

Matrimid-based carbon tubular membranes: The effect of the polymer composition

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ABSTRACT: In this article, we present a development study of new membrane materials and enhancements of productive membranes to improve the current performance of polymeric membranes. Carbon membranes are a promising material for this matter as they offer an improvement in the gas-separation performance and exhibit a good combination of permeability and selectivity. Carbon membranes produced from the carbonization of polymeric materials have been reported to be effective for gas separation because of their ability to separate gases with almost similar molecular sizes. In this study, a carbon support membrane was prepared with Matrimid 5218 as a polymeric precursor. The polymer solution was coated on the surface of a tubular support with the dip-coating method. The polymer tubular membrane was then carbonized under a nitrogen atmosphere with different polymer compositions of 5–18 wt %. The carbonization process was performed at 850°C at a heating rate of 2°C/min. Matrimid-based carbon tubular membranes were fabricated and characterized in terms of their structural morphology, thermal stability, and gas-permeation properties with scanning electron microscopy, thermogravimetric analysis, Fourier transform infrared spectroscopy, and a pure-gas-permeation system, respectively. Pure-gas-permeation tests were performed with the pure gases carbon dioxide (CO₂) and N₂ at room temperature at a pressure of 8 bar. On the basis of the results, the highest CO₂/N₂ selectivity of 75.73 was obtained for the carbon membrane prepared with a 15 wt % polymer composition. © 2015 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* **2015**, *132*, 42394.

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INTRODUCTION

Depletion of natural gas quality has become a main concern in the gas processing industry because of the increasing impurity content over the years. Among these impurities, carbon dioxide (CO₂) is considered crucial for the industry. Stringent CO₂ removal is necessary because its high content would decrease calorific values and cause pipeline corrosion, and this would directly affect the processing and maintenance cost if it failed to be removed efficiently.¹ Several processes have been implemented as solutions; these include cryogenic distillation, pressure swing adsorption, and amine absorption in natural gas purifications. These well-matured processes offer a high CO₂ recovery, and their reliability and adaptability have been proven in various industries. However, conventional processes suffer

from a high processing cost, high solvent loss, and large space occupation.

Separation with polymeric membranes is a high-performance separation method and provides lower operating costs compared to other separation techniques. This technique is widely used for wastewater treatment, gas separation, seawater desalination, distillation, and dialysis. Polymeric membranes have the potential to replace the conventional separation method because of their modest energy and modular equipment requirement compared to those of conventional methods. Although polymeric-membrane-based gas separation offers a low energy requirement and low capital cost,² the Robeson tradeoff limit between the permeability and selectivity is the main obstacle that hinders the exploration of the polymeric membrane process.

The limitations suffered by polymeric membranes have encouraged researchers to develop new classes of membranes. Among various classes of membranes, carbon membranes have been considered as very promising candidates for gas separation. Carbon membranes can be prepared from various types of polymer precursors via heat-treatment (carbonization) processes. During carbonization, the benzene ring and other functional groups present in the polymeric precursor will be broken. This results in amorphous materials consisting of disordered sp^2 -hybridized condensed hexagonal sheets with pores formed from packing imperfections; this serves an idealized pore structure, and a superior membrane emerges.³ Carbon membranes have the potential to be applied under rigorous systems, such as those at excessive pressure, at excessive temperature, and with a high presence of absorbing components. Most polymer precursors used to fabricate carbon membranes are commercially expensive materials, and some of them are even exclusively obtained on the laboratory scale. The choice of commercially available polymers as precursors is very important in the development of high-quality carbon membranes.⁴

However, because carbon membranes are brittle, they require a strong support to sustain their structure. Although the majority of reported carbon membranes have used a flat substrate, a tubular support hardly draws similar attraction even though it is mechanically stronger against a compressing pressure and has a higher membrane area per unit module volume. Because the development of a tubular supported carbon membrane is a relatively a new approach, it is important to identify the most ideal precursor, which would result in a high-performance tubular carbon membrane.⁵ Polyimides are known to exhibit a high permselectivity for various gas pairs, especially for CO_2/N_2 ,⁶ and a high chemical resistance and thermal stability. Many researchers have reported that Matrimid 5218 is one of the best material choices for membrane-based CO_2 separation because of its attractive combination of gas permselectivity and permeability.⁷

