

Biogas upgrading using micro-porous hollow fiber membrane contactors

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Abstract

In this paper, the behavior of micro-porous hollow fiber membrane contactors has been studied for biogas upgrading under different operational conditions. Physical solvents, as deionized water and sodium chloride solution, were initially used. In these cases, carbon dioxide absorption was dependent upon liquid phase flow. Sodium hydroxide was subsequently used as a comparative chemical absorption solvent. In this case, CO₂ mass transfer increased by increasing gas velocity which was attributed to the excess of reactive hydroxide ions present in the solvent. Under optimum conditions, with two membrane contactors operating in series, it was possible to obtain a gas stream with more than 99% of pure methane using deionized water as solvent. The same yields were obtained with NaOH as solvent, but in this case working with just one membrane module, that is, reducing by half the available surface area for molecules diffusion. At constant liquid to gas flow ratios, better separation behavior was targeted at pressures higher than 2.0 barg. When dealing with solvent reuse without regeneration, NaOH is the only adsorbent allowing a high number of solvent recirculation cycles. This aspect could be very interesting for the economy of the process.

Keywords

Biogas, biomethane, gas-liquid absorption, membrane contactors, biogas upgrading

Introduction

Biogas production is growing due to the increasing installation of anaerobic digesters for organic waste treatment, mainly in Europe, promoted by the current environmental legislation. 17,700 biogas plants operated in 2018, only in the EU (EBA, 2018). The demand for biogas, and other renewable fuels, is increasing all over the world with growing concern about environmental problems related to the high greenhouse gases (GHGs) emission from fossil fuel combustion (Pelletier et al.; Yann et al., 2017). In the last years, there is also an increasing demand for methane-rich biogas (biomethane) since the use of this product as vehicle fuel or its injection to the natural gas grid are becoming common practices (Ferreira et al., 2019). Furthermore, and according to previous studies (Adelt et al., 2011), the specific GHG emissions associated with the production of biomethane amount to 44.6 g CO₂eq/kWh, corresponding to an overall GHG emission reduction of 82% compared with natural gas. These figures convert biomethane in a key option on the way to sustainable and renewable energy supplies.

To produce biomethane, the biogas has to be first cleaned to remove the trace elements and then upgraded to remove CO₂ and adjust the calorific value to the foreseen uses. By separating CO₂ from biogas, the calorific value of the gas stream increases and both, the volume of the transported gas and its corrosive tendency decrease (Siagian et al., 2019). Biogas upgrading is thus a key issue today to enrich biomethane to produce a natural gas substitute. Nowadays, there are more near 500 biogas upgrading plants only in Europe (EBA, 2018) and new plants are continually being built around the world due to the increasing demand on biomethane. There will soon be more than 1,000 biomethane production plants operating in thirty-four countries (Cornot-Gandolphe, 2019).

Five technologies are mainly used in biogas upgrading plants, some of them commercially available and others at pilot or demonstration level: water absorption, pressure swing adsorption (PSA), chemical scrubbing, physical scrubbing and membranes separation. Water absorption has been traditionally the most popular upgrading technology with 40% share years ago, followed by PSA and chemical scrubbing with 20% share each (IEA, 2013). However, nowadays, membrane separation competes for the first place with water scrubbing. Use of membranes has increased to make this one of the market leading technologies in biogas upgrading since 2014/2015 (Fig. 1).

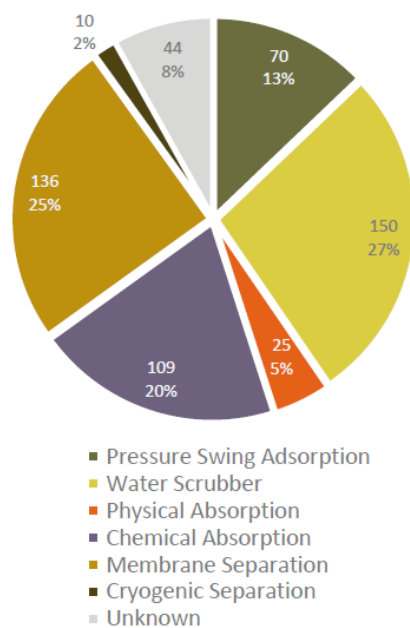


Fig. 1 Use of biogas upgrading technologies, Europe-wide (Source: Adapted from EBA, 2018)

In the case of CO₂ separation from biogas, the main advantages of membrane-based technologies are moderate energy requirement, low cost, flexible operation, and modularity (Siagian et al., 2019). More interestingly, the membrane-based systems are of high interest to be used due to the smaller footprint and lower weight compared to absorption processes (Gilassi et al.).

