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# Biomethane Production and Applications

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## Abstract

Biomethane production generally involves the cleaning to remove minor unwanted components of biogases such as hydrogen sulfide ( $\text{H}_2\text{S}$ ) and moisture ( $\text{H}_2\text{O}$ ) and upgrading in a process that involves the removal of carbon dioxide ( $\text{CO}_2$ ) to increase the concentration of  $\text{CH}_4$  to 95–99% and reduce  $\text{CO}_2$  concentration to 1–5%, with little or no hydrogen sulfide ( $\text{H}_2\text{S}$ ). Biomethane gas is a flexible and easy to store fuel having similar properties and applications as natural gas with no need to modify the settings for natural gas devices and equipment. Biomethane can be used for industrial and domestic applications ranging from thermal and power generation and feedstock for processes like the Fischer-Tropsch (FT) for fuel manufacturer and direct power generation in hydrogen or biogas fuel cells like production of green hydrogen. Therefore, biomethane promises to play a leading role in the energy transition through hydrogen, electricity, and other renewable fuels production. Biomethane production by biogas upgrading methods include the pressure swing adsorption, which has an option of temperature swing adsorption, absorption techniques based on amine, membrane separation, cryogenic separation, and biological separation. The technology adopted may depend on factors such as costs, quality of products, location, and technology maturity and requirements.

**Keywords:** biogas, biogas cleaning, biogas purification, biomethane, anaerobic digestion, biogas enrichment

## 1. Introduction

The production of biogas has been growing and so is the demand for upgraded biogas for applications like vehicle fuel or injection to the natural gas grid. Biogas has to be upgraded to facilitate efficient use in these applications by removal of carbon dioxide which is inert yet it constitutes a significant portion of raw biogas at the expense of methane [1, 2]. Biogas is a mixture of gases produced by action of microorganisms through anaerobic digestion which is a complex process made up of four stages i.e.: hydrolysis, acidogenesis, acetogenesis and methanogenesis leading to biogas. The composition of biogas is influenced by the type of feedstock used and anaerobic digestion process control [3]. Other than production in anaerobic digesters, biogas can also be produced from landfills and through biomass thermal pyrolysis and

gasification processes. The mixture produced generally consists of 30–75% methane ( $\text{CH}_4$ ), 25–55% carbon dioxide ( $\text{CO}_2$ ) and other constituents or impurities like hydrogen ( $\text{H}_2$ ), oxygen ( $\text{O}_2$ ), nitrogen ( $\text{N}_2$ ), hydrogen sulfide ( $\text{H}_2\text{S}$ ), water ( $\text{H}_2\text{O}$ ) and ammonia ( $\text{NH}_3$ ), dust particles, siloxanes, aromatic and halogenated compounds, which are often in tiny quantities [4–6].

Anaerobic digestion is a sustainable process used for simultaneous treatment and production of biogas energy resource. Biogas is a renewable energy resource produced by anaerobic digestion and has methane as the main component with impurities like hydrogen sulfide, carbon dioxide water vapor, siloxanes, hydrocarbons, oxygen, ammonia, carbon monoxide and nitrogen. The energy content of biogas is reduced by the impurities while others cause operational challenges to combustion systems like corrosion [7, 8]. This makes it important to apply different technologies to remove these harmful and undesirable impurities [9]. There are many established and developing physicochemical technologies for biogas operation and maintenance costs, energy requirements, efficiency of removal and other parameters [9, 10].

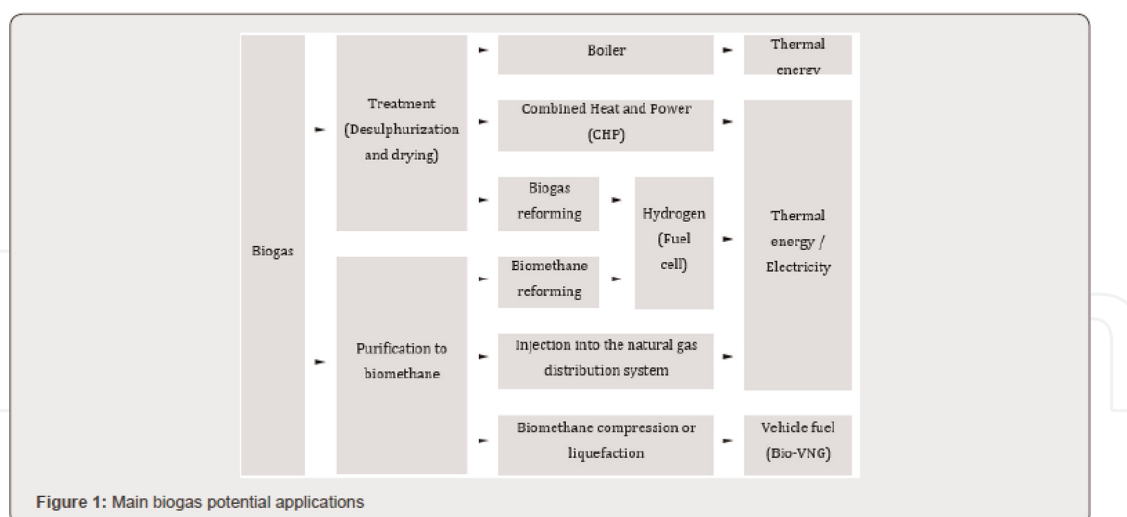
Biomethane or upgraded biogas is made by qualitative processing of raw biogas through several steps to remove impurities mainly carbon dioxide. Biomethane as a fuel provides new opportunities at different levels for the society. In this chapter, various methods of biomethane production are described and compared [11, 12]. There is growing interest in the use of biomethane as a renewable substitute of natural gas in applications like transport fuel which has created demand for biogas upgrading. This chapter is a critical review that summarizes the state-of-the-art technologies used in cleaning and upgrading. Covered in the review are the description of biomethane production methodologies, scientific and technical outcomes related to them, bio-methanation efficiency, challenges and feasibility of the technologies [8, 13].

Biogas is a renewable energy carrier that can be exploited directly as a fuel or as a feedstock for production of hydrogen or synthesis gas. The main constituents of biogas are carbon dioxide ( $\text{CO}_2$ ) and Methane ( $\text{CH}_4$ ), but there are quantities contaminants like such as hydrogen sulfide ( $\text{H}_2\text{S}$ ), ammonia ( $\text{NH}_3$ ), moisture and siloxanes whose existence and composition is a function of the source of biogas like landfills, anaerobic fermentation of manure [14, 15]. The contaminants have the following undesirable effects to biogas applications a fuel;

- i. They can be detrimental to any biogas thermal or thermo-catalytic conversion device (e.g., corrosion, erosion, fouling); and
- ii. Generation harmful environmental emissions. It is therefore important to include biogas purification steps upstream of its final use processes [9].

Other than methane which is the main energy source in biogas, raw biogas has impurities that are noncombustible while others are harmful to the equipment and environment and should therefore be removed to make it suitable for wide range of applications in heat and power generation [8]. The treatment and purification/upgrading pathways and applications are summarized in **Figure 1** below.

From **Figure 1**, it is shown that biogas treatment mainly involves desulfurization and drying of raw biogas. Making it an ideal feedstock for applications like boiler fuel, cogeneration (CHP) and biogas reforming for production of hydrogen and other fuels and as a fuel for direct combustion processes like boilers for heat and power production.



**Figure 1.**  
 Main biogas potential applications.

Purification on the other hand leads to the production of biomethane which is an ideal feedstock for biomethane reforming for hydrogen, production of compressed liquid bio-natural gas and liquefied bio natural gas as well a direct substitute for natural gas fuel applications including injection to natural gas pipelines [2, 6, 16].

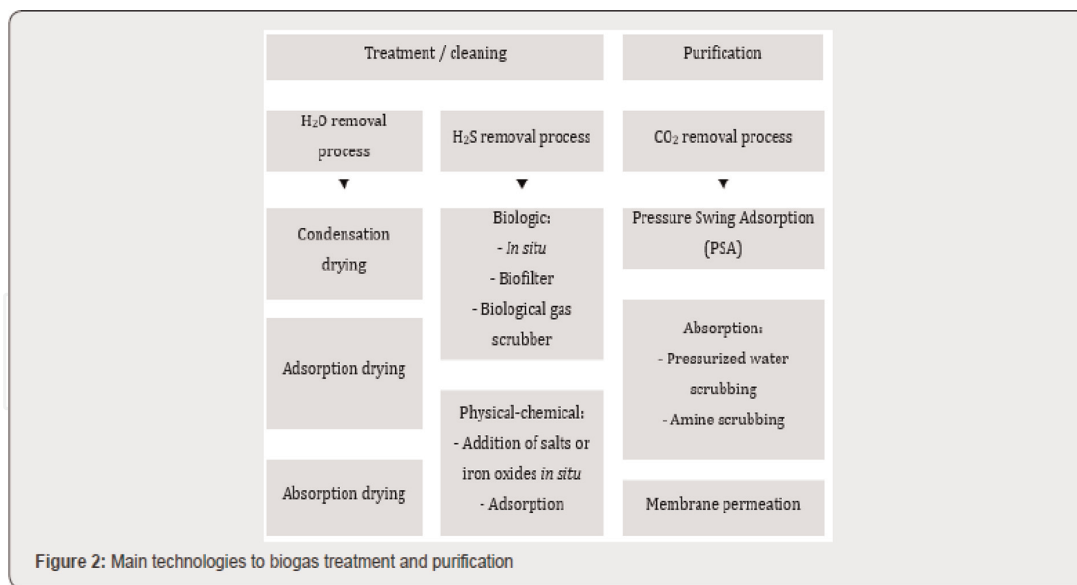
## 2. Technologies for biogas cleaning, drying and purification

Controlling the level of impurities in biogas is essential for success of its recovery. The implementation of treatment and purification technologies must consider the requirements of each specific application of biogas. These technologies aim to adjust the calorific value and remove contaminants that affect the quality of biogas and the useful life of the equipment. The most demanding techniques aim to purify biogas to obtain biomethane. Currently, different techniques that allow the treatment and purification of biogas are commercially available [12, 17, 18].

There are many technologies available at commercial and laboratory scale for the treatment and purification of biogas. These methods include condensation, absorption and adsorption processes for raw biogas. Hydrogen sulfide ( $H_2S$ ) removal can be done by biological in situ, ex situ, biofilter and biological gas scrubbing techniques. Purification which involves mainly removal of  $CO_2$  can be done by absorption i.e. amine and pressurized water scrubbing and amine scrubbing, membrane permeation and cryogenic method [2, 19–21]. **Figure 2** summarizes the various methods for treatment/cleaning and purification of raw biogas.

From **Figure 2**, it is noted that treatment or cleaning mainly involves removal of moisture and  $H_2S$  while upgrading or purifications mainly targets the  $CO_2$  for removal. The choice of biogas treatment and purification technology is a function of factors like the amount of biogas produced, its composition, the level of purification required, and process costs in terms of capital, energy consumption and operational expenditure (CAPEX and OPEX). In biomethane requirements, a combination of processes is used, as no technology can remove all contaminants from biogas [1, 2].

There are simpler and cheaper technologies available for treatment of biogas with the objective of cleaning biogas for sensitive applications [2]. The degree of biogas



**Figure 2.**  
Main technologies to biogas treatment and purification.

treatment depends on the intended application and the initial composition of raw biogas. Most common treatment involves the removal of H<sub>2</sub>O and H<sub>2</sub>S [4, 12].

## 2.1 Moisture (H<sub>2</sub>O) removal

Biogas may contain moisture concentration of between 3 to 10%. Water removal is usually carried out at an early stage of the treatment, to protect the downstream equipment against corrosion and allow the biogas feedstock to fulfill the requirements of subsequent purification steps. **Table 1** is a summary of advantages and disadvantages of processes for H<sub>2</sub>O removal from biogas.

From **Table 1**, the strengths and weaknesses of moisture removal techniques from biogas are presented. Moisture can be removed from raw biogas by condensation, adsorption and absorption. Condensation has high energy consumption and is expensive in terms of investment and maintenance, but the process is simple and effectively removes hydrocarbons and oil particles. The adsorption process has low operating costs, has high removal rate and the adsorbent materials are regenerated as main advantages, although it has high investment costs and requires the prior removal of oil. The Absorption process of moisture removal from raw biogas effectively eliminates hydrocarbons, and like the adsorption method has high removal rates and absorption materials can be regenerated. However, the method has high investment costs, making it only viable at high biogas flow rates. The regeneration of adsorption materials is done at high pressure and temperature making operations and maintenance expensive [7, 22].

### i. Condensation

In condensation, separation of steam and water from biogas is affected through by use of cyclone separators. Condensation of water can be improved further by cooling biogas below the dew point of the gas. For this purpose, cooling pipes are installed with a slope and a purging system to collect the condensate collected [1, 6].

Process	Advantages	Disadvantages
Condensation drying	<ul style="list-style-type: none"> <li>• Simple process, suitable for any biogas flow;</li> <li>• Elimination of hydrocarbons and oils particles;</li> <li>• Application as pre-treatment in all systems.</li> </ul>	<ul style="list-style-type: none"> <li>• High energy consumption;</li> <li>• Requires installation of long tubes with slope and freeze-resistant;</li> <li>• High investment and maintenance costs.</li> </ul>
Adsorption drying	<ul style="list-style-type: none"> <li>• Adsorbent materials can be regenerated;</li> <li>• High removal rate, which allows the process to be applied to any type of biogas use;</li> <li>• Low operating cost.</li> </ul>	<ul style="list-style-type: none"> <li>• Requires prior removal of particles and oil;</li> <li>• High investment cost;</li> <li>• Suitable for small or medium biogas flows.</li> </ul>
Absorption drying	<ul style="list-style-type: none"> <li>• Materials can be regenerated;</li> <li>• High removal rate, which allows the process to be applied to any type of biogas use;</li> <li>• Elimination of hydrocarbons particles.</li> </ul>	<ul style="list-style-type: none"> <li>• High investment cost;</li> <li>• Economic viability only for high biogas flow rates;</li> <li>• Absorbent material regeneration carried out at high pressure and temperature.</li> </ul>

**Table 1.**  
*Advantages and disadvantages of the main H<sub>2</sub>O removal processes from biogas.*

### ii. Adsorption

Cylindrical reactors containing adsorbent materials are used in adsorption drying process. Commonly used adsorption materials are silica gel, activated carbon, aluminum oxides, magnesium oxides and zeolites. The adsorption materials are installed in a fixed bed, that can be exchanged and regenerated when it gets saturated. The system can also operate alternately with two columns, where one has the adsorption material at room temperature and pressure between 6 bars and 10 bars, and the other column is a standby unit where regeneration is done [1, 23].

### iii. Absorption

In absorption drying biogas flows through an absorption tower, in countercurrent with a solution of glycol or other hygroscopic materials. In the process moisture or steam and hydrocarbons are chemically absorbed. This method was originally used to dry natural gas. Absorption operations take place at high pressure of between 20 and 40 bars, while regeneration occurs at around 200°C [1, 2].

