

Development of Short Cut Design Method for CO Removal System

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ABSTRACT

Hydrogen-rich reformat produced by reforming primary fuels in the fuel processor system, contains significant amount of CO, which is the main culprit for the performance reduction in Proton Electrolyte Membrane Fuel Cell (PEMFC) performance by CO poisoning on the Platinum catalyst at anode. Thus the reformat from the fuel processor system must be purified by removing CO before being used in the PEMFC stack. The objective of the present study is to design conceptually a CO removal system consisting of a mesoporous tubular ceramic membrane (TCM) and a pressure swing adsorber (PSA) in series. The first section of the present study describes the design of the TCM and the second section describes the design of the PSA. The designs for both units are based on new shortcut design methods developed by the authors. The effect of some important parameters such as the temperature, pressure and membrane area on the degree of separation is investigated. It is found that theoretically, the TCM-PSA system capable of reducing the CO concentration from 2000ppm to less than 10ppm.

Keywords: PEMFC, Design, tubular ceramic membrane, pressure swing adsorber.

ABSTRAK

Hidrogen yang dihasilkan di unit pemprosesan bahan api mengandungi kandungan karbon dioksida yang mampu menjejaskan prestasi sel bahan api. Karbon dioksida merupakan racun kepada mangkin platinum pada bahagian anod di stak sel bahan api. Dengan itu karbon dioksida di dalam aliran pembentukan semula hidrogen perlu dipindahkan sebelum aliran tersebut memasuki stak sel bahan api. Objektif utama kajian adalah untuk merekabentuk konsep sistem pemindahan karbon dioksida dengan menggunakan kaedah reka bentuk pintas. Dua unit alat penulenan digunakan secara selari di dalam kajian ini, iaitu modul membran seramik dan penjerap tekanan berayun. Bahagian pertama kajian ini akan menerangkan mengenai reka

bentuk membran diikuti dengan reka bentuk sistem penjerap. Kajian ini juga melihat kepada kesan parameter-parameter penting seperti suhu, tekanan dan luas membran terhadap darjah pemisahan membran. Hasil reka bentuk menunjukkan kedua-dua unit membran dan penjerap berupaya untuk mengurangkan kepekatan karbon dioksida daripada 2000bpj kepada kurang daripada 10bpj.

Kata kunci: PEMFC, reka bentuk, modul membran seramik, penjerap tekanan berayun.

INTRODUCTION

The reformat from fuel processor units consists of hydrogen, carbon dioxide, carbon monoxide and residues of hydrocarbons. Since the anode catalyst of a proton electrolyte membrane fuel cell (PEMFC) is platinum base, which is easily poisoned by CO. A CO management system is required to lower the CO concentration to acceptable levels. In general, the CO content in the product hydrogen has to be below 10ppm in order to be used as the anode gas for the PEMFC. The main objective of this study is to develop a short-cut design method for CO removal system. Two separation units were introduced in this paper namely, the tubular ceramic membrane, TCM and pressure swing adsorber, PSA. Both membrane and adsorber will be operated in parallel to gain the product purity as 99.9% hydrogen and CO less than 10ppm. The effect of some important parameters such as the temperature, pressure, and membrane thickness on the degree of separation were investigated.

THEORETICAL BACKGROUND

Tubular Ceramic Membrane, TCM

There has been a considerable growth in the research and development of organic and inorganic membrane materials for the separation of gas mixture during the last 30 years. The interest in membrane process to separate gas mixtures using inorganic membrane has increased considerably. The advantage of inorganic membranes compared to organic membranes is essentially their thermal and chemical stability. Both porous and non porous (dense) inorganic membranes (Mandal et al. 2002) can be used as selective gas separation barriers. The separation factors for porous membranes are however much lower than those of Pd alloys, unless the pores are of molecular dimension. However due to hydrogen embitterment, a phenomenon in which dissolved hydrogen tends to cause lattice expansion in the metal; eventually causing it to rupture on repeated pressure and temperature

cycling. In addition, palladium (and Pd-alloy) requires a high pressure differential (10-20 bar) and a relatively high temperature (300-400°C) (Jayaraman & Lin 1995).

A basic principle for selecting inorganic materials for dense ionic conducting ceramic membranes is described for high temperature permeation/reaction experiments. The new developments of ceramic membranes have given rise to many possibilities for catalytic membranes reactors (Othman 2001; Qi & Lin 2001). The main advantages of these membranes over polymeric membranes are: (1) greater fluxes and stability at high temperatures, (2) the porosities are as high and controlled; (3) stable and narrow pore size distributions; (4) mechanically stable and can withstand high pressure drops, (5) resistant to corrosive chemicals; (6) catalytic materials that are deposited by impregnation can also be dispersed on ceramics and thus they can have high catalyst surface/volume ratios; (7) thermal and chemically stable; (8) long lifetime (Heizel et al. 2002; Lee et al. 2003).

Although the porous membrane for gas separation has been studied in terms of preparation (Lira & Paterson 2002; Zhao et al. 2002) and applications such as for membrane reactor; and biological processes (Mohammadi & Pak 2002), the mechanism of the gas transport on the membrane and the membrane design is not well known since it involved complex equations. Due to that, this paper will present a shortcut method of designing porous membrane system. The novel approach of this paper is that the required membrane area could be expressed as the product of the height of a transfer unit, HTU and the number of transfer unit, NTU. HTU described the dimension parameter while NTU represent the dimensionless parameter.

