



## Review

## Biogas as a renewable energy fuel – A review of biogas upgrading, utilisation and storage



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## ABSTRACT

Biogas upgrading is a widely studied and discussed topic and its utilisation as a natural gas substitute has gained a significant attention in recent years. The production of biomethane provides a versatile application in both heat and power generation and as a vehicular fuel. This paper systematically reviews the state of the art of biogas upgrading technologies with upgrading efficiency, methane (CH<sub>4</sub>) loss, environmental effect, development and commercialisation, and challenges in terms of energy consumption and economic assessment. The market situation for biogas upgrading has changed rapidly in recent years, making the membrane separation gets significant market share with traditional biogas upgrading technologies. In addition, the potential utilisation of biogas, efficient conversion into bio-compressed natural gas (bio-CNG), and storage systems are investigated in depth. Two storing systems for bio-CNG at filling stations, namely buffer and cascade storage systems are used. The best storage system should be selected on the basis of the advantages of both systems. Also, the fuel economy and mass emissions for bio-CNG and CNG filled vehicles are studied. There is the same fuel economy and less carbon dioxide (CO<sub>2</sub>) emission for bio-CNG. Based on the results of comparisons between the technical features of upgrading technologies, various specific requirements for biogas utilisation and the relevant investment, and operating and maintenance costs, future recommendations are made for biogas upgrading.

## 1. Introduction

Biogas is produced by anaerobic degradation of organic compounds and could be the substitute for natural gas and fossil fuels. It contains mostly three components, which are methane (CH<sub>4</sub>), carbon dioxide (CO<sub>2</sub>) and nitrogen (N<sub>2</sub>). However, other trace species exist as well, which are hydrogen sulphide (H<sub>2</sub>S), hydrogen (H<sub>2</sub>), nitrogen (N<sub>2</sub>), ammonia (NH<sub>3</sub>), oxygen (O<sub>2</sub>) and carbon monoxide (CO). Furthermore, typical biogas is saturated with water, dust particles, siloxanes, aromatic and halogenated compounds [1,2], but the amounts of these trace

compounds are very low compared to CH<sub>4</sub> and CO<sub>2</sub>. Various biogas sources with their impurities levels are shown in Table 1.

Biogas can play a major role in the developing market for renewable energy and it is estimated that biogas usage in the world will be doubled in the coming years ranging from 14.5 gigawatts (GW) in 2012 to 29.5 GW in 2022 [7,8]. The demand for renewable fuels is increasing with growing concern about environmental problems due to the high greenhouse gases (GHGs) emission from fossil fuel combustion [9–12]. Purified biogas can be used in various applications such as the production of electricity, heat and steam generation in household and

**Abbreviations:** AS, amine scrubbing; AMDEA, activated methyl diethanolamine; Bio-CNG, bio-compressed natural gas; CA, cellulose acetate; CSP, chemical scrubbing process; CNG, compressed natural gas; CHP, combined heat and power; CMS, carbon molecular sieve; CNT, carbon nanotubes; CCM, carbon cryogel microspheres; CS, cryogenic separation; CXM, carbon xerogel microspheres; DEA, diethanol amine; ESA, electrical swing adsorption; EU, European Union; GW, gigawatts; GHG, greenhouse gas; HPWS, high pressure water scrubbing; ISS, inorganic solvent scrubbing; IEA, international energy agency; LCA, life cycle assessment; LBM, liquefied biomethane; LNG, liquid natural gas; MMM, mixed matrix membrane; MOF, metal-organic framework; MEA, monoethanol amine; MDEA, methyl diethanol amine; Mtoe, million tons of oil equivalents; MS, membrane separation; NGV, natural gas vehicle; NYT, neapolitan yellow tuff; NMP, N-methyl pyrrolidone; OPS, organic physical scrubbing; PSA, pressure swing adsorption; PSf, polysulfone; PI, polyimide; PC, polycarbonate; PDMS, polydimethyl siloxane; PEG, polyethylene glycol; PZ, piperazine; SGC, Swedish gas technology centre; TSA, temperature swing adsorption; VOC, volatile organic compounds; ZIFs, zeolitic imidazolate frameworks

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**Table 1**  
Guidelines for impurities removal for specific biogas applications.

Biogas	CH <sub>4</sub> (%)	CO <sub>2</sub> (%)	N <sub>2</sub> (%)	O <sub>2</sub> (%)	H <sub>2</sub> S (ppm)	Benzene (mg m <sup>-3</sup> )	Toluene (mg m <sup>-3</sup> )	Ref.
Landfills	45–62	24–40	1–17	1–2.6	15–427	0.6–35.6	1.7–287	[2–4]
Sewage digesters	58–65	33–40	1–8	< 1	0–24	0.1–0.3	2.8–11.8	[2,5,6]
Organic waste digesters	60–70	30–40	1	1–5	10–180	0.1–1.1	3–7	[2]

industry, injection into the natural gas grid as well as a vehicular fuel. Biogas production in Europe was estimated at 6 million tons of oil equivalents (Mtoe) in 2007 and it is expected to increase to 23 Mtoe by 2020 [13] and [14,15]. As a result, 60% reduction in GHGs emission is expected after 2017 [16]. In addition, the European Union (EU) countries have set a goal of supplying 20% of European energy demand using renewable energy systems (RES) by 2020 and at least 25% of bioenergy will be produced by biogas [17,18]. Besides, it is estimated that the world share of bio-compressed natural gas (bio-CNG) in all vehicular fuels will rise up from 2% today to 27% in 2050 [19,20]. Global climate changes caused by CO<sub>2</sub> emissions are currently debated around the world. Therefore, greener sources of energy are being required as alternatives to replace fossil fuels [21–24]. Different technologies for biogas upgrading have been developed to date and some of them are commercially available. The technologies that are used commercially for biogas upgrading today are pressure swing adsorption, high pressure water scrubbing, organic solvent scrubbing, amine scrubbing, membrane separation and cryogenic separation which are briefly described in the next section. The selection of suitable technology by considering the efficiency and economy of a specific application is important [25,26].

