

Investigation of Carbon Dioxide and Nitrogen Gases Permeability through PEBAX 1657/PEG/graphene Oxide Nanoparticle Mixed Matrix Membrane

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ABSTRACT

This study investigated gas permeation of neat and blend polyether block amide (PEBAX) 1657 membranes. Polyethylene glycol (PEG) and graphene oxide (GO) were added to PEBAX membrane matrix at different concentration to study their effects on the morphology, permeability and selectivity of the membranes. The prepared membranes were characterized by Fourier Transform Infrared Spectroscopy (FTIR), Scanning Electron Microscopy (SEM) and X-ray diffraction (XRD). The permeabilities of pure gases CO₂ and N₂ were carried out using a constant pressure-variable volume method. The influence of PEBAX 1657 membranes with different weight fractions of PEG and GO on the gas permeation performance were studied. The modified membrane with PEBAX/PEG (40 wt.%) displayed the optimum gas separation performance with a CO₂ permeability of 103.9 Barrer and selectivity of 56.3 for the CO₂/N₂ gas pair at 10 bar and room temperature. The permeability of this membrane was twofold compared to the pristine PEBAX, under the same temperature and pressure conditions, showing promising prospects for CO₂ capture.

Keywords: Mixed matrix membrane, PEBAX, Graphene oxide nanoparticle, Permeability, Selectivity

1.0 INTRODUCTION

As it is well known, CO₂ is one of the greenhouse gases whose major emissions sources are oil refineries, power generation, industrial growth and the combustion of fossil fuels. Membrane technology has shown great potential in CO₂ capture from economic and environmental perspectives [1, 2]. Several traditional technologies (absorption and adsorption) have been demonstrated effectively to capture CO₂. However, most of them are expensive to operate and also energy-intensive. Membrane separation processes, because of their outstanding features, lower capital investment, simplicity, lower energy consumption and smaller

space for equipment installation, are one of the best choices to take the place of the former methods [2, 3].

Lately the strategies on material development are centered on the incorporation of nanoparticles and modifiers. Polyether block amide (PEBAX) is a copolymer, composed of poly amide as hard segments and poly ether as soft segments in the polymer chains [4]. PEBAX 1657, consisting of 40 wt.% of polyamide 6 (PA6) and 60 wt.% of polyethylene oxide (PEO). Table 1 presents physical properties of PEBAX 1657. The performance of PEBAX has been studied by several researchers and the beneficial effect of PEBAX to enhance the CO₂ separation performance was shown.

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Table 1 Properties of PEBAX 1657 [4]

Mechanical properties	Dry Condition	Unit
Density	1.14	gr/cc
wt.% PA crystallinity in PA block	40	%
T _m polyamide block	207	°C
T _g polyether block	-55	°C

Ghadimi *et al.* [5] modified PEBAX with polyethyleneglycol diacrylate (PEGDA) and found that the permeation coefficient of CO₂ increased due to the excellent affinity between Ethylene oxide (EO) and CO₂ compared to those of pristine PEBAX. Simultaneously, permeability of CH₄ and N₂ through the blend membranes decreases by reduction of their fractional free volume (FFV), whereas permeability of H₂ remains constant. Therefore, it can be used in separation of CO₂ from light gases. Reijerkerk *et al.* [6] found that for a PEBAX 1657 blend membrane containing 50 wt.% PDMS-PEG additive, CO₂ permeability was increased by a factor 5 and whereas, a slight decrease in the selectivity of CO₂/H₂ was observed (at 35 °C and 4 bar). CO₂ sorption experiments revealed that the increase in permeability is dominated by an increase in gas diffusivity. So, ethylene oxide (EO) unit in PEGs has been proved to be an effective group to achieve high CO₂ permeability and CO₂/light gas selectivity. Yave *et al.* [7] combined PEG into poly ether block amide and found that the solubility of CO₂ increased due to the excellent affinity between EO and CO₂.

Wang *et al.* [8] applied PEG and carbon nanotubes (CNT) to improve separation performance of PEBAX 1657 membranes. In this work three components hybrid membranes containing PEBAX-PEG and multi walled carbon nanotubes (MWCNT) were prepared and their CO₂ capture

properties were evaluated. Results showed that high molecular weight PEGs in polymeric membranes decrease CO₂ permeability and CO₂/N₂ selectivity, while low molecular weight PEGs increase the CO₂ permeability and CO₂/N₂ selectivity compared with pristine Pebax membrane. The low molecular weight PEG400 and PEG600 tend to inhibit crystallization of Pebax. The incorporation of high molecular weight PEG increases the crystallinity. As previously stated, in order to improve the membrane performance, some modifications have also been made. Nano composites based on different fillers such as graphene have attracted more attention. Graphene tends to agglomerate, because of the intrinsic van der Waals forces; that can weaken the reinforcement of the nano composites. Therefore, the functionalization of graphene, such as oxidation, can make GO hydrophilic and reduce agglomeration [9, 10].

Zhao *et al.* [9] prepared PEBAX/GO and found that the CO₂ permeability of this mixed matrix membranes with 3.85 vol.% GO decreased by about 70%. Because of the impermeable nature of GO, the gas permeability of the PEBAX/GO MMMs remarkably decreased with the increasing GO content. A multi perm-selective mixed matrix membrane (MMM) was fabricated by incorporating versatile GO nanosheets modified by both PEG and PEI into PEBAX matrix for CO₂ capture by Li *et al.* [10]. Compared with pristine PEBAX membrane, the CO₂ permeability and CO₂ gas selectivity of the PEBAX/10 wt.% PEG-PEI-GO membrane were increased by about 2.7 folds and 2.4 folds respectively.

2.0 EXPERIMENTAL

2.1 Materials

PEBAX 1657 was purchased from Arkema (France) and PEG 600 was purchased from Tetrachem (China). Ethanol 99.99 wt.% was provided by Razi company (Iran) and the deionized water was supplied by zolal-chem company (Iran). CO₂ and N₂ gases (purity 99.9%) were purchased from the Aboughadare company (Iran).

2.2 Membrane Preparation

2.2.1 Dense Film Preparation

PEBAX polymer granules were added to an ethanol/water (70/30 wt.%) mixture [8, 11, 12]. The polymer solution was prepared at a concentration of 3 wt.% PEBAX 1657 at about 80 °C and stirred for 2 h. The bubble-free polymer solution was cast on flat-bottomed petri dishes. After the solvent was evaporated for 48 h at ambient conditions, membranes were stripped off from the petri dish and dried in a vacuum oven at 50 °C for 6 h to remove residual solvent.