Figure 1 shows the cross-flow pattern of a porous membrane module. The local permeation rate of component A over differential membrane area, dA_M at any point in the stage is given as,

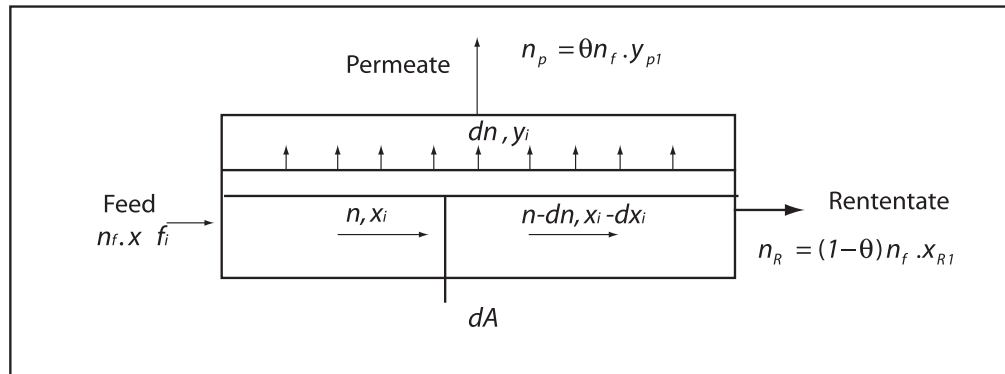


FIGURE 1. Cross flow model for membrane module

$$-ydn = \frac{K_1}{l_M} (P_F x - P_P y) dA_M \quad (1)$$

$$-(1-y)dn = \frac{K}{l_M} [P_F(1-x) - P_P(1-y)] dA_M \quad (2)$$

with y and x is the mole fraction of local permeate and rejection stream along the stream, K_1 is the permeability of component A through the membrane, l_M is the thickness of the membrane, P_F and P_P are the pressure in the feed and product of the membrane, respectively.

Dividing equation (1) by (2) and taking

$$r = \frac{P_P}{P_F} \text{ gives,} \quad (3)$$

$$\frac{y}{(1-y)} = \frac{\alpha^* (x - ry)}{(1-x) - r(1-y)}$$

where α^* is defined as the ideal separation factor. Assuming the pressure ratio, r and the ideal separation factor, α^* are constant, then the differential rate of mass transfer of component A across the membrane is given as

$$y_1 dn = d(nx_1) = x_1 dn + ndx_1 \quad (4)$$

$$\frac{dn}{n} = \frac{dx_1}{y_1 - x_1} \quad (5)$$

While for the actual separation factor, α is defined as:

$$\alpha = \left(\frac{y_1}{x_1} \right) \left(\frac{x_2}{y_2} \right) \quad (6)$$

Equation (6) combines with equation (5) to eliminate y_1 and obtain,

$$\frac{dn}{n} = \left[\frac{1 + (\alpha - 1)x_1}{x_1(\alpha - 1)(1 - x_1)} \right] dx_1 \quad (7)$$

When the downstream pressure is not negligible, combining the definition of r with equation (6), $\alpha_{1,2}$ can be expressed as below:

$$\alpha_{1,2} = \alpha * \left[\frac{\left(\frac{x_2}{y_2} \right) - r\alpha_{1,2}}{\left(\frac{x_2}{y_2} \right) - r} \right] \quad (8)$$

Substituting $y_1 + y_2 = 1$, and $x_2 = x_2 y_1 + x_2 y_2$ into equation (8)

With combine equation (8) and (9) and replace x_2 with $(1-x_1)$, we obtain the separation factor as:

$$\alpha_{1,2} = \alpha * \left[\frac{x_1(\alpha_{1,2} - 1) + 1 - r\alpha_{1,2}}{x_1(\alpha_{1,2} - 1) + 1 - r} \right] \quad (10)$$

where $\alpha_{1,2}$ are defined as the actual separation factor for component 1 and 2.

For a constant value of separation factor, $\alpha_{1,2} = \alpha$, for entire range of cut $\theta = np/nf$ the integration of equation (5) shall give:

$$\frac{n}{n_R} = (1-\theta) \left[\left(\frac{x_1}{x_{1R}} \right)^{\left(\frac{1}{\alpha-1} \right)} \left(\frac{1-x_{1R}}{1-x_1} \right)^{\left(\frac{\alpha}{\alpha-1} \right)} \right] \quad (11)$$

The mole fraction of component 1 in the final permeates and the total membrane surface area is obtained by integrating the values obtained from solving equations (3), (5) and (7):

$$y_{P,1} = \int_{x_{F,1}}^{x_{R,1}} y_1 \frac{dn}{\theta n_f} \quad (12)$$

Combining equation (7), (11), (12) and the definition for α , the integral shall give:

$$y_{1P} = x_{1R} \left(\frac{1-\theta}{\theta} \right) \times \left[(1-x_{1R})^{\left(\frac{\alpha}{\alpha-1} \right)} \left(\frac{x_{1F}}{1-x_{1F}} \right)^{\left(\frac{\alpha}{\alpha-1} \right)} - x_{1R}^{\left(\frac{\alpha}{\alpha-1} \right)} \right] \quad (13)$$

where x_{1F} is the mole fraction of component 1 in feed streams while x_{1R} and y_{1R} are the local permeate and retentate in the membrane unit and can be solved using ingenious transformation equations (Geankoplis 1993). For multi component gas separation,

$$\sum y_{P,1} + y_{P,2} + \dots y_{P,i} = 1.0 \quad (14)$$

and

$$\sum x_{R,1} + x_{R,2} + \dots x_{R,i} = 1.0 \quad (15)$$

where i is defined as number of component

Here cross-flow models for porous membrane are developed by considering mass balance at a differential element of the cross-flow module and then integrating the expression over the whole module to get the module length. The local permeation rate of component A over differential membrane area dA_M at any point in the stage is given as,

$$y_1 dn = \frac{K_1 dA_M}{I_M} [x_1 P_F - y_1 P_P]$$

$$A_M = \int_{\text{Retentate}}^{\text{Feed}} \frac{I_M y_1 dn}{P_{M,1} (x_1 P_F - y_1 P_P)}$$

$$= \frac{I_M}{P_F K_1} \int_{x_{1F}}^{x_{1R}} \frac{y_1 dn}{(x_1 - r y_1)} \quad (16)$$

Since the modelling is rated-based, the length of both modules could be expressed as the product of the height of a transfer unit (HTU) and the number of transfer unit (NTU).

