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Energy Analysis of Different Municipal Sewage Sludge-**Derived Biogas Upgrading Techniques**

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Biomass-derived energy sources are rising their importance since both public opinion and legislation are currently calling for a sustainable development. Biogas is an energy source which can come from municipal sewage sludge digestion thus coupling the advantages of being a renewable energy source and of allowing a smart waste reutilization. In order to fully exploit the biogas potential as vehicle fuel or natural gas substitute, biogas itself must be treated in order to obtain biomethane.

Biogas upgrading, i.e., the treatment for CO₂ removal, can be performed by several techniques, each one characterised by a different energy demand. Since no clear guidelines are given in literature for choosing among different biogas upgrading processes, this work presents a quantitative analysis, from an energy view point, of water scrubbing, MEA (monoethanolamine) scrubbing, and MDEA (methyldiethanolamine) scrubbing when applied to obtain biomethane from municipal sewage sludge-derived biogas. Heat and electrical power consumptions of each of the above mentioned processes have been obtained by means of process simulation with commercial packages (such as Aspen Plus®). The aim of the work is the energetic comparison among these different techniques. Such a comparison can help in assessing the impact of the biogas purification step on the energy balance of the whole biomethane production process.

1. Introduction

The development of technologies for the production and the exploitation of renewable energy sources is currently pushed by legislation and/or public investments worldwide, especially in those regions where the highest energy consumption is located such as, e.g., the European Union (Eur-Lex, 2009), USA (US DoE, 2013) and China (Perkowski, 2012).

Among the available renewable energy sources, biogas plays a minor role in the overall bioenergy sector but its market is growing very rapidly: starting from \$17.3 billion in global revenue in 2011, this market is expected to double by 2022, hitting \$33.1 billion in that year (Navigant Research, 2012a).

Biogas can be obtained as a byproduct in the anaerobic digestion of the sludge produced in the wastewater treatment: the main goal of the anaerobic digestion is not the biogas production but the pathogen reduction and stabilization aiming at obtaining manageable biosolids (King County Department of Natural Resources and Parks, 2012). Thus biogas is a renewable energy source obtained in a process for the valorisation of a "waste" biomass. Its uses vary from electricity and heat generation (principally) which is the main use in Europe and Italy as well (Tricase and Lombardi, 2009) - to the injection in the natural gas grids and the utilisation as vehicle fuel after proper treatment (Navigant Research, 2012b). As for the use of biomethane as vehicle fuel the two leading Countries are Sweden and Germany (Tricase and Lombardi, 2009).

In order to obtain a biomethane suitable for to the injection in the natural gas grids or as vehicle fuel, an upgrading process to remove CO₂ is required as the final step in the purification sequence. Until 2008, water scrubbing and PSA technology were the main techniques used for the upgrading, but lately chemical (amine) scrubbers, and to a minor extent also membrane separation units, have increased their market share. In particular amine scrubbing - along with PSA and water scrubbing - has become an established technology for biogas purification (Bauer et al., 2013; Niesner et al., 2013). The amines typically used are

MEA, DMEA and aMDEA (Bauer et al., 2013; Petersson and Wellinger, 2009). Since, at the best of our knowledge, no clear guidelines are given in literature for choosing among different biogas upgrading processes, this work presents a quantitative analysis, from an energy view point, of water scrubbing, MEA (primary amine) scrubbing, and MDEA (tertiary amine) scrubbing when applied to obtain biomethane from municipal sewage sludge-derived biogas. Heat and electrical power consumptions of each of the above mentioned processes have been obtained by means of process simulation with commercial packages (such as Aspen Plus[®]) (Aspen Technology, 2012). The aim of the work is the energetic comparison among these different techniques. Such a comparison can help in assessing the impact of the biogas purification step on the energy balance of the whole biomethane production process.

2. Simulation of the upgrading process

In order to have a reliable comparison, both water scrubbing and chemical scrubbings have been applied to the same biogas stream. The King County South Treatment Plant (Renton, Washington) has been taken as reference: this plant produces 51.9 kmol/h of biogas at 35°C and 1.04 bar. The biogas is saturated with water and, on a dry basis, its molar composition is 61% methane and 38.5% CO_2 (Butler, 2013). In this work, it has been supposed that the final biomethane (on a dry basis) must contain at least 98% v/v of methane.

