# **Bio Methane Enrichment of Landfill Waste Biogas**

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# Abstract

Landfills are increasingly becoming an attractive source for gas generation, a renewable source of energy that can be used for heating and cooking purposes at both household and industrial level. In this study, a variety of landfill waste with a capacity to generate biogas with 60% bio methane composition was studied for potential bio methane (CH<sub>4</sub>) enrichment. Physical and chemical absorption, membrane separation and cryogenic separation were studied as the bio methane enrichment methane. The methods were ranked in terms of product purity, ease of use and adaptability and the adsorption and membrane separation methods were superior. However, chemical absorbed was then chosen as the bio methane enrichment method due to its low energy requirements of 0.6-1.9 CH<sub>4</sub> percentage of total amount of CH<sub>4</sub> required for heating.

Keywords: Biogas, bio methane enrichment, municipal waste, upgrading methods

#### **1. Introduction**

Landfills are a good source of biogas. The biogas produced from landfills is rich in biomethane (CH<sub>4</sub>) typically ranging between 35 and 75% vol and have higher heating value of 15-30 MJ/Nm<sup>3</sup> (Abatzoglou and Boivin, 2009). However, this biogas must be further upgraded to achive a higher bio methane content. The bio methane (CH<sub>4</sub>) enrichment process is mainly to separate the non-combustible carbon dioxide (CO<sub>2</sub>) in the biogas after other trace impurities have been removed to produce bio methane of high quality >95% CH<sub>4</sub>. The bio methane can be used for various uses such a fuel, cooking and heating purposes. There are numerous methods that can be used to upgrade biogas to bio methane and these include absorption, adsorption, membrane and cryogenic techniques (Ramarajand Dussadee, 2009). The choice of the chosen technique largely depends on some important factors such as biogas composition, available resources such as water, electricity and space, the target purity of CH<sub>4</sub> as well as the volume of biogas to be upgraded.

Absorption: Absorption is a diffusional operation in which some components of biogas in the gas phase are absorbed by the liquid they are in contact with either concurrently or counter currently. The region separating the two phases is called the interfacial region. During absorption, the separation principle is critically based on the solubility of the solute (biogas impurities) in the solvent (Hoyer et al., 2016). There are two types of absorption processes which are determined by the reaction between the solute and solvent which are physical and chemical adsorption processes.

*Physical Absorption Process:* Physical absorption process depends largely on the degree of solubility of the solute in the solvent without any chemical reaction. An example is using pressurised gas scrubbing by means of water as the absorbent is a physical absorption process (Ofori-Boateng and Kwofie, 2009). Organic solvents such as polyethylene glycol-dimethyl ether and propylene carbonate can also be employed during physical absorption. Organic solvents are more effective in absorbing  $CO_2$  and can be operated at low pressure with good chemical stability.

*Chemical absorption:* The chemical absorption process is based on the reactivity of the chemical reagent used as absorbent to chemically react with  $CO_2$  molecule and thus removing it from the biogas feed stream (Biernat and Samson-Brek, 2011). Chemical absorption is more advantageous over physical scrubbing due to its capacity to absorb more  $CO_2$ . Chemical absorption is generally performed using amines solutions and alkaline reagents which include mono-ethanolamine (MEA), di-methyl ethanolamine, di-ethanol amine, deglycol amine and diisopropanol amine.

Membrane purification: Membranes are discrete and thin semi-permeable barriers that selectively separate a feed mixture containing two or more species from one another. The species that moves through the barrier is called permeate and the rejected gases are called retentate. Gases can be separated on two types of membranes; dense membrane (non-porous) and porous membrane. The transportation of gases through dense membranes occurs via solution diffusion while for porous membranes; Knudsen flow, selective diffusion and molecular sieving are the predominant processes. The transportation of gases through membranes takes place when a driving force is applied to the gas species (Zhao et al., 2010). This driving force is mostly due to pressure difference or concentration difference across the membrane. The accurate design and optimization of a gas separation system using polymer membrane depends on the possibility of predicting correctly the membrane transport properties. A number of membrane materials, polymeric and inorganic, exist for  $CH_4/CO_2$  separation. However, polymeric membranes are mostly used for industrial scale application due to their economic advantages over inorganic materials.

