Oxygen Enrichment from Air Through Multilayer Thin Low-Density Polyethylene Films

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ABSTRACT: Several multilayer thin low-density polyethylene (LDPE) films were fabricated by blown thin film having a thickness of 7 μm and an area of 130 cm^2. They were characterized for their oxygen-enrichment performance from air by a constant pressure–variable volume method in a round permeate cell with an effective area of 73.9 cm^2. The relationship between oxygen-enrichment properties, including oxygen-enriched air (OEA) flux, oxygen concentration, permeability coefficients of OEA, oxygen, nitrogen, as well as separation factor through the multilayer LDPE films, and operating parameters, including transfilm pressure difference, retentate/permeate flux ratio, temperature, as well as layer number, are all discussed in detail. It is found that all of the preceding oxygen-enrichment parameters increase continuously with an increase of transfilm pressure difference from 0.1 to 0.65 MPa, especially for the trilayer and tetralayer LDPE films. The oxygen concentration and separation factor appear to rapidly increase within the retentate/permeate flux ratio below 200, and then become unchangeable beyond that, whereas the OEA flux and the permeability coefficients of OEA, oxygen, and nitrogen seem to remain nearly constant within the whole retentate/permeate flux ratio investigated, especially for the monolayer and bilayer LDPE films. The selectivity becomes inferior, whereas the permeability becomes superior, as the operating temperature increases from 23 to 31°C. The highest oxygen concentration was found to be 44.8% for monolayer LDPE film in a single step with air containing oxygen of 20.9% as a feed gas and operating pressure of 0.5 MPa at a retentate/permeate flux ratio of 340 and 23°C. The results demonstrate a possibility to prepare an oxygen-enriching membrane directly from air, based on the easily obtained thin LDPE films. © 2002 John Wiley & Sons, Inc. J Appl Polym Sci 83: 3013–3021, 2002; DOI 10.1002/app.2331

Key words: multilayer thin LDPE film; oxygen-enriching membrane; oxygen enrichment from air; oxygen-enrichment operation; gas separation; multilayer film

INTRODUCTION

Polyethylene (PE) has been widely used in many areas such as light industry, meters, toys, packing, printing, architecture, and agriculture because it possesses good mechanical and chemical properties and high transparency. As a general material, its development seems to have been accomplished; however, as a high-performance material, PE is believed to display some potential applications in some special aspects. For example, ultrahigh strength fiber spun from ultrahigh molecular weight PE has been woven into bullet-proof clothing, protective gloves, and kneepads, showing excellent protection against bullets or knives, like Kevlar fiber. It was also reported that PE exhibited a medium oxygen over nitrogen permselectivity, with the oxygen permeability co-

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efficient 2.93 × 10^{-10} mL (STP) cm/cm^2 s^{-1} cmHg^{-1} and oxygen over nitrogen separation factor P_{O_2}/P_{N_2} 3.06 at 25°C.[1] The permeation of other gases through PE film was previously investigated.[2–9] Additionally, microporous hollow fiber membrane from polyolefins has been melt-manufactured for the preparation of artificial lungs.[10–12] Because of its advantages of being easily obtained and processed and being more flexible than most other thermotropic aromatic copolyesters,[13–21] as well as having a much lower cost than that of polyvinylpyridine,[22] polypyrrolone,[23] polyimide,[23] and even traditional gas-separation membrane polymers such as polydimethylphenylene oxide,[24] polymethylpentene,[25] polysiloxane,[26] and cellulose derivatives,[27–29] PE would possess a better comprehensive performance and potential application for gas separation. Moreover, thermotropic aromatic copolyester is believed to be an excellent gas-barrier material for packaging applications.[30,31]

As we know, membrane separation has an industrial potential because of its economy of energy and operating facility.[32] Unfortunately, the application of membrane for air separation has not always been realized in wide scope because of the lack of high-performance membrane materials. So far, it has taken a lot of effort to seek out high-performance membrane materials for air separation,[30,33–36] although few of the desired materials of practically applicable worth are available. As a matter of fact, high-performance membrane materials for air separation have indeed been synthesized, although the synthetic method is too complicated to achieve in industry. For instance, the synthetic process of the cobalt complex used for air separation includes several steps and the final products (oxygen carrier) must be kept in an oxygen-free atmosphere.[33,34] Furthermore, the stability of the membrane made of the cobalt complex is very poor because it will be quickly degraded by oxygen in air. To some extent, it is significant to fabricate membranes from ready-made materials.

As mentioned earlier, LDPE could be one of the oxygen-enrichment materials having fairly good comprehensive performance. However, oxygen-enrichment properties from air have not yet been reported. The objective of this study is to gain insight into the effects of operating parameters, including transfilm pressure difference, retentate/permeate flux ratio, temperature, and layer number, on the permeate flux of oxygen enrichment air (OEA), oxygen concentration in the OEA, permeabilities for OEA, oxygen, and nitrogen, as well as oxygen/nitrogen separation factor, thus to comprehensively obtain oxygen-enrichment properties from air through LDPE film. The LDPE film is considered useful for air separation for the first time.