EXPERIMENTAL

Materials

Matrimid 5218 was selected as the polymer precursor. It was dried overnight at $80^\circ C$ to remove any moisture. *N*-Methyl-2-pyrrolidone, purchased from Merck (Germany), was used as the solvent. Tubular ceramic membranes with a nominal cutoff of 1 kD (TAMI) were used as supports for the fabrication of the carbon tubular membrane. The tubular supports, 8 cm in length, 1.3 cm in diameter, and 0.3 cm in thickness, were purchased from Shanghai Gongtao Ceramics Co., Ltd. The tubular supports, with an average pore size of $0.2 \mu m$ and a porosity of 40–50%, were made by TiO_2 (4.5–5.5 mm) with a coating of ZrO_2 (2–3 nm) on the inner surface.

Carbon Membrane Preparation

The polymer precursor solution was prepared by the dissolution of different polymer compositions of 5–18 wt % Matrimid 5218 in *N*-methyl-2-pyrrolidone for 7 h with mechanical stirring. The mixture was sonicated to remove all bubbles from the solution. The supported polymer membranes were prepared by the dip-coating of the tubular support into the polymeric

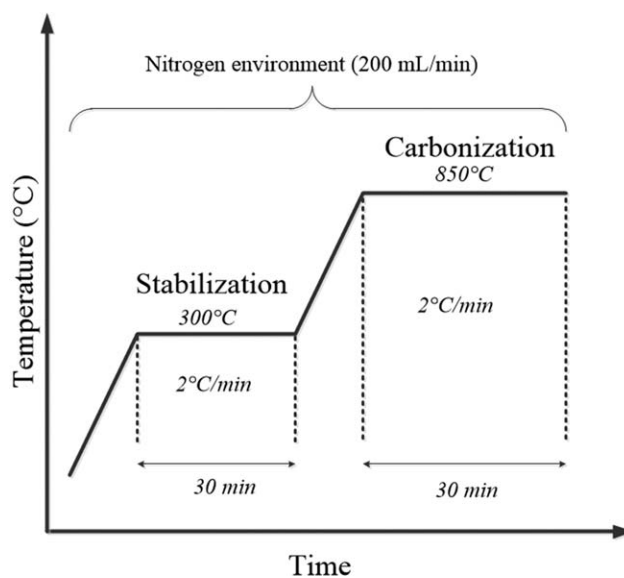


Figure 1. Carbonization protocol.

solution for 15 min. The supported membranes then underwent aging at $80^\circ C$ for 24 h. The membranes were then immersed in methanol for 2 h before they were placed inside an oven at $100^\circ C$ for 24 h to allow the slow removal of the solvent. Subsequently, the supported membranes were placed at the center of the Carbolite horizontal tubular furnace to undergo carbonization process. The carbonization process was performed at a temperature of $850^\circ C$ under nitrogen gas flow (200 mL/min). A heating rate of $2^\circ C/min$ was applied throughout the process. After each heating cycle was complete, the prepared membranes were cooled naturally to room temperature. The detailed carbonization protocol is illustrated in Figure 1. Additionally, free films of these polymer compositions were prepared according to a similar procedure for the supported carbon membranes for characterization purposes.

Membrane Characterization

Thermal analysis was performed via thermogravimetric analysis (TGA) at a heating rate of $10^\circ C/min$. Unsupported films were crushed into small flakes before the analysis. The thermal behavior of the prepared membrane was determined by the placement of these flakes through thermal stability toward temperature changes from 40 to $900^\circ C$. The TGA experiments were performed with a PerkinElmer TGA-7 with an N_2 atmosphere and a flow rate of $20 cm^3/min$. Scanning electron microscopy (SEM) was used to observe the cross sections of the prepared carbon membranes and to estimate fluctuation in the membrane dimensions, as a consequence of the heat-treatment process. The unsupported film was also used for this analysis. Attenuated total reflectance/infrared spectroscopy analysis with universal attenuated total reflectance (Single Reflection Diamond for the Spectrum Two, PerkinElmer, L1600107) was conducted to observe the existence of the functional group in the prepared membranes.

Pure-Gas Measurements

The performance of the membrane was characterized by two important parameters, which were permeance and selectivity.