2.2.2 MMMs Preparation

PEBAX 1657 blend membranes were prepared by the addition of different amount of PEG to PEBAX 1657 solution and stirred for 1 h at room temperature. The prepared solution was poured on a petri dish and dried in the same way as the dense membrane described earlier.

GO was synthesized from graphite by Hummer method [13]. To prepare MMMs, PEBAX/PEG solution was prepared similar to the method was illustrated previously. Then specified amount of GO was dispersed in deionized water and was sonicated until well dispersed to form a homogeneous

suspension. The resultant suspension and PEBAX/PEG solution were mixed and stirred for 5 h at room temperature. The nanocomposite membranes were dried in the same manner as the pristine PEBAX. All membrane thicknesses were in the range of 65-85 μm [9, 10].

2.3 Membrane Characterization

FTIR spectra of synthesized membranes were obtained by a Bruker (Vertex 80) spectrometer. All spectra were obtained from 4000-500 cm⁻¹.

X-ray diffraction (Bruker D8 advance) with 40 KV was used to evaluate crystallographic structure and intermolecular distances between the intersegmental chains in the presence of modifiers. The crystalline properties of membranes were measured with a wide-angle X-ray diffractometer with Cu Kα radiation (λ=1.54 Å).

The morphology and dispersion of GO nanoparticle in the membrane samples were examined by scanning electron microscopy (SEM) using a TESCAN model VEGA TS5136 MM microscope operated at 30 KV.

2.4 Gas Permeation Measurement

Theoretically, the solution-diffusion model was used to describe the gas permeation mechanism through dense polymeric membranes [14]. The pure gases CO₂ and N₂ permeation experiments were carried out using the constant pressure/variable volume method at three different pressures of 6, 8 and 10 bar at 25 °C. The circular cell membrane with effective area of 19.64 cm² was applied. The permeate side was maintained at atmospheric pressure. The gas permeability was calculated by the following equation.

$$P = \frac{Q.L}{\Delta p.A} \quad (1)$$

where P is the permeability coefficient expressed in Barrer [$1 \text{ Barrer} = 10^{-10} \text{ cm}^3 (\text{STP}) \text{ cm cm}^{-2} \text{ s}^{-1} \text{ cmHg}^{-1}$] [15], Q is the volumetric flow rate of permeated gas (cm^3/s), L is the membrane thickness (cm), Δp is the pressure difference across the membrane (cmHg) and A is the effective membrane area (cm^2).

The diffusion coefficient D (cm^2/s) was evaluated by the following formula [14, 16, 17]:

$$D = \frac{L^2}{6\theta} \quad (2)$$

where θ is the diffusion time lag (s) extrapolated from the plot of pressure with time at steady state to the time axis and L is the membrane thickness (cm). The solubility coefficient S [$\text{cm}^3 (\text{STP})/(\text{cm}^3 \text{ cmHg})$] was obtained by Equation (3) [18]:

$$P = D.S \quad (3)$$

where P is the permeability (Barrer). The CO_2/N_2 ideal selectivity (α_{ij}) is given by the following equation:

$$\alpha_{\text{CO}_2/\text{N}_2} = \frac{P_{\text{CO}_2}}{P_{\text{N}_2}} = \frac{D_{\text{CO}_2}}{D_{\text{N}_2}} \times \frac{S_{\text{CO}_2}}{S_{\text{N}_2}} \quad (4)$$

where P_{CO_2} and P_{N_2} are permeability of carbon dioxide and nitrogen respectively. $D_{\text{CO}_2}/D_{\text{N}_2}$ is the diffusivity selectivity and $S_{\text{CO}_2}/S_{\text{N}_2}$ is the solubility selectivity of CO_2/N_2 [17].

3.0 RESULTS AND DISCUSSION

3.1 Fourier Transform Infrared Spectroscopy (FTIR)

Figure 1 shows the FTIR spectrum of GO. The characteristic peaks at 3346 cm^{-1} corresponds to C-OH. The stretching vibrations at 1728 and 1224 cm^{-1} are assigned to C=O and C-O. Analysis of FTIR spectrum suggested

that there were at least some hydroxyl and carboxyl groups in GO [9, 10, 19, 20].

The FTIR spectra of pristine PEBAX, PEBAX/PEG40 wt.%, PEBAX/PEG40 wt.%/GO0.8 wt.% and PEBAX/PEG40 wt.%/GO0.2 wt.% are shown in Figure 2. For the neat PEBAX, the characteristic peaks at 3307.46 and 1125.91 cm^{-1} were the stretching vibration of N-H and C-O. The peak at 1728.45 cm^{-1} is assigned to C=O and the peak at 1457.88 cm^{-1} corresponds to the hydrogen bonded C=O in H-N-C=O. As it can be observed in Figure 2, the characteristic peaks of GO were not shown clearly in the nano composite membranes, which might be because they were so weak that they were overlapped by those strong Pebax peaks. On the other hand, because there was some Pebax matrix on the GO surface, the GO spectrum may not have been detected and only the Pebax spectrum was shown [9, 10, 20].

3.2 SEM

The cross-section morphologies of the pristine PEBAX and MMMS membranes are shown in Figure 3. As shown in Figure 3a, the membrane structure is without any defects and the neat PEBAX membrane is much smoother than that of blended (Figure 3b) and nanocomposite membranes [14]. It is evident in Figure 3c that GO nanoparticles are well dispersed throughout the polymer.

3.3 XRD

Generally, when a polymer contains an amorphous region, the peak from X-ray diffraction is broad although the narrow peak with strong intensity indicates the polymer has a crystalline region [24, 25]. XRD pattern of GO is shown in Figure 4. It has been reported that the

characteristic peak of GO was at 2θ of about 9.45° [10, 21, 22].

Figure 5 shows the XRD patterns of blended and nano composite membranes. PEBAX/PEG membrane revealed peaks at $2\theta=12.25^\circ$, 20.1° and 23.8° which peak at 23.8° attributed to the crystalline region of the PA segment. Both blended (PEBAX/PEG 40 wt.%) and nano composite (PEBAX/PEG 40 wt.%/GO 0.2 wt.%) membranes showed similar XRD patterns. No characteristic peak of GO

was observed in nanocomposite membrane which indicated that GO was fully exfoliated and dispersed in the polymer matrix properly [10].

The intensity of the diffraction peak at $2\theta = 23.8^\circ$ of nano composite (PEBAX/PEG 40 wt.%/GO 0.2 wt.%) membrane was higher than that of blended (PEBAX/PEG 40 wt.%) membrane; this indicated that GO functioned as a nucleating agent and induced the increase of crystallinity [9].