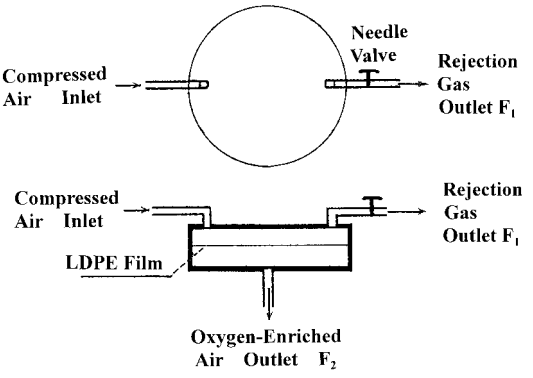

Figure 1 Scheme of permeation cell for investigation of oxygen enrichment from air through thin LDPE films.

EXPERIMENTAL

Thin LDPE films prepared by the conventional blow-molding process at 180–210°C were commercially received. The monolayer thin film thus obtained had an uniform thickness of 7 μm. The bilayer, trilayer, and tetralayer thin LDPE films used for investigation of oxygen enrichment from air were fabricated by a lamination process, and they had the thickness of 14, 23, 29 μm, respectively. The oxygen-enrichment measurements were always made with the same LDPE film samples and, for multilayers, stacked in the same order in the permeability cell.

Oxygen-enrichment measurements were performed by a constant pressure–variable volume method in the permeation cell shown in Figure 1.[36] The permeation cell, which had an effective permeate area of 73.9 cm^2, was designed to be circular in shape with double O-ring seals above and below the evaluated membrane side so as to be guaranteed airtight at an applied high pressure. The feed side of the cell was connected with an inlet pipe and an outlet pipe at the opposite borderline, respectively, and the interval between the inlet and the outlet was designed to be as long as possible. The outlet pipe of the feed side was equipped with a needle valve to adjust rejection flux conveniently. Multilayer thin LDPE films were characterized at a given transfilm pressure difference from 0.1 to 0.65 MPa, a given retentate/

permeate flux ratio, defined by rejection flux F_1 over permeate flux F_2, from 0 to 900 or so, and a given temperature from ambient to 60°C. Feed gas was compressed air, with oxygen content of 20.9 vol %, directly from an air compressor. The rejection flux F_1 and the permeate flux F_2 through the multilayer LDPE films were calculated by measuring the change of the rejection air volume or the permeated OEA volume at fixed operating parameters. The oxygen concentration in OEA permeated was determined with a QF 1901–type gas analyzer by means of oxygen absorption with a copper–ammonium hydroxide–ammonium chloride Cu—$NH_3 \cdot H_2O$—NH_4Cl mixed solution. After the absorption of oxygen, the residual gas was still absorbed with 5 vol % sulfuric acid aqueous solution saturated by sodium chloride, to remove gaseous ammonia from Cu—$NH_3 \cdot H_2O$—NH_4Cl solution during oxygen absorption. The calculated equations are as follows:

$$F_1 = V_1/t_1 \tag{1}$$

$$F_2 = V_2/t_2 \tag{2}$$

$$F_1/F_2 = V_1t_2/V_2t_1 \tag{3}$$

$$Q_{OEA} = 273.15V_2/(t_2AT) \tag{4}$$

where V_1 or V_2 is the volume (mL) of rejection air or OEA permeated in the testing time (s) t_1 or t_2; Q_{OEA} is the OEA flux [mL (STP)/s cm^2], and the Q_{OEA} value was corrected with standard state; A is the effective permeated area (cm^2), with a magnitude of 73.9 cm^2; T is the ambient temperature (K).

Based on the preceding original data and the oxygen concentration Y_{O_2} in OEA, the permeability coefficients [mL (STP) cm/cm^2 s^{-1} cmHg^{-1}] of OEA, oxygen, and nitrogen, P_{OEA}, P_{O_2}, and P_{N_2}, respectively, as well as separation factor P_{O_2}/P_{N_2} were calculated by using the following equations:

$$P_{OEA} = Q_{OEA}L/\Delta P \tag{5}$$

$$P_{O_2} = Q_{OEA}Y_{O_2}L/\Delta P_{O_2} \tag{6}$$

$$P_{N_2} = Q_{OEA}(1 - Y_{O_2})L/\Delta P_{N_2} \tag{7}$$

$$P_{O_2}/P_{N_2} = Y_{O_2}\Delta P_{N_2}/[(1 - Y_{O_2})\Delta P_{O_2}] \tag{8}$$

where L is the thickness (μm) of the multilayer LDPE film; ΔP is the transfilm pressure difference (cmHg); and ΔP_{O_2} or ΔP_{N_2} is the partial pressure difference (cmHg) of oxygen or nitrogen across the film.

