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Research paper

Vehicle fuel from biogas with carbon membranes; a comparison between simulation predictions and actual field demonstration

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Abstract

The energy contents of biogas could be significantly enhanced by upgrading it to vehicle fuel quality. A pilot-scale separation plant based on carbon hollow fiber membranes for upgrading biogas to vehicle fuel quality was constructed and operated at the biogas plant, Glør IKS, Lillehammer Norway. Vehicle fuel quality according to Swedish legislation was successfully achieved in a single stage separation process. The raw biogas from anaerobic digestion of food waste contained 64 ± 3 mol% CH_4, 30–35 mol% CO_2 and less than one percent of N_2 and a minor amount of other impurities. The raw biogas was available at 1.03 bar with a maximum flow rate of 60 $Nm^3\ h^{-1}$. Pre-treatment of biogas was performed to remove bulk H_2O and H_2S contents up to the required limits in the vehicle fuel before entering to membrane system. The membrane separation plant was designed to process 60 $Nm^3\ h^{-1}$ of raw biogas at pressure up to 21 bar. The initial tests were, however, performed for the feed flow rate of 10 $Nm^3\ h^{-1}$ at 21 bar. The successful operation of the pilot plant separation was continuously run for 192 h (8 days). The CH_4 purity of 96% and maximum CH_4 recovery of 98% was reached in a short-term test of 5 h. The permeate stream contained over 20 mol% CH_4 which could be used for the heating application. Aspen Hysys® was integrated with ChemBrane (in-house developed membrane model) to run the simulations for estimation of membrane area and energy requirement of the pilot plant. Cost estimation was performed based on simulation data and later compared with actual field results.

Keywords: Biogas upgrading; Pilot-scale demonstration; Membrane separation: Process simulations

1. Introduction

Industrial development and increased human population have globally led to a rising demand for energy with a growth rate of about 2% per year [1,2]. Oil and gas are the major sources of energy today and the complexity of the fossil fuel energy market always involves the risk of energy crisis emerging for political or the other reasons, like in the mid-1970s, and then recently in 2015 [1–3]. Renewable energy sources like biogas, wind and solar are inexhaustible compared to fossil fuels which are decreasing continuously over time. Although the reserves of fossil fuels are still significant, the questions related to climate change will enforce a change of energy usage. Renewable technologies are making relatively fast progress and expected to increase significantly (30–80%) in 2100 [2,4].

Biogas is a valuable renewable energy source and forms naturally, e.g. under anaerobic conditions such as in small lakes or flooded fields, in the sediments of the sea floor, and in the stomachs of ruminants. It can be produced by microbial digestion of organic material (agricultural waste, manure, municipal waste, sewage, food waste, etc.) in the absence of

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oxygen [5,6]. The major components are methane (CH_4) and carbon dioxide (CO_2) with traces of H_2S, some other gases and vapors [7,8]. The most common applications of biogas are for heating, combined heat and power generation and usage as a vehicle fuel. Other applications which are studied or tested are injection into the natural gas grid and H_2 production for fuel cells.

A study on different utilizations of biogas reports that biogas upgrading to fuel quality gives the highest portion of exportable energy with a medium range (10%) energy demand [9]. In the aforesaid statement, upgrading was done with membrane process. Sweden is one of the countries that widely developed biogas as an energy source after the energy crisis in the 1970s, and today is the leading nation when it comes to biogas as vehicle fuel with projected yearly consumption of 1 TWh in 2020, in comparison with 100 GWh in 2002 [10,11]. A simplified configuration of sustainable energy production is shown in Fig. 1 [6].

Biogas is widespread throughout the world in abundance where there is farming, and people are living. To use biogas as vehicle fuel, it must be upgraded to certain required specifications. The corrosive components (water vapor and sulphur) present in biogas must be removed. The CO_2, which is one of the major components in the biogas, needs to be separated from the biogas because it dilutes/lowers the heating value of the gas. This results in reduced burning capacity which, in return, affects the performance of the engine [12].

Both CO_2 and H_2S present in raw biogas yield corrosive products in the presence of moisture (carbonic acid and sulfurous acid). For extracting the energy carrier (methane) from biogas, membrane separation constitutes one of the attractive separation technologies and is the focus of the current study. A pilot-scale membrane separation unit was installed at a biogas (from food waste) production field in Norway to obtain the fuel quality bio-methane according to the Swedish fuel quality standards [13,14].

The membrane is a perm selective barrier which separates two phases and restricts transport of various molecules in a selective manner. In the case of biogas upgrading, membrane separation is based on the difference in permeation rate of methane and carbon dioxide due to the difference in molecular size, shape, and interaction with membrane material. The process to produce biomethane should be inexpensive and simple to control. Commercially available membranes for CO_2/CH_4 separation are mostly polymeric membranes, and these membranes do not have high enough separation factor (selectivity) to achieve a high recovery and purity of CH_4 in a single stage [15]. The amount of energy required for biogas upgrading is a key factor when selecting a technology for this purpose [16,17]. In this work, carbon membranes were applied in the pilot plant as an economically possible separation solution. Most of the work reported on carbon membranes was done at laboratory scale, and very limited work has been reported for these membranes on real gas industrial exposure. Carbon membranes have shown promising separation properties at laboratory scale, and these membranes are stable at high pressure and temperature [18–25]. The novelty of this work is to assess regenerated cellulose-based carbon hollow fiber membranes with high CO_2 permeability and CO_2/CH_4 selectivity in pilot scale biogas upgrading application. Biogas upgrading process, with the reported carbon membranes, is an energy efficient process and high purity methane (vehicle fuel quality) with high recovery can be obtained in a single stage separation process.

Biogas upgrading process consists of two main stages: (1) pretreatment process to remove trace components (H_2O, H_2S) to meet the fuel standards and (2) the membrane process to separate CO_2. A detailed description of the biogas plant can be found in the references [16,17,23]. This article reports the carbon hollow fiber pilot-scale module design used for biogas upgrading to vehicle fuel quality and testing of these membranes at the biogas field, Glør IKS, in Norway.

