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Testing of Gas Permeance Techniques of a Fabricated CO2 Permeable Ceramic Membrane for Gas Separation Purposes

N. C Nwogu[1]
Centre for Process Integration and Membrane Technology
School of Engineering
The Robert Gordon University, Riverside East Garthdee Road
Aberdeen AB10 7GJ – United Kingdom

M.N Kajama[2]
Centre for Process Integration and Membrane Technology
School of Engineering
The Robert Gordon University, Riverside East Garthdee Road Aberdeen AB10 7GJ – United Kingdom

E.P. Okon[3]
Centre for Process Integration and Membrane Technology
School of Engineering
The Robert Gordon University, Riverside East Garthdee Road
Aberdeen AB10 7GJ – United Kingdom

H. Shehu[4]
Centre for Process Integration and Membrane Technology
School of Engineering
The Robert Gordon University, Riverside East Garthdee Road Aberdeen AB10 7GJ – United Kingdom

E. Gobina[5]
Centre for Process Integration and Membrane Technology
School of Engineering
The Robert Gordon University, Riverside East Garthdee Road
Aberdeen AB10 7GJ – United Kingdom

Abstract: ***A CO_2 permselective ceramic membrane has been developed for CO2 separation at room temperature and feed pressure between 1-5bar for CO_2 capture applications in flue gas and natural gas streams. Gas permeation tests were carried out to determine the selectivity and permeability potentials of the silica modified membrane. Results obtained show that the membrane has high CO_2 permeance of 3.43X 10-6 mol/sec.m2.pa in comparison to N_2 and CH_4 gases at 5bar. At a low pressure of Ibar, gas permeance, molecular weight and gas kinetic diameter as determinant factors through the modified membrane were in the order of $CH_4 > N_2 > CO_2$ confirming the dominance of two flow mechanisms, Knudsen diffusion and molecular sieving. However with further increment in the pressure to 5bar, a change in the flow mechanism occurred. From the results also a remarkable regression fit of R2 = 0.9983, 0.9902 and 0.999 in gas flux was obtained for N_2, CO_2 and CH_4 respectively.***

Keywords: Carbon dioxide, ceramic membrane, gas separation, selectivity & permeability.

I. INTRODUCTION

The atmospheric concentration of CO2 is a key contributor linked to the greenhouse effect. The majority of the CO2 is as a result of fossil fuel consumption and its management is a big challenge and concern to mankind. Curtailing CO2 emissions has become an issue of global deliberations and top discussions on climate change mitigation. One solution to prune down CO2 atmospheric concentration is to utilize conventional methods of gas separations from flue gas and natural gas point sources. Due to its manageable modular size, eco-friendliness, energy saving features and long life span in operating conditions, membrane technology has demonstrated excellently well for application in gas separation processes especially for efficient removal and purification of CO2 [1] [2] [3]. Organic and inorganic membranes are two major types of membranes for gas separation applications. Polymer dense membranes are made of organic materials and first utilized industrially in the separation of CO2 and H2S. The outcome of the separation process did not yield the expected result due to some limitations exhibited by the membrane [4]. This however was attributed to the membrane poor resistance to high operating pressure and temperature. Porous ceramic membrane however which forms the main class of inorganic membranes have gained much attention due to its inbuilt