Given that the LDPE films used in this study were made industrially in a large scale, the homogeneity of the film thickness was always better than ±3%. The permeability cell was held at a constant temperature (±0.5°C) in an air atmosphere inside a thermostatted housing. The error for the absolute values of the permeability coefficients could be estimated to about ±5%, resulting from the uncertainty of the determination of the transfilm pressure difference and the effective film area and thickness, whereas the reproducibility is better than ±1%.

RESULTS AND DISCUSSION

Influence of Transfilm Pressure Difference on Oxygen Enrichment from Air

Figure 2 shows an influence of transfilm pressure difference on oxygen-enrichment properties from air through monolayer, bilayer, trilayer, and tetralayer LDPE films at 31°C. Obviously, the oxygen concentration and the OEA flux Q_{OEA} through these multilayer LDPE films increase simultaneously with an increase in transfilm pressure difference from 0.1 to 0.65 MPa. The oxygen concentration and OEA flux through all films at the transfilm pressure difference of 0.65 MPa are 1.2–1.4 and 5.1–8.9 times as high as those at the transfilm pressure difference of 0.1 or 0.2 MPa, respectively. Similar results were observed for other multilayer or composite membranes.[36] A concurrent increase in oxygen concentration and OEA flux could be attributed to a combination of an acceleration in the OEA passing rate through the LDPE films with a compression between the thin films as the transfilm pressure difference increases.

It can be also seen from Figure 2 that within the testing transfilm pressure difference, the tetralayer LDPE film has the highest oxygen concentration of 34.2–42.1%, whereas the monolayer LDPE film has the highest OEA flux of 3.1–27 $\times$ 10^{-5} mL (STP)/s cm^2. The fairly good true oxygen-enrichment properties are seldom available in other conventional polymer materials. Theoretically, the respective oxygen permeability and oxygen over nitrogen separation factor across LDPE film at 25°C are 2.93 $\times$ 10^{-10} mL (STP)cm/cm^2

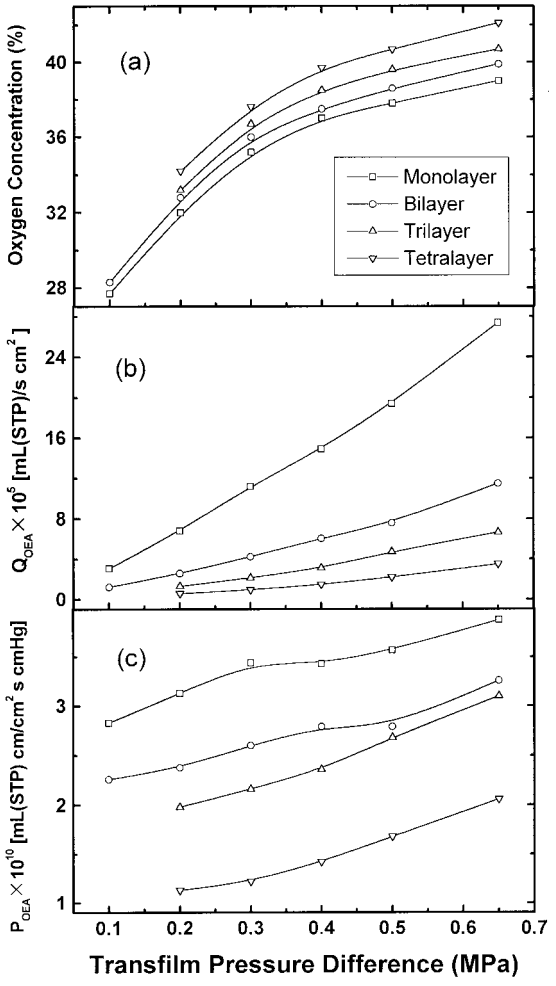

Figure 2 Variation of oxygen concentration Y_{O_2} (a), OEA flux Q_{OEA} (b), and OEA permeability P_{OEA} (c) with transfilm pressure difference at a temperature of 31°C and a retentate/permeate flux ratio from 300 to 500 for multilayer thin LDPE films: (□) monolayer film with thickness of 7 μm; (○) bilayer film with thickness of 14 μm; (△) trilayer film with thickness of 23 μm; (▽) tetralayer film with thickness of 29 μm.

s^{-1} $cmHg^{-1}$ and 3.02, which are not higher than those across other membranes, but its inherent permselectivity for O_2/N_2 could display completely in a true oxygen-enrichment process because of its excellent thin-film–forming ability and flexibility. On the contrary polystyrene, which has the higher theoretical O_2/N_2 separation factor of 6, does not exhibit any true oxygen-enrichment ability at all because no pinhole-free thin membrane can be obtained as a result of its high brittleness.[36] It follows that excellent thin-film–forming ability and flexibility of polymer materials is one of the necessary properties for true oxygen enrichment.

