# A Review on Upgrading Calorific Value of Biogas from the Digester

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Abstract - The consumption of fossil fuel is increasing day by day and is going to deplete soon. Biogas is a clean environment friendly fuel. Biogas produced in the digester from anaerobic digestion of organic waste cannot be used straight off as a fuel. The gases produced in the digester from anaerobic digestion are methane and trace components like carbondioxide, water vapour, hydrogen sulphide, siloxanes, Hydrocarbons, NH<sub>3</sub>, O<sub>2</sub>, CO and N<sub>2</sub>. To use biogas as fuel, its calorific value should be about equal to calorific value of natural gas. Hence calorific value of biogas can be upgraded by removing carbon dioxideand trace components from biogas obtained from the digester. These gases are not completely combustible and will harm, living beings, engine parts and the monuments. Upgrading biogas, two steps are performed: (1) cleaning process to separate trace components of gases and (2) upgrading process to improve calorific value of biogas. This paper reviews the attempt made to upgrade calorific value of a biogas by using different methods for upgrading.

Index Terms - fossil fuel, biogas, Calorific Value

Nomenclature

CV-Calorific value VSA-vacuum swing adsorption PSA-pressure swing adsorption

I. INTRODUCTION

Anaerobic fermentation (AF) of organic waste produces a biogas with high concentration of methane (CH<sub>4</sub>). The biogas formed in AF plants consists of 55–80vol% CH<sub>4</sub>, 20–45vol%CO<sub>2</sub>, 0–1.0vol%H<sub>2</sub>S, 0–0.05vol% NH<sub>3</sub>and it is saturated with water [1]. Methane is the component chiefly responsible for a typical calorific value of 21-24 MJ/m<sup>3</sup> or around 6kWh/m<sup>3</sup>. Biogas is used for cooking, warming, lighting or electricity generation. Larger plants can feed biogas into gas supply networks. The activities of at least three bacterial communities are required by the biochemical chain which releases methane. Initially, during hydrolysis process, extracellular enzymes degrade complex carbohydrates, proteins and lipids into their constituent units.

Next is acidogenesis (or fermentation) where hydrolysis products are transformed to acetic acid, hydrogen and carbon dioxide. The facultative bacteria mediating these reactions exhaust residual oxygen in the digester, thus producing suitable conditions for the last step: methanogenesis, where obligate anaerobic bacteria control methane production from acidogenesis products. Anaerobic digesters are typically planned to run in the mesophilic (20–40 °C) or thermophilic (above 40 °C) temperature zones. Sludge produced from the anaerobic digestion of liquid biomass is often used as a fertilizer[2]. The main impurities arecarbon dioxide, which lowers the calorific value of the gas and hydrogen sulphide which could cause several problems on the engine parts and on human health, it causes corrosion in engine parts (compressors, gas storage tank and engines), while it is toxic after its inhalation. Although  $CO_2$  is a major problem in the biogas and its removal is useful to upgrade the calorific value and the relative density according to the specifications of the Wobbe index [3,4].

The unwanted  $CO_2$  will lower the quality of biogas and contribute to negative effect on biogas compression. After removing  $CO_2$ , biogas can be used as renewable and low carbon fuel substituting natural gas for electricity generation and natural gas vehicle transportation. Methods for biogas upgrading is mainly concentrated on the removal of  $CO_2$  with a little  $CH_4$  loss [5]. After upgradation, the final product is referred to as 'Biomethane', typically holding 95-97%  $CH_4$  and 1-3%  $CO_2$ . Biomethane can be utilized as an alternative for natural gas [6]. Biogas can be utilized for high temperature and steam production, electricity production and/or co-generation, vehiclesfueling, chemicals production and injection into the natural gas grid. Nevertheless, using biogas as a vehicle fuel or injecting it into the natural gas grid is applications that are gaining interest at the international stage. In both the terminal cases, an appropriate upgrading of the biogas i.e. removal of  $CO_2$  and trace contaminants is required. The output gas from the upgrading procedure is generally called Biomethane and it is qualified by an increased content of methane with respect to the entering biogas [7].



