



A Short Review on Separation of H₂/CO₂ using Polymer Membrane

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ABSTRACT

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Polymer is the least expensive and effectively prepared material for membrane manufacture. They demonstrate extraordinary adaptability in their piece and change is a lot simpler that settle on them a superior decision. Membrane gas separation gives numerous advantages over regular procedure in separating H₂/CO₂ either as an independent procedure or incorporated procedure with water-gas shift reactors. Membrane technology involves low speculation cost, working at high pressures and temperatures, lower energy cost as well as high weight retentate CO₂ is transported and put away without the pressure. Materials for gas separation membrane may be delegated metallic (metals or metal compounds), carbon, natural polymers and inorganics (for example zeolite and silicates). Individual class of membrane shows superior properties, however, there are inadequacies in either economy or manufacture techniques.

Keywords:

Membrane for gas separation;

Membrane technology; annealing

condition; operating temperature

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1. Introduction

Membranes with CO₂ permeation and catch at high temperatures with low energy have turned into a significant matter to obtain hydrogen [1,2]. Current procedures utilized for CO₂ permeation need gas to be cooled to surrounding temperatures. The advantage of utilizing membrane could fundamentally change the procedure economy. This will be finished utilizing membrane that is steady at high pressures and temperatures. In procedures intended in separating hydrogen, nonporous polymer membranes usually consist of hydrogen concentrated item stream, the more prominent measures of outstanding gases typically in the retentate. Gases such as O₂, CH₄, CO, and CO₂ normally acts as contaminations in procedure of hydrogen separation. Low hydrogen basic temperature (-240 °C) means that lower solvency of this gas. Low solvency joined with little motor distance across (0.289

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μm) lead to interest for diffusivity specific membranes for this application. Along these lines, the present hydrogen specific membrane is manufactured in the point of expanding the hydrogen diffusivity [3]. Lustrous polymer is the most often utilized materials for making hydrogen particular membrane tasks [4-6]. As example, Figure 1 demonstrates that H_2/CO_2 selectivity can be expressively improved by chemical cross-linking of PBI in solid state, in dissimilar distinction with the previous works where cross-linking PBI in solutions reduced H_2/CO_2 selectivity [7].

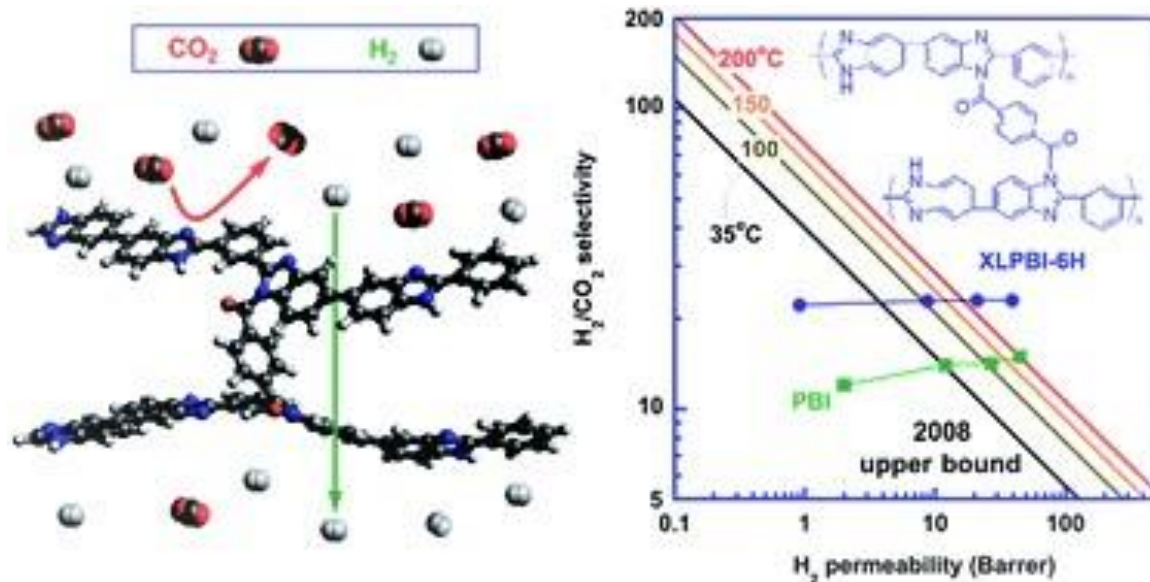


Fig. 1. H_2/CO_2 selectivity improved by chemical cross-linking of PBI in solid state [7]

