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Design and economics of a hybrid membrane-temperature swing adsorption process for upgrading biogas^s

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Highlights

A hybrid membrane-temperature swing adsorption process for upgrading biogas to pipeline quality methane is presented.

Using an objective function a multi-stage process involving temperature swing adsorption and membranes was optimized.

Techno-economic analysis showed that for processing 200 Nm³/h biogas stream, a payback time on the investment was 6.8 months.

Abstract

Processing biogas from wastewater digesters allows recovery of valuable methane and reduction in green house gas emissions. A two-stage membrane process, coupled with a temperature-swing-adsorption (TSA) as pre-treatment, was designed to generate pipeline quality methane. To improve methane recovery and process energy efficiency, the non-product streams of the membrane process were recycled and the permeate of the first membrane stage was maintained at a given pressure as the driving force for second membrane stage. The membrane process design was optimized by minimizing the objective function; the overall processing cost. It was found the membrane approach excels the PSA for producing pipeline quality methane (97% purity) in terms of methane recovery, processing cost and lower emissions. A techno-

economic analysis showed that the payback time for an operation processing 200 Nm³/h of biogas was 6.8 months.

Keywords: process design and optimization, hollow fiber membrane, pressure ratio, recycling, biogas emission, capital and operating cost, economic analysis

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1. Introduction

Huge volumes of biogas are produced daily in wastewater plants around the world. Biogas is produced from the degradation of organic constituents in wastewater [1]. Typically, it contains 55-65% methane, 35-45% carbon dioxide, and some minor components including water vapor, up to a few thousand ppm hydrogen sulfide, and small levels of volatile organic compounds (VOCs) and siloxanes [2]. The earliest strategy in dealing with biogas was burning it as a waste gas and only a small portion of it was utilized to provide heating. Obviously this practice was not environmentally favorable. Sustainable development calls for minimized green house gas emissions from every industrial process, including that for waste management.

Pressure Swing Adsorption (PSA) as a mature technology, has gained wide application in dealing with environmental concerns. Various efficient adsorbents have been made available for removing both the minor species and major components from the biogas mixture. In a PSA process for upgrading bio-methane and recovering the majority of CO₂, the biogas feed stream is usually pressurized to 7-14 bar. The minor components and CO₂ are captured in the respective adsorption towers arranged in series. Finally, the upgraded bio-methane containing ~3 vol.%

CO₂ is injected into the pipeline grid as a source of energy or feedstock [3], and the desorbed CO₂ may be converted into liquid phase for ease of transport to a sequestration point [4].

Compared with the membrane process, PSA has several drawbacks. 1) PSA is an intermittent operation and it needs periodic regeneration of the adsorbents. The intermittent nature of PSA not only increases the operational complexity and labor requirement but also increases the capital cost, since keeping an extra adsorption process within the system is essential to enable the overall operation to be continuous. 2) PSA is not a completely closed system and produces emissions. 3) Membrane based separation tends to be more cost-effective in manufacturing relatively low-purity products. For producing pipeline quality methane and capturing CO₂, membrane tends to outperform PSA [5, 6]. This will be verified later in this work.

Broad commercial successes in gas separation have been achieved by membranes. Processes benefiting from this burgeoning technology have spread from the ammonia synthesis (H₂/N₂ separation) [7], oxygen-enhanced combustion (O₂/N₂) [8, 9], to monomers (e.g., C₂H₄, C₃H₆) recovery and VOCs (e.g., gasoline, CH₃Cl) removal from contaminated air streams [10, 11]. Much of the recent research effort has been directed to natural gas processing and GHG emission reduction (e.g., CO₂/CH₄, CO₂/N₂), which represents a tremendous future market [12, 13]. The major membrane materials tested for the application include cross-linkable polyimide (PI) [14-17], and organic-inorganic composites comprising of cross-linkable PI and carbon molecular sieves or zeolites [18, 19], the typical CO₂/CH₄ selectivity of membrane 30-50, and CO₂ permeance of up to 50 GPU (3.35×10^{-10} (mol/(m² s Pa))) were reported. Recently, a hollow fiber membrane based on polyetherimide-bismaleimide (PEI-BMI) interpenetrating polymer networks

[20] has been synthesized in our laboratory, which displays a typical CO₂/CH₄ selectivity of 55 with a CO₂ permeance of 25 GPU. These attractive properties of the membrane will be used to predict its process performance by simulation.