Process simulation is used to operate the model/limitation of the system before a new system is built or altered. The model can be redesigned, experimented and optimized in a way which would be too expensive or impractical to do in the actual system itself. Aspen HYSYS®, a process modeling tool, was integrated with ChemBrane (in-house membrane model described elsewhere [26,27]) to design the membrane separation process for biogas upgrading. Results obtained at the

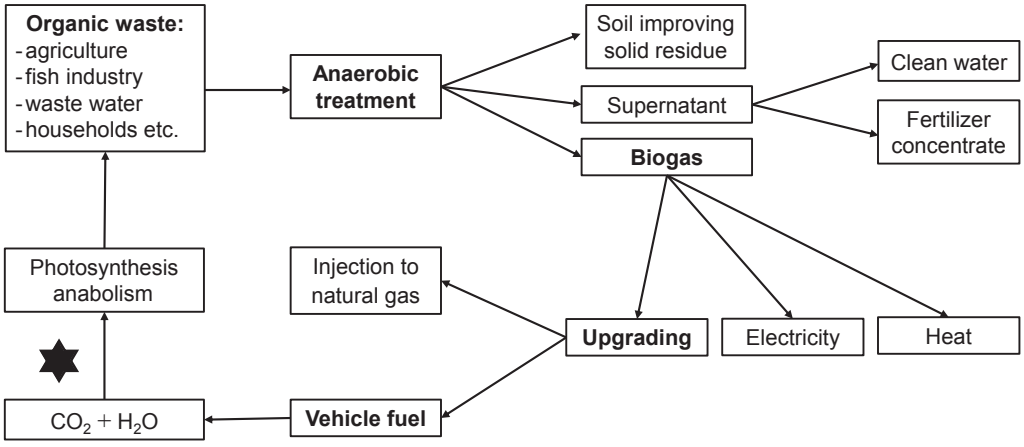


Fig. 1. Simplified configuration of sustainable energy production; turning waste into a resource [6].

membrane production facility were used in the model to optimize the process in the valuation of the required energy and membrane area. Total capital investment and production cost of the process were estimated based on supplier price quotations and simulation data. However, the price for each unit operation is presented as percent of total capital investment and production cost here. This work sums up the membrane module design, simulation predictions, and then, the actual field results in terms of membrane performance and total capital investment/production cost of the process.

2. Experimental

2.1. Carbon hollow fibers (CHF) preparation

The precursor for CHF was prepared using regenerated cellulose acetate (CA) by the dry/wet phase inversion process in a pilot-scale spinning set up delivered by Philos Korea. A dope consisting of CA mixed with N-methylpyrrolidone (NMP) and polyvinylpyrrolidone (PVP) was used to spin CA hollow fibers. CA hollow fibers were deacetylated batch-wise with a mix solution of NaOH in short chain alcohol. Then the deacetylated dried CA hollow fibers were carbonized at 550 °C under N_2 flow in a tubular 3-zone furnace. The carbonization protocol had a heating rate of 1 °C min^{-1} with several dwells and the final temperature of 550 °C for 2 h. Process details were described elsewhere [22].

2.2. Biogas composition and vehicle fuel quality

The raw biogas feed originates from microbial anaerobic digestion of food waste. Raw biogas composition is shown in Table 1. Untreated biogas was fed to a biological H_2S scrubber, and a slip stream of the treated biogas was fed to the membrane pilot plant.

For the biogas to be used as vehicle fuel, it must meet certain quality requirements/standards. Norway does not have its own fuel quality legislation yet, therefore, Swedish standards were used to acquire vehicle fuel with carbon membrane separation process as both countries have an alike climate. The requirements for clean biogas used as vehicle fuel according to Swedish standards is shown in Table 2. According to the legislation, an odorant must be added into flammable gas to ensure that the gas can be smelled below 20% of the lower explosion limit. Tetrahydrothiophene was used as an odorant to the upgraded biogas in this study.

Table 1
Composition of raw biogas obtained from anaerobic digestion of food waste.

Component	Food waste (mole%)
Methane (CH_4)	64 ± 3%
Carbon dioxide (CO_2)	30–35%
Nitrogen (N_2)	< 1%
Oxygen (O_2)	ca. 0%
Hydrogen sulfide (H_2S)	1000 ppm
Water (H_2O), 35 °C	ca. 32 g Nm^{-3}

Table 2
The requirement for vehicle fuel quality; Swedish legislation [13,14].

Components	Standard
CH_4 (Vol%)	96–98
H_2O (mg Nm^{-3})	< 32
Dew point (°C)	−60 °C at 250 bar (g)
$CO_2 + O_2 + N_2$ (Vol%)	< 4
O_2 (Vol%)	< 1
H_2S (ppm)	< 23

