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Biogas Upgrading Technologies: A Mini-Review

Dolores Hidalgo[1,2*], Martín-Marroquín J M[1,2] and Coronas F[1,2]

[1]CARTIF Technology Centre, 47151 Boecillo, Valladolid, Spain

[2] ITAP Institute, University of Valladolid, 47011 Valladolid, Spain

Abstract

Biogas production is growing nowadays due to the increasing installation of anaerobic digesters for organic waste treatment, mainly in Europe, promoted by the current environmental legislation. Depending on the final use, different biogas treatment steps are necessary since the biogas is mainly composed of methane, but also carries carbon dioxide and other minority substances such us hydrogen sulfide, oxygen, nitrogen, water, ammonia, siloxanes and particulates.

In the last years, there is also an increasing demand for methane-rich biogas (biomethane) since the use of this product as vehicle fuel or its injection to the natural gas grid are becoming common practices. This supposes the biogas has to be first cleaned to remove the trace elements and then upgraded to remove CO_2 and adjust the calorific value to the foreseen uses. Biogas upgrading is thus a key issue today to enrich biomethane to produce a natural gas substitute.

This paper evaluates not only the biogas upgrading technologies that are commercially available today (as pressure swing adsorption, scrubbing using different types of absorbers, and gas permeation membranes), but also those that are under development trying to achieve lower investment and operational costs and, at the same time, higher methane content in the upgraded biogas (as membrane contactors, cryogenic upgrading, ecological lungs or in situ methane enrichment). They differ in functioning, the quality requirements for the incoming gas, the efficiency and their operational bottlenecks.

Process technology, binomial energy-economy and environmental impacts will also be aspects to take into consideration in this paper.

The review clearly suggests that water scrubbing technology, following by pressure swing adsorption methods are the most common, well established and conventional upgrading methods. On the other hand, membrane technology, especially membrane contactors, offers progressive research potential.

Keywords: Anaerobic Digestion; Biogas Cleaning; Biogas Upgrading; Biomethane; Landfill Gas; Natural Gas Substitute; Organic Waste

[*]Corresponding Author: Dolores Hidalgo, Parque Tecnológico de Boecillo, 205. 47151 Boecillo, Valladolid, Spain; Tel: +34 983 546504; Fax: +34 983 546521; E-mail: dolhid@cartif.es

Introduction

Biogas is produced during anaerobic digestion of organic substrates (in industrial anaerobic digesters, landfills, or directly in the nature) and it can be utilized as a renewable energy source substituting fossil fuels. The nature of the raw materials and the operational conditions used during anaerobic digestion, determine the chemical composition of the biogas [1, 2]. Raw biogas consists mainly of methane (CH_4, 40-75%) and carbon dioxide (CO_2, 15-60%). Trace amounts of other components such as water (H_2O, 5-10%), hydrogen sulfide (H_2S, 0.005-2%), siloxanes (0-0.02%), halogenated hydrocarbons (VOC, < 0.6%), ammonia (NH_3, <1%), oxygen (O_2, 0-1%), carbon monoxide (CO, <0.6%) and nitrogen (N_2, 0-2%) can be present and might be inconvenient when not removed [3]. Waste containing higher organic concentration leads to a biogas with higher methane gas content [4].

Depending on the final use foreseen for the biogas different treatments will be necessary [5, 6]. For some applications, such as its use in combine heat and power plants, biogas will have just to be cleaned to remove water and trace elements. But for other more demanded applications, where it is important to have a gas stream with high energy content, for example vehicle fuel or grid injection, the biogas will also need to be upgraded after cleaning.

Upgrading of biogas is defined as removal of carbon dioxide from the stream, resulting in an increased energy density since the concentration of methane is increased above 95% [5]. It is also possible to call the upgrading process as a process of improving biogas up to the level of substitute natural gas or compressed natural gas [7].

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Upgrading of biogas has gained attention in the last years due to rising fossil origin fuels prices and increasing targets for renewable fuel quotes in most of the developed countries. These technologies can also be used to help waste management facilities fulfil the European Commission Directive EC 31/1999 (European Commission, 1999) which mandates that all landfill gas must be captured and flared, though it is encouraged to use the gas in energy recovery [8]. As a consequence, new plants are continually being built around the world, as shown by the fact that the number of upgrading plants was around 401 in 2014 [9] when this number was only 100 in 2009 [5]. Most of these plants are located in Europe, being Germany and Sweden the forerunner countries.