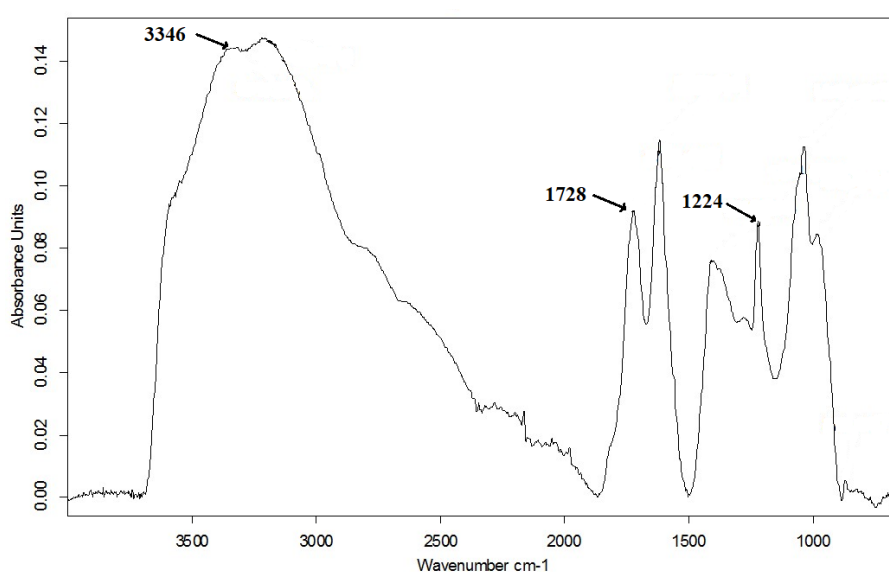


Figure 1 FTIR spectrum of GO

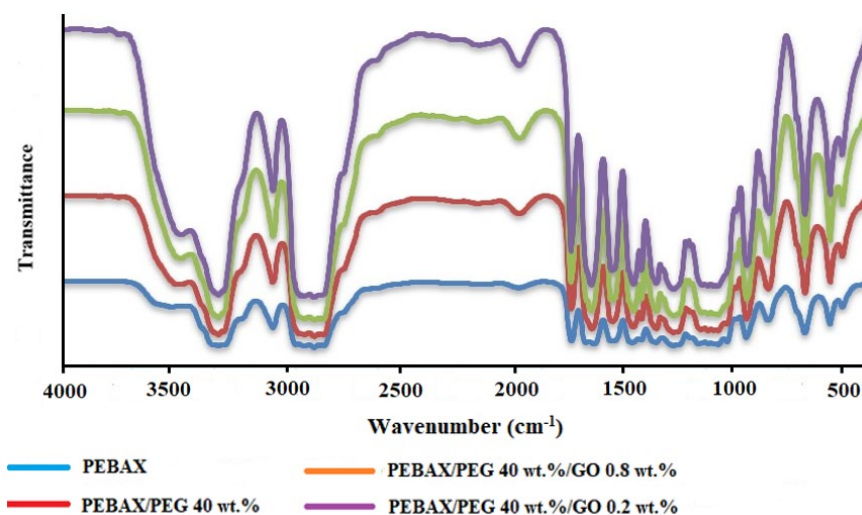


Figure 2 FTIR spectra of pristine PEBAX, PEBAX/PEG 40 wt.%, PEBAX/PEG 40 wt.%/GO 0.8 wt.% and PEBAX/PEG 40 wt.%/GO 0.2 wt.%

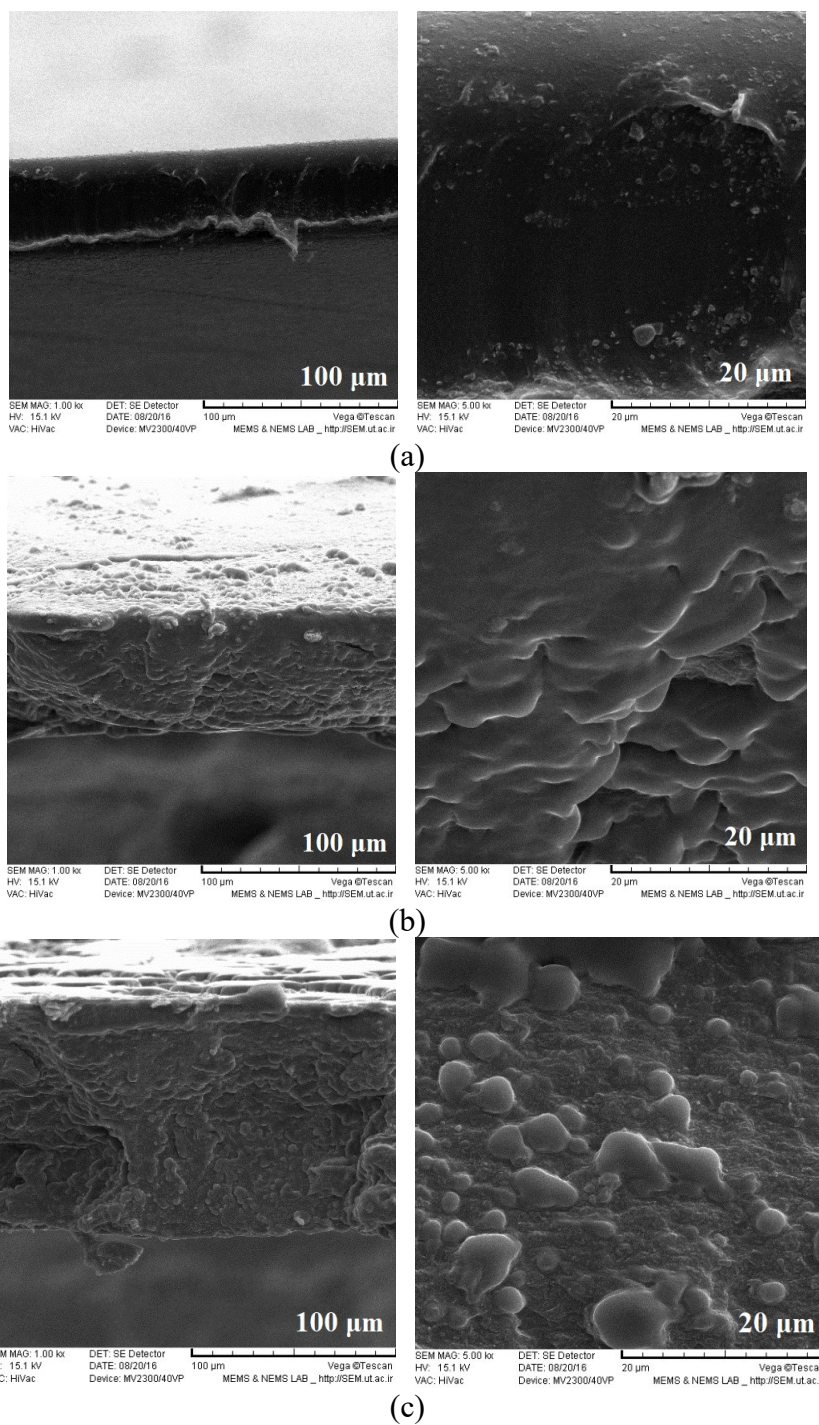


Figure 3 SEM images for the cross sectional of samples at different magnifications; (a) neat PEBAX, (b) PEBAX/PEG 40 wt.% and (c) PEBAX/PEG 40 wt.%/GO 0.2 wt.%