The required membrane area is given by:

$$A_M = n_T \pi D L \quad (17)$$

with n_T is defined as number of tubes, D is the membrane diameter and L gives the membrane length. Equations (16) and (17) could be rewritten in the form of NTU and HTU by

$$L = NTU \times HTU = \frac{A_M}{n_T \pi D} = \frac{I_M}{P_F K_1 n_T \pi D} \times \int_{x_{1R}}^{x_{1F}} \frac{y_1 dn}{(x_1 - r y_1)} \quad (18)$$

Substituting equations (11) and (13) to equation (18), the membrane length is given as below:

$$L = \frac{I_M n_F}{P_F K_1 n_T \pi D_M} (1-\theta)^2 \left[\left(\frac{x_1}{x_{1R}} \right)^{\left(\frac{1}{\alpha-1} \right)} \left(\frac{1-x_{1R}}{1-x_1} \right)^{\left(\frac{\alpha}{\alpha-1} \right)} \right] \times \int_{x_{1R}}^{x_{1F}} \frac{y_1}{(1-ry_1)} \left[\frac{1+(\alpha-1)x_1}{x_1(\alpha-1)(1-x_1)} \right] dx_1 \quad (19)$$

$$\text{where } HTU = \frac{I_M n_F}{P_F K_1 n_T \pi D_M} \quad (20)$$

$$\text{and } NTU = \int_{x_{1R}}^{x_{1F}} \frac{y_1}{(x_1 - r y_1)} \left[\frac{1+(\alpha-1)x_1}{x_1(\alpha-1)(1-x_1)} (1-\theta)^2 \right] \left[\left(\frac{x_1}{x_{1R}} \right)^{\left(\frac{1}{\alpha-1} \right)} \left(\frac{1-x_{1R}}{1-x_1} \right)^{\left(\frac{\alpha}{\alpha-1} \right)} \right] dx \quad (21)$$

with x_i and y_i are defined as local retentate and permeate of component 1, respectively at any point along the membrane and x_i can be estimated as below:

$$x_{i,1} = x_{0,1} - \left(\frac{x_{F,1} - x_{R,1}}{n} \right) \text{ assuming } x_{0,1} = x_{F,1}$$

and $x_{i,1}$ is the local retentate is iteration of x from 1 to n section while $. dx = x_{n-1,1} - x_n$. For local permeate, $y_{i,1}$, can be solved for each x_i using equation (3).

The term HTU is a dimension parameter that characterizes the physical appearance of a membrane. Principally the HTU for porous membrane describes the design parameter for a membrane unit. As can be seen in equation (20), it varies with the feed flow rate, n_F , membrane thickness, l_M , and decreases with feed pressure, P_F , permeability of gas, K and diameter of a membrane, D_M . The stage cut, does not affect the HTU value and it is constant along the membrane. While the solution of the integral representing the NTU of gas separation is calculated based on the mole fraction of component 1 in permeates and retentate stream along the membrane at different $. From equation (21), it was observed that the term of NTU highly depends on the local retentate of component 1, which is from x_F to x_R .$

Permeability of Membrane

There are three types of flow in porous membranes, differing in porous size. If the pores are relatively large, from 0.1 to 10 μm , gases permeate the membrane by convective flow, and no separation occurs. If the pores are smaller than 0.1 μm , then the pore diameters are the same size as or smaller than the mean free path molecules. Diffusion through such pores is governed by Knudsen diffusion, and the transport rate of any gas is inversely proportional to the square root of its molecular weight. The possible permeability for multi component gas in ceramic membrane is expressed as below assuming no molecular diffusion occur:

$$K_{Kn} = \frac{\epsilon}{z\tau_{Kn}RT} \left(\frac{1}{\frac{1}{D_{A,B}} + \frac{1}{D_{Kn}}} \right) \quad (22)$$

The diffusivity in the Knudsen flow region, D_{Kn} , is given as:

$$D_{Kn} = \frac{4}{3} d_{pore} \frac{\epsilon_p}{\tau_{Kn}} \sqrt{\frac{RT}{2\pi M}} \quad (23)$$

where d_{pore} is membrane pore diameter, ϵ_p is volume fraction in the pores, τ_{Kn} is the tortuosity, M is the molecular weight of the gas and T is the isothermal operating.

For multi component gas, the diffusivity of gas differs according to the mole fraction of the gas in the mixture as below:

$$D_{A,mix} = \frac{1}{\frac{x_B}{D_{AB}}} \quad (24)$$

The diffusivity of a binary gas mixture in the dilute gas region at low pressure predicted as below (Othman 2001):

$$D_{AB} = \frac{0.00143 T^{1.75}}{PM_{AB}^{0.5} \left[(\sum v)_A^{0.33} + (\sum v)_B^{0.33} \right]^2} \quad (25)$$

where D_{AB} is the diffusivity of component A and B, T is temperature, P is absolute pressure and M_{AB} is the average molecular weight for component A and B system that can be predicted by equation (26). The above equation considers the intermolecular forces of attraction and repulsion between molecules and also the different sizes of molecules A and B obtained by correlation using the sum of atomic volumes, v . The above equation can be used for mixtures of non-polar gases or for polar-non polar mixtures. As can be seen the equation shows that D_{AB} is proportional to $1/P$ and $T^{1.75}$.

$$M_{AB} = \frac{2}{(1/M_A) + (1/M_B)} \quad (25)$$

Pressure Swing Adsorber, PSA

PSA processes for separation and purification of gaseous mixtures have become important unit operations in the chemical process industry. A large variety of binary and multi component gas mixtures are commercially separated using this technology (Rajasree & Moharir 2000). In this study, the PSA unit is cooperated in parallel with membrane unit to remove the existing CO

in the stream before entering the fuel cell stack. Here we use a PSA, because it is found that PSA has a number of advantages in terms of product purity, product cost, operating cost and capital cost, product recovery and high energy efficiency (Iyuke et al. 2000; Jain et al. 2003).

A PSA process is quite complicated and there are many parameters to be decided and not many general easy-to-use design rules available so far. Due to that this paper will introduce a shortcut design method to design a PSA. The conventional design method by Collins depends on breakthrough curves data of a bench scale adsorber, is clearly not suitable as a short design method. In this paper, a novel short-cut method depending only on the linear adsorption isotherm and gas adsorbent mass transfer is presented. The method uses the Klinkenberg solution of the breakthrough curve for a linear driving forces mass transfer and a linear equilibrium isotherm. A relationship between the dimensionless time, τ , and the dimensionless bed length, ξ , which are function of the bed Daud utilisation parameter, δ (Wan Ramli 2002) are introduced here.