Upgrading biogas generates new possibilities for its use, but also increases the costs associated with biogas production. It is therefore important to count on optimized upgrading processes in terms of low energy consumption and high efficiency able to avoid, or unless minimize, losses of methane in any stream leaving the upgrading treatment, what is important from both, economic and environmental perspective.

Several commercial techniques for biogas upgrading exist today and, on the other hand, new techniques are continuously under development. The main unit operation used are absorption (water scrubbing and amine scrubbing), adsorption (pressure swing adsorption) and membranes (gas permeation). A common factor of all of these techniques is that the removed CO_2 is normally released back into the atmosphere.

In some cases, if its quality is high enough, it can be used for industrial purposes such as increasing the CO_2 concentration for photosynthesis in greenhouses or for carbonation in food production [10].

The latest developments in biogas upgrading are reviewed in this paper, assessing their advantages and shortcomings and pointing out research tendencies.

Cleaning of Biogas

Biogas cleaning is the first necessary step in biomethane production. Although this paper is focused on biogas upgrading technologies, it is necessary first to have an overview of existing cleaning technologies that always will act as a pretreatment in the whole process to prevent corrosion and mechanical wear of the upgrading equipment itself. Table 1 shows the main characteristics and pros and cons of the most common biogas cleaning technologies.

Table 1: Pros and cons of biogas cleaning technologies. Adapted from Ryckebosh et al., [3].

Technology	**Pros**	**Cons**
Removal of water		
Condensation method Demister Cyclone Moister trap Water taps	Higher HC's dust and oil are removed Simple techniques Often used as pretreatment before other techniques	Atmospheric pressure: dew point minimum 1ºC Gas at higher pressure to reach lower dew point (minimal -18ºC) but freezing can occur
Adsorption dryer Silica Aluminum	High removal: dew point -10 till -20ºC Low operational cost Regeneration possible	More expensive investment : pressure 6-10 bar Dust and oil need to be removed in advance
Absorption with glycol	High removal: dew point -5 till -15ºC Higher HC's and dust are removed Not toxic or dangerous	More expensive investment: high pressure and 200ºC for regeneration Higher gas volumes (>500 m^3/h) to be economical
Absorption with hygroscopic salts	High removal efficiency Not toxic or dangerous	No regeneration done
Removal of H_2S		
Biological with O_2/air (in filter/scrubber/digester)	Cheap investment and exploitation: low electricity and heat	Concentration H_2S still high (100-300 cm^3/m^3)

	requirements, no extra chemicals or equipment required Simple operation and maintenance	Excess O_2/N_2 in biogas implies difficult upgrading or additional cleaning Overdosing air results in explosive mixture
$FeCl_3/FeCl_2/FeSO_4$ (in digester)	Cheap investment: storage tank and dosing pump Low electricity and heat requirements Simple operation and maintenance Compact technique H_2S not in biogas wire No air in biogas	Low efficiency (100-150 cm^3/m^3) Expensive operation (iron salt) Changes in pH/temp not beneficial for the digestion process Correct dosing is difficult
$Fe_2O_3/Fe(OH)_3$-bed	High removal efficiency: >99 %	Sensitive for water
Rust steel wool impregnated wood chips or pellets	Mercaptanes are also captured Cheap investment Simple	Expensive operation costs, Regeneration is exothermic: risk of ignition of chips Reaction surface reduced each cycle Released dust can be toxic
Absorption in water	H2S <15 cm^3/m^3 Cheap when water is available (not regenerative) CO_2 is also removed	Expensive operation: high pressure, low temperature Difficult technique Clogging of the absorption column possible
Chemical absorption NaOH $FeCl_3$	Low electricity requirement Smaller volume, less pumping, smaller vessels	Expensive investment and operation More difficult technique Not regenerative
Chemical absorption $Fe(OH)_3$ Fe-EDTA Cooab™	High removal efficiency: 95-100 % Cheap operation Small volume required Regenerative Low CH_4 losses	Difficult technique Regeneration through oxygenation CO_2 /H_2CO_3 (using EDTA) leads to precipitation Build up of thiosulfates from chelates g H_2S (using EDTA)
Membranes	Removal of >98 % is possible CO_2 is also removed	Expensive operation and maintenance Complex
Biological filter	High removal possible: >97 % Low operational cost	Extra H_2S-treatment to reach pipeline quality O_2/N_2 in biogas implies difficult and additional upgrading steps
Adsorption on activated carbon (Impregnated with KI 1-5 %)	High efficiency (H_2S <3 cm^3/m^3) High purification rate Low operation temperature	Expensive investment and operation CH_4 losses