## 2.2 H<sub>2</sub>S removal process

Hydrogen sulfide is (H<sub>2</sub>S) is a gaseous chemical found in many fuel gases, biogas, natural gas, syngas, coke oven gas, landfill gas, refinery gas, and wastewater steams among others etc. [24]. Hydrogen sulfide is flammable, toxic, and extremely hazardous and should therefore be captured and removed from biogas. The challenge and need to H<sub>2</sub>S has led to the development of different materials and methods over the years for its removal. Some alkanolamines are used as absorbents and while metal oxides are used as adsorbents [17, 18, 25]. The removal of H<sub>2</sub>S from fuels is imperative in terms of both safety and economics. The main challenge of H<sub>2</sub>S in application of

biogas is its inherent tendency to form an acidic solution in the presence of water which leads to pipelines and equipment corrosion [8, 25].

H<sub>2</sub>S removing process is divided into two broad levels or categories on the basis of intended application for produced biogas. Hydrogen sulfide (H<sub>2</sub>S) removal is normally done by means of biological or physical- chemical processes, and can be classified as external or internal depending on whether it is done outside or inside the anaerobic bio-digester. The first level involves production of biogas with, H<sub>2</sub>S concentration of below 500 ppm, and can reach as low as 100 ppm. The second level involves reduction in H<sub>2</sub>S concentrations less than 0.005 ppm, which are typical specifications and requirements for biomethane gas [4, 24].

### *2.2.1 Biological removal of H<sub>2</sub>S*

There are various established methods for biological removal of H<sub>2</sub>S from raw biogas. They include in situ, biofilter, biological scrubber, and ex-situ techniques.

#### *i. In Situ methane enrichment*

Situ is a biotechnology based on the direct injection of pure air or oxygen and in the process, the bacteria that oxidize H<sub>2</sub>S develop with the presence oxygen, leading to the biological removal process of H<sub>2</sub>S, to produce sulfur (S) which leaves the digester via the digested. The microorganisms are widely found in the anaerobic environment present in bio-digesters [4]. In situ desorption technology is yet to be fully developed even though it has been around for over 20 years. In-situ is based on the greater solubility of CO<sub>2</sub> over CH<sub>4</sub> in water. The process set up includes an anaerobic digester linked or connected to an external desorption unit. Sludge transported to an aerated desorption column from the digester. Nitrogen or air flowing in counter-current mode and dissolves the CO<sub>2</sub> from the sludge in the desorption unit. The sludge desorbed sludge is pumped back into the digester to reabsorb more CO<sub>2</sub>, and the sludge as the sludge is continuously recycled in the desorption column. It is possible to strip out H<sub>2</sub>S with dissolved CH<sub>4</sub> and CO<sub>2</sub> from the recirculating sludge by applying large quantities of air or N<sub>2</sub>, causing reduction in the H<sub>2</sub>S and CO<sub>2</sub> concentration [26, 27].

Therefore, in Situ is a biotechnology which works by direct injection of pure air or oxygen and causing bacteria to oxidize H<sub>2</sub>S leading to biological removal H<sub>2</sub>S, to produce sulfur (S) which exits the digester via the digested. These microorganisms are widely present in anaerobic environment in bio-digesters [4].

#### *ii. Biofilter*

In the bio filter technology biogas is passed through a column having a synthetic material, in the form of a biofilm. The parallel or countercurrent flow system is used to maintain the humidity and nutrients, that are essential for the microorganisms that degrade of H<sub>2</sub>S [4]. The purification system consists of a bioreactor where sulfur-oxidizing bacteria like the Thiobacillus, Pseudomonas and Acidithiobacillus are immobilized on a carrier. In the process, moisturized biogas is injected from the bottom of the bio filter and forced through a moist, packed bed with microbial

biofilm which purifies biogas. The bed material is used to supply nutrients or nutrient solution added from the top occasionally. Oxygen whose concentration is 5–10% of volume is supplied by injecting air directly into the gas stream [3].

### iii. Biological gas scrubber

In biological gas scrubber, a two-stage system is used to remove H<sub>2</sub>S. In the first stage H<sub>2</sub>S scrubbing column, applies sodium hydroxide solution while activated sludge is used in the second stage which is injected with air, because the microorganisms used are aerobic, leading to the solution regeneration [4]. Bio scrubber system is applied in the removal of compounds like ammonia, amines, hydrocarbons, hydrogen sulfide and odorous contaminants. Bio scrubbing system consists of two reactor units with the first reactor as absorption tower where the pollutants are absorbed in a liquid phase before it goes to the second reactor which is activated sludge. Degradation occurs in the activated sludge reactor where microorganisms like *Thiobacillus* and *Thioalkalivibrio*) grow in suspended flocks. The effluent generated is recirculated back to the absorption tower. In the removal of H<sub>2</sub>S, a sedimentation tank is installed after the second reactor for collection of elemental Sulfur with O<sub>2</sub> being used as the oxidant. Optimal microbial growth and activity are maintained addition of oxygen, nutrients and pH regulation together with continual purging of by-products and excess biomass out of the system [3].

### iv. Ex-situ

Ex- situ biogas cleaning and upgradation relies on supply of carbon dioxide from external sources and hydrogen in an anaerobic reactor, which eventually contributes to their conversion to methane. The ability of ex situ process to manage high concentrations of influent gases, reduces retention time to about 1 hour leading to a smaller device for upgrading. Depending mainly on the reactor used, the ex-situ technology can produce methane with final purity of 79–98%, the main challenge facing this technology is low gas–liquid mass transfer rate [26]. Therefore, ex-situ is more of an upgrading than cleaning method although it can do both by design. The advantages and disadvantages of the biological H<sub>2</sub>S removal processes are summarized in **Table 2** below.

From **Table 2**, the three discussed biological methods for H<sub>2</sub>S removal have significant differences in terms of use of chemicals, operation and maintenance costs and product quality. Biofilter and biological gas scrubber techniques need external oxygen injection. The main advantage of in-situ method is that it has low investments and maintenance costs and does not require chemicals.

#### 2.2.2 Physical-chemical removal of H<sub>2</sub>S

These techniques involve use of salts or iron oxides or sulfide precipitation is used to remove H<sub>2</sub>S inside the digester. Iron oxides or salts are added that react with H<sub>2</sub>S, to produce non-soluble compounds, like iron sulfides, that precipitate and are removed



Process	Advantages	Disadvantages
In situ	<ul style="list-style-type: none"> <li>• A simple process</li> <li>• Low investment and maintenance cost</li> <li>• Does not need chemicals</li> </ul>	<ul style="list-style-type: none"> <li>• Oxygen injection may affect the anaerobic digestion and can oxidize methane</li> <li>• Potentially explosive mixtures may occur</li> <li>• Cannot achieve biomethane purification level</li> </ul>
Biofilter	<ul style="list-style-type: none"> <li>• Enables the removal of ammonia</li> <li>• Does not require chemicals</li> <li>• The injection of oxygen is external to the digester hence no negative effect to the digestion</li> </ul>	<ul style="list-style-type: none"> <li>• Requires nutrients renewal hence more operation and maintenance cost</li> <li>• Only suitable for small biogas flows</li> <li>• Injection of air at high levels through the biofilter is not suitable for biomethane production</li> </ul>
Biological gas scrubber	<ul style="list-style-type: none"> <li>• Oxygen introduced is external to the process and has no negative impact to the digestion</li> <li>• Good for high biogas flow rates</li> <li>• The process can attain purity requirement for biomethane</li> </ul>	<ul style="list-style-type: none"> <li>• Uses chemicals</li> <li>• High operations and maintenance costs</li> <li>• The process needs fresh water introduction</li> </ul>
Ex situ	<ul style="list-style-type: none"> <li>• Can be used to attain high methane purity levels needed for biomethane</li> <li>• Requires smaller devices due to lower retention time</li> </ul>	<ul style="list-style-type: none"> <li>• The process relies on carbon dioxide supplied from external sources hence an extra cost</li> <li>• The process has low gas-liquid mass transfer rate</li> </ul>

**Table 2.**  
*Biological process for removal of H<sub>2</sub>S (summary by the author).*

together with effluents from the biodigester. Through direct dosing, chemicals are added to a reactor installed in the biogas line. The H<sub>2</sub>S adsorption process is achieved by retention in a solid form having a large surface area or in materials with high internal porosity. Activated carbon and iron oxides are the common adsorbent materials applied in the process. Activated carbon enable production of low concentrations of H<sub>2</sub>S based on the catalytic oxidation of H<sub>2</sub>S on the surface of the activated carbon, which is easy to impregnate with catalysts that speed up the reaction and improve the process capacity [4]. The advantages and disadvantages of each physical- chemical H<sub>2</sub>S removal process is summarized in **Table 3**.

From **Table 3**, it is noted that there are broadly two main approaches of physical-chemical methods of H<sub>2</sub>S i.e. addition of salts or iron oxides in situ and adsorption. The addition of chemicals does not be used to attain biomethane quality although the process is simple and cheap. The adsorption method is moderate in cost, attains high removal rate for H<sub>2</sub>S to attain biomethane level of purity but incurs high energy costs and high operation and maintenance cost related to replacement of the adsorbent.

### 3. Biogas upgrading methods

Production of biogas and use has several environmental, social and economic benefits. It is a source of renewable energy, and its production is also considered as a manure production factory. Biomethane has wider industrial applications hence biogas up-gradation is desirable [2]. The main drivers of biogas up-gradation is rapid increment in the price of fossil fuels and growing concerns over global climate change due to greenhouse gas emissions. Biomethane has opened a new window for the replacement of natural gas from the energy mix. There are multiple biogas

Process	Advantages	Disadvantages
Addition of salts or Iron oxides in situ	<ul style="list-style-type: none"> <li>✓ The process is simple and cheaper</li> <li>✓ No need for oxygen injection</li> <li>✓ Low maintenance costs</li> </ul>	<ul style="list-style-type: none"> <li>✓ Cannot attain biomethane purity levels</li> <li>✓ Forms precipitates within the digester and hence handling issues</li> <li>✓ The process uses chemicals</li> </ul>
Adsorption	<ul style="list-style-type: none"> <li>✓ Has moderate investment costs requirements</li> <li>✓ Has got high rate of removal</li> <li>✓ Can attain biomethane quality and standards</li> <li>✓ Oxygen injection does not affect the use of doped activated carbon</li> </ul>	<ul style="list-style-type: none"> <li>✓ The process has high energy consumption</li> <li>✓ Extra cost incurred to renew absorbent in form of operation and maintenance</li> <li>✓ Extracted sulfur cannot be used</li> </ul>

**Table 3.** *Advantages and disadvantages of physical-chemical methods for H<sub>2</sub>S removal (summary by the author).*

up-gradation technologies which are available on commercial scale while others are still developing and are at laboratory scale. The technologies that are widely accepted have attained prominence on their operational efficiency and reliability, merits and demerits and future outlook [10, 18, 21].

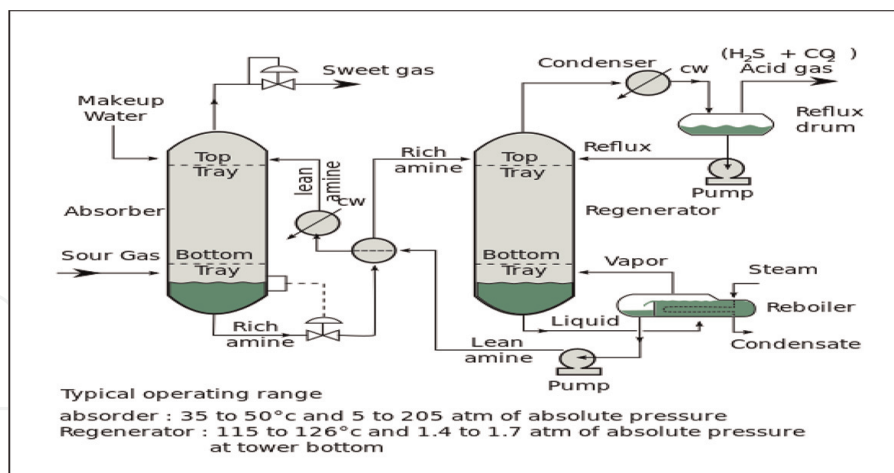
Biomethane production requires more complex and expensive techniques compared to biogas treatment methods aimed at attaining high degree of purity for biogas. Biogas upgrading combines biogas treatment and purification processes to remove other gases from biogas, thence separate methane (CH<sub>4</sub>) and effectively increasing its heating value. Purification of biogas involves removal of carbon dioxide which is mandatory for biomethane to substitute natural gas in pipeline system for natural gas distribution and use as a fuel for applications like vehicle fuel [28].

### 3.1 Chemical scrubbing

Chemical scrubbing systems use aqueous organic or inorganic compounds to bind the CO<sub>2</sub> or H<sub>2</sub>S molecules existing in biogas. The most commonly used scrubbing systems use organic compounds, namely aqueous amine solutions like diethanolamine (DEA), monoethanolamine (MEA) or methyl diethanolamine (MDEA). Most scrubbing systems using amine solutions have an absorber unit maintained between biogas pressure of 1–2 bars which is injected to the tank bottom with amine solution flowing from the top and a stripper. The solute and solvent (CO<sub>2</sub>) undergo a reversible exothermic chemical reaction with the product amine solution which is rich in CO<sub>2</sub> and H<sub>2</sub>S which proceeds to the stripper for regeneration operating at a pressure of 1.5–3 bars and temperature of 120–160°C [6].

The heat in the stripper disrupts the chemical bonds formed in absorption phase and which creates steam having CO<sub>2</sub>. Upon cooling this steam, the CO<sub>2</sub> is released while the condensate is recirculated back to the stripping column. Some commercial systems can cope with biogas with H<sub>2</sub>S content of up to 300 ppm/v, but H<sub>2</sub>S poisons the amine, cause corrosion and increase the system energy requirements hence the need to remove H<sub>2</sub>S before amine scrubbing [3, 25].

The advantages of the chemical scrubbing using amine solutions include high selectivity of the amines by CO<sub>2</sub> and the substantial extraction compared to other methods e.g. two times more CO<sub>2</sub> per unit volume is absorbed compared with water.



**Figure 3.**  
 Chemical scrubbing with amine solution [29].

The process has significantly low energy requirements mainly because of exothermic reactions and low process pressure operations i.e. 1–2 bar for absorption column and 1.5–3 bar in the stripping column. The draw backs include high energy requirements for solvent regeneration, expensive amine solvents and losses of solvents to evaporation which increase operation costs [3]. **Figure 3** shows the process of chemical scrubbing using amine solution.

From **Figure 3**, it is noted that the main system elements for the chemical scrubbing with amine solution are the adsorption column, heater, a cooler, a stripping column, and a heating medium for the stripping column which may be hot water, oil or steam.

