Journal of Applied Sciences

ISSN 1812-5654

Journal of Applied Sciences 14 (12): 1309-1313, 2014
ISSN 1812-5654 / DOI: 10.3923/jas.2014.1309.1313

Effects of Operating Pressure and Temperature on the Oxygen Diffusion through Hollow Fiber Ceramic Membrane

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Abstract: Dense ceramic membrane has become a major selection for its potential to give high oxygen enrichment from surrounding air. The separation through dense ceramic membrane has overcome several limitations of polymeric membrane material and therefore it is capable of performing efficient means of oxygen production. Experiments were carried out to study the potential of ceramic membrane for oxygen system on oxygen/nitrogen mixtures, where the effects of operating pressure and temperature were observed and discussed. The oxygen concentration in the permeate stream increases when the operating pressure increased for each set of air flow rate of 20, 30 and 40 LPM. The permeate oxygen concentration at 40 LPM and 25 psi was found to be 5.30 ppm where as at 20 LPM and 150°C and was 5.13 ppm.

Key words: Air separation, hollow fiber ceramic membrane, flux, permeate, oxygen concentration

INTRODUCTION

Pervaporation is the most promising technology in the molecular-scale liquid/liquid separation existing in industry for being highly selective, economical, safe and eco-friendly (Jiang *et al.*, 2009). In pervaporation, the chemical potential gradient results from a vapour pressure difference across the membrane is the driving force for mass transfer. The permeate side is kept under high vacuum in order to maximize the driving force for mass transport across the membrane (Ribeiro and Borges, 2004). Maximum driving force is needed since almost all the permeating component has to get through the membrane to give pure product purity.

Based on similar separation mechanism, reverse pervaporation is only the opposite phase change of the pervaporation. The concept study of this project is to separate the gaseous component of the air through the most suitable membrane into a liquid phase. The separation through the membrane is solution diffusion mechanism since the difference between less oxygen concentration in feed inlet and the high oxygen concentration in the atmosphere applied. Extensive research has been done by many experts to develop the existing membrane for gas separation. Many types of membrane material to be used in membrane based separation process are available but their limitation in terms of selectivity is the limiting factor to give an effective separation process. In this perspective, dense ceramic membrane with porous support is chosen since it has been recognized to have enormous potential to be applied broadly in the gas separation industry particularly in air separation process. Therefore, this project will allow determining the potential use of membrane used as compared to the other type of membrane and thus study the effect of feed pressure and temperature to the reverse pervaporation process.

The feed pressure must be higher than the permeate pressure, otherwise no separation would occur. The partial pressure difference will provide the driving force for the separation. The driving force and flux will be reduced if the permeate side applied with high partial pressure. Besides that, if the porosity of the sublayer is too small, it can results in high pressure loss on the permeate side thus increase the tendency for capillary condensation to occur. Therefore, the selectivity of membrane is very important factor indeed (Li, 2007). The diffusion routes involves from high to a lower chemical potential with respect to the concentration. Vapour permeation and pervapouration are among the processes which make use of concentration difference as the driving force. The solution diffusion mechanism for gas separation through non-porous membrane determines by the permeabilities of various gases, as well as the selectivity.

The performance of oxygen through dense membrane requires high pressure of air to obtain high oxygen permeation flux. Although, thinner membrane results in

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increasing of oxygen permeation flux, however the mechanical strength of the thinner perovskite/dense membrane will be very poor. As a matter of fact, an increasing in pressure will results in higher flux. Therefore, the rate permeation of oxygen through the membrane will be higher when high pressure is applied because the oxygen from the atmosphere can permeate easily through the membrane to the feed with low concentration of dissolved oxygen in water (Hashim *et al*., 2010; Krishnaiah *et al*., 2011, 2013).

MATERIALS AND METHODS

Preparation of deoxygenated water: One liters of distilled water was put into a conical flask. Nitrogen gas tank head tube was immersed into the distilled water in conical flask. Oxygen probe also immersed into the distilled water and initial reading of dissolved oxygen was taken. The control valve of nitrogen gas tank head was opened slowly to relief its gas safely and nitrogen gas was let passed through uniformly into the distilled water in the conical flask. The oxygen probe was used to measure the dissolved oxygen concentration. The control valve of nitrogen gas tank was closed when oxygen probe show the reading of dissolved oxygen concentration in distilled water are lower than 1 ppm. Then the prepared distilled water was tightly closed as fast as can before transfer to feed tank to prevent oxygen from atmosphere dissolves into the prepared distilled water.

