CFD Modelling of CO_2 Removal from Natural Gas Using Graphene Based Membrane

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The modeling of CO_2 removal from CO_2/CH_4 mixture is performed using the Ansys Fluent 15.0 software. The effect of thickness of polyethersulfone-reduced graphene oxide-zeolitic imadozole framework 8 (PES/RGO-ZIF8) membrane and the feed pressure on the ability of membrane to remove CO_2 are investigated. The increment of thickness resulted in the increase of CO_2 removal. Meanwhile, the increase of feed pressure causes the CO_2 permeance to reduce. Various parameters should be manipulated in order to obtain more accurate and reliable result of membrane's performance in gas separation process

Keywords— membranes, permeability, thickness, feed pressure.

I. Introduction

Generally, carbon dioxide (CO_2) does not pose any harmful threats when it exists as a minor component in the air. Nevertheless, if the percentage of CO_2 are higher than the threshold limit, it would cause serious damages to the environment and human (IPCC, 2005). Besides that, CO_2 can be found in the natural gas reservoirs and it also can be produced from the reaction of certain activity such as gasification of coal, anaerobic digestion of biogas and fossil fuel combustion (Zhang et al., 2013).

This study focuses on the CO_2 removal from natural gas. The main purpose of it, is to prevent corrosion of the pipeline as CO_2 tends to be corrosive in the present of water (Yeo et al., 2012). Apart from that, it helps to reduce the CO_2 content released into the atmosphere since CO_2 is one of the major component of greenhouse gases (GHG) that contributed in the global warming phenomenon (George et al., 2016). Datta and Sen (2006) discovered that through the removal of CO_2, the transportability and calorific value of natural gas are improved.

The conventional methods developed to remove CO_2 from natural gas such as absorption, adsorption and cryogenic separation were found to be inefficient and non-cost effective (Yeo et al., 2012). This situation triggered the researchers to explore into the membrane technology for gas separation process. Some of the benefits of using membrane are the simplicity of the separation process, low maintenance required for the process and the equipment used is compact and lightweight (Yeo et al., 2012). There are various types of membrane developed over the decade for industrial usage such as polymeric, inorganic, and mixed matrix membrane.

Inorganic membrane is suitable to be used for separation process that operated at high pressure and temperature. Based on the study performed by Li et al. (2005) and Zhu et al. (2006), zeolite membrane which is categorized as porous inorganic membrane are preferred for the CO_2 separation from CH_4. This is because the membrane shows a remarkable selectivity compared to polymeric membranes and the good chemical resistance towards CO_2 property. Depending on the zeolite material composition, the CO_2/CH_4 selectivity could be affected. Despite the excellent performance in gas separation, zeolite membranes have a few lacking points such as the difficulty in processing and handling, and the very high fabrication cost (Iarikov & Oyama, 2011)

The advantages of polymeric membrane over inorganic membrane are low fabrication cost, the ability to reproduce and easy to fabricate. However, problems of selectivity loss and plasticization occurred as attempts to increase the permeability of polymeric membranes were made (Jusoh et al., 2016). Throughout the decade, various types of membrane have been developed to satisfy the optimum performance of gas separation. Composite organic–inorganic membranes or mixed-matrix membranes consist of inorganic particles incorporated into a polymer matrix (Iarikov and Oyama, 2011). He et al. (2018) mentioned in their study regarding the good transport properties of mixed matrix membrane possessed. The separation performance could improve significantly with the addition of a small quantity of inorganic filler in the polymer matrix.

Lately, there are quite a number of studies on the usage of metal organic frameworks (MOFs) as filler materials due to their high surface area, high pore volume, low density, and narrow pore size distribution (Li et al., 2018). Due to easiness of synthesis, tunable pore size, and thermal, mechanical, and chemical stability, zeolitic imidazolate framework-8 (ZIF-8) has been widely used in polymeric membrane to investigate the gas separation performance. Meanwhile, GO has been studied as an emerging membrane material because of its high aspect ratio (>1000), good compatibility with polymers, and thermal and mechanical properties (Li et al., 2015). However, the main problem for GO-polymer membranes is the relatively low gas permeability (Dong et al., 2016). Thus, the idea of combining GO and MOF as filler in polymeric was established to overcome each filler's drawbacks.