This work is intended to demonstrate the advantage of membrane technology over PSA in upgrading bio-methane and simultaneously capturing CO₂ from the biogas stream. A two-stage membrane system with a dedicated pre-treatment process was designed. Some energy-efficiency measures have been considered in the process design in an effort to maximize the potential of the membrane stages for the overall process. Particular attention has also been given to include all aspects of capital and operating costs of the system to ensure a reliable evaluation of the process economics, including determination of optimum operating conditions. Techno-economic analysis will show that processing biogas into an energy resource is more profitable by implementing membrane technology.

2. Overview of membrane process designed

The overall membrane process designed is shown in Fig. 1. The pre-treatment shown in the top part removes impurities including H₂S, H₂O, VOCs, and siloxanes. In order to overcome the transport resistance across the adsorption beds in series, a pressure of ~1.4 bar (all pressures in this work are absolute) was applied to the stream with a gas blower. A high-efficiency iron-oxide adsorbent was selected for feed desulphurization, and the process chemistry involved is given below:

The sulfur-free feed is then admitted into the temperature-swing-adsorption (TSA) tower, where all the other gaseous impurities are removed by the packed adsorbents (i.e., activated alumina and activated carbon) [6]. The treated feed contains less than 150 ppm water vapor, 4 ppm H₂S, and ppb levels of VOCs and siloxanes, which are not corrosive to the gas compressor, piping and membranes of the subsequent system.

The membrane system consists of three Blocks. The membrane in Block 1 (A1) serves to generate a CO₂-enriched permeate, which is further enriched to 99.0 vol. % in Block 3 by the second-stage membrane (A3). It should be noted that the gas permeate leaving Block 1 is not compressed before entering the membrane in Block 3. An intermediate pressure is simply maintained there as a driving force for the second-stage permeation (A3). This feature is, in part, responsible for the higher process energy-efficiency as will be shown in Section 6.2.1.

Both Blocks 1 and 2 belong to the first-stage membrane operation, and the retentate pressures are controlled at the same level. The retentate of Block 1 is moderately enriched CH₄, which is further upgraded to 97.0 vol.% as pipeline methane in Block 2. Permeate from Block 2 and residue from Block 3 are compressed and recycled back to the feed stream. With recycling, both CH₄ losses and CO₂ emissions can be minimized. The CO₂ rich stream is specified to 99% purity, yielding a fixed methane loss of 0.67% and a CO₂ recovery of 95.5%. The permeate pressure of module A3 is fixed at atmospheric pressure. The 99% CO₂ permeate from module A3 could be recovered at higher than atmospheric pressures in order to reduce the recompression cost of CO₂ for the ease of transport to a sequestration point, but is not considered in this study. Recovery of CO₂ at pressures above ambient, while saving on the compressions costs for transport, would have a significant impact on the pressure ratio of the feed to permeate and

greatly reduce the recovery and increase the volume of the recycle loop and compressor costs at this point.

3. Process simulation

3.1. Process membrane performance

Four assumptions were made to facilitate the process simulation, which include 1) the transporting properties of the membrane is independent of the variation in gas compositions throughout the separation, 2) there is no pressure drop in the plug-flow feed stream, 3) no concentration polarization takes place in separation, and 4) no coupling transport exists between the two components of the biogas. Based on the equations of gas permeation, mass balance, and Hagen-Poiseuille [21], an Excel-based simulator has been developed using the finite element method and accounting for changing feed compositions as a function of position and recovery in the membrane module. The validity of the algorithm was tested with good agreement ($<0.5\%$) with the process mass balance and literature data.

3.2. Cost assessment specifications

For making a reliable economic analysis, various costs (the depreciated capital cost, operating cost, and emissions cost) should be taken into account. Since the non-product gas streams are recycled in the process design and the emission cost incurred in pre-treating ppm levels of gas impurities are not significant, consideration of capital and operating costs is adequate for this work. For convenience, processing cost is defined as the summation of the capital and operating costs.

Based on the flowchart in Fig. 1, all the significant cost items are listed in Table 1. Given a feed and a set of operating conditions (i.e., the temperature, feed flow-rate, upstream, intermediate and downstream pressures), the membrane area needed is computed with the simulator. The membrane cost, C_1 , (expressed in US\$/h) is calculated from the total membrane area ($A_1+A_2+A_3$), membrane price at 20 US\$/m² [22], and a typical membrane life-span of 5 years (25 years for all equipment and mechanical structures involved) [23]. It has also been noticed that higher membrane prices were used in some simulation work, where expensive membranes (e.g., ceramic membrane) were used. With the protection of a properly selected pre-treatment system, the estimated membrane lifespan of 5 years is generally believed to be reasonable.