Three types of membrane modules exist; hollow fibre modules; spiral wound modules and envelope type modules. Hollow fibre is commonly used in biogas upgrading processes due to its high packing density, low investment cost and operating cost. However, pre-treatment process is always required when hollow fibre is used because it is very susceptible to fouling by hydrogen sulphide ( $H_2S$ ) and it is difficult to clean. Besides the fouling effect on equipment,  $H_2S$  can also have a corrosive effect and must be removed from the biogas (Horikawa et al., 2004).

Cryogenic separation: Cryogenic separation uses the different temperature related properties of the gas species to separate them from the gas mixture (Neisner et al., 2013). The process starts with compression of raw biogas to 26 bar and then cooled to -26 °C for removal of H<sub>2</sub>S, SO<sub>2</sub>, halogens and siloxane. During cryogenic separation, the raw biogas is cooled down step-wisely to temperature where  $CO_2$  in the gas can be liquefied and separated through several heat exchangers. The compressed biogas is dried in advance to prevent freezing. Pure  $CO_2$  has a desublimation temperature of -78.5 °C at atmospheric pressure while CH<sub>4</sub> condenses at -161 °C. Depending on the temperature of the process different purity can be reached, the lower the temperature, the higher the product purity. However, the presence of CH<sub>4</sub> in the biogas mixture affects the physical properties of the gas thus requiring higher pressure and much lower temperature to condense CO<sub>2</sub>. The two main working process cycles of cooling systems as used in the cryogenic biogas upgrading are open loop process cycle and the closed loop process cycle. In the open loop process cycle biogas is first compressed to a high pressure causing a rise in temperature. This creates a good physical property for the biogas to be heat exchanged with lower temperature heat sink. After the biogas has been cooled, it is expanded through a turbine. The biogas can this way reach a low enough temperature to begin the desublimation of  $CO_2$ . In the closed loop process cycle, biogas is not compressed before being heat exchanged thus resulting in temperature difference between the biogas stream and the heat exchanger medium. Since the biogas temperature is not increased through compression, it is not possible to use air as a heat sink therefore a cooling agent mostly  $N_2$  is required to cool the biogas before expansion in a turbine. This decreases both the pressure and temperature which leads to the sublimation of CO<sub>2</sub>. A summary of the bio methane enrichment methods is given in Figure 1 which also includes pressure swing adsorption (PSA).

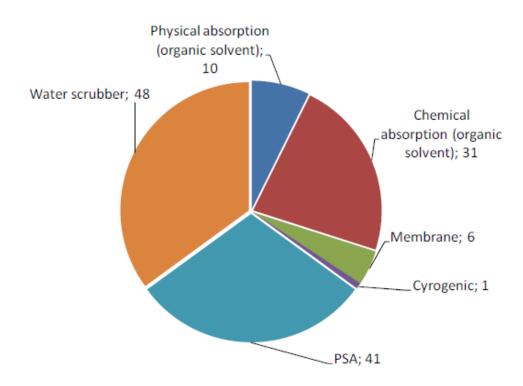


Figure 1. Summary of the bio methane upgrading technologies being applied in Europe (Kaparaju, 2012).

In this study, these three major technologies for enriching biogas are compared and ranked for determining the most effective one that can be used to enrich landfill waste bio methane.

# 2. Materials and Methods

#### 2.1 Determination of biogas production potential from landfill waste

A chosen landfill in South Africa as a case study. The municipal waste was characterized and then quantified for potential biogas production.

#### 2.2 Process selection for bio methane enrichment

The three major technologies for bio methane enrichment absorption, membrane separation, cryogenic were ranked to select the most suitable upgrading technology based on environmental sustainability as the main goal. Four conditions were considered namely environmental, product purity, economics and energy demand, and ease of use and adaptability to the local municipality. The weight of each criterion against the desired goal is as presented in Table 1.

Environmental	Product purity	Economics and energy demand Ease of use and adapta		daptability
Weighted Factors	41%	38%	10%	11%

Table 1. Selection criterion for biogas upgrading technology

#### 2.3 Bio methane upgrading steps

Upgrading biogas to bio methane involves two major steps, namely cleaning and  $CH_4$  enrichment. To some extent, many of the techniques used for removing  $CO_2$  during enrichment can also remove other acid gases and impurities from biogas. Nevertheless, it is often recommended that biogas be cleaned before the enrichment process, since these acidic gases can cause operational problems in the upgrading plant, increase maintenance cost, reduce equipment efficiencies and life span. The cost of cleaning is dependent on the composition and volume of the biogas to be treated but generally it is in the range of 30-100% of the  $CH_4$  enrichment process capital cost. Hence, it is necessary to briefly examine the cleaning of biogas separately, after which upgrading techniques will be discussed in detail.