Gas-gas membrane-based separation systems has been widely investigated for CO₂/CH₄ separation from natural gas (Liu et al., 2018; Nikolaeva et al., 2018). This technology could also be applied for post-combustion capture (CO₂/N₂ or CO₂/N₂/O₂ separation from flue gases (Sekizkardes et al., 2018)) and pre-combustion capture CO₂/H₂ separation (He et al., 2017). In all the cases, a dense membrane is used as selective barrier between the feed gas and the permeate gas streams. For biogas upgrading, CO₂ permeates through the membrane while CH₄ retains on the inlet side as retentate. Most of the biogas upgrading plants operating today with membrane technology are gas-gas modality (Cornot-Gandolphe, 2019).

Gas-liquid membrane-based absorption systems, also called membrane contactors, have been traditionally used in the food industry for liquid gasification or degasification processes but today this technology also offers promising perspectives in biogas upgrading at low pressure (Belaissaoui et al., 2016; Fougerit et al., 2019; McLeod et al., 2013;

Ryckebosch et al., 2011) which combines membrane with conventional phase contacting operation such as absorption, and hence the benefits of both technologies can be fully utilized (Belaissaoui and Favre, 2018; Siagian et al., 2019.). The utilization of a membrane contactor as a mass transfer equipment for the CH₄ recovery from biogas has been described in detail by Rongwong et al. (2017). Hydrophobic membranes separate the gas from the liquid phase. Hollow fiber membrane modules are preferentially used since they give high specific surface area (He et al., 2018; Siagian et al., 2019; Tantikhajongosol et al., 2018; Zak et al., 2018). Specific molecules from a gas stream diffuse through the membrane and are absorbed on the other side by the solvent flowing in counter current. The solvent must be selected carefully in order to achieve a high separation performance minimizing operational complexity. Generally, the solvent should have a high absorption capacity for CO₂ and should also be compatible with the type of membrane used. It is highly important to consider solvent-membrane compatibility since the solvent may react with the membrane, which could result in membrane degradation (Foulong et al., 2018). Organic solvents, such as amine solution, are the most popular for CO₂ removal applications in membrane contactors. This solvent provides a high selectivity (Capra et al., 2018) but the use of this type of organic solvent also has several drawbacks: amine solution is corrosive, has high vapor pressure, and needs high energy for regeneration (Babamohammadi et al., 2015). This is why new types of absorbents are attracting researchers' interest to apply them in membrane contactors for CO₂ capture looking for high CO₂ solubility, high selectivity, low vapor pressure, and high thermal stability (Dai et al., 2016). Some examples for interesting proposals are deionized water (DI) (Belaissaoui et al., 2018), ionic liquids (Bazhenovet al., 2018), or even slurry (He et al., 2018).

Unlike gas-gas membrane systems, membrane contactors have a very high selectivity so that it could be applied in a wide range of gas flow rate and concentration without any loss of product gas (Klaassen et al., 2008). The high selectivity is provided by the physical or chemical interaction between the gas stream and the solvent (absorbent). Furthermore, membrane contactors are usually operated under low pressure so the operational costs are lower than for gas-gas membrane systems that operate at high pressure (Siagian et al., 2019). Also, recent studies show that the membrane upgrading technology is slightly more environmentally convenient than all the other upgrading technologies (Florio et al., 2019). But although gas-gas membrane systems have been already commercialized and applied on a large scale, particularly for natural gas sweetening but also for biogas upgrading, gas-liquid membrane systems for these uses are just now emerging and only some pilot and demo plants are currently running (Cornot-Gandolphe 2019). However, all the interesting features of membrane contactors are attracting the increasing attention from researchers for applying this process in CO₂ removal, especially when biogas production units do not reach a scale large enough to support conventional technologies capital expenditures (Fougerit et al., 2019).

Since operating conditions can significantly affect the performance of membrane contactors, this should be a matter of deep study before a broad implementation of the technology. Temperature, pressure, and flow rates, both gas and solvent or liquid streams, should be carefully controlled to avoid gas losses and increase gas absorption. Following this line, this study seeks to find optimal operational conditions to maximize the separation of the two principal biogas components, methane and carbon dioxide, using membrane contactors. The influence of operational parameters such as mass exchange surface, operational pressure and liquid/gas ratio will be studied. This work also evaluates the impact of different solvents (deionized water, sodium hydroxide and sodium chloride) on methane recovery and the effect multiple solvent cycles compared to single-pass operation.

