, and dehydration of azeotropic bioethanol.

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1. Introduction

Membrane researchers have been aware of the trade-off between selectivity (α) and permeability (P) since the early days of membrane gas separation technology. Robeson first formulated this trade-off relationship in his influential paper [1]. You can have high selectivity or high permeability, but it is difficult to have both at the same time. Another important and less appreciated membrane parameter is the pressure ratio (θ), defined as

$$\theta = \frac{p^f}{p^p} \quad (1)$$

where p^f and p^p are the feed and permeate pressures on either side of the membrane. The impact of pressure ratio on membrane gas separation processes is the subject of this paper.

In many materials research studies, pressure ratio is ignored or treated as a freely adjustable parameter and set at a conveniently high value, where its impact on the separation can be neglected. Unfortunately, in industrial gas separation processes, adjustment of the pressure ratio comes at a significant price in terms of compressor capital cost and power usage. The cost of the compressor package may be much more than the cost of the membrane unit, and the cost of electricity used to power the compressor is usually the largest operating expense. For these reasons, pressure ratios used in industrial processes are normally below 20, and are typically in the range of 5–15.

In this paper, we will use two specific applications to illustrate the importance of pressure ratio and its impact on membrane selection.

- The first application is capture of CO₂ from power plant flue gas. Flue gas from combustion of fossil fuels is produced in enormous volumes and contains 5–15 mol% CO₂ at atmospheric pressure [2,3]. The membrane system must remove 50–90% of this CO₂ in a concentrated stream for sequestration. This is a low pressure separation, so large membrane areas are needed. But the key economic concern is the energy consumed by the compression/vacuum equipment that drives the process. This energy use is directly related to the membrane pressure ratio. A competitive process requires an affordably low pressure ratio, and this determines the preferred membrane selectivity.
- The second application is dehydration of azeotropic ethanol/water mixtures in bioethanol plants. This is the final dehydration step in the production of dry bioethanol and is a large potential market for water-permeable vapor permeation membranes [4]. The objective is to break the azeotrope produced by distillation of the ethanol fermentation broth, reducing the water content from 16 mol% (7 wt%) to 0.76 mol% (0.3 wt%). The permeate vapor is usually cooled and condensed to spontaneously create a vacuum on the permeate side of the membrane, enabling relatively high pressure ratios to be achieved. However, the extreme operating conditions (ethanol/water vapor mixtures at 100–130 °C) require expensive membrane module materials, so controlling membrane area is important. We will show that the balance between pressure ratio and selectivity also affects membrane area.

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2. Theoretical basis

Membrane gas permeation is quantified in terms of membrane permeability and selectivity using the solution-diffusion equation:

$$j_i = \frac{P_i}{\ell} (p_i^f - p_i^p) \quad (2)$$

where j_i is the molar flux ($\text{cm}^3(\text{STP})/\text{cm}^2 \text{ s}$) of component i , ℓ is the membrane thickness, p_i^f and p_i^p are the partial vapor pressures of component i on the feed side and permeate side of the membrane, and P_i is the permeability of the membrane material to component i , usually expressed in Barrer (where $1 \text{ Barrer} = 1 \times 10^{-10} \text{ cm}^3(\text{STP})/\text{cm}^2 \text{ s cmHg}$). Because the thickness of the selective layer in composite membranes is often difficult to measure, commercial membranes are typically characterized by their partial pressure normalized flux ($j_i/\Delta p_i$) or permeance (P_i/ℓ), which is expressed in gas permeation units or gpu (where $1 \text{ gpu} = 1 \times 10^{-6} \text{ cm}^3(\text{STP})/\text{cm}^2 \text{ s cmHg}$).

The membrane separation capability, or selectivity, is expressed as the ratio of the permeances or permeabilities of components i and j :

$$\alpha_{ij} = \frac{P_i/\ell}{P_j/\ell} = \frac{P_i}{P_j} \quad (3)$$

where α_{ij} is the selectivity of the membrane for component i over component j .