The cost of module housings, C_2 , is based on the pressures of the first and second-stage separations, which are different. Due to the higher downstream pressure in the first-stage separation, the hollow fiber bundle (100 mm in diameter) can be made much longer than that used in the second-stage, without excessive pressure buildup in the hollow fiber lumen. The nominal membrane areas for the first and second-stage modules are 60 and 30 m², respectively. The prices for the two types of module housings are 1000, and 80 US\$/pc, respectively. The value-selection of these parameters is detailed in the membrane module design in Section 6.3.

An effective rule of thumb for estimating the compressor cost, C_3 , is: 500 US\$ for 1kW compression requirement [24]. The compression is treated as isentropic, with an energy efficiency of $\eta=80\%$ as C_7 [25]. The isentropic compression is assumed to be a 2-stage compression unless the compression ratio (p_2/p_1) is below 5. The cooling water consumption is estimated by the energy balance and the cost is C_8 (Table 1). The prices of electricity, and

cooling water are 0.05 US\$/kWh, and 0.03 US\$/ton, respectively. Empirically, the cost of maintenance and labor, C_9 , is estimated as 10% of the total capital cost.

As a good estimate and general practice [24], the minor capital cost items (C_4 , and C_5) and installation fee (C_6), as a whole, accounts for 60 % of the major capital cost (C_1 , C_2 , and C_3) of the system, namely:

$$\sum_{i=4}^6 C_i = 0.6 * \sum_{i=1}^3 C_i \quad (3)$$

Therefore the processing cost (PC) of the overall system is:

$$PC = 1.6 \sum_{i=1}^3 C_i + \sum_{i=7}^9 C_i + C_{10} \quad (4)$$

where, the three terms are the capital, operating, and pre-treatment cost (C_{10}), respectively.

4. Objective function and optimization

The pre-treatment cost C_{10} is not a variable affecting optimization of the membrane process. This item can thus be evaluated separately without affecting the optimization of operating conditions (i.e. the upstream and downstream pressures p_1 and p_2). An objective function $Y(p_1, p_2)$ can be defined as follows:

$$Y(p_1, p_2) = PC - C_{10} = 1.6 \sum_{i=1}^3 C_i + \sum_{i=7}^9 C_i \quad (5)$$

Obviously, this hourly-cost objective function is feed-flow-rate-dependent, normalizing this objective function for the energy of methane produced yields:

$$YY(p_1, p_2) = \frac{Y(p_1, p_2)}{F^{\text{methane}} y^{\text{methane}} / 27} (\text{US\$} / \text{MMBtu}) \quad (6)$$

where, F is the flowrate (Nm^3/h) of upgraded product methane, y is the volume fraction of methane, MMBtu (10^6 British Thermal unit) is an energy unit (1.055 GJ), which is an equivalence of the energy of 27 Nm^3 methane [2]. The optimum operating condition and minimal processing cost can thus be located by mapping the dependence of the objective function upon the variables p_1, p_2 in a wisely selected variable domain $\Omega(p_1, p_2)$. The constraint is that the methane and CO_2 purity levels be 97% and 99% respectively. In the recycling scheme, there are no methane losses except for that in the CO_2 stream.

5. Process economic analysis

Three indicators are employed to characterize the membrane process economics. The overall specific processing cost (SPC) is defined as follows:

$$\text{SPC} = \frac{PC}{F^{\text{methane}} y^{\text{methane}} / 27} (\text{US\$} / \text{MMBtu}) \quad (7)$$

The overall specific energy cost (SEC) as defined in Eqn (8) refers to the compression cost needed for producing 27 Nm^3 (1 MMBtu) of product methane.

$$\text{SEC} = \frac{C_7}{F^{\text{methane}} y^{\text{methane}} / 27} (\text{US\$} / \text{MMBtu}) \quad (8)$$

The ratio ($R(i/o)$) of input and output energies is defined in Eqn (9):

$$R(i/o) = \frac{(C_7 / 0.05) * 3.6}{(F^{\text{methane}} y^{\text{methane}} / 27) * 1.055 * 10^6} \quad (9)$$

The input energy refers to the compression work, while the output energy represents the energy of the upgraded methane. This index measures the usability of the waste biogas as an energy resource.

6. Results and discussion

6.1. *The impacts of recycling non-product streams and the importance of operating conditions selection*

The performance parameters of the PEI-BMI membrane are given in Table 2. A feed flow-rate of 200 Nm³/h is selected as a reference for simulation. The two-stage membrane processes with and without non-product stream recycling are illustrated in Fig.2. The impacts of recycling the non-product streams on CH₄ and CO₂ recoveries were demonstrated in a two-case study, and the results are summarized in Table 3.

