Morphological Characterization and Gas Permeation of Commercially Available Ceramic Membrane

Ifeyinwa Orakwe, Ngozi C. Nwogu, Edward Gobina*

***Abstract*— This work presents experimental results relating to the structural characterization of a commercially available alumina membrane. A γ-alumina mesoporous tubular membrane has been used. Nitrogen adsorption-desorption, scanning electron microscopy and gas permeability test has been carried out on the alumina membrane to characterize its structural features. Scanning electron microscopy (SEM) was used to determine the pore size distribution of the membrane. Pore size, specific surface area and pore size distribution were also determined with the use of the Nitrogen adsorption-desorption instrument. Gas permeation tests were carried out on the membrane using a variety of single and mixed gases. The permeabilities at different pressure between 0.05-1 bar and temperature range of 25-200^oC were used for the single and mixed gases: nitrogen (N_2), helium (He), oxygen (O_2), carbon dioxide (CO_2), 14%CO_2/N_2, 60%CO_2/N_2, 30%CO_2/CH_4 and 21%O_2/N_2. Plots of flow rate verses pressure were obtained. Results got showed the effect of temperature on the permeation rate of the various gases. At 0.5 bar for example, the flow rate for N_2 was relatively constant before decreasing with an increase in temperature, while for O_2, it continuously decreased with an increase in temperature. In the case of 30%CO_2/CH_4 and 14%CO_2/N_2, the flow rate showed an increase then a decrease with increase in temperature. The effect of temperature on the membrane performance of the various gases is presented in this paper.**

***Index Terms*— Alumina membrane, Nitrogen adsorption-desorption, Scanning electron microscopy, Gas permeation, temperature**

I. INTRODUCTION

THE knowledge of membrane technology could be traced to over six decades ago with the initial aim of employing membranes for the separation of certain gases [1],[2]. The use of ceramic membrane is very significant in environmental gas separation. The term "ceramic membrane" could be seen as a semi permeable barrier which could be porous or dense in structure [3]. These membranes are usually asymmetric in structure, consisting of a few layers of one or more of different materials with graduated pore sizes. The support could be a macroporous support, mesoporous intermediate layer and the membrane the microporous top layer. Aluminium oxide (Al_2O_3), Titanium oxide (TiO_2), Zirconium oxide (ZrO_2), Silicon oxide (SiO_2 etc., are the most common constituents used for the ceramic membranes preparation [4], [5]. It could as well be a combination of the listed metal oxides [3]. An alumina ceramic membrane was chosen for this experiment because of its chemical stability and also because of its narrow pore size distribution [4]. Alumina ceramic membranes in particular have drawn the attention of researchers as a result of its unique properties for example showing high chemical resistance and mechanical strength and are also known to withstand high temperature and pressure [5].

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II. EXPERIMENTAL

1. Materials

A commercial alumina membrane was chosen for this experiment. It was supplied by Ceramiques Techniques et Industrielles (CTI SA) France, and consists of 77% alumina + 23% TiO_2. The membrane is made up of 7mm inner diameter and 10mm outer diameter respectively with a permeable length of 318mm and 45% porosity. Single and mixed gases: nitrogen (N_2), helium (He), oxygen (O_2), carbon dioxide (CO_2), 14%CO_2/N_2, 60%CO_2/N_2, 30%CO_2/CH_4 and 21%O_2/N_2 were used in the permeation tests.

Fig. 1 Tubular ceramic membrane

2.2 Characterization techniques

2.2.1 Scanning electron microscope

Scanning electron microscope (Zeiss EVO LS10 Variable Pressure) was used to observe the structure of the alumina membrane. The alumina membrane sample was mounted on an aluminium disc called stub using a suitable adhesive. The adhesive used was able to anchor the sample to the stub as well as provide electrical continuity. The sample carousel is then loaded with samples to be examined. After examining the sample, it was then positioned on a metal holder and gold-coated using sputter-coating operated under vacuum. The SEM micrographs of the inner, outer, cross sectional area and edge of the membrane were taken at a number of magnifications.