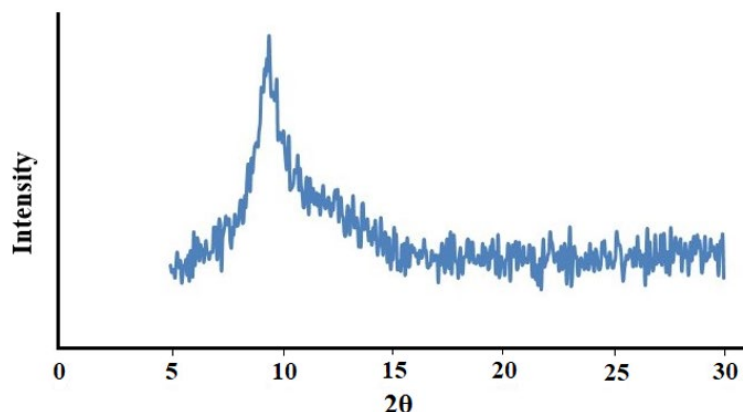


Figure 4 XRD pattern of GO

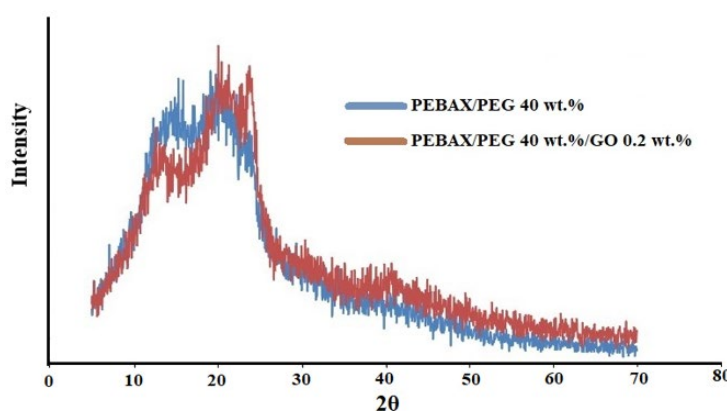


Figure 5 XRD pattern of PEBAX/PEG 40 wt.% and PEBAX/PEG 40 wt.%/GO 0.2 wt.%

3.4 Gas Permeability Performance

Permeability of pure gases N_2 and CO_2 for pristine PEBAX and MMMS membranes was investigated.

3.4.1 Effect of PEG Loading

Figures 6 and 7 show the effect of PEG loading of 0, 20, 30 and 40 wt.% on permeability CO_2 and N_2 respectively. By increasing PEG content, permeability of CO_2 and N_2 increases. The remarkable increase in the CO_2 permeability was mainly attributed to the presence of EO groups (polar ether oxygen) in the polymer matrix. This high CO_2 solubility is ascribed to the favorable interaction of the quadrupolar

CO_2 with the polar ether oxygen in the polymer. Moreover, by increasing EO groups, the PEBAX chain mobility increases further and this leads to higher diffusivity. Also PEG as a plasticizer reduces interchain space between PA and PEO in PEBAX, which will lead to an increase in diffusion rate of permeant CO_2 and N_2 [19, 20, 24]. The permeability of CO_2 is higher than N_2 due to the favorably interaction with CO_2 molecules. N_2 molecules do not interact with EO groups, thus the permeability of N_2 is more governed by diffusivity than solubility [6]. As shown in these Figures, PEBAX/PEG 40 wt.% exhibited the maximum CO_2 and N_2 permeability.

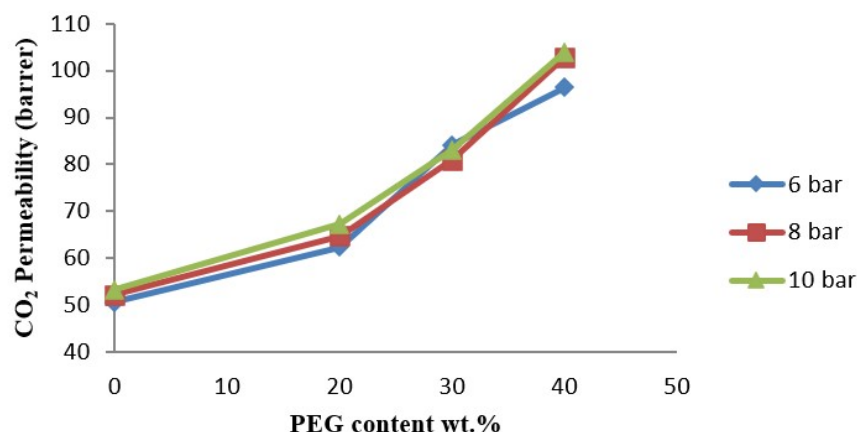


Figure 6 Effect of PEG content on the CO₂ permeability of the membrane at different feed pressure

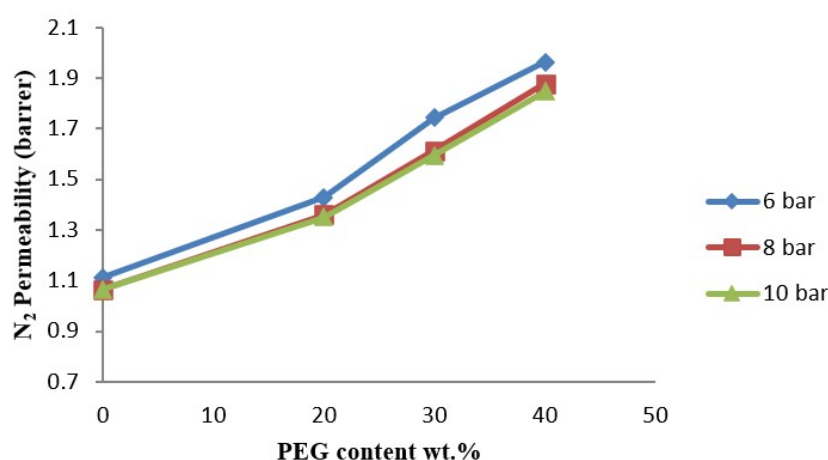


Figure 7 Effect of PEG content on the N₂ permeability of the membrane at different feed pressure

