Performance Evaluation of an Inventive CO_2 Gas Separation Inorganic Ceramic Membrane

Ngozi Nwogu, Mohammed Kajama, Edward Gobina

Abstract—Atmospheric carbon dioxide emissions are considered as the greatest environmental challenge the world is facing today. The tasks to control the emissions include the recovery of CO_2 from flue gas. This concern has been improved due to recent advances in materials process engineering resulting in the development of inorganic gas separation membranes with excellent thermal and mechanical stability required for most gas separations. This paper, therefore, evaluates the performance of a highly selective inorganic membrane for CO_2 recovery applications. Analysis of results obtained is in agreement with experimental literature data. Further results show the prediction performance of the membranes for gas separation and the future direction of research. The materials selection and the membrane preparation techniques are discussed. Method of improving the interface defects in the membrane and its effect on the separation performance has also been reviewed and in addition advances to totally exploit the potential usage of this innovative membrane.

Keywords—Carbon dioxide, gas separation, inorganic ceramic membrane & perm selectivity.

I. INTRODUCTION

ECOLOGICAL concerns have been recently escalating from a local to global scale due to increasing engineering and manufacturing activities. Most critical is the climatic change due to greenhouse gases, such as carbon dioxide, methane, nitrous oxides and fluorinated gases. Especially, a high concentration of carbon dioxide in the atmosphere can seriously give rise to a greenhouse effect, and of specific importance are, therefore, technologies effectively capable to remove atmospheric carbon dioxide, a key contributor to global warming [1], [2]. Capture of carbon dioxide from fossil fuel power plants is gaining widespread attention as a potential technique of regulatory greenhouse gas emissions. The control of these greenhouse gases is undoubtedly the most challenging environmental issue facing the entire globe. Therefore an approach that is of global interest is to regulate CO_2 emissions by capturing from fossil-fuel combustion sources. This would be a fundamental departure from traditional thinking about climate mitigation. With the well-known degree of dependence on fossil fuels on an increasing rate, the application of membrane technology using gas separation

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membranes is another prospective and efficient contender while avoiding greenhouse gas emissions as a potentially attractive alternative have been carefully considered [3]-[7]. This technology has been enhanced further by recent advances in materials evolution in engineering resulting in the invention of new materials with the thermal and mechanical stability required for most gas separations. Membrane technology offers cost-effective capital investment and is relatively less energy consuming. As a result, there is high demand for this technology in environmentally demanding processes leading to an outstanding effect in membrane technological market. To date, most of the membranes reported for CO_2 separation are self-supporting porous polymers. The major drawbacks of these membranes are low mechanical, chemical and thermal stability. In the cases of high temperature or high pressure, these polymers cannot control the shear stress at the interface, thus the separation layer may delaminate from the support with lapse of the operation time. The stability of composite membranes which comprise of numerous layers prepared with different materials of similar pore sizes is however determined by not only the separation layer but also the interface between the separation and support layers [8]. In particular, inorganic gas separation ceramic membranes which exhibit high chemical resistance, high mechanical strength with improved thermal stability and excellent catalytic properties are studied. Therefore to achieve optimization in membrane separation systems performance in an economical manner, the development of a reliable way of dealing with the design of membrane technology is gaining great attention [9]. Pan presented a model for the calculation and prediction of gas separation performance of permeators through an asymmetric membrane. The model was further verified by a large-scale field pilot-plant experiment for helium recovery from natural gas. Results obtained are in harmony with the experimental data [10]. A more comprehensive work on a model design of a high temperature membrane reactor was presented by Rui et al for the recovery and utilization of CO_2 from flue gas stream and for an instantaneous syngas production during CO_2 reaction with methane in the presence of reforming catalysts [11]. The model offers good prospects although could not find its applications in other mixtures involving O_2 in the permeation process. An accurate description of the process behavior in the membrane separation process is therefore an important factor in order to reduce some technical risks which could be encountered especially in relation to traditional separation technique. In addition, designing of process models for membrane gas separation in particular is vital and requires an extremely thorough and careful approach. Thus an accurate

and dependable simulation of the entire system can be employed for the design of the separation process. Subsequently, efforts made towards development of a detailed model for membrane gas separation has not gained enough grounds at the moment and not very readily available in most published literatures, although a limited number of unit models exists in [12]-[14]. In this work, an extension of the model framework in a nanostructured ceramic membrane as a high flux asymmetric gas separation membrane is being studied to validate the experimental results.

