Dip-coating methods for carbon membrane fabrication: Effects of coating-carbonization-cycles on Hydrogen separation prepared from P84/NCC

Norazlianie Sazali[1,2]*, Mohd Syafiq Sharip[1], Haziqatulhanis Ibrahim[1], Ahmad Shahir Jamaludin[1], and Wan Norharyati Wan Salleh[3]

[1]Faculty of Mechanical and Manufacturing Engineering, Universiti Malaysia Pahang, 26600 Pekan, Pahang, Malaysia.
[2]Centre of Excellence for Advanced Research in Fluid Flow (CARIFF), Universiti Malaysia Pahang, Lebuhraya Tun Razak, 26300 Gambang, Kuantan, Pahang, Malaysia.
[3]Advanced Membrane Technology Research Centre (AMTEC), School of Chemical and Energy, Faculty of Engineering, Universiti Teknologi Malaysia, 81310 Skudai, Johor Darul Takzim, Malaysia.

Corresponding author: Norazlianie Sazali at azlianie@ump.edu.my

Abstract. In this paper depicts the manufacture and assessment of tubular carbon membrane equipped from BTDA-TDI/MDI (P-84) polyimide mixes with Nanocrystalline cellulose (NCC). Given earlier investigations, we planned the theory that tubular carbon membrane performance could impose constraints by controlling the carbonization conditions which directed with a heating rate of 3°C/min, a final temperature of 800°C and stabilization temperature of 300°C. The principal purpose of this examination is to acquaint successful dip-coating strategies with produce superior tubular carbon membrane. The coating-carbonization cycles (1, 2, 3, and 4 times) have been considered. This methodology empowers quick and straightforward assessment of dip-coating techniques to yields high separation performance. Gas separation performance of the carbon membranes was adequately carried out by a single gas permeation experiment of H_2, and N_2 to explore the transport component in the carbon membrane separation process. In this case, the most elevated selectivity of 434.68±1.39 for H_2/N_2; side by side with H_2 permeance of 1399.66±5.22 GPU shall accomplish by employing two coating-carbonization-cycles.

Keywords: Coating-carbonization-cycle, P84 co-polyimide, Nanocrystalline cellulose (NCC), Tubular carbon membrane, and Hydrogen.

1 Introduction

Membrane innovation has sociably attracted many reviews in consideration with the gas separation enterprises, for example, hydrogen recovery, air separation, olefin/paraffin separation, CO_2 capture, nature gas dehydration, and many other (1). Carbon membrane innovation is in the process of being created promptly for these reasons. The requests on inorganic membrane have expanded because of the reduced temperature and chemical resistance of natural membrane. The hand-selected molecular sieving system of carbon membrane is especially helpful to accomplish great separation which is smooth between gases with practically relative molecular size (2). Some other way, this approach turns out to be more energy conservation thus progressively efficient by contrasting with other gas separations technique. In this manner, a few specialists have participated in evolving new materials with a few manufacture techniques for carbon membrane (3). One method is to manufacture the carbon membrane as a tubular aided membrane to limit imperfection and manage the cost of high gas separation performance. The fundamental focal point of this examination ponders to give better comprehension of the best dip-coating process parameters to control the morphology of the tubular aided carbon membrane. The primary procedure parameters that will partake is coating-carbonization cycles. Through the author learning, dip-coating techniques are a standout amongst the most significant strategies for generation zone to supplant the conventional spin coating and spray coating methods. It has something to do with the fact of no impediment in supported estimate size, and the usage of polymers is low (4). Hydrogen (H_2) is a decent fuel possibility for clean energy generation since it just generates water as side-effect upon burning . For the moment, steam converting of natural gas is an essential procedure in creating H_2. In any case, filtration of H_2 from the output stream is mandatory for earlier utilisation. As for H_2/N_2 separation, if the molecular size of H_2 is small, H_2 is specialised in permeating through the membrane. In any case, the H_2 gas that penetrates through the membrane ought to be compacted again before it tends to utilise and this is a keen energy procedure (5). In this paper, our target is to manufacture the PI/NCC-based carbon tubular membrane for both H_2 filtration and flue gas treatment. As long as there is a limited number of analyses on tubular carbon membrane from mixing polymer, the tubular membrane design is suitable for industrialised gas separation, and mixing of NCC with PI which can enhance the gas separation performance (6). As we all know, this is the initial endeavour of mixing NCC with PI to create carbon tubular layers for H_2/N_2 separation.