Figure 8 presents the effect of PEG loading of 0, 20, 30 and 40 wt.% on ideal selectivity of CO₂/N₂. The CO₂/N₂ selectivity exhibits an increased trend. The governing permeation mechanism, through dense membranes is solution-diffusion mechanism. According to this mechanism, gas dissolves into the membrane and penetrant gas molecules diffuse through the membrane. In this mechanism, permeability of gases is explained as a product of solubility coefficient (S) of gases and their diffusion coefficient (D). Gas permeation reflects a combination of thermodynamic factors like condensability of gas and its interaction with polymer segments ($S_{\text{CO}_2} > S_{\text{N}_2}$)

plus kinetic factors that are largely governed by penetrant size, polymer segmental mobility and free volume ($D_{\text{CO}_2} > D_{\text{N}_2}$). As a result of increasing the amount of PEG in PEBAX, CO₂ permeability and membrane selectivity ($\alpha_{\text{CO}_2/\text{N}_2}$) increase [5].

3.4.2 Effect of GO Content

Generally, GO incorporation into gas separation membranes reduces the gas permeability of MMMs [14]. Figures 9 and 10 present the influence of the GO content on the permeability of CO₂ and N₂, respectively. The results show that by increasing GO content, the permeability of CO₂ and N₂ was

decreased. Reduction of CO₂ and N₂ permeability illustrates that GO inhibited gas diffusion [9].

The sheet structure of high aspect ratio of GO restricted the diffusion of gas molecules [25]. Moreover, the available diffusion area was decreased because of the replacement of permeable pebax by impermeable GO. The gas diffusivity is closely dependent

on kinetic diameters of gas molecules; thus, the impermeable GO had a larger inhibition on the diffusivity of larger gases. Since the kinetic diameter of N₂ is bigger than that of CO₂, the CO₂ permeability is remarkably higher than N₂ permeability [9, 10].

Figure 11 indicates the effect of GO loading of 0, 0.2, 0.5 and 0.8 wt.% on ideal selectivity of CO₂/N₂.

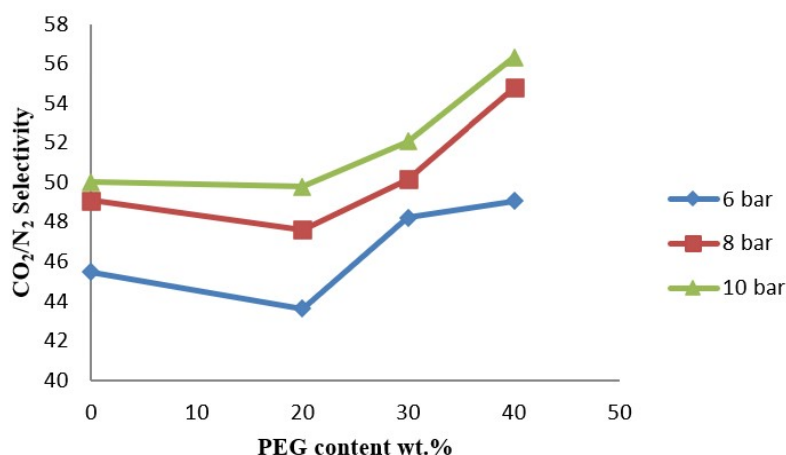


Figure 8 Effect of PEG content on the CO₂/N₂ selectivity of the membrane at different feed pressure

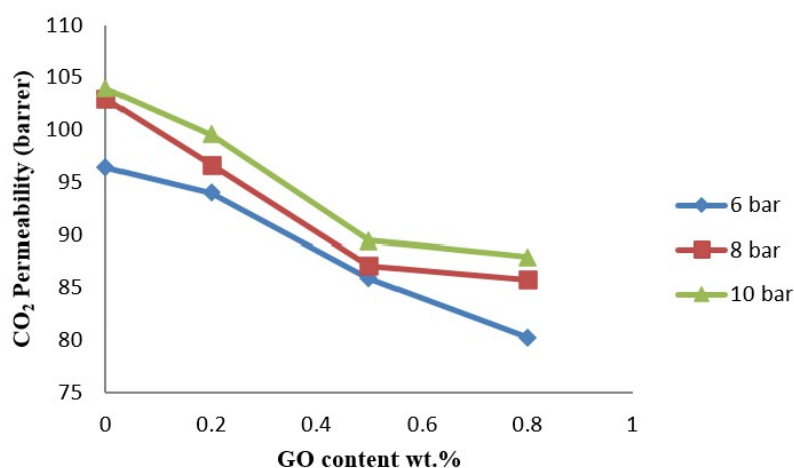


Figure 9 Effect of GO content on the CO₂ permeability of the membrane at different feed pressure

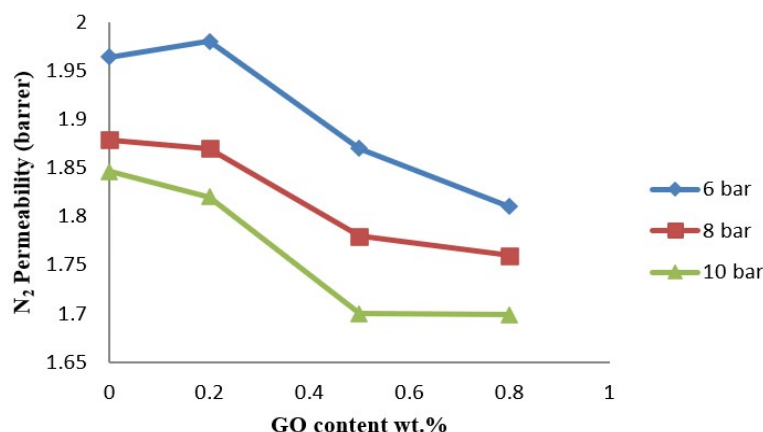


Figure 10. Effect of GO content on the N₂ permeability of the membrane at different feed pressure

As shown in Figure 11, by increasing GO content, the ideal selectivity of CO₂/N₂ has been decreased. The permeability of CO₂ and N₂ has been decreased by the incorporation of GO. So, because of the barrier effect of nano sheets, the CO₂/N₂ selectivity remarkably decreases with increasing GO content [9, 10].

Although CO₂/N₂ selectivity has been reduced with increasing GO content but the ideal selectivity remained reasonably high. The absorbed CO₂ in the membrane surface due to the hydroxyl and carboxyl groups (interaction of CO₂ with polar groups) leads to higher solubility [6, 9, 17, 24]. As discussed above, N₂ has a larger kinetic diameter than CO₂. Therefore, the difference in kinetic diameters

between N₂ and CO₂ leads to the reduction in mobility of polymer chains and restricts the diffusion of larger molecules. This increases CO₂/N₂ diffusivity selectivity.

