The Quantitative Effect of Flow Direction on Gas Permeation in Ceramic Membrane

Muktar M. Ramalan*, Radhakrishna Prabhu*, Idris Hashm*, Priscilla Ogunlude*, Evans Ogoun*, Florence Aisueni*, Ofasa Abunomah**, Ayo Giwa***, and Professor Edward Gobina**

Centre for Process and Membrane Technology, Robert Gordon University, Aberdeen, United Kingdom, e.gobina@rgu.ac.uk
**School of Engineering & the Built Environment, Edinburgh Napier University, United Kingdom, F.MuhammadSukki@napier.ac.uk
***McAlpha Inc, Calgary, Canada, info@mcalpha.com

ABSTRACT

The effect of flow direction in radial porous ceramic membrane has been extensively studied. Understanding fluid flow through membrane has offered utility to a number of industrial processes such as gas separation, catalytic reactions, enhanced oil recovery, and water purification. In these processes the direction of fluid flow affects the magnitude of certain quantities such as of permeation, injection pressure, pressure gradient and mobility. The extent of this effect has been studied for some gases on an individual basis, however, in this research, common industrial gases (i.e. Air, and Carbon dioxide) have been simultaneously investigated thereby offering an opportunity for a comparative study of the effect of flow direction on the respective gases

Keywords: ceramic membrane, flowrate, gas, permeation

1 INTRODUCTION

Membrane have been used in the industry to successfully overcome the challenges of energy efficiency and energy conservation in chemical reactions, space reduction challenges through equipment size and threats posed to the environment because of waste production. Organic synthesis processes and modern chemistry utilise catalysts for their high reaction regioselectivity and stereospecificity some examples of industries that utilize the catalytic membrane reactors are biotechnology sector, pharmaceutical fields, petrochemical, chemical plants, energy and environmental fields (Algieri, al et., 2021). The impact that membrane reactors have on chemistry is that chemical reactions and membrane separation become a single process thereby improving conversion and selectivity as well as space conservation and good cost benefits. The membrane reactors are either polymeric or inorganic as reflected in the operating condition (Westermann, T. and Melin, T., 2009.). The thermal conditions for operating inorganic membranes are higher than that for polymeric ones at up to 1000 degrees Celsius. (Kayvani et al., 2018). The selectivity and permeability observed with inorganic membranes also gives it an edge over the polymeric ones (Robeson, L.M., 2008). The only demerits of the inorganic membranes would be the cost of production and the brittleness (Umukoro, G.E. and Ismail, O.S., 2017). The membranes sometimes come with support to add to their physical strength. These membranes can be made with the following substances. Carbon, silica, alumina, titania, zirconia, palladium, zeolite, and alloy among other substances (Van Veen 1996). Depending on the configuration, the reactor can be Inert Membrane Reactor or Catalytic Membrane Reactor (Fogler, H. Scott. 2005).

2 MATERIAL AND METHOD

Mechanisms for the transport of liquid and gas molecules through porous and dense membrane are shown in the Figure 1. Unlike the solution-diffusion model in which the target gas form a solution with the membrane enabling it to pass though (see Figure 1 (c)) there is no equivalent unified theory (like the Fick's Law) to describe transport in microporous membranes. Microporous membranes have different pore structures and their mechanism of separation differ significantly (see Figure 1(a) and (b)). The membrane used in this study is in tubular shape consist of alumia support. Gas permeabilty and stability examined under defferents operation condition of pressure at steady temperature to measure the behaviours of the variouse gases and to understand the mechanisms involved in their permeation. Figure 2 shows a Schematicdiagram of the experimental set up for gas movment from shellside of the membrane into the membrane bore.
Figure 2 shows the membrane is place in the center of the annulus stainless shell holder and sealed at both ends with graphite ‘’O’’ ring seals at both ends to avoid leakage. Before preceding to the experiemntal, a leak test was conducted by releasing one of the gas into the system and a snoop liquid leak detector was used to detect any leakage. The system can be heated and regulated accorded to desired temperature as shown in figure 3 respectively.