# 3. Results and Discussion

# 3.1 Biogas production potential from landfill organic waste

Assuming that all waste is fed as substrate into an anaerobic digester, the annual biogas potential is calculated to be 14 096 057 m<sup>3</sup> with an energy potential of 291 274 giga joules (GJ) as presented in Table 2 The bio methane composition of the biogas is 60% on average. Anaerobic digestion of the landfill waste is a biological process by which communities of microorganisms consisting of bacteria metabolically break down complex organic molecules in the absence of oxygen to produce the biogas (Ong et al., 2014). Other energetic equivalent of biogas produced from the organic fraction of the municipal waste at a local landfill is presented in Table 1. The theoretical annual  $CO_2$  reduction from diverting this waste into an anaerobic digester is calculated as 124 327 t $CO_2$ eq.

	Tuote 2. Zhengy potential of all ofganie Wable Jaannied				
Type organic	Organic	Quantity	Biogas (m <sup>3</sup> /yr)	Energy (GJ/yr)	Energy
waste	material	organic			production
		(tons/yr)			
Collected	56%	101 426	7 099 820	140 167	48%
refuse					
Restaurants	1%	1 252	97 489	2 106	1%
waste					
Fruit and	9%	16 936	1 318 806	28 486	10%
Vegetable					
Garden waste	34%	61 345	5 579 941	120 516	41%

Table 2. Energy potential of all organic waste quantified\*

# 3.2 Selection of upgrading technology

Physical absorption, chemical absorption, membrane separation and cryogenic technologies were research upon to evaluate their performance characteristics against each criterion. The priority vector of each alternative technology against each criterion were calculated and presented in Table 3. Of the four alternatives investigated, membrane technology is most preferred in satisfying the main goal alongside it adaptability to the local environmental conditions and technical knowhow. The alternative technologies that are also competitive with membrane are absorption with 99% preference to membrane.

Tuble 5. Runnings of the various biogus particular methods						
Environmen	Product purity	Economics	Ease of Tech	Overall	Idealize	d Priority
tal				Priority		
Absorption	0.08	0.13	0.04	0.02	26.9%	99%
Membrane	0.10	0.08	0.03	0.06	27.2%	100%
Cryogenic	0.11	0.09	0.005	0.01	20.6%	76%

Table 3. Rankings of the various biogas purification methods

# 3.3 Energy requirement for upgrading method

The energy requirement of the upgrading process is also a factor to be considered in technology adoption. Physical absorption, adsorption, membrane and cryogenic upgrading techniques are highly dependent on electricity. Table 4 summarises the electricity and energy requirements of four upgrading techniques. The heating value for bio methane (100% CH<sub>4</sub> concentration) is approximately 35 MJ, which is equivalent to 9.7 kWh. This was used to estimate the energy required for upgrading in column 3 of Table 3.

Table 4. Electricity and energy demand of the upgrading techniques

Separation method	Electricity demand (kWh/m <sup>3</sup> bio methane)	Upgrading energy/ CH <sub>4</sub> heating value (%)
Physical absorption with water	0.2-0.5	2.1-5.2
Physical absorption with organic material	0.10-0.33	1-1-3.4
Chemical absorption with amines	0.06-0.18	0.6-1.9
Membrane separation	0.18-0.30	1.9-3.1
Cryogenic separation	0.18-0.63	1.9-6.5

Chemical absorption upgrading energy demand is the least of the four techniques and demand ranges between 0.6-1.9% of CH<sub>4</sub> heating value but requires temperature as high as 120 °C for regeneration when MEA is used as absorbent (Table 4). Generally, absorption processes are best operated at low temperature and high pressure while

desorption process requires an increased temperature hence a heating and cooling system is required. Cryogenic requires the highest demand on electricity which ranges between 1.9-6.5% of CH<sub>4</sub> heating value for the upgrading process. The energy requirement of a cryogenic plant is reported to be about 580.9 kJ/m<sup>3</sup> of bio methane with a heat pump cycle operating between -100 °C to 40 °C. Adsorption technique was also high because of the compression energy required but membrane technique was about the average of all the processes. The energy demand ranges between 1.9-3.1% of CH<sub>4</sub> heating value. From comparisons of both the easy of technology and the energy demand, chemical absorption was then chosen as the bio methane upgrading technique.

