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# Fuel

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# Biogas upgrading via membrane process: Modelling of pilot plant scale and the end uses for the grid injection

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# highlights

 $\triangleright$  Simulation results with an hollow fiber polymeric membrane fueled with simulated biogas.

- $\blacktriangleright$  Feasibility study for the biogas upgrading plant.
- $\blacktriangleright$  Economical analysis for biomethane production from biogas.

# article info

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# **ABSTRACT**

The paper show the techno economical indications for the upgrading process started from biogas with the scope to produce biomethane for the grid injection and delivered to households and industry or alternatively, it can be used as a fuel for CNG-vehicles. The present work give the numerical simulation with a commercial polymeric membrane, PEEK-SEP™ hollow fiber membranes of the PoroGen Corporation, a US based company that specializes in industrial separation process. The membrane, for the numerical simulation, was fueled with methane, carbon dioxide, hydrogen and nitrogen with a composition similar to the real biogas derived from anaerobic digestion of the organic waste. This study will show the feasibility of integrating anaerobic digestion plant with on site polymeric membrane purification system for conditioned biomethane production.

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# 1. Introduction

Several research center have shown the industrial feasibility of upgrading biogas with polymeric membranes [\[1,2\]](#page--1-0), infact at the actually state, the polymeric membranes show a good level of competitiveness with conventional technologies for the separation of  $CO<sub>2</sub>$  and H<sub>2</sub>S from biogas, such as pressure swing adsorbption (PSA), temperature swing adsorbption (TSA) or amine solution both for what concerns the performance and for the operating costs. Membrane technology was used to separate carbon dioxide from the biogas in order to obtain biomethane of suitable quality for placing into the national distribution network [\[3,4\]](#page--1-0). Most of the literature related to the use of polymeric membranes for carbon dioxide removal, however, is directed to natural gas purification [\[5,6\]](#page--1-0).

State of the art polymeric membranes are economically competitive in separating  $CO<sub>2</sub>$  and  $H<sub>2</sub>S$  from the biogas as compared to conventional technologies in both capital and operating costs [\[7,8\].](#page--1-0) However, commercially available polymeric membranes are typically susceptible to degradation by a number of biogas components such as ammonia and thus require extensive feed gas pre-treatment to protect membranes from degradation which increases purification cost, in particular for hydrogen sulfide, ammonia and siloxane removal with other system for cleaning biogas [\[9–14\].](#page--1-0) At the end of the purification process the biogas still contains methane, hydrogen, carbon dioxide and trace of sulphidric acid and ammonia (<100 ppm) that must be removed from the stream to produce biomethane.

Different are the environmental advantages in the use of biomethane, biohydrogen [\[15\]](#page--1-0) or biodiesel [\[16,17\]](#page--1-0) respect to the others fossil fuels, in fact using biomethane in the automotive sector is possible to reduce the  $CO<sub>2</sub>$  emissions from 75% to 200% as shows in [Fig. 1.1.](#page-1-0)

[Fig. 1.1](#page-1-0) shows that using Hydrogen with the mix EU as combustible for automotive sector the  $CO<sub>2</sub>$  emission are greater than the others fuels inclusive the fossil fuels, while with the biomethane is possible to reduce the  $CO<sub>2</sub>$  emission equal to using the electric car, with a production of  $CO<sub>2</sub>$  of about 5 gr/km; at the other hand





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Fig. 1.1.  $CO<sub>2</sub>$  emissions in the automotive sector for different fuels [\[18\].](#page--1-0)

using methane with 20% of biomethane is also possible to reduce of 20% the GHG emissions.

#### 2. Materials and methods

# 2.1. Membrane upgrading unit description

Recently in ENEA Trisaia we have initiated a project for biogas upgrading with polymeric membranes to increase caloric value of the gas and to purify the gas to natural gas pipeline specifications. ENEA has selected membrane technology from PoroGen Corporation as a main component of biogas upgrading system. PoroGen's membrane technology was selected because of the superior membrane chemical durability (membranes do not require specialized pretreatment to protect from aggressive biogas components that can caused degradation of most commercial membrane systems), because of the compact membrane module size and high membrane separation efficiency.

Polymeric membrane modules utilized in the process were provided by PoroGen Corporation, a US based company that specializes in industrial separation process. PEEK-SEP™ hollow fiber membranes composed of poly ether ketone, polymer were used. The membranes are designed to remove acid gases and water vapors from raw natural gas or biogas to improve gas quality.

The membrane modules used for biogas upgrading are shows in [Fig. 2.1.](#page--1-0)

The [Fig. 2.1](#page--1-0) show the polymeric membrane used in this upgrading plant built by PoroGen Corporation, that has a technology based on melt extruded porous poly (ether ether ketone), PEEK, membranes. PoroGen products are made from VICTREX® PEEK high performance polymers and are used in the most demanding separation applications. The VICTREX<sup>®</sup> PEEK polymer was chosen for its outstanding combination of high heat and chemical resistance. Membrane pore size and surface chemistry of each membrane product is tailored to meet a specific separation application. For high precision separation composite membranes are manufactured by depositing an additional ultra-thin separation layer on top of the porous PEEK membrane. Composite membrane technology platform enables rapid commercialization of new applications by tailoring separation layer material characteristics towards the target application.

PEEK-SEP™ membranes can operate at temperatures as high as 200 $\degree$ C and are not affected by aggressive chemicals present in ''real life'' process streams. PoroGen membranes are inexpensive, yet sufficiently durable to be employed in industrial applications (high temperature gas separations, natural gas treatment, and aggressive solvent filtration) under operating conditions in which other polymeric membranes cannot be used.

## 2.2. Separation device and data treatment

#### 2.2.1. Data analysis

The membrane module input–output scheme is reported in [Fig. 2.2](#page--1-0) were the retentate flow R (CH<sub>4</sub> enriched phase) and permeate P are the unique out streams.

Aiming at compare simulation results under different process conditions, the biogas purity (BP) expressed as methane molar fraction in the retentate  $x_{CH4}^R$  was calculated. Also the hydrocarbon yield of the process Y was considered as the ratio between the methane outlet in the retentate phase and the methane inlet flow rate as it follows:

$$
Y = \frac{R \cdot x_{\text{CH4}}^R}{F \cdot x_{\text{CH4}}^F} \tag{1}
$$

In addition, when varying the methane content on the inlet biogas, the process performance was also evaluated by calculating  $\Theta$ parameter as the percentage relative increase of methane molar fraction between the feed and retentate stream:

$$
\Theta = \frac{x_{\text{CH4}}^R - x_{\text{CH4}}^F}{x_{\text{CH4}}^F} \cdot 100
$$
 (2)

In order to estimate the module productivity as defined below, simulations where run by varying the most relevant process parameters:

- The inlet gas composition (from 40% to 70% in methane; step 10%).
- The feed pressure (from 5 to 30 bar; step 5 bar) at constant permeate pressure (0.1 bar).
- The mass flow rate (from 5 to 80 kg/h; step 5 kg/h up to 40 kg/h and step 10 kg/h up to 80 kg/h).

The membrane module input–output scheme is reported in [Fig. 2.2](#page--1-0) were the retentate flow R (CH<sub>4</sub> enriched phase) and permeate P are the unique out streams.

The simulation results were carried out with a model ad hoc for the prediction both of the purity and the recovery in methane [\[19\].](#page--1-0)

# 3. Simulation results with the single stage

# 3.1. Effect of feed pressure

In [Fig. 3.1](#page--1-0) are reported the results for a fixed 50% (mol/mol) in methane feed as a function of total flow rate and at different inlet pressure.

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