Aliphatic polybenzimidazole is first designed by Robeson and Brinker as mention by Chung, Tai-Shung [8]. After two years, Vogel and Marvel [9] orchestrated fragrant polybenzimidazole (PBI) polymer. In the wake of taking consideration from both scholarly world and industry on account of its high warm and mechanical protections, different PBI's are developed. In any case, just a couple of incorporated polymers demonstrated desired properties. To get a thick polymer membrane dissolvability of the polymer is a fundamental matter. Poor solvency is one of the disadvantages of PBI polymer. It is dissolvable in like manner natural solvents only at high temperatures.

Biggest choice of polymers in separating H_2/CO_2 is examined by Orme and colleagues [10]. The researchers attempted to locate an appropriate material that can hinder the transport of CO_2 and provides high hydrogen movements. Among numerous polymers such as polystyrene, polysulfone, poly (vinylidene fluoride) and poly (methyl methacrylate) provide the best execution at 30 °C. Selectivity of the chosen membranes are 2.0, 2.3, 4.0, 2.0 separately. They inferred that the polystyrene is the best material. It shows the best blend for both high permeability and selectivity. Polystyrene, which is used commercially as a membrane polymer material due to its relatively high mechanical properties, high chemical resistance, and high T_g .

Xu *et al.*, [11] inspected the exhibition of fluorine comprising poly(arylene ether)s with huge and cumbersome diphenylfluorene moieties (FBP/6FPT and FBP/6FPPr). The selectivity for H_2/CO_2 is quite low; for FBP/6FPPr membrane and FBP/6FPT membrane which are 1.54 and 1.67 respectively. In any case, the striking point in the research is the high diffusivity selectivity which just about 100 for gas pair of H_2/CO_2 . Large selectivity is the aftereffect of poor solvency selectivity.

Hosseini *et al.*, [12] attempted to show the polyimide polymer performance as a gas separation membrane. Penetrability coefficients for carbon dioxide and hydrogen are 7.00 and 27.16 Bar respectively at 35°C for thick polymer membrane with selectivity of 4.0. Klaehn *et al.*, [13]

contemplated on changing PBI by N-substitution the imidazole nitrogen. Two changes are connected on polymer; silane and natural mixes substitution. Atomic loads of new polymers are higher than unsubstituted PBI. Utilizing little atom natural mixes sub-atomic weight expanded radically. It was added to the crosslinking. Acquired polymers are solvent in tetrahydrofuran. Be that as it may, they couldn't get attractive membrane development characteristics, since membranes are cracked. PBI shows a particular H₂ separation from different gases present in changing blend. In any case, the polymeric material with thick structure anticipates high gas permeability. In defeating the basic downside of this polymer, originating from intermolecular hydrogen holding and chain unbending nature endeavours are carried out. These incorporate creations of carbon membrane or composite membrane, getting ready supported membrane, also membrane basic adjustments [2,14-17].

Han *et al.*, [18] endeavoured to create PBI membrane which has higher gas permeability. Distinctive blended antecedent polymers are utilized to create thermally modified (TR) PBI polymers. Controlled amalgamation of PBI forestalled the high pressing thickness in polymer lattice. Assessment of gas penetration execution of the fabricated membranes is done utilizing unadulterated gas feed containing N₂, CO₂, H₂, CH₄ and O₂. The porousness request of gases is H₂>CO₂>O₂>N₂>CH₄. TR-PBI membranes had increment in gas permeability when contrasted with the antecedent polymers. Besides, at raised working temperatures little gas particles, O₂ and H₂ permeability expanded, however, similar pattern could not be observed for different gases. H₂ porousness does not demonstrate an amazing increment in the temperature range test, however, CO₂ solvency and along these lines its penetrability diminished. These membranes demonstrated worthwhile execution particularly for CO₂ and H₂ separation at 120 °C.

