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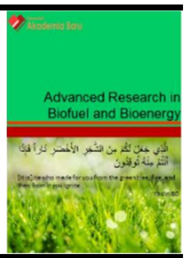

Upgrading Biogas by Membrane Contactor Separation

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ABSTRACT

Biogas consists of mixture of gases, among which methane and carbon dioxide are major composition. The upgrading biogas involves the process of separation methane from other mixture of gases in raw biogas especially carbon dioxide to get high yield of biomethane. The present of other mixture of gases drops the calorific value of biogas and it reduces the quality of biogas. In this research, the membrane contactor separation was used to upgrade the biogas concentration. The main purpose of the research is to maximize the composition of methane in biomethane produced after separation process conducted by determining the condition or parameter during separation process. The parameter involved was inlet pressure which pressure above 2.5 bar had promising the higher biomethane composition obtained, as the inlet pressure increases, the composition of biomethane obtained increases. At inlet pressure 2.5 bar, the composition of methane can achieve 71.2% with 21.1% of carbon dioxide composition at permeate stream. Besides, the type of adsorbents used affect the efficiency of membrane contactor. The adsorbents used for this experiment were molecular sieve 13X and granular activated carbon which 2.5 bars inlet pressure was applied. The molecular sieve had better performance in the separation of methane and carbon dioxide with 64.7% of methane composition at peak compared to the granular activated carbon with 60.0% of methane composition at peak. The operating parameter and type of adsorbents used influenced the performance of membrane as well as the quality of biomethane produced during separation process. As conclusion, process parameter and adsorbents type are crucial components in membrane contactor separation process to upgrade biogas.

***Keywords*:**
Upgrading biogas, membrane contactor, activated carbon and molecular sieve

1. Introduction

Recently, biogas production is a part of biomass energy resources which is growing rapidly to cater the global energy demand. The biogas can be considered as an alternative energy or renewable energy as it can replace fossil fuels and coals for heat and power generation [1]. It also provides high market potential since the source availability of biogas production is still abundant. Before that, the raw biogas obtained should be cleaned or upgraded first.

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Based on the recent study [2], there are increasing trends of the emerging biomethane plants from 2011 to 2015. This proves the potential of biogas plant in the future as the number of biogas plants constructed keep on increasing with more than 20% rate. According to the IEA Bioenergy Task 37 which was reported by researcher[2], most of the biomethane plants are constructed in Germany with 188 plants, followed by Sweden with 59 plants and United Kingdom with 50 plants.

The composition of raw biogas consists of methane (CH_4) with range 50–70% and carbon dioxide (CO_2) in between 30–50%. Other than those gases, the minor gases concentration include; water vapor (H_2O) with 5–10 %, hydrogen sulfide (H_2S) at concentrations of 0-10 000 ppm, siloxanes with concentrations of 0-41 $mg.m^{-3}$, ammonia (NH_3), oxygen (O_2) with concentrations of 0–1%, nitrogen (N_2) with concentration of 0-3% and other hydrocarbon present with concentration of 0-200 $mg.m^{-3}$ [3,4]. The purified biogas should only contain methane gas whereas the others are considered as biogas pollutants. Based on this, the biogas needs to be upgraded since the biomethane used in the industry should contain methane gas more than 97% for the utilization in biofuel energy [5]. The upgraded biogas can be used as a substitute of compressed natural gas (CNG) for transport fuel [6,7], if the biomethane produced meets the quality of CNG.

The upgrading biogas process is one of the crucial process to improve the quality of biogas. In terms of fuel efficiency, the calorific value for biogas is 21.5 MJ/m^3 while the calorific value for pure methane is 39.8 MJ/m^3[6, 8]. Without undergoing upgrading process, the calorific value of biogas drops, as it cannot be used as CNG in fuel industry and power supply [9, 10]. There are limits for the composition of methane needs to have so that it can be applied in both industries. If there is no upgrading biogas process occurred, the usage of biogas narrowed to different industry. The upgrading biogas process also helps to eliminate the greenhouse gases as the methane and carbon dioxide gases are part of it [11]. The biogas available is too abundant and it needs to be reused as other resources or products. By upgrading biogas, the biomethane can be used to replace fossil fuels as fuel new source and also can reduce the abundancy of untreated biogas.

