Separation and Purification Technology 205 (2018) 251–262

Contents lists available at ScienceDirect

Separation and Purification Technology

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

Carbon membranes for oxygen enriched air – Part II: Techno-economic analysis

Shamim Haider, Arne Lindbråthen, Jon Arvid Lie, May-Britt Hägg*

Norwegian University of Science and Technology, NTNU, Department of Chemical Engineering, 7491 Trondheim, Norway

ARTICLE INFO

Keywords:
Oxygen enriched air
Carbon membrane
Simulations
Techno-economic analysis

ABSTRACT

Carbon membrane (CM) separation process for producing oxygen-enriched air (OEA) at a concentration of 50–78 mol% O_2 in a single stage process with no recycle stream has been investigated. This paper (Part II of a two-part study) considers techno-economic analysis for O_2-selective carbon membranes to yield the lowest production cost of "equivalent" pure oxygen (EPO_2) in a single stage separation process based on experimental and predictive membrane performance. Aspen Hysys® interfaced with ChemBrane (in-house developed model) was used to perform the simulations for air separation with CM. Three different approaches with respect to pressure were investigated; (1) feed compression, (2) vacuum on permeate side and (3) combination of (1) and (2). The simulation results and sensitivity analysis showed that with current performance (O_2 permeability: 10 Barrer (*1 Barrer = 2.736E − 09* $m^3(STP)m/(m^2\ bar\ h)$) and O_2/N_2 selectivity: 18), mechanical properties, and cost per m^2 of CM, it is economically most efficient to use the third approach "combination of feed compression and permeate vacuum" to produce EPO_2. A stage cut of 10% was found to be as an average economical optimum when using vacuum pump (approach (2)) to produce OEA. However, the techno-economic analysis for the reported CM showed that a stage cut of 0.15–0.2 was the most cost-effective while using compression approach (1) or (3) to produce EPO_2.

1. Introduction

Cryogenic distillation is the most common technology to produce high purity oxygen (> 99%) at large scale productions (100–300 tons/day). Pressure swing adsorption (PSA) can reach up to 95% oxygen purity and the requirement of sorbents limits the size capacity for small to medium scale plant (20–100 tons/day), mainly due to high capital cost. However, both cryogenic and PSA are considered as energy intensive technologies [1,2]. Therefore, energy efficient methods with low capital investment are required to separate the air into oxygen (as enriched air or pure oxygen) and nitrogen.

Membrane separation is an attractive process alternative due to its simple design, lower energy demand, smaller footprint, good weight efficiency (light weight equipment compared to other technologies), and flexible, modular design. However, compared to conventional technologies commercially available polymeric membranes can not economically produce high purity of O_2. The performance of polymeric membranes is restricted by the trade-off between permeability and selectivity [3]. Polymeric materials may have high permeability for O_2, but rather low O_2/N_2 selectivity (usual range 2–8), and the maximum permeate purity achievable for O_2 with polymeric membranes seems to be 30–60% [4]. Nitrogen of purity up to 99.5% can be produced using polyimide membranes (O_2/N_2 selectivity of 9) [3,5]. Nevertheless, a multi stage separation process with recycle stream is required to achieve high recovery of N_2 which would add more cost and complexity to the system.

High purity oxygen is difficult using membranes because of the high content of nitrogen in the air, (79%) and the relative low selectivity of O_2/N_2 resulting in oxygen enriched air (OEA) in permeate, rather than pure oxygen. Based on the performance of commercially available polymeric membranes, the separation process is competitive only for medium O_2 purity (25–40%) and small-scale plants (10–25 tons/day) [4,6]. OEA is already used for numerous chemical processes (Claus process, the Fluid Catalytic Cracking technology, the oxidation of *p*-xylene to give terephthalic acid) combustion processes (natural gas furnaces, coal gasification), medical purposes, and has more recently also attracted attention for hybrid carbon capture process [7].

Carbon is a class of material that can offer improved performance due to molecular sieving effect. In molecular sieving, the available pore size is below the kinetic diameter of one of the gas components in the feed. This characteristic of the material increases selectivity by reducing the rotational degrees of freedom of nitrogen versus oxygen in the

* Corresponding author.
E-mail address: may-britt.hagg@ntnu.no (M.-B. Hägg).

https://doi.org/10.1016/j.seppur.2018.05.037
Received 23 April 2018; Received in revised form 13 May 2018; Accepted 13 May 2018
1383-5866/

Nomenclature

List of symbols and abbreviations

$Q_{f,i}$	molar flow of i in the feed (mol/s)
P_i	permeance for i ($m^3(STP)m/m^2$ *bar h*)
P_f	feed pressure (bar)
P_p	permeate pressure (bar)
x_{if}	molar fraction of i in the feed side
y_{ip}	molar fraction of i in the permeate side
P	permeance ($m^3(STP)m/m^2$ *bar h*)
A	membrane area (m^2)
EPO_2	equivalent pure oxygen
OEA	oxygen enriched air
n	molar flow rate (moles/day)
T	mass flow rate (tons/day)
CM	carbon membranes
CMC	carbon membrane cost
CC	compressor cost (installed)
VC	vacuum pump cost (installed)
PC	production cost of EPO_2
MRC	membrane replacement cost
EC	electricity cost
CRC	capital recovery cost
LC	labor cost
TLC	total labor cost
VP	vacuum on permeate side
FC	feed compression approach
FC-VP	combined feed compression and vacuum on permeate side

Greek symbols

θ	stage cut (q_p/q_f)
α	membrane selectivity (O_2/N_2)

diffusion (kinetic) transition state. In addition, carbon membranes (CM) offer superior thermal resistance and chemical stability in corrosive environments [8]. Many prior studies have reported higher selectivity and permeability of CM compared to polymeric membranes for air separation [9–13].