II. Scientifically Facilitated Model

A simulation study on mass transfer for a single-stage CO_2/N_2 membrane process with experimental validation is demonstrated. Fig. 1 shows a single asymmetric ceramic membrane operating in co-current flow. We note that the bulk of the permeate exiting the membrane (top layer) Y^1 is not the same as the amount leaving the outer section of the porous layer Y_i. Due to high permeate flux; gas flow resistance through coated layer of the support is minimal. The permeability characteristics of the membrane are not controlled by pressure and gas composition. The membrane is suitable as it can withstand high temperature and pressure due to its durability and high tensile strength. Thus, model is suitable for calculating the performance of binary gases with the high-flux asymmetric membrane and has been verified by experiments for CO_2 recovery from flue gas. All operating conditions in the simulation were applied during experimental validation [10].

Fig. 2 depicts the gas permeation operating parameters. For a single stage membrane process, applicable parameters used in this work under specific operational conditions are pressure, temperature (room) and gas flow rates. Others are selectivity, permeability, membrane surface areas and inlet feed composition. In addition variables that contribute to the membrane performance include the membrane selectivity usually predicted and calculated from the permeance ratio of pure gases using the expression below [15].

$$Selectivity, \alpha = \frac{\text{Permeance of CO}_2}{\text{Permeance of N}_2} \quad (1)$$
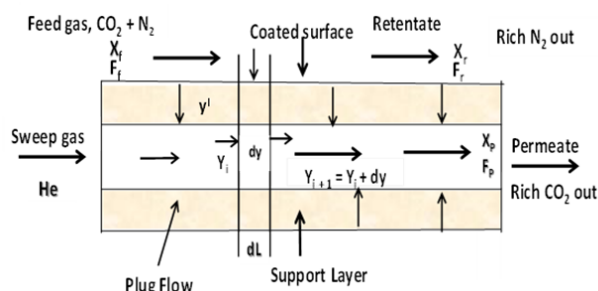

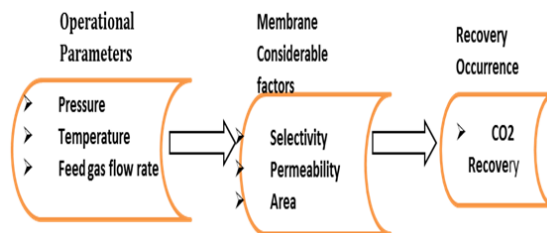Fig. 1 Gas permeation through a Nanostructured Ceramic membrane

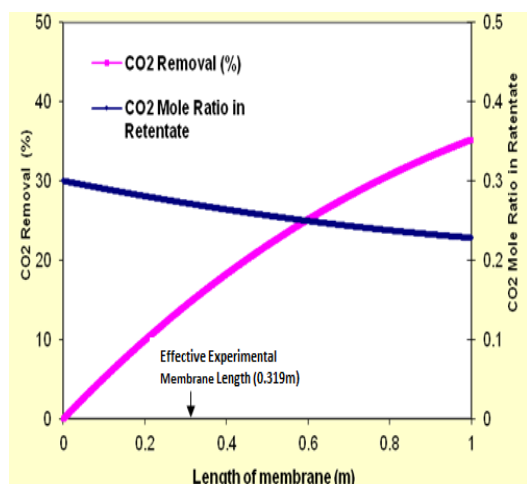Fig. 2 Gas permeation operating parameters

III. Experimental Methodology

The commercial ceramic support used was procured from Ceramiques Techniques et Industrielles (CTI SA), France and made up of 77% α-alumina + 23% TiO2 with an average pore diameter of 6000nm. The support has 20 mm and 25 mm internal and outer diameter respectively, and an effective penetrable length of 318 mm. The feed pressure applied was between 1 to 5 bars. Gas permeation experiments were carried out to examine and test CO_2 and N_2 permeation behaviors as well as the membrane separation performance. Fixed assumptions were made with respect to the membrane process operating conditions. However experimental values obtained from measurements carried out in the laboratory were used as input data in the membrane simulation (model). The selectivity of the membrane was also assumed so as to enable comparison of experimental and model parameters.

IV. Results and Discussions

The input parameters for the model and that of the experiment for validation are shown in Tables I and II. The determination of CO_2 gas percentage recovery is presented graphically in Figs. 3 and 4.

