Influence of the operational parameters in the separation of CO2 from biogas using hollow fiber membrane contactors

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1. 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. Depending on the final use, different biogas treatment steps are necessary since the biogas is mainly composed of methane, but also carries carbon dioxide and other minority substances such as hydrogen sulfide, oxygen, nitrogen, water, ammonia, siloxanes and particulates.

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. This supposes 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. Biogas upgrading is thus a key issue today to enrich biomethane to produce a natural gas substitute.

Nowadays, there are more than 300 biogas upgrading plants only in Europe and new plants are continually being built around the world due to the increasing demand on biomethane. Five main technologies are used in these plants: water scrubbing, the most popular with almost 40% share, followed by pressure swing adsorption, chemical scrubbing, physical scrubbing and membranes separation, in this order. All of them are proven technologies with potential research on process optimization by means of technologies combination, process configuration or membranes and adsorbents development. In this sense, some emerging technologies, as it is the case of membrane contactors, are offering promising results.

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 they are being also applied in biogas upgrading at low pressure [1-4]. Hydrophobic membranes separate the gas from the liquid phase. Specific molecules from a gas stream diffuse through the membrane and are absorbed on the other side by the liquid flowing in counter current.

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, and to evaluate the impact of multiple solvent cycles on methane recovery compared to single-pass operation.

2. Experimental - A commercial bottle of methane and carbon dioxide gases (PR mixture 2CGC - 40%CO_2, 60% CH_4 v/v, X10S, Carburos Metálicos) was used to provide an initial gas feeding to the shell side of the hollow fibre membrane contactor (HFMC, Liqui-Cel® 1.7 x 5.5 MiniModule®, Membrana GmbH, Wuppertal, Germany). The intlet gas flow rate was controlled and measured using a flow meter (F-201CV Series, Bronkhorst Hi.Tec, Ruurlo, the Netherland). A ball rotameter adapted for CH_4 (Tecfluid 2000 Series) has been 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 ºC by room temperature control. The absorbent was passed through the fiber lumen in counter-current mode using a centrifugal pump (max. 10 L h^{-1}, 3.5 bar, GA 170 Milton Roy, Wokingham Berkshire, UK). The HFMC comprised 7400 polypropylene (PP) fibres, with a nominal outer diameter (OD) and length of 300 mm and 0.113 m respectively, yielding a surface area of 0.58 m^2 (based on inner fibre diameter, ID of 220 mm). The fibres were characterised with a nominal pore size of 0.03 mm and porosity of 40%. The fibres were potted in polyurethane fixed in a polycarbonate shell with an ID of 0.0425 m resulting in a packing density of 0.369. At the outlet of the dosing pump, a pulsation dumper (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.

Deionised water was initially used as the physical solvent. Sodium chloride (NaCl) and sodium hydroxide (NaOH) were subsequently used as comparative chemical absorption solvents.
A gas chromatograph fitted with a thermal conductivity detector (Varian CP-4900 Micro-GC, Palo Alto, California, USA) was used to analyse the gas samples during the whole experimentation.
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.

3. Results and Discussion - Deionised water was initially used as the physical solvent. A series of experiments were carried out with two membrane modules operation in series, first keeping constant the gas pressure (P_{Gas}) and the liquid pressure (P_{Liq}) and gradually increasing the flows ratio (Q_{Liq}/Q_{Gas}) (Fig. 1a). A second experiment consisted in keeping constant the Q_{Liq}/Q_{Gas} ratio while P_{Liq} was increased (Fig. 1b).

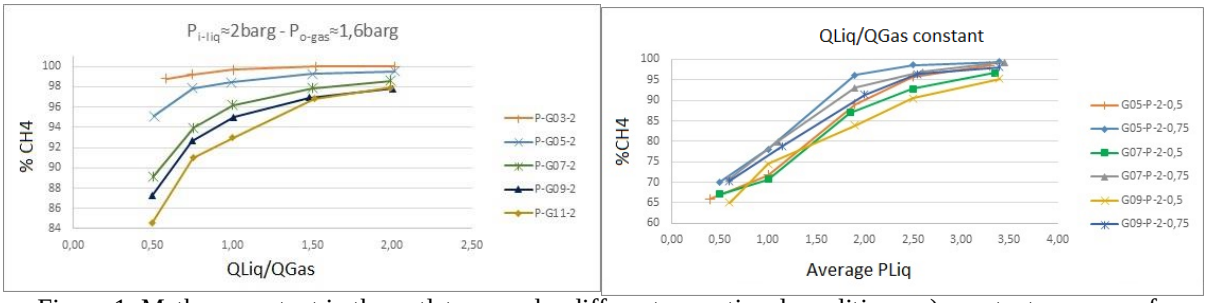

Figure 1. Methane content in the outlet gas under different operational conditions: a) constant pressures for liquid and gas phases; b) constant ration for liquid and gas flows.

Under these conditions, it was possible to obtain a gas stream with more than 99% of pure methane using desionized water as solvent. At constant Q_{Liq}/Q_{Gas} better separation behaviour were targeted at higher pressures (2.0 barg). Carbon dioxide absorption was dependent upon liquid flow (Q_{Liq}). Whilst the highest CO_2 removal from the gas phase was recorded at high Q_{Liq}, selectivity towards CO_2 declined due to low gas residence times. Experimentation with sodium hydroxide and sodium chloride as solvents is on-going.

5. References
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Scientific CV (optional)

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