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# An initial investigation of a nano-composite silica ceramic membrane for hydrogen gas separation and purification

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## Abstract

The effects of trans-membrane pressure difference on hydrogen recovery are investigated on a preliminary basis. The membrane has been developed for high temperature hydrogen separation from CH<sub>4</sub>, N<sub>2</sub>, and Ar gas molecules. Gas permeation and separation performance of the developed membrane was evaluated. The silica fabricated membrane exhibited high H<sub>2</sub> gas flux and much lower fluxes for CH<sub>4</sub>, N<sub>2</sub>, and Ar and showed a rise in permeance with the inverse of the square root of temperature for H<sub>2</sub>. Plots obtained with respect to the effects of gauge pressure on H<sub>2</sub>, N<sub>2</sub>, CH<sub>4</sub> and Ar component gas fluxes through the modified support (silica membrane) at a pressure of 1bar showed that H<sub>2</sub> recorded an almost four-fold high flux value of 0.76 mol/sec.m<sup>2</sup> compared to that of 0.33 mol/sec.m<sup>2</sup> (CH<sub>4</sub>), 0.25 mol/sec.m<sup>2</sup> (N<sub>2</sub>) and 0.22 mol/sec.m<sup>2</sup> (Ar). These results were nearly stable for the temperature range investigated (298K, 373K and 473K). In addition, hydrogen maintained a relatively high permeance value of 1.62 x 10<sup>-5</sup> mol m<sup>-2</sup> s<sup>-1</sup> Pa<sup>-1</sup> at a low pressure of 0.1bar under room temperature (298K) compared to that of N<sub>2</sub>, CH<sub>4</sub> and Ar which were much lower throughout the temperature range studied and in agreement with literature. H<sub>2</sub> separation factor (experimental Knudsen) over CH<sub>4</sub>, N<sub>2</sub> and Ar were quite close to the theoretical Knudsen and can therefore be experimentally applied through further optimization in the separation of H<sub>2</sub> as a key constituent energy for the future.

Keywords: Ceramic membrane, gas separation, permselectivity, gas permeance, dip coating

## 1. Introduction

Hydrogen as an alternative to fossil fuel can pave the way in the fabrication of various operational energy generation devices including the fuel cell [1]. Consequently high-efficiency fuel cell operation from clean hydrogen can be utilized in all energy sectors including application in transportation and distributed power. Hence, fuel cell with clean hydrogen-rich fuel is top among solutions sought for in the transitional process to a CO<sub>2</sub> free emission economy and a pathway to sustainable energy in the nearest future. Production of a cost efficient and sustainable clean hydrogen is among the top challenges which must be surmounted for the evolution from carbon based (fossil fuel) energy economy to hydrogen based economy [2]. Achieving this level of sustainable energy security and economic performance with current technologies has yielded little result in terms of balancing CO<sub>2</sub> emission abatement and the economic growth [3]. Hence the utilization of an affordable technology which will be readily available and can provide economic and clean source of energy can be set out to meet these challenges. An efficient and cost efficient technology for hydrogen/gas separation is highly required that will have a beneficial effect on the overall expenditure of the entire system. Membrane technology at the moment is being

highly applied widely in hydrogen separation because of its simplicity of operation, low energy consumption, option for uninterrupted process and known cost efficiency. In particular, inorganic ceramic membranes have shown evidence of chemical and physical properties, high temperature stability as well as unresponsiveness to sarcastic environment, homogenous pore structure and substantial tensile strength which will conquer the limitations of polymeric membranes [4] [5] [1]. Due to their inherent features, alumina-based ceramic membranes can easily separate small gas molecule like hydrogen. With their gas permeation values larger than polymeric membranes, these kind of membranes have huge prospect for applications in chemical, petrochemical and energy industry where hydrogen separation in severe conditions results to increased productivity as well as process efficiency. Consequently, porous alumina-based ceramic membranes covered with a thin selective layer manufactured by chemical vapour deposition (CVD) or sol-gel methods have attracted great attention for hydrogen separation [6] – [8]. According to most literatures [9] - [12], silica membranes prepared by different methods like CVD or sol-gel, deposited on mesoporous or macroporous supports have been shown to be effective for H<sub>2</sub> permeation with good selectivities. In their work, Oyama and Lee [11] and [12] used chemical vapour