Fig. 1.Wobbeindex and relative density as function of methane content of the upgraded gas [4].

II. METHODS OF REMOVING CO<sub>2</sub>

#### A. Absorption

#### a) Chemical absorption process

Three agents NaOH, KOH and Ca(OH)<sub>2</sub> can be used in chemical upgrading of biogas [8]. The absorption process is based on using aqueous solutions of potassium hydroxide, with the final aim of producing potassium carbonate. The absorption reaction between KOH and the  $CO_2$  produces an aqueous solution of K<sub>2</sub>CO<sub>3</sub>:

$$2 \text{ KOH} + \text{CO}_2 \rightarrow \text{K}_2 \text{CO}_3 + \text{H}_2 \text{O} \qquad (1.1)$$

In this reaction, excess KOH leads to an increased production of potassium carbonate, while excess  $CO_2$  leads to undesired chemical reaction as follows:



Potassium carbonate is a product which has many applications in the chemical industry if adequate quality is available (e.g., crystal industry, special glass production, potassium salts, inks and pigments, detergents, food industry, waste gas treatment). It can be sold as a pulverized solid, or in aqueous solution. The volumetric concentration of  $CH_4$  significantly increases between 85-97%, similar to conventional natural gas quality [9]. This process may have higher energetic penalties since the  $CO_2$  removal in biogas is a bulk removal process. The  $CH_4$  purity in the range 97-98% is obtained[10].

#### b) Physical absorption process

This is one of the easiest and least cost method involves the use of pressurized water as an absorbent. The raw biogas is compressed and fed into a packed bed column from the bottom and the pressurized water is sprayed from the top. The absorption process is, thus a counter-current one. This dissolves  $CO_2$  as well as  $H_2S$  in water, which are collected at the bottom of the tower. The water could be recycled to the first scrubbing tower [8]. This is the simplest method for upgrading biogas. A continuous counter-current type scrubber with gas flow rate of 1.8 m<sup>3</sup>/h at 0.48 bar pressure and water inflow rate of 0.465 m<sup>3</sup>/h. It continuously reduced  $CO_2$  from 30% at inlet to 2% at the outlet by volume [8]. With this technique,  $H_2S$  is removed with  $CO_2$ . Also the purified  $CH_4$  stream (with purity up to 98%) should be dried after leaving the scrubber [10]. The below figure shows that physical absorption of  $CO_2$  in water at elevated pressure. The packed bed scrubbing column with 3500 mm in height and the packing material used was ceramic resching rings. 99%



Fig. 3. Setup for biogas purification and bottling plant [13].

In below setup CO<sub>2</sub> removal ratio increases from 34.6% to 94.2% when liquid/gas ratio was varied from 0.14 to 0.5 at different temperature. At pressure of 1.2 MPa with gas flow rate of 400 L/h and water flow rate 200 L/h, the CO<sub>2</sub>removal ratio obtained is 94.2%. Higher pressure improves CO<sub>2</sub> removal ratio. For better CO<sub>2</sub> removal, both pressure and liquid/gas ratio must be controlled. When the temperature increases from 7°C to 40°C, the CO<sub>2</sub> removal ratio decreases from 85.3% to 52.2%. Hence, lower the

When the temperature increases from  $7^{\circ}$ C to  $40^{\circ}$ C, the CO<sub>2</sub> removal ratio decreases from 85.3% to 52.2%. Hence, lower the temperature in absorption tower is helpful for the improvement of CO<sub>2</sub> removal.

Increasing the water inflow rate, increases the  $CO_2$  removal efficiency. When  $CO_2$  content in the simulated gas increases from 25% to 45% at different inlet water flow rate, the  $CO_2$  removal ratio increases from 24.4% to 83.2%. Finally in this method, 2.6% of  $CO_2$  is present which meets the requirement of  $CO_2$  content in the natural gas for vehicle fuel.