Both water scrubbing and chemical absorptions have been simulated in Aspen Plus® by means of a rate-based approach. Since the correct description of the VLE conditions is a requirement for a reliable process simulation (Gamba et al., 2009; Pellegrini et al., 2010) properly thermodynamic models have been adopted: details about the modelling approach were reported in previous works for water scrubbing (Gamba and Pellegrini, 2013), MEA scrubbing (Gamba and Pellegrini, 2013; Pellegrini et al., 2011a), and MDEA scrubbing (Langè and Quadri, 2011; Pellegrini et al., 2011b). In all cases, as for heat and mass transfer, the built-in correlations of Aspen Plus® has been used (film theory by Lewis and Whitman (1924) plus Onda et al. (1968) correlation).

2.1 Water scrubbing

The water scrubbing has been simulated considering the process adopted at The King County South Treatment Plant (Renton, Washington) where the Binax system (Kohl and Riesenfeld, 1985) is applied in a packed column (packing: Pall rings) with known geometry working at 18.3 bar (Butler, 2013). No water regeneration step is present since the final effluent of the water treatment plant is used for carbon dioxide absorption and the water that leaves the absorption towers is returned to the treatment plant (Butler, 2013). The plant simulation has been carried out in order to determine the solvent (pure water at 20°C) flowrate needed to obtain the desired biomethane and to calculate the electric power needed for the biogas compression. Considering the total required compression ratio it has been supposed to operate a three stage intercooled compression. Figure 1 shows the compression section arrangement.

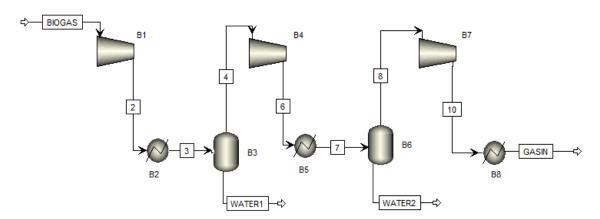


Figure 1: compression section as it has been simulated in Aspen Plus $^{\otimes}$.

After each compression stage the gas is cool down to 35°C and the condensed water is withdrawn from the system. Practically all the water is removed after the first two stages. No water separation is made after the third stage since no further compression is required and the stream "GASIN" of Figure 1 is characterized by a vapour fraction of 0.995.

Table 1 reports the characteristics of the biogas fed to the column (after the compression) and of the obtained biomethane. The required water flowrate for the absorption is 4380 kmol/h. The methane recovery (with respect to the raw biogas) is 95.8%.

Table 1: Raw biogas and obtained biomethane for the water scrubbing

	CH₄ (mol/mol)	CO ₂ (mol/mol)	H ₂ O (mol/mol)	Total flowrate (kmol/h)
Raw biogas	0.6078	0.3836	0.0086	49.5
Biomethane	0.9871 (0.9886 on a dry basis)	0.0114	0.0015	29.2

2.2 Monoethanolamine (MEA) scrubbing

The chemical scrubbing has been simulated considering a conventional process scheme as shown in Figure 2. For what concern MEA scrubbing, both absorption and regeneration have been operated at atmospheric pressure in packed columns (packing: Pall rings). Since MEA is generally used as a 10 to 20% w/w solution in water and the acid gas loading is usually limited to 0.3 to 0.35 moles acid gas per mole of amine for carbon steel equipment (Polasek and Bullin, 1994), in this work a 15% w/w MEA solution has been used and the rich loading, i.e., the loading of the amine leaving the absorption column, has been limited to 0.35. The lean loading, i.e., the loading of the amine fed to the absorption column, has been chosen in order to minimize the heat duty required by the regeneration. Table 2 reports the characteristics of the biogas fed to the column and of the obtained biomethane. The methane recovery (with respect to the raw biogas) is 99.9%. Table 3 shows the result of the minimization procedure for given regeneration column geometry and rich loading.