The selected upstream and downstream pressures p_1 and p_2 for Case I are 20, and 4 bar, respectively. Due to the recycling the feed flow rate increases by 35% as shown at the bottom of the Table 3. Accordingly, the membrane area also increases 34.8 % from 2018 to 2721 m², suggesting that no significant change occurs to the feed composition with the incorporation of two non-product streams. According to Table 3, with the recycling, the CH₄ loss is drastically decreased from 24.9 to 0.7 %, and CO₂ recovery is also significantly increased from 64.7 to 92.2%, which is higher than the 90% recovery required by the United States Department of Energy (USDOE) [24].

The p_1 and p_2 considered in Case II is 30 and 3 bar, respectively. Compared to Case I without recycle, Case II sees a remarkable reduction in membrane area (A_1 and A_2 except A_3). This reduction results from both the increased upstream pressure p_1 and the enhanced pressure ratio

(p_1/p_2) from 5 to 10. It is well-known that a higher pressure ratio will improve the separation efficiency, for a constant selectivity, particularly at high recovery where the local concentration of the faster permeating species, CO_2 , is much lower compared to the inlet. As listed in Table 3, by increasing the p_1/p_2 ratio CH_4 losses decreased from 24.9 to 10.7%, and their CO_2 capture increases from 64.7 to 78.1%. In the case of a membrane system with recycling, a smaller membrane area (A_1 through A_3) is invariably required, and the recycle stream flow rate also decreased from 69.5 to 27.9 Nm^3/h . For accomplishing the same level of processing, Case II displayed much better performance than Case I, irrespective of recycling. This suggests that in addition to having a better process design, an appropriate selection of the operating conditions for the process is equally important.

6.2. Membrane process optimization

6.2.1. Membrane area and compression demands

A wide domain Ω (p_1 , p_2) of operating pressures as shown in Fig. 3 is selected to detect the optimum p_1 and p_2 . The upstream pressure p_1 ranges from 20 to 60 bar and p_2 from 2 to 18 bar. The pressure ratio (p_1/p_2) varies between 2.5 and 20. Higher downstream pressures resulting in a pressure ratio less than 2.5 are excluded due to their inability to deliver acceptable separation efficiency. Twenty-five pressure pairs (p_1 , p_2) in total were tested in the selected region Ω (p_1 , p_2).

The estimated membrane areas under the 25 operating conditions are shown in Fig. 4, where each of the 5 sections corresponds to a selected upstream pressure p_1 . The downstream pressure p_2 in the section decreases from left to right. Invariably, the total membrane area required decreases as p_1 increases. Within a given pressure p_1 , the total membrane initially decreases

dramatically with decreasing p_2 , achieves a minimum and then increases slightly as p_2 further decreases. With a high downstream pressure p_2 , the driving force for the second-stage membrane is greater, and a much smaller membrane area in A_3 is thus observed. For the first-stage membrane, the high downstream pressure p_2 , however, leads to a significantly lower pressure ratio and therefore a poor separating efficiency in the first-stage. Thus greater membrane area for both A_1 and A_2 was needed to achieve the same level of processing. As far as minimizing the total membrane area is concerned, the trade-off between (A_1+A_2) and A_3 should be optimized. It can be seen that No.7, 13, 20 and 24 in Fig. 4 represent the selections leading to smallest membrane area requirement for their respective sections with p_1 at 30, 40, 50 and 60 bar, respectively. According to Fig. 4, it appears that the optimal pressure ratio yielding the lowest membrane area was in the range of 8 to 10. As expected, by operating the membrane system at a higher upstream pressure the membrane area was reduced.

The compression energy demand corresponding to the 25 selected operating conditions (p_1 and p_2) is presented in Fig. 5. Each compression energy demand consists of two parts. One is the primary part required for compression of the feed ($200 \text{ Nm}^3/\text{h}$), and the other for compression of the recycled stream. Again, the merits of recycling of the non-product streams are justified here: provided the membrane system is operated under optimized operating conditions, no significant extra compression energy is required to recycle the valuable CH_4 , and CO_2 from the non-product streams, which would otherwise contribute to emissions and losses.

One important finding in Fig. 5 is that the compression power does not increase linearly with the feed pressure; the ratio of the cost to the driving force decreases at higher pressures, therefore it is beneficial to operate at a higher pressure. This consideration is particularly true for a two-stage membrane operation, where the pressure of the permeate from Block1 in the first-stage will

be sufficient for creating a driving force for the permeation of the second-stage (Block3). In this way, the recompression energy of the intermediate stream can be saved and a second compressor is not required.