3.4.3 Effect of Feed Gas Pressure

The permeability of CO₂ and N₂ at different feed pressures of (6, 8 and 10 bar) are shown in Figures 12 and 13 respectively. The results indicate that by increasing feed pressure the CO₂ permeability increased, whereas for N₂ the permeability reduced. In the case of quadrupolar CO₂, at higher pressures, enhancement of concentration and solubility coefficient of CO₂ dominate and resulting an increase in CO₂ permeability.

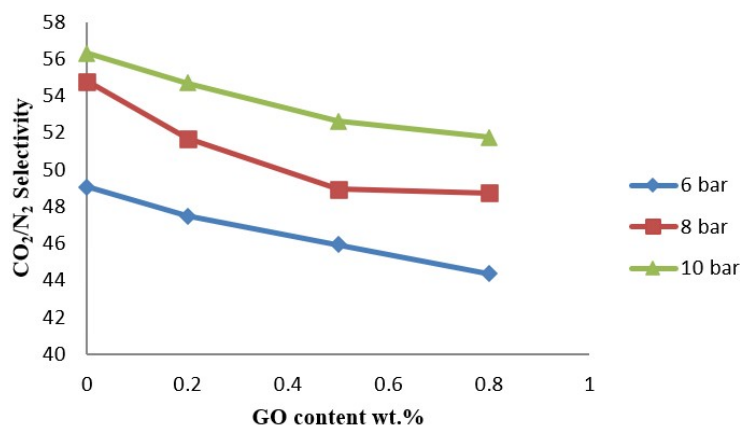


Figure 11 Effect of GO content on the CO₂/N₂ selectivity of the membrane at different feed pressure

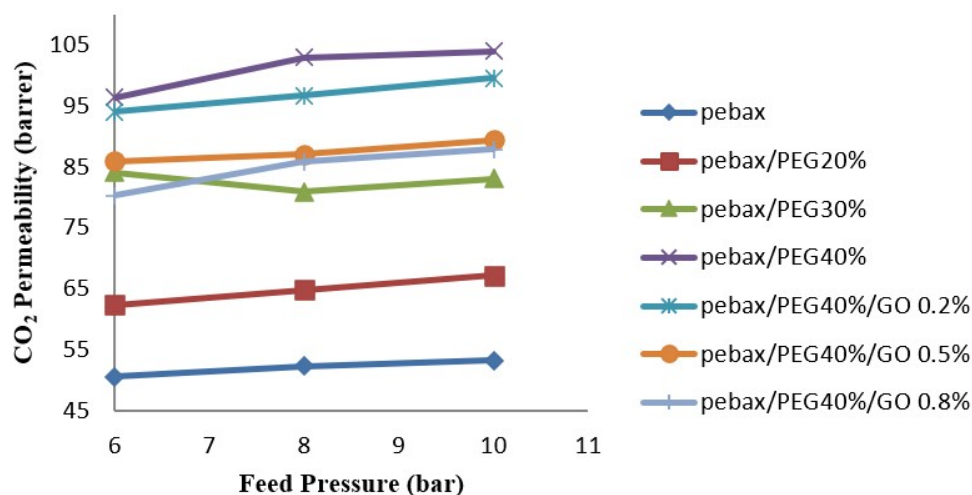


Figure 12 Influence of the feed pressure on the CO₂ permeability of different kinds of membranes

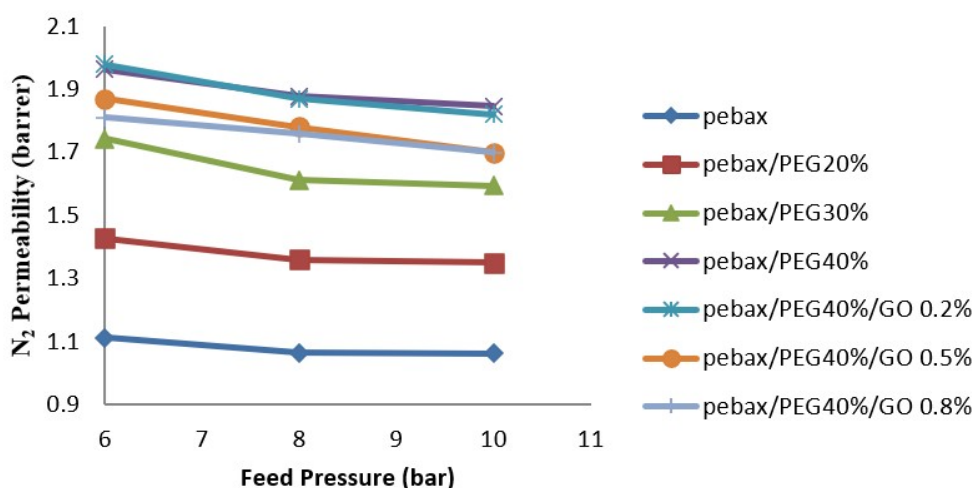


Figure 13 Influence of the pressure on the N₂ permeability of different kinds of membranes

Presence of polar groups in pebax and poly ethylene glycol increases affinity of CO₂ to the polymer matrix. In contrast, N₂ is nonpolar and does not interact with EO units like CO₂, thus the permeability of N₂ decreases with increasing feed pressure [5, 9].

It can be concluded that CO₂ based on its higher critical temperature and condensability and also smaller kinetic diameter compare to N₂, has a greater permeability [5, 10].

Based on XRD results, GO interacts with the polymer chains and increases the crystallinity of pebax and restricts the chain mobility, thus as presented in Figure 13, the increase in crystallinity

made the permeability of larger gas (N₂) to be decreased [9].

The effect of feed pressure, PEG and GO contents on the ideal selectivity of the membranes is shown in Figure 14. The increase in feed gas pressure enhances CO₂/N₂ selectivity in membranes. For example, at the feed temperature of 25 °C, increasing pressure from 6 to 10 bar provides further improvements in the ideal selectivity of CO₂/N₂, from 49.08 to 56.31 (pebax/PEG 40wt.%). This may be attributed to the increase in CO₂ sorption at high pressures [8].

With the increase in PEG content, the permeability of CO₂ and N₂

improves and the ideal selectivity of CO_2/N_2 exhibits an increasing trend. Therefore, it confirms that PEG addition to the pebax has been improved the membrane performance. Application of PEG and GO led to a modest increase in selectivity and displayed a favorable synergy effect on gas separation performance. PEG acted as a path way to increase permeability due to the excellent affinity between EO and CO_2 [6], and impermeable GO acted as a selective barrier to enhance higher selectivity [9]. It can be concluded that existence of PEG and the homogeneous dispersion of GO fillers in the polymer matrix increased the interaction sites between polymer and fillers, consequently an improvement of CO_2/N_2 selectivity is observed. The permeability results of CO_2 and N_2 gases and the ideal selectivity of CO_2/N_2 are given in Table 2.

