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A Novel Performance of Ordered Porous Ceramic Materials in Hydrogen Recovery Processes for Sustainable Energy Potential

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Abstract: ***The main objective of this work is to investigate the permeation behaviour of single gases N_2 and CO_2 and H_2 through a commercial alumina modified ceramic membrane using the dip coating method. The flow rates of the single gases were investigated at room temperature. Results obtained show a linear proportionality of gas permeance to pressure. Experiments show that N_2, H_2 and CO_2 flow rates are clearly governed by Knudsen diffusion. The single gas permeation at varying pressures shows linear fluxes with respect to inverse square root of the molecular weight with H_2 recording the highest flux of 303.785mol.m^2 /sec.***

Keywords: Hydrogen, Porous Alumina Membranes, Gas flux, Knudsen diffusion.

I. INTRODUCTION

An estimated 2 billion tons of CO_2 is currently being emitted per year from existing coal-fired power plants for energy generation through fossil fuel consumption. This is compelling the entire globe to shift away from fossil fuel custom and in pursuit of viable energy sources with desirable characteristics of renewable energy for the future of a habitable environment. [1] [2]. Accordingly, there is a growing evidence and awareness that hydrogen purification and production as an alternative energy source can drive down global reliance on fossil fuel and avoid energy catastrophe in future. In fact hydrogen generation through renewable sources are eco friendly making it the right candidate in the present scenario and termed as a fuel for the future [2]. Currently, processed crude oil is utilized for powering vehicles and combustion engines causing environmental pollution through greenhouse emissions. However, hydrogen has been identified as having the ability to overcome the challenges associated with a sustainable energy future. Utilization of Hydrogen is applicable in various areas, for example, petrochemical and steel industries, chemical plants and in crude oil refining operations and so it is predicted that its necessity in the near future is unavoidable and massive [3]. In the transitional period, fossil fuel feedstock sources can be used for production of hydrogen for a successful evolutionary hydrogen economy through thermo chemical processes as an important technology. This can also be achieved through a traditional natural gas steam reforming involving a water-shift reaction. Moreover, due to CO_2 and H_2 as

the by-products of the reaction, carbon emissions are prevalent, therefore with carbon capture to produce clean hydrogen, decarbonised emission is realistic [4] [5].

The discovery of a process of low energy saving to achieve a reasonable and effective separation performance is an issue of global concern. Due to their profitable and elemental engineering advantages over conventional separation technologies, membrane process are now being utilized for capture of CO_2 emissions from power plants process operations. They are further applied in hydrogen production through selective gas separation technique [1].

Generally, H_2-separation membrane can be designed using materials like silica, alumina and palladium. Furthermore palladium membrane formed on porous alumina support by electro less plating method as reported by some researcher shows excellent H_2-permselectivity, but at a relatively high temperature, a critical and unstable condition occurs. On the other hand ceramic materials are stable at high temperature and yield good results [3].

II. Membrane Separation Process

The conventional significance of the word membrane is allied to the theory of a device which is able to disallow permeation of some constituents in a selective manner. Inorganic membranes can be categorized as dense, porous or composite. This classification is based on the membrane structure in relation to the interaction between the permeating gas molecule and the membrane surface leading to the determination of permeability and selectivity characteristic features as well as the transport mechanism [6]. In a related literature [7], the process of gas separation using porous membranes is primarily based on the differences of the molecules of the permeating gases, simply put that larger gas molecules will usually not pass through membrane of smaller pore sizes. They proposed a new method for the design of meso porous membranes. In contrast some other researcher have proposed that mesoporous membrane material which has a relatively low resistance to gas flow across the membranes is covered with a thin metallic coating which offers composite membranes which have a combination of reasonably high permeation and selectivity features. Moreover some membranes can be fabricated using modified sol-gel techniques to form microporous silica membranes of even smaller pore size with higher selectivity and yields better results. [6].