In chemical scrubbing (CSC), there is reversible reactions between absorbed substances and solvent used. The commonly used biogas upgrading absorption solutions is based on amines i.e. methyl diethanolamine diethanolamine, monoethanolamine, and piperazine. For amine scrubber an absorber tank is used in which carbon dioxide is absorbed from the biogas operating at 20–65°C and 1–2 bar, then followed by a stripper where carbon dioxide is released by heating the stream. Chemical scrubbing with amine facilitates production of high concentration of methane concentration in biomethane greater than  $\text{CH}_4 > 99\%$ . The limitation of chemical scrubbing needs pre-treatment stage, to remove  $\text{H}_2\text{S}$  and has got high operational and investment costs [30].

The process is similar to pressurized water scrubbing, but is a chemical absorption technique. The solution absorbs  $\text{CO}_2$  in biogas, by chemical reaction between amine and  $\text{CO}_2$ . The absorber is maintained at operating pressure 1–2 bar while the stripper maintained 1.5–3 bar. The process is exothermic, causing temperature rise of amine solution and higher efficiency since the reaction between amine and  $\text{CO}_2$  increases with increase in temperature. Hydrogen sulfide ( $\text{H}_2\text{S}$ ) should be removed prior to the reaction to avoid poisoning the amine solution [1, 7].

### 3.2 Organic physical scrubbing

Organic physical scrubbing work on the same principle with water scrubbing with the difference being the use of an organic solvent with higher affinity for  $\text{H}_2\text{S}$  and

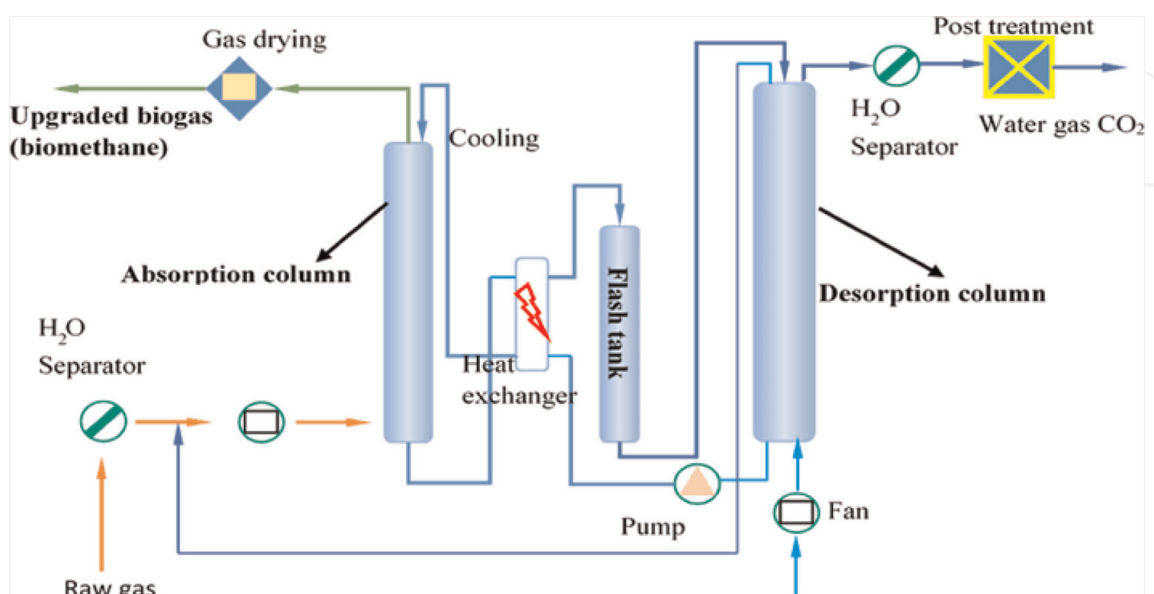
CO<sub>2</sub>. Methanol and dimethyl ethers of polyethylene glycol (DMPEG) mixtures are all used in biogas upgrading. The process simultaneously absorbs hydrogen sulfide, carbon dioxide, and water due to their higher solubility in polyethylene glycol than methane. Examples of commercially available organic physical scrubbing products Selexol® and Genosorb®. These products exhibit high hi solubility of NH<sub>3</sub> and CO<sub>2</sub> compared to H<sub>2</sub>O. Selexol® can absorb three times more CO<sub>2</sub> than water hence lower liquid requirements which requires a smaller upgrading [3, 25].

The challenge associated with high solubility of carbon dioxide in organic solvents is difficulty to regenerate organic solvents. Higher solubility of H<sub>2</sub>S compared to CO<sub>2</sub> in Selexol® leads to increased separation temperatures during the regeneration of the solvent hence higher energy consumption. It is therefore advice able to remove H<sub>2</sub>S before the gas is treated with the solvent. The Selexol process may also be configured to remove H<sub>2</sub>S selectively, or non-selectively in order to remove both CO<sub>2</sub> and (H<sub>2</sub>S) [25].

In the first stage, raw biogas is compression and cooled to (7–8 bar, 20°C), before injection to the bottom of the absorption column. Since temperature affects Henry's constant the organic solvent is cooled down before it is fed to the column. The desorption column is used to regenerate the organic solvent by heating it to 80°C and reducing pressure to 1 bar. This leads to final methane content of 96–98.5% and less than 2% CH<sub>4</sub> losses, in an optimized full-scale plant [3]. **Figure 4** shows that the organic physical scrubbing method.

From **Figure 4**, it is noted that the main elements of the organic scrubbing method are Sulfur absorber, CO<sub>2</sub> absorber, H<sub>2</sub>S concentrator, H<sub>2</sub>S stripper, stripper reboiler reflux pump and reflux accumulator. In organic physical scrubbing, CO<sub>2</sub> in raw biogas is absorbed in an organic solvent e.g. a mix of dimethyl ethers of polyethylene glycol [29].

Concerns over the environment has motivated a shift from the use of conventional solvents to green solvents. This includes the use of deep eutectic solvents (DESs), consisting of two or more components, which are mainly hydrogen bond donors and acceptors [9, 25]. It desirable for the solvents to have a lower melting point, very low



**Figure 4.**  
*Organic physical scrubbing [1].*

vapor pressure and preferably be biodegradable. It is through selection of best fit hydrogen bond donors and hydrogen bond acceptors and donors, that the DESs can be appropriately engineered to yield desired thermodynamic and physical characteristics. It is also possible to remove other biogas contaminants by appropriate process modifications [3, 9].

### **3.3 Pressure swing adsorption (PSA) and vacuum swing adsorption (VSA)**

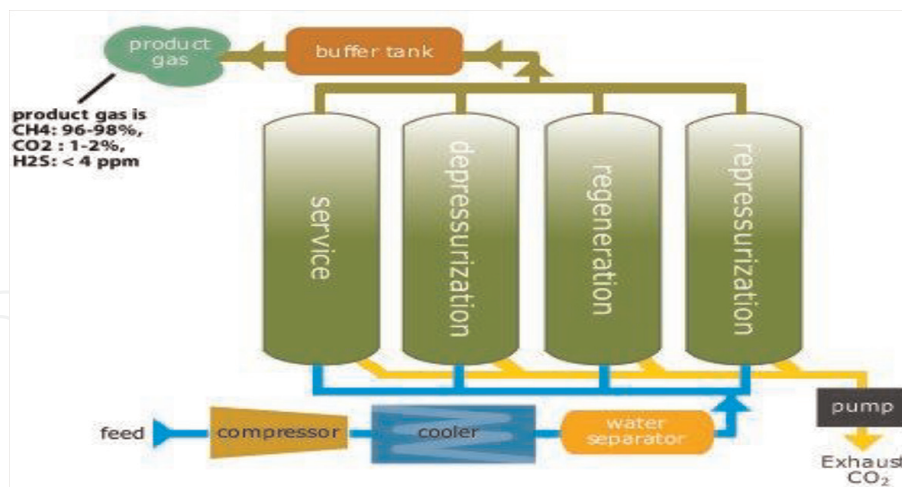
The principle upon which the pressure swing adsorption (PSA) is the adsorption of  $\text{CO}_2$  compared to  $\text{CH}_4$  under conditions of high pressure due to differences in molecular characteristics and the affinity of the adsorbent material used. The vacuum swing adsorption (VSA) is based on the same principle is except that it operates under vacuum during the desorption step. Materials used as adsorbent matter PSA is required to have high surface area, e.g. alumina, silica gel, activated carbon, zeolite, polymeric sorbents and carbon molecular sieves [10, 31].

Pressure swing adsorption is a technique that works by selective adhesion of one or more components of the mixture, on the surface of a micro-porous solid. In this case the material for biogas upgrading is typically equilibrium-base adsorbents. The adsorbent pores allow an easy penetration of the carbon dioxide molecules but filters the larger methane molecules. The molecular sieve materials used include zeolites and activated carbon which act as the adsorptive materials for biogas upgrading. The process requires a pretreatment step because the materials used in PSA plants foul in the presence of raw biogas impurities. The pressure swing technology can achieve 95–99% methane purity for upgraded biogas which meets the typical technical specifications for the grid injection [25]. The main limitations of the pressure swing process are the pre-treatment requirement and extensive process control making the process expensive. To reduce operational costs, the temperature swing adsorption (TSA) is used instead. Temperature swing adsorption works at constant pressure and needs thermal energy to regenerate the adsorbent material making it suitable in applications having cheap heat source [30, 31]. The pressure swing method is illustrated in **Figure 5**.

From **Figure 5**, it is noted that the main processes and elements of the compressor for raw biogas, chambers for absorption, depressurization, desorption, pressurization, and vacuum pump for extraction of vent gases [10]. The characteristics of PSA unit include feeding pressure, cycle time, purging pressure, adsorbent, and column interconnectedness among other things [29].

Pressure swing adsorption (PSA) is a dry method used to separate gases on basis of their properties. Raw biogas is compressed to an elevated pressure and supplied to an adsorption column that retains  $\text{CO}_2$  but leaves  $\text{CH}_4$ . Once the column material is saturated with  $\text{CO}_2$ , pressure is released hence  $\text{CO}_2$  is desorbed and fed to the off-gas stream. Multiple columns can be applied columns are needed for continuous operation allowing them to be closed and opened consecutively [29].

The Pressure Swing Adsorption (PSA) technology makes use of the ability of porous adsorbent medium to adsorb specific molecules out of raw biogas and then release through the application of different pressure levels. For the case of raw biogas upgrading [1, 29]. For the biogas upgrading process, the operation is based on the different molecular dimensions of  $\text{CO}_2$  which is 0.34 nm, methane  $\text{CH}_4$  with 0.38 nm. The application of adsorbent material with cavities of 0.37 nm facilitate retention of  $\text{CO}_2$  in the pores, as methane flows out with no retention. The most utilized adsorbent materials are the Zeolites and activated carbons due to their high efficiency [1].



**Figure 5.**  
Pressure swing adsorption system [29].

The pressure swing adsorption process takes place in vertical columns that are packed with adsorbents. The process has four steps; adsorption, depressurization, desorption and pressurization in the listed order. Biogas passes through in the pressurized column, while CO<sub>2</sub>, N<sub>2</sub>, O<sub>2</sub> and H<sub>2</sub>S are adsorbed by selected material. Hydrogen sulfide and siloxanes are irreversibly adsorbed onto adsorption material and should therefore be removed, together with moisture before injection into the PSA system. It is recommended to use multiple adsorption columns to ensure continuous operation. Once the saturation of adsorbent material is saturated, biogas is allowed into the next column, as regeneration is done for the saturated column. The adsorption column is depressurized to about atmospheric pressure (PSA) or kept under vacuum (VSA). A mixture of CH<sub>4</sub> and CO<sub>2</sub> with high content of CH<sub>4</sub> methane content is released and recycled to the PSA inlet. Biomethane produced can attain purity of 96–98%; but up to 4% CH<sub>4</sub> can be lost within the off-gas stream [10, 25].

The Pressure Swing Adsorption (PSA) technology makes use of the ability of porous adsorbent medium to adsorb specific molecules out of raw biogas and then release through the application of different pressure levels. For the case of raw biogas upgrading [1, 29]. For the biogas upgrading process, the operation is based on the different molecular dimensions of CO<sub>2</sub> which is 0.34 nm, methane CH<sub>4</sub> with 0.38 nm. The application of adsorbent material with cavities of 0.37 nm facilitate retention of CO<sub>2</sub> in the pores, as methane flows out with no retention. The most utilized adsorbent materials are the Zeolites and activated carbons due to their high efficiency [1].

Recent development of the PSA/VSA focus on optimization of adsorption materials and technology. New methods include vacuum swing adsorption system that applies amine-containing nanogel particles supported by carbon fiber having a honeycomb shape whose primary application is the capture of CO<sub>2</sub> from flue gas with potential use in biogas upgrading. In this method, the size of the column and operational costs are reduced by using a rotating design and honeycomb carbon fibers as supportive material, while the combination with amine-containing nanogel particles, increases the recovery of CO<sub>2</sub> [10, 31]. The Amine-containing nanogel particles also reversibly uptake and release CO<sub>2</sub> at lower regeneration temperature of about 75°C which limit the degradation and volatility of amine used [31].

### 3.4 Absorption techniques

In the absorption techniques, purification and enrichment processes are based on the solubility of constituent gases in biogas in a selected liquid. The commonly used liquids are water or organic solvent like methanol, N-methyl pyrrolidone, and polyethylene and glycol ethers are for absorption of CO in physical absorption plants installations. Amine scrubbing is widely applied for chemical absorption. Water scrubbing has two main applications, namely pre-treatment e.g. before PSA and for the removal of H<sub>2</sub>S in actual upgrading. The main limitation of these technique is that it requires significant plant size to achieve high final concentration of methane [30].

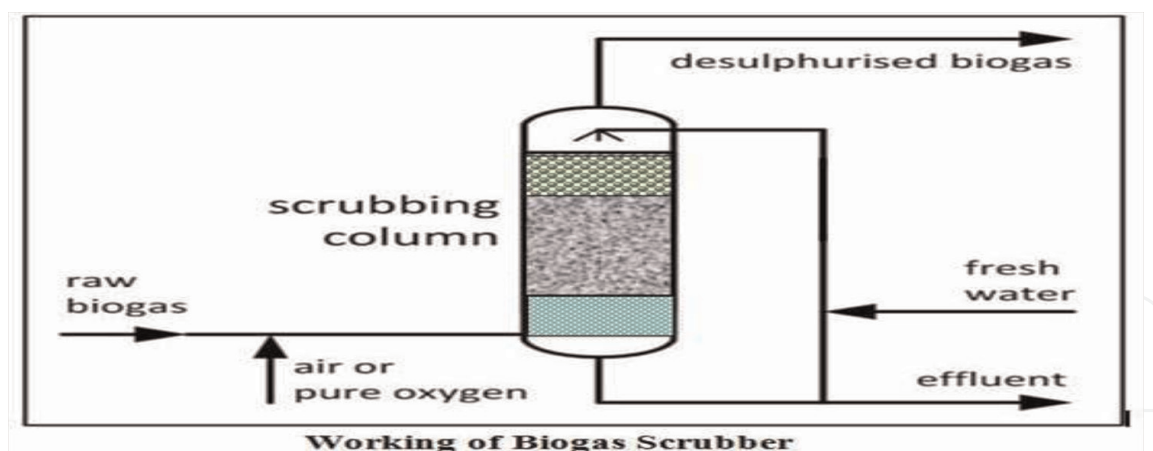
#### 3.4.1 Pressurized water scrubbing

Water scrubbing is the most common technology for both biogas cleaning and upgrading. Pressurized water scrubbing depends on the separation of CO<sub>2</sub> and H<sub>2</sub>S from raw biogas as a result of increased solubility of CO<sub>2</sub> compared to CH<sub>4</sub>. Based on Henry's law, CO<sub>2</sub> solubility in water at 25°C is about 26 times greater than the solubility of methane [13]. Raw biogas is first compressed to 6–10 bar, ND up to 40°C then injected into the absorption column from the bottom side of the tank, while water is supplied from the top while water is supplied from the top side of the column then it flows in the counter-current flow of the gas. The absorption column of the system is filled with random packing material for increased gas-liquid mass transfer [25].