In 1991, Bhide and Stern calculated the membrane performance required to produce OEA at a cost competitive to cryogenically produced oxygen at \$ 40–60/ton of equivalent pure oxygen (EPO_2) [4]. They showed that none of the today's polymers can reach the \$ 40–60/ton EPO_2 target. To reduce the capital and production cost (PC) of the membrane-based process, both selectivity and permeability must be improved. Higher O_2/N_2 selectivity is required to reach the high purity of O_2 with a lower driving force (partial pressure ratio) hence, the operating cost will be reduced. A higher permeability of O_2 will cut the required membrane area for the separation, therefore, low capital investment is needed. Much academic research is focused on producing highly selective membranes, but if the membranes then have too low permeabilities they are most likely not an optimum choice for the application in focus. The carbon membranes reported here were experimentally documented to have a high selectivity 18 for O_2/N_2 and the permeability of O_2 was increasing exponentially with increase in operating temperature without significant loss in the selectivity [14]. Hence it was found that the separation process with these carbon membranes may be optimized to achieve high purity O_2 with reasonable capital investment and production cost. Predicting the cost of carbon membrane modules is difficult because of the lack of commercial precedent. Based on pilot scale production cost of regenerated cellulose-based CM, this paper focuses on the techno-economical analysis of CM-based air separation process to investigate the viability of CM in OEA market.

In order to obtain OEA economically with polymeric membranes, feed compression is not considered a viable solution due to the high energy cost. Some studies have concluded that applying vacuum on permeate side corresponds to the lowest energy requirement whatever the membrane stage cut is ($\theta = q_p/q_{f,}$ defined as the ratio of permeate flow rate to feed flow rate), because only the permeate stream has to be processed which is a small portion of the feed [4,7]. However, in this study we will compare the three compression approaches; feed compression, permeate vacuum, and a combination of both feed compression and permeate vacuum. The simulation results and sensitivity analysis show that with the present performance, mechanical properties, and cost per m^2 of CM, it is more economical to use a combination of feed compression and permeate vacuum for small scale OEA production.

This study is comprised of two parts. Part I of this study [14] describes laboratory testing of carbon membranes for air separation and regeneration techniques (thermal, chemical and electrical) to achieve a stable performance of the membrane. The CM was shown to exhibit single gas O_2/N_2 selectivity of 18 and O_2 permeability of 10 Barrer *(1 Barrer = 2.736E − 09* m^3*(STP)m/(*m^2 *bar h))* at 68 °C. The permeability of O_2 was increasing exponentially with increase in operating temperature without significant loss in the selectivity. Part II of this study (discussed here) examines the economic viability of carbon membranes in the air separation market. Aspen Hysys® interfaced with Chembrane, an in-house built membrane model was used for the simulations. Single stage configuration with CMS membrane was optimized to attain a simple process with minimum cost to produce OEA. The separation properties of the prepared CM were predicted to achieve high O_2 permeability, between100 and 300 Barrer, by keeping the selectivity constant. It has been documented that the assumption on the higher O_2 permeability without sacrificing the O_2/N_2 selectivity, may be achieved by adding the nano particles in the precursor [15,16] or operating the membranes at elevated temperature. The separation process was optimized with respect to installed energy and membrane area to achieve low production cost and total capital investment (TCI). The sensitivity of the process towards membrane area, energy, membrane life time, and membrane cost was investigated in the current study.

Simulation results indicated that these membranes may produce 78% O_2-enriched permeate stream and at the same time obtain 15% O_2 (hypoxic) retentate stream in a single-stage process when using combination of feed compression and vacuum on permeate side. Although retentate stream usage is not considered in the economic calculations, the retentate stream may be used as hypoxic air (Air containing 15 vol% O_2 is named as hypoxic air, and over the last years use of the hypoxic air has increased in venting system to reduce the fire hazards. Further, in multifunctional buildings, electrical appliance rooms and computer rooms use of hypoxic air have been found to be essential to societal important functions [17].

2. Background on membrane model and process simulations

Chembrane, an in-housed membrane model, based on mass transfer equations for co-current, counter current, and a perfectly-mixed flow configuration, was interfaced with Aspen Hysys® V9. The thermodynamic fluid package that uses sour Peng-Robinson equation of state was used to perform all the simulations for air separation with CM. For a shell fed module, based on MemfoACT AS module design [18], the counter-current configuration explains real behavior of gas flow as the best. Therefore, counter-current configuration was also used in the current study. However, other configurations and details of the model can be found elsewhere [19].

A representation of membrane module counter-current