Materials and methods

A commercial bottle of synthetic biogas (methane and carbon dioxide gases, PR mixture 2CGC - 40%CO₂, 60% CH₄ v/v, X10S, Carburos Metálicos) was used to provide an initial gas feeding to the shell side of the hollow fiber membrane contactor (HFMC, Liqui-Cel® 1.7 x 5.5 MiniModule®, Membrana GmbH, Wuppertal, Germany) (Fig. 2). The inlet gas flow rate was controlled and measured using a flow meter (F-201CV Series, Bronkhorst Hi.Tec, Ruurlo, the Netherlands). A ball rotameter adapted for CH₄ (Tecfluid 2000 Series) was installed just before the venting of the gas to measure outlet gas flow. Liquid phase to be used as absorbent was stored in a 30 l PVC tank and maintained at 24-26 °C by room temperature control. The absorbent was passed through the fiber lumen in counter-current mode using a dosing pump (max. 10 l/h, 3.5 bar, GA 170 Milton Roy, Wokingham Berkshire, UK). The HFMC comprised 7,400 polypropylene (PP) fibers, with a nominal outer diameter (OD) and length of 300 mm and 0.113 m respectively, yielding a surface area of 0.58 m² (based on inner fiber diameter, ID of 220 mm). The fibers were characterized with a nominal pore size of 0.03 mm and porosity of 40%. The fibers (X50) were potted in polyurethane fixed in a polycarbonate housing with an ID of 0.0425 m resulting in a packing density of 0.369. At the outlet of the dosing pump, a pulsation damper (0.65 L Pmax 50 bar inox / EPDM, Hidracar SA, Spain) has been installed to smooth the flow profile of the liquid line, avoiding the effect of water hammer on the connecting line with membrane modules, especially sensitive to cyclic pressure waves. A flow meter (20-250 l h⁻¹, ± 2%, Type 8031, Bürkert, Germany) has been also installed. Also, a configuration of valves has been designed to allow the passage of liquid through two membrane contactors arranged in series or only by one, making a bypass to the second.

Among all the different assays liquid phase circulated inside the fibers while gas phase passed through the shell. Both fluids were fed in counter-current, liquid through bottoms and gas through heads. Liquid pressure typically was fed 0.3 barg upper gas phase. A backpressure valve controlled the pressure in the liquid phase while the gas rotameter (TECFLUID –Series 2000) assured the correct gas pressure in the pilot plant.

Synthetic biogas and biomethane samples were taken in 1l Tedlar bags (VERTEX Technics S.L.) from the plant entrance and from the plant vent, respectively. A gas chromatograph fitted with a thermal conductivity detector (Varian

CP-4900 Micro-GC, Palo Alto, California, USA) was used to analyze the gas samples during the whole experimentation. Four samples per experience in each assay were performed, and first of them was discarded as considered as flush. pH of the inlet and outlets was also monitored (Crison PH 25).

Deionized water (DI) was initially used as the physical solvent. Sodium chloride (NaCl) and sodium hydroxide (NaOH) 1M solutions were subsequently used as comparative absorption solvents. Both absorbents were based on deionized water using NaCl 99.5% (Scharlab, Spain) and NaOH 98.5% pellets (Sigma-Aldrich, US), respectively.

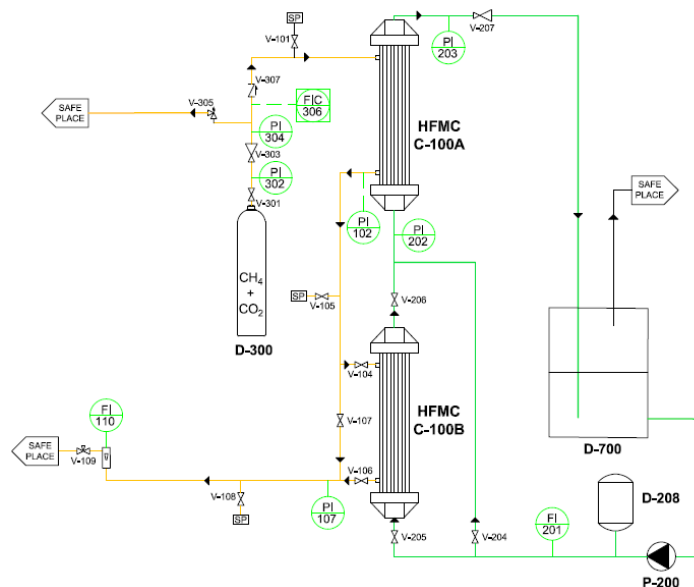


Fig. 2 Schematic of the experimental setup (HFMC, 0.03 mm pore size)