IEA Bioenergy, an international collaboration on bioenergy under the International Energy Agency (IEA), has investigated various renewable energy tasks, concentrating on updating the raw biogas for various applications but did not intend to give detailed descriptions of the technical or economic performance of the technologies [27,28]. The Swedish Gas Technology Centre (SGC) is another group focused on biogas upgrading research and it has published several reports on commercially available technologies [29,30]. Biogas purification and upgrading have also been a highlighted topic in scientific articles in recent years [8,23,31–39]. Xiu and Shahbazi [36] summarised the state of the art technology for producing and upgrading bio-oil, with the focus on the hydrothermal liquefaction process. Abatzoglou and Boivin [31] reviewed biogas purification with the focus on the removal of contaminants, such as H<sub>2</sub>S, NH<sub>3</sub>, and siloxanes, but the removal of CO<sub>2</sub> was only briefly mentioned. Weiland [32] presented an overview of the complete biogas production and consumption chain but did not focus on currently available upgrading technologies. Bekkering et al. [33] studied the current status and future options of biogas upgrading technologies but did not present the technical performance and economic report on various upgrading technologies. Ryckebosch et al. [35] reviewed different biogas upgrading technologies with the focus on their operating conditions, drawbacks, and efficiency. Pertl et al. [34] and Starr et al. [37] applied life cycle assessment (LCA) to biogas upgrading. Bauer et al. [38] found that the market shares for biogas upgrading technologies have been changed rapidly in recent years, amine scrubbing is continuously achieving significant market shares, and a competition between pressure swing adsorption (PSA) and high pressure water scrubbing (HPWS). Kárászová et al. [8] reviewed membrane separation processes for biogas and found that membrane gas permeation is able to compete with classical biogas upgrading technologies. However researchers still need to solve the challenges in using membrane for the removal of volatile organic compounds (VOC) and siloxanes from raw biogas. Sun et al. [23] encouraged more researches on membrane separation process for economical biogas upgrading and its utilisation as a vehicular fuel as it is more beneficial for the environment. Chen et al. [39] revealed that hybrid processes for biogas

upgrading are more efficient, where membrane separation is combined with absorption, adsorption, and cryogenic technique. This combined separation processes can improve the performance and reduce the operational cost of the process. Although the production of biogas is a well-established technology, its commercial utilisation as a vehicular fuel is still limited because high purification is needed.

Meanwhile, the existing reviews and studies have explored the concepts and comprehensively investigated the techno-economic performance of biogas upgrading technologies, their developments, energy requirements, market shares, environmental analysis, utilisation of upgraded biogas, and conversion and storage of bio-CNG. Purified bio-CNG is a substitute for CNG for automobiles [16,40]. Biogas utilisation as a vehicular fuel is beneficial since the vehicles using bio-CNG have CO<sub>2</sub> emission 80% less than those using fossil fuels [41,42]. Sweden and Germany are among the countries that already used bio-CNG as vehicular fuel in the form of pure methane or mixed with natural gas [8]. Also, the total global warming related to the bio-CNG utilisation is approximately 20% less than that of CNG [16]. Bio-CNG storage is also critical and important step as it affects vehicle filling time, mass of gas, temperature, entropy generation, and energy consumption [43]. Therefore, one of the main contributions of this work is to provide insights and guidelines regarding the biogas upgrading technology selection based on the specific utilisation, efficiency, investment cost, and operational and maintenance cost. In addition, this work also summarizes the biogas utilisation and its conversion into bio-CNG and highlights the potential benefits of bio-CNG as a vehicular fuel. Bio-CNG has been explored as an alternative to fossil fuels. Lastly, various methods of storage for bio-CNG are comprehensively discussed and compared. Finally, based on the investigation and recommendations main conclusions are drawn in this work.

## 2. Biogas upgrading and purification technologies

The technologies currently developed and available on the industrial scale for the upgrading of biogas include adsorption, absorption (physical and chemical), membrane separation, and cryogenic. These technologies are primarily used for CO<sub>2</sub> separation while the pre-upgrade stage is required to reduce the high concentrations of contaminants such as H<sub>2</sub>O, H<sub>2</sub>S, and siloxanes. Further classification of these upgrading technologies is shown in Fig. 1. Also, major strengths and weaknesses of these existing upgrading technologies are summarised in Table 2.

### 2.1. Pressure swing adsorption (PSA)

Adsorption process involves the transfer of solute in the gas stream to the surface of an adsorbent material due to physical or van der Waals forces. In pressure swing adsorption (PSA), some undesirable gases like CO<sub>2</sub> are separated from biogas under elevated pressure using adsorbent materials. Later, the pressure is reduced to desorb the adsorbed gases [44,45]. Carbotech, Acrona, Cirmac, Gasrec, Xebec Inc., and Guild Associates are well-known companies which develop and commercialise this technology at low and high capacity (flow rate of 10–10,000 m<sup>3</sup>/h of biogas). In PSA, H<sub>2</sub>S gas removing is a primary step because it is considered as toxic to the process and adsorption of this gas is normally irreversible [46]. Fig. 2 shows a simplified process flow diagram for a PSA process.

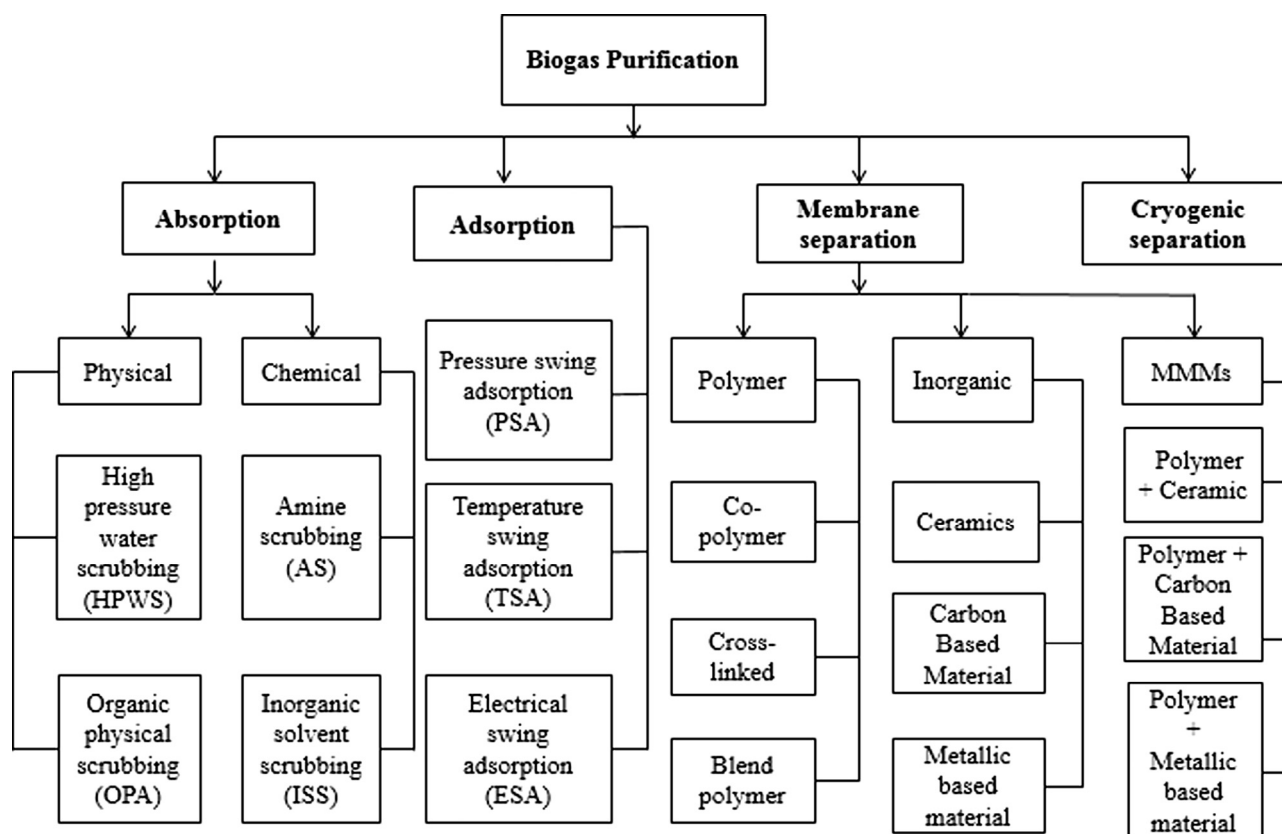


Fig. 1. Current technologies for biogas upgrading.