features of high stability and deliverability under unfavourable working and operational environment especially high resistance when in contact with acidic chemicals. Conversely, for a commercially viable inorganic membrane, two parameters must be achievable. The permeability and selectivity characteristics of the membrane towards the permeating gas mixtures are of interest. Three basic make up of a multilayer ceramic membrane, comprises of a fresh support of large pore diameter with high tensile strength, a middle layer whose pore size diameter is smaller than that of the support and a topmost thin layer. One importance of the deposition of mesoporous layer is to cover up for the very large pores present in the macro porous layer for pressure control along the gas flow line during operations. Micro porous layer actually determines, reduces and controls the pore size of the membrane needed in yielding the desired result. [5] [6] [7]. Moreover, dip coating as a suitable method of membrane formulation has been reported by a number of researchers involving the immersion of a fresh support for surface repairs of pinhole defects [8] [9] [10] [11]. For the purpose of this study, modification process through dip coating in a silica based solution is utilized producing a composite silica membrane [12]. Notably, flow behaviour of gas molecules across porous ceramic membranes is important in membrane technology. This flow is controlled by three factors: contact between the gas molecule and the membrane material used for membrane design, the membrane structure and the gas properties. Consequently a composite material made from ceramics membrane has been tailored through nanotechnology, an economical and very efficient technology. The nano structured inorganic membrane selectively separates the gas of interest from other gases in the stream and allows it to flow through the coated thin layer of the membrane for eventual safe recovery [13]. Knudsen diffusion and viscous diffusion are two main permeation mechanisms to determine the transport behaviour of gases across a porous membrane [14]. However, for porous membranes of smaller pores of about 1nm, molecular sieving type of mechanism takes place. Viscous flow is characterized by the gas molecular collusion whereby the membrane pores are larger than the mean free path. For collusion between gas molecules and the membrane pore walls, Knudsen separation mechanism is dominant. In addition, the flow is molecular weight dependent [15] [14].

II. MEMBRANE FOR EFFICIENT AND SELECTIVE GAS SEPARATION

Gas separation through a membrane works on the theory of permeation based on permeability and selectivity. For gas separation processes, a gas stream at an elevated pressure is allowed to pass through a membrane separator. Interestingly as reported by so many researchers, the gas of interest is then captured at the permeate side of the membrane at a reduced pressure as shown in Figure 1. In addition the level of separation is also dependent on the solubility and diffusivity capacity of the gas molecules through the membrane as well as the driving force which is a function of the pressure drop through the entire system [16] [17] [18]. Fig 1 is a simplified diagram of gas separation theory. As observed the desired gas molecule is being adsorbed on the surface of the membrane from the region of high pressure through that of lower pressure area.

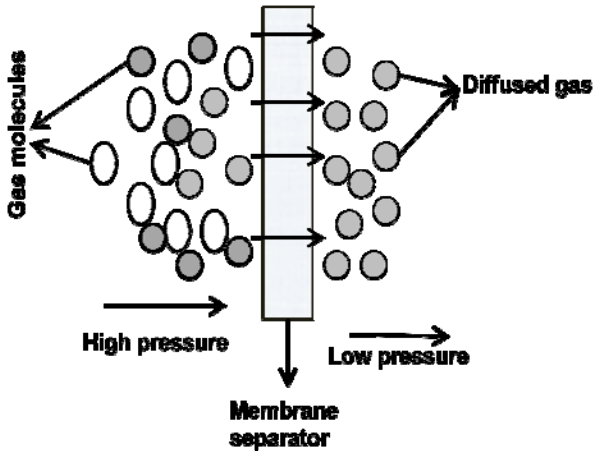

Fig 1: Schematics of gas separation theory in a membrane separator [18] [19].

III. EXPERIMENTAL

In this work, a porous ceramic α-alumina support of 6000nm average pore size was modified and used to carry out the permeation experiment. The support was supplied by Ceramiques Techniques et Industrielle (CTI SA) France and has a specification of 25mm outer diameter, 19mm inner diameter and an effective total length of 318mm. The porous alumina support was modified before the experiment through a dip-coating process in a silica-based solution. The outcome of a five times successive dip-coating resulted in the formation of a thin film separating layer on the outside of the ceramic support. The modification process was done to reduce the membrane pore size to enhance the selectivity. Figure 2 is the inorganic ceramic support while Figure 3 depicts support picture after modification.