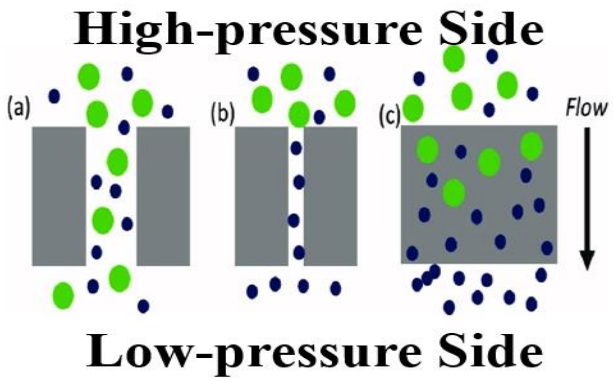

Figure 1: Flow in porous and dense membrane systems –(**a**) Knudsen diffusion through pores, (**b**) molecular sieving and (**c**) Solution diffusion through dense membrane

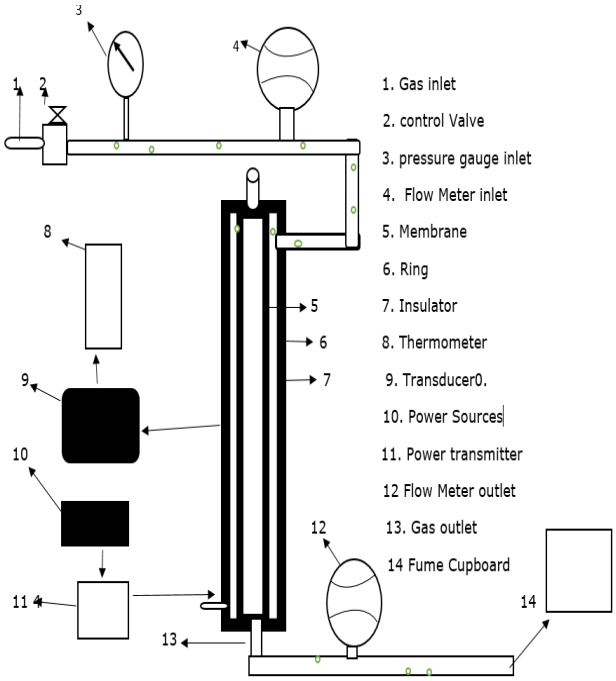

Figure 2 Schematicall diagram showing the experimental set up for gas movment on annulus shell of the membrane

3. EXPERIMENTAL PROCEDURE

An experimental approach was adopted for the study and series of gas experiments were conducted for Air and Carbon Dioxides in a radial inorganic ceramic membrane. The membranes are made of multiple layers of pore sizes (15, 200 and 6000nm), such that the most constricted pore was coated on the outer layer, while the inner layers are made of larger pore sizes and serve as (support).

The gases were injected into the shell-side of the membrane in a normal orientation and allowed the gases to permeate through the support membrane at a different pressure. The steady state permeation was recorded for operating conditions of 0.2, 0.6, 1.0, 1.4, 1.8, 2.2, 2.6, and 3.00 bar at 100 degrees C, respectively.

The effect of transmembrane pressure drops on flux and permeance was determined using the data obtained from the experiment. The inlet pressure used was in atmospheric Pressure (Atm.) for the operation condition, the inlet and outlet pressure from the experiment were recorded in Bar and then converted to Pascal (1bar is equivalent to 100.000pa). The pressure drop was calculated by divided the outlet and inlet pressure in Pascal, the average pressure is the sum of inlet and outlet pressure divided by 2 in Pascal, and the inverse average pressure is the reciprocal of average pressure. The data obtained for flowrate from the experiment was in litre per minute LPM then are converted to meter cubed per second (M^3/S). The membrane surface area was obtained through computing using the equation ($2\pi rh + 2\pi hr^2$). The Flux was calculated by divided flowrate M^3/S with the surface area (M^2), and likewise permeance was calculated by divided flux M^3/M^2S with pressure drop Pa respectively.

4. RESULTS AND DUSCUSSION

In this work, permeation experiments for air & carbon dioxide flows through nanoporous membranes of different average pore sizes were conducted and the effect of the pore size and pressure differential on the permeation behaviour of the gases were analysed in Darcy's law.

The complex structure of ceramic membrane has been the motivating factor behind numerous researches on the transport mechanisms involved in Gas separation

Darcy's law explained the proportional relationship between instantaneous discharge rate through a porous medium, the viscosity of the fluid and pressure drop at a stained distance. The flow of gas through a porous membrane is subject to transport mechanism and component material balance, The Darcy law has been used in so many ranges of application e.g., flow of oil, water and gas in a porous media, In Darcy's law the discharge rate is present as q and the proportionality constant κ, is called the absolute permeability, and is used to characterize the porous medium. Therefore, the Darcy formula for linear displacement is given by equation 1.

$$q/A = Q_0 = -\kappa\Delta P/\mu\delta \quad (1)$$

Where:

q =fluid volumetric flowrate (m^3s^{-1})

A = cross-sectional area of the porous medium perpendicular to the flow (m^2)