2.2.2 Nitrogen Adsorption-desorption

Nitrogen Adsorption-desorption (Quantachrome ®ASiQwin™) instrument was used for the structural study of the membrane. A fragment of the $\gamma -$ alumina membrane was crushed into small pieces and put into a cell, and degassed for about 4 hours at 338K. This was done to remove any moisture and impurities trapped in the membrane. The analysis of the degassed sample was then carried out overnight on the nitrogen adsorption desorption instrument at a temperature of 77K. The nitrogen adsorption desorption isotherms in the case of a non-reactive gas is based on the amount of gas adsorbed as a function of the relative pressure (P/P_o), where P is the applied pressure and P_o is the saturated pressure [5].

2.2.3 Gas permeation test

Gas permeation test were carried out using the single and mixed gases: nitrogen (N_2), oxygen (O_2), carbon dioxide (CO_2), 14%CO_2/N_2, 60%CO_2/N_2, 30%CO_2/CH_4 and 21%O_2/N_2 respectively at temperature range of 25°C - 200°C. The experiment was carried out by regulating the flow rate of the gases using a control valve recorded on a pressure gauge in the range of 0.05-1 bar through the membrane reactor. The permeate flowrate was recorded using a flow meter (Cole Parmer model). Fig 2 shows a schematic diagram of the gas permeation setup.

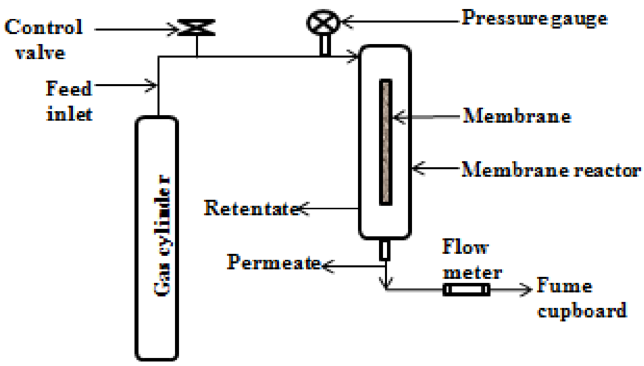

Fig. 2 Schematic diagram of the Gas permeation setup

The permeance of the gas was obtained from the mathematical expression [6]

$Qi=Fi/A\ \Delta Pi$

Where Qi = Permeance ($mol.s^{-1}m^{-2}Pa^{-1}$) of gas i
Fi= Flow rate ($mol.s^{-1}$) of gas i
A= Surface Area of the membrane (m^2)
ΔPi=Pressure difference of gas *i* across the membrane

III RESULTS AND DISCUSSION

3.1 SEM characterization

The SEM photographs of the mesoporous alumina membrane as well as its surface morphology could be seen in Figs 3a and 3b respectively. The SEM photographs show that the ceramic membrane had an asymmetric structure. It was also observed that the membrane does not have a smooth surface and particles were packed in a way to obtain a porous membrane. It was as well noted that the particles were of different shapes, smaller particles were deposited among larger particles and thus the effects of defects are minimised. The result shows the mesoporous membrane used was defect free.