Dip-coating of porous tubular alumina ceramic with a solvent of PI-copolyimide mixes with nanocrystalline cellulose (NCC) was utilised to manufacture carbon membrane. As one of the writer philosophies, the manufacturing by a composite of NCC is the initial examination; henceforth bring the reproducible way of NCC itself. Past investigation proposed that cellulose is one of the natural linear polymers of b-(1/4)-D-glucopyranose that is owing novel highlights, for example, nontoxicity, high biocompatibility, and biodegradability (6). Additionally, it is likewise, has excellent mechanical strength with durable adsorption manner. Truth be told, the one of a kind

local property owe by the cellulose has pull in specialist in including cellulose substances as a supplement substance inside the mixing materials (7). The hydrophobized cellulose inferred so as to keep away from muster of cellulose started from the surface hydroxyl cluster. Past investigation expressed that, NCC is one of the hydrophobized cellulose subordinates for lean or thin cellulose that frequently utilize. Amid membrane manufacture, it is essential to keep away membrane from any fractures and pinholes flaw so as to keep up great membrane selectivity. Therefore, the decision of material to aid this process is a crucial job. An exemplary aid ought to have a deformity-free surface with low coarseness, high porosity and small pore estimate. In this examination, tubular support produced from alumina ceramic is utilised because of its high physical quality, high gas diffusivity and tolerance to the high-temperature carbonization treatment amid membrane fabrication (9). Although the vast majority of the analysts in this field would prefer to pick support materials with a deformity-free microporous. Several even on Nanoporous surface layer to guarantee useful invention by steer clear of deformities generation, the uprising expense joined by this best quality will help in the long run keep down the capability of carbon membrane applications. In the present study, an affordable tubular macro porous Al_2O_3 is chosen as the membrane support material to acknowledge cost sparing in the fabrication process. Nevertheless, the issue of the vulnerability of carbon membrane established from the low-cost assist should initially suppress so as to guarantee reproducibility and versatility of the membrane. Dip-coating is considered the least difficult and most possibly applied manufacture method in the market. Numerous works are in progress to enhance this system (9). Hence, this paper will review about dip-coating state because of less research on dip-coating process conditions.

2 Experimental section

2.1 Materials

Polyimide BTDA-TDI/MDI (P-84), polyvinylpyrrolidone (PVP), and microcrystalline cellulose (MCC) which acquired from Sigma Aldrich whereas N-methyl-2-pyrrolidone (NMP) obtained from Merck (Germany). All synthetic compounds were utilized with no more than that of purification. Nanocrystalline cellulose (NCC) is presently not financially accessible and was equipped already in our investigation (7). Porous tubular ceramic assist (TiO_2) with 8cm long, 13mm in external diameter, 10mm internal diameter with the standard pore size of 0.2µm (porosity of 40-half) that bought from Shanghai Gong Tao Ceramics Co., Ltd.

2.2 Preparation of carbon membrane

Polymer mixture comprise of 15wt.% of P-84 out of a NMP is set up under consistent mixing at 80°C. 7wt% of NCC was bit by bit added to the mixture and keeps mixing until homogeneous mixture was acquired. The mixture was sonicated to eliminate trapped bubbles from the mixture. Then, the mixture is put aside for 12 hours.

