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Journal of
Applied
Chemical
Research
jacr.kiau.ac.ir

Journal of Applied Chemical Research, 17, 2, 26-40 (2023)

Performance Investigation of the Membrane PEEK/PVA Modified with ZnO Nanoparticle for CO_2 Separation

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(Received 17 Aug. 2022; Final revised received 10 Nov. 2022)

Abstract

The characterization of morphological, particle size, mechanical, and thermal on the impact of membrane Polyetheretherketon/Polyvinylalcohol (PEEK/PVA) composite was modified with zinc oxide nanoparticles for Permeability and Selectivity of CO_2 properties. Separation process was done at high pressures since the selectivity of CO_2 boosted from 94.0 at 1 bar with increasing feed pressure. Also, the permeability of CO_2 for membrane PEEK/PVA/ZnO nanoparticle modified with PEEK/PVA composite was performed. The increase in CO_2 permeability to the membrane PEEK/PVA from 26.4 to 82.6 occurred upon modifying the surface area of the membrane PEEK/PVA/m-ZnONPs (1.0 wt%, 2.0 wt%, and 3.0 wt%), and increasing the percentage of this additive in the polymer matrix. The increase was due to the presence of cavities developed in the process of surface modification, and the increased of free volume and solubility of CO_2 inside the polymer membrane.

Keywords: CO_2 gas, Membrane, Permeability, Selectivity, Separation.

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Introduction

In recent years, gas separation is progressively performed by separation methods such as absorption/adsorption, and cryogenic distillation by novel technologies like membrane systems [1-4]. Moreover, industries are seeking novel methods to tackle environmental pollution, energy consumption, and costs. Therefore, the use of membrane approach in industries like natural gas sweetening got the better of other techniques [4,5]. Over several years, this technology, in comparison with other conventional gas separation processes, faced rapid growth [6]. The matter of gas separation in many professions and different applications is of significant importance [7,8].

Separation processes have drawn the attention of various industries due to their advantages of requiring lower energy, having compact structure, having lower operating and maintenance costs, being user-friendly, most important of all, having the most negligible impact on the ecosystem [9,10].

The efficacy of metal nanoparticles on the separation of CO_2 pure gas for two nanocomposite membranes of polyesters has been studied. The net gas permeability test illustrates that the permeability improves along with the rising silica content. This might be related to the increase in the free volume of the polymer network caused by the separation of the molecular chain [11]. Reported polysulfone (PSF) MMMs with mesoporous silica MCM-41 for gas separation. gas permeability of PSF is enhanced by incorporating mesoporous MCM-48. Researchers showed that mesoporous materials increased the permeability of the polysulfone MMMs without decreasing selectivity due to good compatibility with the polymer matrix. Shimizu et al. reported that the addition of amine functionalized SBA-15 was effective for enhancing CO_2/ CH_4 selectivity [12]. They studied the combination of polyimide and zeolite as a polymer membrane. For this object, they prepared a mixed network membrane made up of polyimide and 20 wt% zeolite and then analyzed the permeability data for gases including O_2 ,CO_2, N_2 and CH_4. The permeability of N_2 and O_2 gases decreased, but it increased for CH_4 and CO_2. This change in permeability is strongly influenced by the changes in the permeability coefficient.

Matteucci et al studied the permeability of CH_4 ,N_2 and CO_2 by adding TiO_2 nanoparticles to poly (1-trimethylsilyl-1-propyne) (PTMSP). Results showed that permeability of these gases increased more than 4 times in comparison with the pure polymer [13]. Also, they studied the effect of TiO_2 nanoparticles on 1, 2-polybutadiene (PB) in another research. Obtained results showed that in a membrane containing 27 vol.% TiO_2 nanoparticles, permeability coefficients of CO_2, CH_4 ,N_2 and H_2 gases increased 3 times in comparison with the pure polymer.