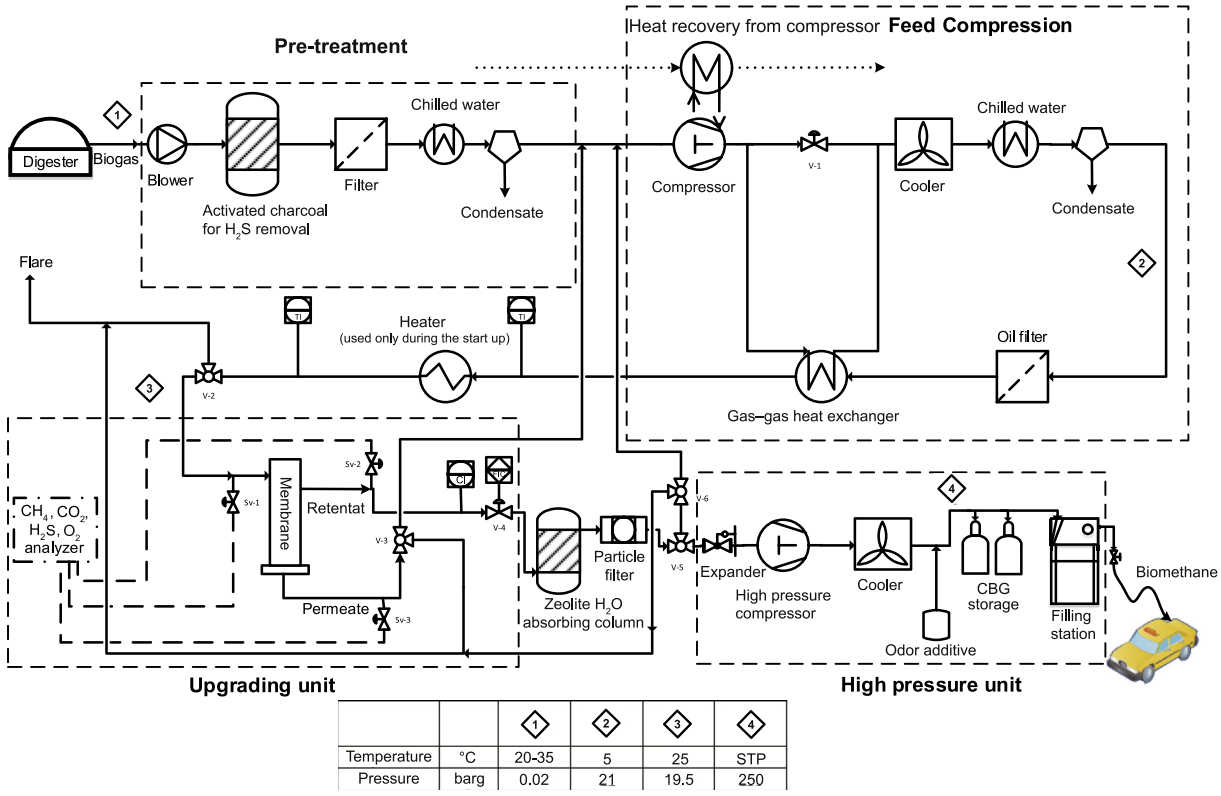
2.3. The carbon membrane pilot plant

The high performance of carbon membranes measured, at laboratory scale, indicated that CO_2–CH_4 separation process may have a very high recovery of methane (> 98%) and high CH_4 content (96%) in a single stage process. The carbon membrane production cost is higher compared to polymeric membrane technologies, as the production process is not yet fully optimized at commercial scale. But the high recovery results in a lower operational cost of the process which compensates to a certain extent for the cost of the membrane.

2.3.1. Biogas upgrading process

In principal, the raw biogas is compressed, bulk water is removed by means of a chiller (dew point: 4 °C at 1 bar), gas is reheated and then led through the membrane system. Carbon membranes are more selective for CO_2 relative to CH_4, therefore, in the biogas upgrading process CO_2 from the feed biogas passes through the membrane (low pressure side/permeate) and CH_4 remains on the high-pressure side (retentate). Hence, the retentate is the desired product. The ratio of permeate flow rate and feed flow rate is defined stage cut (q_p/q_f). Biomethane purity in the retentate stream depends on (1) CO_2/CH_4 selectivity, (2) pressure ratio on both sides of the membrane and (3) stage cut. Carbon hollow fiber membranes possess high CO_2/CH_4 selectivity and can be operated at high pressure, thus a sufficiently high-pressure ratio can be achieved if required. However, the necessity is determined by process design and economic considerations. H_2S and water need to be removed from the biogas stream prior to the membrane and this is done in pre-treatment section as shown in Fig. 2. Pre-treatment is a vital part of the process to meet the fuel standards and enhance the life time of the upgrading plant together with membranes.

The process flowsheet of the upgrading process with essential components is shown in Fig. 2, and operating conditions are shown in Table 3. The biogas upgrading pilot plant, containing carbon hollow fiber membrane, was operated to achieve fuel quality biomethane. The plant was designed to process 60 Nm^3 h^{-1} of raw biogas at pressure up to 21 bar. The initial tests reported here were performed for the feed flow rate of 10 Nm^3 h^{-1} at feed pressure 21 bar and vacuum on permeate side. The raw biogas was available at 1.03 bar and a blower was used to increase the pressure up to 1.3 bar. An activated charcoal system was used to remove most of the H_2S and bring it down to 5 ppm in the biogas stream. To ensure the



		1	2	3	4
Temperature	°C	20-35	5	25	STP
Pressure	barg	0.02	21	19.5	250

Fig. 2. Process flowsheet of biogas upgrading pilot plant based on carbon hollow fiber membrane.

Table 3
Operating conditions used in biogas upgrading pilot-plant.

Parameter	Value	Unit
Feed flow rate	10	$Nm^3\ h^{-1}$
Biogas pressure at blower inlet	1.03	bar
Biogas pressure at blower outlet	1.3	bar
Biogas pressure at compressor inlet	1.3	bar
Biogas pressure at compressor outlet	21	bar
Permeate pressure	0.1	bar
Temperature	20–25	°C
CH_4 in product	96	%
CH_4 recovery	98	%
H_2S in biogas feed	< 5	ppm