The tubular aid was immersed into the polymeric mixture for 45minutes and experienced 'aging' at 80 °C inside the fume hood. The membranes were then submerged in methanol for 2 hours before being set inside an oven at 100 °C for 24 hours to permit moderate expulsion of the solution. Carbon membranes were set up via carbonization procedure of the supported polymeric membranes. The supported polymeric membranes were positioned at the focal point of the Carbolite horizontal tubular furnace to experience carbonization operation. The carbonization procedure executed at various carbonization temperature of 800°C under Argon condition (200ml/min). The heating rate of 3°C/min was connected all through the procedure. In the wake of finishing each heating cycle, the provided membrane was cooled generally to room temperature. Carbon membrane without supported was produce by employing an identical procedure for characterization purpose.

2.3 Pure gas permeation measurements

The carbon tubular membrane was experimental in a gas penetration framework as portrayed in our past examination (10). The carbon tubular membrane laid inside a tubular stainless-steel module of 14 cm long. The membrane was adjusted with elastic O-rings to enable the membrane to the position in the module without spillages. The porousness of four pure gases with various molecular sizes; H_2 (2.80), and N_2 (3.64) as a result carbon membrane created from various coating cycles at a feed pressure of 8 bar. On account of the molecular diameter who is nearly identical, the gas separation by gas compound containing H_2, CH_4, CO and CO_2 is an intricate issue. Thus, gain insight into developed of porous carbon structure is expected to manage the pore sizes in the membrane. The permeance, P/l (GPU) and selectivity, α of the membranes were determined and the selectivity characterized as the penetration proportion of fast gas permeation to moderate gas permeation.

3 Results and Discussions

3.1 Gas permeation measurements

Dip-coating procedures might have profoundly recommendable for reasonable applications because of their relative simplicity. The impact of coating circumstances additionally demonstrates a few consequences for the moving or transfer properties of the carbon membranes (10). It expressed that fitting manipulation of the coating cycles created carbon membrane with high solidness while coating time improved the membrane selectivity and decreased the membrane's porousness. Amid dip covering process, the interfacial stress among polymer and aiding which diminishes the Tg of the polymer subsequently will restrain the development of complicates structure (7). Later in the carbonization process, distinction in carbon microstructure are because of the contrast at homogeneous polymer surface. In the dip-coating technique, the gases permeability came to very nearly a consistent incentive after two coatings and can be considered that two times coating cycle are the best while though in film casting

which required three coatings. Liu stated four successive dip-coating is essential to create their membranes (11).

Table 1. Gas separation performance of carbon membranes produced from different coating-carbonization cycles.

Carbon membrane			
Sample (CM-PI/NCC)	**Permeance (GPU)**		**Selectivity**
	H_2	**N_2**	**H_2/N_2**
1 cycle	1306.27±3.22	3.05±1.21	428.29±1.87
2 cycles	1399.66±5.22	3.22±3.21	434.68±1.39
3 cycles	1105.21±2.39	2.71±1.03	407.835±1.76
4 cycles	988.91±2.94	2.55±2.71	387.81±2.44

Further, growing the number of coatings to four brought about roughly no adjustment in permeance for dip-coating and film casting systems. Nonetheless, for the pouring technique, H_2 permeance was diminished relentlessly after four successive coatings (12). The operation of the manufactured membranes for the control of dip-coating status considered. The selectivity of the equipped membrane by dip-coating was higher than that for the film casting method a role as notified by past examination. The coating carbonization cycles have prompted an expansion of the membrane selectivity. Enlarging the number of coatings to four brought about roughly comparative permeance for dip-coating and film casting systems. Selectivity of the equipped membrane by dip-coating was higher than other coating procedures because of better permeation of PI/NCC mixture into membrane surface pinholes. For the strategies, as the number of repeated coatings expanded, the exposed pores or deformities on the membrane surface were additionally clogged or blocked and thus, enhance production was attained. Besides, a correlation of film casting and dip-coating strategies demonstrated improve inclusion of deformities by the coating layer in the dip-coating procedure than that in film casting (13). In respect to the optimum manufacture states, the aided carbon membrane shall examine so as to decide their gas permeances. Sad to say, all membrane may experience the small insignificant despite the fact that the membrane was seen to be without fracture or gap. Amid the dip-coating process, the residue from the surrounding air could have debased with the aided membrane which brings about little pinholes and infinitesimal defects on the membrane.