TABLE I
INPUT PARAMETERS IN THE MODEL

Feed:		
Volume flow rate	5	m^3/day
Mole flow rate	0.024	mole/s
Pressure	1013250	Pa
Temperature	298	K
Mole ratio of CO_2	0.3	
Mole Ratio of N_2	0.7	
Membrane Module:		
Effective length:	1	m
OD of membrane tube	0.01	m
ID of membrane tube	0.006	m
ID of module shell	0.0243	m
Selectivity of CO_2/N_2	10	
Permeability of CO_2	0.0000004	$mole/m^2.s.pa$
Permeability of N_2	0.00000004	$mole/m^2.s.pa$
Sweep Gas:		
Volume flowrate	50	m^3/day
Mole flowrate	0.028	mole/s
Pressure	121325	Pa
Temperature	298	K

Fig. 3 CO_2 removal and mole concentration in retentate in relation to membrane total length
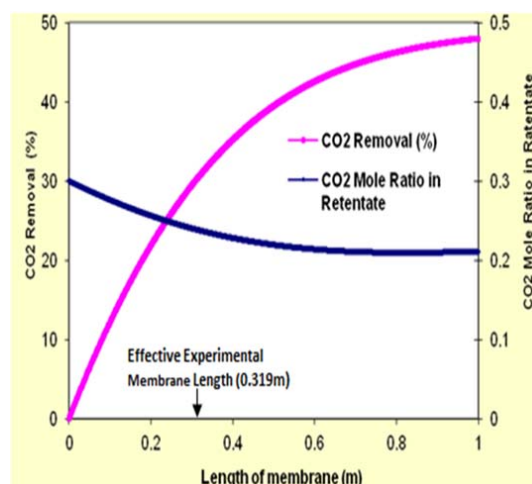
Fig. 4 Experimental CO_2 Removal and Mole Concentration in Retentate vs Membrane length

TABLE II
GENERATED RESULTS FROM THE MODEL

L	A (m²)	CO_2 in feed	CO_2 in Permeate	N_2 in feed	N_2 in Permeate	CO_2 Removal %
1	0.0314	0.228	0.713	0.772	0.287	35.20

TABLE IV
GENERATED RESULTS FOR EXPERIMENTAL VALIDATION

L	A (m²)	CO_2 in feed	CO_2 Permeate	N_2 in feed	N_2 in Permeate	CO_2 Removal%
0.319	0.0251	0.238	0.736	0.761	0.264	48.02

The plots display the CO_2 removal (%) and its mole ratio in the retentate in relation to the membrane length. For the model, an industrial prototype membrane length of 1m was used. Results generated from the simulation input lead to a CO_2 removal of 35.30% under the prevalent conditions and in the presence of a sweep gas as shown in Table II.

TABLE III
EXPERIMENTAL VALIDATION OF SIMULATION RESULT

Feed:		
Volume flow rate	5	m³/day
Mole flow rate	0.024	mole/s
Pressure	1013250	Pa
Temperature	298	K
Mole ratio of CO_2	0.3	
Mole Ratio of N_2	0.7	
Membrane Module:		
Effective length:	0.319	m
OD of membrane tube	0.025	m
ID of membrane tube	0.0198	m
ID of module shell	0.0548	m
Selectivity of CO_2/ N_2	10	
Permeability of CO_2	0.0000004	mole/m².s.pa
Permeability of N_2	0.00000004	mole/m².s.pa
Sweep Gas:		
Volume flow rate	50	m³/day
Mole flow rate	0.028	mole/s
Pressure	121325	Pa
Temperature	298	K

As can be observed in Table III, also, the temperature, pressure, and volume flow rate were assumed constant; this was done for the purpose of comparison. For the membrane module, the membrane parameters used were obtained from actual measurements done during experimental procedures using a nanostructured hybrid ceramic membrane. These values replaced that of the model. In addition, the membrane selectivity in the model was used as a guide for the validation. Thus results generated from the plot in Fig. 4 with respect to the experimental input at an effective membrane length of 0.319m. Results generated from the experiments carried out for gas separation lead to a CO_2 removal of 48.02%. This is illustrated in Fig. 4 and Table IV.