Furthermore, the solubility coefficient of the gases increased by adding the nanoparticles to the polymer, while the permeability coefficients decreased by addition of the nanoparticles [14]. Statistical modeling has been utilized as a method to evaluate the effect of various variables including adding nanoparticles zinc oxide (ZnO), aluminum oxide (Al_2O_3), titanium dioxide (TiO_2) to PMP membrane on both structural characteristics of the membranes and performance of the polymeric membranes [15]. Recently, blending polymers with inorganic nanomaterials has occupied an impressive platform of research because of their simplicity, stable performance and mild operating conditions [16].

ZnONPs amongst different nano-materials have grown in popularity due to their availability, photocatalytic activity, self-cleaning quality, ease of preparation, and affordability. At higher concentrations, ZnO results in the formation of agglomerates. Therefore, procuring a hybrid with fine NPs dispersion is a challenge [17,18]. The construction, of agglomerates is because of the high surface energy of ZnO, which provides a driving force for developing of particle bonding.

The most appropriate method to attach a proper organic group on the surface atoms is silane coupling agents [19,20]. This surface modification not only supports the NPs, against accumulation but also helps their compatibility with the other phase [21]. Poly ether block amides are composed of two different parts of polyethylene oxide as amorphous or soft phase and polyamide part as crystalline or hard phase. In other words, these polymers have an amorphous part that causes the permeability of ions, gases and metals, as well as adhesion between additives and the polymer, and the crystalline part provides the task of creating mechanical resistance in the structure of the polymer [22]. Therefore, the use of polyether ether ketone membranes in the treatment of oily wastes Polyether ether ketone and polyether imide membranes were prepared by phase inversion method [22]. This unique characteristic makes it possible to fabricate a layer of hydrophilic polymer brushes on PEEK surface via ultraviolet (UV)-initiated graft polymerization, and preparing Polyetheretherketon/Polyvinylalcohol nanocomposite modified with zinc oxide nanoparticles synthesis especially as an effective membrane and alternative to exorbitant or noxious membranes for the elimination of gases from natural gas attracted our attention [23,24].

In this study, a new macromolecular was generated based on the membrane PEEK/PVA/ZnO nanoparticle. Then, the functionalized ZnO nanoparticle (1.0 wt%, 2.0 wt%, and 3.0 wt%) into membrane PEEK/PVA /ZnO nanoparticle modified with PEEK/PVA composite. A detailed examination of the contact membrane PEEK/PVA/ZnO nanoparticle as an important parameter on the permeability and selectivity of CO_2 gas and the adsorption mechanism. That the CO_2 selectivity and permeability of the mixed matrix membranes by the membrane PEEK/PVA/ZnO nanoparticle. Sorption studies, FE-SEM, and gas permeation measurements to characterize the structure, and properties of the mixed matrix membranes were based on membrane PEEK/PVA/ZnO nanoparticles.

Experimental

Reagents and materials

Merck Chemical Co and Aldrich Chemical Co provided us with all the materials, and solvent n-methyl pyrolidine (n-MP) were provided and used Co. (Tehran, Iran) provided us with the ZnO nanoparticles with the average particle size of 25–30 nm. To guarantee complete drying, ZnO nanoparticles were dried at 500°C for almost five h before utilization. The coupling agent, Poly ether ether ketone (PEEK) from Chines Company, and membrane PEEK/PVA/ZnO nanoparticle modified with Polyvinyl alcohol (PVA) from Merck Company.

Instrumentation

Scanning electron microscopy (FESEM: MIRA III, TESCAN Company, of the Czech Republic) under an acceleration voltage of 26kV helped us in analyzing the morphology of samples. With the help of infrared spectroscopy (FTIR, Nicolet Co, USA), the functional groups on the surface of zinc oxide modified nanoparticles were determined. Instron (6025K) to do the tensile test of nanocomposites was based on the ASTM-D3039 standards at room temperature on specimens 150 mm long, 25 mm wide, and 3 mm thick, with a machine speed of 5 mm/min. Also, the measurement of flexural strength was done according to ASTM-D790 standard using instron (6025K) on specimens 90 mm long, 10 mm wide and 3 mm thick.