Results and discussion

The type of solvent used, the number of contactors, the pressure of the liquid and gas phases and the relationship between flow rates have been the parameters selected as defining for the characterization of the biogas enrichment process in this work. Repetitions (i.e. duplicates) under particular experimental settings were carried out occasionally, resulting in relative deviations <5%. Temperature is also an important parameter to be considered. According to Zak et al. (2018), CH₄ recovery and separation factor increase with decreasing gas feed temperature, mainly between 20 °C and 25 °C, most likely attributed directly to the decreased solubility of CO₂ in the membrane. However, for temperatures below 20 °C, CH₄ recovery and the separation factor were the highest and become more or less constant. On the other hand, He et al. (2017) state that high temperature in the liquid phase generally results in high molecular activity associated with reduced viscosity and surface tension. Therefore, according to these authors, increasing temperature can improve the CO₂ mass transfer rate in membrane contactors. In this work, all the experiments were conducted at ambient temperature (19-21°C) for gas and liquid phase that is sufficient to achieve reasonable CO₂ absorption performance. The influence of the variation of the temperature of gas and liquid phases on this specific process will be studied in future works of this research group. Finally, in this membrane absorption study, no wetting phenomena (penetration of absorbents into membrane pores) were observed due to short duration of the experimental runs.

3.1 System performance with deionized water as solvent

A set of experiments was carried out using deionized water as solvent, first, with one membrane module operating alone and, later, with two membrane modules operating in series, in all the cases with a single-pass solvent use. The first experiment consisted in keeping constant the gas pressure (P_{o-gas}) and the liquid pressure (P_{i-liq}) and gradually increasing the liquid/gas flows ratio (L/G). The L/G ratio varied in the range 0.5 - 2 for different biogas flows (0.3 – 1.1 l/min). For each experience, gas flow was fixed with the controller and liquid flow was increased from 0.5G to 2G. Selected pressure for the liquid phase was around 2 barg and for the gas phase was approx. 1.6 barg. Biogas upgrading results are shown in Fig. 3. For all the conditions assayed, the better performance of the system was achieved at the lower gas flow (0.3 l/min) and at the higher liquid/gas ratio (L/G=2). It is shown that high gas flow rates lead to short absorption time and thus, low removal efficiency, in agreement with He et al. (2018). On the other hand, and according to Fougetit et al. (2019), as the inlet gas flow increases, the CO₂ concentration will be higher, thus increasing the mass transfer potential. However, this increase in mass transfer does not mean a higher quality of the outlet gas in terms of methane percentage reached. Furthermore, although the increase of liquid flow rate (equivalent to liquid velocity) typically decreases the mass transfer resistance in liquid phase and enhances the absorption performance, the pressure drop along the membrane module increases leading to the increase of membrane wetting, according to Tantikhajongosol et al. (2018). These authors

revealed that the negative influence of wetting on CO₂ absorption is more pronounced than that of liquid velocity at low pressure difference between the gas and the liquid phases. Contrary results were observed when the pressure difference increases. Then, the CO₂ removal increases with the increasing liquid flow rate. This could be due, according to these authors, to the fact that the reduction of mass transfer resistance in liquid due to the increasing liquid velocity is able to increase the absorption performance membrane compensating the negative effect of the wetting phenomena.

Under these operational conditions, biomethane purity was 97.34% (v/v) when operating with one contactor and 99.98% (v/v) when operating with two contactors in series. These maximum yields were obtained when the gas and solvent flow rates were 0.3 l/min and 0.6 l/min, respectively. And then the values decreased with the increase of the biogas flow rate, which mainly results from the insufficient CO₂ absorption due to the short contact time at high gas flow rates. It is also clear the influence of the available mass exchange surface in the system performance, related in this case with the number of membrane modules connected. It is also observed that both, the gas phase and liquid phase (absorbent) flow rates have significant influence on CO₂ removal. Increasing the absorbent flow rate at a fix gas flow rate results in higher CH₄ content in the outlet gas, suggesting better process performance. According to He et al. (2018), higher solvent flow rates lead to faster reactions between the free active absorbent molecules and CO₂ per unit time, leading to better absorption performance. pH of the liquid phase dropped in all experiments around two units due to the CO₂ absorption. The most pronounced pH drop, from 6.01 to 3.69, was achieved in the experiments carried out with two contactors at the lower gas flow (0.3 l/min) and at the lower liquid/gas ratio for that run (L/G=0.58), that coincides with the lower liquid flow and, thus, the maximum CO₂ concentration reached in the solvent.