Experimental setup and procedure: Experimental set-up as shown in Fig. 1 consists of a feed tank, pump, oxygen detector/probe, water bath machine, control valve and hollow fibre ceramic membrane column. Every connection was taped by Teflon tape to ensure tight to avoid unwanted source of oxygen to dissolve into the water and to prevent any leakage from happen. The adjustable flow rate pump was used in this experiment. The water and oil baths machine were used to control the temperature of deoxygenated distilled water. The experiment for effects of operating pressure is carried out with constant temperature (40°C), three different air flow rate and 60% pumping rate. The operating pressure change is controlled from the retentate stream, simply by controlling the valve, each at 5, 10, 15, 20 and 25 psi. For the effects of operating temperature, the pressure is kept constant at 10 psi, 60% pumping rate and at three different air flow rate. The temperature is set at 40, 80, 120 and 150°C. Before the experiment starts, the hot air system is pre-run for 2 h to ensure the piping are heated and thus reduce the heat loss during the experiment. The prepared distilled water was ensured less than 1 ppm (mg L^{-1}) dissolve oxygen before run the experiment.

In this experiment, the prepared deoxygenated distilled water has a 0.80 ppm oxygen concentration. The prepared distilled water from the feed tank was pumped into the ceramic membrane column by flow pump regulator. The speed of the pump depends on the speed set manually on it. The pump rate used in the experiment is set constant at 60%. The concentration of dissolved oxygen of product collected is measured using the Dissolved Oxygen Meter at a different feed and hot air flow rate. The oxygen concentration in the permeate stream is recorded in every ten, 10-300 min period. Each experiment for different operating parameters is repeated at different feed flow rates to ensure inaccuracy of measurement. The air flow rates used are changed from 20, 30 and 40 L min^{-1} (LPM). The air supplied from the compressor will pass through a heating coil bathed in the water and oil baths for heating purpose. The temperature of the air is regulated by the temperature of the oil and water baths.

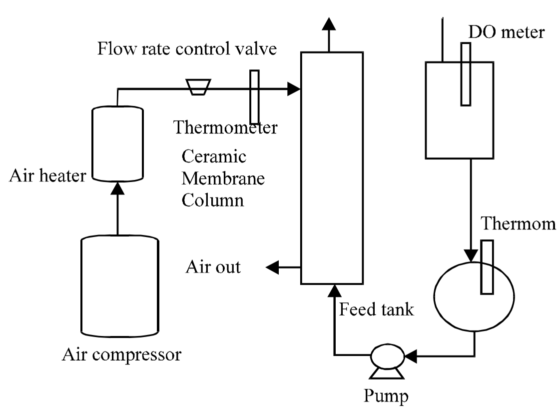

Fig. 1: Equipments set up for lab scale oxygen enrichment system

Calculation of flux: The oxygen flux is:

$$F = \frac{Cf\text{-}Ci}{A\ mg\ cm^{-2}\ sec}$$

where, F is flow rate of water, C_f is final concentration, C_i is initial concentration, A is surface area of membrane. The weight of oxygen, mg was converted into volume by gas law.

RESULTS AND DISCUSSION

Effects of pressure: The experiment was conducted to observe the effect of operating pressure to oxygen permeation. The permeation period was taken for 300 min

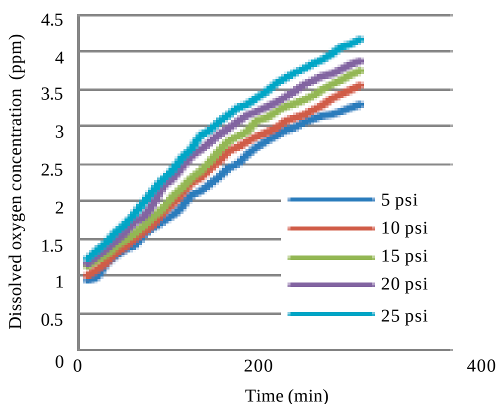

Fig. 2: Effect of operating pressure on oxygen concentration at 20 LPM air flow rate

