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Research paper

Assessment of membrane plants for biogas upgrading to biomethane at zero methane emission

Gianluca Valenti [a, *], Alessandro Arcidiacono [a], José Antonio Nieto Ruiz [b]

[a] Politecnico di Milano, Dipartimento di Energia, Via Lambruschini 4A, 20156 Milano, Italy
[b] Universidad Politécnica de Madrid, Calle Ramiro de Maeztu, 7, 28040 Madrid, Spain

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ABSTRACT

In the future energy infrastructure there is a considerable potential for biogas and, in particular, for biomethane as a natural gas substitute. Among the alternatives of upgrading biogas to biomethane, this work focuses on membrane permeation. Taking cellulose acetate as membrane material and spiral-wound as membrane configuration, five layouts are assessed. All layouts have the same biogas plant rated at 500 m^3/h (STP), yet they may adopt: (i) one- or two-stage permeation, (ii) permeate or residue recycle, and (iii) a water heater or a prime mover (internal combustion engine or a micro gas turbine) to exploit residues as fuel gas. Since residues are consumed, all layouts have zero emission of methane into the atmosphere. The membrane material is modeled considering the phenomenon of plasticization; the membrane modules are described by a crossflow permeation patterns without pressure drops. The results indicates that specific membrane areas range from 1.1 to 2.4 m^2h/m^3 (STP), specific energy from 0.33 to 0.47 kWh/m^3 (STP), and exergy efficiencies from 57.6% to 88.9%. The splitting of permeation over two stages and the adoption of water heater instead of prime movers is a convenient option. The preferred layout employs a single compressor, a two-stage membrane permeation at 26 bar, a water heater fueled by the first-stage permeate, and a second-stage permeate recycle. Assuming a biomethane incentive of 80 €/MWh_{LHV} and a project life of 15 years, the total investment of this plant is 2.9 M€, the payback time 5 years and the net present value 3.5 M€.

1. Introduction

The future energy infrastructure will be based likely and largely on renewable sources. In this scenario, there is a considerable potential for biogas production from anaerobic digestion of agricultural byproducts, animal manure and slurry. Holm et al. [1] estimate that at least 25% of the whole bioenergy produced in Europe can originate from the digestion of wet biological materials.

The composition of a biogas depends strongly on the organic substrate and the digestion conditions. Typically, biogas has two main constituents, methane and carbon dioxide, and other minor components, water, hydrogen sulfide, nitrogen and oxygen, as well as ammonia and other organic components in very low quantities. The bulk presence of carbon dioxide reduces significantly the calorific value of the gas, whereas the minor components may lead to critical operational problems, like corrosion and clogging. Thus, biogas upgrading to a higher quality combustible gas, the so-called biomethane, requires removing most of that carbon dioxide and of the minor components. Their removal may be achieved by a variety of processes. Ryckebosch et al. [2] provide an accurate review of a large number of these processes, including the membrane systems that are the focus of this assessment. According to the review, the main advantages of the membrane technology are simple construction, easy operation and high reliability, while the general disadvantages are a low selectivity and the possibility of requiring multiple stages.

Technologies for the carbon dioxide separation differ by physical principle, plant layout, removal effectiveness, energy requirements, investment costs, operational efforts as well as the amount of methane that may be unrecovered. Among these technologies, membranes are recognized to be simple, reliable and modular. However, they may require multiple stages to achieve high purities at low methane losses, they cannot recover completely the methane, and they have not found yet a large market diffusion. Nevertheless, market share will predictably increase as membrane

* Corresponding author.
E-mail address: gianluca.valenti@polimi.it (G. Valenti).
URL: http://www.gecos.polimi.it

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materials are improving.

The present paper focuses specifically on agricultural biogas due to the larger potential of this substrate compared to others. Agricultural biogas plants are usually small, located in rural areas, and operated by very few technicians.[1] In this rural context, the biogas upgrading process must be simple and reliable. Naturally, the membrane technology is chosen for the scope. Among all possible materials and modules, cellulose acetate spiral-wound membranes are selected because, despite they are not the best performing option, they are common and robust, as demonstrated by UOP Separex™ and former Grace Membrane Systems technologies [3, Table 2], meeting the mentioned requirement of simplicity and reliability set for the installation in the agricultural sites.

Five layouts of the membrane plant, alternatively connected to the same biogas plant, are assessed here from energy, economic and exergy perspectives. All plants produce pipeline-quality biomethane, inject it into a mid-pressure (5–10 bar) natural gas pipeline, and avoid emitting methane into the atmosphere. The plants differ by:

- number of membrane stages (one or two),
- presence and type of recycles (permeate or residue),
- type of system utilizing the residue gas as fuel gas to avoid the methane emission (internal combustion engine, micro turbine and heater).