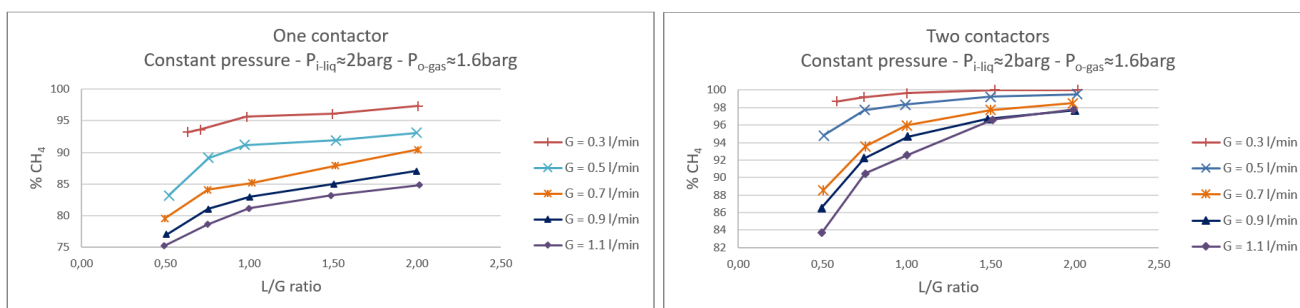


Fig. 3 Methane content in the outlet gas under constant pressures for liquid and gas phases: left) one contactor; right) two contactors in series.

A second experiment consisted in keeping constant the L/G ratio while P_{i-liq} was increased in order to determine the influence of operational pressure in biogas upgrading (Fig. 4). Pressure is an important variable which plays a key role in the liquid-gas equilibrium (Nemestóthy et al., 2018). In this specific case, according to the membrane contactor supplier, maximum operation pressure should be 4.2 barg, although for the economy of the process it is convenient to work at the lowest possible pressure as long as the process performance is acceptable.

For the experimentation, 0.5, 0.7 and 0.9 l/min of gas were consecutively fed in the pilot plant operated with two contactors in series. Liquid phase flow was adjusted accordingly and each assay was performed at L/G = 0.5 and then repeated at L/G = 0.75. Liquid pressure was progressively increased from 0.5 barg up to 3.5 barg. During all the assays liquid pressure was always 0.3 barg upper than gas pressure in order to avoid system malfunction. Experimental results are shown in Fig. 4.

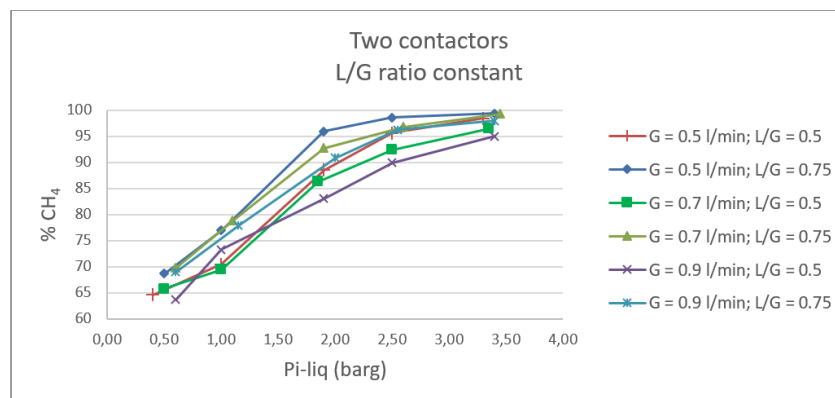


Fig. 4 Methane content in the outlet gas under constant L/G ratio at different operational pressures.