At equilibrium conditions the adsorption isotherm of an adsorbate on a particular adsorbent could be expressed in the form of Henry's Law:

$$q = Kc \quad (27)$$

where q is the concentration of the component in the adsorbed phase and c is the concentration of the component in the gas phase in equilibrium with the adsorbed component. The adsorption of the component in a packed bed of adsorbent is given by:

$$-D_L \frac{\partial^2 c}{\partial z^2} + \frac{\partial(uc)}{\partial z} + \frac{\partial c}{\partial t} + \frac{(1-\varepsilon)}{\varepsilon} \frac{\partial \bar{q}}{\partial t} = 0 \quad (28)$$

where D_L is the axial dispersion coefficient, u is the interstitial velocity, and ε as the bed porosity. If the mass transfer of the component to the adsorbent follows a linear driving force model than:

$$\frac{\partial \bar{q}}{\partial t} = k(q^* - \bar{q}) = kK(c - c^*) \quad (29)$$

If axial dispersion is neglected ($D_L = 0$) and the velocity is constant, then the solution of equation (28) expressed as a breakthrough curve is given as

$$\frac{c}{c_F} \approx \frac{1}{2} \left[1 + \operatorname{erf} \left(\sqrt{\tau} - \sqrt{\xi} + \frac{1}{8\sqrt{\tau}} + \frac{1}{8\sqrt{\xi}} \right) \right] \quad (30)$$

where c_F is the feed concentration of the component and

$$\xi \approx \frac{kKz}{u} \left(\frac{1-\varepsilon}{\varepsilon} \right) \quad (31)$$

$$\tau \approx k \left(t - \frac{z}{u} \right) \quad (32)$$

The mass transfer coefficient k is calculated using the correlations of Wakao and Funazkri (Wan Ramli 2002) which is given by:

$$\text{Sh} = 2 + 1.1 \text{Re}^{0.6} \text{Sc}^{1/3} \quad (33)$$

where $\text{Sh} = kd_p/D$ is the Sherwood number, $\text{Re} = Gd_p/\mu$ is the Reynolds number, $\text{Sc} = \mu/\rho D$ is the Schmidt number, D is the diffusivity of the component in the carrier gas and G is the mass flux of gas. The diameter of the bed, D_p is given as follows:

$$D_p = \left(\frac{4Q_F}{\varepsilon\pi u} \right)^{0.5} \quad (34)$$

where Q_F is the volumetric flow rate of feed gas. The length of the adsorber is estimated as the length of adsorbent fully saturated with the adsorbate in equilibrium with the component in the gas. The mass balance for this case gives

$$Q_F c_F t_{ideal} = q_F (1-\varepsilon) L_b D_p^2 / 4 \quad (35)$$

or

$$\frac{t_{ideal}}{L_b} = \frac{K(1-\varepsilon) \pi D_p^2}{4Q_F} = \frac{K}{u} \quad (36)$$

By using this relationship between the ideal time and the ideal length, the ratio of Klinkenberg's dimensionless ideal time and Klinkenberg's dimensionless ideal length is given simply by:

$$\frac{\tau_{ideal}}{\xi} = \frac{\varepsilon}{(1-\varepsilon)K} \left[\frac{t_{ideal}}{L_b} - 1 \right] = \left[1 - \frac{\varepsilon}{(1-\varepsilon)K} \right] \quad (37)$$

A bed utilization factor, $\tau/\xi \leq \tau_{ideal}/\xi$ and (38)

δ is defined by $\delta = t/t_{ideal}$ and known as (39) Daud utilisation factor

Combining equations (31), (32),(34) and (39) yields

$$\frac{\tau}{\xi} = \left[\delta - \frac{\varepsilon}{(1-\varepsilon)K} \right] \tag{40}$$

Equation (40) that has not been derived explicitly before, relates τ and ξ with δ in a simple manner. In fact this equation is consistent because if $\delta = 1.0$, then equation (40) reduces to equation (37). Equation (39) is solved simultaneously with equation (30) at c/c_f and various values of δ to get the actual curves of τ and ξ . The solution of τ and ξ is then expressed as a function of the bed utilization factor, δ .

TABLE 1. The physical and mechanical properties of TCM

The composition of reformat (mole %)	Porosity, ε	Tortuosity, τ_{Kn}	r	Module Type
H ₂ :0.728, CO:0.2, CO ₂ :0.27	0.52	6.7	0.2	Tubular

TABLE 2. The physical and mechanical properties of PSA

The composition of reformat (mole %)	Porosity, ε	Mass transfer coefficient, k	Rate constant, K	δ
H ₂ :0.955, CO:0.0005, CO ₂ :0.04	0.43	0.02 m/s	6500	0.2-0.3

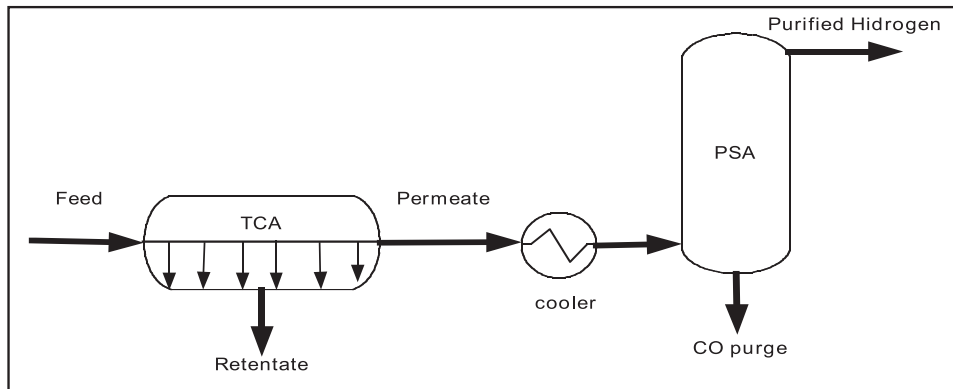


FIGURE 2. Schematic diagram of CO removal system

RESULT AND DISCUSSION

In this study, the transport of hydrogen (component 1) and CO (component 2) were considered in the TCM. The following assumptions are introduced in the formulation of the theoretical framework: (1) The transport process is isothermal (2) There are no chemical reaction.