Calculated oxygen-enrichment parameters such as permeability coefficients of OEA, oxygen, and nitrogen (P_{OEA}, P_{O_2} and P_{N_2}), as well as separation factor P_{O_2}/P_{N_2} are shown in Figure 2(c) and Figure 3. It can be seen that P_{OEA}, P_{O_2}, and P_{N_2} exhibit similar increasing regularities as the transfilm pressure difference increases. Of all the preceding permeability coefficients curves, P_{O_2} curves show the highest increased slopes, indicating that oxygen has the fastest acceleration passing through the film with an increase of pressure. Compared with the literature values at 25°C, all these multilayer LDPE films show higher P_{O_2} and P_{N_2} values as a result of higher testing temperature. P_{O_2} through the trilayer and tetralayer LDPE films exhibits a continuous increase with an increase in pressure, in which the P_{O_2} values at the transfilm pressure difference of 0.65 MPa are 1.6 and 1.8 times, respectively, as high as those at the pressure difference of 0.2 MPa. It is interesting that the increasing rates of

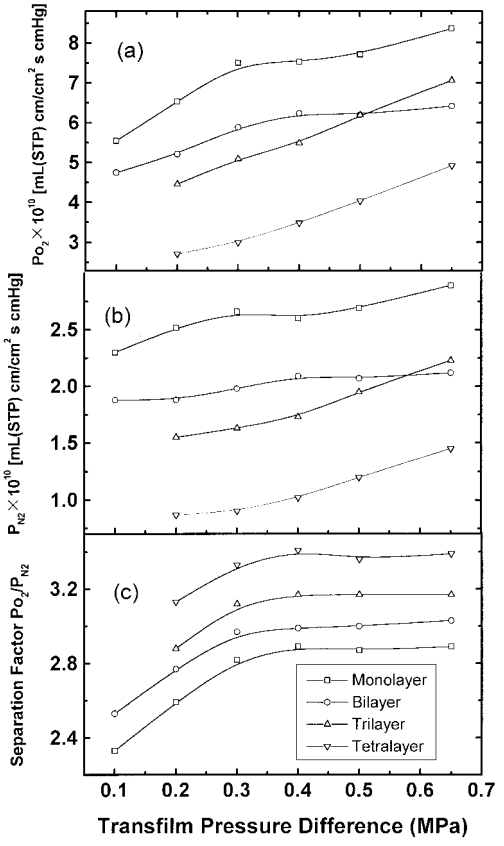

Figure 3 Variation of oxygen permeability P_{O_2} (a), nitrogen permeability P_{N_2} (b), and separation factor P_{O_2}/P_{N_2} (c) with a transfilm pressure difference at a temperature of 31°C and a retentate/permeate flux ratio from 300 to 500 for multilayer thin LDPE films. For symbols, see Figure 2 legend.

P_{OEA}, P_{O_2}, and P_{N_2} across the monolayer and bilayer LDPE films obviously slow down at high pressure differences. These could be explained with the compaction between porous support and the LDPE thin films. The compaction should also exist in the trilayer and tetralayer LDPE films, although this compaction could be counterbalanced by many more wrinkles within the films. Thus P_{OEA}, P_{O_2}, and P_{N_2} across the trilayer and tetralayer LDPE films increase steadily.

Figures 2(a) and 3(c) show some similar changes of oxygen concentration and separation factor with an increase in pressure difference. The oxygen concentration and separation factor increase rapidly, initially, and then slow down as the pressure difference increases, which is coincident with other multilayer or composite membranes.[36] The highest O_2/N_2 separation factor in this study at 31°C is 3.39, corresponding to an oxygen concentration of 42.1% when feed gas is air containing 20.9% oxygen. It follows that thin LDPE film has the potential application for a true oxygen-enrichment process if the OEA flux could be further enhanced by adding a porous supporter.

These results show that the permeability coefficients for oxygen and for nitrogen in LDPE increased with increasing transfilm pressure difference. Furthermore, the values of P_{O_2} and P_{N_2} decreased as the number of stacked LDPE layers used was increased, being the lowest for a tetralayer and the highest for a single layer. As a result, the ideal separation factor P_{O_2}/P_{N_2} (i.e., the selectivity of the LDPE films to oxygen relative to nitrogen) was also found to be dependent on transfilm pressure difference and on the number of stacked LDPE layers used. These observations suggest that the LDPE layers used may have contained defects such as pinholes. Therefore, the transfilm oxygen and nitrogen flux of single LDPE layers may have been enhanced and oxygen/nitrogen selectivity lowered because of a faster and less gas-selective transport mechanism through the defects than by solution–diffusion, such as Knudsen or Poiseuille flow. Solution–diffusion is the common gas transport mechanism through nonporous polymer membranes. By stacking two or more LDPE layers on top of each other the values of P_{O_2} and P_{N_2} decreased, although the oxygen/nitrogen selectivity of the stack was increased because of the low probability that pinholes in adjacent layers would coincide.

