A review on biogas purification through hydrogen sulphide removal

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Abstract. Biogas has been recognized as a clean and renewable form of energy that is produced from biodegradable organic materials via an anaerobic digestion. In fact, biogas has been well expected to substitute current conventional sources of energy. The main composition of biogas includes methane (CH4) and carbon dioxide (CO2), which is accompanied by different contaminants in varied quantities such as ammonia (NH3), water vapour (H2O), hydrogen sulphide (H2S), methyl siloxanes, nitrogen (N2), oxygen (O2), halogenated volatile organic compounds (VOCs), carbon monoxide (CO) and hydrocarbons. Biogas has been widely utilized as either engine fuel or starting material for chemicals, hydrogen and/or synthesis gas productions. For certain applications, a certain purity degree of biogas is required. Technically, the presence of trace components in biogas has been known to negatively affect engine performance. Therefore, a removal of contaminants, particularly H2S and CO2, is expected to significantly improve biogas quality, thus enhancing its performance in various applications. Besides, the removal is performed to meet standardized gas specifications for its usage as vehicle fuel or being injected into a natural gas grid. In practices, different methods for biogas cleaning and upgrading have been recognized, in which differ in terms of functions, efficiency, and required quality of input gas. This study, therefore, aims at reviewing various H2S removal-based biogas cleaning techniques during and after digestion process.

Keywords: anaerobic digestion; biogas purification; H2S removal; upgrading; cleaning; biogas quality

1. Introduction

The production of biogas from various biological wastes through an anaerobic digestion or landfilling has been growing worldwide. Besides, it has been considered as a solution for numerous energy deficiency scenarios. In general, biogas has been recognized to offer economical and environment friendly characteristics, by which it is known as the most efficient energy production technology compared to other biological or thermo-chemical routes of energy conversion processes [1,2]. Through three major biological processes, biogas being produced consists of mainly methane (CH4, 40 to 75 %) and carbon dioxide (CO2, 15 to 60 %). Besides, trace amounts of other components such as water
(H₂O, 5 to 10 %), hydrogen sulphide (H₂S, 0.005 to 2 %), siloxanes (0 to 0.02 %), halogenated hydrocarbons (VOC, < 0.6 %), ammonia (NH₃, <1 %), oxygen (O₂, 0-1 %), carbon monoxide (CO, < 0.6 %) and nitrogen (N₂, 0 to 2 %) may present and cause unintended consequences [3]; [4]; [5]. Table 1 summarizes typical properties of biogas generated from anaerobic digestion plants and landfill sites [6,7].

Practically, CO₂ is a recalcitrant gas that reduces the density and decreases the calorific value of biogas. It is, however, not toxic and corrosive as H₂S. In fact, H₂S is known to be harmful to the environment and corrosive to numerous metal parts of engines, pumps, gas storage tanks, compressors, valves and reduce the lifespan of process equipment [3,7,8]. Besides, some impurities may increase emissions and hazardous risks for human health, hence requiring the contaminative components to be removed before any eventual utilization [6].

Depending on the end uses of biogas, different treatment steps are necessary. For some applications where it is important to have high energy content in biogas, e.g., vehicle fuel or grid injection, the biogas needs to be pretreated [9]. It is therefore important to have an optimized process in terms of low energy consumption and high efficiency, adding high methane content in the upgraded biogas [6]. Basically, two steps involved in biogas treatment to produce a higher fuel standard include cleaning and upgrading. In the cleaning, harmful and toxic compounds such as H₂S, N₂, O₂, Si, H, VOCs, CO, and NH₃ are removed from biogas, while upgrading focuses on adjusting CO₂ content to increase the calorific value of biogas [6,10].

