Available online at www.sciencedirect.com
ScienceDirect
Energy Procedia 82 (2015) 971 – 977
Energy Procedia

ATI 2015 - 70th Conference of the ATI Engineering Association

Bio-methane generation from biogas upgrading by semi-permeable membranes: An experimental, numerical and economic analysis

Caterina Micale[a]

[a]LAR Laboratory - Engineering Department – University of Perugia,Via G. Duranti 93, Perugia 06125, Italy

Abstract

The possibility of upgrading biogas produced by anaerobic digestion of the organic fraction of municipal solid waste (OFMSW) to bio-methane, was investigated with the aid of an experimental apparatus and a numerical model. Different compression pressure and three types of membranes, cellulose acetate (CA), polyamide (PI) and polyaryl-ether-ketone-ketone (PEKK), were investigated. The biogas production and composition turned out to be of about 107 NL/kg OFMSW with a CH_4 and CO_2 content of 60.22%v/v and 38.52%v/v, respectively. The upgrading process requested a membrane surface ranging from 1-1.5m^2h/m^3 to 3.5-6.5 m^2h/m^3 in the case of CA and PI, respectively, whereas for PEKK it ranged from 5 to 14.2m^2h/m^3.Methane content in the upgraded gas was not lower than 95%. The methane losses in all the analyzed scenarios were around 1% and the upgrading costs ranged between 0.08-0.18 €/Nm^3.

Peer-review under responsibility of the Scientific Committee of ATI 2015

Keywords:Anaerobic Digestion, Biogas ,Membranes, Organic fraction of munuicipal solid waste, Upgrading.

1. Introduction

The Anaerobic Digestion (AD) of the biodegradable fraction of municipal solid waste (OFMSW) is a widely exploited process both for energy production and for biological reactivity reduction before recovery and/or disposal operations [1-4]. The biogas produced during AD process results mainly composed by methane (60%v/v) and carbon dioxide (40%v/v) [5, 6], presenting a good energy potential. Currently biogas is mainly used for burning in combined heat and power (CHP) unit for the production of electrical energy and heat [7,8]. Never the less, this solution resulted to be affected by a limited value of the electrical efficiency, generally lower than 40% [9]. A new frontier for the energetic exploitation of the

Peer-review under responsibility of the Scientific Committee of ATI 2015
doi:10.1016/j.egypro.2015.11.854

biogas is currently represented by the upgrading of biogas into bio-methane [10, 11]. The upgrading process of the biogas consists in the removal of CO_2 and other compounds [12] to obtain a gas with an higher CH_4 concentration (≥95%v/v) [13, 14], and consequently higher LHV, that can be used for natural gas (NG) substitution and injected into the NG grid [14].In industrial practice there are several methods for CO_2 separation. Processes based on chemical and physical absorption resulted fully proven [15], but also characterized by high energy consumption and investment costs, making these solutions suitable only for larger-sized facilities[16] (i.e. > 4MW thermal). Another promising industrial solution for biogas upgrading is represented by the permeation through membrane-based technology [17]. This technology showed suitable features for being exploited also for lower-sized AD plants. The membrane acts as a molecular sieve keeping the biggest molecules like CH_4 and letting smaller molecules like CO_2 go, exploiting the partial pressure of the gasses as driving force [18,19]. The membrane modules are compact, simple to use and requiring low maintenance[20].Moreover, the membrane-based technology presents easiness of scaling-up, and for its simplicity it is very promising in particular for lower-sized facilities, up to 1.5-2 MWt, that represent the majority of the anaerobic digestion plants currently operating [21]. However some pre-treatments are necessary to maintain high efficiency of the membrane separation modules and to produce bio-methane in compliance with the required technical specifications [22].

Nomenclature

AD	Anaerobic Digestion
CA	Cellolose Acetate
MSW	Municipal Solid Waste
OFMSW	Organic Fraction of MSW
PEKK	Polyaryl-ether-ketone-ketone
PI	Polyamide
$\alpha_{CO2/CH4}$	Selectivity
p_{Ri}	Partial pressure of i-esm gas in retentate side
p_{Pi}	Partial pressure of i-esm gas in permeate side
P_i	Permeability i-esm gas
J_i	Flux though the membrane of i-esm gas
s	Membrane thickness
j	membrane section