Synthesis of membrane PEEK/PVA/ZnO nanoparticle

A soluble casting method was employed to fabricate the separation membranes. Therefore, a few grams of membrane and a few ml of n-methyl pyroledine (n-MP) solvent were mixed and stirred for 3 hours through the instrumentality of a mechanical stirrer which ended in a uniform solution. They provided a homogeneously composite membrane with different quantities of m-ZnO NPs (1.0 wt%, 2.0 wt%, and 3.0 wt %). First, 0.1 g of polymer was dissolved in 2 mL n-methyl pyroledine (n-MP) solvent and second, under constant stirring for

12 h and at 25°C, m-ZnO was separately dispersed in 2 mL n-methyl pyroledine (n-MP) solvent. Then, to obtain the desired weight percentages of m-ZnO from 1.0% to 2.0%, the solutions were mixed. The provided membrane PEEK/PVA/ZnO nanoparticle solutions were taken into glass Petri dishes to remove the solvent, and afterward they were dried under vacuum at 60°C for 12 h and 130°C for 3h, membrane PEEK/PVA/ZnO nanoparticles, with Polyetheretherketon and Polyvinylalcohol (PEEK/PVA) composite modified zinc oxide nanoparticle.

Procedure of the permeability of gas by membrane

The dissolution-permeation mechanism results from the difference in thermodynamic activity in the upstream and downstream faces of the membrane (Figure 1). The difference in activity causes the difference in concentration, which leads to penetration in the direction of reducing activity. The difference in the permeability of different gases is caused by the difference in mobility and the mutual influence of the physical chemistry of gas and polymer.

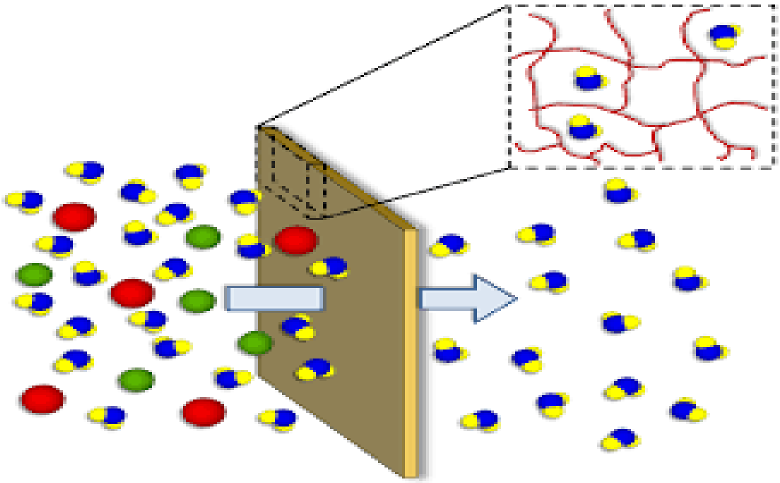

Figure. 1. Schematic representation of the permeability of gas by membrane.

Measurement and calculation method

With the help of a constant pressure method (a soap bubble discharge gauge), the permeability of CO_2 in the membrane PEEK/PVA/ZnO nanoparticle was obtained. The trial entailed a gas seepage cell in which one to the membrane and the other side to the gas feed. By making use of a regulator, and using, a barometer. The following formula to determine the gas permeability in the membrane was used [25,26].

$$\frac{P_i}{l} = \frac{1}{A\Delta P}\frac{dv_i}{dt} \qquad (1)$$

Pi in the aforementioned formula stands for the permeability of gas i, the volume of gas discharged into soap bubble discharge or volume of Permeability gas into the membrane in cubic centimeters is shown by vi, effective membrane surface area in square centimeters by A, t refers to bubble motion the column in seconds, and 1P shows the pressure drop across the membrane in centimeters of mercury. In GPU, reports on the asymmetric membranes developed from the permeability division of the membrane thickness are available. Also, via using the following equation, the optimal selectivity of the two gases is calculable:

$$\alpha_{i/j} = \frac{\left(\frac{P_i}{l}\right)}{\left(\frac{P_j}{l}\right)} \qquad (2)$$

Where α (i / j) is the optimal selectivity of gas i over j, Pi / l is the gas i permeability, and Pj / l is the gas j permeability. The excessive dependence of gas transfer properties of the organic/inorganic lattice membranes to the membrane structure is beyond doubt. In contrast, the membrane structure is dependent on the surface area of contact between the particles and the polymer.