Fig 2: Tubular inorganic ceramic support

Fig 3: Modified Support

The hybrid composite membrane has been characterized by scanning electron microscopy (SEM). Figure 4 and Figure 5 shows the SEM images of the composite membrane cross section and the surface view respectively. The structural make-up of the cross section with the pores are more exposed and obvious while the surface view is narrowly interweaved and improved at the surface. Membranes with smaller particle sizes results in tremendous gas selectivity as a consequence of restrictions related to small particles [9].

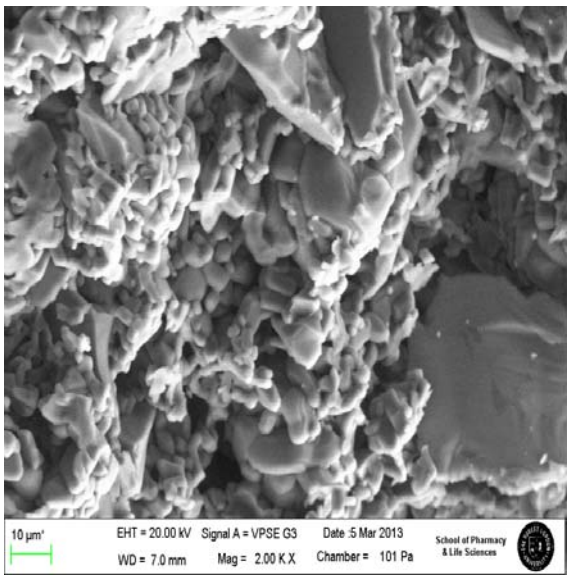

Fig 4: SEM image of composite membrane cross sectional view

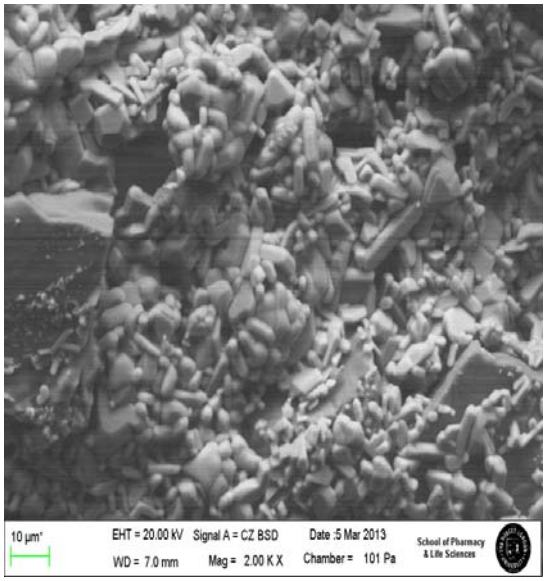

Fig 5: SEM image of composite membrane surface view

A schematic of the gas permeation experimental set up is shown in Figure 6. The system consists of gas cylinders which deliver the pure gases, the membrane and membrane reactor with gas inlet, retentate outlet and permeate outlet. Graphite seals are used at both ends to prevent gas leakage and pressure tight system. Additionally, the gas flow rates are measured by using a digital flow meter while maintaining a pressure difference across the membrane. The permeation test was carried out using carbon dioxide, Nitrogen and Methane single gases with a purity of at least 99.9%. The gases were supplied by BOC, United Kingdom. The feed pressure range used to determine the gas volumetric flow was between 1 and 5 bar at room temperature [20].