deposition method to deposit silica on  $\gamma$ -alumina supports with pore diameter of 4 nm and obtained silica membranes with high selectivities of hydrogen for temperatures less than 900 K. Yan et al. [13] used chemical vapour deposition to add silica layer to a macroporous  $\alpha$ -alumina support which had 110–180 nm pore diameter with three layers of  $\gamma$ -alumina. They obtained a membrane with selectivity for  $H_2/N_2$  of 100–1000 and a  $H_2$  permeance of  $10^{-8} - 10^{-9} \text{ mol m}^{-2} \text{ s}^{-1} \text{ Pa}^{-1}$  at 873 K. Oyama et al. [14] reported that preparing a composite layer by silica and an inorganic oxide like zirconia ( $ZrO_2$ ) gives rise to membrane stability with superior permeation properties. Nomura et al. [15] found an improvement of steam stability of a silica membrane. Although this membrane had a good selectivity for  $H_2/N_2$  of over 800 at 773 K, the permeance was low and in the order of  $2 - 7 \times 10^{-8} \text{ mol m}^{-2} \text{ s}^{-1} \text{ Pa}^{-1}$ . In another work, [14] Lee et al. [16] prepared membranes with silica layer on the outer surface of a mesoporous alumina support and obtained a  $H_2$  permeance of  $1.2 \times 10^{-7} \text{ mol m}^{-2} \text{ s}^{-1} \text{ Pa}^{-1}$  and a selectivity of  $H_2$  to  $CH_4$  of 2800 at 873 K. Gu and Oyama [17], and [18] effectively produced a defect-free  $\gamma$ -alumina multilayer membrane with an ordered structure by successively placing boehmite sols of different particle sizes on a macroporous alumina support. They now coated the surface of the  $\gamma$ -alumina with a thin-silica by chemical vapour deposition method. The resulting novel silica-on-alumina membranes had excellent permeability of  $5 \times 10^{-7} \text{ mol m}^{-2} \text{ s}^{-1} \text{ Pa}^{-1}$  and good selectivity for hydrogen over  $CH_4$  of 1500–5900 at 873 K. The resulting supported composite silica–alumina membrane has high permeability for hydrogen in the order of  $10^{-7} \text{ mol m}^{-2} \text{ s}^{-1} \text{ Pa}^{-1}$  at 873 K with a selectivity of  $H_2$  over  $CH_4$  of 940. However, silica membrane fabrication using similar methods as stated above by deposition on support of large pore (macro porous) are known for their effectiveness with excellent  $H_2$  permeation selectivity outcomes [19][20] [11][12]. Again, the difference in the flow rate and permeation features during the separation processes involving multiple component gases makes silica ceramic membrane a good contender for  $H_2$  gas separation [21]. However the main features in the selection of silica based membranes are their hydrogen separation ability and large hydrogen permeability [17].

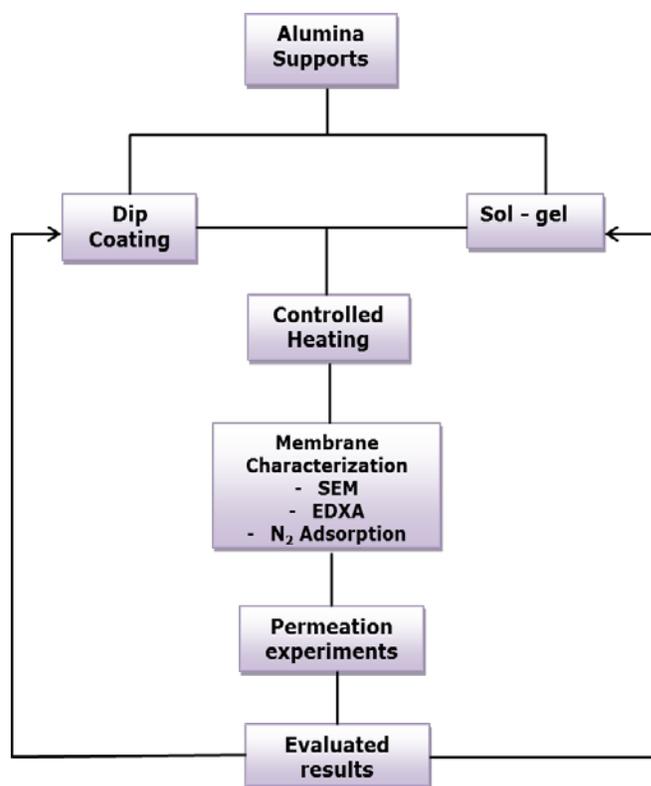
In this work, a preliminary experiment has been performed using a novel nano-porous silica ceramic membrane manufactured through another alternative, dip coating method. The membrane separates hydrogen gas from  $N_2$ , Ar, and  $CH_4$ . In particular, the membrane can be applied to separate hydrogen at low-pressure from gas streams and also at elevated temperatures. The process of preparation of the membrane and its description, pure gas fluxes and permeance through the membrane, as well as its industrial application are reported. The morphology of the silica membrane layer is analysed by scanning electron microscopy. However, the transport properties of the

membranes were obtained in the temperatures of 298K, 373K and 473K and at pressure differences ranging from 0.1 to 1 bar.

## 2. Experimental methods

### 2.1 Preparation of a nano-composite ceramic membrane

The nano-composite membrane in this work was prepared by depositing a very thin, dense layer composed of silica based solution on a macro-porous alumina support for the purpose of modification. Figure 1 shows the schematic diagram of the sequential steps that are employed to carry out synthesis and characterization of a membrane before permeation tests takes place.



**Figure 1: Membrane preparation steps**

The commercial alumina (SCT, France) tube, i.d. = 7 mm, o.d. = 10 mm) with a nominal pore size of 15 nm and surface area of  $0.0062 \text{ m}^2$  was used as the support and has been modified by dip coating technique. This dipping process used a silica-based solution comprising of 900mls of 2-methylbutane, 100mls of silicon elastomer and 10mls of a curing agent. The mixture of Iso pentane, silicone elastomer and curing agent (which prevents a cross linking between silica molecules) were mixed together thoroughly using a magnetic stirrer for homogeneity purposes. A fresh ceramic

support was immersed into the solution prepared and left for about 30mins; it was then withdrawn and placed on a rotating device for approximately 1hour for air drying. After this, it was put in an oven at a temperature of 65°C for another 2 hours. This process is a patented innovation [22] [23]. In addition the entire procedure was done to achieve the desired affinity of the membrane for the gas of interest, in this case hydrogen.

