Advanced Membrane Design for Improved Carbon dioxide Capture

N. C. Nwogu* and E. Gobina**

Centre for Process Integration and Membrane Technology, School of Engineering, the Robert Gordon University, Aberdeen AB10 1FR, UK
n.c.nwogu@rgu.ac.uk*, e.gobina@rgu.ac.uk**

ABSTRACT

A nano-structure tubular hybrid inorganic membrane capable of stripping carbon dioxide from flue gas stream was designed and tested at laboratory scale to improve compliance to various environmental regulations to cushion the effect of global warming. Single gas separation experiments using silica modified ceramic membrane was carried out to investigate individual gas permeation behaviors at different pressures and membrane efficiency after a dip coating method. Four gases; Nitrogen (N$_2$), Carbon dioxide (CO$_2$), Oxygen (O$_2$) and Methane (CH$_4$) were used. Plots of flowrate versus pressure were generated. Results show that the gas flow rate increases with pressure drop. However at above a pressure of 4bar, the flow rate of CO$_2$ was much higher than the other gases, indicating dominance of a more selective adsorptive type transport mechanism.

Keywords: ceramic membrane, carbon dioxide capture, permeability, selectivity, gas transport mechanisms

1 INTRODUCTION

About one third of the overall CO$_2$ emission globally comes from energy creation. Its decrease and management is vitally critical and an important factor to ease global warming [1]. The position of CO$_2$ in global warming is a present day ecological concern and need urgent attention in the provision of technologies that will curtail the emission of CO$_2$ [2]. CO$_2$ removal from flue gas stream recorded success with conventional technology like absorption using glycol, amine and methanol under low temperature, process with hot potassium carbonate, reaction with calcium oxide and the use of polymer membrane. These processes employ low temperature resulting in energy losses due to high temperature recuperation as well as cooling of the gas stream [3]. Owing to the burning desire to abate global warming especially at the rate of CO$_2$ emission and its concentration in the atmosphere through flue gases today, inorganic ceramic membrane with distinct characteristics should be a key issue. Cost effective, energy-saving, high chemical resistance, cheap materials are needed in the development of membrane module for manufacturing purposes. Porous inorganic membrane conquers some if not all of the inbuilt limitations. They can tolerate higher temperature and basically limit the connection between selectivity and permeability. If properly designed the pore size and its distribution decide its selectivity while the volume fraction porosity regulates and establishes permeability [3]. Porous inorganic membrane also shows exceptional evidence of physical and chemical properties, including unresponsiveness to sarcastic environment, stability under high temperature, homogeneous pore structure and reasonable fluxes. [4] Some membranes today have been used for decades in CO$_2$ capture, but because the membrane is used for natural gas at very high pressure they are unsuitable for CO$_2$ capture from flue gases. Design and fabrication of porous ceramic membrane consists of several layers of different materials namely: Aluminium oxide (Al$_2$O$_3$), Titanium Oxide (TiO$_2$), Zirconium Oxide (ZrO$_2$), Silicon dioxide (SiO$_2$), Silicon carbide, Zeolite or a mixture of two materials applied on an underlying porous stainless steel, α-alumina, γ-alumina, zirconium, zeolite supports [5]. The manner in which gas molecules flow across the membrane referred to as permeation mechanism is a significant fact in membrane technology. This flow generally is influenced by three factors: the gas properties, morphology of the membrane and the material used for membrane design. Consequently a hybrid material made from ceramics membrane has been well thought out by means of Nanotechnology. The technology is eco-friendly, economical and very efficient. Its applicable in all forms of CO$_2$ elimination from other gases and its effectiveness increases comparatively to the CO$_2$ concentration in the flue gas stream feed. Nanotechnology is among the range of technologies paying attention to explore carbon-capturing technology [6]. Membrane separation of gases is a highly complex process and therefore the material used for its preparation should exhibit a long-lasting characteristic, stability and modified in an advanced manner to be adapted to separate specific gases.

However, the innovation is that a so called agent is immobilized in the membrane porous network, thus assisting to attract the CO$_2$, and enhancing its transport across the membrane. This is in complete contrast to the more common and older method of using a filter that separates directly between CO$_2$ and other gases.
2 EXPERIMENTAL

The gas separation experiment was performed using a membrane support and a nano-structured membrane deposited on a macro porous tubular filter employing a repeat dip-coating technique [6]. The gases used for the gas transport tests included Nitrogen (N$_2$), Carbon dioxide (CO$_2$), Oxygen (O$_2$) and Methane (CH$_4$) respectively. The experiment was carried out to investigate the single gas permeation behaviors of the gases listed above with the permeation done separately and individually.