compressor safety, in form of scale formation or deposition of charcoal inside the compressor, a filter was present after H_2S removal system to remove the entrained particulates of activated charcoal. Then the water knockout through temperature swing (TS) at 4 °C and 1 bar was introduced just before the feed to the compressor to reduce the water level in the feed gas. The raw biogas was compressed to 21 bar with the mechanical reciprocating compressor (oil-free with 4-radial cylinders and external oil lubrication pump). Another chiller and an oil filter shown after compressor were part of the compression unit. They were not installed separately. In the case of carbon membranes, less than 40% relative humidity (RH) is satisfactory [21,28] as the performance of carbon membrane deteriorates at higher RH, so (partly)drying is needed. However, as a precaution, a heater was introduced just before membrane unit. The compressor oil, in case of any leakage from lubrication side, may deposit on the membrane surface and thus have a deleterious effect on the membrane performance, therefore several oil filters were used downstream to remove oil from the compressed feed gas. The compressed biogas then entered a cylindrical multi-module (shown in Fig. 3) containing 24 medium sized carbon hollow fiber modules (≈ 0.5–2 m^2 each). A single stage separation configuration was successfully tested to obtain 96% CH_4 and a significant amount of data was collected. The membrane feed gas temperature was regulated by an electric heater, and the pressure was controlled by a modulating valve (v-4 in Fig. 2, a globe valve with K vs. 2.5, supplied by Samson). The membrane pressure, temperature, flow of the

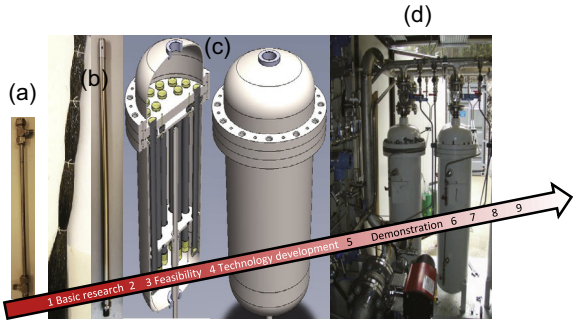


Fig. 3. Technology readiness level according to the EU commission/Up-Scaling from lab to pilot-scale; (a) lab scale module, (b) medium sized module, (c) Multimodule, (d) Membrane Pilot plant.

two outlet streams, permeate and retentate, were monitored with instruments as shown in Table 4. Online infrared analyzers (GD10P from Simtronics ASA) were used to monitor the composition of permeate stream only. However, another online gas analyzer (SSM 600C from Pronova) was available to measure composition of only one stream at a time, feed, retentate or permeate. A handheld gas analyzer GA 2000 from Geotechnical Instruments UK, was used at each site to have an estimation of the actual biogas composition. The analyzer can detect: Methane (0–100%), CO_2 (0–100%), O_2 (0–25%), H_2S (0–500 ppm). No more thorough analysis of trace compounds in the gases was performed. To accomplish the dew point: −60 at 250 barg in the final product, a zeolite-H_2O absorbing column was installed followed by a particle filter prior to the high-pressure compressor. The purpose of particle filter was to retain zeolite particles entering high pressure compressor. High-pressure compression up to 250 bar and odor addition was performed before storage of the vehicle fuel.

2.3.2. Multi-module system assemblage

A multi-module system (MMS) was comprised of up to 24 medium sized modules, of which, each module was made up of up to 2000 carbon hollow fibers, which were tested for strength in bundles with effective area ranging from 0.5 to 2 m^2. The outer diameter of the hollow fiber is in the range of 150–300 micron and a wall thickness of 30–50 micron. Feed is on the shell side of the module (outer membrane surface) and permeate flows internally (bore side) along the fibers. The assemblage, testing and performance of each medium sized module are reported elsewhere [25]. The MMS was designed in a way to accomplish maximum efficiency of the membranes. The structural strength, low fouling tendency, membrane replacement and ease to clean the MMS were important considerations for its application in a biogas upgrading plant. The MMS size was 0.324 m in diameter and 1 m in active length and consisting of three parts: (1) the vertical tank having both feed, retentate connecting ports and three legs with screws to secure it to the skid. (2) Middle part to insert the medium sized modules and, consisting of two round plates with holes according to the outer diameter of the medium sized modules. One partition plate on the top to separate the permeate section from feed section and 2nd partition plate between feed and retentate also helping to hold the modules firmly and avoid bumping into each other. (3) The lid on the top with permeate connection. The arrangement of the medium sized modules inside the MMS is shown in Fig. 3c. Photographs of the biogas upgrading plant are shown in Fig. 4. After the assembly and before fitting the lid, each of the medium sized module was tested again for any leakage (fiber breakage) using air pressure and soap water.

Fig. 4. Photographs showing the biogas upgrading membrane plant.

The MMS were pressurized and filled with gas by adjusting the feed, retentate and permeate valves manually in the initial stage. The valves were adjusted to fill gas in such a way that fibers achieve gentle treatment on the surface and there is no excessive pressure difference between feed and retentate across the partition plate inside the MMS.

3. Simulations and cost estimations

3.1. Simulation basis

Based on laboratory results, a single stage membrane configuration without recycle stream was examined and optimized by computer simulations before the execution of the pilot plant operation. The process configuration in simulation software “Aspen HYSYS” is illustrated in Fig. 5.

Then following basis and assumptions were used to simulate the membrane process performance:

Table 4
List of main instruments and measuring range.

Instrument	Model	Supplier	Measuring range	Accuracy
Dedicated product gas analyzer	GD 10P	Simtronics ASA	CO_2: 0–100%	± 3%
Handheld gas analyzer	GA 2000	Geothecnical instruments	CH_4: 0–100%	± 3%
			CO_2: 0–100%	± 3%
			O_2: 0–25%	± 1%
			H_2S: 0–500 ppm	± 5%
Online gas analyzer	SSM 6000C	Pronova	CH_4: 0–100%	± 2%
			CO_2: 0–100%	± 2%
			O_2: 0–25%	± 2%
			H_2S: 0–5 ppm	± 5%
			H_2: 0–1000 ppm	± 5%
Temperature transmitter	TT-Classe A	Officina Orobiche	−30–+350 °C	± 0.15 °C
Dew point transmitter	DP-001	Michell Instruments	−100–+20 °C	± 1 °C
Pressure transmitter	3051S	Emerson	1–275 bara	0.025% of span