The objective of this study is to model the flow of CO_2 removal from CO_2/CH_4 mixture that represent as natural gas stream, and to observe the effect of membrane's thickness and the feed pressure on the performance of membrane in removing CO_2. The type of membrane used is a flat sheet mixed matrix membrane made up from polyethersulfone with two type of fillers which are reduced graphene oxide and zeolitic imidazolate framework.

II. METHODOLOGY

This study made used of the computational fluid dynamics (CFD) method to model the flow of CO_2/CH_4 in the flat sheet membrane and to observe the effect of membrane thickness on the CO_2 removal performance. CFD Ansys Fluent 15.0 is usually used to predict and evaluate the membranes performance by manipulating the parameters such as pressure on the permeate side, temperature, mass flow rate of feed and other relevant parameters related to the study that is going to be performed. This method is preferred because researchers does not have to waste their time and money in developing new set of membranes to obtain the information for each case that they are concerned about.

Modeling Geometry

Dead-end flow membrane cell is suggested for the two-dimensional model used in this work. The mixture of CO_2 and CH_4 is fed into the permeation cell which then moves along the membrane to reach the permeate side that is exposed to the atmospheric pressure. The CFD modeling of this study is based on Gilassi and Rahmanian (2015) with a slight alteration made, where the sudden expansion and contraction at the inlet zone and outlet zone caused by the changes in pipe size, slip boundary condition at membrane's surface, and also the effect of membrane's elasticity are not considered. The modeling geometry and boundary condition is presented as below:

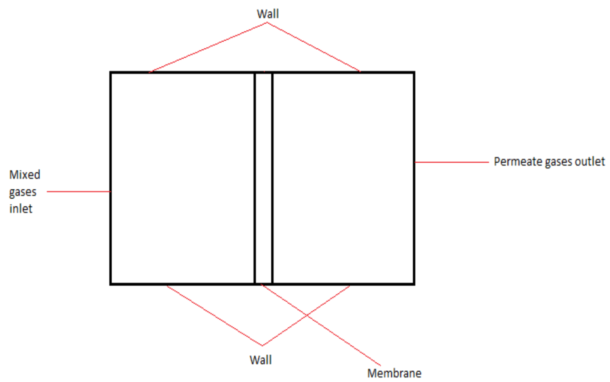

Figure 1: Modeling geometry schematics diagram

Governing Equations

The domain for this study consists of three parts, the inlet zone that contain gases mixture, the membrane and also the outlet zone that contain permeate gases. The conservation of mass equation or mostly known as Continuity equation and the momentum equations are applied to develop the two-dimensional model.

The equation of continuity is expressed as:

$$\frac{\partial u_x}{\partial x} + \frac{\partial u_y}{\partial y} = 0$$

Meanwhile the momentum equation can be written as:

$$\rho\left(u_x\frac{\partial u_x}{\partial x} + u_y\frac{\partial u_x}{\partial y}\right) = -\frac{\partial P}{\partial x} + \mu\left(\frac{\partial^2 u_x}{\partial x^2} + \frac{\partial^2 u_x}{\partial y^2}\right) + \rho g_x$$

$$\rho\left(u_x\frac{\partial u_y}{\partial x} + u_y\frac{\partial u_y}{\partial y}\right) = -\frac{\partial p}{\partial y} + \mu\left(\frac{\partial^2 u_y}{\partial x^2} + \frac{\partial^2 u_y}{\partial y^2}\right) + \rho g_y$$

And the concentration equation is represented as:

$$u_x\frac{\partial C_{co_2}}{\partial x} + u_y\frac{\partial C_{co_2}}{\partial y} + D\left(\frac{\partial^2 C_{co_2}}{\partial x^2} + \frac{\partial^2 C_{co_2}}{\partial y^2}\right) = 0$$

Where P, D_m, ρ, C_{co_2}, u_y and u_x represents the pressure, diffusivity coefficient, CO2 concentration, density and the velocity in the x and y direction.