The transport across the membrane is considered as cross flow and later it enters the PSA unit (refer to Figure 2). Table 1 and 2 summarized the physical and mechanical properties of TCM and PSA. Both design models were solved using Excel sheet and Matlab.

Figure 3 plots the permeability, K for H2 and CO. The permeability is expressed in g-mol.cm.cm-2-s-1 atm-1. The permeability is calculated for 300°C and 500°C. From the result it can be seen that the permeance for H2 and CO are slightly lower for 500°C.

Figure 4 predict the maximum and minimum values of permeance for H2 (yP1) and CO (yp2) using shortcut design method. It is observed that the maximum H2 content of the permeate (97.7 mol%) occurs with the smallest amount of permeate ($\theta=0.01$). The maximum CO content of the retentate (about 10%) occurs with the largest

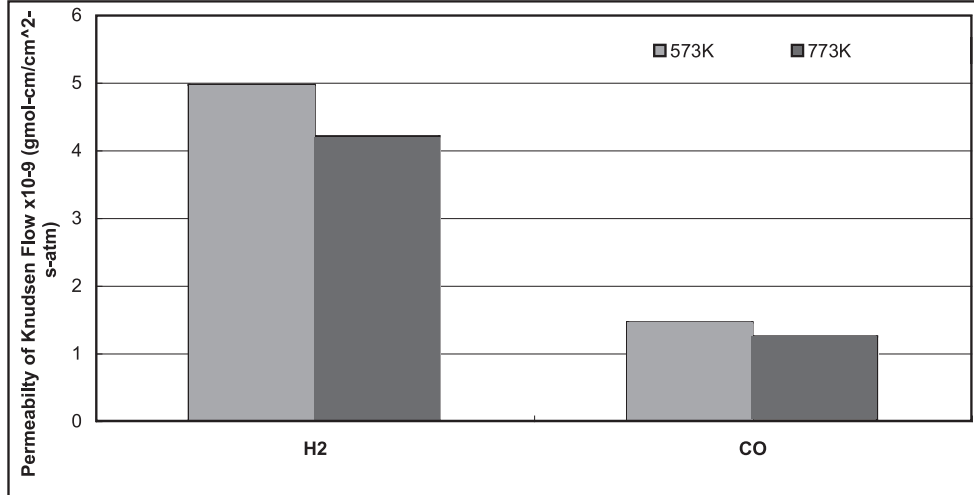


FIGURE 3. Results of permeability for different temperature

amount of permeate ($\theta=0.99$). With a retentate equal to 50 mol% of the feed ($\theta=0.5$), the CO content of the permeate had decreased from 2,000ppm to 500ppm. The permeance of H₂ had increase from 72.8% to 95.69%.

This show that this porous ceramic membrane ($d_{pore} = 1\text{ nm}$) is capable of removing CO and can be use as one of the purification system in the fuel cell system. The result is comparable with studies by Lee et al. (2003) obtained 99% purification of H₂ over CO using porous silica.

is observed the value of NTU increases with cut as the rejection, x_r of hydrogen reduces sharply compared to the decrements of the hydrogen permeation, y_p . As a result, it can be concluded that the NTU described the degree of separation, relatively, in term of local permeate and retentate. The higher the value of NTU, it predicts a higher degree of separation.

Figure 6 plots the surface area of the membrane required for the system based on NTU value for different flow rates. From the result, it can be

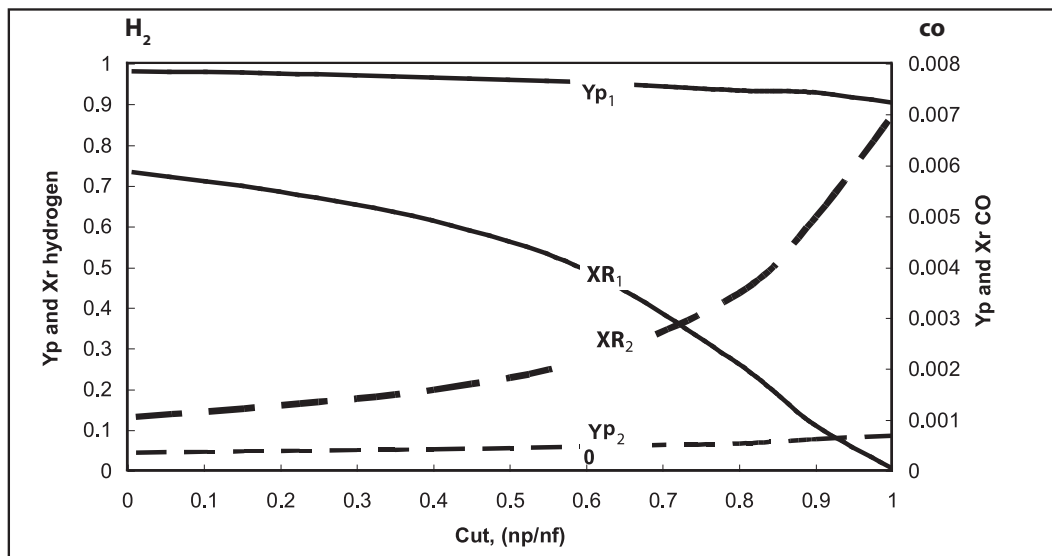


FIGURE 4. Mole fraction of y_p and x_r for H₂ (component 1) and CO (component 2) at different cut, θ .