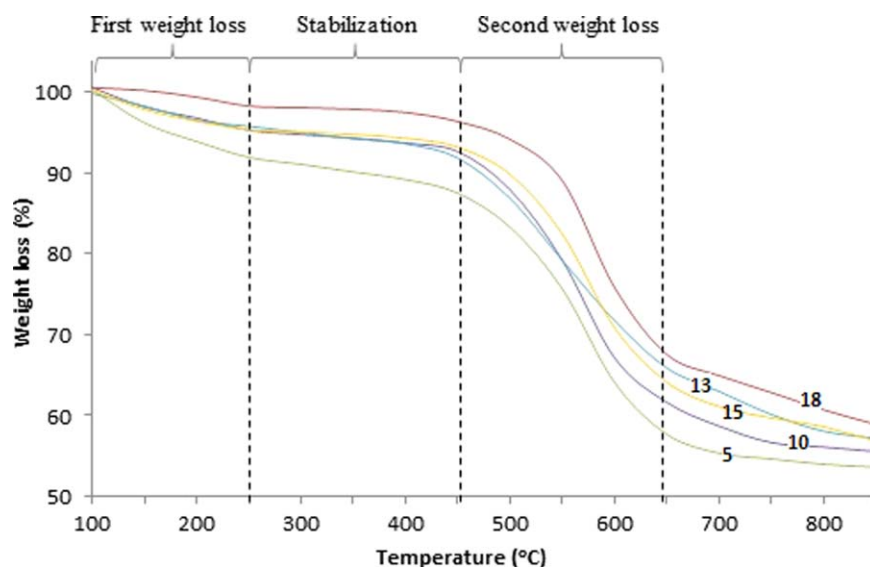


Figure 2. Weight loss of Matrimid-based membranes prepared with various polymer compositions versus the temperature as measured by TGA. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

The carbon tubular membranes were tested in pure-gas-permeation system. An 8-cm carbon tubular membrane was placed inside the membrane module. A tubular stainless steel module 14 cm in length was used to contain the carbon tubular membrane. The membrane was then fitted with rubber O-rings to allow the membrane to be housed in the module without leakage. The pure gases CO_2 (0.33 nm) and N_2 (0.36 nm) were fed into the module at a trans membrane pressure of 8 bar. The permeance [P/l (GPU)] and selectivity (α) of the membranes were calculated with the following equations:

$$(P/l)_i = \frac{Q_i}{\Delta p A} = \frac{Q_i}{\eta \pi D l \Delta p} \quad (1)$$

$$1 \text{ GPU} = 1 \times 10^{-6} \text{ cm}^3(\text{STP}) \text{ cm}^{-2} \text{ s}^{-1} \text{ cmHg}^{-1}$$

where P/l is the permeance of the membrane (GPU), Q_i is the volumetric flow rate of gas i at standard temperature and pressure [cm^3 (STP/s)], Δp is the pressure difference between the feed side and the permeation side of the membrane (cmHg), A is the membrane surface area (cm^2), n is the number of fibers in the module, D is the an outer diameter of the membrane (cm), and l is the effective length of the membrane (cm):

$$\alpha_{ij} = \frac{(P/l)_i}{(P/l)_j} \quad (2)$$

where α_{ij} is the selectivity of gas penetrant i over gas penetrant j and $(P/l)_i$ and $(P/l)_j$ are the permeance of the gas penetrants i and j , respectively.

RESULTS AND DISCUSSION

Characterization of the Polymer and Carbon Tubular Membranes

Thermal Behavior Analysis. Generally, for the fabrication of carbon membranes, polymers with high glass-transition temperatures (T_g s) are favorable choices as precursors. T_g indicates the ability of polymers to withstand high temperature during the carbonization process. In this study, the thermal decomposition

behaviors of Matrimid-based membranes, together with the weight loss during heat treatment, were observed by TGA. We observed that the color of the tubular membranes changed from yellow to black after the carbonization process; this indicated the transformation of the organic material into a carbon material. Figure 2 presents the thermally induced weight loss of Matrimid versus temperature as measured by TGA.