Absorption theory expresses the permeability of gas in glass polymers [27]. This approach, introduces the total concentration of permeates (C) in the glass polymer as a function of pressure (p). The following equation expresses the theory:

$$C= k_D p+C_H bp/(1+bp) \qquad (3)$$

In the abovementioned mathematical statement, K_D stands for the dissolution coefficient of artistic law, and CH shows the cavity saturation constant. Taking into considering the large porosity-to-surface ratio in the porosities. That a place for the interaction between gases, and the polymer network is required and the porosities can provide it, which provokes an increase in the permeability and solubility [26,27].

Investigation of permeability characteristics and selectivity of permeability measurement membranes

The pressure-volume-constant-variable method can be utilized to verify the permeability prosperities of carbon dioxide, and methane gases through polymer membranes. Taking into consideration the steady-state conditions, the polymer membranes in this method for a specific duration of time were pressurized and with the help of a digital flow meter, the volume flow rate through the polymer membrane. The effective surface area of all membranes was estimated to be 56/71 cm^2. The developed polymeric membrane in the membrane tubes, and with the help of a hollow circular rubber ring, was maintained. A gas feed inlet and two outlet portions in the membrane tube. From the two outlets, one is for the gas component leaked through the membrane and the other for the residual gas component. Figure 2, represents the schematic of the devices applied to measure the leakage of carbon dioxide from the polymer membrane [21].

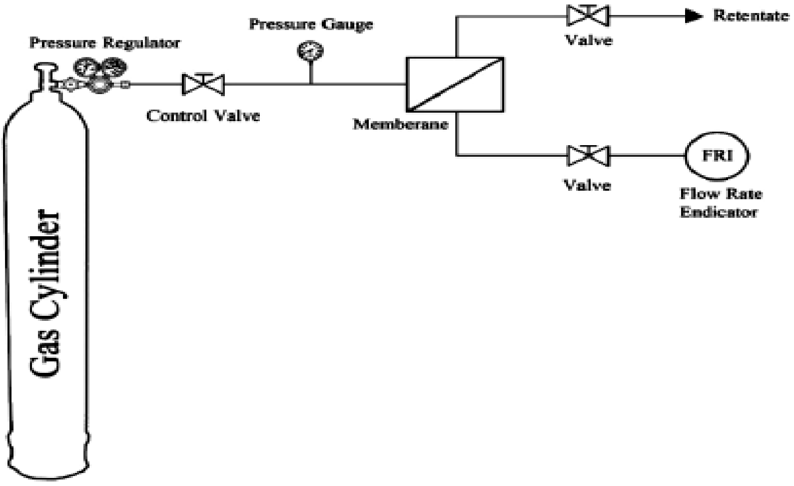

Figure. 2. Schematic permeability gas of the mixed matrix membranes.

Results and discussion

Examination of ZnO Surface Modification Process

Figure 3 represents the FTIR spectra of both unmodified and modified ZnONPs. The curve refers to the FTIR spectrum of membrane PEEK/PVA fundamental, by ZnONPs and represents that of the membrane PEEK/PVA/ZnONPs coupling components. In scrutiny, a strong signal in the range of 1005-1345 cm^{-1} was detectable, which could result from the vibration of the hydroxyl groups on the surface of the nanoparticles [18].

Considering the b curve, it became apparent that the signals in the range 1300-1655 cm^{-1} were associated to the groups CH –CH and $(-CH_2)n,-CH_3$. Therefore, that the alphabetic groups of the components related onto the surface of the nanoparticles were Zn-O. The signal of the 2850 cm^{-1} region to the b-curve of the bond C=C at the component level. Consequently, the thriving coupling of coupling groups to the surface of zinc oxide nanoparticles by the FTIR spectrum. In other terms, surface modification was successfully performed [18,26].