Biomethane is released from the top of the scrubber, the water phase containing the CO<sub>2</sub> and H<sub>2</sub>S are circulated to the flush column, in which the pressure is degraded to 2.5–3.5 bar and while traces of CH<sub>4</sub> dissolved in the water is recovered. On the basis of water re-use single pass scrubbing is often employed when water is from sewage treatment plants and “regenerative absorption”. Water can be regenerated in a desorption column by decompression at pressure, leading to the removal of CO<sub>2</sub> and H<sub>2</sub>S. Water decompression is done by air stripping but where biogas has high concentrations of H<sub>2</sub>S, steam or inert are consumed on desorption process to prevent formation of elemental Sulfur by means of air stripping, which leads to operational problems. The regeneration is desirable of huge water requirement by the system e.g. water flow to upgrade 1000 Nm<sup>3</sup> /h of raw biogas needs 180 and 200 m<sup>3</sup>/h based on pressure and water temperature. Upon drying, in drying stage, the purity of methane formed can reach 99% purity [13, 25].

Pressurized water scrubbing process works on the basis of the fact that carbon dioxide is more soluble in water than methane. It is the simplest and most popular upgrading technology for biogas. It is necessary to remove H<sub>2</sub>S from biogas prior to scrubbing due to its high solubility in water, making its removal difficult. Hence the need to previously remove hydrogen sulfide (H<sub>2</sub>S) from biogas, to avoid corrosion and process efficiency reduction. Biogas is first compressed and fed to the absorption column (scrubber), for cooling to (5°C) and pressurized (4–10 bar) to allow water to absorb CO<sub>2</sub> and other impurities. The flash tank is used for water regeneration in the first phase with the recovery of the absorbed biogas, being recycled by injecting at the biogas inlet. The second phase of regeneration takes place in a second column called a stripper through a countercurrent with air, operating under atmospheric pressure [2, 4]. **Figure 6** shows the water scrubbing system.

The main parts of the water scrubbing system as shown in **Figure 6** are the water separator, a compressor a flash tank, desorption column, a cooler, filter, water and an upgraded biogas dryer for upgraded biogas. A water scrubber is a physical scrubber



**Figure 6.**  
*Water scrubbing system [29].*

which exploits the fact that  $\text{CO}_2$  is more soluble in water than methane. The  $\text{CO}_2$  is separated from the raw biogas and dissolved into the water in the absorption column by application of high pressure of 6–10 bar. The  $\text{CO}_2$  is then released from the water in the desorption column, by addition of air at atmospheric pressure [29].

### 3.4.2 Chemical scrubbing

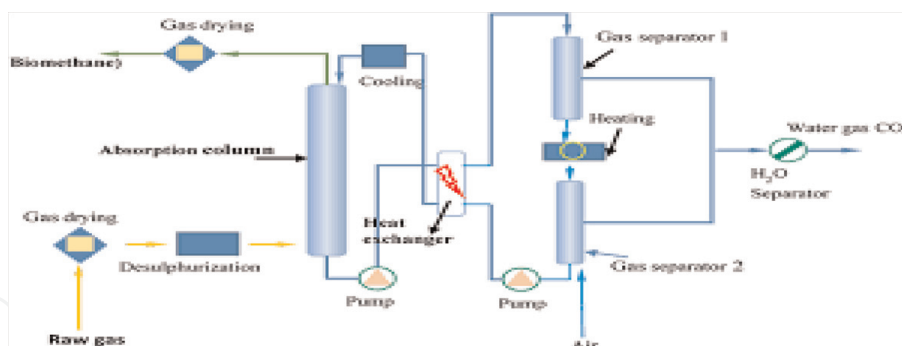
Chemical scrubbing systems use aqueous organic or inorganic compounds to bind the  $\text{CO}_2$  or  $\text{H}_2\text{S}$  molecules existing in biogas. The most commonly used scrubbing systems use organic compounds, namely aqueous amine solutions like diethanolamine (DEA), monoethanolamine (MEA) or methyl diethanolamine (MDEA) [25]. Most scrubbing systems using amine solutions have an absorber unit maintained between biogas pressure of 1–2 bars which is injected to the tank bottom with amine solution flowing from the top and a stripper. The solute and solvent ( $\text{CO}_2$ ) undergo a reversible exothermic chemical reaction with the product amine solution which is rich in  $\text{CO}_2$  and  $\text{H}_2\text{S}$  which proceeds to the stripper for regeneration operating at a pressure of 1.5–3 bars and temperature of 120–160°C [6].

The heat in the stripper disrupts the chemical bonds formed in absorption phase and which creates steam having  $\text{CO}_2$ . Upon cooling this steam, the  $\text{CO}_2$  is released while the condensate is recirculated back to the stripping column. Some commercial systems can cope with biogas with  $\text{H}_2\text{S}$  content of up to 300 ppm/v, but  $\text{H}_2\text{S}$  poisons the amine, cause corrosion and increase the system energy requirements hence the need to remove  $\text{H}_2\text{S}$  before amine scrubbing [3].

The advantages of the chemical scrubbing using amine solutions include high selectivity of the amines by  $\text{CO}_2$  and the substantial extraction compared to other methods e.g. two times more  $\text{CO}_2$  per unit volume is absorbed compared with water. The process has significantly low energy requirements mainly because of exothermic reactions and low process pressure operations i.e. 1–2 bar for absorption column and 1.5–3 bar in the stripping column. The draw backs include high energy requirements for solvent regeneration, expensive amine solvents and losses of solvents to evaporation which increase operation costs [3, 25]. **Figure 7** shows that chemical scrubbing using amine solution.

**Figure 7** demonstrates a chemical scrubbing system using amine solution. It is equipped with the absorption column a heater cooler and a stripping column with a





**Figure 7.** Chemical scrubbing with amine solution [1].

heating medium being hot water, oil or steam. The removal of  $\text{CO}_2$  using reactive systems is not new, but it is less common compared to other technologies like PSA and water scrubbing. The synopsis of features of the chemical scrubbing technology is to use a reagent which chemically binds carbon dioxide molecules for removal from the gas [29].

In chemical scrubbing (CSC), there is reversible reactions between absorbed substances and solvent used. The commonly used biogas upgrading absorption solutions is based on amines i.e. methyl diethanolamine diethanolamine, monoethanolamine, and piperazine. For amine scrubber an absorber tank is used in which carbon dioxide is absorbed from the biogas operating at  $20\text{--}65^\circ\text{C}$  and  $1\text{--}2$  bar, then followed by a stripper where carbon dioxide is released by heating the stream. Chemical scrubbing with amine facilitates production of high concentration of methane concentration in biomethane greater than  $\text{CH}_4 > 99\%$ . The limitation of chemical scrubbing needs pre-treatment stage, to remove  $\text{H}_2\text{S}$  and has got high operational and investment costs [30].

The process is similar to pressurized water scrubbing, but is a chemical absorption technique. The solution absorbs  $\text{CO}_2$  in biogas, by chemical reaction between amine and  $\text{CO}_2$ . The absorber is maintained at operating pressure  $1\text{--}2$  bar while the stripper maintained  $1.5\text{--}3$  bar. The process is exothermic, causing temperature rise of amine solution and higher efficiency since the reaction between amine and  $\text{CO}_2$  increases with increase in temperature. Hydrogen sulfide ( $\text{H}_2\text{S}$ ) should be removed prior to the reaction to avoid poisoning the amine solution [7].

### 3.4.3 Organic physical scrubbing

Organic physical scrubbing work on the same principle with water scrubbing with the difference being the use of an organic solvent with higher affinity for  $\text{H}_2\text{S}$  and  $\text{CO}_2$ . Methanol and dimethyl ethers of polyethylene glycol (DMPEG) mixtures are all used in biogas upgrading. The process simultaneously absorbs hydrogen sulfide, carbon dioxide, and water due to their higher solubility in polyethylene glycol than methane. Examples of commercially available organic physical scrubbing products Selexol® and Genosorb®. These products exhibit high hi solubility of  $\text{NH}_3$  and  $\text{CO}_2$  compared to  $\text{H}_2\text{O}$ . Selexol® can absorb three times more  $\text{CO}_2$  than water hence lower liquid requirements which requires a smaller upgrading [3].

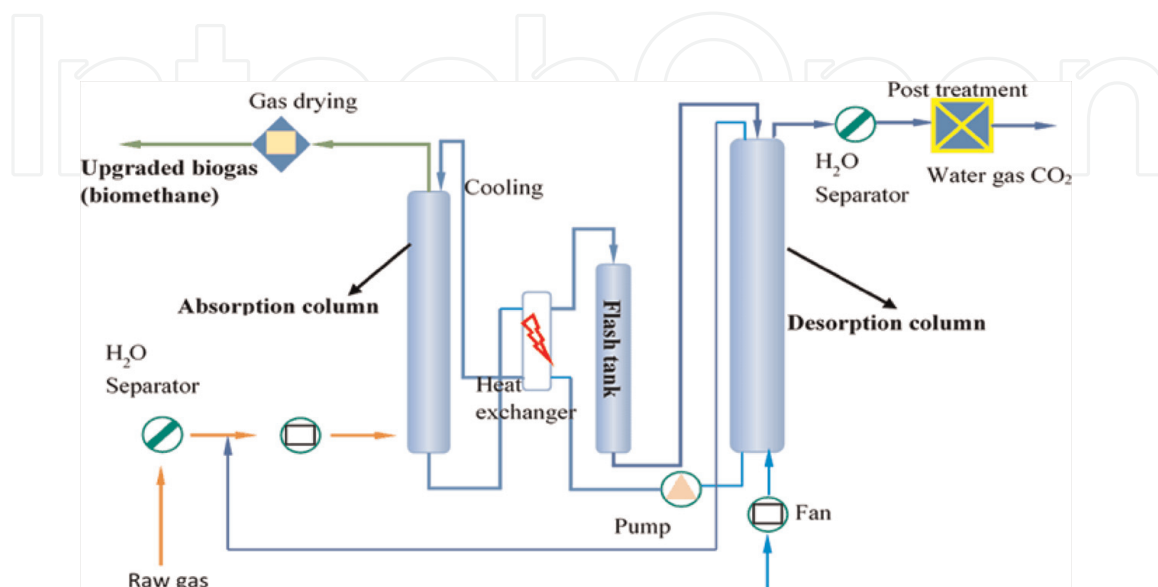
The challenge associated with high solubility of carbon dioxide in organic solvents is difficulty to regenerate organic solvents. Higher solubility of  $\text{H}_2\text{S}$  compared to  $\text{CO}_2$  in Selexol® leads to increased separation temperatures during the regeneration of the solvent hence higher energy consumption. It is therefore advice able to remove  $\text{H}_2\text{S}$

before the gas is treated with the solvent. The Selexol process may also be configured to remove H<sub>2</sub>S selectively, or non-selectively in order to remove both CO<sub>2</sub> and (H<sub>2</sub>S [25]).

In the first stage, raw biogas is compression and cooled to (7–8 bar, 20°C), before injection to the bottom of the absorption column. Since temperature affects Henry's constant the organic solvent is cooled down before it is fed to the column. The desorption column is used to regenerate the organic solvent by heating it to 80°C and reducing pressure to 1 bar. This leads to final methane content of 96–98.5% and less than 2% CH<sub>4</sub> losses, in an optimized full-scale plant [3, 25]. The organic physical scrubbing method is shown in **Figure 8** below.

The main elements of an organic physical scrubbing system as shown in **Figure 8** are the Sulfur absorber, CO<sub>2</sub> absorber, a compressor H<sub>2</sub>S concentrator, a reflux pump and accumulator, and stripper reboiler. An organic solvent is used to absorb the CO<sub>2</sub> in raw biogas organic in physical scrubbing method in a process that is theoretically similar to water scrubbing, based on the Henry's law. These solvents include a mix of dimethyl ethers of polyethylene glycol. The relative solubility of the biogas components depends on the solvent used e.g. the solubility of carbon dioxide is much higher in the organic solvent than in water, meaning that the Henry's constant for carbon dioxide is higher. CO<sub>2</sub> has a solubility of 0.18 M/atm in Selexol which is about 3 times higher than in water. CO<sub>2</sub> is about 17 times more soluble than methane in the Genosorb solvent which is a smaller difference than for water, in which CO<sub>2</sub> is 26 times more soluble than methane [29]. These differences in solubility have technical and economic implications.

Concerns over the environment has motivated a shift from the use of conventional solvents to green solvents. This includes the use of deep eutectic solvents (DESs), consisting of two or more components, which are mainly hydrogen bond donors and acceptors [9]. It desirable for the solvents to have a lower melting point, very low vapor pressure and preferably be biodegradable. It is through selection of best fit hydrogen bond donors and hydrogen bond acceptors and donors, that the DESs can be appropriately engineered to yield desired thermodynamic and physical characteristics. It is also possible to remove other biogas contaminants by appropriate process modifications [3, 9].



**Figure 8.**  
*Organic physical scrubbing [3].*

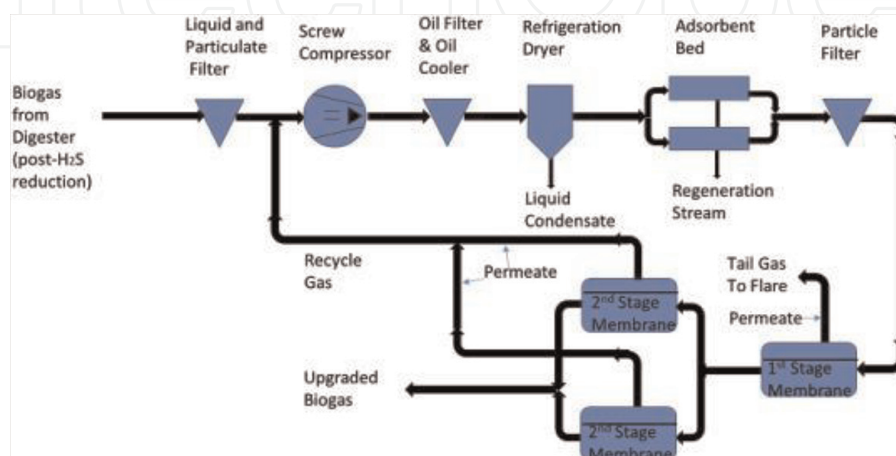
### 3.5 Membrane separation

One alternative to the conventional absorption in biogas upgrading is membrane technology which make use of membranes which can be made from polymeric materials like cellulose acetate [3]. The membrane is a filter with ability to separate components in raw biogas to the molecular level. Membranes came into use in the 1990s and were initially built with less selective membranes and were applied in applications with lower recovery demand for the methane [29]. The membrane separation technology is a viable alternative to the conventional absorption-based biogas technology. The process relies on the selective permeability properties of membranes which effectively enable separation of biogas components. The various components of raw biogas have different relative permeation rates which can be ordered hierarchically from the slowest to fastest permeation as follows;  $C_3H_8$ ,  $CH_4$ ,  $N_2$ ,  $H_2S$ ,  $CO_2$  then  $H_2O$  [13, 25]. This is demonstrated in **Figure 9** below.