Currently recognized techniques for biogas upgrading include water scrubbing, physical and chemical adsorptions, cryogenic separation, membrane technology, biological upgrading and in-situ

| Table 1. Parameter and composition of biogas produced through anaerobic digestion (AD) and landfilling [6,7,9,37] |
|-------------------------------------------------|-----------------|---------------|-----------------|-------------------------------------------------|
| Parameters                                      | Unit            | Biogas from AD | Landfill Gas    | Impact on Biogas Utilization                      |
| Lower heating value                             | MJ/Nm³          | 23             | 16              | Corrosion in compressors, gas storage tanks and engines due to reaction with H₂S, NH₃, CO₂ to form acids |
| Density                                        | KWh/Nm³         | 6.5            | 4.4             | Decreasing calorific value, anti-knock properties of engines and corrosion |
| Methane number                                  | MJ/kg           | 20             | 12.3            | Corrosion, fooling in cavern storage, the risk of explosion |
| Methane                                        | Kg/Nm³          | 1.1            | 1.3             |                                                   |
| Heavy hydrocarbon                               | Vol (%)         | >135           | >130            |                                                   |
| Water vapour                                   | Vol (%)         | 1 to 5         | 1 to 5          | Emission, anti-knock properties of engines and corrosion |
| Hydrogen                                        | Vol (%)         | 0              | 0               |                                                   |
| Carbon dioxide                                 | Vol (%)         | 0              | 1               |                                                   |
| Oxygen                                         | Vol (%)         | 0              | 1               |                                                   |
| Hydrogen sulphide                              | ppm             | 0 to 4000      | 0 to 100        |                                                   |
| Ammonia                                        | ppm             | 100            | 5               |                                                   |
upgrading methods. Meanwhile, techniques for biogas cleaning are distinguished into wet desulphurization (e.g. physical and chemical absorption, wet oxidation) and dry desulphurization (e.g. membrane separation, molecular sieve, pressure swing adsorption (PSA), fixed-bed adsorption method, Claus oxidation process) [11].

Recently, biogas cleaning and upgrading have been facing significant challenges due to the site-specific and case sensitive characteristics of the selected technology. In general, the selected technology relies on numerous factors, including specific biogas utilization requirements and local circumstances. The technology should also be efficient in terms of energy consumption and operating costs [6]. According to [12], investment cost is a critical issue, by which a cheapest technology that meets required conditions for biogas utilization will be preferred. This study focuses on biogas cleaning by the removal of H$_2$S to optimize biogas production for certain usages.

2. Hydrogen Sulphide on Biogas Production

Hydrogen sulphide is naturally contained in biogas due to biogas-producing protein-rich organic materials. The concentration of H$_2$S in biogas is varied from 100 to 10,000 ppm depending on the feedstock [13,14]. Technically, H$_2$S is formed under anaerobic conditions due to the role of sulphate (SO$_4^{2-}$) as an electron acceptor while organic compounds are biologically degraded [13]. Primary mechanism for H$_2$S production is the reduction of S-containing compounds (mainly proteins) and anionic species (particularly SO$_4^{2-}$) contained in the feedstock under anaerobic conditions by sulphate-reducing microorganisms [13,14,15,16]. Meanwhile, inorganic sulphur, particularly sulphates, can also produce H$_2$S through a biochemical conversion [14].

Furthermore, hydrogen sulphide has been known to raise environmental concerns due to their conversion to harmful sulphur dioxide (SO$_2$) and sulphuric acid (H$_2$SO$_4$) [7,14,17]. In fact, H$_2$S at 1000-3000 ppm concentration may cause an instantaneous death due to a reaction between H$_2$S and enzymes in the bloodstream that inhibits cellular respiration, resulting in pulmonary paralysis, sudden collapse, and death [13,18,19]. Besides, the combustion of fuels containing H$_2$S at a high concentration has been discovered to increase SO$_2$ emissions, which is a precursor of acid rains and brings serious damages to the vegetation and civil constructions. The gas is also known to be highly corrosive for most equipment, including pipelines, pump, compressors, gas storage tanks and engines, and act as a strong poison for fuel cells and reformer catalysts [7,13]. Due to the potential problems that hydrogen sulphide can cause; desulphurization is recommended to be conducted early in a biogas upgrading process. Practically, various H$_2$S removal technologies largely depend on the end uses, composition, variability and volume of biogas being treated, the concentration of H$_2$S present in the gas, and the absolute quantity of H$_2$S to be removed [14].