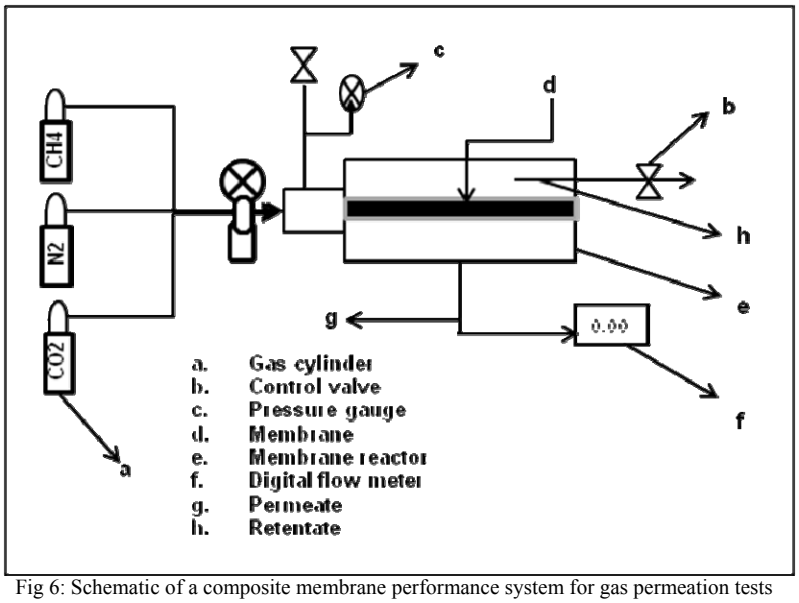

Fig 6: Schematic of a composite membrane performance system for gas permeation tests

IV. Results and Discussions

The permeance of three single gases CO_2, N_2 and CH_4 were found to increase linearly with pressure drop. A plot og gas permeance as a function of the differential pressure with respect to CO_2,N_2 and CH_4 gases is shown in Figure 7. As can be observed, the behaviour in terms of the flow mechanism is a non-Knudsen diffusion mechanism. It can be seen also that CO_2 gas permeated faster than CH_4 and N_2 which ordinarily should not be the case due to its lower molecular weight compared to others. This result suggests that the transport mechanism present is active and may have been initiated as an outcome of the membrane modification. Therefore, there is a strong indication of an adsorptive surface flow mechanism between the gas molecules and the surface of the membrane pores. It was also noticed that even at an elevated pressure of 5bar, CO_2 still exhibited the highest permeance.

The flux of the individual gases as a function of the feed pressure was investigated. The graphs are shown in Figures 8, 9 and 10. Of all the gases CO_2 gas recorded the highest flux of 1.717mol/sec.m^2 at a relatively low pressure. Most importantly, the results obtained for all the gases shows a remarkable regression fit of R^2 = 0.9983, 0.9902 and 0.999 for N_2, CO_2 and CH_4 respectively.

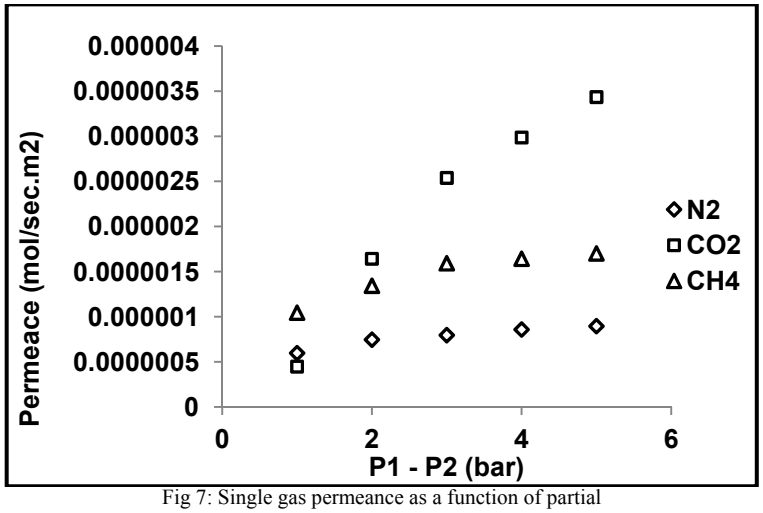

Fig 7: Single gas permeance as a function of partial differential pressure at 293k