6.2.2. Minimal processing cost $YY(p_1, p_2)$ and optimum operating conditions

The processing cost $YY(p_1, p_2)$ is plotted against the downstream pressure p_2 in Fig. 6, where the upstream pressure p_1 is treated as a parametric variable and a typical processing cost curve exhibits a minimum. When operating at a lower p_1 , e.g., 20 bar, a larger membrane area is required. The cost associated with this larger membrane area is not countered by increasing the feed pressure until p_1 reaches around 40 bar, above which the decrease in membrane area does not compensate for the increasing compression cost. Fig. 6 shows that only a well-balanced membrane system can bring the processing cost down to its minimal value.

To some degree, the right selection of p_2 may be even more important than that of p_1 . This is because p_2 , as the intermediate pressure of the 2-stage membrane process, impacts on the separation performance of both the first and second-stages. When $p_2 >^*p_2$ (optimal p_2 of the curves), the increased processing cost stems from the increased membrane area ($A_1 + A_2$), due to the smaller driving force and smaller first-stage pressure ratio, and certainly this small pressure ratio also incurs more compression demand for the increased recycle volume. When $p_2 <^*p_2$, the unduly increased membrane area in A_3 is responsible for the observed rise in the cost. Interestingly, it can be seen that with the increase of the upstream pressure from 20 to 60 bar, the processing cost becomes less sensitive to the variation of downstream pressure p_2 , as seen by the wider “trough” of each curve for p_1 . This suggests that as the membrane system is run at a higher

upstream pressure less stringent process control is needed to maintain the process near the most economical operating zone.

The minimal processing cost and the optimal downstream pressure p_2 in Fig. 6 were determined by a polynomial fitting of the processing cost. The minimal cost and associated p_2 values are plotted against the corresponding upstream pressure p_1 in Fig. 7. Again using a polynomial fitting, the minimal processing cost is obtained with the optimum operating conditions p_1 and p_2 , such that $YY(p_1, p_2)$: $p_1 = 43.5$ bar, $p_2 = 5.0$ bar, and $YY(p_1, p_2) = 0.67$ US\$/MMBtu. The optimal value for p varies almost linearly with the upstream pressure p_1 . However, the processing cost is relatively insensitive to p_1 when $p_1 > 40$ bar. As shown in Table 4, the individual contributions to the processing cost are: electricity with the biggest share at 67.2%, the total capital cost consisting of the membrane modules and compressor is only about 17.6% with installation, maintenance and cooling water comprising the balance. It is clear that opportunities for cost-reductions are mainly on operational aspects.

Fig. 8 shows the contours of the processing cost as a function of the upstream and downstream pressures, p_1 and p_2 . A wide range of upstream pressures, from 39 to 58 bar, produces only a small increment of 2% of the minimal processing cost. The same increase in processing is predicted for a downstream pressure, p_2 , ranging from 4.3 to 6.2 bar. Obviously, this wide tolerance of the cost to p_1 obviates the exact control of the intermediate pressure, and simultaneously ensures that the 2-stage membrane process can be operated near the minimum cost. In practice the intermediate pressure can be controlled by placing a permeate reservoir as a buffer between Blocks 1 and 3.

6.2.3. Impact of electricity price

According to a report of global energy statistics [26], the price of electricity varies considerably across the world. North America enjoys the lowest price (0.05US\$/kWh) while some European countries pay more than double and most Asian countries are in between. The impact of the electricity price on the processing cost is depicted in Fig. 9. Naturally, at a higher electricity price, a higher processing cost is observed. At the lowest electricity price, e.g., 0.04US\$/kWh, the membrane process favors a higher feed pressure to offset the membrane and module costs. The optimum p_1 is around 60 bar. As the electricity price increases, the optimum upstream pressure decreases to 30 bar at 0.10US\$/kWh. The optimum upstream and downstream pressures are plotted as a function of electricity price in Fig. 10. An interesting finding is that despite the changes of both p_1 and p_2 , the p_1/p_2 ratio almost remains constant within 8.5-8.7. This constant ratio offers a simple empirical method to estimate the downstream pressure p_2 given a upstream pressure p_1 .

6.3. Membrane modules design

It is well-known that rational membrane module design is important not only in achieving better membrane performance but also in saving compression energy by eliminating the excessive pressure buildup, which may exist in the bore of fine hollow fibers. A useful design criterion, $\Delta = 8\sqrt{\frac{2RTD_0\mu JL^2}{D_i^4 p_2}} < 0.4$, has been reported [27] which acts as a guideline for the design of industrial scale gas separation membrane modules. All the symbols in the inequality are specified in Table 5. By selecting proper values for each of the operating parameters (T , p_2) and module design parameters (D_o , D_i and L), which satisfy the inequality, less than 5% loss of the gas permeation driving force can be ensured.