	Compact technique High loading capacity	H_2O and O_2 needed to remove H_2S H_2O can occupy the binding places of H_2S Regeneration at 450ºC Residue present till 850ºC
Removal of siloxanes		
Absorption with organic solvents	High removal efficiency (97%)	Complete removal not possible
Absorption in strong acid	High removal efficiency (<95%)	Corrosion Environmental issues Hazardous chemicals
Absorption in strong base	Not used due to CO_3^{-2} precipitation.	Corrosion CO_3^{-2} precipitation Hazardous chemicals
Adsorption on silicagel	High removal efficiency (<95%) Higher removal capacity vs activated carbon (50% extra) Regeneration possible (95% desorption at 250ºC)	High pressure needed Moisture decreases efficiency
Adsorption on activated carbon	High removal efficiency (95%) Regeneration possible (desorption < desorption with silicagel at 250ºC)	High pressure needed (higher adsorption capacity) Moisture decreases removal efficiency
Cryogenic separation	High removal efficiency (99.3% at -70ºC) Removal of several impurities	Expensive investment and operation (high pressure and low temperature)

Removal of Water

Biogas always contains water vapor that have to be removed to avoid corrosion in pipelines and equipments. Water can be eliminated by physical separation (cooling and compression) and chemical drying (absorption and adsorption).

Refrigeration or cooling, preceded or not by compression, is the simplest way of removing excess water vapor. The condensed water can be then separated using demisters, cyclones or water traps [3]. Adsorption using alumina, silica or zeolites is the most common technique [11]. Also hygroscopic salts or triethylene glycol can be used.

Removal of H_2S

H_2S concentration in the biogas can be reduced by preventing its migration to biogas during digestion process or by treating the gas stream [12, 13]. In the first case, the addition of iron ions to the digester precipitates the iron sulfide that will leave the rector with the digestate. In the second case, adsorption on activated carbon, iron oxide or hydroxide [14], washing with sodium hydroxide or biological treatment [15] are all well-known options. H_2S can be also separated from the biogas by leading the gas by a semi-permeable membrane [16].

Removal of Siloxanes

The removal of siloxanes is mainly necessary for landfill gas and biogas originated by digestion of municipal waste since its origin is in cosmetics, pharmaceuticals and anti-foam products. Siloxanes can be removed by cryogenic separation (cooling), adsorption on activated carbon, silica gel or activated aluminum [17] and physical absorption in liquid mixtures of organic solvents [18], strong base or strong acid.

Removal of oxygen / air

Oxygen and nitrogen can be present in the biogas if air has intruded the digester or the gas collector (quite often in landfills). Oxygen and nitrogen removal is expensive because membranes, activated carbon, pressure swing adsorption or catalytic oxidation [19] should be use. Air introduction prevention by careful monitoring is the best option [3].

Removal of Ammonia

Ammonia is present in the biogas when the original substrate is rich in nitrogen. It could be removed from the biogas by a washing process with diluted acid or with activated carbon, but it is usually separated when the biogas is upgraded, without a separate cleaning step.

Removal of Particulates

Mechanical filters are commonly used to separate particulates from the biogas.

Removal of Halogenated Carbon Hydrates

Halogenated carbon hydrates are typically found in landfill gas and they can be removed with activated carbon.

Biogas Upgrading Mature Technologies

Depending of its intended use biomethane consists typically of 97-99% methane and 1-3% CO_2. Several technologies for biogas upgrading to produce biomethane are commercially available [20]. Some of them are described below and reviewed in terms of recent developments.

Absorption

Carbon dioxide is more soluble than methane and the absorption technique is based on this fact. Absorption is usually carried out in a packed absorption column. Biogas is introduced to the bottom of the column and flows up. The selected liquid enters the column at the top and flows downward, so that mass transfer occurs in a counter-flow way. Purified biogas (biomethane) leaves column at the top and liquid saturated with CO_2 is let out at the bottom. Examples of the absorption technology using different types of absorbents are water scrubbing, organic physical scrubbing and chemical scrubbing.