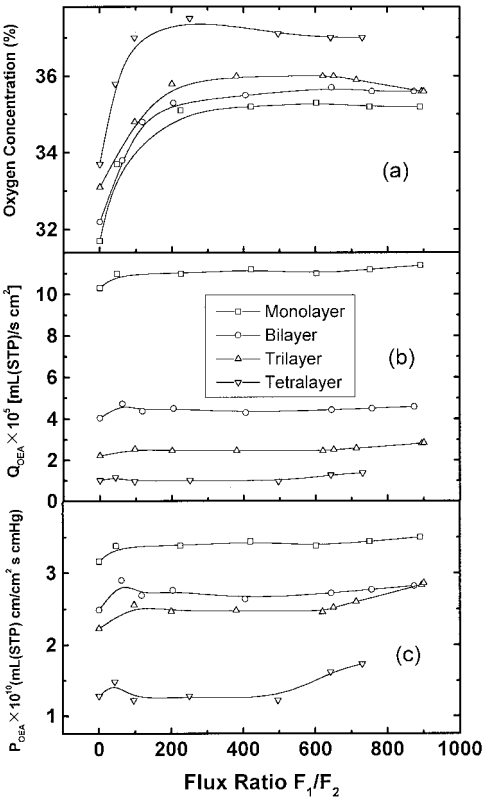

Figure 4 Variation of oxygen concentration Y_{O_2} (a), OEA flux Q_{OEA} (b), and OEA permeability P_{OEA} (c) with a retentate/permeate flux ratio at the transfilm pressure difference of 0.3 MPa and a temperature of 31°C for multilayer thin LDPE films. For symbols, see Figure 2 legend.

Influence of Retentate/Permeate Flux Ratio on Oxygen Enrichment from Air

The influence of retentate/permeate flux ratio on oxygen enrichment from air across the multilayer LDPE films at a fixed transfilm pressure difference of 0.3 MPa is shown in Figures 4 and 5. It can be seen from Figures 4(a) and 5(c) that both oxygen concentration and separation factor across the multilayer films increase rapidly in the retentate/permeate flux ratio range from 0 to approximate 200, and then substantially keep constant from the retentate/permeate flux ratio 200 up to 900 or so. It is apparent that the lower air separation ability at the retentate/permeate flux ratio below 200 is caused by concentration polarization in the feed air. With an increase in the retentate/permeate flux ratio from 0 to 200, the extent of the concentration polarization reduces and the air is refreshed step-by-step; thus, the oxygen concentration and separation factor gradually increase until the retentate/permeate flux

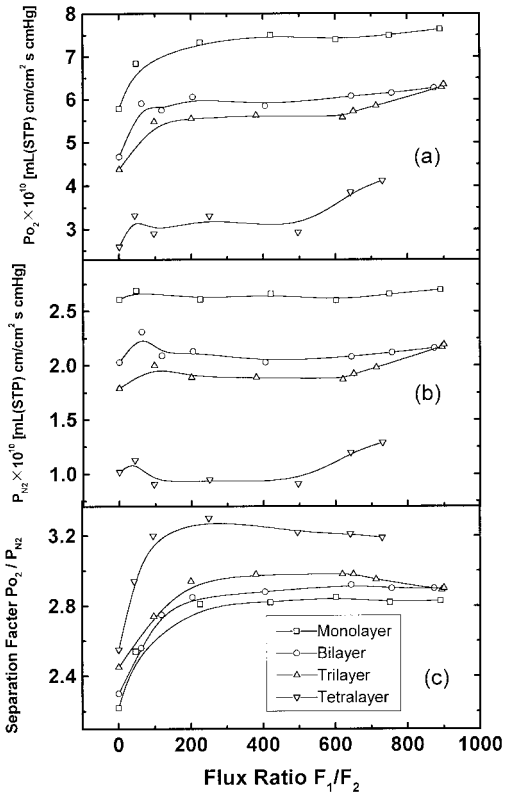

Figure 5 Variation of oxygen permeability P_{O_2} (a), nitrogen permeability P_{N_2} (b), and separation factor P_{O_2}/P_{N_2} (c) with a retentate/permeate flux ratio at the transfilm pressure difference of 0.3 MPa and a temperature of 31°C for multilayer thin LDPE films. For symbols, see Figure 2 legend.

ratio of about 200 is attained. Beyond that, the feed air is further refreshed but the concentration polarization has already been eliminated; therefore, the oxygen concentration and the separation factor no longer increase and essentially maintain fixed values, especially for monolayer and bilayer LDPE films. It must be noted that some small declines in oxygen concentration and separation factor across the trilayer and tetralayer LDPE films are observed in Figures 4(a) and 5(c). Considering the obvious rises in permeabilities of OEA, oxygen, and nitrogen from Figures 4(c), Fig. 5(a) and (b), it could be inferred that pinholes form after several measurements at high pressure differences. Undoubtedly, many wrinkles on thin film surfaces sandwiched in between the two films would be responsible for the defects in the films. These same wrinkles lead to worse stability of trilayer and tetralayer LDPE films than those of the monolayer and bilayer LDPE films in the later testing period.