A long-lasting, cost-effective and energy saving technology of CO_2 capture from flue gas has resulted from different researchers finding solution to the problem of climate change. Consequently, a novel inorganic ceramic support modified with silica has shown exceptional evidence of chemical and physical stabilities. The membrane can withstand high temperature and resistance to harsh environment, homogenous pore structure, high tensile strength and proven to be a viable substitute to conventional amine methods in H_2 gas separations even at a relatively low temperature and pressure. Apparently this results in a well-defined permeance and selectivity features which surmount the limitations of polymer membrane [8]. This fundamental membrane features will be used in this article to tackle the questions on how to solve global energy security problems through gas separations for hydrogen recovery. Figure 1 shows a simplified concept of membrane separation process with three main sections, namely the feed inlet, retentate and the permeate side.

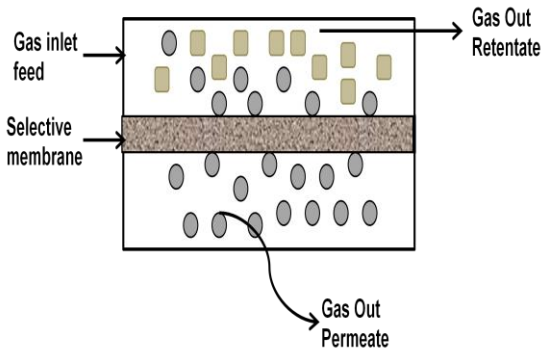

Fig 1: A simplified concept of membrane separation process

III. EXPERIMENTAL

In this study, gas transport through a porous inorganic ceramic membrane was performed using single gases namely hydrogen (H_2), nitrogen (N_2) carbon dioxide (CO_2), at room temperature (298K) and varying gauge pressure of 1-5(Bar). A 15nm pore size commercial alumina ceramic membrane with an effective permeable length of 358mm, outer and inner diameter of 10mm and 7mm respectively and membrane surface area of 0.0062m^2 wasnused for the permeation experiments. Figure 2 depicts a nano-stuctured composite after modification through dip-coating technique.

Fig 2: Nano-structured Composite

The separation of hydrogen from other gases by means of membrane technology is essential especially in the production of pure hydrogen in an efficient and economical manner. Figure 3 shows a schematic diagram of carbon capture for hydrogen production through a membrane. In order to achieve hydrogen of high purity from the products of the water–gas shift reaction, for example separation of H_2 from either CO or CO_2 is required. This can then be carried out in the permeations cell as shown in Figure 3.

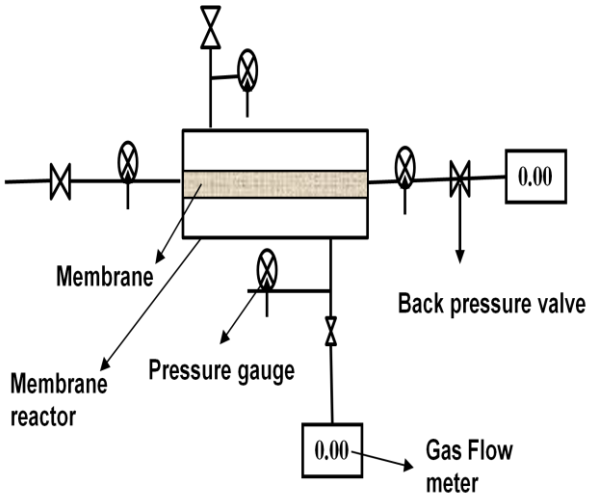

Fig 3: Gas permeation experimental set up for Hydrogen Production and Purification through a Membrane inside a membrane reactor

IV. RESULTS AND DISCUSSION

Gas permeance $mols^{-1}m^{-2}$ Pa^{-1} can be calculated using the formula in equation 1 below

$$P_i = \frac{F_i}{A_m \Delta P_i}$$

Where, P_i = gas permeance for gas i (mol m^{-2} s^{-1} Pa^{-1}), = F_i gas flow rate for gas *i* in moles per second A_m = membrane surface area (m^2), ΔP_i = pressure difference of gas *i* in bar [9].

The graph in figure 4 is a plot of the flow rate of H_2, CO_2, and N_2 gases versus pressure drop. From the investigation, an increase in the pressure drop increased gas flow rate. Furthermore, hydrogen gas of lower molecular weight permeated faster with increase in feed pressure through the ceramic inorganic membrane than the other three single gases. The high flow rate of hydrogen suggests its relatively smaller molecular weight in comparison to the other gases which provides the ease in movement within the pore network of the membrane.