The diffusivity and solubility coefficients of CO_2 and N_2 gases are summarized in Table 3 and 4, respectively. As shown in Table 3, the diffusion coefficient of both CO_2 and N_2 gases extend to a maximum for the blend pebax/PEG40wt%/GO0.2wt%. Added GO to the polymer matrix can be located between pebax polymer chains and led to the highly tortuous diffusion path which consequently reduced the permeability [9, 10, 26]. Moreover, as shown in Table 4, with the incorporation of 40% PEG the higher solubility coefficient for CO_2 while for N_2 reached a maximum for the neat membrane. The hydroxyl and carboxyl groups on GO and EO groups on PEG had a stronger interaction with CO_2 than N_2 , therefore there is an increase on CO_2/N_2 solubility selectivity [6, 9, 10].

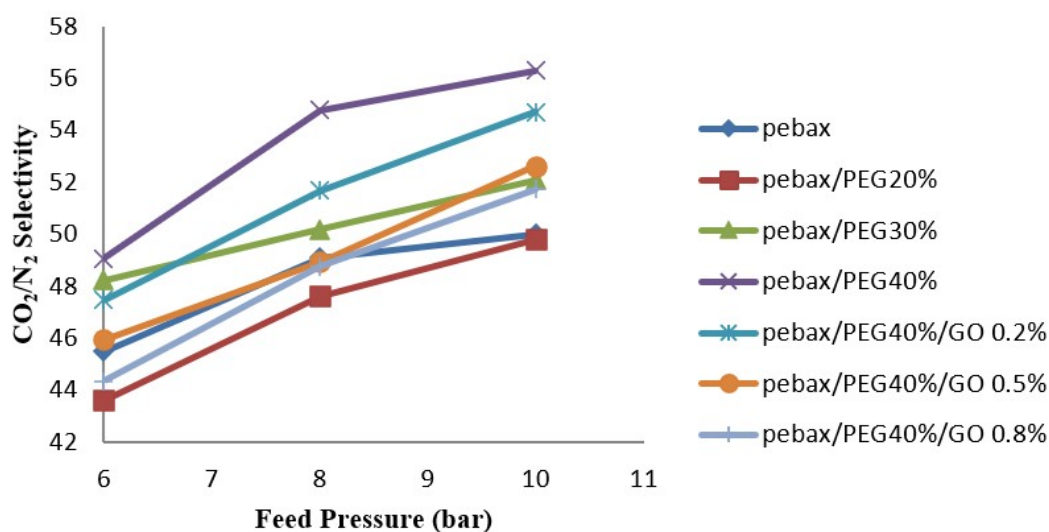


Figure 14 Effect of pressure, PEG and GO contents on the CO_2/N_2 selectivity of the different kinds of membranes

Table 2 Permeation properties of the prepared membranes

membranes↓	Permeability (Barrer) at 6 bar		Permeability (Barrer) at 8 bar		Permeability (Barrer) at 10 bar		Selectivity at 6 bar	Selectivity at 8 bar	Selectivity at 10 bar
	N ₂	CO ₂	N ₂	CO ₂	N ₂	CO ₂	CO ₂ /N ₂	CO ₂ /N ₂	CO ₂ /N ₂
gases→									
pebax	1.113	50.62	1.065	52.286	1.064	53.218	45.4807	49.0948	50.0169
pebax/PEG20%	1.429	62.34	1.36	64.78	1.35	67.22	43.6249	47.6324	49.7926
pebax/PEG30%	1.744	84.117	1.613	80.935	1.595	83.072	48.2322	50.1767	52.0828
pebax/PEG40%	1.964	96.392	1.879	102.941	1.846	103.949	49.0794	54.7849	56.3104
pebax/PEG40%/GO 0.2%	1.98	94	1.87	96.64	1.82	99.54	47.4747	51.6791	54.6923
pebax/PEG40%/GO 0.5%	1.87	85.9	1.78	87.1	1.7	89.44	45.9358	48.9325	52.6118
pebax/PEG40%/GO 0.8%	1.81	80.29	1.76	85.8	1.699	87.91	44.3591	48.75	51.7422

Table 3 Gas diffusivity coefficients of the membranes at 25 °C

Pressure	6 bar		8 bar		10 bar	
membranes	D _{N₂} ^a	D _{CO₂} ^a	D _{N₂} ^a	D _{CO₂} ^a	D _{N₂} ^a	D _{CO₂} ^a
Pebax	185.743	418.647	169.749	206.476	308.051	385.226
pebax/PEG20%	196.626	114.509	217.211	116.370	285.883	184.170
pebax/PEG30%	281.184	699.405	294.459	144.128	332.814	713.250
pebax/PEG40%	1021.826	988.046	652.166	2321.83	740.326	1589.106
pebax/PEG40%/GO 0.2%	1860.838	3720.812	4060.307	6053.184	886.747	1881.606
pebax/PEG40%/GO 0.5%	790.694	1229.348	615.705	449.554	562.635	1401.778
pebax/PEG40%/GO 0.8%	644.934	419.6413	465.420	768.599	839.239	1754.981

^a Diffusivity coefficient (μm²/s)**Table 4** Gas solubility coefficients of the membranes at 25 °C

Pressure	6 bar		8 bar		10 bar	
membranes	S _{N₂} ^a	S _{CO₂} ^a	S _{N₂} ^a	S _{CO₂} ^a	S _{N₂} ^a	S _{CO₂} ^a
Pebax	0.0059	0.1209	0.0063	0.2532	0.0035	0.0048
pebax/PEG20%	0.0057	0.0976	0.0063	0.5567	0.0047	0.0047
pebax/PEG30%	0.0051	0.1203	0.0055	0.0443	0.0048	0.0035
pebax/PEG40%	0.0017	0.5444	0.0029	0.5615	0.0021	0.0021
pebax/PEG40%/GO 0.2%	0.0011	0.2240	0.0030	0.1258	0.0025	0.0025
pebax/PEG40%/GO 0.5%	0.0025	0.0699	0.0004	0.1937	0.0030	0.0030
pebax/PEG40%/GO 0.8%	0.0029	0.0216	0.0038	0.0142	0.0020	0.0501

^a Solubility coefficient [μm³(STP)/(μm.bar)].

3.4.4 Performance Comparison of Pebax-based MMMs

Table 5 compares the performance of the present study and similar investigations in other literatures. In the present work, appropriate CO₂/N₂ ideal

selectivity has been achieved by adding PEG and GO fillers.