| Technology                  | H$_2$S tolerance (ppm) |
|-----------------------------|------------------------|
| Heating and Stirling engines| <1,000                 |
| Kitchen stoves              | <10                    |
| Internal combustion engine  | <500 (depends on the type of the engine, it can be <50) |
| Turbines                    | <10,000                |
| Micro-turbines              | <70,000                |
| Fuel cells                  |                        |
| PEM                         | <1                     |
| PAFC                        | <20                    |
| MCFC                        | <10                    |
| SOFC                        | <1                     |
| Natural gas upgrade         | <4                     |
The recommended level of H₂S in biogas being produced is between 0.02-0.05 % (w/w) (200-500 ppm), while H₂S-free biogas is more desirable [13,20]. The H₂S requirement varies based on application, equipment and vendor. Table 2 outlines the typical tolerance of H₂S levels for different biogas utilization equipment [14].

3. Biogas Purification with H₂S Removal
In general, H₂S removal methods are distinguished into physical-chemicals and biotechnological ones. In fact, hydrogen sulphide can be removed either in a digester, from crude biogas, or during an upgrading process [14]. In the past two decades, biotechnological methods have been attracting more attention due to its considerably higher efficiency (> 99 %) and lower operating cost than physical-chemical methods, while they do not generally produce secondary streams that require specific treatments [14].

Technically speaking, the cleaning of biogas is required since the energy content of biogas is in a direct proportion to the methane concentration. Hence, H₂S removal in the cleaning process is expected to increase energy content of the gas [9]. Various techniques have been developed to purify biogas from H₂S contamination. This review on existing desulphurization techniques is conducted to identify the most promising option(s) for biogas application. Existing biogas cleaning techniques, especially those based on H₂S removal, are summarized in the following sections.

4. H₂S Removal During Digestion
H₂S can be treated directly in a digester vessel. Existing methods to remove H₂S in the digester include air/oxygen dosing to the digester and adding iron chloride into the digester. In practices, sulphides either react with metal ions to form insoluble metal sulphides or are oxidized into elementary sulphur [3]. In fact, H₂S removal during digestion has been considered to have a low investment cost since the only required equipment is a dosing system. Since its operational costs strongly depend on the amount of hydrogen sulphide formed by the digestion processes, however, the use of protein-rich substrates and other sulphur-containing molecules will eventually increase the total costs significantly [21]. By adding 5 to 10 % air/oxygen to the digester, H₂S content of biogas can be reduced from 2000 to 3000 ppm to 50 to 100 ppm. In other experiments, the H₂S content can be reduced from 800 ppm to 10 ppm [21]. The air/oxygen dosing technique is based on biological aerobic oxidation of H₂S into elemental sulphur by a group of sulphide-oxidizing microorganisms (Thiobacillus), which are autotrophic and use CO₂ from the biogas to cover their needs of carbon [3]. The following reaction occurs in the biogas:

\[ 2\text{H}_2\text{S} + \text{O}_2 \rightarrow 2\text{S} + 2\text{H}_2\text{O} \]  

Furthermore, the limitation of this method includes requirement of an additional treatment to maintain the quality of gas pipeline, while the addition of air/oxygen may also reduce the effectiveness of following upgrading and cleaning processes [21]. Other than air/oxygen, iron chloride in the form of liquid FeCl₂ or FeCl₃ in solid form can also be dosed directly into the digester or through an influent-mixing tank. They react with H₂S present in the biogas to form FeS (Equation 2 and 3). The method is very efficient in reducing high concentrations of H₂S and causes no air content in biogas. The addition of iron chloride, however, can cause a change in pH/temperature in the digester, which is not beneficial for anaerobic digestions [3].