Figure 1: Tubular ceramic support (Top) and nano-structured composite (bottom).

Figure 2: Permeation cell

Figure 2 shows the permeation cell set up. The components of the cell includes: 1. Gas cylinder, 2. pressure valve, 3. pressure gauge, 4. membrane reactor, 5. retentate, 6. permeate and 7. flow meter respectively.

2.1 Morphology of support

The scanning electron microscopy was used to verify the morphology of the support. The image is shown in figure 3 and 4 respectively.

Figure 3: SEM Membrane cross-section

Figure 4: SEM Membrane Inner surface

3 APPLICABLE THEORY

The manner in which gas molecules flow across composite membrane comprising of the membrane and the support with respect to the pressure will be determined. In addition, a distinction between viscous and knudson flow mechanisms in porous membrane to determine flaws in the
membrane before and after coating. A schematic illustration of the transport process is shown in figure 5.

![Composite membrane diagram](image)

**Figure 5:** Schematic diagram of gas transport through a coated support membrane.

Values of gas permeation rate of the toplayer from figure 5 were calculated using the formula [7]

\[ J_{TL} = \frac{F}{(P_f - P_b) \times S} \]  \hspace{1cm} (1)

Where \( F \) (l/min) is the flowrate through the membrane and \( S \) (m\(^2\)) is the membrane surface area of the flow pathway. \( P_f \) (bar) and \( P_b \) (bar) are absolute feed pressure and borderline pressure between the support and the toplayer. \( P_b \) can be calculated from equation (2) below

\[ P_b = \frac{-C_{\text{diff}} + \sqrt{C_{\text{diff}}^2 + 2D_{\text{diff}}(C_{\text{diff}}P_f + \frac{1}{2}D_{\text{diff}}P_f^2 + Z)}}{D_{\text{diff}}} \]  \hspace{1cm} (2)

where \( C \) and \( D \) are constants relative to membrane support characteristics and \( Z \) given below for gas permeation through the composite membrane.

\[ Z = \left[ C_m + \frac{1}{2}D_m \times (P_f + P_p) \right] \times P_f - P_p \]  \hspace{1cm} (3)

### 4 RESULTS AND DISCUSSION

Accordingly, **Figure 6** presents a plot of flow rate against pressure and the trend explains the proportionality for both parameters.

![Flowrate graph](image)

**Figure 6:** Graph of Flowrate of N\(_2\), CO\(_2\), O\(_2\) & CH\(_4\) versus Pressure

**Figure 7** shows the plots of the ratio of flow rate of CO\(_2\) to that of N\(_2\) in relationship to the ratio of the molecular weight of CO\(_2\) to that of N\(_2\). The trend confirms that at pressure of 2bar and above more CO\(_2\) was obtained from the separation.

![Ratio graph](image)

**Figure 7:** Ratio of flowrates and Molecular Weights of CO\(_2\) and N\(_2\) at different pressures

Figure 8 also demonstrated that at pressure of 3bar and above more of CO\(_2\) was separated from O\(_2\) while **Figure 9** illustrates that from 2.4 bars and above more CO\(_2\) will be removed from CH\(_4\).
Figure 8: Ratio of Flowrates and Molecular Weights of CO₂ and O₂ at different Pressures

Figure 9: Ratio of Flowrates and Molecular Weights of CO₂ and CH₄ at different Pressures.

Figure 10: Membrane defect determination

As expected, the layer showed only minor dependence of permeance with average pressure suggesting the absence of defects. For the support overall results have shown that both viscous and Knudsen flow can have a significant effect on the single gas transport. The dip coating technique had a significant effect toward achieving a zero level membrane deficiency. However, real flue gas will consist of mixtures and work is currently ongoing to obtain mixed gas transport and selectivity.

REFERENCES


5 CONCLUSION

Using the combination of Knudsen flow and viscous flow, it has been possible to extract the transport of the thin membrane layer in a composite hybrid system.

**Corresponding Author – Email: e.gobina@rgu.ac.uk; Tel: +44(0)1224 262348; Fax: +44(0)1224 262444