The membrane separation system is demonstrated in **Figure 9** showing stage wise removal of  $CO_2$ ,  $O_2$ ,  $H_2O$  from raw biogas leaving methane in purified form leaving the membrane. This process can also remove  $CO_2$ ,  $H_2O$  and hydrogen and parts of the oxygen from biogas. The permeation rate depends on the size of molecules hydrophilic through a typical membrane typically made of a glassy polymer [29].

The membranes used in separation are selectively permeable barriers that are designed allow some molecules to pass through but stop or block other with process drivers being the pressure, relative concentration, temperature, and electric charges of the molecules. The membranes used are of three major types, namely polymeric membranes, inorganic membranes, and mixed matrix membranes [2]. Inorganic membranes have got higher mechanical strength, chemical resistance and thermal stability making them more popular. Mixed matrix membranes are the mostly used type of membrane separation in industrial applications. The polymeric and inorganic membrane separation technologies need pre-treatment since  $H_2S$  negatively affect medium –term performance. As a strategy to recover up to 99.5% methane, a multi-stage membrane strategy is adopted. The penetration of the membrane technology of separation is high costs and low reliability [2, 30].

The membrane permeation technique works on the basis of the difference in permeability of between the various constituents of biogas. The action of a membrane facilitate separation in which methane is retained, while carbon dioxide and other



**Figure 9.** Operation of membrane separation [29].

constituents penetrate through the membrane. Three types of membranes are currently used for biogas purification; polymeric, inorganic and mixed matrix membranes. The process is not meant to remove  $H_2S$  and  $H_2O$ , however, they should not be allowed to affect the performance of the membrane. By introducing multiple stages of membranes,  $CH_4$  concentration above 98% and with low operational cost can be attained [3, 4].

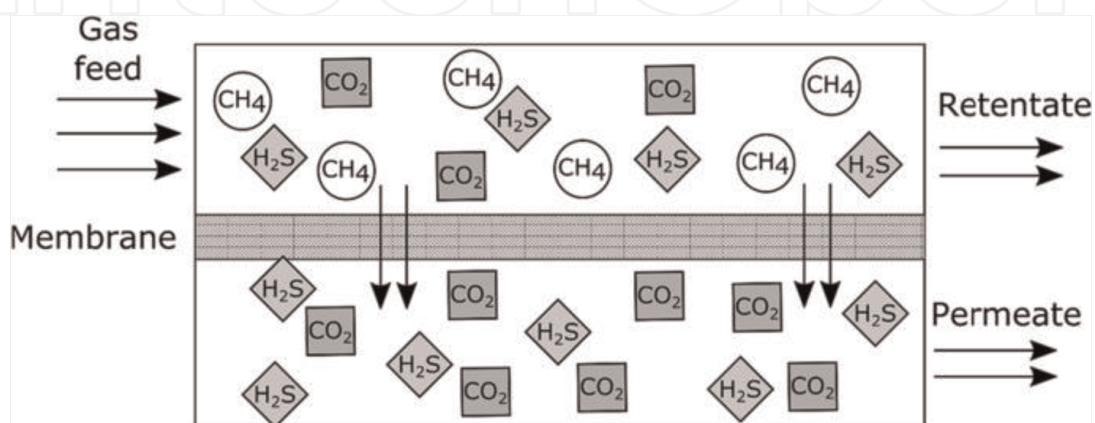
From **Figure 10**, it is noted that a gas is fed into a membrane separator where the impurities mainly  $H_2S$ ,  $CO_2$ , are isolated from raw biogas to exceed as permeates while larger methane molecules exceed as retentate i.e. biomethane.

The membrane separation is divided into wet (gas–liquid) and dry (gas–gas) techniques. Biogas is usually pressurized to 20–40 bars or 6–20 resulting in  $CH_4$  abundant gas which will on one side of the membrane with the higher pressure. The  $CO_2$ , some  $H_2S$  and a significant amount of methane of 10–15% diffuses to the lower pressure side. Contaminants like water, siloxanes,  $NH_3$ , VOCs and  $H_2S$  are removed before membrane separation to avoid corrosion and clogging. There are different configurations of gas-gas units i.e. single-pass membrane unit or multiple stage membrane units with internal recirculation of permeates and retentates. For one system, about 92% purity of biomethane can be attained while multiple stages can attain 96% or more methane purity [3, 25].

The cold-membrane and cryogenic technologies combination is an interesting approach that can be used in upgrading of biogas. Polyimide and polysulfone membranes can be used in biogas upgrading process to attain up to 98% methane purity, based on simulation results. The process has relatively lower energy requirement of about 1.6 MJ/kg  $CH_4$  which is lower than energy requirements of a standard membrane process which is about 2.4 MJ/kg  $CH_4$ . The process can further lower energy requirements to 0.8 MJ/kg  $CH_4$  if the process is coupled with liquefied methane regasification [3, 8].

### 3.6 Cryogenic separation

Cryogenic separation technology is done by a gradual reduction in the temperature of raw biogas causing liquefaction of  $CH_4$ ,  $CO_2$  and other constituent parts to ensure methane meets quality standards for Liquefied Natural Gas (LNG). Raw biogas is initially dried and compressed to 80 bars followed by stepwise cooling to  $-110^\circ C$  leading to gradual removal of impurities like., siloxanes,  $H_2O$ ,  $H_2S$ , halogens etc. and



**Figure 10.**  
*Principle of membrane separation [6].*

CO<sub>2</sub> which is the main impurity in biogas to obtain almost pure biomethane with purity (> 97%) [13, 32].

The physical principle behind cryogenic technique is based on the fact that the gases like carbon dioxide, hydrogen sulfide liquefy and solidify under different pressure and temperature conditions. Therefore, the cryogenic plants operate at very low temperature (−170°C) and high pressure (80 bar). Biogas purification is done by cryogenic technology, with lower methane losses but the process is expensive. The cryogenic process can be used in the production of liquefied natural gas (Bio-LNG) [6, 30].

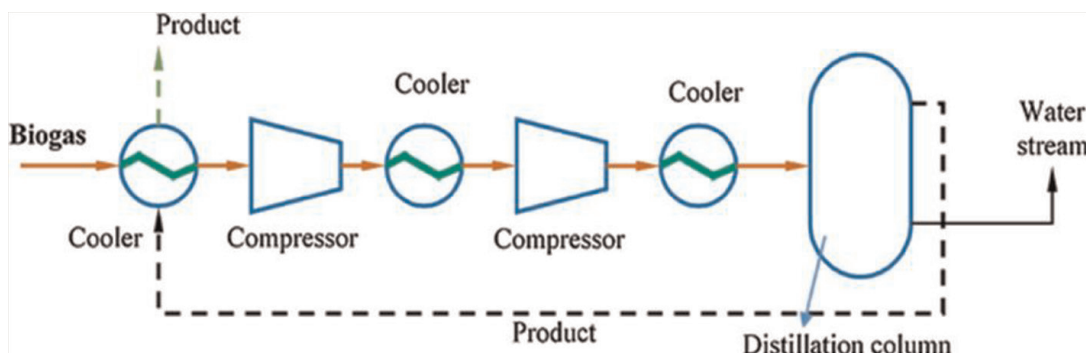
The cryogenic separation process involves the separation of different gas components based on the basis of their different boiling points by gradually reducing the temperature. The process begins by compressing biogas to 80 bars then reducing the temperature to −25°C. At this temperature and pressure, components that are removed from raw biogas are moisture, halogens, siloxanes and H<sub>2</sub>S. Reducing temperature to −55°C, liquefies most of the present CO<sub>2</sub>, then further reduction to −85°C is the last step which removes the remaining CO<sub>2</sub> in solid form. However, to avoid operational challenges like pipe clogging, ice formation and heat exchanger clogging, the impurities like H<sub>2</sub>S, water, siloxanes and halogens are by practice removed prior to cryogenic separation [6].

The purity of biomethane produced with cryogenic upgrading be over 97% with methane losses of less than 2%. The limitation of cryogenic upgrading the high investment and operation costs, methane losses and need for pretreatment to remove impurities. There are additional variants and configurations like cryogenic distillation or cryogenic adsorption [2, 3]. The cryogenic process is shown in **Figure 11**.

From **Figure 11**, we note that the cryogenic separation method is characterized by successive compression and cooling at different pressure to liquefy and isolate the different components of raw biogas. Water is removed in the distillation Column of the process.

Although the cryogenic separation process is quite promising with interesting performance and results, the method is still under development with just few facilities operating at commercial scale. The process limitations so far are high costs of investment and operation costs, methane losses and clogging derived from increased concentration of solid CO or and presence of other impurities [13, 28].

The cryogenic processes take advantage of the low temperatures to achieve their goals. By allowing component gases in raw biogas to liquify. A process does not have to operate below a fixed temperature level for it to be considered “cryogenic”., but since the However, since the processes involved in this process are done well below



**Figure 11.** Cryogenic separation system [6].

–55°C, the common gases in raw biogas can be liquefied and separated which forms the basis for cryogenic biogas upgrading [29].

### 3.7 Biological upgrade techniques

These processes apply biological separation via hydrogenotrophic methanogenesis consisting of hydrogenotrophic-methanogens to convert CO<sub>2</sub> and H<sub>2</sub> into CH<sub>4</sub> but commercialization of the processes is limited by several challenges [30]. The positive side is that it requires low investment and operating costs particularly in terms of electricity and head demand. It does not require any chemical products or additional equipment. Easy operation and maintenance. The process leads to high concentrations of hydrogen sulfide while O<sub>2</sub>/N<sub>2</sub> excess necessitates additional cleaning while air overload creates an explosive mixture [33].

Although the physicochemical techniques dominate the biogas upgrading market, biological methods have been riding for the last 20 years. Biological technologies for Sulfur treatment in biogas are classified into chemotrophic and photosynthetic types. The advantage of biological techniques is that end products are non-hazardous i.e. sulfur or sulfate with efficiency being same or higher than that of physicochemical technologies while the Sulfur recovered can be used as a raw material for production of sulfuric acid, fungicides and Sulfur fertilizers [33, 34].

#### 3.7.1 Chemotrophic removal of hydrogen sulfide

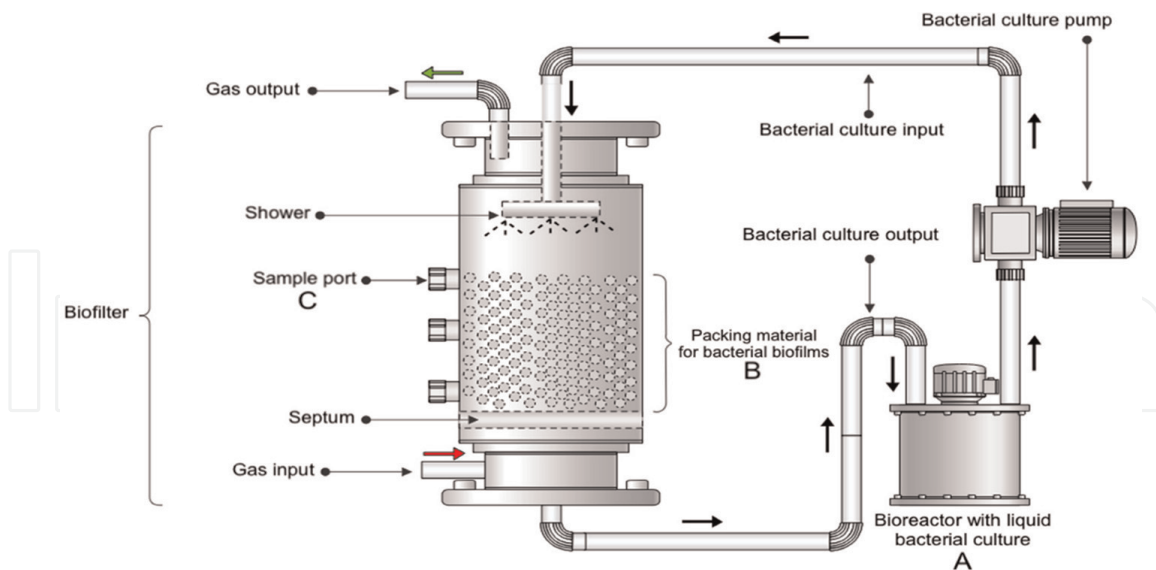
In this process, the Chemotrophic Sulfur oxidizing bacteria, also known as colorless Sulfur bacteria, is the ideal microbial group used for biodegradation of H<sub>2</sub>S. The bacteria are used to oxidize the reduced Sulfur compounds like. Sulfide, polysulfide, elemental Sulfur, thiosulfate, and sulfite to gain chemical energy and utilize CO<sub>2</sub> as a carbon source. Biodegradation of Hydrogen sulfide is done aerobically using where the electron acceptor is O<sub>2</sub> and anaerobically where the electron acceptor is NO<sub>3</sub>. The bacteria include genera Thiobacillus, Acidithiobacillus, Sulfolobus, Thiovulum, Thiostrix and Thiospira [3, 24].

#### 3.7.2 Biofiltering

Biofilters remain the simplest type of gas desulphurization systems. The purification system consists of a bioreactor where sulfur-oxidizing bacteria like the Thiobacillus, Pseudomonas and Acidithiobacillus are immobilized on a carrier. In the process, moisturized biogas is injected from the bottom of the biofilter and forced through a moist, packed bed with microbial biofilm which purifies biogas. The bed material is used to supply nutrients or nutrient solution added from the top occasionally. Oxygen whose concentration is 5–10% of volume is supplied by injecting air directly into the gas stream [3, 24]. **Figure 12** shows the biofiltering system.

The biofiltering system as demonstrated in **Figure 12** consists of peristaltic pumps for biogas and air, a bioreactor and biogas storage.

Factors influencing the operation of biofilters include the bed medium, moisture content of biogas, gas temperature, the pH, nutrient and oxygen levels, and the development of biofilm. A good or suitable bed material should have large specific area and porosity, create small pressure loss, light in specific weight and cheap. The bed material should absorb gas odor and but retain its nutrients, contain indigenous microorganisms and water i.e. moisture content between 40 and 60%. Suitable bio



**Figure 12.**  
*Biofiltering system [29].*

filter materials include natural organic materials like composts, coconut fiber, woodchips/bark, and peats mainly because of their native microorganism consortia and good level of performance [3, 35].