\[ 2\text{Fe}^{3+} + 3\text{S}^{2-} \rightarrow 2\text{FeS} + \text{S} \]  

\[ \text{Fe}^{2+} + \text{S}^{2-} \rightarrow \text{FeS} \]  

The addition FeCl₂ or FeCl₃ into a digester is related to the hydrogen sulphide equilibrium between liquid and gas, while the requirement for iron can be determined by following Equation 4 ([14]; [22]).

\[ \text{Fe} = \beta \frac{\nu \text{Fe}}{\nu \text{M}_s} \left( \frac{S_{\text{agg}}}{f_{\text{H}_2\text{S}}} + \frac{S_g \Delta H_2}{1000} \right) \cdot \rho_{\text{H}_2\text{S}} \cdot V_{\text{biogas}} \]
Where Fe is iron ions (g/d), $H_2S_{aq}$ is total dissolved hydrogen sulphide (g/m$^3$), $f_{H2S}$ is portion of the total sulphur dissolved as $H_2S_{aq}$, $\Delta H_2S_g$ is amount of $H_2S$ removed from biogas (ppmv), $V_{substrate}$ is flow of substrate (m$^3$/d), $V_{biogas}$ is biogas flow (m$^3$/d), $P_{H2S}$ is $H_2S$ density (g/l), $M_{Fe}$ is iron molecular mass (g/mol), $M_s$ is sulphur molecular mass (g/mol), and $\beta$ is factor of over dosing which is 1.7–2.3 [22] or 3–5 [23].

5. $H_2S$ Removal After Digestion

On the other hand, conventional post-digestion $H_2S$ removal methods include adsorption desulphurization (e.g. metal oxides and zeolite), biological desulphurization, and chemical scrubbing. In practices, each method offers both advantages and disadvantages for the process of $H_2S$ removal from biogas.

5.1. Adsorption

5.1.1. Adsorption using iron oxide.

As one of the oldest methods still in practice, iron oxides technically remove hydrogen sulphide by forming insoluble iron sulphides. In a process so-called “iron sponge” [3,14], hydrogen sulphide reacts with iron oxide to form iron sulphide. Either rust steel wool, impregnated wood chips, or pellet can be used in the process to form reaction beds. In fact, this iron oxide removal technique has been recognized as being simple and may reach up to 99.98 % effectiveness in achieving less than 1 ppm of $H_2S$ concentrations (related to 1,000 ppm $H_2S$ in the raw gas stream). However, this process is largely chemical intensive, limiting its usages due to high operating costs and continuous streams of accumulated waste materials. Chemically speaking, iron oxide reacts with $H_2S$ in biogas according to the following reaction:

$$Fe_2O_3 + 3H_2S \rightarrow Fe_2S_3 + 3H_2O \quad (5)$$

In practices, the reaction is slightly endothermic and optimal to occur between 25-50°C [3]. Besides, the regeneration of iron oxide by using oxygen is possible for a limited time and occurs based on the following reaction:

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

5.1.2. Adsorption using a molecular sieve and activated carbon.

Applying molecular sieves (a.k.a. zeolites), which are highly porous, has been known as being effective to separate various compounds in gas streams. Technically, the selective adsorption is achieved by using molecular sieves with different mesh sizes and/or by adjusting gas pressures [14]. The zeolites have a large surface area with a high degree of localized charges, which allows a strongly adsorbing polar or polarizable compounds such as $H_2S$ and other sulphur compounds [24]. In general, the adsorption is dependent on hydrogen bonding between hydrogen and hydrogen groups in the sulphur and zeolitic molecular sieve, which is reversible in certain conditions, forming varied structures with different strengths [11]. Currently, 194 unique zeolite frameworks and over 40 naturally occurring zeolites have been recognized to function in the removal of $H_2S$ [24].