In this process, high methane concentration can be achieved (95–99%). However, this process needs an extensive process control and requires high investment and operational costs [44,47]. Also, impurities in the raw biogas stream affect the efficiency of the process [48]. To reduce  $\text{CH}_4$  losses in the desorbed gas, the system should be designed in such a way that desorbed gases recirculate to PSA process. Temperature swing adsorption (TSA) and electrical swing adsorption (ESA) are the other types of adsorption (Fig. 1). The temperature in TSA is enhanced at constant pressure, while in PSA, the pressure is reduced [49,50]. TSA requires thermal energy to regenerate the adsorbent material while in PSA compression energy is used. Thus, TSA could be a better option if a cheap source of energy is available. Carbon cryogel microspheres (CCM) and carbon xerogel microspheres (CXM) have been identified as efficient adsorbent materials for TSA process due to their highly porous and stable structure [51]. In ESA, the regeneration is carried out by passing electricity through the saturated adsorbent and the heat generated by Joule effect facilitates the release of  $\text{CO}_2$  [52]. Although this process has the potential to reduce the cost of  $\text{CO}_2$  capture compared to TSA and PSA, electrical conductivity is required for the adsorbent used. Activated carbon has been developed as a new type of semiconductor adsorbent with large surface area and micropore volume [53,54].

### 2.1.1. Adsorbents

By a proper choice of adsorbent, the process can remove  $\text{CO}_2$ ,  $\text{H}_2\text{S}$ , moisture, and other impurities from biogas either selectively or simultaneously. The adsorbent materials are the core of PSA process thus their engineered selection is very important to achieve high selectivity of  $\text{CO}_2$ . Molecular sieve materials such as zeolites and activated carbon are commonly used adsorptive materials for biogas upgrading [55]. The pores of these adsorbents are responsible for easy penetration of  $\text{CO}_2$  while retaining  $\text{CH}_4$  molecules. This is due to different sizes of  $\text{CO}_2$  and  $\text{CH}_4$  molecules and also their adsorption capacity [56,57]. Cationic

zeolites are commercially available adsorbents for biogas upgrading due to their small pores. Zeolites denoted with the LTA (Linde type A) IZA code, like Linde 4A and 5A types, are reported in the literature to be used for the upgrading of biogas and in particular, for the separation of  $\text{CO}_2$  from  $\text{CH}_4$  [58–61]. Recently, 13 X molecular sieves are applied successfully to upgrade biogas with moderate resistance to humidity [54]. Pandey and Fabian [62] used zeolite-Neapolitan Yellow Tuff (NYT) for the purification of natural gas and reported that 0.4 kg of  $\text{CO}_2$  could be adsorbed per kg of NYT at ambient conditions. During this process, the  $\text{H}_2\text{S}$  was also reduced. Zeolitic imidazolate frameworks (ZIFs), a sub-class of metal-organic frameworks (MOFs) have recently gained much attention to be used for the adsorption of  $\text{CO}_2$  due to their intrinsic characteristics such as tuneable pore size, large pore volumes, and large surface area [63]. Biogas can also be upgraded using silica, alumina, activated carbon, or silicates, which are also known as molecular sieves [64].

The adsorbent material used for this process is desired to have the following properties:

- (1) The surface of the adsorbent should be basic so can be easily attracted by  $\text{CO}_2$  acidic gas. These types of materials are known as equilibrium-base adsorbents because main driving factor is the differences of interaction forces between  $\text{CO}_2$  and  $\text{CH}_4$  with the surface of the adsorbent.
- (2) The pores of the adsorbent should be modified in such a way that  $\text{CO}_2$  (kinetic diameter of 3.4 Å) can easily penetrate into their structure while larger  $\text{CH}_4$  molecules (kinetic diameter of 3.8 Å) are constrained to pass through them. These materials are reported as kinetic adsorbents since its main selectivity is due to diffusion constrain.
- (3) Material should be easily regenerated or desorbed with low energy demand.
- (4) It should have a good moisture removal capacity.

### 2.2. Absorption

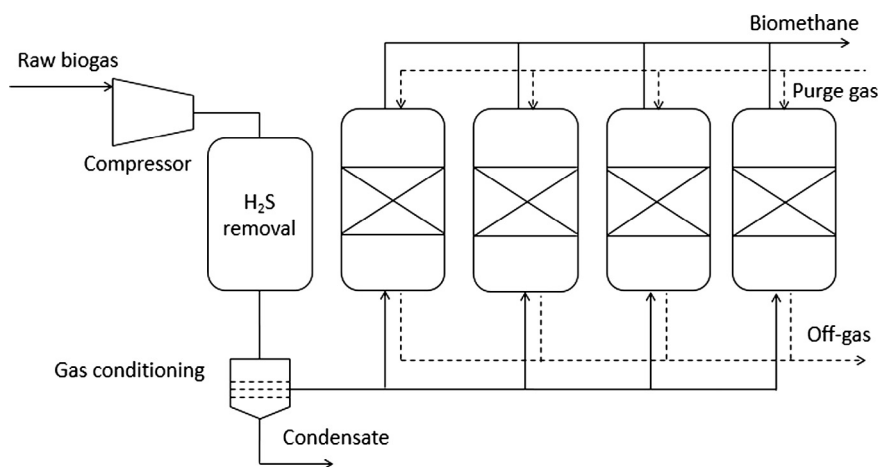
Absorption depends on the solubility of various gas components in a