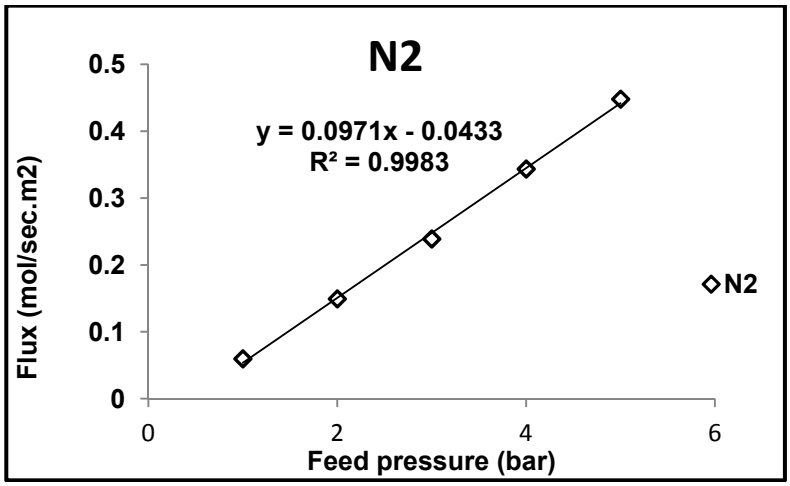

Fig 8: Nitrogen flux versus feed pressure at 293k

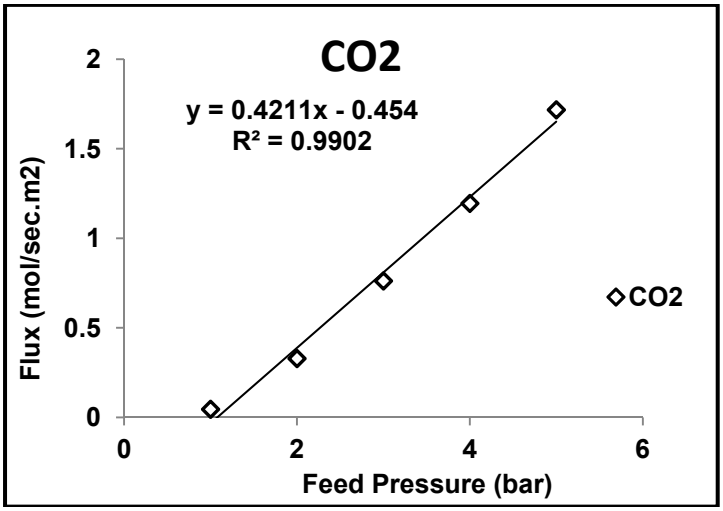

Fig 9: Carbon dioxide flux versus feed pressure at 293k

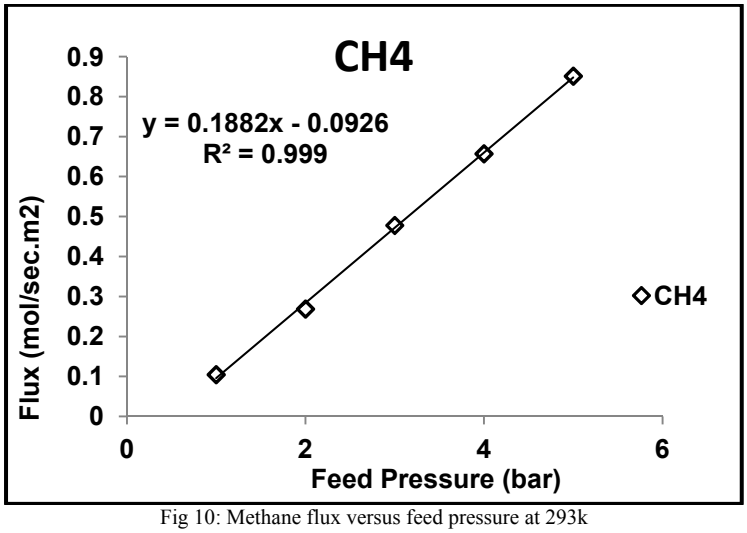

Fig 10: Methane flux versus feed pressure at 293k

The relationship of the inverse square roots of the gas molecules and their kinetic diameter at 1and 3 bar as a function of gas permeance were calculated and values plotted in Figures 11, 12, 13 and 14. As can be observed, Figure 11 and Figure 12 have very similar trend. For Figure 11 and Figure 12, a linear proportionality is seen in their relationship of inverse square root of gas molecular weight as a function of gas permeance at a pressure of 1bar. Generally, gas permeance rate is in the order, CH4 > N_2 > CO_2 with the order of their molecular weight as CO_2 > N_2 > CH_4 (44, 28 and 16) respectively. Similarly for Figure 13, an order of increasing kinetic diameter follows as CH_4 > N_2 > CO_2 (3.8, 3.64 and 3.3). An analysis of these results confirms the