A picture of a commercial alumina membrane arrangement is shown in Figure 2.



(a)

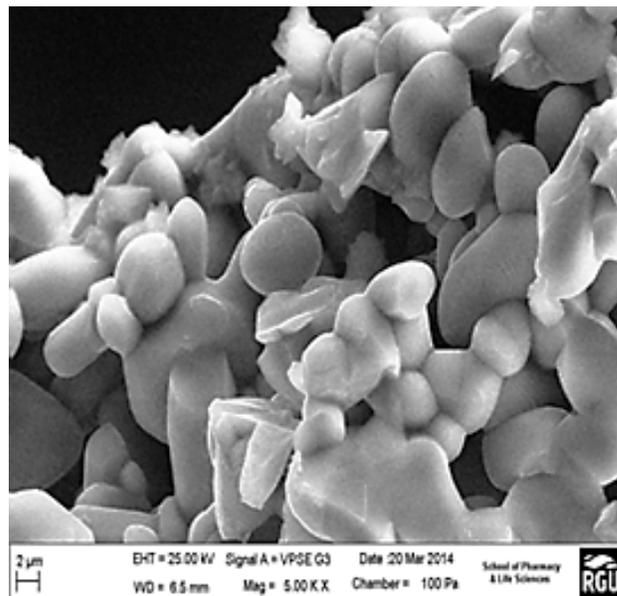


(b)

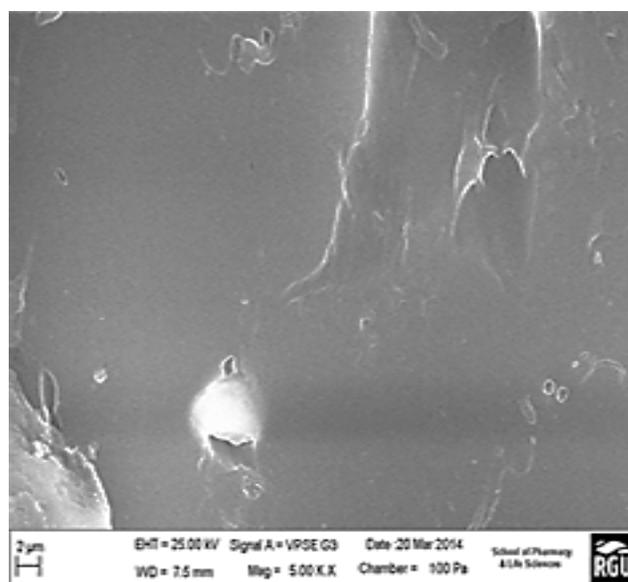
**Figure 2: A photograph of the commercial alumina membrane arrangement (a) Membrane outer surface (b) Membrane inlet ports**

### 2.2 Morphological characterization of the membrane

Surface SEM images obtained from the porous support and top selective composite layers are shown in Figures 3 and 4. Figure 3 depicts a porous alumina graded structure while Figure 4 shows a layer which is coated on the surface of a macro-porous alumina support forming a thin silica layer obtained through dipping. The support has a mean pore size of about 15 nm which is in agreement with the nominal value reported by the supplier. As shown in Figure 4, the top selective composite layer deposited on top of intermediate multilayer has a uniform structure. These SEM images indicate that gas permeation through various layers of the membrane can be rate limiting because of a fundamental difference in the structural make-up of the membrane layers.



**Figure 3: SEM image showing a porous alumina support**



**Figure 4: SEM image of a thin silica layer deposited on outside layer of support**

### 2.3. Gas permeation test

The experimental set up for gas permeation is shown in figure 5. The modified ceramic membrane was inserted inside a tubular metallic casing which was wrapped up with a heating tape connected to a power controller. Permeation tests were carried out to investigate the transport through the thin top selective layer with a nano-structure. Prior to gas permeation test, the temperature was allowed to stabilize. Membrane temperature, gas flux and pressure drop were important factors considered for separation. Gas permeation experiment was performed at a temperature range 298K –

473K and pressure between 0.1 – 1bar. Single gas flow of four carrier gases namely hydrogen (H<sub>2</sub>), nitrogen (N<sub>2</sub>), argon (Ar) and methane (CH<sub>4</sub>) were passed individually through the shell side of the membrane while obtaining the flow rate at the permeate side of the membrane to determine their fluxes and the membrane selectivity characteristics. The flow rates of the permeated gases were measured using a flow meter while thermocouples and pressure gauges were used to measure the temperature and the pressure respectively. The retentate side was closed while the experiment was being carried out.