Table 2: Raw biogas and obtained biomethane for the MEA scrubbing

	CH₄ (mol/mol)	CO ₂ (mol/mol)	H ₂ O (mol/mol)	Total flowrate (kmol/h)
Raw biogas	0.5797	0.3659	0.0544	51.9
Biomethane	0.9173 (0.9871 on a dry basis)	0.0120	0.0707	32.8

Table 3: Results for the minimization procedure of the regeneration reboiler duty for the MEA scrubbing

Lean amine loading	Rich amine loading	Reboiler duty	Reboiler temperature	Amine flowrate [kmol/h]
loading	loauling	[MW]	[K]	[KITIOI/II]
0.204	0.348	3.91	374.0	2620
0.225	0.345	3.59	373.8	3120
0.246	0.349	3.27	373.6	3650
0.266	0.348	3.20	373.3	4580
0.287	0.348	3.38	372.9	6200

It has to be highlighted that also the regeneration column geometry influences the reboiler duty. The choice of the column geometry should come from an economic optimization. Even though the economic analysis is beyond the scope of this work, the minimization procedure of the reboiler duty has been repeated considering a smaller regeneration column than that used in the simulations reported in Table 3 (same diameter but with a 1/6 of the packing volume) in order to verify the correct order of magnitude of a reasonable reboiler duty. The results of this minimization procedure are reported in Table 4.

Table 4: Results for the minimization procedure of the regeneration reboiler duty for the MEA scrubbing with the modified regeneration column geometry

Lean amine	Rich amine	Reboiler duty
loading	loading	[MW]
0.204	0.348	4.87
0.225	0.345	4.34
0.246	0.349	3.83
0.266	0.348	3.60
0.287	0.348	3.61

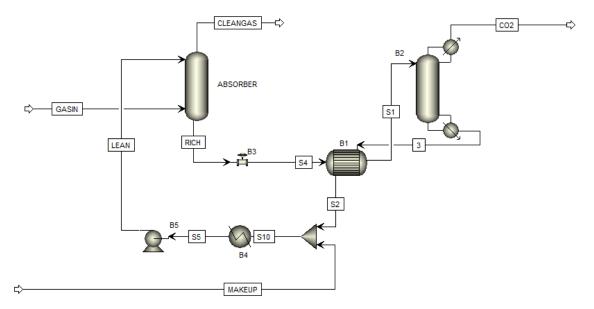


Figure 2: process flow scheme of the chemical scrubbing as it has been simulated in Aspen Plus[®].

2.3 Methyldiethanolamine (MDEA) scrubbing

In the case of MDEA scrubbing, in order to achieve the desired biomethane purity, it has been necessary to operate the absorption process in pressure using a 50% w/w MDEA solution in water. The absorption column geometry is the same as in the case of water scrubbing and the absorption pressure is 2.7 bar (one stage gas compression is required). The regeneration is operated at atmospheric pressure (Ruggeri, 2013). The rich amine loading has been kept lower than 0.5 moles acid gas per mole of amine. Even though loadings as high as 0.7 to 0.8 are practical in carbon steel equipment (Polasek and Bullin, 1994), in order to keep the corrosion rate of the equipment, which depends on the amine loading (Vergani, 2013), as lower as possible with a reasonable amine recirculation rate, the 0.5 value has been chosen. As for the MEA scrubbing, the same procedure has been followed to decide the lean amine loading and the regeneration column geometry. Table 5 reports the characteristics of the biogas fed to the column (after the compression) and of the obtained biomethane. The methane recovery (with respect to the raw biogas) is 99.98%. Table 6 shows the key parameter values for the MDEA scrubbing.

