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Analysis of permeate pressure build-up effects on separation performance of asymmetric hollow fiber membranes

Prodip Kundu, Rabitah Zakaria [1], Amit Chakma, Xianshe Feng *

Department of Chemical Engineering, University of Waterloo, 200 University Avenue West, Waterloo, Ontario, Canada N2L 3G1

HIGHLIGHTS

- More permeance underestimation at lower feed pressures.
- Impact of pressure build-up on membrane performance is actually lower at higher pressures.
- Membrane performs close to its full potential if operated at high feed pressures.
- Percentage loss in driving force is less at higher feed pressures.

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ABSTRACT

The study rectifies some perceptions about pressure build-up in hollow fiber membranes. It is a general intuition that operating at higher pressures permeates more gases, and sometimes the membrane module is tested or characterized at lower pressures to reduce gas consumption. It is also perceived that higher pressure build-up occurs at higher feed pressures, and membrane performance deteriorates at higher feed pressures. In this study, the apparent and intrinsic permeances of H_2 and N_2 in asymmetric cellulose acetate-based hollow fiber membranes were evaluated from gas permeation experiments and numerical analysis. It was shown that though the permeate pressure build-up increases as the feed pressure increases, the effect of the permeate pressure build-up on the membrane performance is actually reduced at higher feed pressures. Membrane performs close to its intrinsic separation properties if it is operated at high feed pressures, under which conditions the effect of pressure build-up on the membrane performance is minimized. The pressure build-up effect was further investigated by evaluating the percentage loss in the driving force due to permeate pressure build-up, and it was found that percentage loss in driving force is less at higher feed pressures than that at lower feed pressures.

1. Introduction

An important advantage of hollow fiber membrane modules is their ability to pack a very large membrane area into a single module. Hollow fibers enable substantially higher membrane packing than are possible with either of the flat-sheet or spiral-wound designs. For high-pressure gas separation applications, hollow fine fibers have a major segment of the market, and can withstand high-pressures of 1000 psig or more in shell-side feed modules (Baker, 2004). Operating at a higher pressure consumes more gas, and therefore sometimes membrane modules are tested or characterized at lower pressures to reduce gas consumption. There is a permeate pressure build-up in the lumen side for shell feed applications and can become enough to seriously affect the membrane performance. It is also commonly believed that there is little or negligible pressure build-up for short fibers, though the pressure build-up increases for longer fibers. It is thus often perceived that the effect of the pressure build-up is significant only for long fibers, especially at high feed pressures. This, however requires rectification and interpretation, and these perceptions are sometimes misleading.

Gas permeability in glassy polymer membranes may be described by the dual-mode model especially for condensable gases and vapors. However, the permeability of permanent gases in glassy polymers is normally not a strong function of gas pressure, and pressure-independent permeability is often assumed in process design and analysis. The hollow fiber membranes used in gas separation are often very fine. For shell feed configuration, the permeate pressure build-up on the lumen side of these fine fibers reduces the driving force across the fiber wall available for permeation, and this loss can become enough to seriously affect the membrane performance. Because of the pressure build-up in the fiber lumen, the permeances calculated

* Corresponding author. Tel.: +1 519 888 4567; fax: +1 519 746 4979.
E-mail address: xfeng@uwaterloo.ca (X. Feng).
[1] Current address: Fakulti Kejuruteraan Proses dan Makanan, University Putra Malaysia.

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from pure gas permeation experiments (apparent permeances) based on permeate outlet pressure do not necessarily represent the true permeances (intrinsic property) of the membrane unless the effect of the permeate pressure build-up in the fiber lumen is insignificant. A number of examples can be cited from the open literature where gas permeance was found independent of pressure difference for flat homogeneous membranes but a dependent function for hollow fibers made from the same material. The permeate pressure build-up is significant for long fibers, fine fibers and for high pressure applications. On the other hand, to minimize gas consumption, lab tests are often performed at low feed pressures on miniature fiber modules with short fibers (Pan, 1986). However, the percentage loss in the driving force at low pressures can still be enough to deteriorate the actual permeation performance. On the contrary, the reduction in the driving force at high gas pressures is in fact less critical as the membrane permeance will be closer to its intrinsic permeance at a higher pressure. Further, the permeate pressure build-up is not always negligible for short fibers. The hollow fiber length is usually more than 1 m for industrial applications and a considerable pressure build-up in the lumen can occur, resulting in a discrepancy in the measured membrane performance if this effect is neglected. For the separation of hydrogen from nitrogen or methane, or carbon dioxide from natural gas, the permeation fluxes are considerably high, and hollow fiber modules can develop excessive permeate-side pressure drops. On the other hand, for the production of nitrogen from air, the gas permeance is relatively low, from 1 to 2 GPU, and pressure drops are not likely to be a big problem (Baker, 2004). However, depending on the module geometry, the effect of permeate pressure build-up could be significant for slow permeating gases as well. Moreover, the potted section of the hollow fibers does not take part in permeation but contributes additional pressure drop, and this section adds up driving force loss which can influence the apparent permeance calculated from the permeate flow.