According to the thermogravimetric curve, the first weight loss occurred at 100–250°C for all of the prepared membranes; this was attributed to the residual solvent evaporation (boiling point of *N*-methyl-2-pyrrolidone = 202–204°C). The first weight loss was more prominent at a low polymer composition. The weight of the samples remained almost constant from 250°C up to approximately 450°C. The second weight loss started at a temperature of 450°C and ended around 650°C. This was attributed to the evolution of $-\text{CO}$, $-\text{CO}_2$, and $-\text{CH}_4$ during the heat-treatment process; this caused a cleavage of the benzene ring of the Matrimid-based membrane.⁸

Furthermore, the TGA profile also provided information on the stabilization process. On the basis of TGA, we found that the most appropriate temperature for the stabilization process was at temperatures above 250°C, where all of the residual solvent was evaporated. The stabilization temperature should also have not exceeded T_g of the Matrimid polymer ($T_g = 328^\circ\text{C}$) to prevent a melting stage during the carbonization process.⁹ In this study, the stabilization temperature was set at 300°C throughout the experiment. In addition, degradation of Matrimid ended at 650°C, which indicated a minimum temperature for the carbonization process. A thermally stable structure was created in the membrane matrix by crosslinking during the stabilization step, which led to the formation of defect-free carbon membranes after carbonization. This result was in agreement with the literature reported by Briceño *et al.*¹⁰

Morphological Structure. The morphological structure of the Matrimid-based carbon membranes were viewed via SEM. The

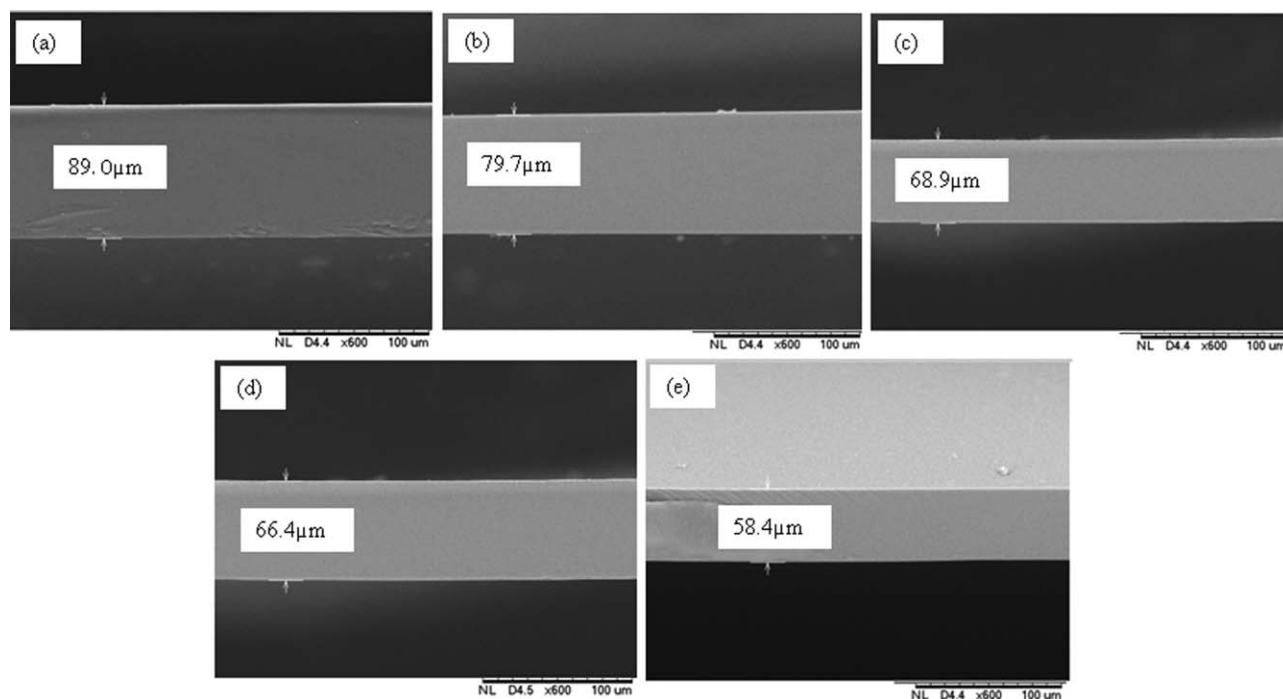


Figure 3. SEM images of the cross sections of carbon membranes prepared with the following polymer compositions: (a) 5, (b) 10, (c) 13, (d) 15, and (e) 18 wt %.