3. CO₂/N₂ separations

By way of illustrating the importance of pressure ratio, consider the one-stage CO₂/N₂ separation process shown in Fig. 1. For CO₂ to permeate the membrane, the partial pressure of CO₂ in the permeate must be less than the partial pressure of CO₂ in the feed, so we can write:

$$p^p \times \text{permeate CO}_2 \text{ concentration } (C_{\text{CO}_2}^p) \leq p^f \times \text{feed CO}_2 \text{ concentration } (C_{\text{CO}_2}^f) \quad (4)$$

Hence

$$\frac{C_{\text{CO}_2}^p}{C_{\text{CO}_2}^f} \leq \frac{p^f}{p^p} = \theta \quad (5)$$

or

$$\text{membrane enrichment} \leq \text{pressure ratio } (\theta) \quad (6)$$

This simple relation can also be written as

$$\frac{C_{\text{CO}_2}^p}{\theta} \leq C_{\text{CO}_2}^f \quad (7)$$

In the separation shown in Fig. 1, the feed contains 10 mol% CO₂. From Eq. (5), this means that, at a pressure ratio of 5, the permeate can contain no more than 50 mol% CO₂. Thus, the rest of the permeate must be nitrogen, the slower permeating component. It follows that the membrane area required to permeate a specific volume of CO₂ at a specific pressure ratio is determined more by the permeation rate of the slow component (nitrogen) than by that of the fast component (CO₂). In the limit that the permeation rate of the slow component drops to zero (an infinitely selective membrane), no slow component permeates, so an infinite membrane area is required.

The impact of membrane selectivity on membrane performance at a particular pressure ratio is illustrated graphically in Fig. 2; two examples are shown here, one is at pressure ratio of 5, and the other is at pressure ratio of 10. The membranes are assumed to have the same CO₂ permeance of 1000 gpu, and the selectivity is varied by changing the nitrogen permeance. The feed CO₂ concentration is assumed to be 10 mol% and the feed flow rate is 1000 m³(STP)/h. This plot and others shown later in this paper could have been calculated using analytical solutions derived by Pan and Habgood in the 1970s [5–7]. However, in this paper, these plots have been generated using differential element membrane code written at MTR and incorporated into a computer process simulation program (ChemCad 6.3, ChemStations, Austin, TX).

First consider the curves in Fig. 2 calculated at a pressure ratio of 5. The plot in Fig. 2(a) shows the permeate CO₂ concentration as a function of increasing membrane selectivity. At a selectivity of 1 (no separation), the permeate concentration is the same as the feed. As the selectivity increases, the permeate concentration increases, reaching about 40 mol% CO₂ at a selectivity of 40. As the selectivity increases further, the permeate concentration approaches the limiting value of 50 mol% given by Eq. (5). Fig. 2 (b) shows the relative membrane area required to permeate a fixed amount of CO₂ at the same conditions used in Fig. 2(a). This plot has an exponential form where the required membrane area increases rapidly as membrane selectivity increases. It follows that at a pressure ratio of 5 the optimum selectivity under the chosen conditions is between about 20 and 40. A higher selectivity produces a slight increase in permeate CO₂ concentration, but at the expense of a very large increase in membrane area (and cost).

Consider now the curves in Fig. 2 calculated at a pressure ratio of 10. As the membrane selectivity increases, the permeate CO₂ concentration increases approaching a limiting value of 100 mol% given by Eq. (5). As before, the membrane area increases with increasing selectivity. Comparing the curves for the two pressure ratios, it is clear that increasing the pressure ratio, if economically possible, has a very beneficial effect: at the same selectivity and CO₂ permeance, a smaller membrane area is needed to produce the same amount of permeate, which has a higher concentration of CO₂. However, even at

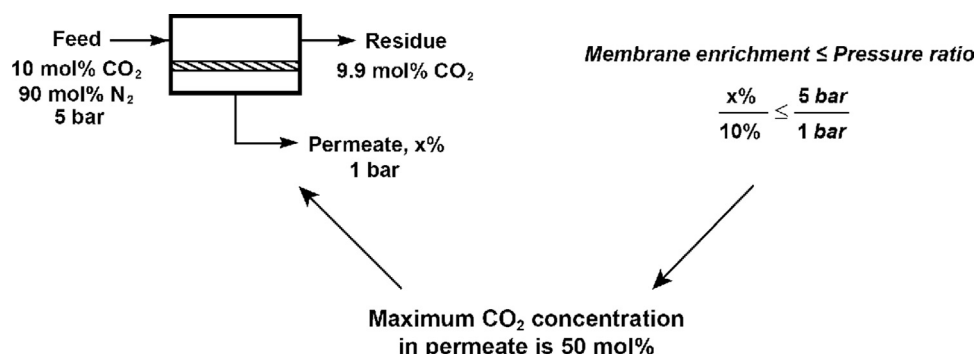


Fig. 1. Separation of CO₂ from nitrogen by a CO₂-selective membrane at a pressure ratio of 5 (5 bar feed, 1 bar permeate). The permeate can never contain more than 50 mol% CO₂, even when the membrane is infinitely selective.

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