Polyimide such as Matrimid is a standout amongst the most contemplated polymers in separating hydrogen. It is picked in light of the fact that the formation of membranes offers high porousness, selectivity, dissolvable obstruction also warm security [1,19]. Smahi *et al.*, [20] has arranged hydrogen particular polyimide membranes. The permeability of thick membranes are 1.14 Bar for CO₂ and 3.0 Bar for H₂ at 35 °C. With the aim to advance the membranes performance, they orchestrated half breed imide-siloxane copolymers. The membranes arranged from this copolymer demonstrated a superior execution in separating gas, the selectivity of H₂/CO₂ expanded between 2.63 to 3.5 at 35 °C.

2. Influencing of Annealing Conditions

Annealing is a warmth treatment that connected on membranes to expel the lingering dissolvable caught inside the membrane and to delete the warm history. Type and states of the annealing strategy give various effects on membranes relying upon the structure. Toughening at various temperatures capable to alter the inherent membranes properties. Fu *et al.*, [21] reviewed the impact of remaining dissolvable on the thick polyimide membranes at 25 °C and 150 °C under vacuum environment about 0.5 to 9 days. Low temperature is insufficient to remove leftover dissolvable. They referenced that at 150°C the portability of the polymer chains is higher, consequently, the dispersion of dissolvable is quick. Expanding lingering dissolvable sum, expanded the permeability for both O₂ and N₂, which means after a basic worth staying dissolvable plasticizes membrane.

Ismail and Kusworo [22] explored the impact of this wonder by getting ready polyethersulfone/polyimide mix and strengthened them at temperatures higher and beneath its glass transition temperature. The permeability of O₂ and N₂ are declining by expanding toughening temperature. At first, tempering temperature of 150°C and 240°C are utilized. In any case, the selectivity of acquired membrane is far lower compared to the unannealed membrane. Expanding the temperature to 280°C enhanced the selectivity nearly by factor 5. They translated that

toughening above T_g of the polymer is a successful strategy to delete the leftover dissolvable engravings. Hacıoğlu *et al.*, [23] revealed the impact of tempering time on thick polycarbonate membrane. Annealing is utilized for 8, 24, 72, 154 hours at temperature of 50°C. Denser structures are obtained because of tempering. For the following 24 hours, the permeability is declined, yet selectivity expanded. Then again, permeabilities of different gases remained practically steady somewhere in the range of 72 and 154 hours tempering. They inferred that 72-hour tempering is sufficient to crush dissolvable engravings inside the membrane structure.

Macchione *et al.*, [24] considered the impact of staying dissolvable on Hyflon® AD 60X membranes. They arranged thick gas separation membranes and the membranes are heated under vacuum at temperature of 70°C to 200°C gradually. They tested both high temperatures strengthened membranes and unannealed membranes utilizing CH₄, H₂, CO₂, O₂ and He. It is presumed that the size sieving properties of membranes are enhanced in the wake of toughening because of decreased chain versatility. The dispersion coefficient of helium remained steady, however, the dissemination coefficient of CH₄ diminished by very nearly one request of extent.

Kruczek *et al.*, [25] contemplated an elective tempering strategy utilizing sulfonated polyphenylene oxide. Extra warmth treatment after dissolvable vanishing is definitely not a legitimate strategy for the total expulsion of ensnared remaining dissolvable in the fabricated membranes as a result of the sulfonic bunches disintegration at high temperatures. In this manner, they chose to expel the remaining dissolvable by long haul pervasion of CO₂ across the membrane. The CO₂ pervasion rate expanded in the initial 3 days of the research, however no adjustment in permeation estimations of CO₂ are seen in 3 to 63 days. They inferred that there is no plausibility for dissolvable nearness in the membrane structure following 63 days. To exhibit the temperature and time reliance of toughening, Joly *et al.*, [26] explored the impact of dissolvable staying in 6FDA–mPDA polyimide membrane. The fabricated thick membranes are warmth treated at 200 °C for various durations. The warm treatment for a longer period gave lower dissemination coefficients, however, higher penetrability and solvency coefficients is achieved for N₂ and CO₂ gases. The engraving of dissolvable inside the membrane is dispensed at longer periods. Additionally, in the extent of the research, they considered various solvents for membrane creation. They presumed that high molar volume solvents leave engravings inside the structure.