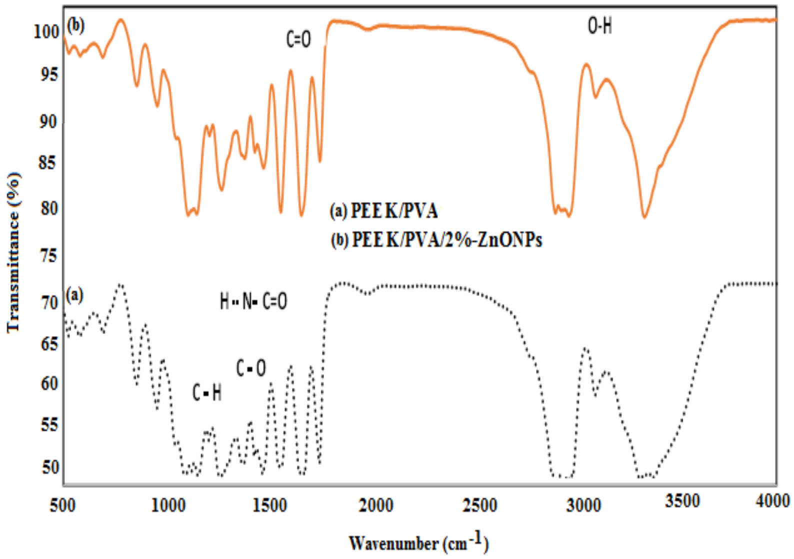

Figure 3. (a) FT-IR spectra of the aromatic membrane PEEK/PVA (b) FT-IR spectra of the membrane PEEK/PVA/ZnO nanoparticle.

X-ray diffraction

In Figure 4, the XRD patterns of PEEK/PVA, and PEEK/PVA/ZnONPs. It is apparent that there is a lack of diffraction signal in the range of 2θ angle barring one crystalline signal suggesting the presence of a small proportion of crystalline phase. In Figure 4, ZnONPs the apparent signals are at 100, 002, 101, 102, 110, 103, 200, 112, 201, and 202 which prove the presence of a rutile phase as good evidence of the crystalline form of this compound. The characteristic signals of ZnONPs and PEEK/PVA in the XRD patterns revealed that the preparation process and surface modification could not change the morphology of NPs [18,28].

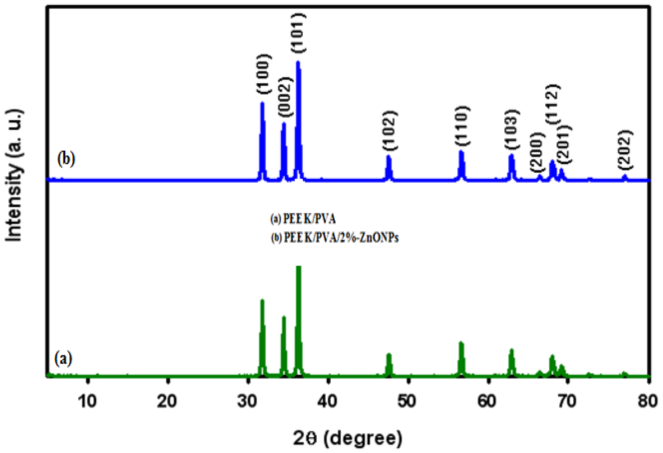

Figure 4. XRD patterns of the surface ZnO nanoparticles (a), membrane PEEK/PVA/2.0% ZnO nanoparticles (b) membrane PEEK/PVA/1.0% ZnO nanoparticles (c) of membrane PEEK/PVA (d).

Morphology of prepared membranes

In SEM images, filamentous fibers exist on the pure membrane surface. Also in Figure 5, SEM images of membrane PEEK/PVA/ZnO nanoparticles display the result of this modification on the morph of the provided membrane.

Surface structure of the membrane PEEK/PVA/ZnO nanoparticles, metamorphosed into pseudo-flowering from fibres. Additionally, in this image, the transparent layers on the membrane surface confirm the hardening of the polymer-particle interface an indistinct separation when the modification was performed [29,30]. The nanoscale size and rounded shape of the m-ZnO nanoparticles have been shown in these images to be in the range of 40–60 nm.