Table 5: Raw biogas and obtained biomethane for the MDEA scrubbing

	CH₄ (mol/mol)	CO ₂ (mol/mol)	H ₂ O (mol/mol)	Total flowrate (kmol/h)
Raw biogas	0.6000	0.3786	0.0214	50.2
Biomethane	0.9632 (0.9873 on a dry basis)	0.0123	0.0244	32.8

Table 6: Main parameters for the MDEA scrubbing

Lean amine loading	Rich amine	Reboiler duty	Reboiler temperature	Amine flowrate
	loading	[kW]	[K]	[kmol/h]
0.046	0.493	321	366.3	319

3. Results and discussion

As shown in Section 2, all three purification processes ensure a biomethane purity higher than 98% v/v on a dry basis. Amine scrubbing allows a higher methane recovery. For what concerns the water content of the biomethane, the gas obtained from the water scrubbing has the lowest water molar fraction while that obtained from the MEA scrubbing has the highest one. This has an impact on the subsequent dehydration process biomethane has to undergo. As for the solvent recirculation, MDEA scrubbing presents the lowest request while MEA and water scrubbing have solvent needs of the same order of magnitude (but water scrubbing works at 18.3 bar instead of atmospheric pressure). MEA scrubbing is affected by the higher heat of reaction with CO_2 that limits the extent of the amine regeneration in order to contain the reboiler

duty and causes a relatively high solvent recirculation (in this case it is also enhanced by the choice of using a dilute amine solution).

In Table7 heat and electrical power consumptions of each upgrading technique are reported.

Table 7: Heat and electrical power consumptions of each upgrading technique

	Electrical power consumption [kW]	Heat duty [MW]
Water scrubbing	213.78	1
MEA scrubbing	1	3.20
MDEA scrubbing	58.78	0.32

Electrical power consumption is due to biogas compression and solvent recirculation (or water pumping in the case of water scrubbing) and thus MEA scrubbing has a negligible electric power consumption since both absorption and regeneration are run at atmospheric pressure. Heat duty is due to the solvent regeneration: water scrubbing has no regeneration section as already stated in Section 2, while MEA scrubbing is characterized by a heat consumption an order of magnitude higher than that of MDEA scrubbing.

On the other hand, the MEA scrubbing process has the advantage of avoiding the biogas compression and requires a smaller absorption column. As earlier mentioned, the column geometry for the water scrubbing is known while in the case of chemical scrubbing the volume of the column has been adjusted, together with the amine flowrate, in order to achieve the desired biomethane purity with an optimal amine usage (i.e., maximum possible rich amine loading). MDEA and water scrubbings require the same absorption column while for the MEA scrubbing process, a packing volume 45 times smaller is enough for the service. It must be point out that the accurate column design is beyond the scope of this work and a careful evaluation must be performed in order to decide between packed and tray columns especially for MEA which requires a lower reaction hold-up since primary amines are characterised by a high rate of reaction with carbon dioxide.

4. Conclusions

In this work the heat and electrical power consumptions of water scrubbing, MEA scrubbing, and MDEA scrubbing have been quantified. Water scrubbing requires the highest electrical power consumption (due to biogas compression and water pumping) while MEA scrubbing requires the highest heat consumption. MDEA scrubbing is between the two above mentioned processes. As for chemical scrubbing both the extent of the regeneration and the dimensions of the regeneration columns have been chosen taking into account a reasonable minimization of the reboiler duty. MEA scrubbing is characterized by a heat consumption an order of magnitude higher than that of MDEA scrubbing. As for the investment costs, MEA scrubbing should be favoured since no biogas compression is required and a smaller absorption column is needed.

As future developments of this work, the MEA scrubbing with a 20 or 30% w/w water solution could be studied as well as an economic analysis could be performed (considering also several biogas flowrates to be treated) in order to decide what is the best process from an economic view point at different scales.

References

Aspen Technology, 2012, Aspen Plus V8.0.

Bauer F., Hulteberg C., Persson T., Tamm D., 2013, Biogas upgrading - Review of commercial technologies, SGC Rapport 2013:270 http://vav.griffel.net/filer/C_SGC2013-270.pdf accessed 11.12.2013.

Butler R., 2013, Private communication.

Eur-Lex, Access to Europen Union Law, 2009, Directive 2009/28/EC of the European Parliament and of the Council of 23 April 2009 on the promotion of the use of energy from renewable sources and amending and subsequently repealing Directives 2001/77/EC and 2003/30/EC http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=CELEX:32009L0028:EN:NOT accessed 11.12.2013.