The membranes for biogas upgrading could be classified by module structure [20], but the most important feature of the membrane is the material of which it is made [23, 24]. The most diffused types are: polymeric, inorganic, mixed matrix membranes [25]. The polymeric membranes are the most widely diffused in biogas upgrading [23, 25]. The main parameters affecting membranes performances for biogas upgrading are represented by pressure drop, permeability and surface area [14, 16]. In general high operating pressure leads to higher upgrading efficiency and lower membranes surfaces but also to higher compression costs. Among the numerous polymers available PI [25], CA [25] and PEKK membranes [26] present high selectivity in terms of CO_2/CH_4, and the efficiency of separation could be further increased operating in a multi stage membrane separation process [14, 16, 23]. In this study the quality of the biogas obtained from the anaerobic digestion of the organic fraction of municipal solid waste was evaluated with the aid of an experimental equipment. On the basis of these data PI, CA and PEKK-based membranes performances were numerically investigated for biogas upgrading at different operating conditions. The comparative study was also integrated with an economic analysis.

2. Materials and methods

2.1 AD experimental set up

To evaluate the production and the composition of biogas generated from OFMSW, 9 runs of a SADB process were simulated in an experimental apparatus (Fig. 1) [4]. This apparatus consists of pilot scale SADB reactor, with a gastight, static, steel, cylindrical reactor of 100 liters (Fig. 1), with a removable top.

Process temperature was maintained at mesophilic values (35°C±2°) by a thermal band (TECAM; 400W) powered by a potenziometer (AEG-1phase-230V) controlled by a temperature detector resistance (Pt100) inserted inside the reactor volume (Fig.1). The OFMSW and the inoculum were put inside the reactor in a ratio of 1:1 by weight. The liquid fraction was collected at the bottom of the reactor and recirculated to maintain the optimal humidity conditions. The biogas produced was collected from the reactor top, piped to a dehumidifier vessel and then to a thermal flow meter with a measuring range of 0-10L/h (0.1% FS). CH_4 and CO_2 concentration in biogas %v/v were determined by infrared sensors (±1%) whereas $O_2$and H_2S and other compounds concentration %v/v were included in the remaining fraction (*i.e.* global imbalance 100%).

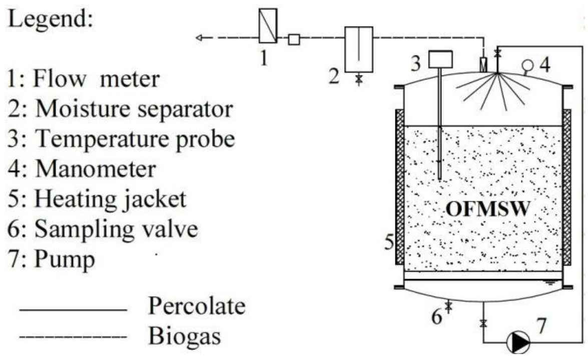


Fig. 1. Solid Anaerobic Digestion Batch experimental apparatus.

2.2 Membrane module

The upgrading process of the biogas based on membrane technology was simulated through a mathematical model. A two-stage upgrading scheme using hollow fibers membrane was considered (Fig.2). As demonstrated by [14, 16, 23], this solution showed high separation efficiency and economic viability.

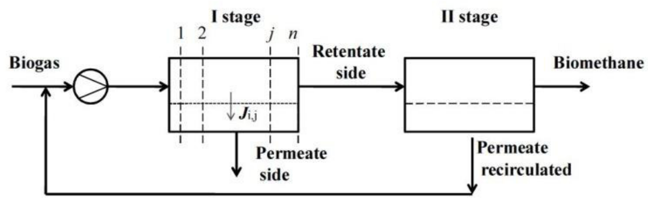


Fig. 2. Two stage membranes upgrading scheme.

The biogas at the membrane inlet was assumed to be already pretreated for the removal of such compounds like H_2S, water and ammonia. Post-compression stage for the upgraded gas utilization was out of the scope of the study. The biogas is compressed to p_R and conveyed into the first module. The I stage modules return two distinct output streams: a retentate, with an higher concentration of CH_4, and a permeate, mainly composed of CO_2. The retentate was piped to the II stage where the remaining amount of CO_2 was definitively removed from the retentate that was returned with a methane concentration ≥ 95%v/v. Pressure losses through the membranes and the effects on separation efficiency of other traces components were disregarded. The membrane film is divided into *j* sections and for each of them the solution-diffusion model [18] was adopted. So the specific flux of CH_4 or CO_2 through the *j-esm* membrane section could be expressed by Fick law (eq.1). The specific permeate output flow (J_i), for each gas, so, is given by the sum of the *j-esm* fluxes (eq.1). The retentate, for the *j-esm* section, is given by the difference between the inlet flux in the *j-esm* section (*j-1* retentate), and the *j-esm* permeate flux ($J_{i,j}$).