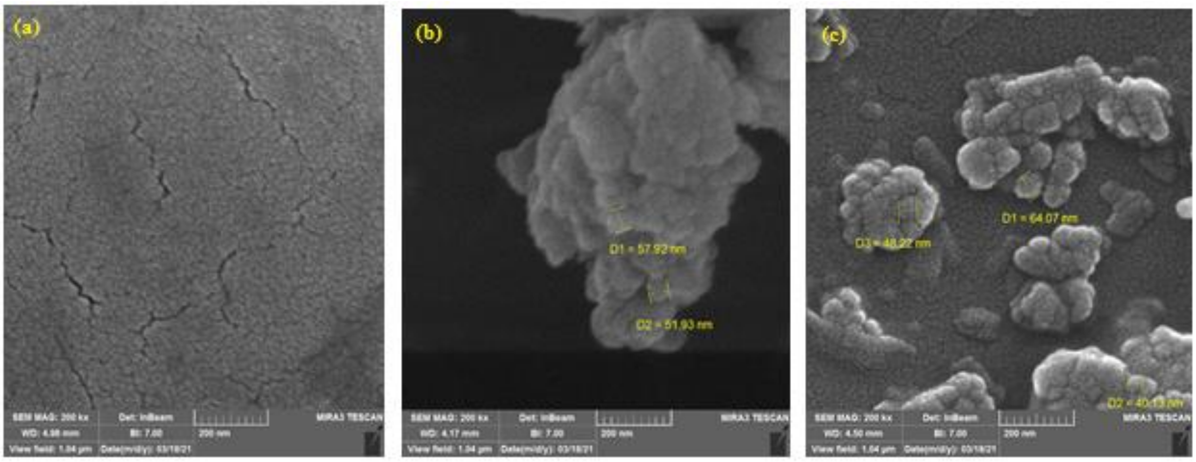

Figure 5. SEM images of Surface (a) membrane PEEK/PVA (b) ZnO nanoparticles (c) membrane PEEK/PVA/ZnO nanoparticles.

Outcomes of the stretch test

The outcomes of tensile strength in are seen in Figures 6 and 7, and those of the elongation to fracture of the specimens in the tensile test are demonstrated. As displayed in Figure 6, with each weight of the filler phase, the tensile strength increases, and with each type of membrane PEEK/PVA, the weight percent of the filler phase increases. Membrane PEEK/PVA unmodified with zinc oxide nanoparticles and modified zinc oxide nanoparticles were analyzed. However, at the same weight percent, the membrane PEEK/PVA modified with ZnO nanoparticles showed greater tensile strength, possibly because of lower interaction between membrane PEEK/PVA and the polymer phase in the modified ZnO nanoparticles [31].

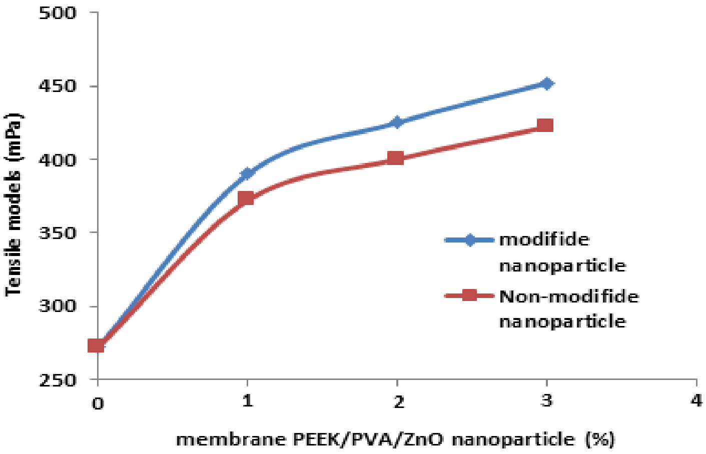

Figure 6. Influence of composition of different percentages and types of Filler ZnO nanoparticles on tensile models of membrane PEEK/PVA.