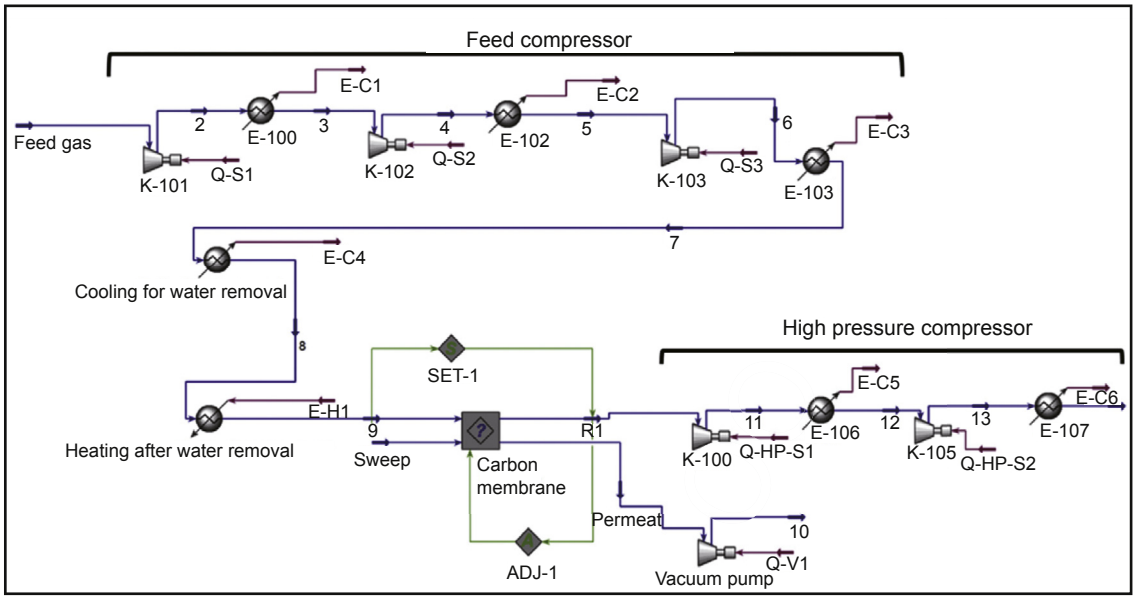


Fig. 5. Single stage process for biogas upgrading with carbon membranes.

Table 5
Process operating conditions used in the simulations.

Feed composition	35–40% CO_2, balance CH_4
Feed flow rate ($Nm^3\ h^{-1}$)	60
CO_2 permeability (Barrer[a])	300
CO_2/CH_4 selectivity	100
CH_4 purity in product (%)	96
Feed pressure (bar)	21
Permeate pressure (bar)	0.1
Temperature (°C)	25
Flow pattern in membrane module	Countercurrent

[a] 1 Barrer = 2.736E-09 (m^3(STP).m)/(m^2.bar.h).

- Countercurrent gas flow pattern without sweep on permeate side was used in all hollow fiber membrane modules.
- In-house made membrane simulation model (Chembrane) was integrated into 6 V Aspen Hysys®. This model, developed at NTNU, uses fourth-order Runge-Kutta method to calculate the flux along membrane length, and then iteration over permeate values to converge to a solution.
- CO_2 composition (40–45%) balance with CH_4, was considered in the feed biogas stream entering the membrane system. Process conditions used in simulations are shown in Table 5. Permeabilities and selectivities used in simulations are also shown in the same table.
- The adiabatic efficiency of the compressors was modeled as 75%.

3.2. Economic parameters

The economic calculations may differ considerably, as they are justified by the data available and cost model. An economic evaluation was performed to assess the total capital investment and production cost of the biogas upgrading plant. A single stage biogas upgrading process with installed carbon

Table 6
General assumptions for economic calculations [23].

	Values	Units
Energy prices		
Electricity	0.06	€ kWh^{-1}
Vehicle gas	0.33	€ Nm^{-3}
Methane content	60	%
Financial assumptions		
Membrane cost	161	€ m^{-2}
Installed compressor cost (CC)[a]		€ 7100 × $(HP)^{0.82}$
High pressure compressor cost (CBGC)[a]		$C_{comp,ins} = 912.\ (W_{comp})^{0.9315}.\ f_m.\ f_i.\ f_{inst}$ [29]
Internal rate of return (IRR)	5	%
Depreciation	15	Yrs
Operating percentage	96	%
Total hours in operation	8409.6	h/yr
Normal supervision	416	h/yr
Membrane life time	5	Yrs

[a] Cooling system was included in the compression unit.

membranes with given performance, cannot recover all the CH_4 coming from the biogas. Consequently, this CH_4 will be lost in the permeate stream. Therefore, the cost of the lost CH_4 was not included in the cost of upgrading process. General assumptions used to evaluate the economics of the upgrading unit are presented in Table 6. A detailed description of the economic analysis and net present value are reported elsewhere [23].

4. Results and discussion

4.1. Multimodule membrane system

The first trial was run relatively quickly, using one MMS comprising medium sized membrane modules of low permeance, and a feed flow rate of 4 $Nm^3\ h^{-1}$ was applied. The CO_2/CH_4 selectivity obtained in this run was quite low, and high permeance was recorded compared to the values estimated from individual module testing and MMS results at the production facility. Two reasons were considered: firstly, the trial was run for too short time, and it is unlikely that the permeate stabilizes so quickly, therefore, relatively low selectivity was obtained in the beginning. Stabilizing the permeate concentration for the low permeance modules may take days, due to long residence time on the permeate side of the MMS. Secondly, due to fiber breakage as carbon hollow fibers being self-supported hold relatively poor mechanical stability.