gas-separation mechanisms of the prepared membrane were found to be highly dependent on the membrane morphology. All of the membranes prepared at various polymer compositions showed a dense structure. This was because further heat induced on the membrane led to densification. At a low polymer composition, rapid solvent evaporation likely led to nonselective carbon membranes, whereas at a high polymer composition, slower phase inversion resulted in slow solvent evaporation and hindered gas permeation because of the thinner dense layer. These phenomena are often depicted by polymer chain entanglement because of the polymer chain reorientation at different polymer compositions.¹¹ Figure 3 shows the SEM images of the carbon membranes prepared from different polymer compositions.

We observed that the thickness of the carbon membrane prepared from 5 wt % polymer was around 89.0 μm . A further increase in the polymer composition systematically decreased the membrane thickness from 89.0 (5 wt %) to 58.4 μm (18 wt %). Because of the slower phase inversion, the thickness of the polymer composition was reduced with increasing polymer composition. This was due to adequate time for polymer chain reorientation and a tendency to produce a thinner dense layer. This structure was generated during the dip-coating process during the fabrication of the tubular membrane. Aside from the membrane thickness, no apparent difference was observed in the membrane structure at different polymer compositions. As the membranes were established as supported membranes, the different polymer composition and the thickness of the selective layer influenced the final gas-permeation results of the membrane.¹²

Fourier Transform Infrared (FTIR) Results. The molecular orientation of the Matrimid-based membrane was measured via

FTIR spectroscopy. Figure 4 shows the FTIR spectra of the polymer membrane prepared from different polymer compositions. The chemical structural changes took place through the transformation from polymer to carbon, and this was observed by FTIR spectroscopy. The bands in the range 719.97–722.24 cm^{-1} indicated the presence of C–H aromatic monosubstituted benzene. The wavelengths from 1718.08 to 1780.65 cm^{-1} were attributed to asymmetric and symmetric C=O stretching vibrations of imide groups. The bands observed at 1363.12 to 1363.47 cm^{-1} for all of the compositions proved the presence of aliphatic C–N stretching of the imide groups, whereas the aromatic C=C bending vibration was indicated at 1506.71–1508.35 cm^{-1} . C–N–C transverse stretching was indicated at 1093.57–1099.30 cm^{-1} for the polymeric precursor membrane. A previous study stated that the amide group was characterized by bands around 1661 cm^{-1} (stretching of C=O in the amide group) and 1539 cm^{-1} (stretching of C–N and/or bending of N–H in the amide group).¹³

Figure 5 shows the FTIR spectra of the carbon membrane prepared with different polymer compositions. Compared to its polymeric counterpart, no apparent bond was observed in the FTIR ranges between 2500 to 600 cm^{-1} . The decomposition and breakage of the chemical structure occurred as the carbonization temperature increased. Similarly, Ismail and David¹⁴ characterized the transformation of other polymer structure according to the thermal history. As the polymer composition increased throughout the carbonization process, a significant reduction in the peaks was observed for all of the carbon membranes.¹⁵ It was observed that because the structural compaction became closer to the attenuated total reflectance crystal interface, the transmission intensity decreased. In short, the