4 Conclusions

From this examination, the impact of dip-coating conditions on PI/NCC carbon membrane was explored. As a result, there are couple of outcomes can be made:

1. The enlarging of carbonization-covering-cycles from 1 to 2 would upgrade pore configuration because of disintegration preceding carbonization method. Thus,

the existence of NCC as added substance gives better pore auxiliary properties due to their nanocrystalline structure.

2. In this context, the carbon membrane carbonised at 800°C demonstrates the most encouraging outcome to H_2/N_2 selectivity of 1399.66±5.22 with H_2 permeance of 213.56±2.17

3. The positive findings acquired in this examination show the capability of crystalline cellulose, particularly NCC, for thermally labile added substance warrants for further study.

Acknowledgement

The authors would also gratefully acknowledge the financial support from the Ministry of Education and Universiti Malaysia Pahang under the Fundamental Research Grant Scheme (Project Number: Rdu1703253 and Rdu191105).

References

1. Sołowski G, Shalaby MS, Abdallah H, Shaban AM, Cenian A. Production of hydrogen from biomass and its separation using membrane technology. Renewable and Sustainable Energy Reviews. 2018;82:3152-3167.
2. Sazali N, Salleh WNW, Ismail AF, Kadirgama K, Othman FEC, Ismail NH. Impact of stabilization environment and heating rates on P84 co-polyimide/nanocrystaline cellulose carbon membrane for hydrogen enrichment. International Journal of Hydrogen Energy. 2018.
3. He X. Techno-economic feasibility analysis on carbon membranes for hydrogen purification. Separation and Purification Technology. 2017;186(Supplement C):117-124.
4. Li P, Wang Z, Qiao Z, Liu Y, Cao X, Li W, et al. Recent developments in membranes for efficient hydrogen purification. Journal of Membrane Science. 2015;495:130-168.
5. Ismail NH, Salleh WNW, Sazali N, Ismail AF, Yusof N, Aziz F. Disk supported carbon membrane via spray coating method: Effect of carbonization temperature and atmosphere. Separation and Purification Technology. 2018;195:295-304.
6. Salleh WNW, Ismail AF. Carbon membranes for gas separation processes: Recent progress and future perspective. Journal of Membrane Science and Research. 2015;1(Issue 1):2-15.
7. Sazali N, Salleh WNW, Ismail AF, Kadirgama K, Othman FEC. P84 Co-Polyimide Based-Tubular Carbon Membrane: Effect of Heating Rates on Helium Separations. Solid State Phenomena. 2018;280:308-311.
8. Sazali N, Salleh WNW, Ismail AF, Nordin NAHM, Ismail NH, Mohamed MA, et al. Incorporation of thermally labile additives in carbon membrane development for superior gas permeation performance. Journal of Natural Gas Science and Engineering. 2018;49:376-384.
9. Sazali N, Salleh WNW, Ismail AF, Ismail NH, Yusof N, Aziz F, et al. Influence of intermediate layers in tubular carbon membrane for gas separation performance. International Journal of Hydrogen Energy. 2018.

10. Sazali N, Salleh WNW, Nordin NAHM, Ismail AF. Matrimid-based carbon tubular membrane: Effect of carbonization environment. Journal of Industrial and Engineering Chemistry. 2015;32:167-171.
11. Liu R, Qiao, X. & Chung TS. The development of high performance P84 co-polyimide hollow fibers for pervaporation dehydration of isopropanol. Chemical Engineering Science,
2005: 60, 6674-6686.
12. Ismail NH, Salleh WNW, Sazali N, Ismail AF. (2017). Effect of intermediate layer on gas separation performance of disk supported carbon membrane. Separation Science and Technology. 52(13): 2137-2149.
13. Ismail NH, Salleh WNW, Sazali N, Ismail AF. (2015). The Effect of Polymer Composition on CO2/CH4 Separation of Supported Carbon Membrane. Chemical Engineering Transactions 45: 1465-1470.