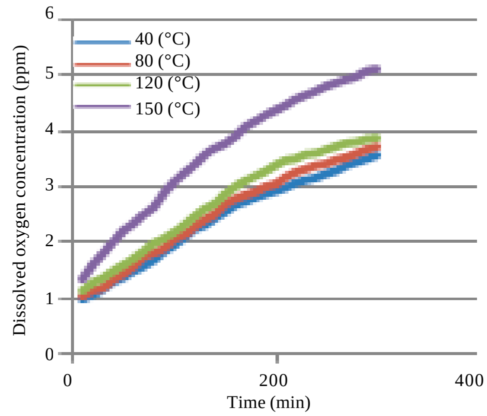

Fig. 3: Effect of temperature on oxygen concentrations at 20 LPM air flow rate

with constant temperature of 40°C and 60% pumping rate. The data recorded was used to plot dissolved oxygen concentration against permeation period.

Figure 2 shows a typical plot of the dissolved oxygen concentration in permeates stream and its dependence on feed pressure. It is evident from the Fig. 2 that the dissolved oxygen concentration in the permeate stream increases as the pressure increased from 5, 10, 15, 20 and 25 psi.

It was found from the experimental result that the oxygen concentration is more prominent at higher operating pressure and feed flow rates. At a feed flow rate of 20 LPM, the permeate oxygen concentration increased from 3.42 to 4.07 ppm when the pressure was increased from 5 to 25 psi in a period of 300 min. When the feed flow rate is increased to 40 LPM, the same pressure change resulted in a permeate oxygen concentration from 4.1-5.3 ppm. As the feed pressure is increased, the driving force increases as well because the feed has a higher pressure than the permeate stream, resulted in the increase of the pressure difference across the membrane and causes the diffusion of larger amount of gas through the membrane (Ismail *et al*., 2001). The partial pressure of a component at the feed side of a pervaporation membrane has always to be higher than that on the permeate side, otherwise no transport will occur and this condition also applied in reverse pervaporation. As presented in the graph, at a pressure of 20 psi and 20 LPM of feed flow rate, the dissolved oxygen concentration obtained after 300 min is 3.99 ppm. Then the experiment at a feed pressure of 20 psi was also repeated at 30 and 40 LPM of feed flow rate which gives oxygen concentration in permeate stream 4.52 and 5.01 ppm, respectively. At higher feed flow rate, the amount of oxygen diffused into the deoxygenated water through membrane increases (Suhaina *et al*., 2004).

Effect of temperature: The experiment to study the effect of operating temperature on oxygen permeation was carried out at four different temperatures. Besides that, the operating pressure was kept constant at 10 psi and at 60% pumping rate.

From the Fig. 3 the dissolved oxygen against permeation time, the amount of oxygen concentration in the permeate increases for every feed flow rate as the temperature is increased. From the results, at 20 LPM feed flow rate, the amount of oxygen permeated into the deoxygenated water at 120°C is 3.87 ppm. At the same temperature the oxygen concentration measured in the permeate stream is 4.91 ppm at 40 LPM. Hence, as the operating temperature and feed flow rate are increased, the permeate oxygen concentration increases as well.

However, this increment is restrained in the range of temperature 40-120°C only. When the feed temperature is further increased to 150°C, a different behavior of oxygen permeation result was obtained. It is found that the oxygen permeation is higher at a lower feed flow rate than at higher feed flow rate. This is because at a temperature of 150°C and higher feed flow, there is no temperature drop in the gas boundary layer. In the liquid laminar flow regime, there is significant temperature drop which reduces the temperature at the wall of the membrane. Under these conditions, the heat transfer rate is determining for the mass transfer which results in the temperature polarization phenomenon. As a matter of fact, increasing the water temperature will decrease the solubility of oxygen into the thus results in decreasing of dissolved oxygen concentration. The high temperature of