In gas separation technology, the method for the upgrading biogas process has been varied in many ways. A lot of gas separation technologies have been introduced which has been improvised from time to time. The current gas separation technologies available that had been introduced which included membrane separation, cryogenic technology, Pressure Swing Adsorption(PSA), water scrubber and amine scrubber [12]. This shows that how important biogas plants in current industry as the improvements are needed to increase the quality of biomethane. The water scrubber process has been used in practice in most commercialize biogas plants . This situation reflects that the water scrubber process more commercialized than the targeted separation process in this study, which is membrane separation process. As the water scrubber technology has been used and introduced earlier than membrane separation technology, this does not prove that the latter technology is not as great as the water scrubber. In term of operation process, the membrane separation is cost-effective, simple and easy to scale up [13]. The membrane separation process does not require large space which also helps to reduce the capital investment [13].This proves that membrane separation process is suit to be used in industrial process. A lot of potential and advantages can be rediscovered from the membrane separation technology. Because of this, the research studied more extensive in membrane separation technology and to prove that this technology is one of the best technology in upgrading biogas.

The study is focused on maximizing the yield of biomethane at end of the separation process by determining the process parameters in filtration and analyzing the membrane characteristics. The parameters involved are the operating pressure and the type of absorbents material can be used in membrane separation. These parameters can be useful for the database in research and development especially in upgrading biogas via membrane separation. The membrane used for this study is ceramic membrane which included with two types of adsorbent materials; activated carbon and molecular sieve. The adsorption study included as the type and arrangement of adsorbent used, contribute to the membrane separation operation. While the operating parameter used to study and determine the process parameters that contribute to maximize the biomethane production.

2. Experimental Method

2.1 Materials and Equipment

The raw biogas obtained was in the form of crude biogas which was supplied by the biogas production company such as Cenergi Sdn Bhd. Two types of adsorbents were used for this project, molecular sieve 13X and granular activated carbon. The molecular sieve used was in desiccant type which the component ratio of 1:1:2.8 with sodium oxide, aluminium dioxide and silicates respectively.The arrangement of adsorbent types were shown in Fig. 1. The type 1 arrangement involved activated carbon which fully filled the membrane vessel including the inside of ceramic membrane. While the Type 2 arrangement replaced the activated carbon with molecular sieve. The Type 3 arrangement utilized the mixer of both molecular sieve and the activated carbon.

Fig. 1. The Type 1, 2 and 3 arrangements of adsorbent from left to right

All the final composition of biogas was measured using gas analyzer. This project was using ceramic membrane which incorporated molecular sieve and activated carbon in order to be in form of membrane contactor. The ceramic membrane was placed inside the membrane vessels that had one inlet for feed gas and two outlets for permeate and retentate stream. Others equipment included pressure gauges and flow meters to observe the inlet pressure.

2.2 Method

There were two experiments conducted for this study which was setup as in Fig. 2. The first experiment was conducted to determine the operating parameter which was inlet pressure for upgrading biogas using membrane contactor. The second experiment was to compare the type of adsorbent materials that can be used in methane and carbon dioxide separation.

The first experiment started with inlet pressure 0.5 bar for Type 1 adsorbent arrangement. Both the composition of biogas in the permeate and retentate stream were measured for one minute. This step was to ensure the reading of biogas was stable and accurate. Then, the experiment was repeated with inlet pressure 1 bar up until 2.5 bars. As the Type 1 arrangement was done, the experiment was repeated with Type 2 and 3 adsorbent arrangement. The result of the experiment was recorded, tabulated and analysed.

The second experiment was conducted at 2.5 bars for each type of adsorbents arrangement. During separation, the biogas composition in both permeate and retentate stream were measured every ten seconds up until one minute by using biogas analyser. The result obtained was recorded and tabulated.