Besides zeolites, activated carbon (AC) has been widely applied for $H_2S$ removals if low concentrations are required. Technically, AC provides a catalytic surface for oxidation processes to elemental sulphur and sulphate, which significantly enhances the removal capacity of $H_2S$. In practices, the AC must have 20–30% of moisture content and a certain volume of oxygen. In the presence of oxygen, the following reaction takes place:

$$2H_2S + O_2 \rightarrow \frac{1}{2} S_8 + 2H_2O \quad (7)$$

In short, the adsorption of hydrogen sulphide on AC is catalytic, in which carbon acts as the catalyst. The carbon is often impregnated with alkaline or oxide coatings to increase reaction rate. Besides potassium iodine, sodium hydroxide, sodium carbonate, potassium hydroxide and metal
oxides are the most commonly employed coatings. In general, impregnated product is expected to enhance the capacity for H₂S removals from a normal value at 10–20 kg H₂S/m³ carbon for untreated carbon to 120–140 kg H₂S/m³ for impregnated carbon [14].

5.2. Absorption/Scrubbing
During an absorption process, H₂S is removed in water or other organic solvents such as methanol and ethers of polyethylene glycol (physical absorption), while in a chemical absorption the water solubility of H₂S is enhanced by utilizing alkaline water or an oxidation to more water-soluble compounds [14]. The absorption process has been acknowledged to deliver a high removal efficiency (up to 99 %), small footprints and an ability to handle a wide concentration range of pollutants. Technically, absorption processes are suitable for approximately 100–10,000 m³/h flow rate and 8–30 g/m³ concentration of pollutants [14].

However, these processes require considerably high investment costs and high consumptions of water and/or chemicals. In general, absorption-based processes are not feasible for low-flow and low-pressure applications in small biogas plants due to expectedly increasing costs of operation at high pressure, higher energy required for recirculation pumps and regeneration vessels, and higher media costs. Nevertheless, some of them, e.g. iron-chelated process, are feasible for small biogas systems [14,25]. For large-scale biogas plants, on the contrary, these methods are economically more feasible.

Furthermore, the most widely applied absorption process is water scrubbing that consists of two types of processes, i.e. single pass absorption (Figure 1) and regenerative absorption (Figure 2) [1,3,26]. In single pass absorption, an approximately 150 l/Nm³ of water consumption are required, which is considerably high. Due to the high consumption of water and a considerably large power consumption associated with pumping and circulating flows, however, the water scrubbing does not offer competitive advantages to other methods.

Next, adding chemicals into absorption column has been acknowledged to improve absorption process, resulting in lower water and energy consumptions due to a reduced pumping [3]. Besides, physical absorption by applying organic solvents instead of water has been stated as being comparable to water scrubber technique. Figure 3 exhibits a basic scheme of the chemical/physical absorption. In general, commonly applied liquids for chemical absorption include NaOH solution and several chelated iron salt solutions, e.g. FeCl₂, and Fe(OH)₃. Technically speaking, the oxidation of hydrogen sulphide into elemental sulphur is attained by the reduction of soluble ferric chelated iron [Fe³⁺] into ferrous chelated iron [Fe²⁺] according to the following reaction:

Absorption: \( \text{H}_2\text{S} + 2\text{Fe}^{3+} \rightarrow \text{S} + 2\text{Fe}^{2+} + 2\text{H}^+ \) (8)

Regeneration: \( 2\text{Fe}^{2+} + 0.5\text{O}_2 + 2\text{H}^+ \rightarrow 2\text{Fe}^{3+} + \text{H}_2\text{O} \) (9)

Despite the high efficiency of chemical absorption at up to 99.9 %, challenges in using the system may include foaming and plugging problems [14].

5.3. Biological purification

5.3.1. Biofilter and biotrickling filters.
Biofilters and biotrickling filters are technically a set of fixed-bed reactors where microorganisms are immobilized (Figure 4). In the process, contaminated gases flow through a porous material, in which biological oxidation occurs after the gases have diffused into the microorganisms-synthesized biofilm [27]. Therefore, H₂S in biogas is transferred from the gas phase into biofilm, where it is used as an energy source by the microorganisms to produce sulphur or sulphate [14,27].