The plant was stopped, and MMS was opened to check the fiber breakage. Each medium-sized module was tested using air pressure and soap water to find the leakage in the modules as shown in Fig. 6. Many broken fibers were found which ultimately were manually clogged using epoxy “Loctite 3090” and the procedure in detail is reported somewhere else [25]. It was considered that vibration from compressor could break the fibers as many of the broken fibers were found close to the support legs of MMS where vibration effect was at maximum. Therefore, the membrane skid was damped down to reduce the vibration amplitude defecting the brittle fibers and operation was started again.

Several measurements over a period of some days were made. One MMS with an estimated membrane area of 2.5 m^2 was tested for four days (cumulative operation time) to determine the membrane performance. Fig. 7 presents the results of this module with cumulative operation time. Feed pressure was gradually increased to obtain a required pressure of 21 bar. Depending on the composition of the raw biogas, H_2S was removed upstream of the feed gas compressor. All measurements were made at 21 bar feed pressure and vacuum on the permeate side. CH_4 contents in feed, retentate and permeate streams are shown graphically along with flow rates in Fig. 7. The concentration of CH_4 in retentate stream increased to 78 (maximum CO_2/CH_4 selectivity of 7) in the beginning but suddenly started decreasing and the flow of the product stream (retentate) reduced. A very high permeate flow with no CO_2/CH_4 selectivity was measured which indicated fiber breakage in the module. The broken fibers were clogged, and the operation was started up again. This time membranes were showing some selectivity (during 50–65 h plant operation time), but the value was very low as compared to the laboratory results. After few hours, some fibers broke again and the same composition as feed was detected on permeate stream, hence, the plant operation was stopped once more. Hence the first three operations were not successful with respect to achieve high selectivity due to the fiber breakage problems as shown in Fig. 9.

These initial problems of fiber breakage were solved, and in the fourth test the pilot plant was run for eight days at stable conditions and measurements were done periodically both by an online infrared analyzer and a portable analyzer. The plant was working as expected by giving required vehicle fuel quality as shown in Fig. 8. The results in Fig. 8 show that CH_4 concentration in the feed, retentate and permeate streams and flow rate of each stream was almost constant during the cumulative test period of 192 h. The concentration of CH_4 in the product stream was 96 mol% (CH_4 loss: 2–4%) throughout

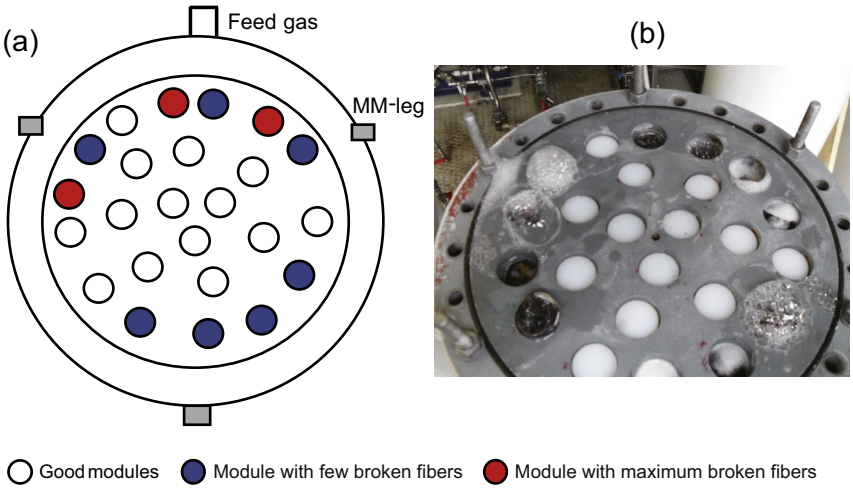


Fig. 6. (a) Arrangement of small modules into a big multi-module (MMS) (b) leakage testing and manual clogging of each module inside MMS.

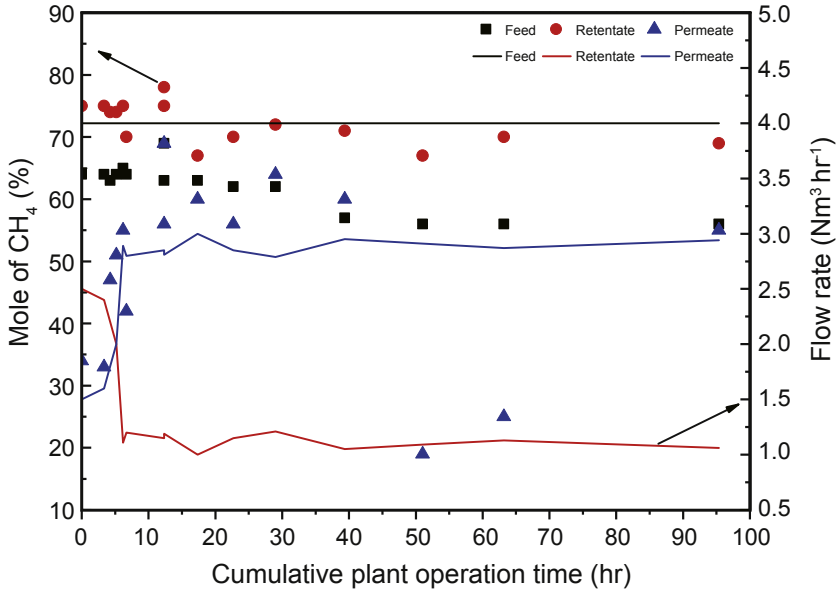


Fig. 7. Results of one MMS, tested for feed flow: 4 $Nm^3\ h^{-1}$.