1-CO<sub>2</sub> cylinder; 2-air compressor; 3-gas buffer tank; 4-absorption tower; 5-rich solution tank; 6-heat exchanger; 7-desorption tower; 8-air blower; 9-water pump; 10-lean solution tank

## B.Adsorption

PSA and VSA are technologies that use a column filled with a molecular sieve to separate some gas species from a mixture of gases under pressure [15,16]. To separate  $CO_2$ ,

activated carbon, silicagel, alumina or zeolite are typically used. The molecules are adsorbed in a reversible way in the cavities of the molecular sieve, so it is possible to create a cyclic batch process where the adsorber can be regenerated. Adsorption is performed at a relatively high pressure (800 kPa) while desorption is performed at lower pressure [17]. Normally four vessels are used, each working on a different stadium: adsorption, depressurization, desorption and pressurization [11]. Before the PSA or VSA unit, H<sub>2</sub>S must be taken out in order to prevent poisoning of the molecular sieve. In addition, the PSA process requires dry gas; so it is necessary to dry raw biogas before the upgrading process. PSA and VSA are similar systems, with the only difference that VSA requires a supplementary vacuum pump in this case the differential pressure is fixed at a lower absolute pressure [12].



Fig. 5. Set-up of (vacuum) pressure swing adsorption [4].

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The efficiency of this process is up to 98% [8]. One of the advantage of the PSA process is that is can be adapted to biogas upgrading in any part of the world since it does not depend on the availability of cold or heat sources [10].

#### C.Membrane separation processes

The use of membranes for gas cleaning is a well established technology in chemical industries. The membrane is a porous material that lets some gases permeate through its structure. The most commonly used materials are hollow fibers made of different polymers [10].



#### a) Gas-gas separation

Gas is pressurized (2000 to 3600 kPa) and cleaned to remove hydrocarbons,  $H_2S$  and oil vapor. Acetate-cellulose membranes separate small polar molecules as  $CO_2$ ,  $H_2S$  and moisture. With this technique, it is possible to purify biogas to maximum 92%  $CH_4$  in one step. With more steps (two or three), a gas with even more than 96%  $CH_4$  is achieved. The off-gas still contains 10-25% of  $CH_4$ , so that it has to be flared or used in a steam boiler [19,20]. Besides operation at high pressure, a pilot study has been carried out at low pressure (800 kPa) membrane separation system. Trace components need to be removed before the membrane-unit i.e. by an active natural gas can be achieved, compared to high pressure membrane separation techniques [21].

#### b) Gas-liquid separation

Gas-liquid absorption membranes have been developed recently and they consist in a micro porous hydro-phobic membrane that separates the gaseous from the liquid phase. The gas stream molecules which are able to diffuse through the membrane will be absorbed on the other side by the liquid. This process involves gas and liquid flowing in counter current. The gas is pressurized, so liquid is prevented to flow to the gas side. This process operates at atmospheric pressure (100 kPa), with advantages on the point of view of construction and operating cost. The removal of  $CO_2$  is carried out with an amine solution and biogas with more than 96%  $CH_4$  can be obtained in one step. Furthermore the amine solution can be regenerated by heating. During this process, a pure  $CO_2$  flow is released and can be sold for industrial applications [19].

#### D. Cryogenic separation

It is possible to produce bio-methane by cooling and compressing the biogas, since different impurities liquefy at different temperature and pressure conditions. The raw gas is compressed in multiple stages, up to 8000 kPa. To prevent freezing during the cooling steps, the gas is dried before. After the compression, the biogas is cooled until -45 °C. The condensed  $CO_2$  is removed and treated to recover the  $CH_4$ . The biogas is cooled further to -55 °C; afterward is expanded to 800-1000 kPa reaching the temperature of -110 °C. In this condition, there is solid  $CO_2$  and a gaseous phase with more than 97%  $CH_4$ , which is collected and heated before leaving the process [4,11,8].

In the Netherlands, a cryogenic system to upgrade biogas to natural gas quality wasimplemented in four steps[20]. The incoming biogas is first compressed to 1000 kPa and cooled to -25 °C. Moisture, H<sub>2</sub>S, SO<sub>2</sub>, halogens, siloxanes and other trace components are removed from the biogas. From there, the biogas is further cooled till -55 °C. Liquefied CO<sub>2</sub> is taken out from the gas mixture so the calorific value is increased. The remaining gas stream is further cooled till -85 °Cwhere CO<sub>2</sub>convertsinto solid form. Three vessels are used: one is for upgrading the biogas, set at -85 °C, one is kept at intermediate temperatures (-85 °Cto -60 °C) and one vessel is set at -60 °Cto liquefy the CO<sub>2</sub>. The gaseous phase is depressurized and can be injected after removing odour into the natural gas grid. The results show high efficiency and low methane losses [20].