In Figure 15, the optimum gas permeability performance is represented by Robeson's 2008 the upper bound line, it is very close to the line. This indicates that the CO₂

permeability and CO₂/N₂ selectivity are improved. The blend membrane doped with 40 wt.% PEG displays the optimum performance with a CO₂

permeability of 103.95 Barrer and CO₂/N₂ ideal selectivity of 56.31.

Table 5 Comparison of the present investigation with other relevant studies

membranes	Temperature & Pressure	CO ₂ Permeability Barrer	Selectivity CO ₂ /N ₂	References
Pebax (1657)/1 wt.% Sulfocalix [6] arene	25 °C & 6 bar	224.6	66.3	2
Pebax (1657)/80 wt.% PDMS-PEO-600	25 °C & 2 bar	1629	32.1	3
Pebax (1657)/10 wt.% PEG (200 g/gmol)	35 °C & 4 bar	127	52.6	6
Pebax (1657)/10 wt.% PDMS-PEG (200 g/gmol)	35 °C & 4 bar	179	48	6
Pebax (1657)/30 wt.% PEG (250-500 g/gmol)	30 °C & 0.3 bar	606	43	7
Pebax (1657)/10 wt.% PEG (600 g/mol)	25 °C & 2 bar	179	52.3	8
Pebax (1657)	35 °C & 7 bar	126.8	56.2	9
Pebax (1657)/GO	35 °C & 7 bar	38.3	54.8	9
Pebax (1657)/10 wt.% PEG-PEI-GO	30 °C & 2 bar	1330	45	10
Pebax (1657)	25 °C & 1 bar	76.2	58.44	11
Pebax (2533)	25 °C & 1 bar	257	28.23	11
Pebax (1657)/SSMMP-20%-[bmim][Tf ₂ N]	25 °C & 4 bar	192.2	68.3	12
Pebax (2533)	25 °C & 2 bar	351	35.1	15
Pebax (2533)/35 wt.% ZIF-8	25 °C & 2 bar	1287	32.2	15
Pebax (1657)/3 wt.% MoO ₃	35 °C & 10 bar	209.5	245	16
Pebax(2533)/10 wt.% Nitrogen-doped porous carbon	25 °C & 4 bar	457	64.7	18
Pebax (1657)/0.8 wt.% Imidazole graphene oxide	25 °C & 8 bar	76.2	105.5	22
Pebax (1657)/8 wt.% PVC + silica nano particles	25 °C & 1 bar	29	76	23
Pebax (1657)/0.1 wt.% A-prGO	25 °C & 4 bar	47.5	105.6	26
Matrimid/5 wt.%GO/5 wt.%CNT	30 °C & 2 bar	38	81	27
Pebax (1657)/8 wt.% SiO ₂	25 °C & 2 bar	-	137	28
Pebax (1657)/15 wt.% Glycerol	25 °C & 10 bar	50.42	222.7	29
Pebax (1657)/15 wt.% Glycerol + 1.5 wt.% Cu	25 °C & 10 bar	63.6	200	29
Pebax (1657)/40 wt.% PEG (600g/gmol)	25 °C & 10 bar	103.95	56.31	This study
Pebax (1657)/40 wt.%PEG(600g/gmol)/0.2 wt.%GO	25 °C & 10 bar	99.54	54.69	This study

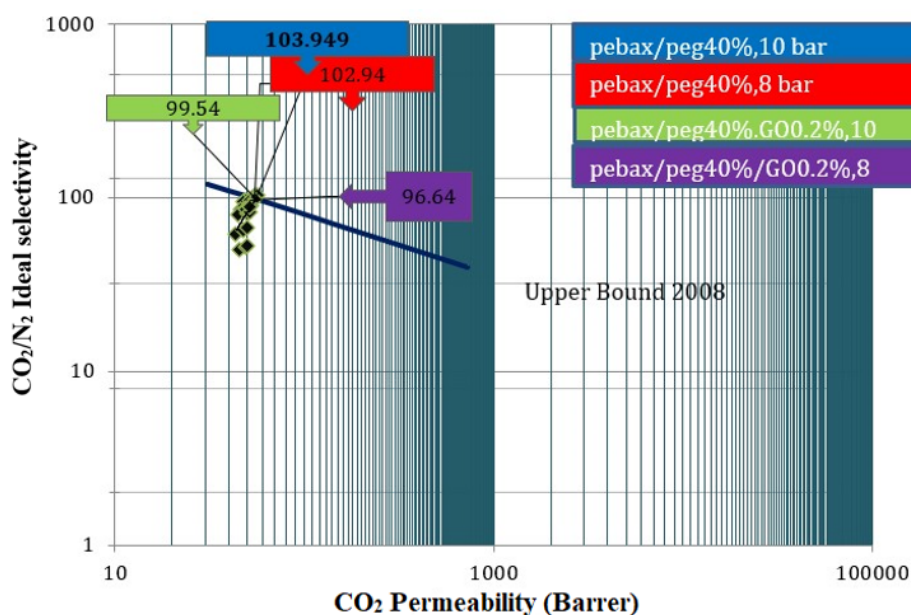


Figure 15 Robeson plot for CO₂/N₂ separation

4.0 CONCLUSIONS

Nano composite membranes were fabricated by incorporating poly ethylene glycol (PEG) and graphene oxide (GO) into the pebax matrix and their CO₂ and N₂ permeation properties were investigated. The influence of incorporation of GO fillers and PEG into the polymer matrix was studied. Membranes were prepared with the solution casting method. SEM images showed that the membrane structure is without any defects and the neat PEBAX membrane is much smoother than that of blended. As confirmed by XRD analyses, both blended and nano composite membranes showed similar XRD patterns. No characteristic peak of GO was observed in nanocomposite membrane which indicated that GO was fully exfoliated and dispersed in the polymer matrix properly. The permeability results of pure N₂ and CO₂ showed the simultaneous increase of CO₂, N₂ permeability and selectivity in PEBAX/PEG blend membranes due to the excellent affinity between EO and CO₂. Meanwhile, nano composites

comprising GO had lower permeability. The difference in size and solubility of CO₂ and N₂ gases is the reason for excellent CO₂ permeability and CO₂/N₂ selectivity by the prepared membranes. An increase in the feed pressure provides higher permeability of CO₂ and CO₂/N₂ selectivity. Pebax/40 wt.% PEG exhibited best performance with a CO₂ permeability of 103.949 Barrer and CO₂/N₂ ideal selectivity of 56.3104 at 10 bar and room temperature conditions.

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CONFLICTS OF INTEREST

The authors declare that there is no conflict of interest regarding the publication of this paper.

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