Q_i = fluid Volume flux ($m^3\ m^{-2}s^{-1}$)

κ = absolute permeability (m^2)

ΔP = pressure difference (Pa) across the distance L (m) parallel to the direction of flow

μ = the fluid viscosity (Pa-s)

δ = finite distance (m)

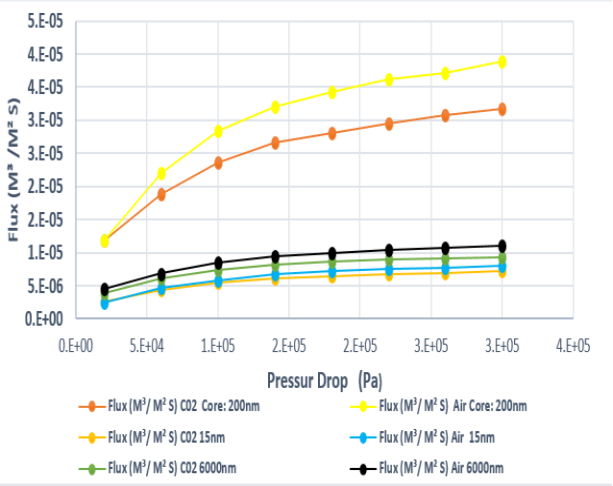

Figure 3 Display a typical graph plot showing the effect of pressure on flux for Air and CO2 gases on a membrane pore size 200, 15 and 6000nm at **100^0** C

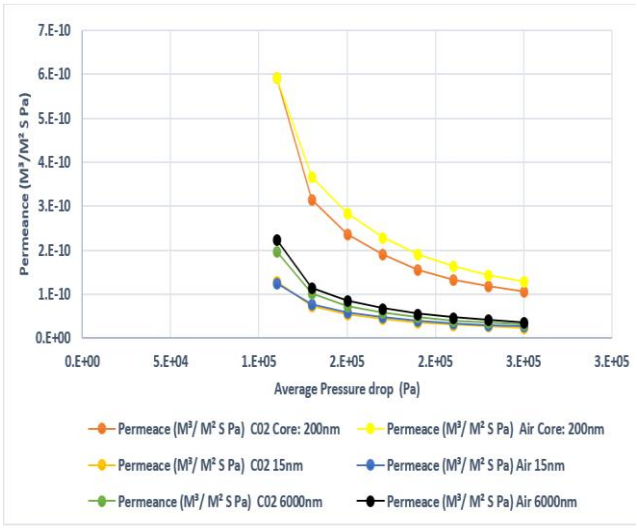

Figure 4 Display a typical graph plot showing the effect of Average Pressure (Pa) on Permeance (M^3/M^2 S Pa) for Air and C02 gases on a membrane pore size 200, 15 and 6000nm at 100^0 C

The resistance encounytered by a gaseous species as it is tries to go through the pore space of a porous membrane is depended on its molecular properties, its interaction with the material tconstituting the walls of the pores, and the pore structure. Gaseous transport in pores can occure through various mechanisms, each contributing to the overall transport rate of a particular specie.

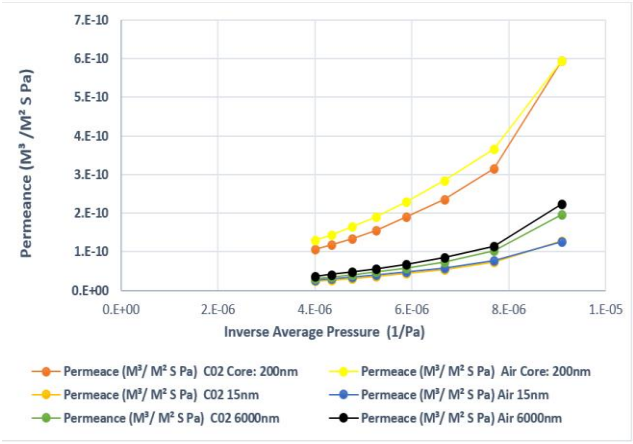

Figure 5 Display a typical graph plot showing the effect of Inverse Pressure on Permeance (M^3/M^2 S Pa) forAir and CO2 gases on a membrane pore size 200, 15 and 6000nm at 100^0 C