#### E.Biological methane enrichment

In this experiment, different methanogens using only CO<sub>2</sub> as a carbon source and H<sub>2</sub> as an energy source were examined. The selection between mesophilic and thermophilic operation temperatures is typically based on whether the completion of reaction or the rate of reaction is of primary concern. Thermophilicmethanogens exhibit rapid methanogenesis, while mesophilic bacteria give more complete conversion of the available CO<sub>2</sub>. The organism works optimally at temperatures of 65-70 °C and has a specific requirement for H<sub>2</sub>S, so both unwanted components are removed. A synthetic biogas of 50-60% CH<sub>4</sub>, 30-40% CO<sub>2</sub> and 1-2% H<sub>2</sub>S was mixed with H<sub>2</sub> to a final mole fraction of H<sub>2</sub>: CO<sub>2</sub>equaling 0.79:0.21. The gas mixture was fed to the hollow fiberspacked with organisms. This biological system can effectively remove CO<sub>2</sub>and H<sub>2</sub>S, while approximately double the original CH<sub>4</sub>mass. Alternative





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physicochemical treatment methods only get rid of the contaminating gas components, without changing  $CH_4$  mass. The purified biogas contains about 96%  $CH_4$  and 4%  $CO_2$ , while  $H_2$  and  $H_2S$  were not detected.

## III. METHODS OF REMOVING H<sub>2</sub>S

## A. Removal of $H_2S$ during digestion

 $H_2S$  can be treated directly in the digester vessel and the most used techniques to interfere with its formation are adding air/oxygen or iron chloride [4, 11].

## a)Air/oxygen dosing to the biogas system

This technique is based on the biological aerobic oxidation of  $H_2S$  to elemental sulfur by a group of specialized microorganisms. The following reaction occurs in the biogas:



Elemental sulfur and sulfate is formed, which cause corrosion in solutions. A small amount (2-6%) of  $O_2$  needed for the reaction to occur, is introduced in the biogas system by an air pump. A reduction of H<sub>2</sub>S concentrations down to 20-100100 cm<sup>3</sup>/m<sup>3</sup> and a removal efficiency of 80-99% can be achieved [15,19,8]. Safety steps have to be adopted to avoid overdosing of air in the digester. Biogas in air (6-12%) is an explosive mixture. Safety must be considered so that anaerobic conditions remain present in parliamentary procedure to prevent the digestion process from being inhibited [19,8].

## b)Addition of iron chloride into the digester

Iron chloride reacts with the H<sub>2</sub>S present in the biogas to form FeS as solid precipitated particles. These are the chemical reactions:

$$2Fe^{3+} + 3S^{2-} \rightarrow 2FeS + S$$
 (1.4)  
 $Fe^{2+} + S^{2-} \rightarrow FeS$  (1.5)

The presence of  $H_2S$  in the biogas is avoided due to precipitation of FeSand therefore this method can achieve a reduction of  $H_2S$  concentration in the biogas down to 100 cm<sup>3</sup>/m<sup>3</sup> at standard condition [22].

## B.Removal of $H_2S$ after digestion

## a) Adsorption using iron oxide or hydroxide

Hydrogen sulfide reacts easily with iron oxide, iron hydroxide and zinc oxide and forms iron sulfide or zinc sulphide respectively. This procedure is frequently concerned to as "iron sponge" because rust-covered steel wool may be employed to form the reaction bed. Iron oxide and iron hydroxide react with  $H_2S$  in the biogas according to following reactions