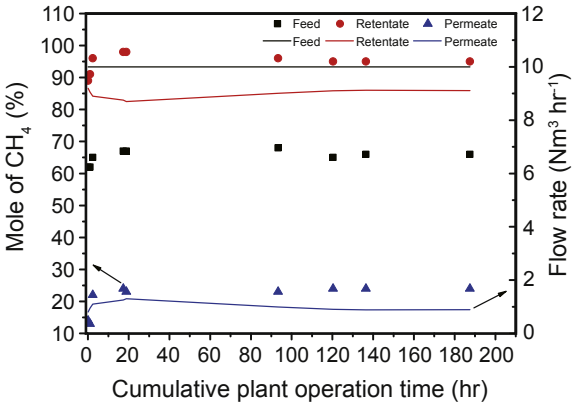


Fig. 8. Carbon membrane separation process for biogas upgrading; Flow rates as "solid lines" and CH_4 contents as "dots" in the graph.

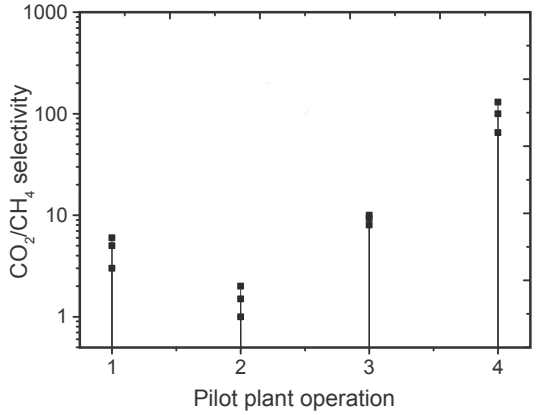


Fig. 9. CO_2/CH_4 selectivity during different operational runs of pilot plant. For the three first runs there were some fiber breakage, hence low performance resulted.

this time, and maximum selectivity for CO_2/CH_4 was measured 130 as shown in Fig. 9. The Robeson plot shows the trade-off between permeability and selectivity for gas pairs through a membrane. For the gas pair CO_2 and CH_4, it shows clearly that both high purity and high recovery cannot be attained in a single stage with a polymeric membrane [30]. Therefore, a two-stage system with recycle may usually be needed to achieve high purity and recovery of the product. But a high-performance membrane (showing both high CO_2 permeability and CO_2/CH_4 selectivity), may achieve both high purity and recovery of the product in a single stage process with optimized process conditions. Thus, the feed pressure of 21 bar (against the 0.1 bar in permeate) the required methane purity (96%) and recovery (98%) of the product was achieved in a single stage process (estimated through simulations before installation). Fig. 9 presents the CO_2/CH_4 selectivity achieved during the different set of operations. As already mentioned, the first three operations were not successful and very low selectivity was achieved due to the fiber breakage problem. This was however resolved and the plant was working as expected by giving required quality vehicle fuel (plant operation 4 in Fig. 9). The effective membrane area was significantly reduced due to the manual clogging process of broken fibers. The modules with a high number of damaged fibers were later replaced by the good performing modules. After installation of the good modules, the membrane area that was lost due to clogging was only about 1–2% of the total membrane area.

4.2. Installed energy and cost of the plant

The energy values for the biogas upgrading unit were assessed for 60 $Nm^3\ h^{-1}$ plant capacity and values for the compressed natural gas unit (CNG) were considered for 40 $Nm^3\ h^{-1}$. Cost assessment is very important before the implementation of the plan. Normally the membrane cost for

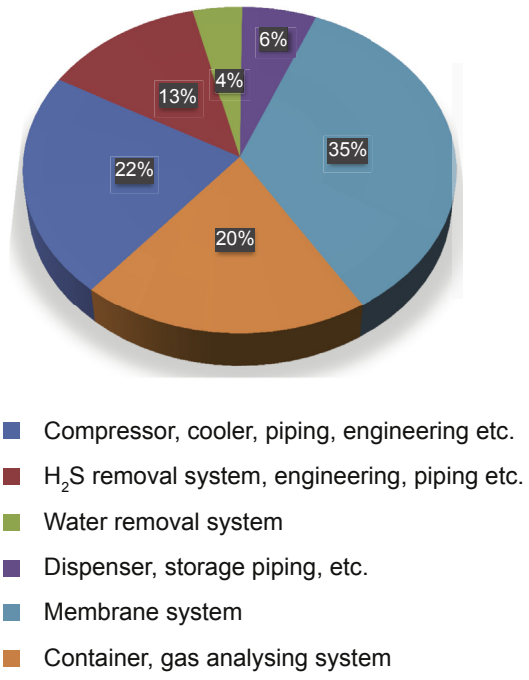


Fig. 10. Contribution of each unit in the biogas upgrading pilot plant.

polymer membrane modules is about 10–25% of the total cost [23], but the scenario is quite different in case of carbon hollow fibers. The carbon membrane production process is not optimized for commercial scale, and continuous process is not yet developed to produce the hollow fibers and to construct the modules. Therefore, a batch process was used on pilot-scale production plant which adds up the cost in terms of material usage, energy consumption, man power and working hours. Hence, the estimated membrane cost based on "small scale membrane preparation" data was 161 € m^{-2}, contributing about 35% of the total capital investment as shown in Fig. 10. Moreover, the economic assessment depends on the method of analysis and assumptions used to evaluate the final results.