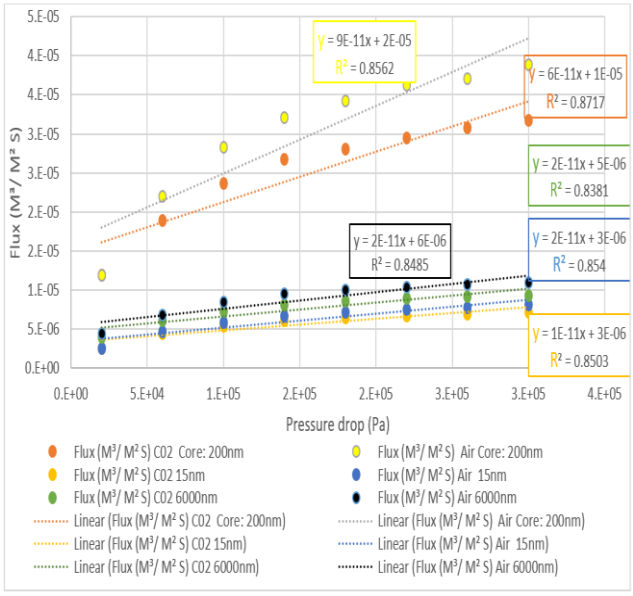

Figure 6. Display a typical graph plot showing the effect of Flux on pressure (Correlation R^2) for Air and C02 gases on a membrane pore size 200, 15 and 6000nm at 100^0 C

The results indicate that the direction of fluid flow affects the permeation magnitude of the respective gases. The degree of directional flow disparity is highest for Air gas and lower for CO2 in all the membranes (figure 3) the permeation flow rate is the function of molecular weight and atomic radius of the gas molecules therefore the direction and magnitude, the force exerted upon unit mass of the fluid by gravity and by the gradient of the fluid pressure differed for each gas. Pressure gradient is main driving force for this process.

The results obtained satisfied the conditions required to divide the flow stage into Darcy and Non-Darcy flow. Below the steady state permeation, the result obeyed Darcy Equation as separate permeabilities of the two gases (Air, & CO2) increases with pressure. As a result of this, the gas tends to move faster thereby increasing its volumetric flux (i.e., the rate at which it flows per unit area in the system regardless of the temperature difference). Above the steady state permeation, Darcy Equation is not obeyed and in the graph, the region is termed Non-Darcy State and it is characterized with a plateau. This implies that the direction and rate of flow of gas across ceramic membrane is established by pressure gradient and are independent of temperature gradient as refelected in figure 3.

Figure 4 shows a plotted graph average pressure against permeance, which shows the magnitude of the degree at which membrane permit gas to permeate, as seen in the graph the permeance of individual gases decreased with the increase in average pressure in the membrane, as the average pressure increase, the radius of the membrane tends to increase as will, at the result of that the selective layer area increase and thickness decrease (Laguntsov N I et al. 2017) therefore the deformation of the porous support has influences the permeability to reduced, since its thickness area and the porosity differs over entire thickness of the support, which lead to decrease in the permeability of the porous support as shown in figure 4.

In figure 5, the effect of inverse average pressure on permeance for different membrane at the same temperature was observed, the inverse avreage pressure increase also the permeate increaese with time. The Air permeability is larger than CO2 permeability by several times to one order of magnitude for in all the membranes especially in 200nm core as reflected in the graph figure 5, therefore the gas permeability increases with increasing pore pressure, the gas permeability slightly increases with increasing pore-pressure gradient across the membrane. The inverse relationship as was observed between the permeance and molecular weight of the individual gases confirm the Darcy's flow mechanism.

In figure 6 the correlation (R^2) between molecular weight and degree of directional disparity was found to increase as injection pressure increases, the correlation R2 (i.e. is the transport flux of material through the membrane per unit driving force per unit membrane thickness) as seen in figure 6, the permeance across the radial ceramic membranes for Air and CO2 increases linearly with absolute differential pressure (square coefficients R^2 ranging between 0.8485 and 0,8717 for Air and CO2, for membrane 200, 25, and 6000nm shows that the membrane were found to have significant positive correlation to both.

CONCLUSIONS AND FUTURE WORK

The good performance and stability of the novel radial inorganic membranes make these materials promising candidates in applications such as gas reforming.

The information from this study has significant implication to industrial processes. It has been demonstrated that better permeation is achieved where flow direction is from constricted to less constricted layers. This also apply to applications where flowrate, flux, and permeability are to be optimised. The knowledge of the molecular weight as well as the atomic size of the molecule to be separated is essential in the selection of the membrane pore size

The permeance and permeabilty decrease with increase in pressure and a better permeance for the gases occure at pressure higher than 1.0 bar. The future experiemnt on defferent temperature range from 200, 300, 500, for permeability using the same techniques to confirm Darcy's and Klinkenberg is on going.

5. ACKNOWLEDGEMENTS

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