Figure 4 also confirms the membrane performance with respect to the gases that permeates faster than the others through the membrane.

A clear dissimilarity is illustrated in Figure 5. Even though the transport mechanism supported Knudsen diffusion, an increase in the pressure resulted in a decrease in gas permeance. This analysis is in agreement with a study on 'Gas permeation through PTFE porous membrane [10]

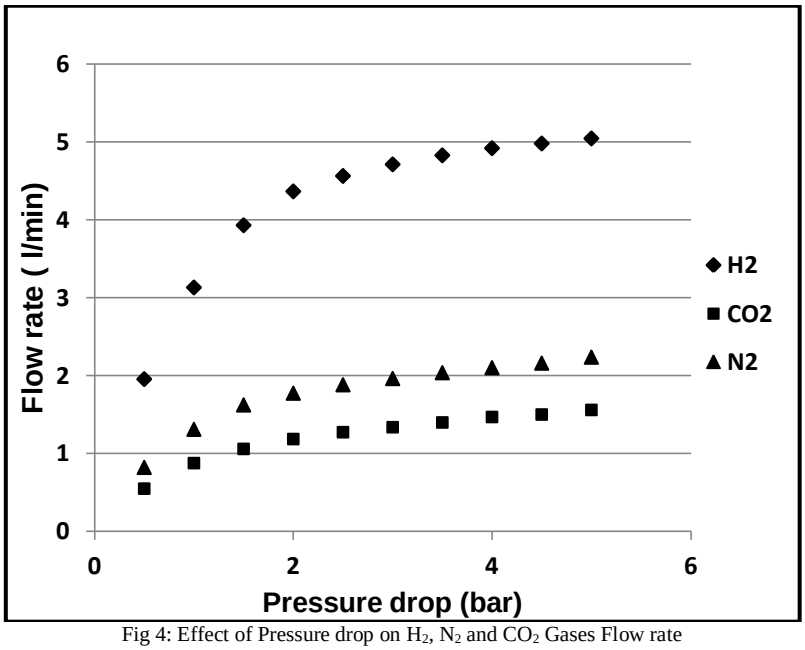

Fig 4: Effect of Pressure drop on H_2, N_2 and CO_2 Gases Flow rate

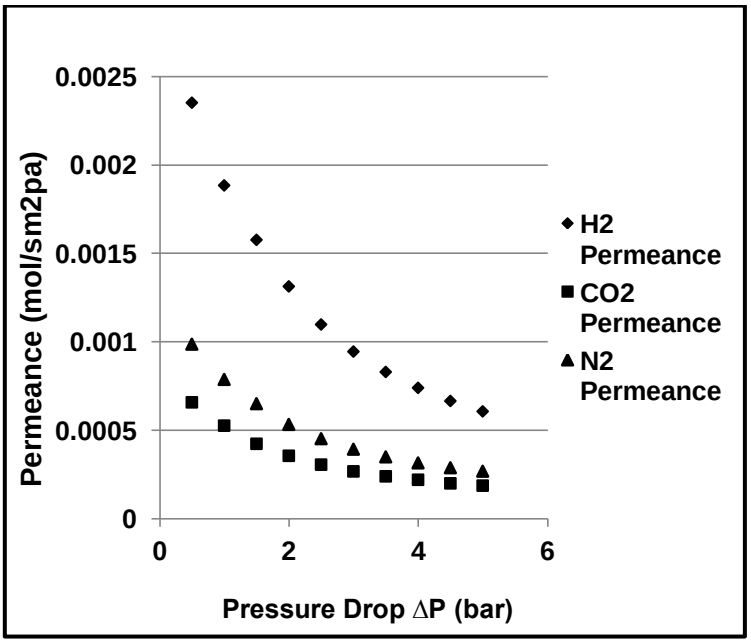

Fig 5: Effect of Pressure drop on H_2, N_2 and CO_2 Gases permeance

Figure 6 presents the flux of H_2, N_2 and CO_2 as a function of inverse of the square root of their molecular weights. The experiment was carried out at varying pressures of 1, 3 and 5bars. Flow rates of the permeating gases were measured and recorded using a flow meter. The dip-coated membrane displayed a linear prportionality dependene to the inverse square root of molecular weight. This is in accordance with results of a study on synthesis and characterization of mesoporous silica membrane via dip-coating and hydrothermal deposition technique [11]. As expected the analysis of the result is dominated by knudsen diffusion regime. H2 gas with a lower molecular weigh of 2 had the highest flux nearly three times more than that of N_2 and CO_2 at a relatively low pressure.