$$J_i = \sum_{j=1}^{n} J_{i,j} = \sum_{j=1}^{n} \frac{P_i(p_{R\,i,j} - p_{Pi,j})}{s} \qquad [cm^3/cm^2*s] \qquad (1)$$

The amount of *i-esm* gas that cross the membrane ($J_{i,j}$) depends on membrane Permeability (P_i) referred to the gas, membrane film's thickness (s) and the partial pressure difference of the gas among retentate ($p_{Ri,j}$) side and permeate side ($p_{Pi,j}$) (eq.1). The partial pressure varies depending on the gasses concentration. The membrane permeability depends closely on the membrane material. In this study the performances of three types of membranes were analyzed: CA [16], PI [25] and PEKK [26]. The P assumed for each type and the relative selectivity $\alpha_{(CO2/CH4)}$ were reported in Table 1. The $p_{Ri,j}$ coincides with the feed pressure and was imposed at 10,15 and 20 bar. The economic analysis was performed assuming a reference AD facility with a biogas production of 200 Nm^3/h. According to [13, 27, 28] the membrane useful life was assumed to be of 5 years [28]. Data of the economic model were reported in Table 2.

Table 1. Features of membrane materials assumed in the mathematical model.

Membrane material	P Barrer ($cm^3cm /cm^2 s cmHg$)		$\alpha(CH_4/CO_2)$	Reference
	CO_2	CH_4		
CA	6.3E-10	2.1E-11	30	[16]
PI	1.10E-09	3.03E-11	36.3	[25]
PEKK	2.17E-10	5.63E-12	38.5	[26]

Table 2. Data for the economic analysis.

Capital costs	Cost	Unit	Operating costs	Cost	Unit
Membrane (C1)	55	€/m^2	Compression cost	0.08	€/kWh
Compressor, valves and piping (C2)	1,500	€/kW	Labour and maintenance	10% of capital cost	€/anno
Housing	20,000	€			
Pre-treatment	369	€/Nm^3 biogas			
Other instrumentations	60% of (C1+C2)	€			
Design	10%of capital cost	€			

3. Results and discussion

The mean biogas production and composition evaluated by experimental tests turned out to be of 106.81 NL/kg (±43.3) with a CH_4 and CO_2 content respectively of 60.22%v/v (σ±4.1) and 38.52%v/v (σ±3.5). The O_2 was absent, H_2S and other gasses represent only the 1.25%v/v (σ±0.98). The mean biogas composition turned out to be in accordance with other data referred to biogas production plants from OF of the waste as reported in [3, 4, 6].

The upgrading process by PI membranes resulted to be the most advantageous (Fig.3-a) with a specific surface need ranging between about 3m^{2}h/m^3, for a compression pressure of 10bar, and of about 1 m^{2}h/m^3 for a compression pressure of 20 bar.The CA membrane (Fig.3-a) turned out to be less advantageous with a surface need of about 6.5, 3.5 and 1.5 m^{2}h/m^{3}respectively for 10, 15 and 20 bar. If operated at 10 bar PEKK requested a specific exchange surface of about 14.2 m^{2}h/m^3 for a pressure of 10bar, 6.5 m^{2}h/m^3 for a pressure of 15 bar and about 5 m^{2}h/m^{3}for a pressure of 20 bar (Fig.3-a). The CH_4 content in the upgraded bio-methane was in all cases higher than 95%. Methane concentration in the outlet stream > 97 %v/v was detected only for the CA and PI membranes when operated at 15 and 20 bar. The two-stage upgrading system turned out to be a good solution because the methane losses by the first permeate flow, were quite limited in all the scenarios (*i.e.*<1%). By increasing the compression pressure, the need of specific exchange surface was reduced for all the membrane types because of the raising in the driving force that lead to an enhance in the CO_2 passage through the membrane. The effects of pressure highlight a net divergence in the scenario with 10 bar among the PI, CA and PEKK: the higher the compression pressure, the lower the divergence.