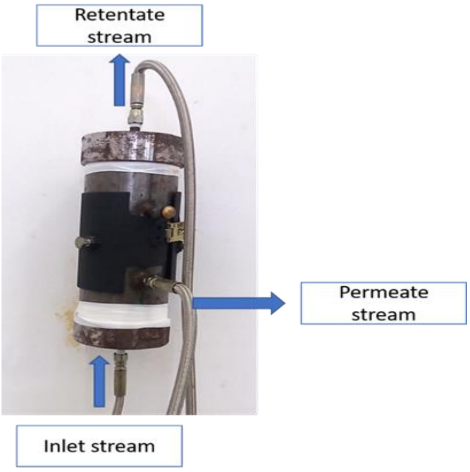

Fig. 2. Experimental set-up of membrane contactor separation process

3. Result and Discussion

3.1 Determining the Inlet Pressure

Based on Fig. 3, permeate 1 and retentate 1 referred as streams in Type 1 arrangement, permeate 2 and retentate 2 referred as streams in Type 2 arrangement, and permeate 3 and retentate 3 referred as streams in Type 3 arrangement. The permeate 2 stream shown the best performance for the separation process as the methane content in permeate stream is higher than in the retentate stream with the more than 70% of methane gas at 2.5 bars. It attained the highest composition of methane and the lowest composition of carbon dioxide at 2.5 bars. This clearly showed that molecular sieve performed well in permeate stream as at high pressure applied, the methane content was increasing while the carbon dioxide content was decreasing. The membrane separation process becomes effective when the composition of methane increasing in one stream while the composition of carbon dioxide increasing in other stream as the pressure inlet increasing. Thus, the molecular sieve is the best adsorbent for membrane contactor separation at 2.5 bars pressure inlet applied.

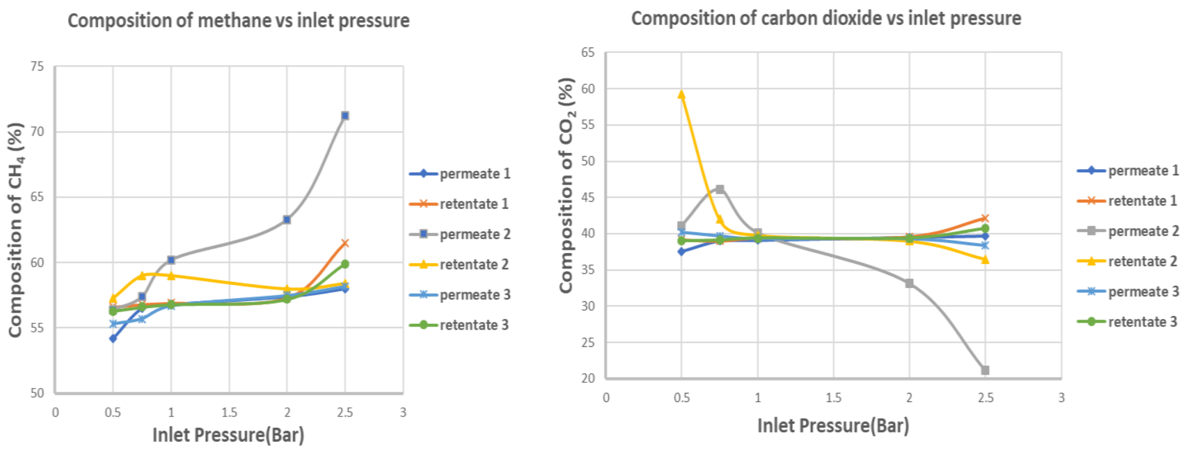

Fig. 3. The summarized results for determining the inlet pressure

All these difference in composition value for all type of adsorbents were influenced by the adsorption rate of carbon dioxide along with the membrane. According to [14], the adsorption rate of molecular sieve 13X is higher than activated carbon at low pressure (below 5 atm). Above 5 atm, the adsorption rate of activated carbon is higher than the molecular sieve. This parameter reflects on the mass transfer rate of carbon dioxide to adsorbent. The carbon dioxide composition should be decreased in both outlet stream as the mass transfer between adsorbate and adsorbent increased when the pressure increased. As the composition of carbon dioxide decreased, the methane composition should be increased. Comparing the type of arrangement of adsorbents, Type 2 arrangement with molecular sieve gave the best outcome data as the composition of methane increased to 71.2% while the carbon dioxide composition with 21% as the inlet pressure at 2.5 bars when comparing the composition of both gases in the inlet stream. The inlet pressure can be higher than 2.5 bars in order to achieve higher methane content around 90% with higher carbon dioxide being adsorbed by adsorbents [15]. The difference in membrane gas separation with the membrane contactor was showed in this data as the methane content high in permeate stream rather than retentate stream

3.2 Comparing the Type of Adsorbents

The data was collected on two types of arrangements only, which was Type 1 and 2 since the gas analyzer broken down before starting the other arrangements. Based on Fig. 4, permeate 1 and retentate 1 referred as streams in Type 1 arrangement while permeate 2 and retentate 2 referred as streams in Type 2 arrangement. By referring to the both graphs in the Fig. 4, both type of arrangements had steady trends, as the time taken for the separation process increasing, the composition of both gases constant after 20 seconds experiment conducted. The composition of methane had constant amount after 20 seconds at the range 57 to 60% except for the permeate 2 stream with 65% methane composition While the carbon dioxide content constant at around 40% after 20 seconds.