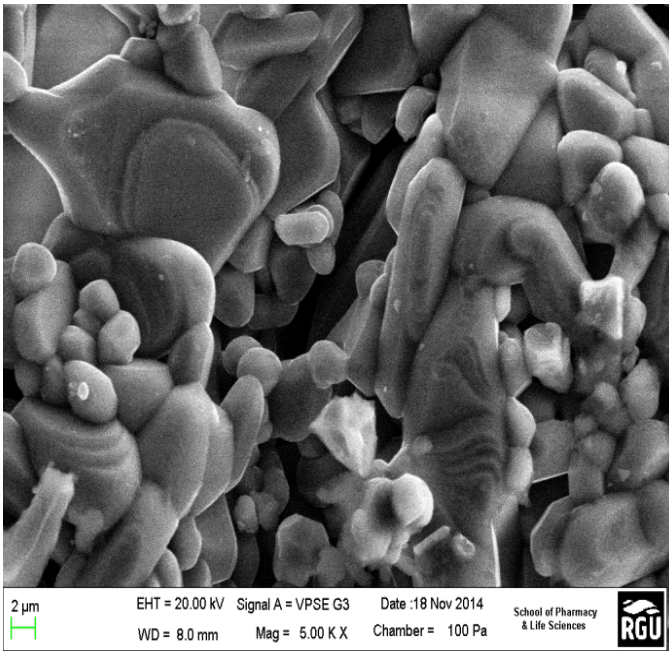

Fig. 3a Outer section of the alumina membrane

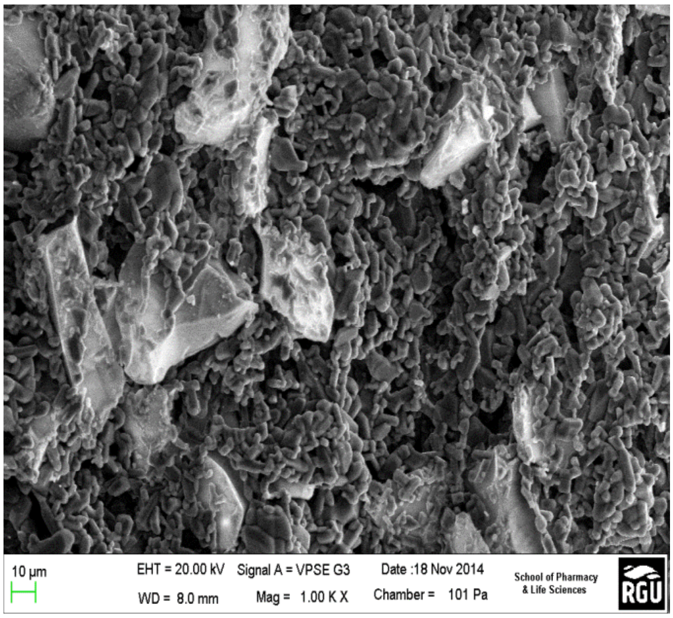

Fig. 3b Inner section of the alumina membrane

3.2 Nitrogen Adsorption Desorption characterization

The nitrogen adsorption desorption instruments generates physiosorption isotherms as well as calculates and provides

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vital information for example: surface areas, pore diameter, pore volume etc. Fig 4a shows a linear isotherm obtained which corresponds to the type IV isotherm explained in [5], and which shows characteristics of a mesoporous membrane undergoing capillary condensation.

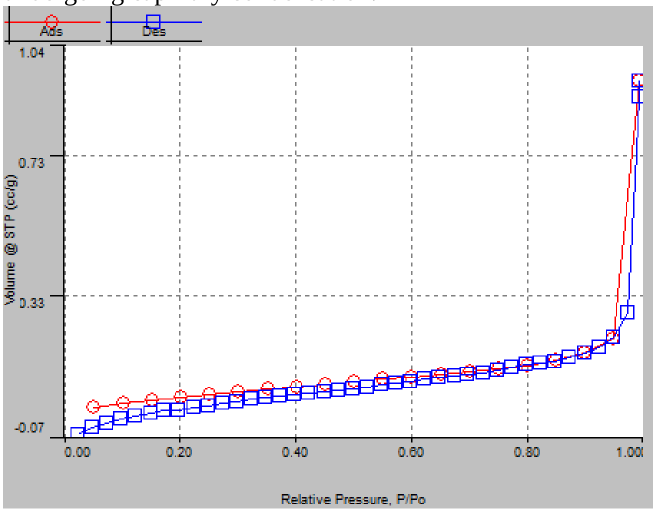

Fig. 4a Physisorption isotherm of the alumina membrane

From the linear isotherm in Fig 4a, a nitrogen desorption isotherm was generated using the Barrett, Joyner and Halenda (BJH) method as shown in Fig. 4b below. The graph calculated the surface area of the alumina membrane to be 0.192m^2/g, pore volume 0.001cc/g and pore diameter 3.305nm.

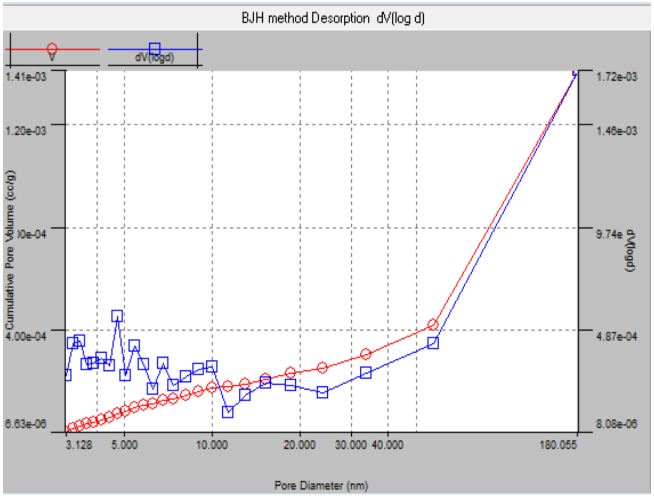

Fig. 4b BJH desorption Isotherm

3.3 Gas permeation analysis

Fig 5a represents a plot of flow rate of single gases at 0.5 bar pressure against various temperatures. It was noted from the graph, that the different gases behaved differently at various temperatures. O_2 showed a monotonic decrease in flow rate as the temperature increased, while nitrogen and carbon dioxide showed a drop in flowrate, then an increase and finally a decrease in flowrate as the temperature was increased.