All layouts are first optimized economically, as a function of the operational parameters, and then compared against each other. The objective is to determine the strategic layout with the best overall performances. The exergy analysis is employed to identify the major sources of thermodynamic losses.

Methodologically, all layouts are modeled with a bottom-up approach through three levels:

- membrane material (here cellulose acetate as said),
- membrane module (spiral-wound),
- membrane process (five alternative layouts).

The models of membrane plants include all the necessary operational units (compressors, air coolers, and fuel gas utilizers) that constitute the upgrading process. The biogas plant is instead defined in an approximate manner because of its minor interest from the overall process perspective. All models are implemented in a Matlab code developed for the purpose.

To the best knowledge of the authors, the novelty of this paper resides first in the types of considered layouts, which may employ a micro gas turbines as opposed to the commonly considered internal combustion engines or which may combine multiple membrane stages with a heater. Moreover, the biogas plant and the membrane plant are simulated altogether, while most of the studies focus only on the latter. Finally, the simulations do not consider only mass and energy balances, but also an economic assessment over the entire plant lifetime and an exergy evaluation, both of which are original.

The following sections provide in sequence: a general bibliographic review on membrane materials, modules and processes; a description of the considered plants as well as of the numerical models; the numerical results along with their discussion and, ultimately, the conclusions and the future developments.

[1] Here biogas plant refers to the system that converts wet biomass into dehumidified and desulfurized biogas, which could be used in internal combustion engines, while membrane plant to the system that upgrades that biogas into biomethane by membrane technology.

2. Bibliographic review

The next paragraphs review those studies that provide an overview on the topic or that focus on the three scales of the problem: materials, modules and process. For completeness, the sections include cellulose acetate and polyimide as well as spiral-wound and hollow-fiber membranes, despite this assessment covers solely cellulose acetate spiral-wound membranes.

2.1. Overview

Baker [4] describes the membranes from the lower scale of solution-diffusion mechanism to the larger scale of commercial plants. Membrane modules for carbon dioxide removal from methane-rich streams are technically viable. They turn economically competitive for flow rates smaller than 3500 m^3/h at the Standard Temperature and Pressure (STP) conditions of 0 °C and 1 bar. This statement is in agreement with the plant size adopted in this work of 500 m^3/h (STP), as outlined in Section 3.

A review of biogas upgrading by membrane technology is provided by Scholz et al. [5]. According to the authors, available materials are suitable for harsh working conditions of high pressure (around 25 bar) and of chemically aggressive components (H_2S, in particular when H_2O is also present). Single-stage permeation processes are not able to simultaneously produce a high CH_4 purity and obtain a high CH_4 recovery. Hence, multistage layouts become mandatory. Both Baker [4] and Scholz et al. [5] highlight the need for utilizing or even flaring the permeate gas. Sharing this observation, the present work considers only layouts that utilize entirely the permeate.

2.2. Materials

There are many materials proposed for biogas upgrading. According to Basu et al. [6], the most common for commercial applications are polymeric materials, in particular cellulose acetate and polyimide. Harasimowicz et al. [7] report that the high permeability of the polyimide membranes to H_2O and H_2S makes them useful for biogas processing without special pre-treatment, while cellulose acetate membranes, which are sensitive to water vapor, require water removal. The biogas upgrading layouts considered here comprise both H_2O and H_2S abatements.

A problem of polymeric membrane is plasticization, which is the sorption of CO_2 in the polymer matrix that causes a higher polymer chain mobility and, ultimately, a higher mass transport of all gases. Lee et al. [8] describe the effect of plasticization in cellulose acetate membranes by a modified dual-mode theory with concentration-dependent diffusivities, whereas Kanehashi et al. [9] illustrate the plasticization in a polyimide membrane. Both these materials show an appreciable plasticization at pressures above the threshold of 10 atm, which can be raised to 30 atm by special material treatment. The work of Lee et al. [8] is taken as reference for the material modeling (Section 4.1).

2.3. Modules

Commercial membranes for gas separation applications are usually spiral-wound or hollow-fiber modules. The later modules can be designed in coflow, counterflow and crossflow arrangement, while the former in crossflow. Their mathematical models can be solved analytically for simplified cases or numerically for general applications. The analytical solution for crossflow gas permeation of binary mixtures under simplifying condition is presented first by Weller and Steiner in 1950 [10], and extended by Stern and Walavender in 1969 [11], who also correct the membrane area