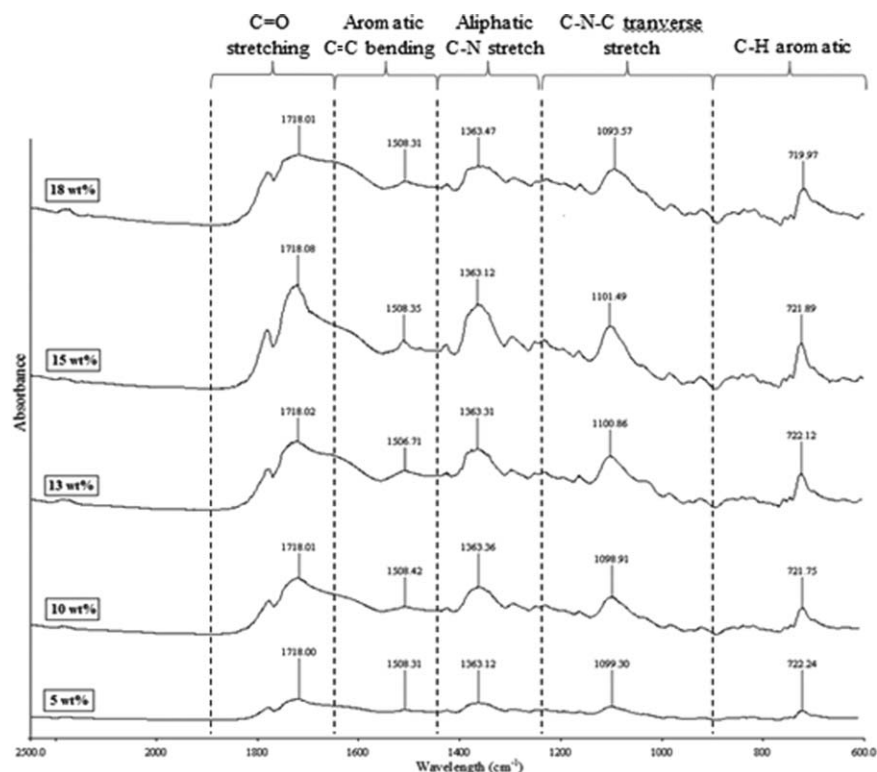


Figure 4. FTIR analysis of the polymeric precursor membrane prepared with different polymer compositions.

carbonization of the polymeric precursor still occurred regardless of the different polymer compositions used.

According to FTIR spectroscopy and TGA, this indicated that the Matrimid-based polymeric membrane underwent thermal degradation during the heat-treatment process. During the carbonization process, polymeric membranes can degrade via a number of mechanisms, which are generally involved in random scission, crosslinking, and side group elimination. Random scission results from the formation of free radicals along the backbone of the polymer and can occur at random locations in the

chain. Crosslinking generally occurs after some stripping of the substituent and involves the creation of bonds between two adjacent polymer chains. These processes are very important in the formation of chars because they generate a structure with a higher molecular weight that is less easily volatilized.¹⁶ In the chain elimination reaction, the bonds connecting side groups of the polymer chain to the chain itself are broken. The products of this reaction are generally small enough to be volatile. Specifically, the weight loss of the polymeric membranes after the carbonization process is attributed to the release of ammonia (NH₃), hydrogen cyanide (HCN), methane (CH₄), hydrogen

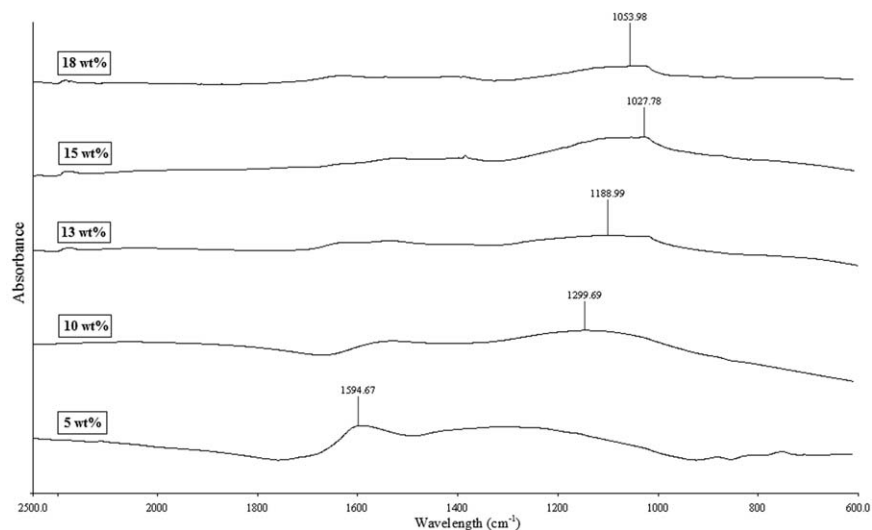


Figure 5. FTIR analysis of the carbon membranes prepared with different polymer compositions.