As can be seen in Figure 4(b), the OEA flux through all multilayer films substantially remains constant in a retentate/permeate flux ratio range from 100 to 900. However, peaks appear at the retentate/permeate flux ratio of about 50 for most multilayer films, and these peaks become obvious in P_{OEA}, P_{O_2}, and P_{N_2} against retentate/permeate flux ratio curves. The trend could be explained by the fact that oxygen is a fast gas but nitrogen is a slow gas. Thus a rapid increase in oxygen concentration with the retentate/permeate flux ratio in the range from 0 to 200 would result in the increase of oxygen flux, consequently resulting in the increase of OEA flux and the permeability coefficients in the same retentate/permeate flux ratio range. On the other hand, the preceding permeability parameters would decline slightly, given that the actual transfilm pressure difference is slightly dropped with the retentate/permeate flux ratio changing from 0 to 150. Thus, the two opposite influential factors would lead to the occurrence of the above-noted peaks.

From an engineering viewpoint, a great retentate/permeate flux ratio is not acceptable for true oxygen-enrichment because the enhancement of the retentate/permeate flux ratio means the enhancement of the rejection flux, leading to the consumption of more energy. Certainly, an extremely small retentate/permeate flux ratio cannot attain OEA with higher oxygen concentration. Apparently, there must be an optimum retentate/permeate flux ratio. For LDPE multilayer film at a transfilm pressure difference of 0.3 MPa, the optimum retentate/permeate flux ratio is found to be about 200. A very similar result was observed at a transfilm pressure difference of 0.5 MPa.

Influence of Layer Number on Oxygen Enrichment from Air

With increasing the layer number from 1 to 4, the oxygen concentration continuously increases and the OEA flux decreases continuously and remarkably in the examined pressure difference range of 0.1 to 0.65 MPa, as indicated in Figure 2(a) and (b) as well as in Figure 4(a) and (b). These behaviors are coincident with the other multilayer membranes[36] and are easily understood. Figure 6 presents a relationship between OEA flux and oxygen concentration for multilayer LDPE films at the ambient temperature of 31°C in the pressure difference range of 0.1 to 0.65 MPa. It is quite evident that the monolayer LDPE film exhibits the highest OEA flux of 2.7×10^{-4} mL (STP)/s cm^2, whereas the tetralayer LDPE film exhibits the highest oxygen concentration of 42.1% at the ambient temperature. Although the

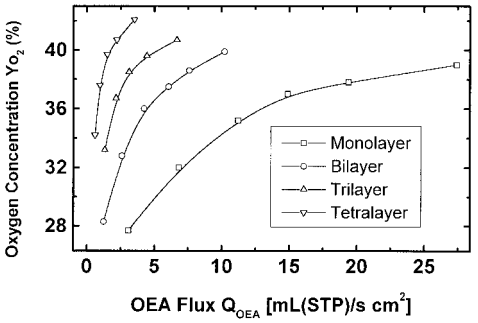

Figure 6 Plots of oxygen concentration Yo_2 and OEA flux Q_{OEA} through multilayer thin LDPE films at a transfilm pressure difference from 0.1 to 0.65 MPa, a retentate/permeate flux ratio from 300 to 500, and a temperature of 31°C. For symbols, see Figure 2 legend.

maximum OEA flux and the maximum oxygen concentration cannot be available by the same multilayer film, monolayer LDPE film gives much more magnitude on OEA flux and a little bit less magnitude on oxygen concentration than that of the other three multilayer LDPE films. This monolayer LDPE film is supposed to exhibit better comprehensive capability of oxygen enrichment from air. It is appreciated that further improvements could be done for the monolayer LDPE film by means of fabricating this film with other porous support membranes.