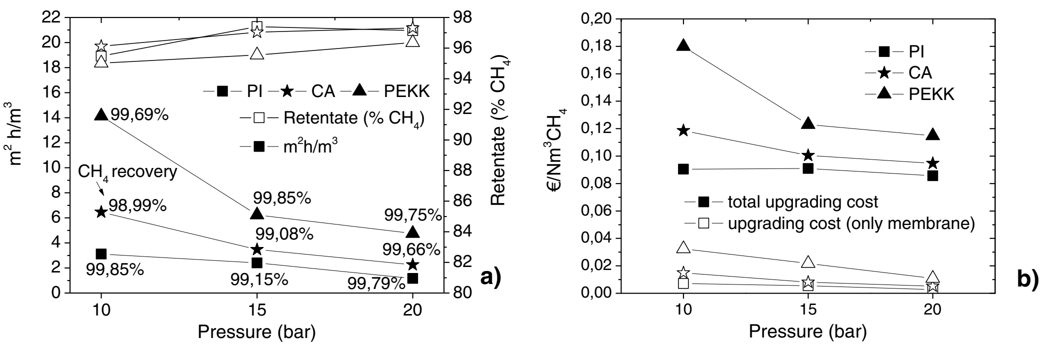


Fig. 3. Performances of PI, CA and PEKK membrane for different compression conditions. Methane recovery fraction, specific surface and methane concentration in the retentate (a), upgrading costs (b) .

The membrane cost (Fig.3-b) in the PI and CA scenario represent about 5-12%of the total upgrading cost that ranged from 0.08 to 0.12 €/Nm^3. For PEKK scenario, membrane cost ranged between 9% and 18% of the total upgrading cost that ranged from 0.11 to 0.18 €/Nm^3. Similar results were obtained by [16] with a specific area of 1.92 m^2h/m^3 for a compression pressure of 20 bar and a CH_4 content in the upgraded gas >95%. For CA membranes [13] reported a specific surface of 3.5 m^2h/m^3 for an operating pressure of 16 bar with a CH_4 recovery >98%. For a feed pressure of about 20 bar, using the most common membranes, instead, [27] reported a specific area demand of 1.27 m^2h/m^3 with a CH_4 content in the biogas of 98% and CH_4 losses in the permeate of 4.3%. In the same study [27] the running costs and energy costs are respectively of 0.012 €/Nm^3 and 0.084 €/Nm^3 but in this case also a further post-compression stage is considered. In the research reported by [14] on similar membrane, with a double stage configuration, the costs for the upgrading range between 0.10 and 0.12 €/Nm^3.

Conclusions

The upgrading of biogas to bio-methane, from AD of the organic fraction of urban waste, could be a suitable way to enhance the reduction of traditional fossil fuel consumption as natural gas. The membrane-based technology can represent a modular, simple and viable solution useful in particular for the medium-small sized AD facility. In particular CA, PI turn out to be most advantageous compared to PEKK in all pressure scenarios. In particular PI and CA performances are quite similar for compression pressure of 15 and 20 bar, instead in the case of 10 bar PI turn out to be the best solution. In all the scenarios however the economic analysis shows affordable upgrading costs if compared with other upgrading technologies which present higher investment costs and higher costs for energy demand.

References

[1] Liu X., Gao X., Wang W., Zheng L., Zhou Y., Sun Y., 2012. Pilot-scale co-digestion of municipal biomass waste: focusing on biogas production and GHG reduction. Renewable Energy 44, 463-468.

[2] Di Maria F., Sordi A., Micale C., 2013. Experimental and life cycle assessment analysis of gas emission from mechanically–biologically pretreated waste in a landfill with energy recovery. Waste Management, 33(11), 2557-2567.

[3] Cavinato C., Bolzonella D., Pavan P., Fatone F., Cecchi F. , 2013. Mesophilic and thermophilic anaerobic co-digestion of waste activated sludge and source sorted biowaste in pilot- and full-scale reactors. Renewable Energy, 55:260-265.

[4] Di Maria F., Gigliotti G., Sordi A., Micale C., Zadra C., Massaccesi L., 2013. Hybrid solid anaerobic digestion batch: biometane production and mass recovery from the organic fraction of solid waste. Waste Manag.& Research, 31(8):869-873.

[5] Di Maria F., Gigliotti G., Sordi A., Micale C., Zadra C., Massaccesi L., 2013. Hybrid solid anaerobic digestion batch: biometane production and mass recovery from the organic fraction of solid waste. Waste Manag.& Research, 31(8):869-873.

[6] De Laclos H.F., Desbois S., Sanit Joly C., 1997. Anaerobic digestion of municipal solid organic waste: Valorga full scale plant in tilburg, The Netherlands. Water Science Technology, 36:457-462.