Parameters influencing the process include bed medium, moisture content, temperature, pH, contact time, nutrient and oxygen levels. Bacteria used for the H₂S removal are normally aerobic, hence requiring a continuous oxygen supply. The conventional way of supplying oxygen into a biofilter/biotrickling filter is taken by a direct air injection (4 to 10 %) into the gas stream [14].
The primary difference between biofilters and biotrickling filters is their carrier material, i.e., organics in biofilters and inert in biotrickling filters. The most commonly used bed materials in biofilters are peat, compost and wood bark, while inorganic inert carriers such as ceramic, pozzolan, marble are also used, which may present some advantages compared to organic carriers [27]. On the other hand, the packed bed in biotrickling filters is usually made up of an inert material, e.g., a randomly dumped plastic packing, structured plastic packing, open-pore synthetic foam, lava rock, glass or rock wool, Tire derived rubber particles (TDRP), glass beads or ceramics.

Furthermore, the use of biofilms generated by single bacterial cultures in biofilters or biotrickling filters has been suggested to improve filtration efficiency and removal capacities. It also leads to the shortening or a complete elimination of bacterial lag phase, as well as to increase efficiency in the removals of H₂S and other organosulphur compounds. In general, various types of chemoheterotrophic microorganisms, e.g., *Pseudomonas acidovorans* DMR-11 and *Pseudomonas putida*, have been recognized to deliver degrading activities on H₂S and organic sulphur compounds.
In particular, bacteria from genus *Acidithiobacillus*, including *A. thiooxidans* that uses H$_2$S as an energy source, offer an advantage due to their low nutritional requirements ([28][29][30]). Other bacterial species that have been investigated for the removal of sulphur reduced compounds are *Thiobacillus denitrificans* [31], *A. ferrooxidans* [32], and *T. novellus* [33].

5.3.2. Bioscrubber.

Then, bioscrubber is technically stated as a three-phase, fluidized-bed bioreactor consisting of two sub-systems, i.e. an absorption tower where pollutants are absorbed into a liquid phase, and a bioreactor, which is a kind of activated sludge unit [27][34]. In a bioscrubber, gas contaminations are removed by both physical and biochemical processes. Heterotrophic or autotrophic microorganisms living in a bioreactor are responsible for the significant conversion of odorous compounds contained in the aqueous phase [35], while microorganisms growing in suspended flocks in the water degrade the...
pollutants. Then, effluents produced in the unit are re-circulated over the absorption tower. Nutrient additions, oxygen and pH are continually monitored and controlled to maintain active microbial growths and high biological activity. Excess biomass and by-products are continually purged from the system. An advantage offered by this technique compared to biofilters includes no injection of oxygen or nitrogen into biogas stream. Meanwhile, its disadvantage includes a higher specific cost. In general, H₂S content in biogas is reduced to about 10-100 ppmv, while in some cases low ppmv levels may also be achieved. Then, gas flows normally range from 200 to 2,500 m³/h [36].

6. Conclusion
Cleaning and upgrading biogas quality is important in order to meet the requirement of biogas as an alternative energy source substituting the conventional fossil fuels. These processes are needed so that biogas can be used such as to produce heat and stream, electricity, vehicle fuel, and feedstock for the production of bio-based chemicals and substrate in fuel cells. The enhancement in technology for cleaning and upgrading biogas in the future is expected. Although the production of biogas is a mature technology that is established worldwide, the commercial utilization is still limited as the gas need to be purified before on-site use. Although various cleaning and upgrading technologies have been developed, technology can be chosen according to the highest achievable methane content and the type of technique that is implemented depends on desire product, economical and possibly ecological issues. The presence or the absence of supplier for the technology in a particular country could also determine the possibility of chosen technique.

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Author Contribution
D. Andriani is contributed as the main contributor of this work. All authors read and approved the final paper.

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