Hibshman *et al.*, [27] examined the impact of toughening on gas separation of incompletely hydrolyzed polyimide membrane. Toughening at 400 °C and afterward diminishing the temperature by extinguishing caused cross-linking inside the structure. This is apparent with the aftereffects of FTIR-ATR, TGA-MS. Membrane is described by single gas permeability for CO₂, CH₄, N₂, O₂ and He at 35°C. The permeability expanded just about 2 to 3 creases. Results are credited to modify neighbourhood atomic movements on the polymer chain. Diffusivity predominant pervasion is observed. In any case, selectivity declined.

To decide the impact of annealing temperatures on membrane execution, previous researcher analyzed integrated fluorinated, 6FDA based polyamide–imide to separate CH₄ from streams containing plasticizers such as H₂S and CO₂ using thick membranes toughened at different temperatures [28]. 200°C is observed as the most appropriate annealing temperature.

3. Effect of Operating Temperatures

In literature, the vast majority of the researches related to gas separation membranes are carried out at low working temperatures. Indeed, Robeson [29] exchange trade-off is plotted utilizing estimations done at temperatures ranging from 25°C to 35°C. The impact of temperature on upper bound line are examined as of late. On the off chance that the point is getting elite membranes

particularly for high temperature applications and to completely investigate the performance of arranged membranes, it is essential to study using higher temperatures. Versatility of polymer stuffed structure and intersegments are temperature subordinate [30]. Figure 2 shows the diagrams of operating temperature effect on disk-carbon membrane reported by Ismail and *et al.*, [31]. At raised temperatures, the particles diffusivity and segmental movements increment causes higher transportation rates. Explicit gas atoms that have higher enactment energies are more impacted by the temperature [32]. Ostwal *et al.*, [33] contemplated the vehicle properties of nitrogen and carbon dioxide with poly(fluoroalkoxyphosphazene) membrane. Expanding temperature from -15°C to 30°C expanded the porousness of CO₂ in the range of 142 Bar to 336 Bar. Ideal selectivity of CO₂/N₂ modified from 21 to 12 at 3 bar feed weight. Solvency coefficient of CO₂ decayed as temperature increment then again, diffusivity coefficient demonstrated a contrary pattern.