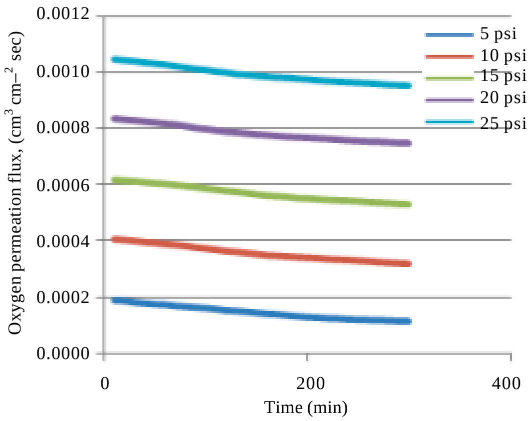

Fig. 4: Effect of pressure on oxygen permeation flux dependence at 20 LPM air flow

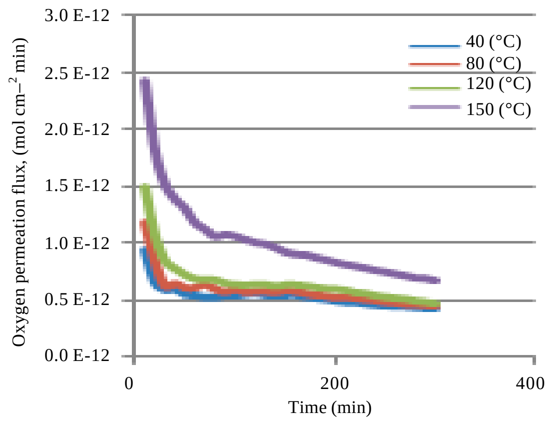

Fig. 5: Effects of operating temperature on oxygen permeation flux at 20 LPM

the feed helps in transporting the mass (oxygen) across the membrane. The high temperature will act as an energy source for the transporting of the oxygen through the membrane as it will achieve the activation energy as in the Arrhenius equation. The increasing flux with increasing of operating temperature leads to a higher diffusion rate of oxygen gas. The temperature dependence of the oxygen permeation flux at different feed flow rate is determined by the amount of oxygen concentration increased in the permeate stream. The increasing of oxygen permeation flux with increasing temperature attributed to the promotion of the oxygen diffusion and the oxygen surface reaction rates to pass across the membrane.

Flux dependence on pressure: Figure 4 shows the relationship between the flux and operating pressure. It can be seen that the flux increases as the operating pressure increased. The partial pressure difference across the membrane increases as the higher operating pressure thus makes an increasing flux of oxygen enrichment. Besides that, there is a slight decreasing of flux at different feed flow rates under the same operating pressure. In this case, it can be deduced that a slight increment in feed flow rate has a small effect on the oxygen flux.

Flux dependence on temperature: Oxygen permeation flux at different operating temperatures and at 20 LPM air flow rate is shown in Fig. 5.

Figure 5 shows that the flux value increases slightly from 40-120°C. However, when the feed temperature is further increased to 150°C, a distinct acceleration of the oxygen permeation flux is observed. When the experiment repeated at 30 and 40 LPM of feed flow rate, it is also found that the oxygen permeation flux increased which is proven that the dependence of permeation flux on operating flow rate as well. Hence, it is an evident that higher diffusion rate of the oxygen can be achieved and the enlarged free volume between the molecular chains is responsible for the increase of oxygen flux (Li *et al*., 1996).

CONCLUSION

The potential use of dense ceramic membrane for oxygen enrichment by reverse pervaporation has been analyzed by the studying of the effects of operating pressure and temperature. The results showed that the permeate oxygen concentration increased with increasing in operating pressure from 5-25 psi. In this pressure interval, there is no decreasing value of oxygen concentration. As the pressure increased, the oxygen permeation flux also increased but the flux values decreased with permeation time. The decreasing value of the flux calculated over time is due to the reduction of concentration gradient between the permeate and feed stream. The effects of operating temperature on oxygen permeation shows that the oxygen enrichment in permeate increases when the temperature is increased. However, the results show that the highest permeate oxygen concentration obtained at highest temperature and lower air flow rate, 20 LPM due to temperature polarization. Hence, it can be concluded that dense ceramic membrane have a good potential in oxygen enrichment and can be used for industrial purpose.

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