Table I. Gas-Permeation Properties of the Matrimid-Based Carbon Tubular Membrane

Polymer composition (wt %)	Polymeric membrane			Carbon membrane		
	Permeance (GPU)		Selectivity (CO ₂ /N ₂)	Permeance (GPU)		Selectivity (CO ₂ /N ₂)
	CO ₂	N ₂		CO ₂	N ₂	
5	2.160	0.792	2.727	101.990	1.960	52.036
10	2.357	0.843	2.796	115.300	2.110	54.645
13	3.320	0.915	3.628	137.990	2.160	63.884
15	6.497	1.007	6.452	171.350	2.260	75.819
18	3.920	0.944	4.153	145.670	2.175	66.975

(H₂), nitrogen (N₂), oxygen (O₂), carbon monoxide (CO), CO₂, benzene, phthalimide, and others.^{17,18} It was stated that cross-linking was dominant over the scission reaction in the Matrimid-based polymeric membrane because of the thermally stable properties. In fact, the greater the crosslinking density was, the higher T_g became. As the temperature increased above T_g , the polymer became denser in chain packing and density.⁶

Gas-Permeation Measurements

The carbonization process was performed by the heating of the Matrimid-based polymeric membrane under nitrogen flow from room temperature to the final carbonization temperature with different polymer compositions. Table I shows the gas-permeation properties of the Matrimid-based polymeric and carbon tubular membranes.

For the polymeric membrane, the highest permeance and selectivity readings were observed at the composition of 15 wt %, whereas the lowest permeance and selectivity readings were at 5 wt % Matrimid. The precursor composition affected the polymeric membrane gas-permeation results, depending on the different rates of solvent evaporation. The solvent evaporation of the membrane has been deeply described in polymeric membrane development.¹⁹ In general, a low polymer composition produces a membrane with a large pore size because of fast solvent evaporation. In contrast, at a high polymer composition, a slower phase inversion produces slow solvent evaporation and hinders gas permeation because of the small pore size.²⁰

We observed that the prepared carbon membranes showed a high CO₂ permeability and CO₂/N₂ selectivity. The gas-permeance values of the selected gases were in this order: CO₂ (3.3 Å) > N₂ (3.64 Å). The permeance values decreased with increasing kinetic diameter of the gases; this indicated that a molecular sieving mechanism dominated for carbon membrane-transport processes. This performance was due to the micropore presence within the carbon membrane, which allowed smaller molecules to permeate easily across the membrane and to discriminate against larger molecules.²¹ This was the result of the breakage of the benzene ring and other functional groups and created a bimodal rigid pore distribution.²²

The results indicate that the gas permeation across the carbon membrane was highly dependent on the precursor polymer composition. The CO₂ and N₂ permeation showed a systematic increase for carbon membranes prepared from 5 to 15 wt %

polymer compositions and began to diminish at 18 wt %. The increasing trend suggested that the micropore formation and carbon structure of the carbon membrane became rigid and compact, and some of the pores might have changed into closed pores during the carbonization process. The results reveal that the most suitable polymer composition for the preparation of carbon membranes was 15 wt % Matrimid. Similar results of the same composition were reported by Favvas's *et al.*,²³ who used carbon hollow-fiber membranes based on the Matrimid 5218 precursor. A higher composition of polymer solution induced a higher porosity.

The selectivity of the prepared carbon membrane at 18 wt % was approximately 67.00 with a CO₂ permeance of 145.67 GPU. The reduction of the CO₂ permeance for the carbon membranes was often related to the structural compaction occurrence and entanglement of the polymer chain when the polymer composition exceeded the ideal composition. Consequently, poor structural arrangements induced higher mass-transport resistance and hindered the permeation of the gases. Significant reductions in the CO₂ permeance and N₂ further suggested that the severe structural arrangement at a macroscopic level led to a lower CO₂/N₂ selectivity.²⁴

CONCLUSIONS

Carbon tubular membranes for gas separation were successfully fabricated by the carbonization of Matrimid. The structure and properties of the prepared carbon tubular membranes were tuned by the adoption of different polymer compositions. We found that the thickness of the dense structure decreased as the polymer composition increased. FTIR analysis indicated that the decomposition and breakage of the chemical structure occurred as the carbonization temperature increased. The carbon tubular membranes prepared from 15 wt % Matrimid had a higher separation performance for CO₂/N₂ systems than those prepared from lower and higher polymer compositions. The highest CO₂/N₂ selectivity of 75.73 was achieved when 15 wt % Matrimid was used in the polymer solution formulation.

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