V. CONCLUSION

The objective of this work is to access the feasibility of membrane process for CO_2 capture from flue gas application, especially flue gas with lower CO_2 feed concentration. Permeability, selectivity and membrane surface area are significant parameters for membranes performance. Each of these parameters as identified should be taken into account by membrane inventors. Hence the capture target is high degree of CO_2 separation. This simulation analysis, has been conducted while bearing in mind the representative process design and operation parameters which reflect the scopes of a real flue gas treatment capacity. In this context, use of helium as sweep gas contributed towards increasing the separation efficiency of the membrane. Operating conditions such as CO_2 molar fraction in the feed gas, pressure and temperature dramatically influence the degree of separation. In principle, a single-stage process consumes less energy than a multi-stage membrane system will become more competitive with respect

to energy consumption. It can however be concluded from this work that by the development of simple model, it is possible to attain higher CO_2 recovery of 48% from flue gas mixture. However, permeation tests are on-going to certify the permeability of gases and durability of the model under extreme process conditions.

NOMENCLATURE

F_f = Flow rate of gas component in the feed side (l/min)
F_r = Flow rate of gas component in the retentate side (l/min)
F_p = Flow rate of gas component in the permeate side (l/min)
X_f = Mole fraction of gas component in the feed side (%)
X_r = Mole fraction of gas component in the retentate side (%)
X_p = Mole fraction of gas component in the permeate side (%)
Y^I = Bulk of the permeate exiting the membrane (top layer)
Y_i = Bulk of permeate exiting the outer section of porous layer

REFERENCE

[1] Takaba H, Mizukami K, Kubo M, Stirling A, Miyamoto A. The effect of gas molecule affinities on CO 2 separation from the CO 2/N 2 gas mixture using inorganic membranes as investigated by molecular dynamics simulation. Journal of Membrane Science. 1996; 121(2):251-259.

[2] Niswander R, Edwards D, DuPart M, Tse J. A more energy efficient product for carbon dioxide separation. Separation Science and Technology. 1993; 28(1-3):565-578.

[3] Rao AB, Rubin ES. A technical, economic, and environmental assessment of amine-based CO2 capture technology for power plant greenhouse gas control. Environmental science & technology. 2002; 36(20):4467-4475.

[4] Herzog H, Drake E, Adams E. CO2 capture, reuse, and storage technologies for mitigating global climate change. A White Paper. 1997

[5] Nwogu NC and Gobina G. Advanced Membrane Design for Improved Carbon Dioxide Capture. Abstracts of Papers of the American Chemical Society: Amer Chemical Soc 1155 16th St, Nw, Washington, Dc 20036 Usa; 2013.

[6] Nwogu N, Kajama M, Okon E, Shehu H, Gobina E. Testing of Gas Permeance Techniques of a Fabricated CO2 Permeable Ceramic Membrane for Gas Separation Purposes. 2014.

[7] Nwogu NC, Gobina E, Kajama MN. Improved carbon dioxide capture using nanostructured ceramic membranes. Low Carbon Economy. 2013; 4(03):125.

[8] Kluiters S. Status review on membrane systems for hydrogen separation. Energy Research Centre of the Netherlands: Petten, The Netherlands. 2004; 168.

[9] El-Halwagi MM. Synthesis of reverse-osmosis networks for waste reduction. AIChE Journal. 1992; 38(8):1185-1198.

[10] Pan C. Gas separation by permeators with high-flux asymmetric membranes. AIChE Journal. 1983; 29(4):545-552.

[11] Rui Z, Anderson M, Lin Y, Li Y. Modeling and analysis of carbon dioxide permeation through ceramic-carbonate dual-phase membranes. Journal of Membrane Science. 2009; 345(1):110-118.

[12] Krovvidi K, Kovvali A, Vemury S, Khan A. Approximate solutions for gas permeators separating binary mixtures. Journal of Membrane Science. 1992; 66(2):103-118.

[13] Qi R, Henson MA. Modeling of spiral-wound permeators for multicomponent gas separations. Industrial & Engineering Chemistry Research. 1997; 36(6):2320-2331.

[14] Coker D, Freeman B, Fleming G. Modeling multicomponent gas separation using hollow-fiber membrane contactors. AIChE Journal. 1998; 44(6):1289-1302.

[15] Bounaceur R, Lape N, Roizard D, Vallieres C, Favre E. Membrane processes for post-combustion carbon dioxide capture: a parametric study. Energy. 2006; 31(14):2556-2570.