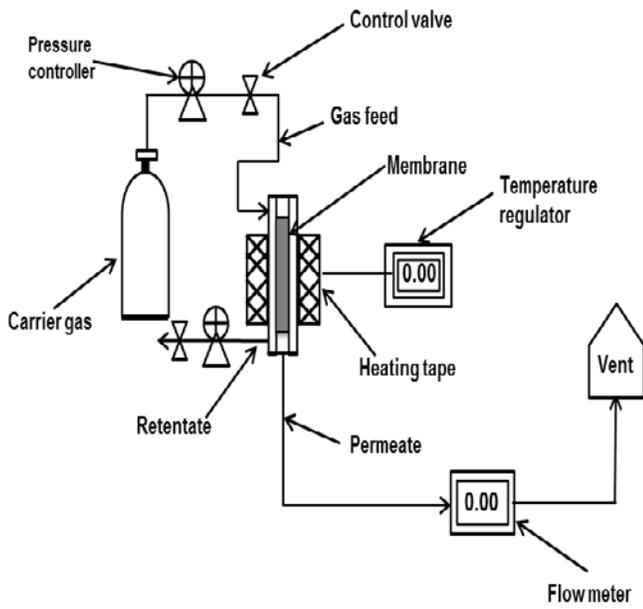


Figure 5: The experimental set-up for hydrogen gas separation

### 3. Results and discussions

#### 3.1. Effect of feed pressure on gas flux

A distinctive feature is observed in gas flux through the composite membrane (see Figures 6 - 8). This was done by determining the flux  $J$  (mol m<sup>-2</sup> s<sup>-1</sup>), the governing equation is given as:

$$\text{Flux, } J = \frac{\text{flow rate } \left[ \frac{\text{mol}}{\text{sec}} \right]}{\text{membrane surface area (m}^2\text{)}} \quad (1)$$

Where  $J$  is the gas flux across the membrane, mol.s<sup>-1</sup>m<sup>-2</sup> given as the ratio of gas flow rate to the membrane surface area measured as 0.0062m<sup>2</sup>.

The plots in figures 6, 7 and 8 presents the effects of gauge pressure on H<sub>2</sub>, N<sub>2</sub>, CH<sub>4</sub> and Ar component gas fluxes through the modified support at temperatures of 298 K, 373 K and 473 K respectively. As can be noticed from the graphs, H<sub>2</sub> recorded the highest flux as expected due to its lower molecular weight whereas Ar with a higher molecular weight of 40 recorded the lowest flux. Methane (CH<sub>4</sub>) and N<sub>2</sub> with molecular weights of 16 and 28 respectively are seen to fall between the lowest flux (Ar) and highest flux (H<sub>2</sub>) with CH<sub>4</sub> flowing faster than N<sub>2</sub>. From the plots also, we can generally say that the results present a better representation of the linear relationship between flux and pressure since the regression lines are well-aligned showing higher correlation of almost 1 for all gases. Hence, it can be inferred that Knudsen type of transport mechanism is dominant as the smaller molecules tend to collide more towards the membrane pore wall than having interaction with one another. This further confirms that the mechanism in the flow is molecular weight dependent as can be noticed. The statistical error determination for the effect of pressure on the H<sub>2</sub>, N<sub>2</sub>, CH<sub>4</sub> and Ar fluxes for the range of temperatures and pressures investigated are shown in Figures 6a, 7a and 8a.

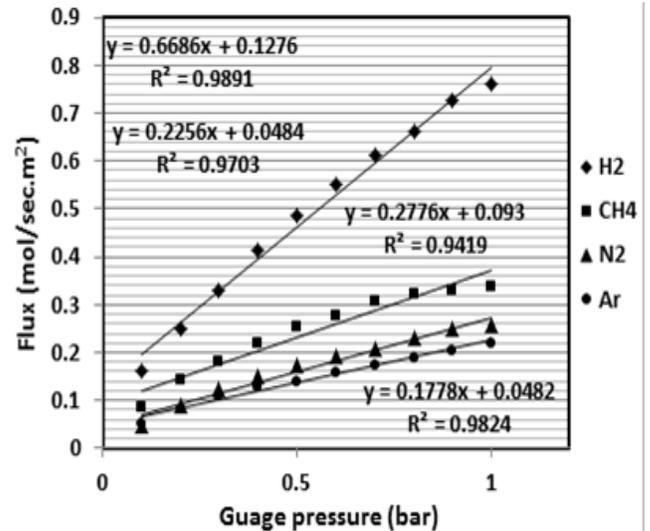


Figure 6: Pressure dependence of H<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub> and Ar gas fluxes at 25°C

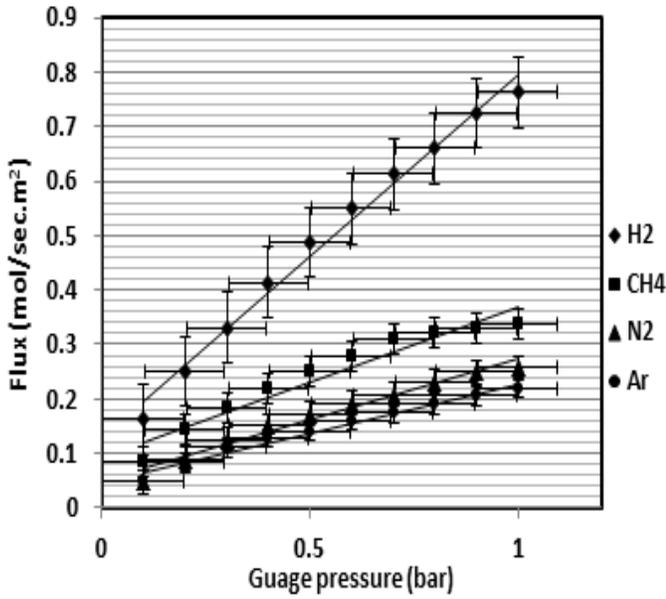


Figure 6a: Statistical error determination of the effect of gauge pressure on H<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub> and Ar gas fluxes at 25°C