The benefits of using biofilters are reduced operating costs, no chemical requirements. The main limitations in use of bio filters in purification of biogas are media acidification by the sulfuric acid formed from  $H_2S$ , degradation and inefficient mixing. Solutions to these challenges include using a carrier with alkaline properties, adding alkaline. Biofilters are also not suitable for high loading rates due to limited buffering capacity and limited control capability for moisture, and pH during high airflows [3, 25].

### 3.7.3 Bio trickling filters

The general mechanism of biotrickling filtration is same as bio filters, except for the use of inert packing bed material hence the need for continuous supply of the nutrient solution. Plastic supports, activated granular carbon or porous ceramics, are materials commonly used to provide support for biofilm formation. Advantages of bio trickling over traditional filters include better process stability, better control and regulation of the pH and temperature, low flow resistance, less space and continuous nutrient supply. The continuous washout of products of acidic reactions solves the problem of buffering and acidification common in bio filters. However, the challenge of continuous nutrient supply leads to excessive growth of biomass and clogging of anaerobic zones. Commercially available bio trickling systems include BioSulfurex® (DMT Environmental Technology), Biopuric process (Biothane Corporation), Bidox® (Colsen B.V.) and BiogasCleaner® (BioGasclean) [3, 5].

### 3.7.4 Bio scrubbing

Bioscrubber system is applied in the removal of compounds like ammonia, amines, hydrocarbons, hydrogen sulfide and odorous contaminants. Bio scrubbing system consists of two reactor units with the first reactor as absorption tower where the

pollutants are absorbed in a liquid phase before it goes to the second reactor which is activated sludge. Degradation occurs in the activated sludge reactor where microorganisms like *Thiobacillus* and *Thioalkalivibrio*) grow in suspended flocks. The effluent generated is recirculated back to the absorption tower. In the removal of H<sub>2</sub>S, a sedimentation tank is installed after the second reactor for collection of elemental Sulfur with O<sub>2</sub> being used as the oxidant. Optimal microbial growth and activity are maintained by addition of oxygen, nutrients and pH regulation together with continual purging of by-products and excess biomass out of the system [3, 36].

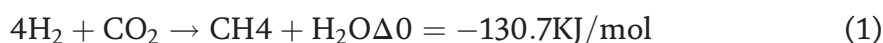
No injection of N<sub>2</sub> and O<sub>2</sub> is required by bio scrubbers. They are used to handle load fluctuation loads better as well as stable performance due to easy control conditions. The main disadvantage of the bio scrubbing process is high initial costs. Commercially available bio scrubbing systems used for H<sub>2</sub>S removal from are THIOPAQ® process and Sulfothane™ which are very similar. In the process, the gas is injected to the absorption tower where counter flow of alkaline solution absorbs H<sub>2</sub>S from raw biogas. The formed sulfide-containing goes to a micro-oxygenated reactor. The chemotrophic sulfur oxidizing bacteria, are dominated by haloalkaliphilic *Thioalkalivibrio* which convert absorbed sulfide to elemental Sulfur [3, 37].

### 3.7.5 Phototrophic Sulfur removal with anoxygenic bacteria

Phototrophic Sulfur removal uses bacteria with ability to utilize light as an energy source to remove Sulfur compounds from the environment e.g. anoxygenic phototrophic sulfur bacteria, purple non-Sulfur bacteria, cyanobacteria, and phototrophic members of phylum Chloroflexi and Heliobacteria. Some bacteria like the anoxygenic phototrophic sulfur bacteria can oxidize hydrogen sulfide to elemental Sulfur through an oxygenic photosynthesis. Anoxygenic phototrophic Sulfur bacteria consist of two families namely Chlorobiaceae (green sulfur bacteria) and Chromatiaceae (purple Sulfur bacteria). The purple and green sulfur bacteria use light as an energy source, and use reduced Sulfur compounds as electron donors for photosynthetic CO<sub>2</sub> reduction. Sulfide oxidation produces globules of elemental Sulfur. The Chromatiaceae store Sulfur outside of their cells while the Chlorobiaceae store Sulfur inside. The green Sulfur bacteria utilize bacteriochlorophyll *c*, *d*, or *e* found in special light-harvesting organelles (chlorosomes) that allow the growth under the lower intensity light (25–80 lx). The photosynthetic pigments in purple Sulfur bacteria, are bacteriochlorophyll *a* or *b* and various carotenoids i.e. spirilloxanthin, rhodopinal, spheroidene, and okenone) [25].

### 3.7.6 Chemoautotrophic methods

The chemoautotrophic biogas upgrading methods rely on the hydrogenotrophic methanogens which use H<sub>2</sub> to convert CO<sub>2</sub> to CH<sub>4</sub> based on the following Eq. (1):



To make this reaction renewable requires that the source of hydrogen used should be derived from renewable sources hence the need to apply renewable electricity to hydrolyze water for H<sub>2</sub> generation. This facilitate storage of the surplus energy generated by solar and wind to create a new technology called power to gas (P2G). Variable renewable sources need buffering to enable energy delivery when it is dark with no solar and the wind is still. Storage batteries are widely used to store electricity



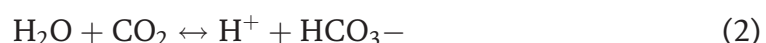
but have drawbacks like capacity limitations, high production cost, and use of toxic materials. Hydrogen (H<sub>2</sub>) is a clean energy resource that can be produced by electrolysis of water. As a renewable energy carrier, H<sub>2</sub> has some disadvantages like very low volumetric energy density about 10.88 MJ/m<sup>3</sup> compared to CH<sub>4</sub> which has 36 MJ/m<sup>3</sup> making hydrogen storage a challenge in terms of space requirement. This makes integration of P2G technology for conversion of H<sub>2</sub> to CH<sub>4</sub> really attractive as it integrates wind or solar energy technology as well as biogas technology [25, 37].

Biogas upgrading makes use of existing facilities of the biogas plants which reduces the initial investment cost. The process of chemoautotrophic does not separate or absorb the CO<sub>2</sub>, instead it is converted to methane (CH<sub>4</sub>) leading to significant increase in the final energy value of the output “wind gas” (wind gas is methane produced using the surplus energy from wind turbines) or “solar gas” (which is methane produced using surplus solar energy). This technology acts as a precondition for the sustainability of the ambitious biogas development strategy of decoupling the biogas production from the biomass availability. Hydrogen assisted biogas upgrading configurations are classified into in-situ, ex-situ and hybrid designs. The in-situ and ex-situ processes have been experimentally proven with several research undertaken unlike the hybrid concept which is still under development [13, 36].

### 3.8 In-situ biological biogas upgrading

This method was earlier own presented as one of the raw biogas cleaning technique not aimed at producing biomethane grade biogas. Situ is a biotechnology based on the direct injection of pure air or oxygen and in the process, the bacteria that oxidize H<sub>2</sub>S develop with the presence oxygen, leading to the biological removal process of H<sub>2</sub>S, to produce sulfur (S) which leaves the digester via the digested. The microorganisms are widely found in the anaerobic environment present in bio-digesters [4, 37]. In situ desorption technology is yet to be fully developed even though it has been around for over 20 years. In-situ is based on the greater solubility of CO<sub>2</sub> over CH<sub>4</sub> in water. The process set up includes an anaerobic digester linked or connected to an external desorption unit. Sludge transported to an aerated desorption column from the digester. Nitrogen or air flowing in counter-current mode dissolves the CO<sub>2</sub> from the sludge in the desorption unit. The sludge desorbed sludge is pumped back into the digester to reabsorb more CO<sub>2</sub>, and the sludge as the sludge is continuously recycled in the desorption column. It is possible to strip out H<sub>2</sub>S with dissolved CH<sub>4</sub> and CO<sub>2</sub> from the recirculating sludge by applying large quantities of air or N<sub>2</sub>, causing reduction in the H<sub>2</sub>S and CO<sub>2</sub> concentration [26, 27].

In the in-situ concept, H<sub>2</sub> is injected into a biogas reactor so that it is coupled with the endogenous CO<sub>2</sub> from anaerobic digestion in the digester for conversion into CH<sub>4</sub> by autochthonous methanogenic archaea. The process can yield methane with purity of 99% if operational parameters like the pH are fully monitored to values above 8.5, as a result of the removal of bicarbonates which inhibits of methanogenesis [13]. CO<sub>2</sub> dissolved in the liquid phase of the reactor dissociates to H<sup>+</sup> and HCO<sub>3</sub><sup>-</sup> ions Utilization of carbon dioxide (CO<sub>2</sub>) leads to reduction in H<sup>+</sup>, which causes concomitant increase in the fermentation pH [13]. The reaction is summarized in Eq. (2) below.



Studies on in-situ biogas upgrading reactors show some process inhibition of methanogenesis from bicarbonate consumption which proves the argument that

conventional biogas production systems need a pH of 8.5 as the threshold for optimum bio-methanation process for mesophilic and thermophilic activities. Co-digestion with acidic waste can be applied to mitigate the pH increase and alleviate the technical limitations. Another solution to the challenge is application of pH control which enable upgrading to almost pure biomethane [13].

Oxidation of the Volatile Fatty Acids (VFA) and alcohols is thermodynamically feasible in where  $H_2$  concentration is very low. High  $H_2$  levels ( $> 10$  Pa) inhibit bio-digestion and, and promote accumulation of electron sinks like lactate, ethanol, propionate, and butyrate. VFA degradation will cease if there is introduction of sudden and high  $H_2$  concentrations in the reactor which causes system imbalanced or even, fatal deteriorated as a result of excess acidification caused by VFA accumulation. Injection of  $H_2$  in batch reactors at a concentration more than stoichiometric amount for hydrogenotrophic methanogenesis leads to accumulation of acetate, due to stimulated homoacetogenic pathway, and/or decreased methanogenic activity of acetoclastic archaea. However, upon longer term  $H_2$  exposure, there is increase in hydrogenotrophic population which improves the utilization capacity of  $H_2$  and reverts the inhibition [13, 25].

Solubilization of  $H_2$  to the liquid phase is another important parameter since it must cross the interface between the liquid and gas for it to be available for the microorganisms. Hence aqueous solubility of most gasses is rather low, limiting the gas-liquid mass transfer and which retards performance the bioreactor. Therefore, the material and module type used to inject  $H_2$ , use of gas recirculation flows and the reactor designs are important aspects of the implementation of sufficient in-situ biogas upgrading. Studies in Batch experiments showed that the rate of uptake of  $H_2$  decreases rapidly at  $CO_2$  concentrations  $< 12\%$  and maximum  $CH_4$  purity attained was 89%. Studies in continuously fed reactors using hollow fiber membranes for  $H_2$  injection in a reactor treating cattle manure and cheese realized 96%  $CH_4$  purity of final gas. Studies in up flow anaerobic sludge blanket reactor, using a hollow fiber membrane in an external degassing unit and realized 94% methane purity [13, 36].

### **3.9 Ex-situ biological biogas upgrading**

Ex-situ biogas upgradation relies on supply of carbon dioxide from external sources and hydrogen in an anaerobic reactor, which eventually contributes to their conversion to methane. The ability of ex situ process to manage high concentrations of influent gases, reduces retention time about 1 hour leading to a smaller device for upgrading. Depending mainly on the reactor used, the ex-situ technology can produce methane with final purity of 79–98%, the main challenge facing this technology is low gas-liquid mass transfer rate [26].

Through studies, it has been established that the operating temperature significantly affects bio-methanation efficiency e.g. enriched thermophilic culture resulted in  $> 60\%$  higher  $H_2$  and  $CO_2$  bioconversion when compared to mesophilic culture in batch. In q typical study, increase of operating temperature from 55 to 65°C showed significant increase in efficient of bio-methanation operation Other than temperature, an adaptation period is needed for microorganisms to efficiently ferment the  $CO_2$  and  $H_2$  gasses e.g. it was established that operating a mesophilic trickle-bed reactor with immobilized hydrogenotrophic culture for 8 months, improved output to  $CH_4$  content of over 96%. Similar results are experienced for bio-methanation efficiency under thermophilic conditions [25].

The reactor type and application of gas recirculation or liquid mixing are important design parameters for biogas upgrading system. Up flow in series or bubble column reactors realize over 98% methane purity, even when H<sub>2</sub> is injected through conventional spargers instead of advanced membrane modules. The trickle bed reactor systems yield higher CO<sub>2</sub> and H<sub>2</sub> conversion efficiency to achieve as high as 98–99% methane purity, due to the formation of biofilm of mixed anaerobic consortia which act as good biocatalyst for the process. High stirring speed or diffusion devices with pore sizes generate gas-bubbles which are able to mix the reactor yield better kinetics and gas quality [13].

The bio filter technology involves passing the biogas through a column having a synthetic material, in the form of a biofilm. The parallel or countercurrent flow maintains the humidity and nutrients, that are essential for the microorganisms that degrade of H<sub>2</sub>S [4].

In biological gas scrubber, a two-stage system is used to remove H<sub>2</sub>S. In the first stage H<sub>2</sub>S scrubbing column, applies sodium hydroxide solution while activated sludge is used in the second stage which is injected with injected with air, because the microorganisms used are aerobic, leading to the solution regeneration [4].

## **4. Applications of biomethane**

Biomethane has superior properties compared to biogas and is attractive substitute of natural gas. It has both industrial and domestic application like use as cooking gas, cogeneration can be packed in containers/cylinders as compressed biomethane and can be injected to natural gas mains for distribution. The main challenge is the cost of processing which is a function of technology used. Bio-CNG, which is a methane-rich compressed fuel in form of biomethane. Bio-CNG is made from pure biogas with more than 97% methane composition pressurized to 20–25 MPa. Compressed bio-CNC is similar in properties to regular CNG in terms of its fuel properties, economy, engine performance, and emissions. Like regular CNG, bio CNG has high octane number, and yields high thermal efficiency. It can therefore substitute the regular compressed natural gas in gas pipelines and other applications including fuel for natural gas power plants [28, 36, 38, 39].

### **4.1 Hydrogen production**

Hydrogen is an ideal raw material for a sustainable energy transformation, but with the challenge being where and how to get hydrogen from renewable sources. Renewable hydrogen can be produced using renewable energy sources and usually produced via water electrolysis [40]. Biogas has applications beyond electricity and biomethane production, as because, through steam reforming, it can be used to manufacture green hydrogen, in a process where a catalyst refines and separates the hydrogen from the gas stream [6, 40]. The most common method used to manufacture hydrogen is by steam-reforming of natural gas, followed by pressure-swing adsorption to remove impurities. However, small reformers like those used for combined heat and power with biogas plants are in commercial operation [40]. The biogas has low heating coefficient due to high composition of carbon dioxide and water vapor interfere with the combustion process [2, 40]. Upon removal of carbon dioxide and water molecules, the methane (CH<sub>4</sub>) can be used for hydrogen synthesis and bio-fuel production. Methane can be split to hydrogen molecules (H<sub>2</sub>) in a process that can be

done in a steam/methane reformer. In the process, high pressure and temperature steam is combined with the methane (CH<sub>4</sub>) to produce flow of hydrogen molecules and CO molecules [2, 6, 33].