Given a fibre packing density of 50%, a 100 mm diameter membrane module will contain 20,000 fibers. Using the design criterion, a higher downstream (permeate) pressure allows for a longer module length. This suggests that length of the modules used in B3, and B1 and B2 should be different. Based on the physical properties of the hollow fiber, and the downstream pressure p_2 as shown in Table 5, the calculated active lengths of the fibers are 1000 for the B3 module and 2000 mm for the B1 and B2 modules, which possess a nominal membrane area of 30 and 60 m², respectively. The major reason for the difference in the module housing prices is in the material of construction. Stainless steel is selected for the high pressure modules, B1 and B2, while the less costly poly (vinyl chloride) can be used for B3 which operates at a lower pressure.

6.4. *Techno-economic analysis*

The breakdown of the capital cost of the pre-treatment is shown in Fig. 11(a). The total capital cost is 73.8 kUS\$; nearly 50 % of which goes to the adsorption towers and adsorbents. This, to some degree, explains the large “footprint” of TSA as an intermittent separation technology. Since TSA needs periodic intensive heating for adsorbent regeneration, the cost of the heat exchanger was found to be greater than that of the gas blower. A surprising finding was that the membrane system costs only 25% more than that of the pre-treatment, Fig. 11(b). The nature of continuous operation and superior separation efficiency of the membrane system under the optimum operating condition is responsible for this result. The small “footprint” of the membrane system is to some degree reflected in the smaller percentage (38%) of the membrane module share in the total cost. The optimized membrane process operates at a high feed pressure, resulting in a compressor cost that coincidentally is comparable to the membrane cost. Fig. 12 gives an overview of the total capital investment. Again, due to the requirement of two sets of adsorption towers in the pretreatment process design, the cost of adsorption is comparable to the

investments for the membranes/modules and the compressor. Installation and piping typically represents the greatest cost, and the smallest fraction comprises the supporting facilities, e.g., heat exchanger, water separator and cooling water.

A summary of the techno-economic analysis for producing 97% methane at 99% recovery is given in Table 6. As confirmed by the three indicators (equations 7-9), membrane technology shows an excellent potential for processing waste biogas. The R(i/o) value of 0.031 means that for every unit of input energy, there is a return of 32.3 units. This corresponds to a specific energy cost of 0.45 US\$/MMBtu. The specific processing cost is 0.89 US\$/MMBtu, compared to the typical pipeline natural gas price of 8 US\$/MMBtu, and the higher incentive price for biomethane 12 US\$/MMBtu [27]. This analysis shows that the membrane based process compares favourably to the typical processing cost of 5-7 US\$/MMBtu by PSA [28, 29] where the CH₄ recovery is only 90 %.

The gross profit generated by the pipeline methane based on the natural gas price is about 309.5 kUS\$/yr, which could be increased up to 464.2 kUS\$/yr if assessed using an incentive biomethane price (12US\$/MMBtu). By including the operating and the yearly depreciation of the whole system, the net profit earned from the pipeline methane is 275.7 kUS\$/yr. The estimated payback time of a potential capital investment of 156.2 kUS\$ for treating 200Nm³/h biogas was 6.8 months.

7. Conclusions

A two-stage membrane process integrated with a TSA-based pre-treatment was designed for processing wastewater plant biogas to yield 97% methane purity with 0.67% losses and a 99%

purity CO₂ stream and 95.5 % recovery. CH₄ and CO₂ losses were minimized by recycling the non-product streams created in the membrane process.

In an optimized two stage membrane process, the first stage is composed of two module blocks and a single block in the second stage, the feed pressure for the first stage was 43.5 bar. The permeate of the first stage was retained at an optimized pressure of 5.0 bar as the feed pressure to the second stage. This configuration eliminated the need for a second compressor. Though the optimum operating conditions change with such factors as electricity price, the ratio of the upstream and downstream pressure for the first-stage membrane was found almost constant (8.5-8.7).

Based on the current membrane performance, membrane technology clearly outperforms PSA in terms of the specific processing cost at 0.89 US\$/MMBtu compared to a typical range of 5~7 US\$/MMBtu for PSA. The specific energy consumption of the overall process is 0.031 J/J of recovered methane, or 1.69 MJ/kg of methane, using 19.3 kg methane/MMBtu. Techno-economic analysis showed that for processing 200 Nm³/h biogas stream a capital investment of 156.2 kUS\$ was needed. With an annual net profit of 275.7 kUS\$ in processing this stream a payback time of the investment was 6.8 months.