Figures 5 to 7 characterized by NTU for ceramic membranes that are given in logarithm scale. Figure 5 gives the relation between NTU and cut, θ , for different r . Comparing Figures 4 and 5, it

observed that the surface area of the membrane is inversely proportional with permeance. At low cut, with high permeate, the surface area of membrane are small. This is due to the

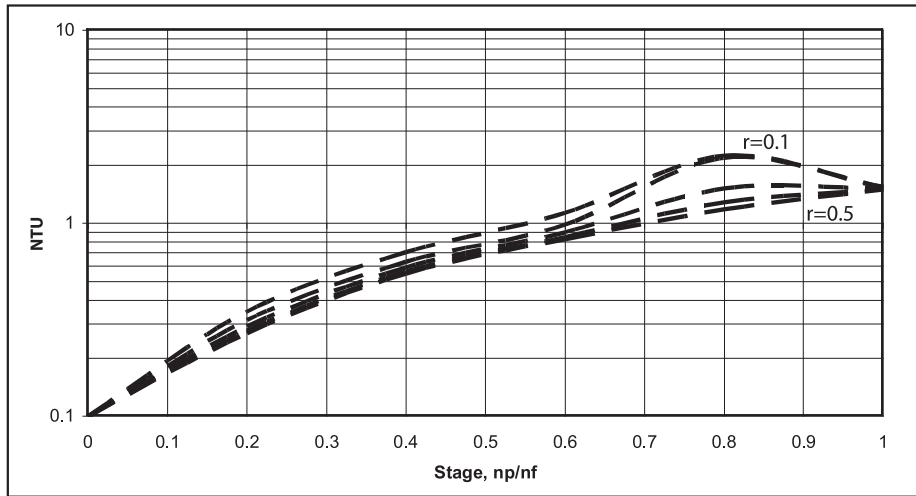


FIGURE 5. The NTU value versus cut, θ , at different values of r

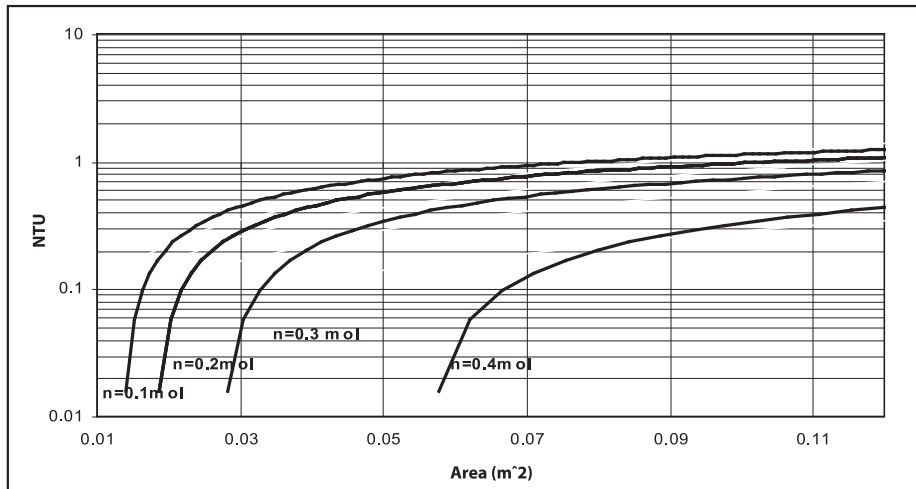


FIGURE 6. Results of the NTU and A_M for ceramic membrane for different feed flowrate

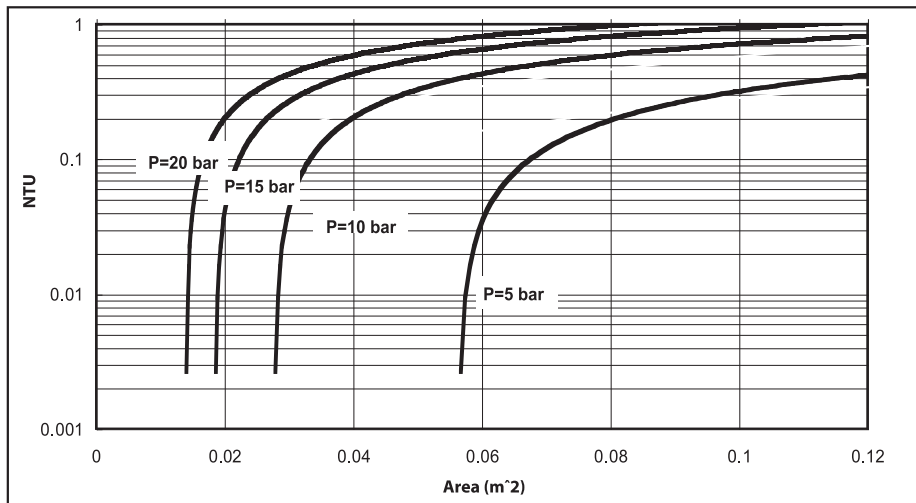


FIGURE 7. Results of the N_T and A_M for ceramic membrane for different pressure

competition of molecules to pass through the membrane are less in low flow rate and H_2 are separated easily while Figure 7 plots the effect of pressure on the membrane surface area. It was found that pressure plays an important role in designing a membrane. The higher the pressure, the smaller the surface area of membrane required. This is because the molecules are easily separated at high pressure.

The following Figures 8-11 describe the PSA unit in the system. For a PSA, the cycle time for increasing and decreasing pressure is short, so the value of δ is typically small, 0.2-0.3. The length of the adsorbent and the break through time are calculated by obtaining the values of ξ and τ . Figure 8 show the solution for ξ and τ as a function of the bed utilisation factor, δ while Figure 9 plot the breakthrough time and the bed length in the adsorption unit whereby c/c_f

is taken as 0.01. From the plot, the breakthrough time at $\delta = 0.3$ is estimated as 7 min for a bed length of 0.20m with a diameter of 0.25m.

Figure 10 and 11 plots the effect of bed length and diameter towards the interstitial velocity, u and the feed flow rate, Q_f . From Figure 10, it is observed that the length increase linearly with the u while the diameter decrease exponentially. In Figure 11, it can be seen that the diameter of the PSA unit increase exponentially with feed flow rate whereas the length does not change. This is because the length of adsorber only depends to the value of interstitial velocity, u .

Finally, Figure 12 illustrates the concentration of H_2 and CO in mole percentage. As a result, the CO is reduced from 2000ppm to 500ppm after the TCM and further reduced to less than 10ppm after the PSA unit.