 $Fe_2O_3 + 3H_2S \rightarrow Fe_2S_3 + 3H_2O \qquad (1.6)$ 

 $2Fe(OH)_3 + 3H_2S \rightarrow Fe_2S_3 + 6H_2O \quad (1.7)$ 

The iron oxide can be regenerated with oxygen according to the following reaction:

$$2Fe_2S_3 + 3O_2 \rightarrow 2Fe_2O_3 + 6S \qquad (1.8)$$

This reaction is exothermic and therefore a great amount of high temperature is released during regeneration. This may contribute to self-ignition of the wood chips, if air flow and temperature are not carefully checked. Typically two reaction beds are established. One bed is regenerative while the other bed removes  $H_2S$  from the biogas [15]. The formed elementary sulfur remains on the surface and blocks the active iron oxide or hydroxide, restricting the number of cycles that can be performed [19].

## b) Adsorption on activated carbon

 $H_2S$  can also be removed by using activated carbon often dosed with KI or sulfuric acid ( $H_2SO_4$ ) to increase the reaction rate. In biological filters the  $H_2S$  is catalytically converted to elemental sulfur and water:

$$2H_2S + O_2 \rightarrow 2S + 2H_2O \quad (1.9)$$

The biogas must have a certain amount of air before entering the carbon bed, therefore 4-6% air is added. The formed elementary sulfur is adsorbed by the activated carbon. Best efficiency is obtained at pressures of 700-800 kPa and temperatures of 50-70 °C. This temperature is easily achieved through heat generation during compression. If a continuous process is required the system canconsist of two vessels [4,11,19]: one vessel for adsorption and the other for regeneration. Regeneration can be performed with hot nitrogen

(inert gas) or steam. The sulfur is vaporized and, after cooling, liquefied at approximately 130 °C. Typically, the activated carbon is replaced rather than regenerated [4,6,11].

## IV. METHODS OF REMOVING WATER

#### A.Physical drying methods (condensation)

The simplest way of removing excess water vapor is through refrigeration. The minimum dew point reachable is 0.5 °C at atmospheric pressure due to freezing problems on the surface of the heat exchanger. To achieve lower dew point the gas has to be compressed before cooling and then later expanded to the desired pressure [4,11]. The utilization of any form of anti-freezing compound is not advocated. The chemical treatment it is very complicated and adding other compounds would be overly expensive and would need more complexity in the system. In fact, adding any chemicals in the gas requires a control injection system to ensure the correct composition and an absorption tower to re-separate the glycol from the biogas.

Techniques using physical separation of condensed water include:

- Demisters in which fluid molecules are removed by wired mesh (micro-pores, 0.5-2 nm);

- Cyclone separators in which water droplets are separated using centrifugal forces;

- Moisture traps in which the condensation takes place by the expansion. As result the temperature reductions and the water condenses;

- Water taps in the biogas pipe from which condensed water can be removed [4,22].

## B.Chemical drying methods

These techniques are usually applied at elevated pressures. The most usual technique is adsorption by using alumina or zeolites/molecular sieves. Methods based on gas drying include:

- Adsorption of water vapor on silica, alumina or equal chemical components that can hold water molecules (adsorption dryer). Two chemical columns are often used together in order to adsorb the water and to regenerate the adsorber;

- Absorption of water in triethylene glycol. Regeneration of saturated glycol solvent is made out in a specific unit operating at a temperature of 200 °C;

- Absorption of water with hygroscopic salts. The salt dissolves as it takes up water from the biogas. The salt is not regenerated and new salt granules have to be added to replace the dissolved salt [4,11].

## V. CONCLUSION

Biogas produced from animal waste is a valuable energy resource. By combusting waste methane (biogas), a powerful greenhouse gas is eliminated that would otherwise be released. One of the most interesting utilization of Biomethane is as a vehicle fuel. The exchange of conventional fossil fuels with Biomethane can provide up to 80% less greenhouse gas discharges. The cost of Biomethane without incentives is generally not competitive compared with conventional natural gas. For increasing CV of biogas  $CO_2$  must be removed from it and thus obtaining Biomethane.Biomethane, due to its composition (97 to 99 % of  $CH_4$ ), delivers all the right qualities that natural gas has: it is one of the most environmental friendly combustion fuel, it is easy to carry and it can be utilized as fuel for all kinds of vehicle.

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