8. Acknowledgements

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9. List of symbols

- 1 **A, membrane area (m^2)**
- 2 **B, block of membrane modules**
- 3 **C, hourly cost (US\$/h)**
- 4 **D, diameter of hollow fiber (mm)**
- 5 **c, heat capacity of water or gas (J/g.C)**
- 6 **F, gas flux (NM^3/h)**
- 7 **L, length of the hollow fiber (mm)**
- 8 **J, membrane permeance for CO_2 ($10^{-6} \text{ cm}^3/\text{cm}^2.\text{sec.cmHg}$)**
- 9 **p, gas pressure (bar)**
- 10 **PC, processing cost (US\$/h)**
- 11 **R, universal gas constant (J/mol.C)**
- 12 **R(i/o), the ratio of the input and output energy**
- 13 **T, temperature (K)**
- 14 **y, mole percentage of CO_2 in permeate (%)**
- 15 **YY, the objective function (US\$/h)**
- 16 **Δ , dimensionless quantity**
- 17 **μ , gas viscosity (Pas.sec)**

Ω , defined domain of the up-and down-stream pressures

η , the efficiency of isentropic compression

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Figure captions

Fig. 1. Flow-chart of a membrane process designed for upgrading biogas from wastewater plant digesters, with the adsorption and temperature-swing-adsorption as the pre-treatment.

Fig.2. The membrane system without (a) and with (b) the non-product streams recycling.

Fig. 3. The domain of the selected operating conditions considered for process optimization.

Fig.4. The membrane area needed for the processing under the 25 selected operating conditions.

Fig.5. the compression power needed for the processing under the 25 selected operating conditions.

Fig. 6. The plot of processing cost versus the upstream and downstream pressures

Fig. 7. The plot of minimal processing cost and optimal downstream pressure versus the optimal upstream pressure.

Fig. 8. The contours of processing cost for processing 200 Nm³/h of biogas feed stream.

Fig. 9. The effect of electricity price on the processing cost of the membrane system.

Fig. 10. The effect of electricity price on the selection of the optimum operating condition.

Fig. 11. The capital cost of the pre-treatment system (a) and the membrane system (b) for processing 200 Nm³/h of biogas feed stream.

Fig. 12. An overview of the total capital investment for processing 200 Nm³/h of biogas feed stream.

Table 1. Various items considered in the processing cost

Items	Formula
1) Membrane cost:	$C_1 = \sum_1^3 A_i * 20 * \frac{1}{5 * 365 * 24} (US\$/h)$
2) Module housing cost:	$C_2 = [(A_1 + A_2) / 60 * 1000 + A_3 / 30 * 80] * \frac{1}{25 * 365 * 24} (US\$/h)$
3) Compressors cost:	$C_3 = \frac{C_7}{0.05} * \frac{500}{25 * 365 * 24} (US\$/h)$
4) Instrumentation cost:	C_4 (defined collectively with C_5 and C_6 in the Eqn (3))
5) Piping and valves cost:	C_5 (same as the above)
6) Installation cost:	C_6 (same as the above)
7) Compression cost:	$C_7 = \frac{2k}{k-1} RT \left[\left(\frac{p_2}{p_1} \right)^{\frac{k-1}{2k}} - 1 \right] * \frac{V}{22.4 * 3600} * \frac{1}{\eta} * 0.05 (US\$/h)$
8) Cooling water cost:	$C_8 = \frac{c_{gas} \Delta t_{gas} w_{gas}}{c_{water} \Delta t_{water}} * 0.03 (US\$/h)$
9) Maintenance & labor cost:	$C_9 = 0.1 \sum_1^6 C_i (US\$/h)$

10) Pre-treatment system cost: C_{10} (assessed in the section of economic analysis)

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2 Table 2. Membrane properties and feed stream specifications

Membrane	Hollow fiber
Material	PEI-BMI ^a
CO ₂ permeance	25 GPU ^b
CO ₂ /CH ₄ selectivity	55
Operating temperature	303.2 K
Feed flowrate	200 Nm ³ /h
Feed composition	40 vol.% CO ₂ , 60 vol.% CH ₄
^a Polyetherimide-bismaleimide	
^b Gas permeation unit (10 ⁻⁶ cm ³ /cm ² .sec.cmHg)	

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1 Table 3. Case studies: the impacts of recycling the non-product streams