**Table 2**  
Advantages and disadvantages of different biogas upgrading technologies.

Technology	Advantages	Disadvantages
PSA	95–99% CH <sub>4</sub> concentration [39] The humidity of the raw biogas can be removed [44] Less energy demand with low emissions, elimination of nitrogen and oxygen is also possible [55] Clean and water-free gas [48] Relatively fast installation and easy start up [38]	High capital investment and operational costs (due to a number of columns in PSA unit) [39] H <sub>2</sub> S elimination step is needed and tail gas from the process needs to be treated [27,55] Water should be removed before PSA process [44] Susceptible to fouling by impurities in the biogas stream [48] High CH <sub>4</sub> losses when valves malfunction [38]
HPWS	> 97% CH <sub>4</sub> concentration [39] Removal of both CO <sub>2</sub> and H <sub>2</sub> S [55] No special handling and chemicals are required [48] Easy in operation with low CH <sub>4</sub> losses (< 2%) [23] Tolerant for impurities [35] Regeneration of water is possible [32]	High investment and operating costs [39,55] Less efficient [48] Low flexibility toward variation of input gas [23] Slow process [1] High pressure, need higher energy to compress the gas and to pump water [35] Requires a lot of water even with regeneration [32] Clogging due to bacterial growth [39] Corrosion problem due to H <sub>2</sub> S [31]
OPS	> 97% CH <sub>4</sub> concentration [39] Remove organic components such as H <sub>2</sub> S, NH <sub>3</sub> , HCN, and H <sub>2</sub> O [48] Low CH <sub>4</sub> loss [38]	Complex operation with high investment and operational costs [39] Expensive for small -scale applications [48] Reduced operation when dilution of glycol with water [35] Need high energy to regenerate the solvent [38] Solvent is expensive and difficult to handle [8] Solvent regeneration is difficult if H <sub>2</sub> S is not removed first [33]
CSP	> 99% CH <sub>4</sub> concentration with low operational costs [39]  Complete H <sub>2</sub> S removal is possible and can operate at low pressure [55]  High selectivity for CO <sub>2</sub> with low CH <sub>4</sub> loss [48] The process is faster than water scrubbing and solvent is easier to regenerate [1]	High investment cost and heat is required for regeneration of solvent [39,55,67] Problems of contaminant build-up, corrosion, and amine breakdown [48] Waste chemical may require treatment [35]
MS	Less operational and capital investment costs and high CH <sub>4</sub> recovery up to > 96% [39] Small space requirements and available at low capacities [8,30]. Easy maintenance without hazardous chemicals [82] Low maintenance cost [100] Simple and environmentally friendly process [101] Ease of process with low energy consumption [102] Fast installation and start up [55] Highly reliable and cheap process [103]	For high purity product, multiple steps of membrane are required [39] Low CH <sub>4</sub> yield in single step [55] Low membrane selectivity [103] Not suitable for high purity needs [31] Consumes more electricity per unit of gas produced [1]
CS	High purity of CH <sub>4</sub> with 98% concentration [35] CO <sub>2</sub> purity is also high and can be used as a dry ice [1] Low energy and cost is required to obtain highly pure liquefied biomethane (LBM) with less than 1% CH <sub>4</sub> loss [104] Environmentally friendly technique with no chemicals use [46]	High investment, maintenance and operational costs [104] High energy requirements [1] Use of different expensive process equipment [38]

liquid solvent. Raw biogas meets a counter-flow of liquid in a column that is filled with packing material to increase the contact area between gas and liquid. In biogas upgrading, CO<sub>2</sub> is more soluble than CH<sub>4</sub> in

liquid, thus the gas that leaves the column has a high concentration of CH<sub>4</sub> while the liquid leaving the column has an increased concentration of CO<sub>2</sub> [65]. High pressure water scrubbing (HPWS) and organic



**Fig. 2.** Process flow diagram of pressure swing adsorption process.

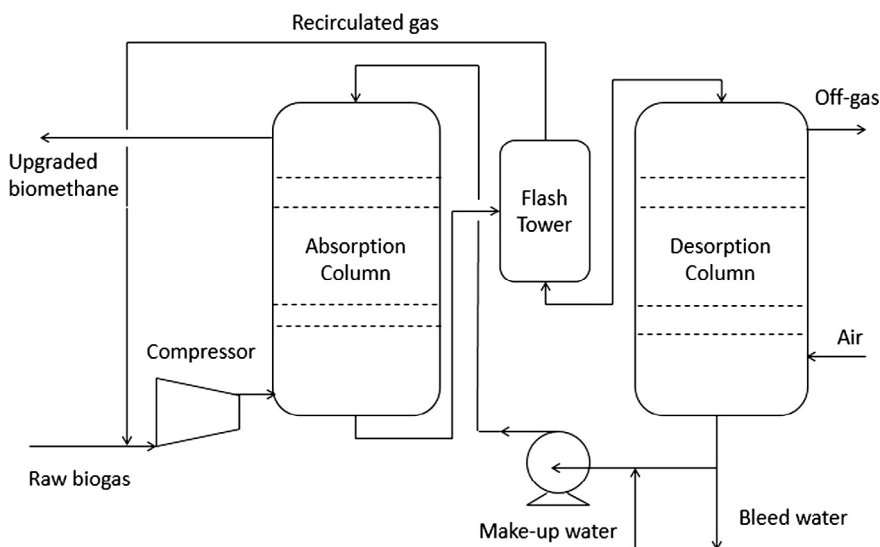


Fig. 3. Process flow diagram of high pressure water scrubbing.

physical scrubbing (OPS) are types of physical absorption, while amine scrubbing (AS) and inorganic solvent scrubbing (ISS) are types of chemical absorption [39] as shown in Fig. 1.

2.2.1. High pressure water scrubbing (HPWS)

High pressure water scrubbing (HPWS) is the most common and well-established technology used to remove CO<sub>2</sub> and H<sub>2</sub>S from biogas since these gases are more soluble in H<sub>2</sub>O than CH<sub>4</sub>. The operating pressure of the HPWS is 10 bar [66] where biogas is fed into the bottom of a packed column while water is fed counter-currently as shown in Fig. 3. The physical absorption of the gases is governed by Henry’s law, which states that at a constant temperature, the amount of any dissolved gas is directly proportional to its partial pressure in the gas stream. It is also observed that solubility of CO<sub>2</sub> can be increased at low temperature [27].

This process also is useful for the removal of H<sub>2</sub>S because H<sub>2</sub>S is more soluble than CO<sub>2</sub> in water [31]. Wastewater leaving the bottom of the scrubber is saturated with CO<sub>2</sub> or H<sub>2</sub>S and a small amount of CH<sub>4</sub> is regenerated and recirculated back to the absorption tower. Regeneration is accomplished by reducing the pressure or by stripping with air in a desorption column. Stripping with air is not recommended when H<sub>2</sub>S concentration level is high since the water rapidly becomes polluted with elementary sulphur which causes corrosion and operational problems. It is preferable to use fresh water if the cheap source of water is available. Produced biomethane is saturated with water so the drying

process is mandatory for final upgrading [67,68]. When a high concentration of H<sub>2</sub>S is present in the gas stream then pre-removing of H<sub>2</sub>S is a mandatory step in water scrubbing process like in PSA [23]. Although it is an eco-friendly and highly efficient process with no special chemicals requirement and high methane recovery (> 97%), high investment and operational cost are required. In addition, high energy consumption is required during water regeneration process, which leads to high costs [69].