As expected, low pressures are not suitable for high biogas upgrading performance. Liquid pressure lower than 2 barg results in moderate CO₂ removal from the gas phase. Higher methane concentration in the outlet gas stream, 99.44%, was achieved at the higher P_{i-liq} assayed, 3.4 barg (G = 0.5 l/min and L/G = 0.75). At the same P_{i-liq} , the worst yield, 95.29%,

was achieved for $G = 0.9$ l/min and $L/G = 0.5$. So, for the different assays at working liquid phase pressure close to 3.5 barg, methane content in biomethane increases when gas flow decreases and L/G ratio increases. At medium liquid phase pressures (around 2 bargs) this effect is more pronounced. It is pointed out that at high pressures (3.4 barg) results converge between 96% and 100, while at medium pressures results trend to diverge (96.13%-83.92%), so that at high pressures biomethane upgrading is less dependent on flow and flow ratios. Nemestóthy et al. (2018) also demonstrated that slight modifications of process pressures could have a great influence on CO_2 separation, showing these authors that the achievable concentration of the methane in the outlet gas seemed to be positively influence by the greater pressure differences between phases. The monitoring of the pH of the liquid phase in the different experiments reveals a pronounced pH drop when $P_{i\text{-liq}}$ increases at liquid pressures lower than 2 barg from pH around 6 to pH 4 ± 0.1 . At higher pressure values the pH of the liquid phase was independent of $P_{i\text{-liq}}$ and it was only slightly affected (± 0.1 pH units) by L/G ratio.

3.2 System performance with sodium chloride and sodium hydroxide as solvents

An experimental set was designed to compare the behavior of the system when it worked with DI, NaCl 1M and NaOH 1M as solvents in a single-pass solvent use way. In a first run, the pilot plant was operated with two membrane contactors at $P_{i\text{-liq}} \approx 2$ barg and $P_{o\text{-gas}} \approx 1.6$ barg. L/G ratio was progressively raised from 0.5 to 2 by increasing L from 0.35 l/min to a maximum of 1.4 l/min at a fixed $G = 0.7$ l/min. The three solvents were tested consecutively with cleaning periods in between of the assays. Fig. 5 shows the results obtained. CO_2 removal from the liquid phase reached $>99\%$ even for low L/G values when NaOH was used a solvent. On the other hand, NaCl was the solvent yielding the worst results. At $L/G = 2$, for example, NaCl achieved 97.9% of CH_4 purity versus 98.5% achieved when using DI and 99.9% when using NaOH. It is clear, thus, that to achieve an equivalent outlet CH_4 purity, the L/G required for NaOH is lower than for DI and NaCl solvents. In agreement with McLeod et al. (2013), the substitution of the DI and NaCl ‘physical’ solvents for a NaOH ‘chemical’ absorbent resulted in a marked CO_2 removal increment and close to 100% CH_4 concentration in the outgas, which remained unchanged when liquid phase flow, L , was varied. This shows a stagnation in CO_2 removal with increasing L , indicating that the process is gas phase controlled in the case of chemical based solvents, as NaOH. However, CO_2 removal from gas phase increases when liquid flow is increased, showing a liquid phase control in the case of physical solvents, as DI water and aqueous NaCl. These findings are consistent with that observed by Zang et al (2019). These authors also reported that solvent concentration had the greatest effect on the efficiency of CO_2 absorption, followed by gas velocity and liquid (absorbent) velocity. This was associated with the increase in chemical reaction rate between the CO_2 and the absorbent. He et al. (2018) also demonstrated that the CO_2 absorption rate increases with the increase of the absorbent concentration. This effect is attenuated at high absorbent concentrations ($< 1\text{M}$) by the relatively high viscosity of the liquid phase that hinder the mass transfer.

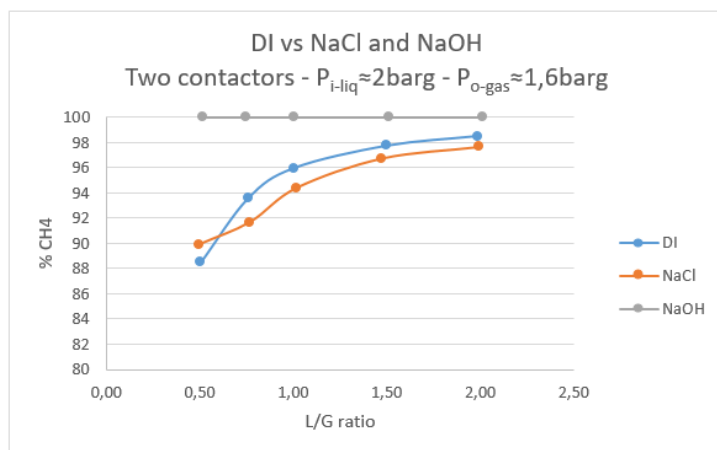


Fig. 5 Process yields operating with two membrane contactors and different solvents (DI, NaOH 1M and NaCl 1M)