[7] Hosseini S.S., Peng N., Chung T.S., 2010. Gas separation membranes developed through integration of polymer blending and dual layer hollow fiber spinning process for hydrogen and natural gas enrichments. Jou.of Membrane Sci., 349:156-166.

[8] Yingjian L., Qi Q., Xiangzhu H., Jiezhi L., 2014. Energy balance and efficiency analysis for power generation in internal combustion engine sets using biogas. Sustainable Energy Technologies and Assessments, 6:25-33

[9]Walla C., Schneeberder W. 2008. The optimal size for biogas plant. Biomass Bioenergy, 32:551-557.

[10] Holm-Nielsen J.B., Al Seadi T., Oleskowicz-Popiel, 2009. The future of anerobic digestion and biogas utilization. Bioresource Technology, 100:5478-5484.

[11] Yang L., Ge X., Wan C., Yu F., Li Y., 2014. Progress and perspectives in converting biogas to transportation fuels. Renewable and Sustainable Energy Reviews, 40:1133-1152.

[12] Rasi S., Lantela J., Rintala J., 2011. Trace compounds affecting biogas energy utilisation – A review. Energy Conversion and Management, 52:3369–3375.

[13]Scholz M., Alders M., Lohaus T., Wessling M., 2015. Structural optimization of membrane-based biogas upgrading processes. Journal of Membrane Science, 474:1-10.

[14] Molino A., Migliori M., Ding Y., Bikson B., Giordano G., Braccio G., 2013. Biogas upgrading via membrane process: modelling of pilot plant scale and the end uses for the grid injection. Fuel, 107:585-592.

[15]Yang L., Ge X., Wan C., Yu F., Li Y., 2014. Progress and perspectives in converting biogas to transportation fuels. Renewable and Sustainable Energy Reviews, 40:1133-1152.

[16]Scholz M., Melin T., Wessling M., 2013. Trasforming biogas into biomethane using membrane technology. Renewable and Sustainable Energy Reviews, 17:199-212.

[17] Baker R.W., Lokhandwala K., 2008. Natural gas processing with membranes: an overview. Ind.Eng.Chem.Res., 47:2109-2121.

[18] Wijimans J.G., Baker R.W., 1995. The solution-diffusion model: a review. Journal of Membrane Science, 107:1-21.

[19] Deng L., Hagg M.B., 2010. Techno-economic evaluation of biogas upgrading process using CO2facilitated transport membrane. International Journal of Greenhouse Gas Control, 4:638-646.

[20] Baker R.W., 2002. Future directions of membrane gas separation technology. Ing.End.Chem. Res.,41:1393-1411.

[21] Di Maria F., Sordi A., Micale C., 2012. Energy production from mechanical biological treatment and Composting plants exploiting solid anaerobic digestion batch: An Italian case study. Energy Conversion and Management, 56:112-120.

[22] UNI/TR11537:2014, Immissione di biometano nelle reti di trasporto e distribuzione di gas naturale.

[23] Bernardo P., Drioli E., Golemme G., 2009. Membrane gas separation. A review/state of the art. Industrial Engineering and chemical Research, 48:4638-4663.

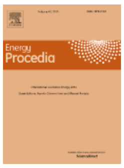
[24] Robeson L.M.,2008. The upper bound rivisited. Journal of Membrane Science, 320:390-400.

[25] Zhang Y., Sunarso J., Liu S., Wang R., 2013. Current status and development of membranes for CO2/CH4 separation: a review. International Journal of Greenhouse Gas Control, 12:84-107.

[26] Wang Z., Chen T., Xu J., 2002 .Gas transport Properties of a series of cardo polyaryleters. Journal of applied Polymer Science, 83:791-801.

[27] Deng L., Hagg M.B., 2010. Techno.economic evaluation of biogas upgrading process using CO2 facilitated transportmembrane. International Journal of Greenhouse Gas Control, 4:638-646.

[28] Shao P., Dal-Cin M., Kumar A., Li H., Singh D.P., 2012. Design and economics of a hybrid membrane-temperature swing adsorption process for upgrading biogas.



Biography

Caterina Micale received the B.Sc. and M.Sc. in Environmental Engineering from the University of Perugia (Italy) in 2009 and 2011, respectively. She is currently a Ph.D. candidate in the Department of Engineering at the same University. Her research interests include biological treatment of waste, renewable energy, landfill improvement, LCA assessment and waste management.