2.2.2. Organic physical scrubbing (OPS)

Organic physical scrubbing (OPS) has the same principle as water scrubbing, but it uses an organic solvent instead of water. Various organic solvents such as methanol (CH<sub>3</sub>OH), N-methyl pyrrolidone (NMP), and polyethylene glycol ethers (PEG) are used to absorb CO<sub>2</sub>. The solubility of CO<sub>2</sub> is five times higher in PEG than in water for the same upgrading capacity [70] which results in lower organic solvent demand and less pumping requirement [39,71]. H<sub>2</sub>S, H<sub>2</sub>O, O<sub>2</sub>, N<sub>2</sub>, and halogenated hydrocarbons are also removed together with CO<sub>2</sub>, but the prior removal of H<sub>2</sub>S is recommended for this absorption process [55]. Selexol® and Genosorb® are examples of trade names for PEG liquids used in organic physical scrubbing [48].

The biogas is compressed to 6–8 bar and cooled before it is injected into the bottom of the absorption column. The organic solvent is supplied to the top of the column to make a counter-current flow of gas and liquid. It is also cooled before being injected into the column to

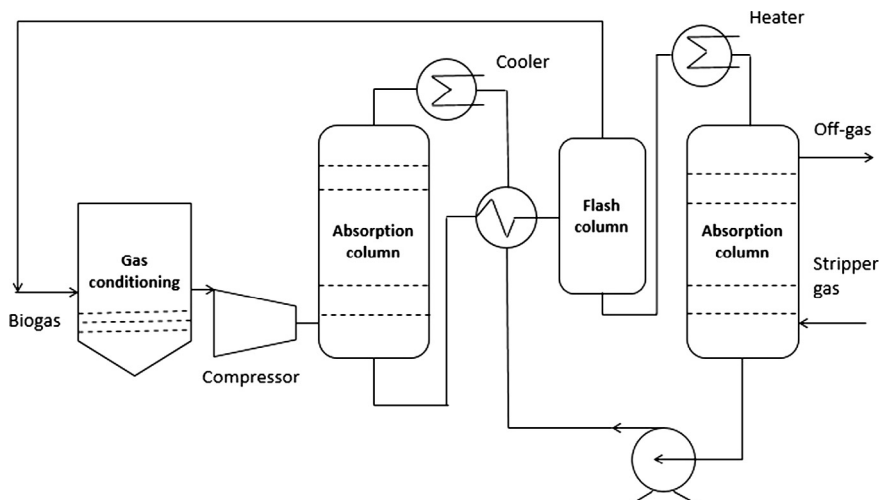


Fig. 4. Process flow diagram of an organic solvent scrubber.

maintain the low temperature in the absorption column around 20 °C (Fig. 4). The organic solvent that leaves the bottom of the absorption column is heat exchanged with the organic solvent that will be injected to the top of the column. Then, the organic solvent is injected into the flash column, where dissolved CH<sub>4</sub> and some CO<sub>2</sub> are released under reduced pressure and circulated back to the inlet of biogas. To regenerate the organic solvent, it is further heated to around 40 °C before entering the desorption column. The solvent is injected into the top of the column and the pressure is decreased to 1 bar. The regenerated solvent is injected to the top of the absorption column. Although this process is characterised by improved efficiency than water scrubbing in terms of CO<sub>2</sub> separation but more energy is required for the solvent regeneration. Also, the cost of the organic solvents is significantly higher than that for water [70].

### 2.2.3. Chemical scrubbing process (CSP)

Chemical absorption involves reversible reaction between absorbed substances and solvent. The most common amines used as solvent for removing acidic gases (CO<sub>2</sub> and H<sub>2</sub>S) are diethanolamine (DEA), monoethanolamine (MEA), and methyl diethanolamine (MDEA) [39]. Nowadays, a mixture of MDEA and piperazine (PZ), called as activated MDEA (AMDEA), is commonly used in this process. The absorption capacity of AMDEA is significantly higher compared to MDEA. The possible reason is the presence of primary and secondary amines in PZ and the tertiary amine in MDEA that gives high reaction rate for CO<sub>2</sub> absorption. Typically, an amine scrubber system (Fig. 5) consists of an absorber, where the CO<sub>2</sub> is absorbed from the biogas and a stripper in which the CO<sub>2</sub> is separated from the waste amine solution by heating under reduced pressure [72]. The raw biogas enters the absorber from the bottom and the amine solution is supplied to the top of the column to make a counter current flow contact. The CO<sub>2</sub> in the biogas reacts with the amine solution and is absorbed. This is an exothermic reaction, which increases the temperature of the absorber from 20–40 to 45–65 °C [73]. Usually, the solubility of CO<sub>2</sub> in H<sub>2</sub>O increases with decreasing temperature [27] but in amine scrubbing (AS), the reaction rate between CO<sub>2</sub> and the amine solution increases with increasing temperature, subsequently gives more absorption of CO<sub>2</sub>. The product gas (CH<sub>4</sub>) exits from the top of the column. The operating pressure of the absorber is 1–2 bar. The liquid from the bottom of the absorber is going through the heat exchanger and pumped to the top of the stripper column, where it is connected with the steam and CO<sub>2</sub> is released. The bottom part of the stripper column is equipped with a reboiler at 120–150 °C where amine solution is boiled. Reboiler provides the heat of reaction for the release of CO<sub>2</sub> from the waste amine solution and regenerates the amine solution. Highly concentrated CH<sub>4</sub> of > 99% gas

is obtained with low operational cost but high investment cost, and significantly a massive heat is required for the regeneration of amine solution [48]. If H<sub>2</sub>S is present in the raw biogas, it will be absorbed by amine solution and higher temperature will be needed for the regeneration to desorb H<sub>2</sub>S. Therefore, it is recommended to remove it prior to the AS process. Another downside of this technology is the requirement to treat waste chemicals, corrosion, and contaminant build-up which makes the process more complex [74].

Inorganic solvent scrubbing (ISS) generally employs an aqueous solution of alkaline salts such as sodium, potassium, ammonium, and calcium hydroxides [75]. The absorption of CO<sub>2</sub> in this alkaline solution is assisted by agitation. The turbulence in the solvent and the contact time between biogas and liquid increase the diffusion of the CO<sub>2</sub> in the solvent [39].

### 2.3. Membrane separation (MS)

During the last 40 years, the membrane-based gas separation process has become the part of market share [76–78]. The membrane acts as a permeable barrier that allows specific compounds to pass through differently and control their permeability based on the applied driving forces such as the difference in concentration, pressure, temperature, and electric charges of different species. Generally, two models are used to explain the membrane separation process such as solution-diffusion and pore-flow model [79]. In the first model, permeates dissolve in the membrane material and then diffuse through the membrane due to concentration difference. Later in this process, permeates are separated by pressure-driven convective flow through small pores [80]. However, the solution-diffusion model is frequently used for gas transportations in polymeric membranes [81]. For biogas upgrading, CO<sub>2</sub> permeates through the membrane while CH<sub>4</sub> retains on the inlet side as retentate (Fig. 6). The membrane gas separation could be more beneficial if the gas flow is low and the inlet CO<sub>2</sub> content is high. These considerations are suitable for a typical biogas upgrading plant [82]. It is a cheap process including low operating and capital costs, less demand for energy, and requires simple and compact membrane equipment set up [83]. Polymeric, inorganic, and mixed matrix membranes (MMMs) are three different types of membrane used for biogas purification as shown in Fig. 1.