Influence of Temperature on Oxygen Enrichment from Air

Figures 7 and 8 present the oxygen-enrichment abilities from air for monolayer and trilayer LDPE films at two temperatures of 23 and 31°C. Over the whole transfilm pressure difference examined in this study, the OEA flux at 31°C is 2 to 3 times and 6 to 9 times as high as in the OEA flux at 21°C, whereas the oxygen concentration at 31°C is 94–99 and 88–93% the size of the oxygen concentration at 21°C for monolayer and trilayer LDPE films, respectively. The detailed dependency of the OEA flux and oxygen concentration on temperature for tetralayer LDPE film is shown in Figure 9. Likewise, the OEA flux increases gradually and the oxygen concentration decreases linearly with increasing temperature from 30 to 60°C. Additionally, a distinct acceleration of OEA flux is observed in the temperature range of 35–55°C. These behaviors are similar to those typically observed for many other composite membranes,[36] because higher temperature is inclined to originate less-discriminating gaps or more free volume in the films. On the other hand, the air will become less condense with rising temperature.

Based on a comparison of the temperature dependency of oxygen concentration and separation factor [see Figs. 7(a) and 8(c)] through the monolayer LDPE film at 23°C and the trilayer LDPE film at 31°C in the transfilm pressure difference range of 0.3 to 0.65 MPa, it was found that both oxygen concentration and separation factor through the monolayer film are higher than those through the trilayer film, suggesting that the affect of increasing temperature of 8°C is greater than that of increasing the layer number to 2. That is to say, the enhancement of the selectivity resulting from an increasing layer number is completely offset by a reduction in the selectivity from an increase in temperature. It is interesting that the OEA flux through the monolayer film is also higher than that through the trilayer film, although the permeability coefficients for oxygen and nitrogen through the former film are lower

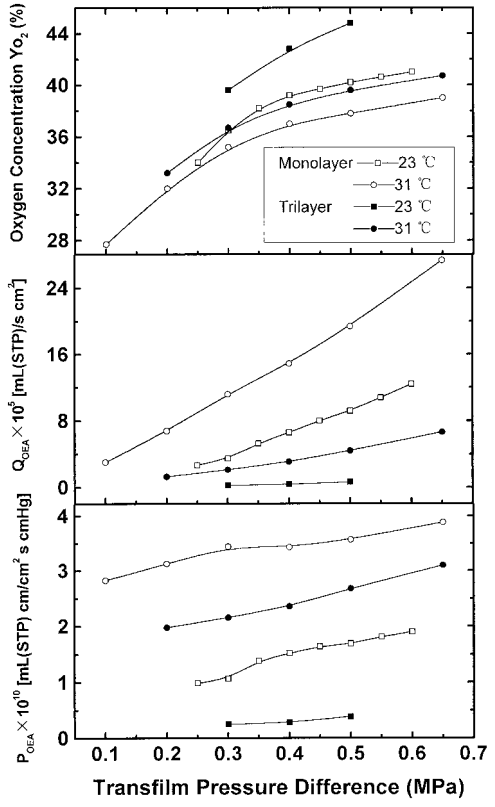

Figure 7 Effect of temperature on oxygen concentration Yo_2 (a), OEA flux Q_{OEA} (b), and OEA permeability P_{OEA} (c) through monolayer and trilayer thin LDPE films at a transfilm pressure difference range of 0.1 to 0.65 MPa and a retentate/permeate flux ratio range of 300 to 500 at two temperatures (□, ■ 23°C; ○, ● 31°C).

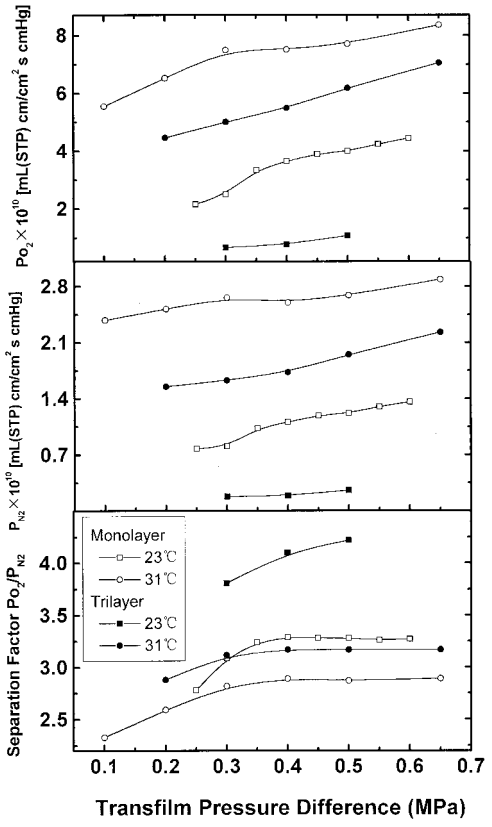

Figure 8 Effect of temperature on permeabilities of oxygen P_{O_2} (a), nitrogen P_{N_2} (b), and separation factor P_{O_2}/P_{N_2} (c) through monolayer and trilayer thin LDPE films at a transfilm pressure difference range of 0.1 to 0.65 MPa and a retentate/permeate flux ratio range of 300 to 500 at two temperatures (□, ■ 23°C; ○, ● 31°C).

than those through the latter film. This implies that both the oxygen concentration and separation factor through the monolayer film are simultaneously higher than those through the trilayer film. A much smaller thickness for the monolayer film would be responsible for this behavior. This phenomenon is seldom observed for many other membranes.[36] From an engineering standpoint the characteristic is a desirable feature for an air-separation membrane. Therefore, it is of practical significance that the film for air separation should be as thin as possible.