dominance of two flow mechanisms. The dominance of Knudsen diffusion and molecular sieving for the investigations carried out on a modified silica membrane.

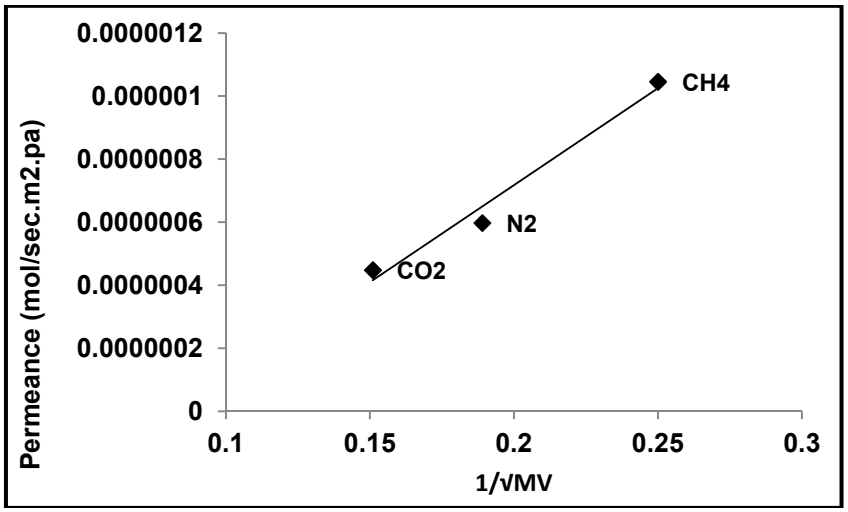

Fig 11: Relationship of the inverse square root of molecular weight on gas permeance at pressure of 1bar and 293k

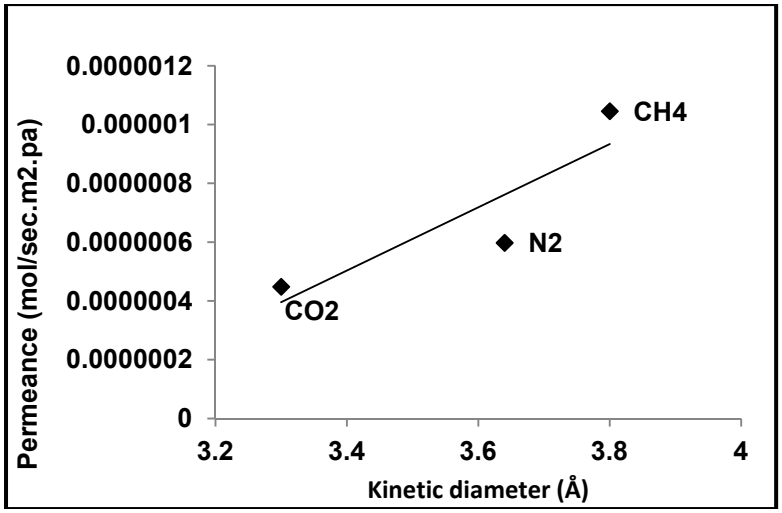

Fig 12: Relationship of gas kinetic diameter on gas permeance at pressure of 1bar and 293k

In figure 13 and Figure 14, this is same relationship under same temperature but at a higher pressure of 3bar. The result suggests non Knudsen diffusion as pressure is increased rather a reasonable interaction between the gas molecules and the surface of the membrane may be occuring. Moreover, the relationship existing between the gas permeance and their kinetic diameter for the membrane may have possible molecular sieving characteristics with respect to an increase in kinetic diameter with pressure, the gas of higher kinetic diameter (CH_4) permeating faster than other.