In a second run, the pilot plant was operated with one membrane contactor, thus reducing by half the available surface area for molecules diffusion. The operational conditions set, in this case, were $G = 0.5$ l/min, $P_{i\text{-liq}} \approx 2$ barg and $P_{o\text{-gas}} \approx 1.6$ barg. Liquid flow, L , was progressively increased from 0.25 l/min to 1 l/min, thus increasing L/G ratio progressively from 0.5 to 2. As can be seen in Fig. 6, CO_2 removal from the liquid phase reached again 100% when NaOH 1M was used a solvent. A recent study reported that the absorbent concentration had the greatest effect on the efficiency of CO_2 chemical absorption, followed by gas velocity and liquid (absorbent) velocity (Zhang et al., 2019). This was associated with the increase in reaction rate between the CO_2 and the absorbent. A high chemical reaction rate can also be obtained by selecting a suitable chemical absorbent.

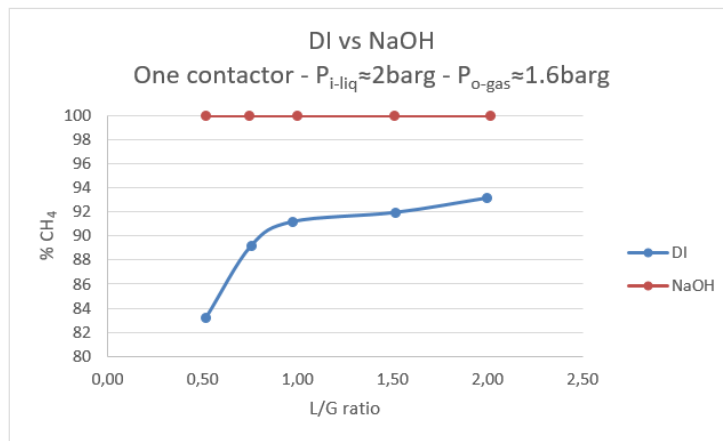


Fig. 6 Process yields operating with one membrane contactor and different solvents (DI and NaOH 1M)

3.3 Solvent recirculation

All the previous assays were carried out with a single-pass solvent use, but, taking into the account the sustainability of the process, from the financial and environmental point of view, solvent recirculation should be considered. A new set of experiments was designed to compare single-pass solvent use to recirculating the solvent in multi-pass operation to enable a greater utilization of the available liquid phase. DI, NaCl 1M and NaOH 1M were used again as solvents. Inlet liquid pressure and gas out pressure were kept constant at $P_{i-liq} \approx 2$ barg and $P_{o-gas} \approx 1.6$ barg during the whole experimentation. Liquid flow and gas flow were also fixed at $L = 1$ l/min and $G = 0.5$ l/min. Percentage of CH₄ in the outlet gas was compared following subsequent solvents uses without regeneration. The three solvents were recirculated in consecutive experiments from zero to five times. Results are shown in Fig. 7. It is observed that, for DI and NaCl solvents, outlet gas content in methane rapidly diminished and the trend is increasingly pronounced as the number of solvent recirculations increases. As a consequence, the capacity to utilize physical solvents in multi-pass absorption without regeneration is very limited so the energy penalty associated with solvent regeneration must also be considered when dealing with economic issues (Martín-Gil et al., 2018). This can be explained by the saturation of the solvent with CO₂ after the recirculations. In comparison, the CO₂ removal and outlet gas quality recorded for NaOH solution following five recirculations was analogous to single pass, demonstrating a sustained excess of highly reactive hydroxide [OH⁻] ions that would theoretically allow a high number of solvent recirculation cycles. The experiments carried out by Galán-Sánchez (2011) under analogous hydrodynamic conditions support this statement, since this author demonstrated that a NaOH solution 0.1 M was sufficient to maintain a >99% CH₄ outlet gas concentration.