The impact strength test

The size of the particles and their distribution and the level of interaction between the filler particles and the matrix phase, are prerequisites for the achievement of the desired behavior [32].

In Figure 7, the bending test results of the specimens can be seen. As evident, any rise in the weight percent of the nanoparticles triggered an increase in the impact strength of both models. However, this increase was remarkably apparent in the case of modified ZnO nanoparticles reinforced. In sum, in the case of unmodified zinc oxide nanoparticles, agglomeration and bigger particle sizes can produce stress-focused areas and weaken the interaction between the nanoparticles and the polymer chains, all of which lower the impact strength of the membrane PEEK/PVA [33].

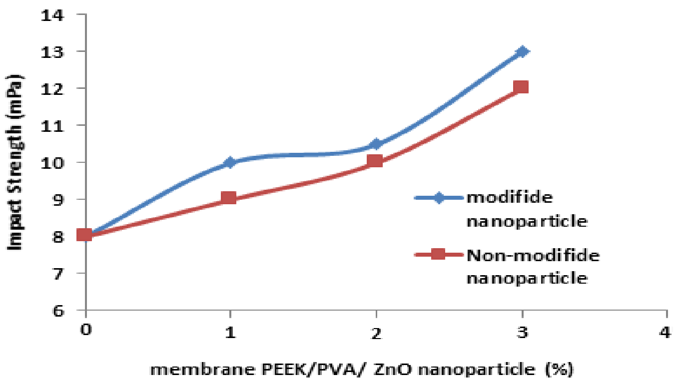

Figure 7. Influence of composition of different percentages and types filler ZnO nanoparticles on impact Strength of membrane PEEK/PVA.

Investigation of permeability properties and selectivity

The permeability and selectivity of carbon dioxide for membrane PEEK/PVA/ZnO nanoparticles. The following diagram demonstrates the influence of these additives on the permeability of carbon dioxide. The surface modification with PEEK/PVA composite and expansion of the percentage of this additive in the polymer matrix can increase the carbon dioxide permeability to the membrane.

With increasing feed pressure, the permeability of CO_2 increased from 94.1 at 1 bar pressure to 42.0 at 8 bar pressure, also, the permeability and selectivity of CO_2 for membrane PEEK/PVA/m-ZnO nanoparticles. The CO_2 permeability to the membrane PEEK/PVA increased from 26.4 to 82.6 upon modifying the surface area of the membrane PEEK/PVA/m-ZnO NPs (1.0 wt%, 2.0 wt%, and 3.0 wt%), and increasing the percentage of this additive in the polymer matrix. Carbon dioxide has the property of softening the membrane and breaking the polymer chains apart membrane polymers, the other hand, it removes the separation. The determining factor in the dissolution-diffusion process is the degree of gas condensation. The determining factor in the dissolution-diffusion process is the degree of gas condensation. Therefore, the high condensability of carbon dioxide has increased its permeability. Figure 9 shows the membranes selectivity of the relative to carbon dioxide [33,34]. Thus, the recognition of the selectivity of CO_2 is in (Table 1). The perfect selectivity of the prepared membranes, as well as the unique properties of the membranes based on the copolymer, is shown in Figures 8 and 9 [35-37].

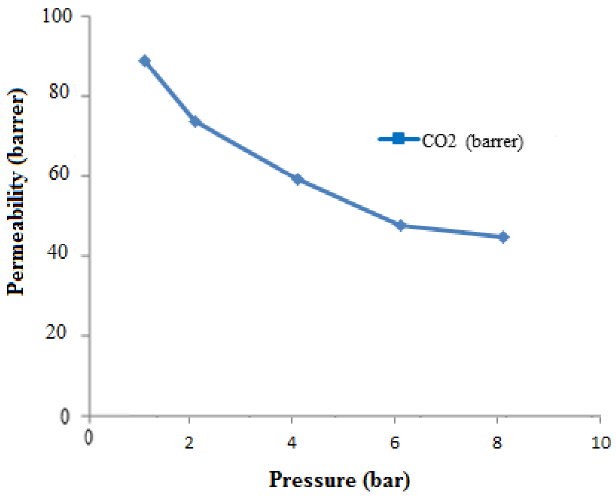

Figure. 8: The display of impact Permeability (barrer) CO_2 gas of the modified ZnO nanoparticles without Pressure percentage.