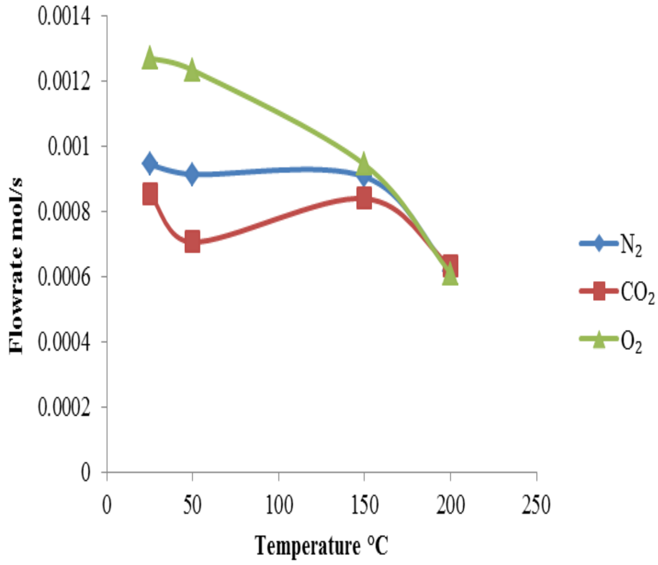

Fig. 5a Graph of flowrates of single gases against temperatures

Fig 5b also represents a plot of flowrate of mixed gases at 0.5 bar pressure against temperature. The slope obtained indicated the presence of initial viscous flow, and then Knudsen flow as the temperature was increased. It was also noted that the different gases behaved differently at various temperatures.

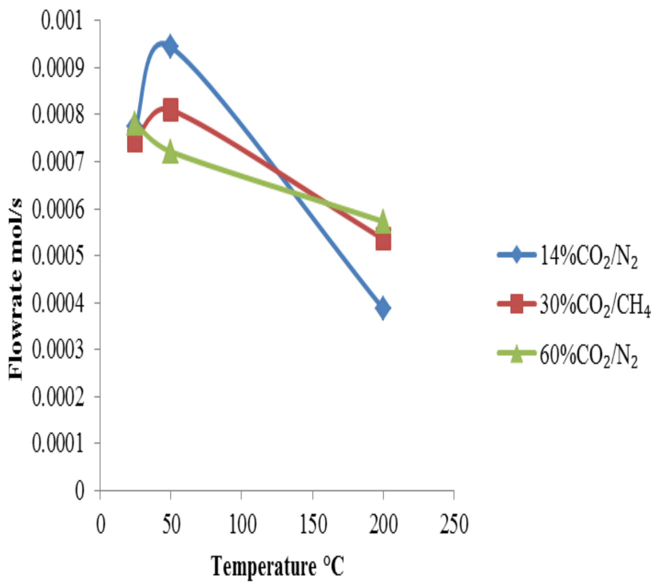

Fig. 5b Graph of flow rates of binary gases against temperatures

IV CONCLUSION

It was observed from the SEM micrographs that the pore looked smaller in the inner surface than at the outer surface. The nitrogen adsorption desorption instrument showed a linear isotherm which corresponded to the characteristics of a mesoporous membrane. It was also concluded from the permeability test carried out that a combination of viscous and Knudsen flow occurred in the mesoporous alumina membrane. This had a significant effect on the transport of gases through the membrane. It was also noted that at higher temperatures, the flowrate of the gases tend to decrease.

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NOMENCLATURE

A Surface area of the membrane (m^2)
F Flowrate ($mol.s^{-1}$)
ΔP_i Pressure difference of gas $_i$ across the membrane (Pa)
Q_i Permeance ($mol.s^{-1}m^{-2}Pa^{-1}$ of gas $_i$)

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