Water Scrubbing

This process also enables to remove simultaneously H_2S, hence pretreatment for H_2S is not necessary according to Beil and Hoffstede [21].

70% reduction of CO_2 at 282.2 K and using a L/G molar ratio of 868 was achieved by Pirola et al., [22] operating at low pressure. Product gas with a methane contents ranging from 83.0% to 92.1% was achieved by Rasi et al., [23] with differing process parameters using a high pressure water absorption column. Other authors reports separation efficiency values of this technology up to 95% 3, [3, 24].

Water scrubbing is the most common upgrading technique and plants are commercially available from several suppliers in a broad range of capacities [5].

Organic Physical Scrubbing

Instead of water, polyethylene glycol and other ionic solvents can also be used. H_2S, CO_2 and H_2O can be absorbed together, since they have a higher solubility in polyethylene glycol than CH_4 [25]. 90% is the maximum CH_4 yield reported for biomethane with this method [7].

Chemical Scrubbing

The most employed solvents in chemical scrubbing are amides (monoethanolamine, diethanolamine or diglycolamine), which in comparison to water can dissolve considerably much more CO_2 per unit volume [7]. This method requires previous H_2S removal sand later amino solution regeneration, usually by heating.

Adsorption

Pressure Swing Adsorption (PSA) separates CO_2 from the biogas using a sieve filled with an adsorbent (activated carbon, silicagel, alumina, zeolite, etc.) under elevated pressure. The concentration of CH_4 can be more than 98% after this upgrading process [26]. PSA can separate CO_2, but also O_2, H_2S and N_2, from CH_4. Effectiveness of simultaneous H_2S and CO_2 capture depends on water content on the biogas. It is usually better to separate the H_2S previously to avoid any potential destruction of the adsorbent when water is present [27].

A variation of PSA is vacuum swing adsorption, based on the same principles.

Membranes

Conventional membrane-based gas separation is commonly done by gas permeation, with a gas phase at both sides of the membrane [28]. Gas-gas separation is a high-pressure system. Membranes can be made of silicone rubber, cellulose acetate or polyimides. Usually membranes are in the form of hollow fibers bundled together.

The raw gas can be purified to maximum 92% CH_4 in one step. When two or three steps are used, a gas with 96% or more CH_4 is achieved [5]. The appropriate cleaning is required for any undesirable compound in biogas [29] and the intensity of the cleaning depends on membrane material [28].

Biogas Upgrading Novel Technologies

At the same time that matures biogas upgrading technologies are looking for continuous improvements, emerging technologies are also being developed. Some of them are mentioned below.

Membrane Contactors

Gas-liquid membrane-based absorption systems, also called membrane contactors, have been traditionally used in the food industry for liquid gasification or degasification processes but today they are being also applied in biogas upgrading at low pressure [30-33]. Hydrophobic membranes separate the gas from the liquid phase. Specific molecules from a gas stream diffuse through the membrane and are absorbed on the other side by the liquid flowing in counter current.

The removal of CO_2, carried out with an amine solution, is very efficient: biogas with 55% CH_4 can be upgraded to more than 96% CH_4 in one step. Water as liquid phase gives lower purification rates [34].

Cryogenic Upgrading

Since CH_4, CO_2 and other impurities liquefy/ sublimate in different temperature/ pressure areas, it is possible to produce biomethane by cooling and compressing the biogas [3]. The biogas has to be first dried, to avoid freezing in the following steps, then compressed and finally cooled. The CO_2 condenses and can be removed. Cooling usually takes place in several steps in order to remove the different gases in the biogas individually and to optimize the energy recovery [5].

This process is interesting when the final product is liquid biomethane. In this case cooling for purification is synergic to further cooling to produce liquid biomethane [35].

Biological Methane Enrichment

Biotechnologies can offer a low cost and environmentally friendly alternative to physical/chemical biogas upgrading. Thus, biotechnologies such as H_2-based chemoautrophic CO_2 bioconversion to CH_4, microalgae-based CO_2 fixation, enzymatic CO_2 dissolution, fermentative CO_2 reduction and digestion with in situ CO_2 desorption have consistently shown CO_2 removals of 80–100 % and CH_4 purities of 88–100 %, while allowing the conversion of CO_2 into valuable bio-products and even a simultaneous H_2S removal [36].