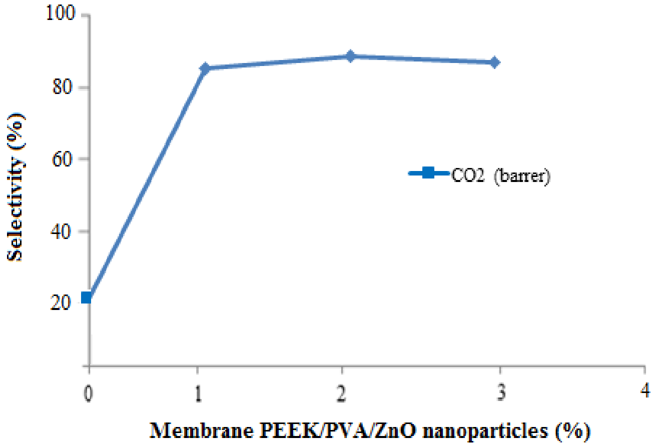

Figure 9. The display of impact selectivity CO_2 gas of the modified ZnO nanoparticles at Pressure (1bar) percentage.

Table 1. Separation outcomes for membrane PEEK/PVA/m- ZnO nanoparticles.

Pressure (bar)	**Permeability (barrer)** CO_2	**Selectivity**	**membrane**
1	94.0	26.4	membranes PEEK/PVA/ZnO nanoparticles
2	78.0	33.15	
4	60.2	77.8	
6	48.0	80.1	
8	42.0	82.6	

Comparison of results for this work with other reported membranes

In order to illuminating the applicability and efficiency of membranes in this work, the results are compared to those of some of the recently reported methods for the reduction of variety of materials by membranes in (Table 2). Obviously, SPEEK/PVA/ZnO nanoparticles show the high Permeability for the gas CO_2 in comparison with literature other membranes.

Table 2. Comparison of results for this work with other reported membranes.

Samples	Membranes	Pressure (bar)	Permeability CO_2 (%)	References
CO_2/CH_4	Silicate-based MMM	2	73.6	[2]
CO_2/CH_4	TPX/AL_2O_3	8	71.73	[5]
CO_2	PEBA/MWCNT	8	92.5	[13]
CO_2/CH_4	PEEK/Zeolite 4A	10	55.0	[14]
CO_2/N_2	PVA/PEG 600	1.5	52.9	[17]
CO_2	PEBA/PEG/CNT	2	44.6	[33]
CO_2	PVA/CNF	8	39.0	[37]
CO_2	PEEK/PVA/ZnO NPs	1	94.0	This Work

Conclusions

The permeability of CO_2 gas for membrane PEEK/PVA/m-ZnO nanoparticles has been investigated at different pressures. The findings revealed that the direct relation of the permeability of CO_2 with pressure was relative to the elasticity of the polymer membrane. In other words, the tensile strain could affect the permeability of polar CO_2 gas at high pressures. When high concentrations of CO_2 gas were absorbed onto the membrane surface (sufficient concentration of CO_2 in the membrane), elongation was observed. Therefore, with an expansion in the solubility of the gas molecules within the polymer matrix, the CO_2 permeability increased. This increase is the result of the presence of cavities due to the increase of free volume and the increase of solubility of carbon dioxide inside the polymer membrane. However, the membranes were high permeable to CO_2. The results showed the excellent permeability of the prepared membranes as well as the unique properties of the membranes based on the membrane PEEK/PVA/m-ZnO nanoparticles.

Acknowledgement

The authors would like to acknowledge and thank the partial support of the Islamic Azad University, Ayatollah Amoli Branch, Iran.

Conflict of interest

The authors declare that they have no conflict of interest related to the publication of this article.

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