Table 3 summarized the design parameters for the base-case for TCM and PSA units.

TABLE 3. Design Parameters for TCM and PSA

TCM	PSA
Feed flow rate, $n_f = 0.7 \text{ m}^3\text{min}^{-1}$	Feed flow rate = $0.3 \text{ m}^3\text{min}^{-1}$
Temperature = 250°C	Temperature = 25°C
Pressure ratio, $r = 0.2$	Pressure = 6 bar
Degree of separation, $\theta = 0.4$	Bed factor, $\delta = 0.3$
Pore size, $d_{\text{pore}} = 1 \text{ nm}$	Time = 7 min
Thickness of Membrane, $l_M = 5 \mu\text{m}$	Height = 0.2 m
Membrane area, $A_M = 0.5 \text{ m}^2$	Bed diameter = 0.25 m

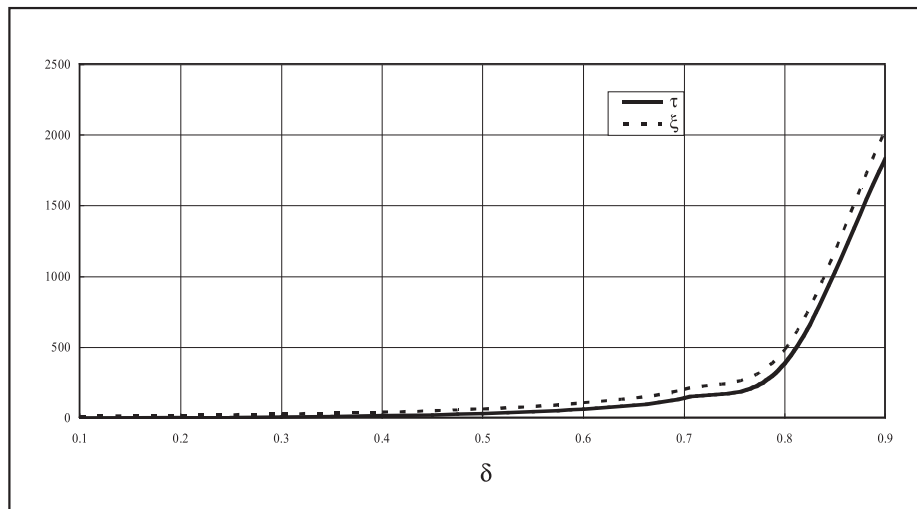


FIGURE 8. The design curve of PSA

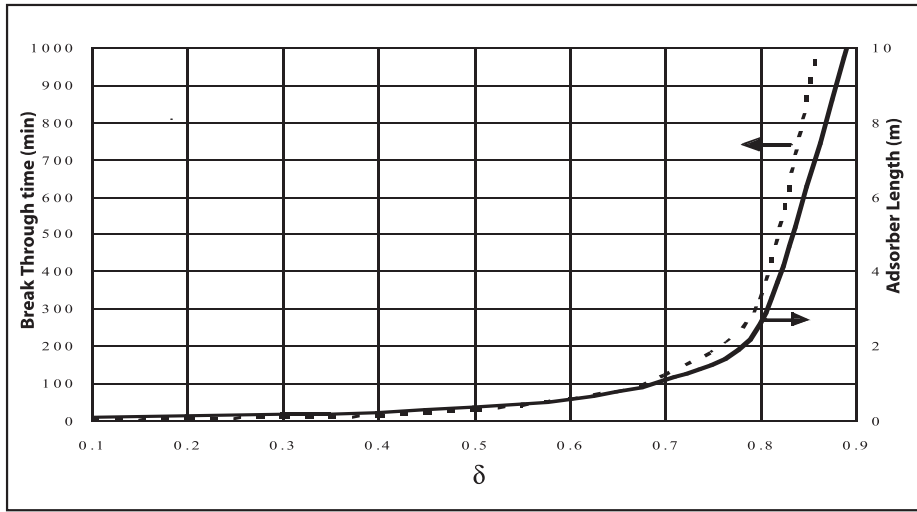


FIGURE 9. Plots show the break through time and the PSA length

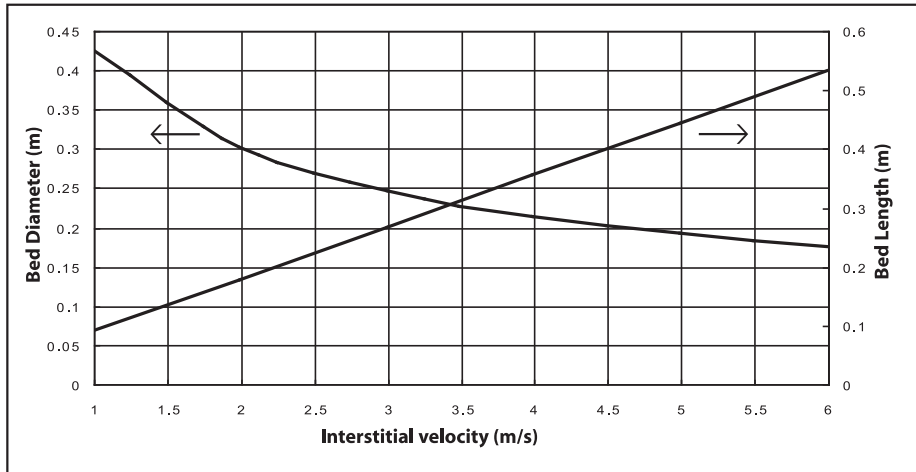


FIGURE 10. The result of adsorber length and diameter for different interstitial velocity