Fig. 10 presents the cost of both upgrading unit and CNG package. Membrane price was assumed based on lab-scale production price, however, other costs mentioned in Fig. 10 were corrected in this paper according to the price quotations obtained from suppliers. The membranes represent the largest capital cost, while the second largest capital investment was of compression unit which made 22% of the total cost. H_2S removal with charcoal was also very costly (13% of the total cost), thus, a biological H_2S removal system is recommended for future studies. By applying these assumptions and available information, a projected net present value (NPV) of 765,189 € was estimated. A detailed description of cost analysis and NPV calculations were reported elsewhere [23]. The total capital investment for a 60 Nm^3 h^{-1} biogas upgrading pilot plant was 297,897 € and total operation & maintenance cost (production/running cost) was predicted to be 5532 € per year. Fig. 10 only presents the contribution of each unit as a percentage value of the total capital investment. The price for high pressure compressor was also added in total capital investment. However, the specific energy of high pressure compressor was 0.13 kWh/(Nm^3 of upgraded biogas) which was not included in running cost in order to make comparison with other studies. Carbon membranes used in current study possessed high performance (selectivity and permeability), therefore, very small area (10 m^2) and low specific energy was required to produce high quality vehicle fuel with high recovery of methane in a single stage separation process. Running cost of the carbon membrane based pilot plant was thus estimated 0.014 €/(Nm^3 of upgraded biogas), which is much lower than 0.05 € Nm^{-3}, the values computed by Deng et al. [15]. They reported an experimental analysis of biogas upgrading process based on CO_2 facilitated transport membranes. The two-stage membrane process with permeate recycle was proven optimal and specific energy consumption of 0.29 kWh/(Nm^3 of upgraded biogas) was estimated. Makaruk et al. [16] investigated different membrane systems for biogas upgrading process and reported specific energy consumption of 0.3 kWh/(Nm^3 of upgraded biogas), whereas Valenti et al. [17] have simulated the optimal value of specific energy from 0.33 to 0.47 kWh Nm^{-3} (depending on the layout). No vacuum pump was used on permeation side in any of the above-mentioned studies. However, biogas upgrading with carbon membranes proved that a single stage membrane system with no recycle stream, can produce vehicle fuel with specific energy consumption of 0.28 kWh Nm^{-3}. Although a vacuum pump was used (vacuum energy is also added in total specific energy usage) on permeate side for carbon membrane system, yet the total energy consumption is still lower than in all the above-mentioned studies. The energy consumption for high pressure compression was not included in any of above-mentioned studies.

4.3. Simulations and field results

To make an economically viable membrane separation process, both high permeability and high efficiency (selectivity) are needed. Carbon membranes reported here showed superior separation performance on laboratory scale experiments compared to polymeric membranes. Hence, a pilot-scale system was simulated based on the experimental results at production facility. The performance of the membranes was almost similar or even higher in some operations for biogas upgrading as compared to simulated values. The total capital investment was quite close to the projected values based on simulations. The membrane cost was considered 80 € m^{-2} in the simulations based on knowledge from pilot-scale production at MemfoACT AS. However, the brittleness of hollow fibers remained a challenge and the total cost of the membrane was almost doubled when required membrane area was in operation at the biogas facility. Hence, the total capital investment and production cost increased because of that extra membrane area. The energy consumption by the compressor and vacuum pump, product (methane) purity and methane recovery were very much comparable with the simulated results.

4.4. Challenges and suggestions

Although the carbon membrane pilot plant successfully obtained the vehicle fuel, there are still challenges that need consideration. The manually sorted and randomly packed hollow fibers of carbon membranes had smaller mass transfer-coefficients than those for regular dense packings (polymeric hollow fibers). Flow through the randomly packed hollow fiber bundle could be highly nonuniform. Membrane effective area was very much reduced due to selective and manual clogging. Furthermore, regions, where fibers come in close contact, may create sections of high pressure drop. The gas velocity through these regions is much lower than the velocities in the regions where fiber spacing is larger, yielding higher mass transfer coefficient in these regions. On the other hand, in high velocity regions, there are increased chances of fiber breakage if any weak point occurs on the fiber surface. It may result in flow-channels formation and hence, bypassing effect which would result in selectivity loss. The MMS design for 24 medium size modules was not most efficient in this development phase of the operation. It could have been easier with individual module housing instead of MMS housing in order to isolate and treat the modules with bad performance separately. The process of dismantling the MMS to take out the medium sized module, finding and clogging of the broken fibers, and again assembling the MMS increased the probability of fiber breakage in neighboring modules inside MMS and the entire process was time consuming as well. The shell-side feed configuration might have damaged the fibers due to high pressure feed flow. Bore-side feed configuration might have been more efficient in the MMS system. The membrane production cost at semi-industrial production plant was about 80 € m^{-2}, but due to a decrease in membrane effective area, the membrane cost doubled for the biogas pilot plant, which ultimately increased the total capital investment and production cost of the plant. The mentioned problems must be solved before a successful hollow fiber membrane module sees the market.

5. Conclusions

A multimodule system containing 24 medium sized modules, was successfully installed and operated at 21 bar feed pressure to obtain vehicle fuel from biogas. The carbon hollow fiber membranes achieved 97 mol% CH_4 with 98% CH_4 recovery in a single stage process. Pretreatment of biogas was performed prior to membrane separation to meet the fuel quality standards according to the Swedish legislation. The pretreatment consisted in removing H_2S with charcoal bed and H_2O removal by temperature swing and zeolite absorption. The plant operation was run successfully for 8 days and membranes used in this study yielded consistent results. It was observed that shell-side feed configuration was not very efficient in the MMS because the fibers could damage or break with high pressure feed flow. A bore-side feed configuration may give better results. Simulations were conducted to estimate area and energy requirement for the pilot plant. Total capital investment and production cost were estimated based on simulated data. The membrane cost was considered 80 € m^{-2} in the simulations based on pilot-scale production at MemfoACT AS. However, the brittleness of hollow fibers remained a challenge and the total cost of the membrane was almost doubled when required membrane area was installed and in operation at biogas facility. Hence, the total capital investment and production cost increased only because of that extra membrane area needed. However, the high recovery resulted in a lower operational cost of the process which compensated to a certain extent for the cost of the membranes. The running cost was much lower (0.014 € Nm^{-3} of biogas upgraded) than the polymeric membranes (0.05 € Nm^{-3}) reported in literature. The energy consumption by the compressor and vacuum pump, product (methane) purity, and methane were very much comparable with the simulated results. As far as carbon membrane-based biogas upgrading is concerned, the future membrane development should focus on improved mechanical properties of the membrane fibers and bore-side feed configuration should possibly be applied.

Conflict of interest

There is no conflict of interest.

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