Xu et al., [37] studied the formation of hydrogenotrophic methanogenic granules specialized for high-rate hydrogenotrophic methanogenesis and biogas upgrading. According to these authors, glucose-acclimated granules seemed to perform in *in situ* biogas upgrading systems.

Ecological Lung

The enzyme carboanhydrase (present, for example, in human blood) can be used to dissolve carbon dioxide from biogas and thereby remove it from the gas, mimicking its activity in human lungs [38]. This is a promising research line that needs further development.

Comparison between the different mature technologies for biogas upgrading

From the energy point of view, water scrubbing and pressure swing adsorption appears to be the less energy demanding technologies when comparing with membranes separation and chemical and physical scrubbing, including here energy demanded for solvent regeneration [39].

Niesner [7] made a comparative analysis of biogas upgrading technologies in reference to investment and operating costs. These authors evidenced the lowest operational cost related to water scrubbing followed by membranes separation, whereas pressure swing adsorption and chemical and physical scrubbing were the most demanding. The lowest investments costs belonged to water scrubbing.

Analyzing the environmental impact of these technologies in relation to the methane losses occurred during the processes the lowest methane losses are reported for chemical absorption and the highest ones relate to membranes [7] unless a multistage membranes system is considered [40].

Table 2 shows the main characteristics and pros and cons of the most common biogas upgrading technologies.

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Table 2: Pros and cons of biogas upgrading technologies. Adapted from Ryckebosh et al., [3].

Technology	**Pros**	**Cons**
Absorption with water	High efficiency (>97% CH_4) Simultaneous removal of H_2S When H2S < 300 cm^3/m^3 Easy in operation Capacity is adjustable by changing pressure or temperature Regeneration possible Low CH_4 losses (<2%) Tolerant for impurities	Expensive investment Expensive operation Clogging due to bacterial growth Foaming possible Low flexibility toward variation of input gas
Absorption (physical) with ionic solvents (as polyethylene glycol)	High efficiency (>97% CH_4) Simultaneous removal of organic S components, H_2S, NH_3, HCN and H_2O Energetic more favorable than water Regenerative Low CH_4 losses	Expensive investment Expensive operation Difficult in operation Incomplete regeneration when stripping/vacuum (boiling required) Reduced operation when dilution of absorber with water
Absorption (chemical) with amines	High efficiency (>99% CH_4) Cheap operation Regenerative More CO_2 dissolved per unit of volume (compared to water) Very low CH_4 losses (<0.1%)	Expensive investment Heat required for regeneration Corrosion Decomposition and poisoning of the amines by O_2 or other chemicals Precipitation of salts Foaming possible
Pressure swing adsorption Activated carbon Zeolites Alumina silicates	Highly efficient (95-98% CH_4) H_2S is removed Low energy use: high pressure, but regenerative Compact technique Also for small capacities Tolerant to impurities	Expensive investment Expensive operation Extensive process control needed CH_4 losses when malfunctioning of valves
Membranes Gas permeation Contactors	H_2S and H_2O are removed Simple construction Simple operation High reliability Small gas flows treated without proportional increase of costs Gas/gas Removal efficiency: <92% CH_4 (1 step) or > 96% CH_4	Low membrane selectivity: compromise between purity of CH_4 and amount of upgraded biogas Multiple steps required (modular system) to reach high purity CH_4 losses Little operational experience

	H_2O is removed Gas/liquid Removal efficiency: > 96% CH_4 Cheap investment and operation Pure CO_2 can be obtained	
Cryogenic separation	90-98% CH_4 can be reached CO_2 and CH_4 in high purity Low extra energy cost to reach liquid biomethane	Expensive investment and operation CO_2 can remain in the CH_4
Biological removal	Removal of H_2S and CO_2 Enrichment of CH_4 No unwanted end products	Addition of H_2 Experimental, not at large scale

Concluding Remarks

Nowadays there are more than 300 biogas upgrading plants only in Europe and new plants are continually being built around the world due to the increasing demand on biomethane. Five main technologies are used in these plants: water scrubbing, the most popular with almost 40% share, followed by pressure swing adsorption, chemical scrubbing, physical scrubbing and membranes separation in this order. All of them are proven technologies with potential research on process optimization by means of technologies combination, process configuration or membranes and adsorbents development. In this sense, some emerging technologies, as it is the case of membrane contactors, are offering promising results.

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