The permeate pressure build-up is more significant for species with a higher permeability than that of the lower permeability species. Thus, we opted for H_2 and N_2 to investigate the effect of permeate pressure build-up in this study because H_2 and N_2 are representative fast and slow gases, respectively, and H_2/N_2 separation by hollow fiber membranes is an important application for H_2 recovery from ammonia synthesis purge gas. In this study, the apparent permeances of H_2 and N_2 were evaluated from gas permeation experiments with short and long fibers. The true separation performance of the hollow fiber membranes was calculated by taking into account the effect of permeate pressure build-up in the fiber lumen (including the potted section). A correlation between the apparent membrane permeance and pressure difference was observed. The observations were explained and the trends were compared with some experimental results reported in the literature. The permeate pressure build-up effect was further investigated by evaluating the percentage loss in the driving force due to the pressure build-up inside the fiber lumen for pure gas permeation of H_2 at different pressures. The gas permeances for a gas mixture permeation were also measured from experiments, and were compared with permeances determined from pure gas permeation experiments. The study rectifies some general perceptions about the pressure build-up with high flux asymmetric hollow fiber membranes. The analysis may also provide an insight into the characteristics of gas permeation through hollow fiber membranes.

2. Materials and methods

2.1. Gas permeation experiment

A short membrane module was prepared by inserting a bundle of 23 asymmetric cellulose acetate-based hollow fibers in a 1/4-in copper tubing. The potted section was about 4 cm in length, and this section was potted with epoxy resin to form a gas-tight tube sheet as shown in Fig. 1. The tube sheet was carefully cut to make the fibers fully open for gas flow. The effective length of the fiber was 23.5 cm, and the outer diameter of the fiber was 200 μm. Pure gas permeation of H_2 and N_2 was conducted (Fig. 2). The feed gas at a specific pressure was admitted to the membrane module to contact with the selective layer of the membrane, and the permeate stream exited the module from the lumen side at atmospheric pressure. The residue end was completely closed. The permeation rate was determined volumetrically using a bubble flow meter. The gas permeance through the membrane can be calculated by

$$J = Q/A\,\Delta p \tag{1}$$

where J is the apparent membrane permeance, which is customarily expressed in units of GPU ($1\ \text{GPU} = 10^{-6}\ \text{cm}^3\ (\text{STP})/\text{cm}^2\ \text{s cm Hg} = 3.35 \times 10^{-10}\ \text{mol/m}^2\ \text{s Pa}$), A the effective membrane area for permeation (excluding potted section) in cm^2, Q gas permeation rate in cm^3 (STP)/s, and Δp the pressure difference across the membrane in cmHg. The permeation selectivity is characterized by the permeance ratio of a pair of gases. A longer membrane module, consisting of 9 fibers with 105 cm in length, was also prepared to determine the effect of pressure build-up on the apparent permeance and the selectivity of the membrane module. Another module, consisting 9 fibers and 87 cm in length, was prepared for mixed gas permeation experiment. For the mixed gas permeation experiments, the gas mixture was prepared in-house by blending hydrogen and nitrogen until the required composition was achieved. The feed gas contains 57.39 mol% hydrogen and the

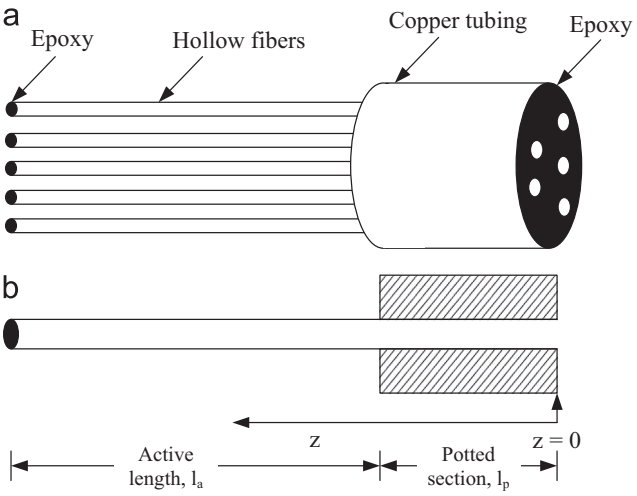

Fig. 1. (a) Schematic of assembled hollow fiber membranes, and (b) active and potted section.

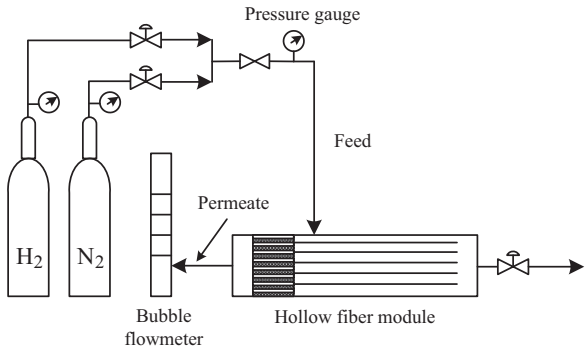

Fig. 2. Apparatus of gas permeation experiment.