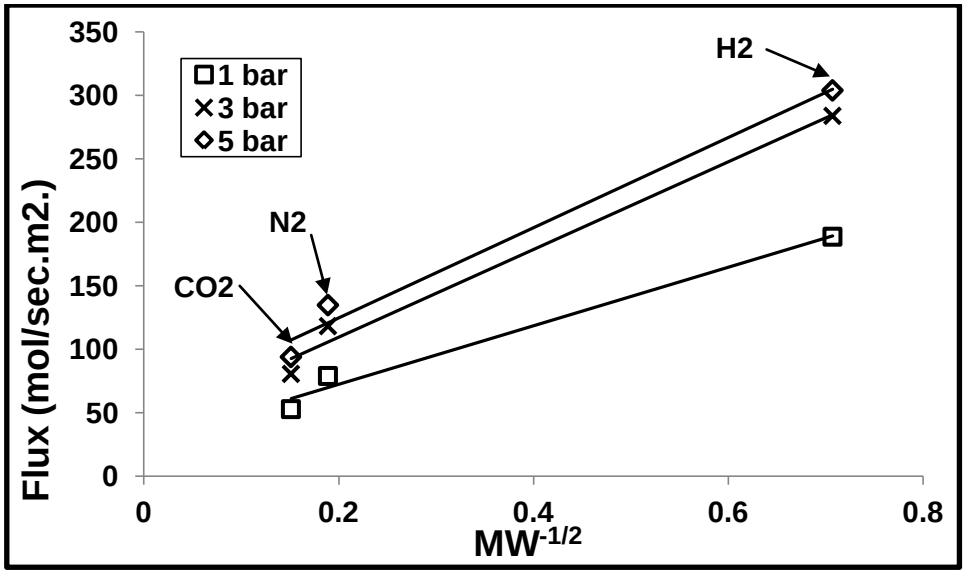

Fig 6: Single gas flux values for the modified alumina support at 1, 3 and 5bar pressure in relation to inverse molecular weight at 293k

Figure 7 and Figure 8 shows results obtained to investigate the membrane molecular sieving behaviour using the data in Table 1 for the kinetic diameter and permeance for H_2, N_2, and CO_2 at 0.5 and 5 bars. As shown, the gas permeance is not in an exact order of their kinetic diameter. H_2 with the lowest kinetic diameter (2.89Å) had the highest permeance compared to other gases but, N_2 gas with the highest kinetic diameter (3.64Å) permeated faster than CO_2 (3.3 Å). The flow mechanism present can be suggested to be non-molecular sieving diffusion regime although H_2 gas behaviour can be said to exhibit molecular sieving characteristics. In addition, no clear difference was observed even when the pressure was increased to 5 bar except for an increase in the gases permeance.

TABLE 1
Single Gas Permeance as a function of Kinetic Diameter

		Pressure (0.5bar)	**Pressure (5bar)**
Gas	**kinetic diameter**	**Permeance**	**Permeance**
H_2	2.89	0.001946	0.000608
N_2	3.64	0.001196	0.000269
CO_2	3.3	0.001024	0.000188