It is through thermochemical processes of hydrocarbons that large-scale hydrogen production is manufactured through the reforming process. Biomethane has significant potential application in hydrogen manufacture as a substitute of fossil natural gas as a raw material for reforming processes. The demand for renewable hydrogen production is set to grow significantly due to concerns over fossil fuels depletion and greenhouse gas emissions, and associated concerns over global climate change. The availability of capital, desired hydrogen amount and purity hydrogen and the composition of available biogas will influence the selection of reforming processes [41].

Biomethane as significant application in fuel cell technologies for applications like power generation. Fuel cells can use hydrogen to generate electric power just like batteries as well as fuel for the fuel for powering fuel cars. Fuel cell technology in power generation is emission free and hence attractive [42]. Biomethane can be used as a source for renewable hydrogen, for stationary fuel cells and power fuel cell electric vehicles (FCEVs). The Hydrogen-powered FCEVs are environmentally attractive since they have no tailpipe emissions other than making them extremely clean as transport option to fossil fuel powered vehicles [37, 41].

Use of biomethane for hydrogen production can increase energy sustainability can be for energy applications like fossil fuels. Hydrogen can be manufactured by autothermal reforming (ATR), electrolysis or methane reforming (SMR) [43]. Biomethane can be used as a substitute of natural gas which will provide a hedge against growing demand for natural gas [41].

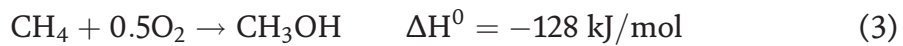
Hydrogen fuel can be used to reduce emissions from engines which are widely used in transportation. Hydrogen fuel cells promise to provide an alternative to internal combustion (IC) engines particularly due to the clean exhaust emissions, renewal nature of the fuel and higher efficiencies. Hydrogen fuel cell vehicles can achieve widespread acceptance except for existing challenges like waste heat removal in mobile applications [44].

#### **4.2 Production of biofuels from biomethane**

The transport sector is important since it accounts for about 14% of the global greenhouse gas emissions [45]. Liquefied biomethane is a feasible fuel for power plants and heavy trucks and can also be used as a raw material for production of other fuels and chemicals like methanol, dimethyl ether, and hydrogen fuel. Biomethane is currently used as a transport fuel many countries with benefits of lower environmental impact compared to fossil fuels and several other processed transport fuels [46].

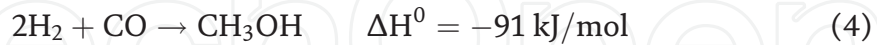
Biofuels include the Bio-CNG which is compressed biomethane similar to (CNG) in properties with industrial, automotive and domestic applications. The process needs removal of impurities like water, N<sub>2</sub>, O<sub>2</sub>, H<sub>2</sub>S, NH<sub>3</sub> and CO<sub>2</sub> to achieve composition of >97% CH<sub>4</sub>, <2% O<sub>2</sub> at 20–25 MPa. Bio-CNG occupies less than 1% of the volume at standard conditions [46, 47].

Biomethane can also be used in the industry as transport fuel by liquefying it to at a high pressure ranging from 0.5 to 15 MPa [4]. In the biological or chemical pathways, biomethane can be converted to methanol, diesel, liquefied petroleum (LPG) and gasoline. Methanol is produced by partial oxidation of methane as shown below;



In another method, methane is biologically converted from to methanol by using methanotrophic bacteria used in methanol production through the action of methane monooxygenase (MMO) enzyme [5].

Methanol can also be produced by reforming methane to syngas then followed by catalytic conversion of syngas to methanol as shown below [6].



Methanol can then be converted to gasoline through methanol-to-gasoline process. Biogas or biomethane can be processed to methanol through dry reforming, steam reforming, partial oxidation reforming, autothermal Reforming (ATR) and the Fischer-Tropsch (FT) Process. Synthesis gas (syngas) is the main product of biomethane reforming process. Syngas is a raw material for production of many long chain hydrocarbons [48].

#### i. Dry reforming

In dry reforming, CO and H<sub>2</sub> are produced by reaction of methane (CH<sub>4</sub>) and carbon dioxide (CO<sub>2</sub>). The process utilizes two greenhouse gases (CH<sub>4</sub> and CO<sub>2</sub>) making it very attractive. Unfortunately, the endothermic reaction reduces the of CO<sub>2</sub> emissions, since CO<sub>2</sub> emitted generate the heat required for the reaction has to be accounted for. Dry reforming is an efficient route for producing synthesis gas yielding a H<sub>2</sub>/CO ratio close to 1 [49]. The disadvantage of dry reforming compared with steam reforming it produces lower syngas ratio (H<sub>2</sub>/CO = 1), The ratio of H<sub>2</sub>/CO ratio is influenced by water gas shift reaction (WGS), which reduces the ratio due to reverse reaction that oxidizes hydrogen to water. In this process, the H<sub>2</sub>/CO ratio is kept between 1 and 2 by partial oxidation of methane through feeding water. This enhances forward water gas shift reaction. The energy demand by the process is lower since partial oxidation is exothermic [48]. The temperature range for dry reforming process 700–1000°C [48].



#### ii. steam reforming and water shift reaction

This process combines methane in biomethane with water vapor generate CO and H<sub>2</sub> in the in the presence of a catalyst. The process is endothermic and takes place between 650 and 850°C, to produce hydrogen yield of 60–70% [49]. Steam reforming takes place between 700 and 900°C. The two-step chemical reaction is shown below;



The process of steam reforming is often followed by a water shift reaction to improve hydrogen generation.

### iii. Partial oxidation reforming (POR)

This process is used to produce hydrogen at reduced energy cost because the process is moderately exothermic compared to steam reforming which is highly endothermic. H<sub>2</sub> and CO are produced by the partial oxidation at atmospheric pressure and between 700 and 900°C partial oxidation reforming. The H<sub>2</sub>/CO ratio of 2 yield is achieved in full conversion with reduced soot formation. Methane react with oxygen to form carbon dioxide (CO<sub>2</sub>) due to decrease in CO selectivity. The combustion is strongly exothermic leading to formation of hot-spots in the reactor bed and coke deposition on the catalyst [49]. In this process, methane is oxidized to syngas as demonstrated below



### iv. Autothermal Reforming (ATR)

Autothermal Reforming is a combination of two processes i.e., POR and SR in the presence of carbon dioxide. In Autothermal Reforming (ATR) partial oxidation takes place in the reactor to produce heat needed for steam reforming in the catalytic zone. The process does not need external heating and the reactor is easy to stop and restart. Compared to partial oxidation reaction, the hydrogen yield is higher and consumes less oxygen compared [49].

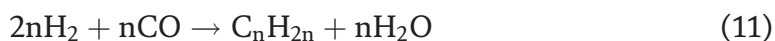
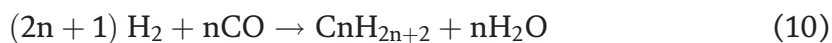
#### 4.2.1 Upgrading syngas

The Syngas produced from the dry reforming has carbon dioxide which should be removed before it is supplied to the Fischer-Tropsch reactor. Amine absorption has high selectivity for carbon dioxide. Other applications of this technology are separation of CO<sub>2</sub> from flue gases, natural gas cleaning and largescale upgrading of biogas. Common solvents used in the process are alkanolamines like monoethanolamine (MEA), diethanolamine (DEA) or methyldiethanolamine (MDEA) [50, 51].

#### 4.2.2 Fischer-Tropsch (FT) process

The Fischer-Tropsch (FT) synthesis, was name after the German inventors Franz Fischer and Hans Tropsch is a process used to manufacture liquid hydrocarbon fuels like coal-to-liquids (CTL) and/or gas-to-liquids (GTL) based on source of syngas. [50, 51]. The biomethane and natural gas conversion to fuels via Fischer-Tropsch synthesis (FT-synthesis) is feasible at industrial scale [48]. The Fischer-Tropsch (FT) process is coverts syngas to products like LPG, diesel, and jet fuels [24, 50].

The Fischer-Tropsch synthesis (FT-synthesis) polymerizes the carbon and hydrogen atoms in syngas or biomethane to create long chain molecules. The process is run over iron or cobalt catalyst at 20–30 bars [14] in an overall exothermic process leading to polymerization of CH<sub>2</sub> to hydrocarbons with long chains called syncrude. The various reactions in Fischer-Tropsch process are summarized below;



Reactors used are include multi-tubular fixed bed, circulating fluidized bed, fixed fluidized bed, and slurry reactor. The reactions for the slurry reactor conditions are 20–30 bar, and 200–300°C while the syngas H<sub>2</sub>/CO ratio of 1–1.8 [48, 50]. For high temperature synthesis, fluidized-bed FT reactors are used to generate light hydrocarbons in form of gaseous hydrocarbons and gasoline and generally have higher output. The catalysts used are Fe and Co which are sensitive to sulfur compounds in syngas [50].

#### 4.2.3 Biofuels from biomethane

Various biofuels can be made from biomethane for the transport sector e.g. methanol, compressed biogas (CBG), hydrogen, liquid biogas (LBG), dimethyl ether and Fischer-Tropsch (FT) fuels [52]. Fuels can be produced by biogas upgrading to biomethane then compressing to make (CBG) or liquefying to make (LBG), or gasification to produce syngas for use in manufacture of hydrogen, methanol, DME and FT diesel [52].

### 4.3 Biomethane for gas and power grids

In many countries, governments have come up with national support schemes to promote biomethane market. Support mechanisms include feed-in support schemes, green gas products and quota obligations as market drivers in Europe. Biomethane production for many countries is based on organic waste as feedstock, but for Germany which dominates Europe's feed-in market is based on energy crops. In Germany, the main driver is the feed-in tariff for renewable electricity through the Renewable Energy Sources Act (EEG). Biomethane support schemes mainly rely on mass balancing systems or 'book and claim'-certificates. Existing mass balancing systems, can contribute to international market development through creation of common standards. As biomethane becomes popular and relevant in the energy systems, integration into power and gas grids and shift away from subsidies to markets and competition with natural gas will become major issues [53].

Biomethane should meet some standard specifications with respect to storage and transport before it can be practical injection into existing natural gas networks. The presence of various components in different concentrations make it difficult to inject biogas to the grid hence the need for upgrading. Pipeline designers should know the exact thermodynamic properties of a gas mixture are, particularly in terms of density, and heating value which may tend to vary greatly in biogas [54].

Biomethane has a very important role to play in the transition to renewable sources of energy. Demand based production of biomethane for power generation directly links the gas grid and the electricity grids which can help in balancing the power grid. The gas grid will shift from fossil fuel distribution to provide energy balancing service provider with short-term as well as seasonal storage options. There is increasing integration of decentralized biomethane feed-in into the gas grid for the gas grid infrastructure thus introducing new challenges. There are examples in Germany of facilities that feed in more biomethane to the local distribution network than the total discharge which leads to the need to compress the excess gas and transfer it to a higher level [24, 53].

Production of biomethane from energy crops has a negative impact on agriculture. On the other hand, use of digestion residues as fertilizers close to biogas production

site improves local nutrient cycles. Production of energy intensive nitrogen fertilizers and use of declining global phosphorous reserves can be avoided by use of bio-fertilizer from the digesters [8, 18].

Biomethane is the most efficient biofuel in terms of fuel production equivalent per area of crop land needed and is therefore expected to a larger role in the fuel/energy market because of government support, growing use in NGVs and reduction in GHG emissions. There is growing awareness of biomethane and a shift in perception from regarding biomethane as a sub-branch of biomass production to an independent renewable energy resource. And legislation and strategies are recognizing biomethane as an independent energy resource [53].

The main sustainability challenge facing biomethane market is cost of subsidies and need for free market competition with fossil natural gas which can be accelerated if the market price of natural gas rises. The European cap-and-trade for greenhouse gas emissions GHGs is another driving factor for the future. Since use of biomethane omits GHG emissions, there will not be compensation or penalties in form of GHG certificates [37, 53].

The evolution of biomethane markets is expected to create their own demand and supply and also enable and exchange between different countries since the green gas product market open to international trade. Each country has tended to create its own set of biomethane support schemes to address individual situations and are therefore designed with to address the priorities and challenges of specific countries. For biomethane market to grow, countries should open up their support schemes to biomethane imported from neighboring countries to encourage international trade in biomethane [53].

#### **4.4 Electricity from biomethane**

Biomethane can be used as fuel for power generation in various prime movers. They include internal combustion engines, gas turbines of varying sizes, fuel cells, among other. The efficiency can be improved through combustion and conversion in set ups like cogeneration and tri-generation schemes [6, 37].

Diesel engines can run on biomethane as a direct substitute of natural gas. Biomethane used can made from biogas upgrading or gasification and methanation schemes [55]. Diesel engines would perform efficiently whether using pure diesel or when running in dual fuel mode as long as the calorific value of fuel is controlled [55, 56].

Electricity from biomethane can be used directly onsite to avoid or limits electricity imports from the grid while excess generated electricity within the design of decentralized power generation systems using a wide range of prime movers for the electric generators e.g. turbines, internal combustion engines, fuel cells, etc. Biomethane can converted to hydrogen fuel for wide renewable applications or used in fuel cells for direct conversion. Various pathways for use of biomethane for power generation are summarized in **Table 4** below.

From **Table 4**, it is biomethane can be used through various conversion technologies with varying characteristics in thermal and electricity generation. The conversion can be done in cogeneration, trigeneration and open conversion systems. Prime movers that can use biomethane include internal combustion engines, gas turbines, fuel cells, and Stirling engines as well as production of fuels for application in transport, heat and electricity generation.

In the transport sector, biomethane has a double role to play in emissions reduction i.e. as a direct fuel substitute of fossil fuels and as feedstock for production of biofuels/



Device/ technology	Application	Remarks
1 Fuel cell	Generation of electricity	Very efficient, reliable but expensive
2 Hydrogen production	Biomethane can be converted to hydrogen for use as combustion fuel or electricity and process chemicals.	Renewable hydrogen process if its biogas or methane. Hydrogen is manufactured by dry reforming, steam reforming, or hydrolysis
3 Bio-methanation	Biomethane can be fed to natural gas supply as substitute for natural gas	Renewable replacement of fossil natural gas is feasible with use of biomethane
4 Diesel engine	Biomethane can be used as a diesel engine fuel in either dual fuel mode or pure gas engines	Diesel engines have more fuel flexibility and efficiency and can easily use easily use biofuels as fossil fuel substitutes
5 Gas/petrol engine	Biomethane can be used as a fuel for petrol or gasoline engines with little or no modification	Less efficient than diesel engines but are easier to convert to biogas fueled engines.
6 Stirling engine	Stirling engines are also called hot air engines.	Stirling engines have fuel flexibility and can run on a wider range of fuels
7 Gas turbine	Based on size, gas turbines can be micro, small, and large gas turbines in open, closed or combined cycle configuration	Turbines are simple in construction, are versatile and can use raw biogas as well as biomethane and easy to operate.
8 Cogeneration	In cogeneration, biomethane is burn to simultaneously produce useful heat and electricity.	Cogeneration with biomethane as a fuel can be applied on various conversion systems like Stirling engines, diesel engines, gas turbines, hydrogen and fuel cells to increase system efficiency
9 Trigeration	Trigeration refers to generation of electricity and both heating and cooling from same fuel/energy resource simultaneously	Trigeration is the most efficient conversion system but more complex and expensive

**Table 4.**  
Summary of biogas to electricity conversion systems and technologies [2, 6].

chemicals through the Fischer-Tropsch (FT) Process e.g. diesel, jet fuel, and gasoline, and through reforming processes to produce hydrogen and methanol [6, 28].