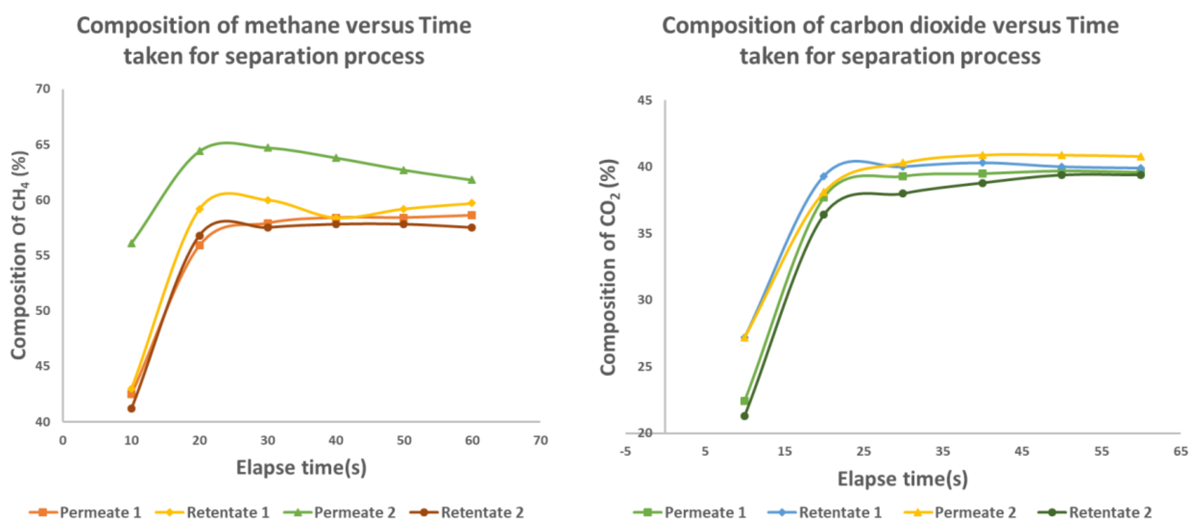

Fig. 4. The summarized results for comparing the type of adsorbents

For overall comparison in the performance of adsorbents, Fig. 4 shown the comparison according to the composition of gases involved. For the composition of methane, the permeate 2 had the highest methane content which peaked at 65% of methane along with the time taken. The other streams had similar composition of methane at the range of 55 to 60%. While the composition of carbon dioxide for all streams had slight difference only with permeate 2 had slightly the highest among the other stream at the end of the experiment. The range for carbon dioxide for all streams were around 37 to 40%. This reflected on the adsorption rate of molecular sieve was higher than activated carbon at 2.5 bars since the composition of methane was the highest in the permeate stream 2.

Generally, the carbon dioxide supposed to be adsorbed quickly on the adsorbent surface at the initial stage of adsorption. The rate became slower as the time increasing, until it reached adsorption equilibrium. Based on Fig. 4, the adsorption process reached its equilibrium after 20 seconds. There was no increment on the composition of both gases after 20 seconds. Comparing the composition of methane from both type of adsorbents, Type 2 which was molecular sieve had higher amount of methane compared to Type 1. As mentioned before, this experiment conducted at 2.5 bars inlet pressure which supposed the highest methane composition should be achieved for all type of adsorbents. Comparing the composition of carbon dioxide, both absorbents had similar value at 40% composition. This clearly showed that Type 2 which was molecular sieve 13X is better adsorbent than Type 1(activated carbon). According to the [14], the mass transfer coefficient of molecular sieve or zeolite 13X was higher than activated carbon. This explained why the composition methane in molecular sieve was higher than in activated carbon.

4. Conclusions

The membrane separation technology has outstanding potential that can be grown more in upgrading biogas industry. A variety improvement can be made such as improvised both the feed parameters and membrane characteristics to increase the quality and purity of biomethane. The inlet pressure for membrane separation can be operated above 2.5 bars in order to achieve high methane composition in membrane contactor, the types of adsorbent used plays vital role in separation process as it affects the efficiency of separation between methane and carbon dioxide. The usage of solid adsorbent still not widely known as the commercialized industry used liquid absorbent in membrane contactor separation. Based on the result obtained, the molecular sieve has big potential to be solid adsorbent in membrane contactor. Those components play crucial roles in membrane separation as its control the product quality. As mentioned earlier, the importance in upgrading biogas should be recognized as biomethane is a part of renewable energy resources, which one of the best candidate to replace the fossil fuels resources. The biogas plant should be introduced and constructed in each country as it can contribute in supplying energy sources to country. The biogas benefits should be utilized at its maximum as it can be used in various industry present.

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