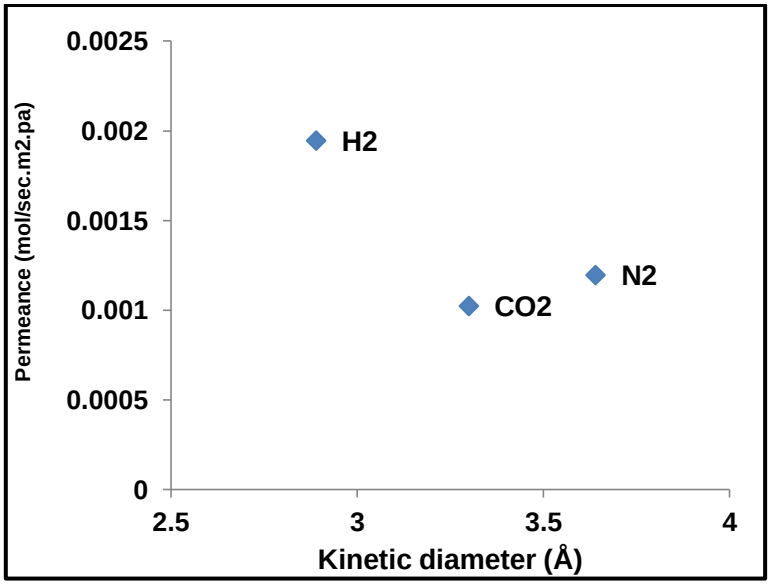

Fig. 7: Molecular sieving behaviour of the ceramic membrane as a function of kinetic diameter at 298k and 0.5 bar

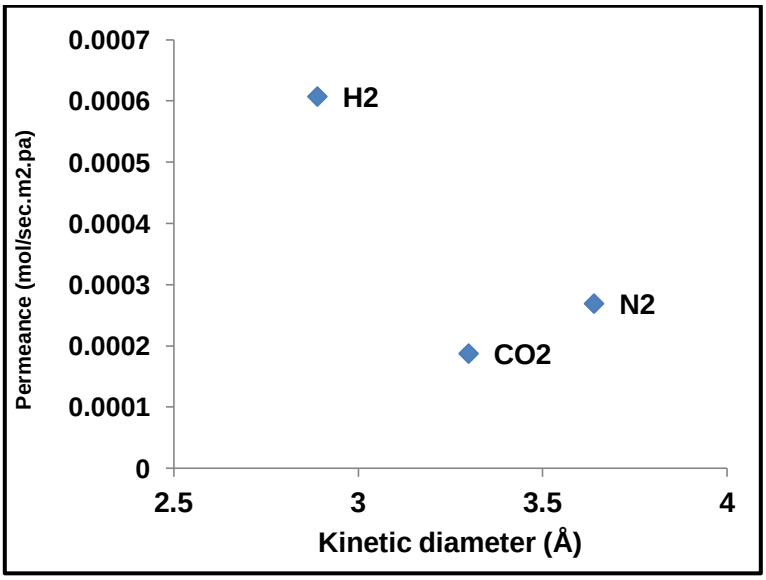

Fig. 8: Molecular sieving behaviour of the ceramic membrane as a function of kinetic diameter at 298k and 5 bar

V. CONCLUSION

A commercial alumina ceramic membrane was fabricated through a dip-coating technique in a silica solution. The silica membrane formed exhibited excellent potentials in hydrogen separation and purification from other gases. In the first subject of investigation in this work, the effect of pressure drop on H_2, N_2 and CO_2 flow rates are presented. It was observed that the gas flow rate increased with increase in pressure with H_2 exhibiting the highest flow rate. For the gas permeance as a function of inverse square root of the molecular weight, the single gas permeation confirmed the governance of Knudsen diffusion with no role played by viscous flow. This high H_2 permeance achieved in the study is attributed to the excellent permeability characteristics of the silica modified membrane and can therefore be recommended in substantial and effective H_2 recovery for various industrial applications.

SYMBOLS

F_i Gas flow rate for component *i* in moles per second

P_i Gas permeance for component *i* (mol m^{-2} s^{-1} Pa^{-1}),

A_m Membrane surface area (m^2)

ΔP_i Pressure difference in Pascal

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CLARIBELLE NGOZI NWOGU is a PhD researcher at the Robert Gordon University, Aberdeen, United Kingdom, having previously obtained Bachelor and Master of Engineering degrees in Petroleum Engineering. She is currently working on 'Advanced membrane design for improved Carbon dioxide capture. By occupation, she is a lecturer and has previously published and co-authored a number of academic/professional papers in international journals. Her research interests are in the areas of design of inorganic hybrid ceramic membrane and multi-channel membrane reactors for Carbon dioxide capture from flue gases. This good team player is a member of various professional associations. She has also made several conference presentations in the United State of America, United Kingdom and Canada.