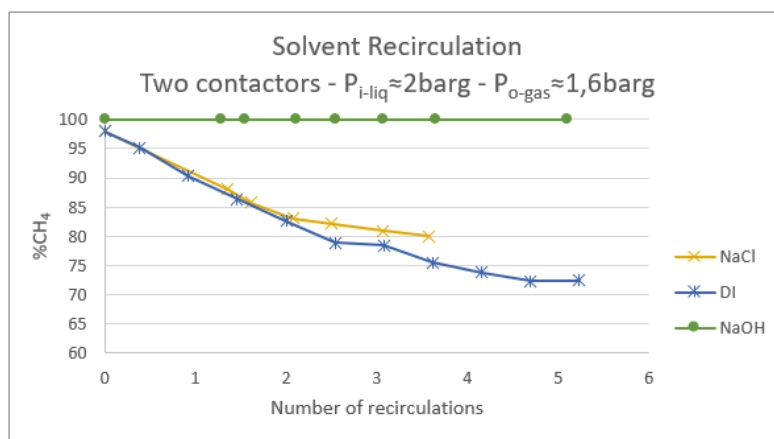


Fig. 7 Process yields operating with two membrane contactors and solvent recirculation at $L = 1$ l/min and $G = 0.5$ l/min

The monitoring of the pH of the liquid phase (Fig. 8) shows a drastic drop at the beginning of the experimentation when DI and NaCl are used as solvents. This indicates that the solvent saturation with CO₂ occurs very quickly. The opposite situation is found when NaOH is used. In this case, the pH decreases very slowly and, as a consequence, the solvent maintains its activity along time.

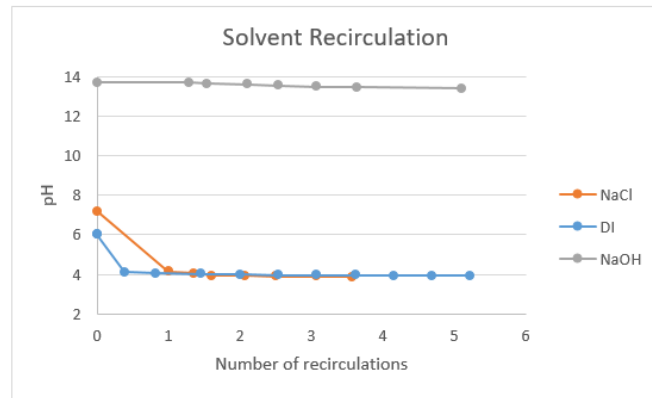


Fig. Liquid phase pH variation with solvent recirculation

4. Conclusions and future prospects

The aim of this study was to evaluate the influence of different operational parameters on biogas upgrading with micro-porous hollow fiber membrane contactors. Through a combined set of experimentations, the following conclusions have been reached:

- Pressure is a crucial parameter. For the pilot system under study, pressures under 1.5 barg are not suitable for biogas upgrading. For intermediate pressures 1.5 - 3 barg biogas purification is highly dependent on biogas flow and L/G ratio. At relative high pressures, above 3 barg, greatest results of biomethane quality were obtained under different operation conditions. Gas and liquid flow and L/G ratio seems to play, in this case, a less relevant role.
- Biogas upgrading showed better performance at higher L/G ratio, but this entails an increase on operation costs. To reduce L/G ratio some action can be implemented such as introducing more mass exchange surface (extra membranes), elevate pressure operation or treat lower raw biogas flows.
- Solvent plays an important role. Minimize solvent usage while keeping the quality of outlet gas supposes an investment cost reduction. NaOH solutions showed better upgrading yields than DI water and HCl solution, even using only one membrane contactor.
- Solvent recirculation without regeneration is not feasible with DI water or NaCl solution. In these cases, solvent is progressively saturated. NaOH allowed several recirculation cycles without showing solvent degradation.
- For physical solvents as DI or NaCl, an enhancement in gas-side methane purity is manifested when liquid phase flow was increased.
- For chemically reactive solvents as NaOH, the process is gas phase controlled, which is illustrated by the negligible gradient recorded for CO₂ separation following an increase in the liquid phase flow.

Hollow fiber membrane contactors for biogas upgrading have a large improvement path until become a stable industrial process. For further development of CO₂ absorption membrane, the challenge is how to improve their durability in operation in the presence of contaminants such as SO₂, usually present in the biogas. Another aspect to be investigated is membrane fouling. Although this phenomenon is reported to be not as severe as that of membrane filtration, in industrial applications where the gas stream contains contaminants such as suspended particles, the membrane fouling becomes a bigger concern. Also wetting phenomenon should be addressed, avoiding the penetration of the absorbent liquid into membrane pores so that a low mass transfer resistance can be achieved. Finally, during biogas upgrading, methane losses occurs. By avoiding this phenomenon upgrading efficiency will improve. Further research on methane slip and measures for its reduction will contribute to the feasibility of the process.

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