Computational Model

For the pressure-velocity coupling, SIMPLE (SemiImplicit Method for Pressure Linked Equations) scheme is used, and all of the spatial discretization used the second order upwind scheme. The flow is considered as steady state incompressible laminar flow. The membrane used for this modelling is a type of polymer – metal organic framework composite membrane which is polyethersulfone-reduced graphene oxide-zeolitic imadozole framework 8 (PES/RGO-ZIF8).

Variables	*Unit*	*Values*
Velocity	m/s	0.0006
Pressure drop	bar	1
Temperature	ºC	30
Diffusivity coefficient	m^2	0.00001
Membrane thickness	mm	0.14
Membrane area	cm^2	19.64
CO_2 volume fraction	-	0.5
CH_4 volume fraction	-	0.5
CO_2 density	kg/m^3	1.7878
***CH_4** density*	kg/m^3	0.6679

Table 1: Operating condition of the permeation cell

III. RESULTS AND DISCUSSION

A. The Effect of Thickness on Membrane's Performance

i) 0.18mm thickness of membrane

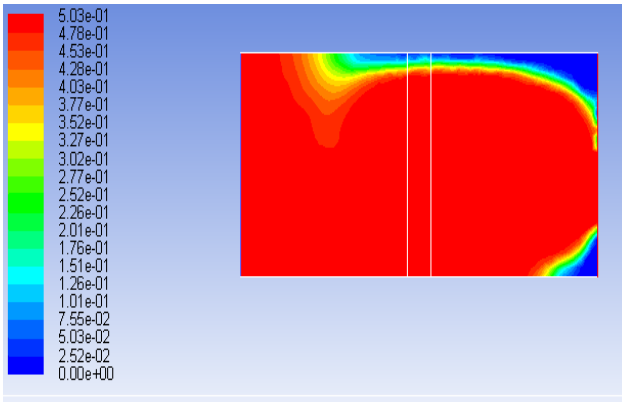

Figure 2: CO_2 concentration gradient along the x-direction

ii) 0.16mm thickness of membrane

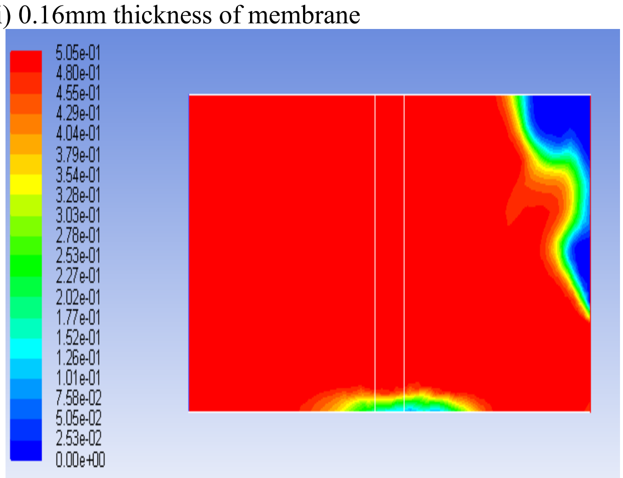

Figure 3: The concentration gradient of CO_2 in x-direction

iii) 0.14mm thickness of membrane

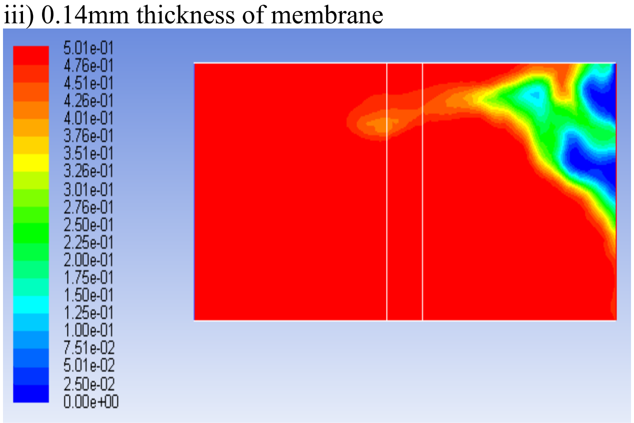

Figure 4: The concentration gradient of CO_2 in x-direction

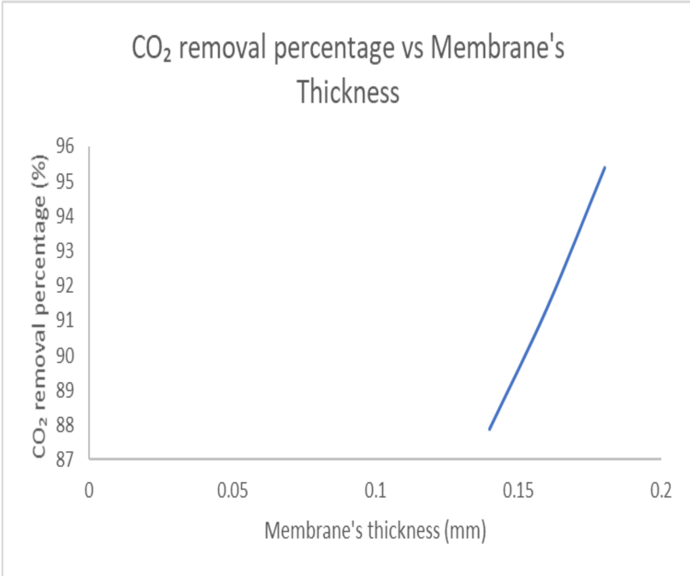

Figure 5: The percentage of CO_2 removal against different membrane's thickness

As shown above in the graph, the CO_2 removal percentage increases with the thickness of the membrane. This is because the membrane's thickness influences the permeability of the gas passing through it. The greater the thickness of a membrane, the higher the permeability of the gas. Thus, reducing the required area of membrane for the separation process, which in turn could help to reduce the cost of membranes development. There is lack of documented study regarding the flat sheet membrane's thickness effect on CO_2 removal from CH_4, therefore the result obtained is compared with the study of single gases permeability and different gas mixture through the membrane of different thickness.

Shen and Lua (2010) discovered that the permeability of single gases (He, N_2, O_2 and CO_2) relies on the membrane thickness. As the thickness of membrane increases from 6 to 309µm the single gases permeability increases by more than three times. Meanwhile, Alsari et al. (2007) stated in their study that for the mixture of O_2 and N_2, the permeability of O_2 decreases as the membrane's thickness increases. On the other hand, N_2 permeability increases along with thickness of membrane. However, for the single gas test performed they found out that the permeability of O_2 and N_2 increases as the thickness increase.

B. Effect of Feed Pressure on Membrane's Performance

As for this part of the study, membrane with a thickness of 0.14mm has been used to test for different feed pressure of 1 bar, 3 bar and 5 bar