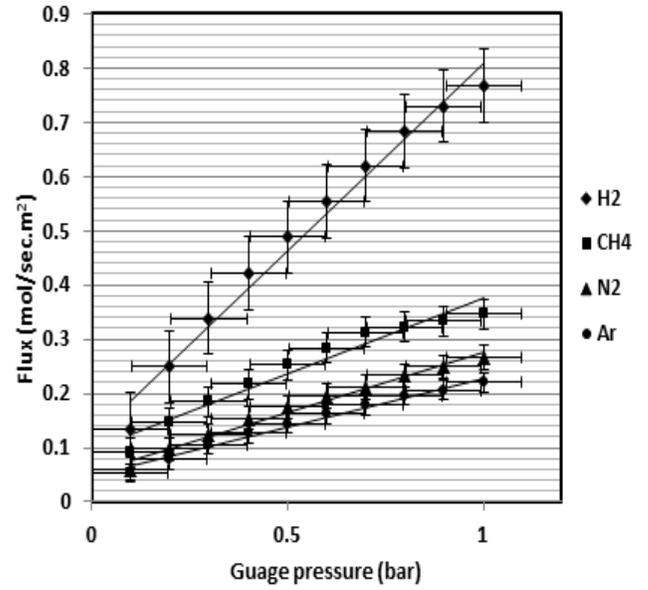


Figure 7a: Statistical error determination of the effect of gauge pressure on H<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub> and Ar gas fluxes at 100°C

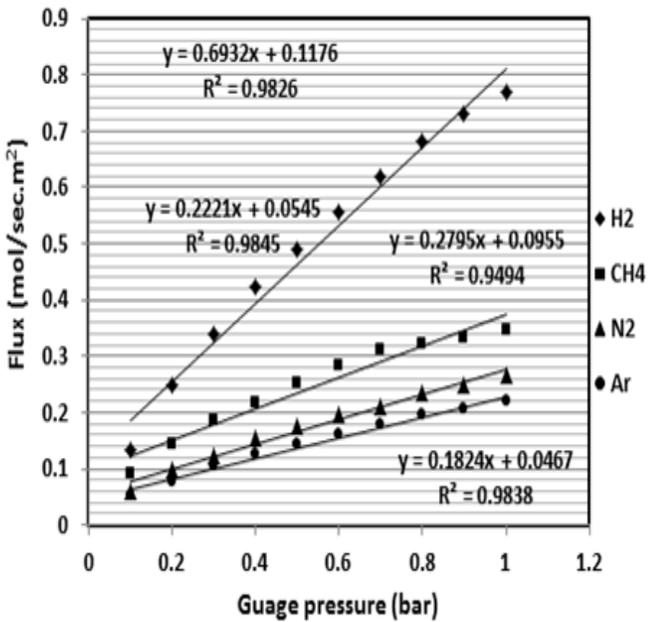


Figure 7: Pressure dependence of H<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub> and Ar gas fluxes at 100°C

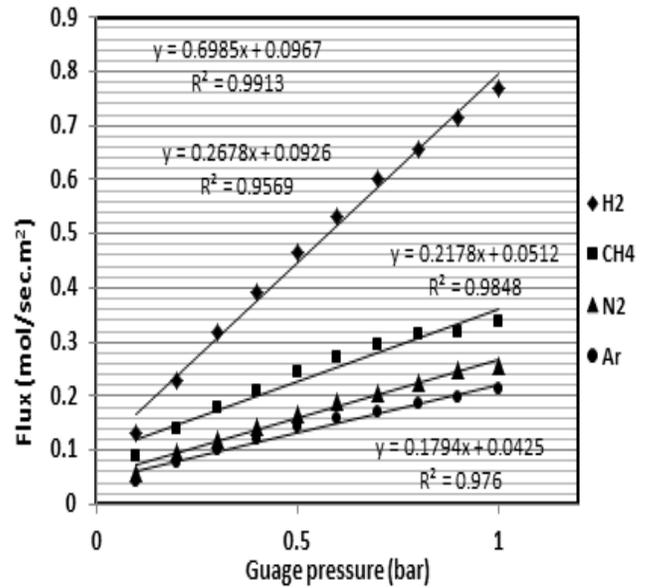
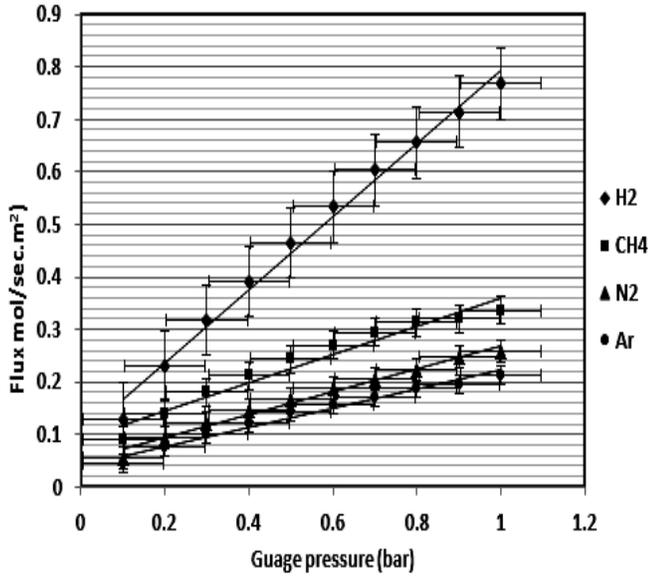