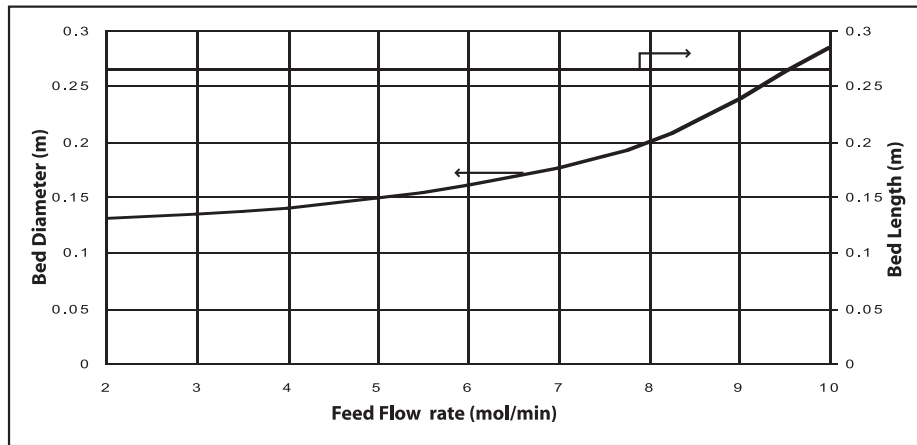


FIGURE 11. The result of adsorber length and diameter for different interstitial velocity

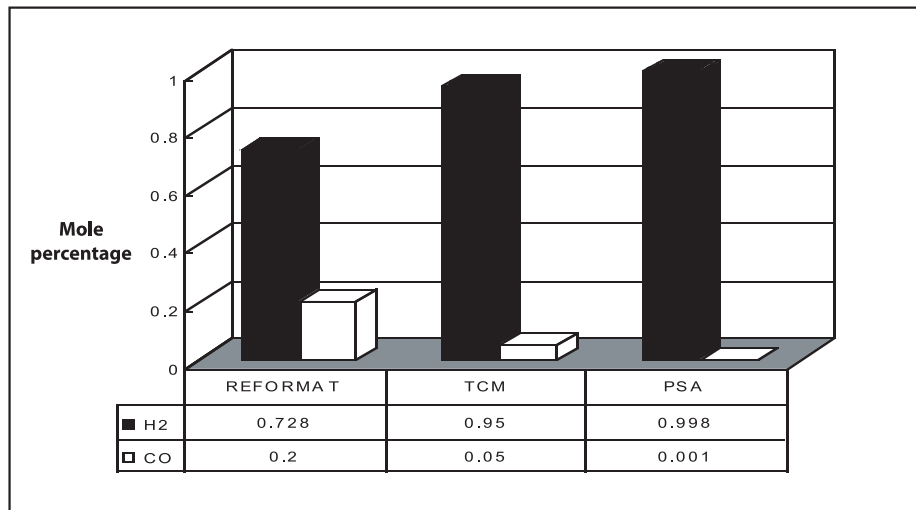


FIGURE 12. Results of the mole percentage of H₂ and CO in TCM and PSA

CONCLUSION

For membrane, it can be concluded that the length and membrane area of membrane modules for cross flow could be determined using the cross-flow model whose solution could be expressed in simplified terms of the NTU and HTU . The integral of NTU is the solution for the local permeate along the along membrane. It was found that, NTU for porous membrane mainly depends on: y_1 and x_1 , local permeate and retantate, cut, θ , actual separation factor, α , and ratio of product and feed pressure, r , along the membrane and relatively predicts the degree of separation while the HTU characterized by the physical property of the membrane.

Besides that, as a PSA process is quite complicated and there are many parameters

to be decided, this study simplified the design by using the Klinkenberg solution of the breakthrough curve for a linear driving force mass transfer and a linear equilibrium isotherm. Relationship between the dimensionless time, τ , and the dimensionless bed length, ξ , that are function of the Daud bed utilisation parameter, δ was introduced. The recommended value of δ , for PSA is about 0.2-0.3 resulting in short adsobers.

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NOMENCLATURE

$A_{M'}$	Asurface area of membrane
c	Concentration in gas phase
c^*	Equilibrium concentration
D	Diffusivity
D_L	Axial Dispersion
D_p	Bed Diameter
D_{AB}	Diffusivity of component A and B
d_p, d_{pore}	Pore diameter
D_t	Tube diameter
G	Mass flux of gas
HTU	Height of heat transfer unit
k	Mass Transfer Coefficient
K	Rate of constant

K_1	Permeability for component 1
K_{Kn}	Permeability by governed by Knudsen flow
L	Length
L_b	Bed Length
l_M	Membrane thickness
M_A	Molecular weight for component A
M_B	Molecular weight for component B
M_{AB}	Average molecular weight for component A and B
n, n_F	Feed flow rate

n_p	Permeate flow rate
n_R	Retentate flow rate
n_T	Number of tube
NTU	Number of heat transfer unit
P	Absolute pressure
P_F	Feed pressure
P_P	Product pressure
q	Concentration in adsorbed Phase
Q_F	Volumetric flow Rate Of Feed
R	Ideal gas constant
Re	Reynolds Number
Rg	Molecule radius
R	Ratio of product pressure to feed pressure
r_p	Pore radius
r_g	Gas molecule radius
Sc	Schmidt Number
Sh	Sherwood Number
T	Temperature
t	Time
t_{ideal}	Ideal Time
u	Interstitial Velocity
x	Mole fraction in local retentate
x_1	Mole fraction of component 1 local rejection
x_{1R}	Total mole fraction of component 1 in rejection flow
x_2	Mole fraction of component 2 local rejection
x_{F1}	Mole fraction of component 1 in feed stream

x_i, X_{Pi}	Mole fraction for i number of components in rejection
x_{R1}	Mole fraction for retentate
y	Mole fraction in local permeate
y_1	Mole fraction of component 1 local permeate
x_{1P}	Total mole fraction of component 1 in permeate flow
y_2	Mole fraction of component 2 local permeate
y_i, Y_{Pi}	Mole fraction for i number of components in permeate
y_{p1}	Mole fraction for permeate

Greek Symbols

α	Ideal separation factor
α^*	Actual separation factor
$\alpha_{1,2}$	Separation factor for component 1 over component 2
ε	Bed porosity
ξ	Dimensionless length
τ	Dimensionless time
τ_{ideal}	Ideal dimensionless time
τ_{Kn}	Tortuosity
μ	Gas viscosity
δ	Bed utilisation parameter
ρ	Gas density
V_A, V_B	Atomic volumes for component A and B
θ	Stage or cut = n_p/n_F
α	Separation factor
α^*	Ideal separation factor

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