Biomethane production and use has less environmental impact, but is still associated with some greenhouse gas emissions like CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O whose quantities depend on the technology applied and the source of biogas or feedstock used. Biomethane use can reduce the negative environmental impact and pollution potential while anaerobic digestion and gasification used to produce biogas and syngas can be used for hygiene and of bio wastes further keeping the environment clean and healthy [57–59].

## 5. Results and discussion

### 5.1 Summary

Biogas is a product of the anaerobic digestion process with many applications as in generation of renewable energy. The main component of biogas with energy value is methane, but has impurities like moisture, carbon dioxide, siloxanes, hydrogen

sulfide, siloxanes, hydrocarbons, oxygen, ammonia, oxygen, carbon monoxide and nitrogen whose presence is undesirable as they reduce the calorific value of biogas and create operational problems in the energy systems. This necessitate biogas cleaning and application of multi-stage technologies to produce upgraded biogas called biomethane. Biomethane gas is a flexible and easy to store fuel with similar properties and applications as natural gas with no need to modify any equipment settings for natural gas devices and equipment.

Technologies that are commercially available for operating biogas upgrading biogas today include amine scrubbers, water scrubbers, PSA units, organic scrubbers and membrane units. Cryogenic upgrading technology though interesting has some important operational challenges that have to be resolved. For medium scale upgrading schemes all the most common upgrading options are feasible. The scrubbing technologies have proved to be effective and efficient and have similar costs of investment and operation. The water scrubber is a preferred choice for many applications due to the simplicity and reliability, but the high purity and very low methane slip from amine scrubbers are notable characteristics. The pressure swing adsorption (PSA) and membrane units, have similar investment costs as the scrubbers. Advances in the membrane technology has also led to low methane slips with this technology, which is a notable progress [1, 6].

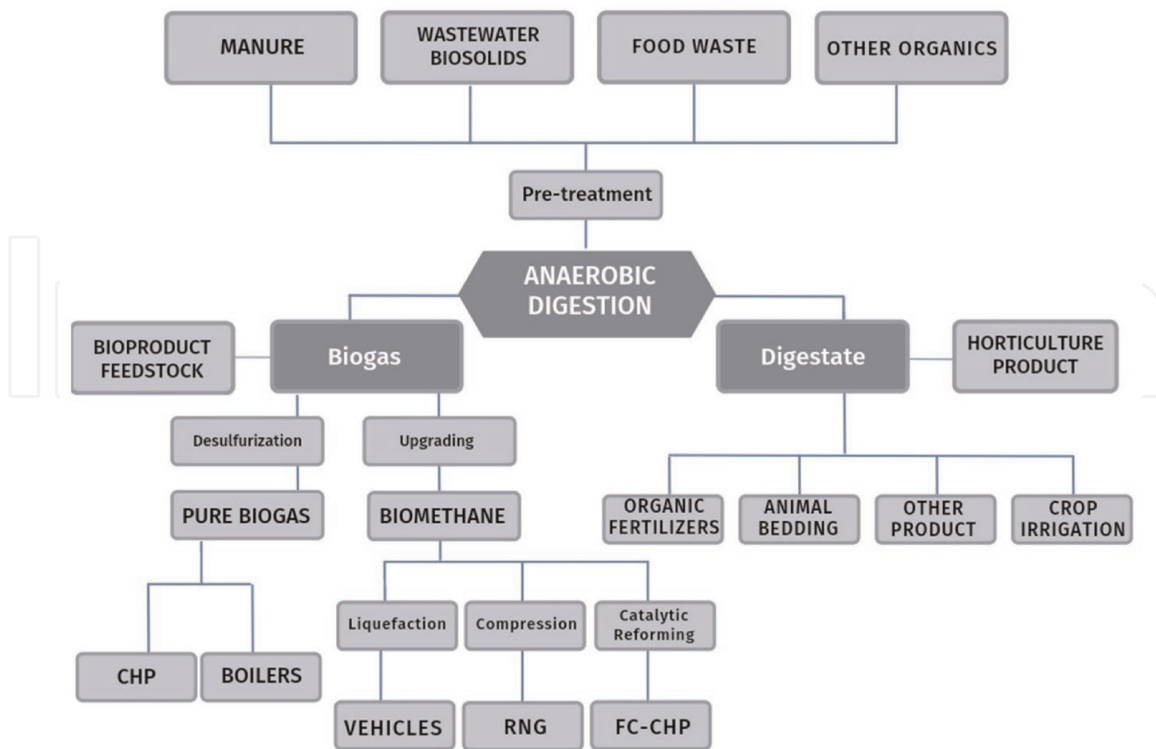
The physical and chemical biogas upgrading technologies are the technologies that are currently mature and have reached near optimum technical and economic feasibility. Their main limitation is huge energy consumption which limit expansion of the biogas market. Upgrading technology selection may depend on factors like costs, quality of products, location, and technology maturity and requirements [26]. It is possible to attain 98% methane content with a water scrubber but the content of oxygen and nitrogen in the raw biogas cannot be separated in the water scrubber and therefore should be controlled. Oxygen and nitrogen can also be transported with water from the aerated desorption column to the absorption column. With methane composition of 50% in raw biogas, composition of oxygen and nitrogen can double in the final product after upgrading [29, 32].

Biological methods are the less explored methods for industrial-scale testing and optimization scenarios, even show they promise significant potential in terms of techno-economic feasibility with new horizons in hybrid renewable energy applications. There also exist large gaps between pilot scale and commercial scale technology applications like hydrate separation, cryogenic separation, biotechnologies, and chemo lithotrophic-based bioreactors. The main limitation to wide scale use of biomethane is high cost remains a major limitation to widespread biogas application [26].

## 5.2 Production pathways for biogas

Biomethane is produced by processing raw biogas through multiple steps and by of methane from other components. The various pathways for biogas and biomethane production and applications are summarized in **Figure 13**.

**Figure 13**, shows various pathways for production of biogas and related by-products, biogas purification, upgrading and various energy applications. The raw materials for biogas production undergo pre-treatment and then fed to the biodigester to undergo anaerobic digestion to produce biogas and digestate. Digestate is applied as green manure, animal bedding, and application like crop irrigation to enhance agricultural production. Raw biogas is purified mainly by desulphurization for



**Figure 13.** Pathways for production and application of biogas and biomethane [6].

applications like boilers, cogeneration and engines. Raw biogas can also be upgraded and applied in catalytic reforming for production of hydrogen and biofuels, liquefied to produce bio-liquified natural gas (bio-LNG) or compressed to produce bio-compressed natural gas (bio-CNG) [2, 6, 35].

### 5.3 Processes comparisons

Biomethane has become an important renewable energy resource for heat and power generation and industrial applications as a feedstock. Applications of biomethane include a transport fuel as a substitute for natural gas, diesel and liquid natural gas, thermal applications i.e. steam and heat generation, combined heat and power, tri-generation, and injection into the natural gas grid upon meeting certain requirements. Biomethane can be manufactured through upgrading of biogas or by gasification followed by methanation process. The approaches in biomethane production have similar efficiencies in biomethane production from the energy output and conservation perspective but since the two technologies have fundamental differences in process and equipment, the cost of the output varies. The main limitations facing biomethane production are the high costs of the process while many biological processes are still under research and development and are yet to be fully commercialized.

Biomethane technology market is a promising venture globally mainly due to existence of mature production, conversion technologies and applications Biomethane remains viable and as a result of abundance in cheap feed stocks supply for anaerobic digestion and gasification. Biomethane has significant flexibility for domestic and industrial scale production and use and is promising to be a leading economical alternative to produce renewable bioenergy.

There are five main biomethane production processes through biogas upgrading. The techniques are pressure swing adsorption which has an option of temperature

swing adsorption, absorption techniques based on amine, membrane separation, cryogenic separation and biological separation. Biogas upgrading can be significantly improved by combining a wide range of methods ranging from biological and physicochemical processes and adaptation of technologies in the field of advanced oxidation or anaerobic phototrophs. The treatment of biogas can theoretically apply biological methods like chemotrophic or phototrophic to remove  $H_2S$  then start upgrading using more efficient physicochemical processes. Purification of biogas is generally a high energy intensive process. But through appropriate choice of a combination of cleaning and upgrading methods based on the methane purity demand saves energy as well as minimize methane loss, in large scale operations. The physicochemical processes are more developed and widely used compared to many biological methods which are still new and not yet commercialized, but they offer significant huge potential in terms of efficiency, feasibility, and technological easiness. Biological methods of upgrading biogas open new horizons for integration of different forms of renewable energy besides electricity, storage advances and decoupling bioenergy production from the availability of biomass resources. The various processes have different benefits and limitation. The water scrubbing can simultaneously remove  $CO_2$ ,  $H_2S$ ,  $NH_3$  and dust but consumes a lot of water. On the other hand, the pressure swing adsorption technique requires biogas pretreatment since water and hydrogen sulfide can damage the adsorbents but, has low energy requirement. The advantage of Amine absorption is low methane loss and produces high quality  $CO_2$ , although it has high energy consumption. The advantage of the membrane permeation systems is their compactness and ease of operation but yields a relatively low methane ( $CH_4$ ) purity.

Biomethane can be used in power generation using prime movers gas turbines, micro turbines, diesel engines, petrol engines, Stirling engines. Other applications are hydrogen production, manufacture of transport fuels, fuel for cogeneration and trigeneration, compression to bio-CNG and LPG, syngas production, methanol production.

## 5.4 Applications

Biogas has multiple applications which include heat and electricity generation. Electricity can be produced from biogas at sewage works, by means of a combined heat and power (CHP) engine, gas turbines, petrol engines, modified diesel engines of dual engines, among others. Biogas and biomethane can also be used as a fuel in automobiles to power an internal combustion engine or a fuel cell in cleaner processes compared to use of fossil fuels [2, 5, 6, 60].

Upgraded biogas or biomethane can attain same properties as natural gas and hence be used as a substitute fuel for natural gas as green natural gas. Biomethane can be injected to natural gas pipelines for use in applications domestic heating and cooking, power generation and feedstock for many industrial processes [6, 35, 61].

## 6. Conclusions

Biogas is a product of the anaerobic digestion process with many applications as in generation of renewable energy. The main component of biogas with energy value is methane, but has impurities like moisture, carbon dioxide, siloxanes, hydrogen sulfide, siloxanes, hydrocarbons, oxygen, ammonia, oxygen, carbon monoxide, and nitrogen whose presence is undesirable as they reduce the calorific value of biogas and create operational problems in the energy systems. This necessitates biogas cleaning

and upgrading to biomethane. Biomethane gas is a flexible and easy to store as a fuel having similar properties and applications as natural gas with little or no modifications to the natural gas equipment.

Biogas upgrading methods can be classified into physical, chemical and biological methods like water scrubbing, physical absorption, pressure swing adsorption, cryogenic separation, membrane separation, chemical scrubbing, chemoautotrophic methods, photosynthetic upgrading and desorption. The physical and chemical upgrading technologies have almost reached optimal level but still have high energy requirements. High-pressure water scrubbing is more economic for small-sized plants, but potassium carbonate scrubbing has high net value for large-sized plants. Therefore, physicochemical methods are technologically ready compared to biological methods which are still new and not yet commercially available, although they offer huge potential in respect to feasibility, technological easiness, and potential. Through biological upgrading new opportunities for integrating different forms of renewable energy are availed besides upgrading including electricity storage advances and decoupling bioenergy production from availability of biomass.

Biogas can be cleaned or purified to remove harmful components like moisture and  $H_2S$  without necessarily upgrading to biomethane which is mainly about the removal of Biogas. removal of  $CO_2$ . Some upgrading methods remove other impurities in addition to  $CO_2$ , while others require upfront removal of  $H_2O$  and  $H_2S$ . Raw biogas cleaning/treatment and upgrading which enables the use of biogas in applications like vehicles fuel or for injection into the natural gas grid as a substitute for natural gas. There have been significant developments over the last few years, in the field of biogas cleaning and upgrading through process improvements and development of new technologies although water scrubbing, PSA and amine scrubbing currently dominate the market. Membrane separation is a technology is while organic physical scrubbers have limited share of biogas upgrading market. Cryogenic upgrading technologies, which are potentially the best choice for combination with liquefaction of biomethane, still face operational challenges that may be resolved.

The market for biomethane globally is promising mainly due to existence of mature production, energy conversion technologies and applications and abundance of cheap feed stocks for anaerobic digestion and gasification. Biomethane has significant flexibility for domestic and industrial scale production and use and is promising to be a leading economical alternative to natural gas.

The main limitations facing biomethane production are high costs of the process while others like biological techniques are still under development. Biogas upgrading can be significantly improved by combining a wide range of methods ranging from biological and physicochemical processes and adaptation of technologies in the field of advanced oxidation or anaerobic phototrophs. The treatment of biogas can theoretically apply biological methods like chemotrophic or phototrophic to remove  $H_2S$  then start upgrading using more efficient physicochemical processes. Purification of biogas is generally a high energy intensive process. But through appropriate choice of a combination of cleaning and upgrading methods based on the methane purity demand saves energy as well as minimize methane loss, in large scale operations. The physicochemical processes are more developed and widely used compared to many biological methods which are still new and not yet commercialized, but they offer significant huge potential in terms of efficiency, feasibility, and technological easiness. Biological methods of upgrading biogas open new horizons for integration of different forms of renewable energy besides electricity, storage advances and decoupling bioenergy production from the availability of biomass resources.

Biomethane can be used in power generation using various available uses like the use of gas turbines, micro-turbines, diesel engines, petrol engines, Stirling engines besides thermal applications as a biofuel for transport and industrial applications. The tracks for production of fuels biomethane are compressing to produce (CBG) or liquefying to make (LBG), hydrogen production, methanol, production, DME, and FT diesel.

Biomethane can be manufactured through upgrading of biogas or by gasification followed by methanation process. The approaches in biomethane production have similar efficiencies in biomethane production from the energy output and conservation perspective but since the two technologies have fundamental differences in process and equipment, the cost of the output varies. High initial and operating costs remain the limiting factor facing the biomethane technology market, while several promising technologies are still under research and development.

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## **Author's contribution**

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## **Availability of data**

The research has provided all data and information used and did not use any undeclared data and information. However, any datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

## **Conflict interest**

The authors declare that they have no conflict of interest.

## **Consent for publication**

The authors have authority to publish the research work.

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