Figure 8: Pressure dependence of H<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub> and Ar gas fluxes at 200°C



**Figure 8a: Statistical error determination of the effect of gauge pressure on H<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub> and Ar gas fluxes at 200°C**

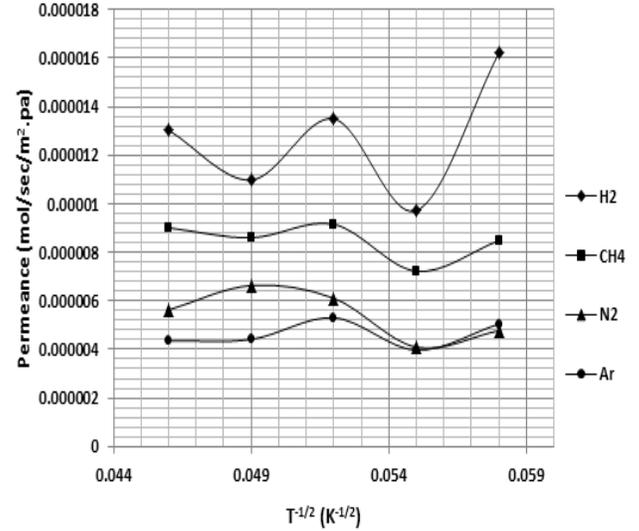
### 3.2. Effect of feed pressure on gas permeance

The performance of inorganic ceramic membranes is typically described by the permeability and permselectivity parameters. Permeability, is the flux normalized against the transmembrane driving force (pressure difference) and the membrane thickness ( $\text{mol} \cdot \text{m} \cdot \text{m}^{-2} \cdot \text{s}^{-1} \cdot \text{Pa}^{-1}$ ). The membrane thickness was not readily available for the silica membranes and so permeance,  $Q$  ( $\text{mol} \cdot \text{m}^{-2} \cdot \text{s}^{-1} \cdot \text{Pa}^{-1}$ ), have been determined instead. Gas permeance was obtained from the following expression:

$$Q = \frac{F}{A \cdot \Delta P} \quad (2)$$

Where  $Q$  is the Permeance ( $\text{mol} \cdot \text{m}^{-2} \cdot \text{s}^{-1} \cdot \text{Pa}^{-1}$ );  $F$  is the molar flow ( $\text{mol}/\text{sec}$ );  $A$  is the membrane area ( $\text{m}^2$ ); and  $\Delta P$  is the pressure difference (Pa) across the membrane. The membrane performance for various gases with inverse of the square root of temperature at 0.1bar and 1bar is given in Figures 9 and 10 respectively. The total available membrane area calculated for the membrane was  $0.0062\text{m}^2$ . Hydrogen displayed a maximum and minimum values between 0.1bar and 1bar but showed a relatively high permeance of  $1.62 \times 10^{-5} \text{mol} \cdot \text{m}^{-2} \cdot \text{s}^{-1} \cdot \text{Pa}^{-1}$  and  $0.76 \times 10^{-5} \text{mol} \cdot \text{m}^{-2} \cdot \text{s}^{-1} \cdot \text{Pa}^{-1}$  respectively. CH<sub>4</sub> permeance was higher than that of N<sub>2</sub> permeances. However, N<sub>2</sub> permeated faster than Ar but had same permeance of  $0.4 \times 10^{-5} \text{mol} \cdot \text{m}^{-2} \cdot \text{s}^{-1} \cdot \text{Pa}^{-1}$  at exactly  $0.055 \text{K}^{-1/2}$ . This is a very remarkable. At a higher pressure of 1bar, H<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub> and Ar permeances through this

membrane were almost constant throughout the temperature range studied while hydrogen still maintained the highest permeance. These results suggest that the gas transport mechanism of the coated support is mainly due to the Knudsen diffusion.