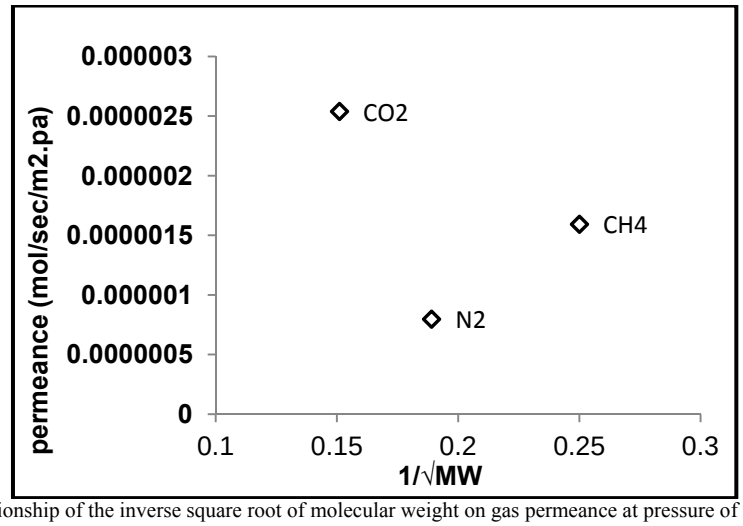

Fig 13: Relationship of the inverse square root of molecular weight on gas permeance at pressure of 3bar and 293k

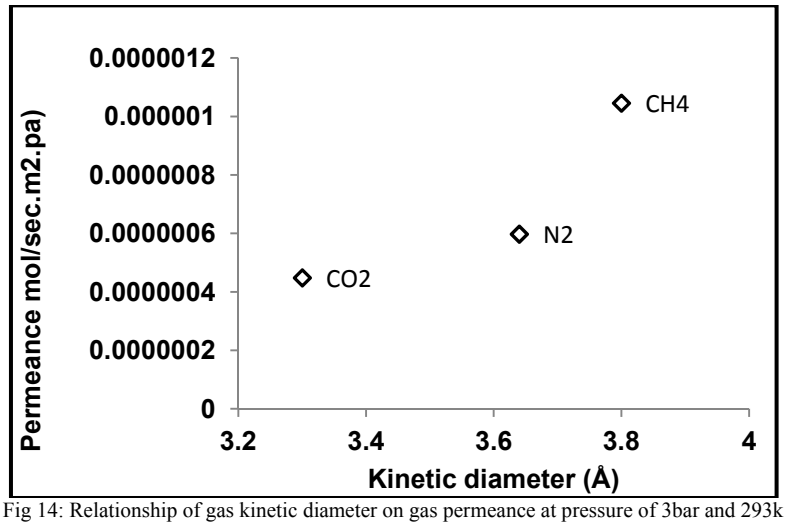

Fig 14: Relationship of gas kinetic diameter on gas permeance at pressure of 3bar and 293k

V. CONCLUSION

An experimental study has been carried out to test for gas permeation through a fabricated CO_2 silica modified membrane for improved gas separations. Results obtained show a non-linear variation of gas permeance with pressure drop with a reasonable flux of 1.711mol/ m^2sec. for CO_2 compared to N_2 and CH_4 gases. The influence of increase in pressure from 1bar to 3bar was observed and reflected in the relationship between the inverse of the square root of the molecular weight as well as the kinetic diameter with a drift from Knudsen flow mechanism to a more adsorptive surface flow mechanism. This is an indication of the membrane having improved selectivity of CO_2 from other gases in addition to its enhanced permeability features exclusively for CO_2.

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AUTHOR(S) PROFILE

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.