#### 3.4 Proposed bio methane enrichment process

The biogas from the municipal waste is first desulphurized to remove hydrogen sulphide ( $H_2S$ ). After that the desulphurized biogas is compressed to 12 bars and then cooled and filtered to remove contaminants. Adsorption is then carried out for the removal of water, remaining  $H_2S$  and other impurities in the biogas. The biogas is then sent for bio methane enrichment applying either absorption or membrane separation. The enriched bio methane at this stage can be sent for combined heat and power (CHP) uses. The other bio methane can be further compressed to 220 bars and distributed as a source of vehicle fuel. The detailed process flow diagram is shown in Figure 2.

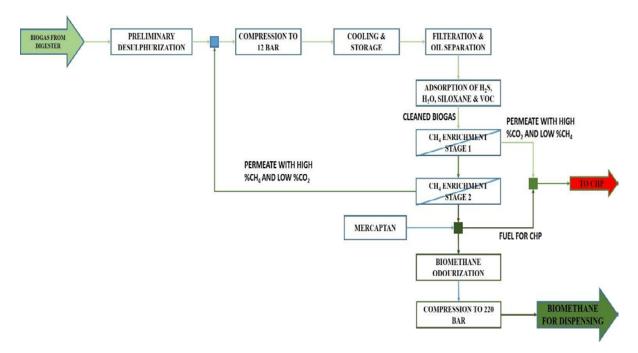


Figure 2. Biogas purification process

#### 4. Conclusion

Potential exist for the conversion of landfill waste to biogas which can be upgraded to bio methane. Biogas upgrading is critical for obtaining high purity bio methane with minimal pollutants. Absorption and membrane separation of the bio methane from the biogas are ideal separation methods in terms of easy of technology and energy usage.

#### References

Abatzoglou, N., and Boivin, S., A review of biogas purification processes. *Biofuels, Bioproducts and Biorefinery, vol.* 3, pp. 42–71, 2009.

Biernat, K., and Samson-Brek, I., Review of technology for cleaning biogas to natural gas quality. *Chemik*, vol. 65, no. 5, pp. 435-444, 2011.

Horikawa, M. S., Rossi, F., Gimenes, M. L., Costa, C. M. M., and da Silva, M. G. C., Chemical absorption of H<sub>2</sub>S for biogas purification. *Brazilian Journal of Chemical Engineering*, vol. 21, no. 03, pp. 415 - 422, 2004

Hoyer, K., Hulteberg, C., Svensson, M., Jernberg, J., and Nørregård, O., Biogas upgrading - Technical review Report 2016:275. 2016 ENERGIFORSK.

Kaparaju. P., Evaluation of potential technologies and operational scales reflecting market needs for low-cost Gas UpgradingSystems. Valorgas

Availablehttp://www.valorgas.soton.ac.uk/Deliverables/111129\_VALORGAS\_241334\_D5-1\_Final\_version.pdf Accessed 19.05.2012.

- Niesner, J., Jecha, D., and Stehlík, P., Biogas upgrading technologies: State of art review in European Region. *Chemical Engineering Transactions*, vol. 35, pp. 517-522, 2013.
- Ofori-Boateng, C and Kwofie, E.M., Water scrubbing: A better option for biogas purification for effective storage. *World Applied Sciences Journal*, vol.5, pp. 122-125, 2009.
- Ong, M. D., R. B. Williams, S.R. Kaffka. (California Biomass Collaborative, University of
- California, Davis). 2014. Comparative Assessment of Technology Options for Biogas

Clean-up. Contractor Report to the California Energy Commission. Contract CEC-500-11-020.

Ramaraj, R.,and Dussadee, N., Biological Purification Processes for Biogas Using Algae Cultures: A Review. *International Journal of Sustainable and Green Energy*. Special Issue: Renewable Energy Applications in the Agricultural Field and Natural Resource Technology. vol. 4, no. 1-1, pp. 20-32, 2015. doi: 10.11648/j.ijrse.s.2015040101.14.

Zhao, Q., Leonhardt, E., MacConnell, C., Frear, C., and Chen, S., Purification technologies for biogas generated by anaerobic digestion. CSANR Research Report 2010 – 001 Climate Friendly Farming.

#### **Biography**

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