Contents lists available at ScienceDirect

Chemical Engineering Research and Design

journal homepage: www.elsevier.com/locate/cherd

Unsteady state cyclic pressure-vacuum swing permeation for low pressure niche gas separation applications

Prodip K. Kundu[1], *Amit Chakma*, *Xianshe Feng*[*]

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

ARTICLE INFO

Article history:
Received 12 October 2015
Received in revised form 19 January 2016
Accepted 24 February 2016
Available online 4 March 2016

Keywords:
Gas separation
Membranes
Membrane process
Permeation
Cyclic operation

ABSTRACT

A relatively higher transmembrane driving force compared to conventional steady state membrane process can be achieved in unsteady state cyclic pressure-vacuum swing permeation process, which accomplishes feed pressurization and permeate evacuation using a single pump. Moreover, a higher feed to permeate pressure ratio can be achieved and the enhanced transmembrane driving force can also be sustained over a longer period of time, which enhances gas separations. Improved separation efficiency in terms of product purity and throughput compared to the conventional steady state membrane gas separation process can be obtained for low pressure niche applications by means of pressure-vacuum swing permeation. CO_2 separation for greenhouse gas emission control, oxygen-enriched air production and methane enrichment from biogas are shown as exemplary model applications to demonstrate the feasibility and effectiveness of this novel process.

1. Introduction

In steady state conventional membrane gas separation process, the permeation rate and permeate concentration do not change with time except at the initial start-up stage as the feed and the permeate pressure are kept constant (Wang et al., 2011). Steady state permeation offers such advantages as easy start-up and shout-down, simplicity of pressure and flow controls, large throughput of permeation, and low maintenance requirements. Elevating the pressure differential across the membrane enhances permeation. The pressure differential across the membrane can be enhanced by feed pressurization and/or permeate evacuation. However, simultaneous use of a compressor and a vacuum pump to increase the transmembrane pressure difference is normally not considered rewarding, especially when "deep" vacuum is applied to suck the permeate from the permeate side.

Membrane gas separations under transient state conditions are rather unexplored. Enhanced separation as compared to steady state operation was reported using unsteady state permeation (Paul, 1971; Higuchi and Nakagawa, 1989; Beckman et al., 1991). According to the solution-diffusion mechanism, the permeability coefficient of a gas is the product of its diffusion coefficient and solubility coefficient (Nunes and Peinemann, 2006). Unfortunately, a trade-off relationship between the permeability and selectivity often exists for most membrane materials. An upper bound appears in permeability vs. selectivity plot above which virtually no or little data exist (Robeson, 1991). This suggests that once the membrane material is fixed, the selectivity characteristics is essentially fixed if a steady state operation is used. However, transient and, in particular, steady cyclic operation of the membrane can be used to alter the selectivity characteristics. The first theoretical study of membrane separation process operating

---

[*] *Corresponding author*. Tel.: +1 519 888 4567; fax: +1 519 888 4347.
E-mail addresses: prodip.kundu@olisystems.com (P.K. Kundu), xfeng@uwaterloo.ca (X. Feng).

[1] Present address: OLI Systems Inc., 240 Cedar Knolls Road, Suite 301, Cedar Knolls, NJ 07927, USA.

http://dx.doi.org/10.1016/j.cherd.2016.02.033
0263-8762/

Nomenclature

A	membrane area [m^2]
J	permeance [mol/m^2 s Pa]
p	permeate side pressure [kPa]
p_V	permeate pressure when the permeate side is evacuated [kPa]
P	feed side pressure [kPa]
Q	quantity of permeate [mol]
t	permeation time [s]
T	temperature [K]
V_F	volume occupied by feed [m^3]
V_M	volume occupied by permeate [m^3]
X	mole fraction of the most permeable component in retentate [–]
Y_M	bulk permeate mole fraction [–]
V1–V5	valve in operation [–]