	Case I		Case II	
	No recycle	With recycle	No recycle	With recycle
Feed flowrate F1 (Nm ³ /h)	200	200	200	200
Upstream pressure p ₁ (bar)	20	20	30	30
Downstream pressure p ₂ (bar)	4	4	3	3
Pressure ratio (p ₁ /p ₂)	5	5	10	10
Membrane area A1 (m ²)	605	827	299	340
Membrane area A2 (m ²)	1015	566	144	167
Membrane area A3 (m ²)	398	1328	710	856
Total membrane area (m ²)	2018	2721	1153	1363
CH ₄ flowrate F2 (Nm ³ /h)	94.9	125.5	112.8	125.5
X2 (vol. fraction)	0.950	0.950	0.950	0.950
Non-product F2' (Nm ³ /h)	31.9	41.8	8.4	10.6
Y2' (vol. fraction)	0.588	0.604	0.499	0.512
CO ₂ flowrate F3 (Nm ³ /h)	52.3	74.5	63.1	74.5
Y3 (vol. fraction)	0.990	0.990	0.990	0.990
Non-product F3' (Nm ³ /h)	20.8	27.7	15.7	17.3
X3' (vol. fraction)	0.491	0.502	0.511	0.521

Recycle stream F4 (Nm ³ /h)	N/A	69.5		N/A	27.9
X4 (vol. fraction)	N/A	0.561		N/A	0.518
CH ₄ loss (%)	24.9	0.7		10.7	0.7
CO ₂ recovery (%)	64.7	92.2		78.1	92.2
Recycle/feed ratio	N/A	0.35		N/A	0.14

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1 Table 4. The item costs of the minimal processing cost.

Cost items	Hourly cost	YY(p ₁ ,p ₂)	Percentage
	(US\$/h)	(US\$/MMBtu)	(%)
Electricity	1.95	0.44	67.2
Membrane module	0.42	0.09	14.5
Compressor	0.09	0.02	3.1
Maintenance	0.08	0.02	2.8
Cooling water	0.06	0.01	2.1
Installation etc.	0.30	0.07	10.3

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1 Table 5. Parameters considered in membrane module design.

Properties		Units	Blocks
Downstream pressure p_2	1.01	bar	B3
	5.0	bar	B1, B2
Outside diameter (D_o)	500	μm	N/A
Inside diameter (D_i)	200	μm	N/A
CO ₂ permeance (J)	25	GPU	N/A
Temperature (T)	303.2	K	N/A
CO ₂ viscosity	0.0153	cp	N/A
Packing porosity	50	%	N/A
No. of fiber	20000		N/A
Bundle diameter	100	mm	N/A
Fiber length (L)	1123	mm	B3
	2511	mm	B1, B2
Active fiber length	1000	mm	B3
	2000	mm	B1, B2
Tube-sheet thickness	2x60	mm	B3
	2x100	mm	B1, B2
Nominal area	30	m ²	B3
	60	m ²	B1, B2
Module housing	PVC		B3
	Stainless steel		B1, B2
Housing price	80	US\$/pc	B3
	1000	US\$/pc	B1, B2

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1 Table 6. Techno-economic analysis report

Techno-economic analysis report				
Description	Sub-description	Amount	Unit	
Feed	Biogas: 60 vol.% CH ₄	200	Nm ³ /h	
Product stream	Pipeline CH ₄ (97 vol.%)	122.9	Nm ³ /h	
	CO ₂ : 99 vol.%	77.1	Nm ³ /h	
Index	SPC	0.89 ^b	US\$/MMBtu	
	SEC	0.45	US\$/MMBtu	
	R(i/o)	0.031		
Pipeline CH ₄ price	Natural gas	8	US\$/MMBtu	
	Upgraded from waste biogas	12	US\$/MMBtu	
Gross profit	Pipeline CH ₄	309.5	kUS\$/yr	
		464.2 ^a	kUS\$/yr	
Operating cost	Electricity	17.5	kUS\$/yr	
	Cooling	0.6	kUS\$/yr	
	Maintenance	0.9	kUS\$/yr	

Depreciation	Membrane module	7.3	kUS\$/yr
	Compressor	1.3	kUS\$/yr
	Instrumentation etc.	0.9	kUS\$/yr
	Pre-treatment system	5.3	kUS\$/yr
Net profit		275.7	kUS\$/yr
Total capital cost		156.2	kUS\$
Payback time		6.8	

^a Profit estimated using the incentive price.

^bThe energy consumption by the periodical regeneration of adsorbents in the pretreatment is assumed to be three times as much as the energy consumed by the blower, which compresses the feed stream from 1 to 1.4 bar.

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 10

(a)

(b)

Figure 11

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Total capital investment (156.2 kUS\$)

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4 Figure 12

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