Most of the commercial membranes are polymeric made from organic materials such as polysulfone (PSf), polyimide (PI), polycarbonate (PC), polydimethyl siloxane (PDMS), and cellulose acetate (CA) [77,84]. They have excellent mechanical strength, easy to fabricate with low cost, and high selective permeation. CA and PI based polymeric membranes are the mostly used commercial membranes for

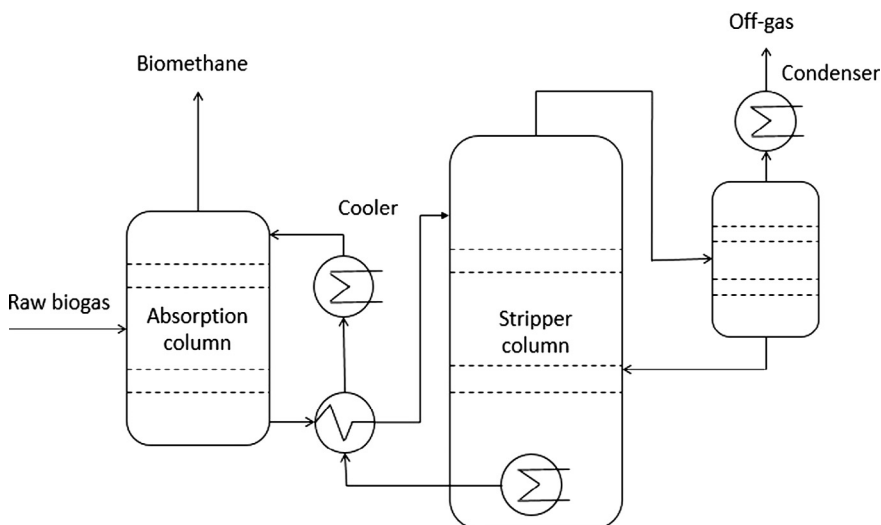


Fig. 5. Process flow diagram of an amine scrubber.

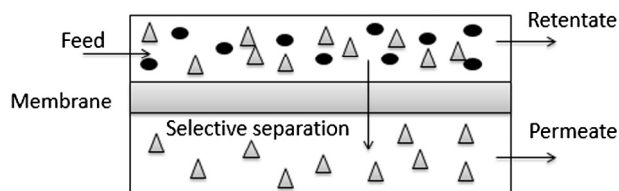


Fig. 6. Scheme of membrane gas separation process.

biogas separation. Polyvinyl amine/polyvinyl alcohol blend membrane was also tested and could achieve 98% CH<sub>4</sub> purity [78]. CA membrane is the first polymeric membrane commercialised for biogas purification [85]. CA is relatively inexpensive due to an abundant and renewable resource of cellulose with attractive separation properties. Therefore, CA membrane has been commercialised for over 30 years. However, CA membranes possess several limitations that restrict its use in gas separation membrane. CA membranes are susceptible to plasticisation ( $P_{\text{plasticisation}} = 8$  bar) [86] due to its -OH rich functional groups that easily dissolve CO<sub>2</sub> within the membrane matrix. PI as a highly crystalline material is attractive for gas separation. It shows high permeability and selectivity as well as high mechanical/thermal stability. Matrimid® is commercially available PI with stiff polymer backbone. Matrimid® is highly rigid and thermally stable, and thus suits the harsh working environment. Despite its prevalence, Matrimid® is significantly expensive and also susceptible to plasticisation ( $P_{\text{plasticisation}} = 17$  bar) [86]. Furthermore, PSf also possesses excellent mechanical strength and high rigidity as well as acceptable gas pair selectivity and thus, becoming one of the most important polymeric membrane materials. Although the separation properties of PSf are still lacking compared to PI-based membrane, PSf is cheaper with high plasticisation resistance ( $P_{\text{plasticisation}} = 34$  bar) [86]. State of the art polymeric membranes are economically competitive in separating CO<sub>2</sub> and H<sub>2</sub>S from the biogas compared to conventional technologies in both capital and operating costs [39]. However, the investigation of polymeric materials for gas separation has been challenged by the upper bound trade-off limit between permeability and selectivity. In fact, highly permeable membranes are commonly accompanied by low gas pair selectivity and vice versa [87].

Inorganic membranes are more advantageous compared to the conventional polymeric membrane as they offer more mechanical strength, thermal stability, and resistance against any chemicals. Mostly, inorganic membranes facilitate permeability and selectivity, exceeding the Robeson upper bound. There are several classes of the inorganic membranes such as zeolite, activated carbon, silica, carbon nanotubes (CNT), and metal-organic framework (MOF). Developing continuous defect-free inorganic membranes, however, is the most crucial step in inorganic membrane fabrication [88]. It is noted that the fabrication of inorganic membranes is a stringent process and requires continuous monitoring due to their fragile structure. For example, a carbon membrane derived from a polymer precursor is often supported by alumina or quartz to provide strength to the membrane before undergoing pyrolysis. The rigid porous materials like carbon molecular sieves and zeolites have difficulties in forming continuous and defect-free membranes for practical applications in spite of their superior gas

separation properties [79,89,90]. It is clear that both polymeric and inorganic membranes have limitations that motivate researchers to develop new membranes. The developments so far are focused on integrating both polymeric and inorganic membranes known as mixed matrix membranes (MMMs). MMMs are composite membranes consisting of polymeric materials as continuous phase with inorganic particle as dispersed phase. It is predicted that the resultant membranes have benefits owing to the combined advantages of both polymeric and inorganic materials and overcome the limitations of each individual phase [91,92]. Significant improvement in MMMs properties is expected due to the superior separation performance of inorganic particles combined with high processability and moderate processing cost of base polymeric membranes. Excellent gas separation properties of MMMs are the main driving force in the development of the novel gas separation membranes [93–95]. The embedment of inorganic filler into polymer matrix would introduce an additional mechanism to improve the membrane performances. For example, using zeolite as a filler for CO<sub>2</sub> removal membrane would provide a molecular sieving mechanism to further discriminate CH<sub>4</sub> while specific interaction with CO<sub>2</sub> provides an improved surface diffusion across the membrane. The presence of particles in the polymer matrix also increases the tensile strength and thermal stability of the membrane. so the membrane can be applied at harsh environment [96–98]. Thus, improvements in biogas separation factor are expected where MMMs are applied. However, more investigations should be performed to enhance filler dispersion and compatibility when it is incorporated into the polymeric phase.