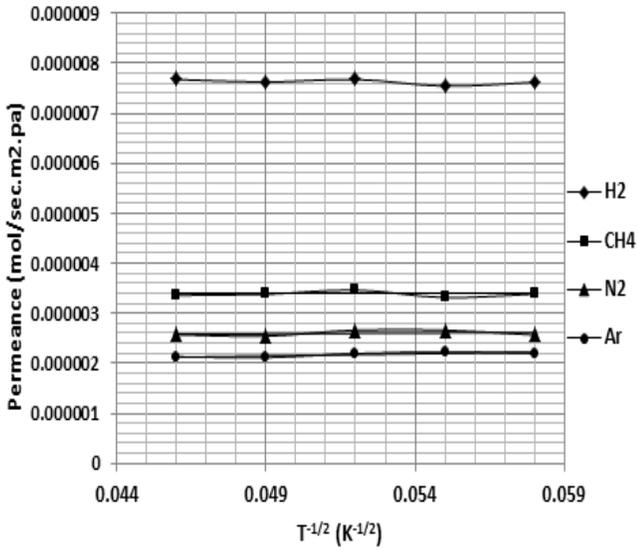


**Figure 9: Relationship of the inverse square root of temperature on gas permeance of the nano composite at pressure of 0.1bar**

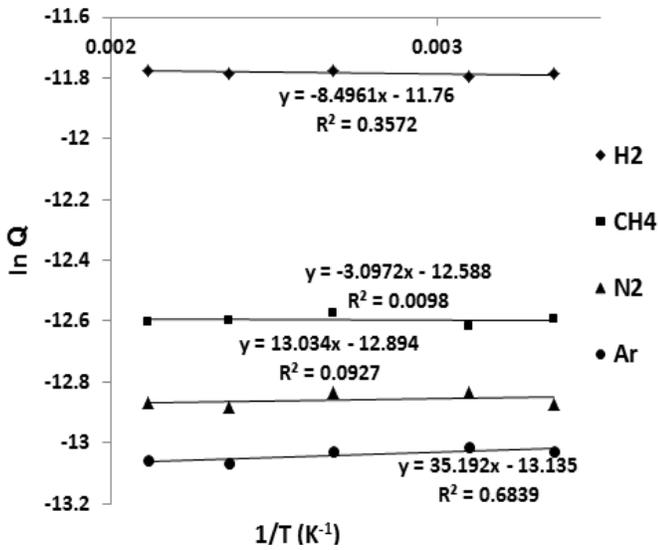
The activation energy of permeation was obtained by fitting the experimental permeance data shown in figure 10 to an Arrhenius type expression [16]:

$$Q = Q_0 \exp\left(\frac{-E_a}{RT}\right) \quad (3)$$

Where  $Q$  is the permeance,  $Q_0$  is the pre-exponential factor ( $\text{mol} \cdot \text{m}^{-2} \cdot \text{s}^{-1} \cdot \text{Pa}^{-1}$ ),  $E_a$  is the activation energy ( $\text{KJ}/\text{mol}^{-1}$ ),  $R$  is the gas constant ( $8.314 \text{J}/\text{mol}^{-1} \cdot \text{K}^{-1}$ ) and  $T$  is the temperature (K). The experimental permeance data do not indicate a good fit to the Arrhenius equation. The activation energy values are presented in Table 3 as shown in figure 10a where the  $\ln Q$  is plotted against the inverse of the temperature. The correlation coefficients obtained are too low and therefore do not suggest either an activated or an adsorptive transport mechanism for these gases.



**Figure 10: Relationship of the inverse square root of temperature on gas permeance of the nano composite at a pressure of 1 bar.**



**Figure 10a: Activation energy plot of permeance vs the inverse of temperature of the nano composite at a pressure of 1 bar.**

### 3.3. Membrane perm selectivity

Permeability,  $S_{xy}$  is the ratio of the permeance of two different gas species as given by:

$$S_{xy} = \frac{Q_x}{Q_y} \quad (4)$$

Where  $S_{xy}$  is the permselectivity of  $x$  to  $y$ ;  $Q_x$  is the permeance of  $x$  ( $\text{mol m}^{-2} \text{s}^{-1} \text{Pa}^{-1}$ );  $Q_y$  is the permeance of  $y$  ( $\text{mol.m}^{-2} \text{s}^{-1} \text{Pa}^{-1}$ )

Tables 1 and 2 shows the experimental selectivity in relation to theoretical Knudsen selectivity for the temperature and pressure range investigated. The theoretical selectivity of component  $x$  over  $y$  is given by equation 5:

$$S_{x/y} = \sqrt{\frac{M_y}{M_x}} \quad (5)$$

where  $M_y$  represents the molecular weight of  $\text{CH}_4$ ,  $\text{N}_2$  or  $\text{Ar}$  while  $M_x$  is the molecular weight of  $\text{H}_2$  being the target gas. The permselectivity achieved using equation (4) is relatively close to those obtained using equation (5).