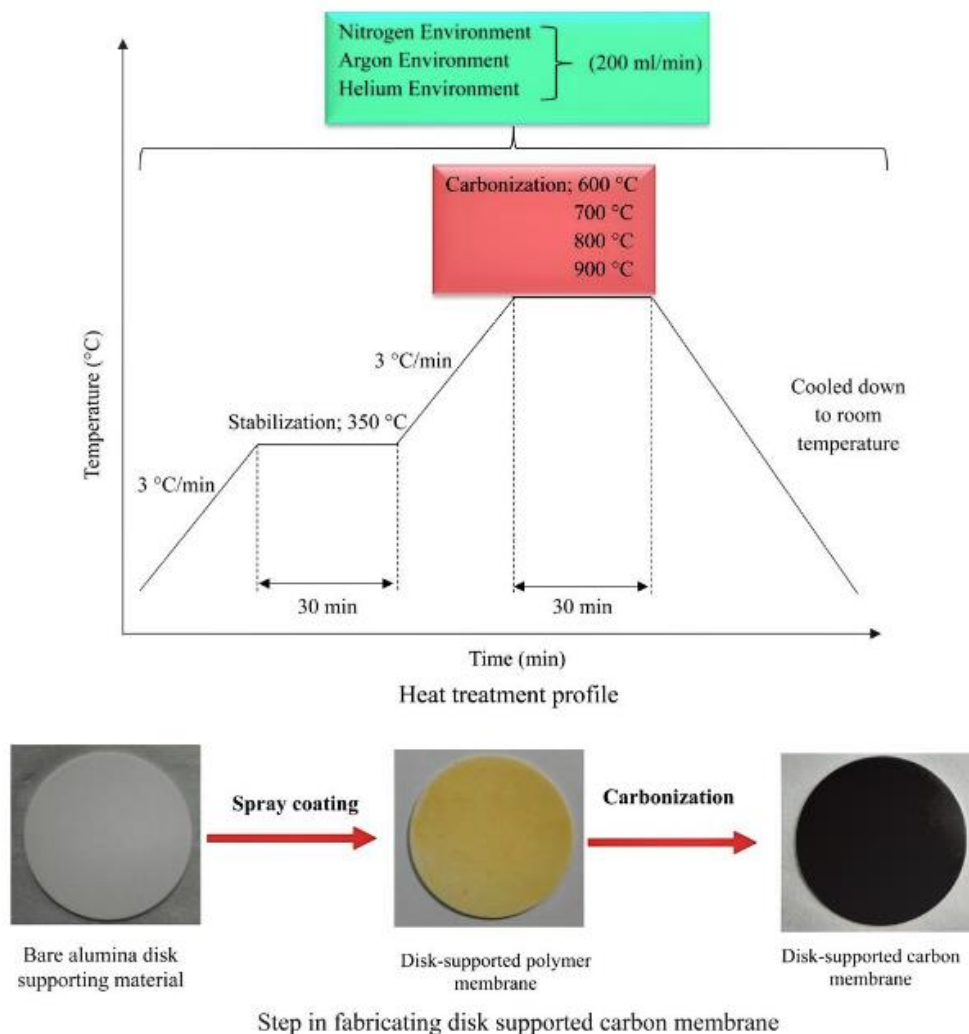


Fig. 2. Diagrams of operating temperature effect on disk-carbon membrane

Costello and Koros [34] researched the adjustment in gas pervasion execution of three diverse polycarbonate polymers as an element of temperature. Tetramethylhexafluoro polycarbonate, tetramethyl polycarbonate and bisphenol-A polycarbonate are tested utilizing N₂, He, CH₄ and CO₂ at temperature ranging from 35°C to 125°C. Permeability of gases are expanded through working temperature increment, however, selectivity declined. The disadvantage in He/N₂ and CO₂/CH₄

selectivity are identified with the lessening in both dissolvability and diffusivity selectivity. Membranes arranged by tetramethylhexafluoro polycarbonate has shown the best performance.

Merkel *et al.*, [35] researched the penetrability of syngas feed containing CO₂, CO, H₂S as well as H₂ and at temperatures switching from room temperature to 240 °C. The reproduced feed is tested utilizing solvency specific rubbery poly(dimethylsiloxane) (PDMS) as well as polished poly(1-trimethylsilyl-1-propyne) (PTMSP) polymer. At room temperature, the two membranes demonstrated higher permeability for CO₂ and H₂S. At raised temperatures, they wound up hydrogen specific. At higher temperatures, penetrability of all gases aside from hydrogen diminished for PTMSP polymer demonstrating negative actuation energies. In any case for PDMS, all gases gave positive actuation energies. The determined initiation energies are most elevated for hydrogen and diminished by the solubility of gases. Mixed matrix membrane is fabricated utilizing polysulfone acrylate-zeolite 3A and concentrated as an element in separating H₂/CO₂. The working temperature altered from 25°C to 55°C. Diffusivity of the two gases expanded with rising temperature due to improved adaptability inside polymer chains. In any case, CO₂ porousness increment is progressively huge thought about the H₂ selectivity which thusly results in lessening of selectivity with temperature [36].

Diaz *et al.*, [37] used C₂H₄, C₂H₆, CH₄, CO₂, O₂, N₂ and H₂ gases to investigate the performance of poly(1,4-phenylene ether-ether-sulfone) membranes which contain ZIF-8 particles as filler. For every single tested gas, permeability expanded because of improved dispersion. Hydrogen is the main atom that displayed endothermic sorption process for both smooth also MMMs, therefore, dissemination prevailing permeation is obtained for arranged MMMs.

Oral *et al.*, [38] utilized four distinct membranes to research the impact of toughening temperature and time on membrane performance. These membranes incorporate smooth PES/SAPO-34 and PES, as well as PES/HMA mixed matrix membranes and PES/SAPO-34/HMA. Pristine PES membrane indicated stable separation results. Be that as it may, same pattern could not be obtained for different kinds of membranes. Subsequently, post annealing is connected at the pressure of 0.2 atm, temperature of 120°C for 7-30 days. After post annealing, membranes demonstrated higher and stable gas penetrations for every single tested gas.

4. Conclusions

The membrane structure is a significant basis in deciding the correct application territory of utilization. The principle point in membrane creation is accomplishing the mix of high selectivity and high gas penetrability for a specific application. Subsequently to develop a membrane with desired properties considering development systems is vital. Numerous endeavours are done to build the membrane performance indicating high porousness and selectivity to have product with higher purity [39]. Then again, it is imperative to get membranes exhibit high performance at extreme industrial working environments.

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