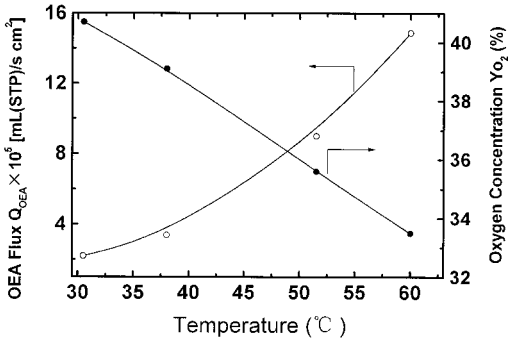

Figure 9 Variation of OEA flux Q_{OEA} (○) and oxygen concentration Y_{O_2} (●) with temperature across tetralayer LDPE film at a transfilm pressure difference of 0.5 MPa and a retentate/permeate flux ratio range from 70 to 470.

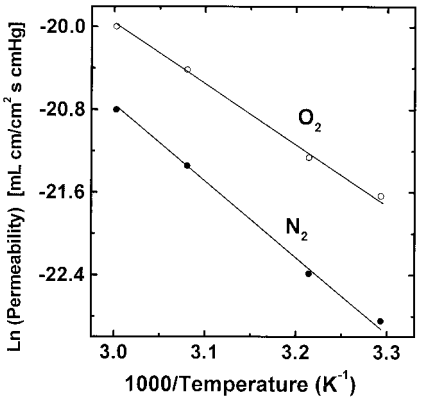

Figure 10 Arrhenius plots of oxygen permeability P_{O_2} (○) and nitrogen permeability P_{N_2} (●) through tetralayer LDPE film at a transfilm pressure difference of 0.5 MPa and a retentate/permeate flux ratio range from 70 to 470.

Note that the separation factor for trilayer LDPE film at 23°C is 3.8–4.2, which is higher than the literature value of 3.02 at 25°C. Besides the reason that a little lower temperature leads to higher separation factor, another reason may be the difference in density between the two LDPE materials.

Figure 10 displays the Arrhenius plots of the permeability coefficients for oxygen and nitrogen in tetralayer LDPE film in an examined temperature range of 30–60°C. It can be seen that the permeability coefficients for oxygen and nitrogen increase linearly with an increase in temperature. The slope of ln P_{O_2} against the reciprocal temperature curve is lower than that of ln P_{N_2} against the reciprocal temperature curve, indicating that oxygen permeability possesses a weaker dependency on temperature than nitrogen. The calculated activation energies of oxygen and nitrogen are 48 and 60 kJ/mol (303–333 K), respectively. These activation energies are higher than the literature values of E_{O_2} = 31.3 kJ/mol (257–313 K)[8] or 42.7 kJ/mol (278–333 K) and E_{N_2} = 49.4 kJ/mol.[1] The diversity of these magnitudes is probably attributable to the difference of the testing method or temperature range. In our ex-

periment, air directly from an air compressor was used as a feed gas, whereas in the literature, either pure gas or $O_2/N_2/CO_2$ (14.3/72.7/13) mixture gas was used as a feed gas. Given the different interactions of oxygen and nitrogen in the three kinds of feed gases, the sensitivity of permeability on temperature is changed. On the other hand, the diversity is also related to its density or fabrication fashion of the film. The film in this study is a tetralayer film, which was laminated with four 7-μm-thick thin films, inducing some gaps between the thin films.

CONCLUSIONS

Multilayer thin LDPE films were fabricated and evaluated for their true oxygen-enrichment performance. It was demonstrated that the OEA flux and the oxygen concentration through the multilayer LDPE films are remarkably influenced by operating pressure, retentate/permeate flux ratio in the range of 0 to 200, layer number, and temperature. The highest oxygen concentration is found to be 44.8%, and the corresponding OEA flux is 6.5×10^{-6} mL (STP)/s cm^2 for trilayer LDPE film in a single step, with air as a feed gas at 23°C and the transfilm pressure difference at 0.5 MPa. The highest OEA flux is found to be 2.7×10^{-4} mL (STP)/s cm^2, and the corresponding oxygen concentration is 39.0% for the monolayer LDPE film in a single step, with air as a feed gas at 31°C and a transfilm pressure difference of 0.65 MPa. These preliminary results demonstrated another characteristic of traditional LDPE thin film with a potential application for air separation.

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