Table 1 and 2 depicts  $\text{H}_2$  theoretical and experimental selectivity over  $\text{CH}_4$ ,  $\text{N}_2$  and  $\text{Ar}$  at pressure of 0.1bar and 1bar respectively at various temperatures. Overall results shows that at a pressure of 0.1bar, the experimental Knudsen selectivity of  $\text{H}_2$  gas to  $\text{CH}_4$ ,  $\text{N}_2$  and  $\text{Ar}$  decreased as the temperature was increased, however the highest  $\text{H}_2$  experimental selectivity was obtained at 298K. This was also the closest to the theoretical Knudsen obtained with  $\text{H}_2$  selectivity to  $\text{N}_2$  having its experimental selectivity value to be 3.41 whereas the theoretical Knudsen selectivity is 3.74. This is presented in Table 1. Further increment to a pressure of 1bar resulted in  $\text{H}_2/\text{Ar}$  and  $\text{H}_2/\text{CH}_4$  experimental selectivity value of 3.61 and 2.28 respectively whereas the theoretical Knudsen selectivity value is 4.47 and 2.8 at 473K respectively. These results are therefore reasonably practicable; nevertheless, there is a possibility that more  $\text{H}_2$  can be recovered from these gases at higher pressure. But use of higher pressures is not practical in fuel cell due to balance of plant limitations.

**Table 1: Relationship between theoretical selectivity and experimental permselectivity pressure of 0.1 bar**

At 0.1 bar				
	Theoretical Knudsen	Experimental Knudsen		
		298K	373K	473K
$\text{H}_2/\text{CH}_4$	2.8	1.91	1.47	1.45
$\text{H}_2/\text{N}_2$	3.74	3.41	2.21	2.32
$\text{H}_2/\text{Ar}$	4.47	3.21	2.54	2.31

**Table 2: Relationship between theoretical selectivity and experimental permselectivity pressure of 1 bar**

At 1 bar				
	Theoretical Knudsen	Experimental Knudsen		
		298K	373K	473K
H <sub>2</sub> /CH <sub>4</sub>	2.8	2.25	2.22	2.28
H <sub>2</sub> /N <sub>2</sub>	3.74	2.97	2.88	2.98
H <sub>2</sub> /Ar	4.47	3.47	3.51	3.61

**Table 3: Correlation of pre-exponential factor and activation energy for Arrhenius gas permeation through membrane at 1.0 bar**

Gas	Pre-exponential factor Q <sub>0</sub> (molm <sup>-2</sup> s <sup>-1</sup> pa <sup>-1</sup> )	Activation Energy (KJmol <sup>-1</sup> )	Sum of least squares Σ(Q <sub>calc</sub> -Q <sub>expt</sub> ) <sup>2</sup>
H <sub>2</sub>	7.8 x 10 <sup>-6</sup>	70.6	1.39 x 10 <sup>-9</sup>
CH <sub>4</sub>	3.4 x 10 <sup>-6</sup>	25.7	2.81 x 10 <sup>-10</sup>
N <sub>2</sub>	2.5 x 10 <sup>-6</sup>	-108.36	1.80 x 10 <sup>-10</sup>
Ar	2.0 x 10 <sup>-6</sup>	-292.59	1.40 x 10 <sup>-10</sup>

#### 4. Conclusion

An initial investigation have been carried out on a composite membrane with a graded structure, successfully synthesized and tested for separating and purifying hydrogen from other gases such as methane, nitrogen and argon at high operating temperatures. This membrane was prepared by depositing a thin layer composed of silica based solution on top of a graded substrate through dip coating technique method. SEM images obtained from surface section of the membrane indicated formation of a thin defect-free top selective layer which is responsible for the improved membrane performance by carefully controlling the operation parameters. The permeation characteristics of the gas molecules investigated confirmed very high H<sub>2</sub> flux of 0.16 mol.s<sup>-1</sup>.m<sup>-2</sup> at low pressure of 0.1 bar with a relatively high permeance of 1.62 x 10<sup>-5</sup> mol m<sup>-2</sup> s<sup>-1</sup> Pa<sup>-1</sup> slightly higher than that obtained in literature and the gas transport mechanism showed Knudsen diffusion. For optimization purposes, further experiment will be carried out at a higher pressure and at elevated temperature to achieve higher H<sub>2</sub> selectivity.

If we compare the performance of the support and the composite silica/alumina, it is observed that gas permeation through the composite membrane has decreased drastically but H<sub>2</sub>/N<sub>2</sub> selectivity has increased from 1.0 to 3.41 which compares with the theoretical Knudsen value of 3.74. We have therefore succeeded in preparing a porous silica

composite membrane with high gas flux. However due to non-uniform pore size, high selectivity of hydrogen could not be achieved. Improvement in the hydrogen selectivity can be through for example, better control in the grain size and washcoating using TiO<sub>3</sub>. Although the measured hydrogen selectivity is low, the flux or throughput is relatively high and a cascade of composite silica membrane is an attractive option for concentrating hydrogen for commercial use including fuel cell. The effect of temperature on the permeance does not support either an activated or adsorptive transport mechanism due to the relatively low correlation coefficient.

#### Nomenclatures

E <sub>a</sub>	Activation energy (KJmol <sup>-1</sup> )
F	Flow rate (mol.sec-1)
J	Flux ( mol.sec <sup>-1</sup> .m <sup>2</sup> )
R	Gas constant (Jmol <sup>-1</sup> .k <sup>-1</sup> )
A	Membrane surface area (m <sup>2</sup> )
M	Molecular weight (g)
Q	Permeance ( mol.sec <sup>-1</sup> .m <sup>-2</sup> .Pa <sup>-1</sup> )
ΔP	Pressure difference (pa)
S <sub>xy</sub>	Selectivity of x to y
T	Temperature (K)

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