Subscripts

1–5	different steps
A	component A
B	component B
F	feed side

in a cyclic transient fashion was performed in 1971 to improve the separation performance when the mobility selectivity is significantly greater than permselectivity (Paul, 1971). Higuchi and Nakagawa (1989) theoretically studied transient permeation with pulsed upstream pressures as described by Paul (1971) for air separation to illustrate the improvement in selectivity over steady state operation. Beckman et al. (1991, 1993) carried out a dynamic process with intermittent feed admission on a continuous basis to exemplify the pulse feeding process for He/CO_2 separation. The permeate and residue were removed periodically and they were synchronized with the feed admission sequence. Corriou et al. (2008) optimized Paul's (1971) mode of operation and claimed that synchronous operation would offer the best performance. Corriou et al. (2008) also reported that cyclic operation could potentially compete with the most selective polymers available at the time, both in terms of selectivity and productivity. Kao et al. (1991) reported a pressure swing scheme similar to pressure swing adsorption to carry out transient permeation where the opposite solubility and diffusivity selectivities were exploited synergistically. LaPack and Dupuis (1994) modified the process to exploit the differences in the rates of either attainment of the steady state permeation or fall-off from the steady state permeation. Ueda et al. (1990) described a cyclic process where feed pressurization and permeate evacuation were completed with a compressor and a vacuum pump, respectively (or with a single pump suitable for both feed pressurization and permeate suction synchronously). Bowser (2004) and Nemser (2005) extended the application of the cyclic process for controlling emissions of volatile organic compounds (VOC) from solvent storage.

Feng et al. (2000) reported a novel process for gas separation called pressure swing permeation, and a bench-scale unit was tested for H_2/N_2 separation to demonstrate the effectiveness of the process. High pressure feed gas was "pushed" periodically on the low pressure permeate side, thereby elevating the permeate pressure close to (or as high as) the feed pressure, which was otherwise impossible to achieve with the conventional steady state permeation. Feng and Lawless (2015) recently received a patent on novel unsteady state pressure-vacuum swing process which accomplishes feed gas pressurization and permeate evacuation alternately with a single pump, thereby enhancing the transmembrane driving force for permeation. The feed and the permeate stream pressures were elevated and lowered, respectively, but not at the same time, using the same pump in a dynamic cyclic fashion without the need of operating two pressure changers (i.e., a compressor for feed pressurization and a vacuum pump for permeate evacuation) simultaneously. The process does not alter permselectivity of the membrane materials, and the enhanced separation is due to (1) the increase in the transmembrane pressure differential for permeation, and (2) the increase in the feed to permeate pressure ratio. The technical feasibility of such an unsteady state pressure-swing permeation process for gas separation has been confirmed experimentally using an automated demonstration unit equipped with a Parker membrane module at the research facility of Monteco Ltd. for oxygen enrichment of air as reported in our earlier study (Chen et al., 2014). The mathematical model formulation, model assumptions, and solution technique have been presented elsewhere (Kundu, 2013; Chen et al., 2014), and the current study is limited to the applications and relative advantages over traditional steady state processes. Work is currently underway for pilot scale tests and economic feasibility evaluations. The current study reports some of the theoretical aspects of that novel dynamic process and exemplifies applications where the cyclic process is best suited. A parametric analysis was conducted to evaluate the effects of critical design and operating parameters on the separation performance. The pressure–vacuum swing permeation appears to be best suited to membrane gas separations that are normally carried out at a relatively low feed pressure. Separation of carbon dioxide from flue gas, oxygen-enriched air production and biogas upgrading could be the potential niche applications for pressure swing permeation. The extent of separation that can be achieved in pressure swing permeation in comparison with the traditional membrane gas separation are presented to exemplify the feasibility and effectiveness of this process to aforementioned applications.

2. Steps of a single pump driven cyclic pressure-vacuum swing permeation

As illustrated in Fig. 1, the pressure-vacuum swing permeation undergoes feed gas pressurization, permeate side evacuation, and residue side venting sequentially. A cycle consists of five operating steps, and the schematic of pressure variations (on the feed and permeate sides) with time are shown in Fig. 2.

Step 1: Step 1 (time t_0 to t_1) is referred to as the "feed pressurization" step. A diaphragm pump (or other commonly used positive displacement pump) can be used to quickly charge the feed side (volume V_F) with a pressurized gas to reach a pressure of $P_1 = P_h$. The pump functions as a vacuum pump alternately during Step 3. Permeation begins to occur under the transmembrane pressure difference. V_M is the volume on the permeate side. Open valves V1, V2; closed valves V3, V4, V5.

Step 2: Step 2 (time t_1 to t_2) is referred to as the "feed admission/permeation" step. In this step, a constant pressure P_h on the feed side is maintained by charging feed continuously.