Gamba S., Pellegrini L.A., 2013, Biogas upgrading: analysis and comparison between water and chemical scrubbings, Chem. Eng. Trans. 32, 1273-1278, DOI: 10.3303/CET1332213.

Gamba S., Soave G.S., Pellegrini L.A., 2009, Use of normal boiling point correlations for predicting critical parameters of paraffins for vapour-liquid equilibrium calculations with the SRK equation of state, Fluid Phase Equilib. 276, 133-141.

- King County Department of Natural Resources and Parks, 2012, 2011 Biosolids Quality Summary http://www.kingcounty.gov/environment/wastewater/Biosolids/DocumentsLinks.aspx accessed 11.12.2013.
- Kohl A.L., Riesenfeld F.C., 1985, Gas Purification, fourth ed, Gulf Publishing Company, Houston, USA.
- Langè S., Quadri C., 2011, Il processo di assorbimento di gas acidi con soluzioni acquose di metildietanolammina, Master of Science Thesis, Politecnico di Milano, Milano, Italy (in Italian).
- Lewis W.K., Whitman W.G., 1924, Principles of gas absorption, Ind. Eng. Chem. 16, 1215-1220.
- Navigant Research, 2012a, Global biogas market to nearly double in size to \$33 billion by 2022 http://www.navigantresearch.com/newsroom/global-biogas-market-to-nearly-double-in-size-to-33-billion-by-2022 accessed 11.12.2013.
- Navigant Research, 2012b, Renewable biogas Methane recovery and utilization in landfills and anaerobic digesters: municipal solid waste, agricultural, industrial, and wastewater market analysis and forecasts http://www.navigantresearch.com/research/renewable-biogas accessed 11.12.2013.
- Niesner J., Jecha D., Stehlík P., 2013, Biogas upgrading technologies: state of art review in European region, Chem. Eng. Trans. 35, 517-522, DOI: 10.3303/CET1335086.
- Onda K., Takeuchi H., Okumoto Y., 1968, Mass transfer coefficients between gas and liquid phases in packed columns, J. Chem. Eng. Jpn. 1, 56-62.
- Pellegrini L.A., Gamba S., Moioli S., 2010, Using an adaptive parameter method for process simulation of nonideal systems, Ind. Eng. Chem. Res. 49, 4923-4932.
- Pellegrini L.A., Moioli S., Gamba S., 2011a, Energy saving in a CO₂ capture plant by MEA scrubbing, Chem. Eng. Res. Des. 89, 1676-1683.
- Pellegrini L.A., Moioli S., Picutti B., Vergani P., Gamba S., 2011b, Design of an acidic natural gas purification plant by means of a process simulator, Chem. Eng. Trans. 24, 271-276, DOI: 10.3303/CET1124046.
- Perkowski J., 2012, China leads the World in renewable energy investment www.forbes.com/sites/jackperkowski/2012/07/27/china-leads-the-world-in-renewable-energy-investment accessed 11.12.2013.
- Petersson A., Wellinger A., 2009, Biogas upgrading technologies Developments and innovations, Task 37 Energy from biogas and landfill gas, IEA Bioenergy http://www.en.esbjerg.aau.dk/digitalAssets/80/80449_iea-biogas-upgrading-report-2009.pdf accessed 11.12.2013.
- Polasek J., Bullin J.A., 1994, Selecting amines for sweetening units, Bryan Research and Engineering, Inc. Technical Papers http://www.bre.com/Support/TechnicalArticles/GasTreating accessed 11.12.2013.
- Ruggeri C., 2013, Confronto tra l'assorbimento fisico con acqua e l'assorbimento chimico con metildietanolammina per l'"upgrading" del biogas, Master of Science Thesis, Politecnico di Milano, Milano, Italy (in Italian).
- Tricase C., Lombardi M., 2009, State of the art and prospects of Italian biogas production from animal sewage: technical-economic considerations, Renew. Energy 34, 477-485.
- US DoE, 2013, Office of energy efficiency & renewable energy < http://energy.gov/eere/office-energy-efficiency-renewable-energy > accessed 11.12.2013.
- Vergani P., 2013, Private communication.