Preremoval of H<sub>2</sub>S from raw biogas is necessary because it can negatively affect the membrane performance. The process for upgrading biogas with membrane technology is shown in Fig. 7. Besides, water, oil droplets, and aerosols are also needed to be removed by a filter before the gas enters the membrane unit [27]. There is a need to develop a system that separates CO<sub>2</sub>/CH<sub>4</sub>, H<sub>2</sub>S/CH<sub>4</sub>, and other traces impurities simultaneously from raw biogas using different membranes. Also, membrane separation process for biogas system should be further explored to facilitate easy operation with low energy demands using a multistage membrane that is more economical for biogas upgrading. The multistage membrane process has lower investment and operating costs with high CH<sub>4</sub> purity in comparison to the single stage process [97]. Chen et al. [39] identified that CH<sub>4</sub> recovery could be improved from 80 to 99.5% using multistage membrane process. Still, in the area of membranes for biogas separation, intensive research has been required to synthesise new membrane materials for efficient biogas upgrading.

#### 2.4. Cryogenic separation (CS)

Cryogenic separation of biogas is based on the principle that various gases like CO<sub>2</sub> and H<sub>2</sub>S liquefy under different pressure and temperature conditions. It operates under a very low temperature (–170 °C) and high pressure (80 bar) condition. The boiling point of CH<sub>4</sub> at 1 atm is –161.5 °C that is much lower than the boiling point of CO<sub>2</sub> which is –78.2 °C and thus allowing the separation of CO<sub>2</sub> from CH<sub>4</sub> by liquefying it [1]. These operating conditions are maintained using a series of compressors and heat exchangers (Fig. 8) [98]. The main drawback of this process is the use of different process equipment, mainly turbines,

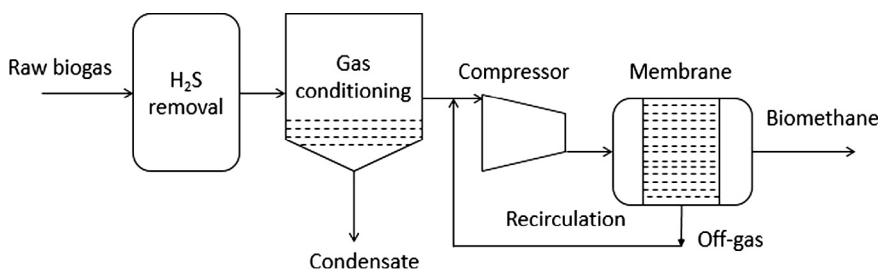


Fig. 7. Process flow diagram of membrane separation.

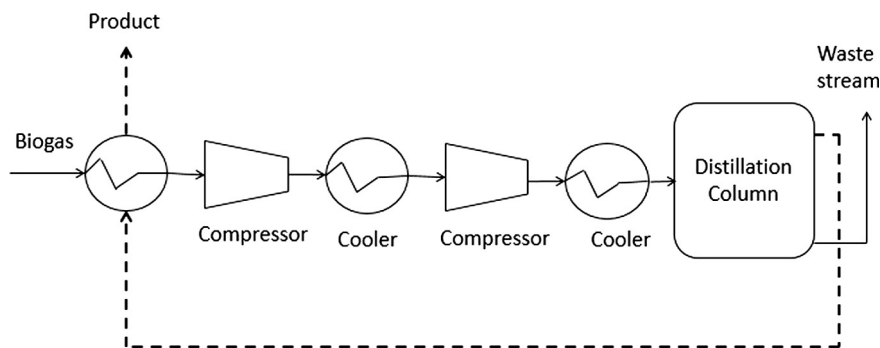


Fig. 8. Process flow diagram of cryogenic separation.

heat exchangers, distillation columns and compressors which raise its capital and operational costs [46] with high energy requirements. Generally, four steps involve in a typical cryogenic system to upgrade biogas to natural gas quality. Firstly, the moisture, H<sub>2</sub>S, dust particles, halogens, siloxanes, and other unwanted components are removed from the raw biogas. In the second step, the biogas is compressed to 1000 kPa and subsequently cooled to -25 °C. In the third step, the biogas is further cooled until -55 °C and liquefied CO<sub>2</sub> is removed from the gas mixture. Finally, the remaining gas stream is further cooled until -85 °C where CO<sub>2</sub> reaches a solid form and removed. The purified gas is depressurised and can be used in various applications. This separation is regarded as new technology, which is still under development [27,35] but some commercial plants are already in operation [35]. Cryogenic separation can be useful if the goal is to produce liquefied biomethane (LBM) and liquid natural gas (LNG) [48]. It is advised to pre-separate H<sub>2</sub>O and H<sub>2</sub>S to avoid plugging of equipment due to the freezing of existed water in the raw biogas [23]. This phenomenon significantly reduces the operating pressure of the system [99].

2.5. Separation of other trace components from biogas

The major components of biogas are CH<sub>4</sub> and CO<sub>2</sub>, but it also contains H<sub>2</sub>S and other sulphur compounds, halogenated compounds, siloxanes, water, ammonia, and other volatile organic compounds (VOCs) in trace amounts [2]. The effects of various impurities in biogas are summarised in Table 3. The moisture in biogas is not acceptable for all applications and needs to be removed before utilisation because it seriously damages the equipment by corrosion, thus the commercial benefit of using biogas reduces. Drying biogas is conducted through condensation and adsorption by silica gel, activated carbon, and aluminium oxide (Table 4). Preremoval of H<sub>2</sub>S is very crucial in all upgrading technologies because H<sub>2</sub>S can cause corrosion in pipelines and

Table 3  
Biogas impurities and their effects [2,35].

Impurities	Impact
H <sub>2</sub> O	Corrosion in compressors, gas storage tanks and engines due to reaction with H <sub>2</sub> S, NH <sub>3</sub> , and CO <sub>2</sub> to form acids Accumulation of water in pipes Condensation or freezing due to high pressure
H <sub>2</sub> S	SO <sub>2</sub> and SO <sub>3</sub> are formed due to combustion, which are more toxic than H <sub>2</sub> S and cause corrosion with water
CO <sub>2</sub>	Reduction in calorific value
NH <sub>3</sub>	Corrosive when dissolves in water leads to an increase in antiknock properties of engines; causes formation of NO <sub>x</sub>
O <sub>2</sub> /air	Explosive mixtures due to high concentrations of O <sub>2</sub> in biogas
Cl <sup>-1</sup>	Corrosion in combustion engines
F <sup>-1</sup>	Corrosion in combustion engines
Dust	Clogging due to deposition in compressors and gas storage tanks
Hydrocarbons	Corrosion in engines due to combustion
Siloxanes	Formation of SiO <sub>2</sub> and microcrystalline quartz due to combustion; deposition at spark plugs, valves, and cylinder heads abrading the surface

Table 4  
H<sub>2</sub>S and H<sub>2</sub>O pretreatment and removal methods [35].