i) 1 bar

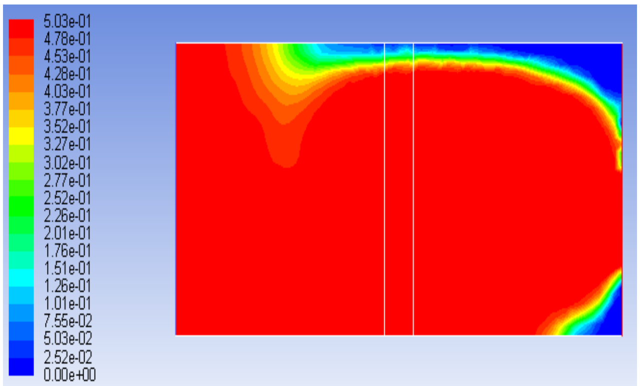

Figure 6: The CO_2 concentration gradient along the x-axis

ii) 3 bar

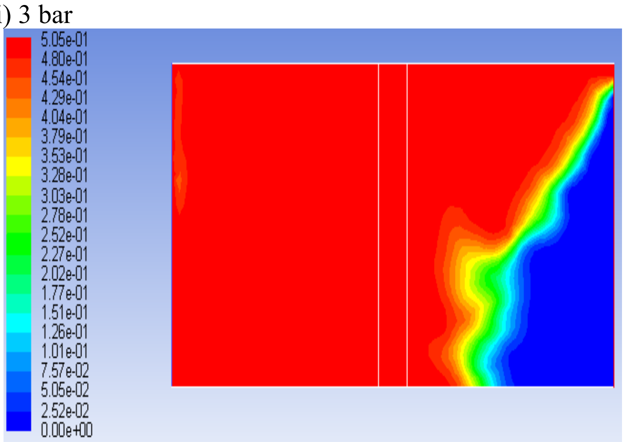

Figure 7: The CO_2 concentration gradient in x-direction

iii) 5 bar

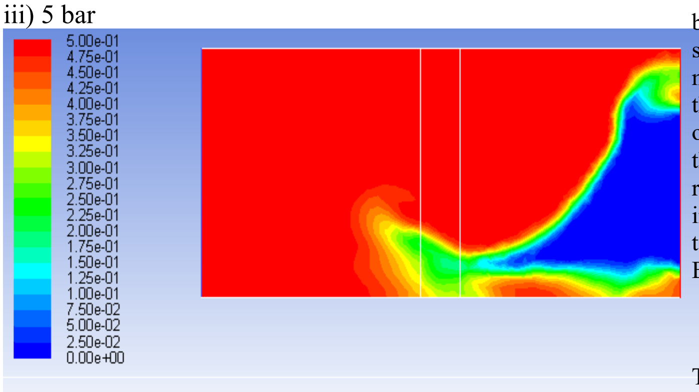

Figure 8: The concentration gradient of CO_2 in x-axis

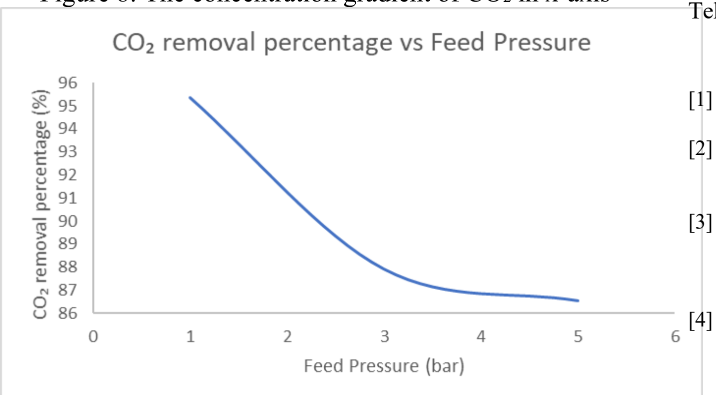

Figure 9: The effect of feed pressure on the CO_2 removal percentage

From the result obtained, it can be seen that the increase in feed pressure causes the removal of CO_2 percentage to decline, which can be justified by the statement made from Dortmundt and Doshi (1999), where the permeability and selectivity of membrane decreases as the feed pressure increases. Yoshimune and Haraya (2013) investigate the effect of total feed pressure on the permeation properties of single and binary CO_2/CH_4 mixture. The permeance of CO_2 slightly decrease due to the increase of total feed pressure, while CH_4 permeances remained nearly constant. They came out with an assumption that the phenomenon can be attributed to the Langmuir-type adsorption effect of CO_2, in which adsorption coefficients decrease with increasing relative pressure.

However, a study conducted by Sridhar et al. (2012) using a plain PEBAX-2533 membrane recorded a contrast result. The CO_2 permeance increases as the pressure was increased from 9.8 bar to 29.4 bar. They concluded that the gradual increase of CO_2 permeance with the increase of feed pressure is attributed to increased sorption of the soluble CO_2 gas in the membrane, since it has a more preferential affinity than CH_4. The conclusion that can be made from the result obtained which is then compared to the previous studies, is that the type of membrane used for gases separation have a significant effect on the permeability and selectivity of the gases due to different physical and chemical properties of the membrane synthesised.

IV. Conclusion

The Computational Fluid Dynamic (CFD) study of graphene based membrane was carried out using the Ansys Fluent 15.0 software. The flow of CO_2 removal from CO_2/CH_4 mixture is modelled using the software and the effect of membrane's thickness and feed pressure on the percent of CO_2 removal was observed. From the result gained, it can be concluded that the thicker the membrane, the better performance of membrane in removing CO_2 from the mixture stream. In the meantime, the increase in feed pressure will cause the CO_2 removal to decline due to the decrease of permeability and selectivity of the membrane. Enhanced study should be

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