H <sub>2</sub> S pretreatment methods	H <sub>2</sub> O removal methods
Air/O <sub>2</sub> dosing to biogas reactor	Condensation
Iron sponge	Demister
Iron oxide	Cyclone
Iron chloride dosing to digester slurry	Moisture trap
Air stripping and recovery	Adsorption dryer
Biological removal on a filter bed	Silica
Membranes	Aluminium
Adsorption on activated carbon	Physical absorption with glycol
Physical and chemical absorption	Absorption with hygroscopic salts
Zinc oxide sorbents	

equipment. Different pretreatment processes are used to remove H<sub>2</sub>S as shown in Table 4. The mentioned H<sub>2</sub>S removal processes are generally effective where the sulphur concentration in biogas is high [105,106]. Siloxanes are the part of various industrial products such as shampoos, paper coating, textile, and detergents [107]. Siloxanes are not decomposed in the activated sludge process and needs to be removed. The most frequently used method for removing siloxanes is adsorption on activated carbon [108–110]. Silica gel is also reported to be effectively used for the removal of organic silicon compounds by adsorption [28,108]. Other possible adsorbents are molecular sieves and polymer pellets. An absorption process using non-volatile organic solvents has also been reported [106]. With water scrubbing, water soluble halogenated compounds, sulphur compounds, and ammonia can also be removed [2]. Membrane separation process is also reported in the literature which based on the principle of selective siloxane permeation by solution and diffusion through dense polymeric membrane material [108]. However, due to high investment cost and moderate operating cost, this technology was no further pursued [76]. The possible reason is that a large number of impurities in biogas has not been tested yet in membrane separation process, therefore these contaminants can have adverse effects on the separation performance and membrane stability [85]. Generally, a precleaning of biogas is required prior to the introduction of the biogas to the membrane process to protect the membrane against harmful pollutants and subsequently prolong the service life time of membrane separation system.

2.6. Life cycle assessment of biogas upgrading technologies

Life cycle assessment (LCA) is an operational tool commonly used to evaluate the energy and environmental performance of a product or system throughout its whole life cycle [111]. LCA takes into account all environmental effects of a product, including exploration of the resources, transport, manufacturing, emissions, and disposal [37,112]. The emission from the entire biogas production to energy conversion measures the environmental impacts for any upgrading technology. Furthermore, LCA concepts and techniques provide the base for the evaluation of GHG emissions [113]. These evaluations mostly focused



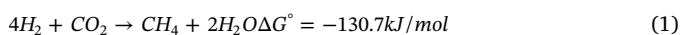
**Table 5**  
Key parameters for LCA analysis [34,37,111,115]

Technology	PSA	HPWS	OPS	AS	MS	CS
Electric energy (MJ/m <sup>3</sup> ), (kWh/Nm <sup>3</sup> )	0.72 0.24	0.97 0.20	– –	– –	1.80 0.19	– –
MJ/ton of CO <sub>2</sub> removed	915	770	1069	433	1264	1275
Methane loss (%)	4	5.13	4	0.1	6	0.65
Upgrading yield (%)	65	68	–	–	65	–
Methane purity (%)	97.5	98	97	99	91	98

on CH<sub>4</sub> production and loss, the energy efficiency of various biogas upgrading technologies, biogas utilisation, and by-products usage. The key parameters of LCA are investigated for various biogas upgrading technologies which are summarised in Table 5. LCA also suggests that all the wastes from various industries should be minimised by converting them into alternative renewable energy sources to provide a clean environment and low GHG emissions [114]. For the GHG aspect, the biogas production and biogas upgrading step have the highest impact due to methane loss and high energy consumption. A large number of technologies for biogas upgrading have been developed in order to improve the overall efficiency to reduce the investment, operation and maintenance costs [111,115]. Despite the increasing interest in these technologies, there is a substantial lack of knowledge on the environmental impacts connected to these upgrading technologies.

## 2.7. Innovative biological biogas upgrading

Biogas upgrading is vital for clean energy production. Although various prevalent and commercial technologies are available for biogas upgrading but significant challenges in terms of energy consumption and operating costs still exist. In this context, an innovative alternative solution for biogas upgrading is the biological method via hydrogenotrophic methanogenesis which opens new avenues for a renewable energy source [116]. In this process, CO<sub>2</sub> and H<sub>2</sub> are biologically converted to CH<sub>4</sub> by the action of autochthonous hydrogenotrophic methanogens without any additional energy input according to Eq. (1).



The concentration of CH<sub>4</sub> in the product gas is > 98%, allowing its exploitation as a substitute for natural gas [117,118]. The environmentally friendly production of H<sub>2</sub> relies on water electrolysis utilising the excess energy from the wind or solar power stations [119,120]. Indeed, it has been reported that in EU countries, more than 26% of the electricity from wind is a temporary surplus which is used for electrolysis of water [116]. The biological biogas upgrading process can be classified into three categories, which are (1) in situ biogas upgrade technology, in which H<sub>2</sub> is transported inside the liquid phase of a biogas reactor and then reacted with the CO<sub>2</sub> contained in the reactor resulting in CH<sub>4</sub> production [121], (2) ex situ biogas upgrade technology, in which CO<sub>2</sub> comes from external sources (e.g., CO<sub>2</sub> off-gas, industrial waste gas, or syngas) and H<sub>2</sub> are introduced into the liquid phase of a reactor containing hydrogenotrophic cultures, resulting in CH<sub>4</sub> production [117], and (3) hybrid biogas upgrade technology, in which in situ and ex situ biogas upgrading are executed together for optimising the process [116].

### 2.7.1. Advantages and challenges of the biological biogas upgrading process

Biological biogas upgrading process has several advantages: (1) It requires lower technical requirements compared to available technologies resulting in reduced operational and investment cost and energy [122]. (2) It gives a high CH<sub>4</sub> final volume [123]. (3) It does not involve CH<sub>4</sub> emissions to the atmosphere, resulting in enhanced life-cycle environmental benefits [124]. (4) It converts surplus electricity to CH<sub>4</sub> which is easily transported and distributed across long distances for various uses such as heating, CHP generation, and vehicular fuel [123].

**Table 6**  
Technical availability and maintenance costs of biogas upgrading technologies [127].

Technology	Technical availability per year (%)	Maintenance cost (€/year)	Cost (€/m <sup>3</sup> )
PSA	94	56,000	0.26
HPWS	96	15,000	0.15
OPS	96	39,000	–
CSP	91	59,000	–
MS	98	25,000	0.22
CS	–	–	0.40

However, practical challenges related to high pH and low gas-liquid mass transfer rate of H<sub>2</sub> bounds its availability for methanogens [122–125]. The use of the hybrid technology addresses the problem of pH enhancement during the in situ technology, while a considerably smaller separate reactor is required for the ex situ technology [116]. Also, gas recirculation flow rate and reactor dimension are the main factors for an effective biogas upgrading process [118]. This technology could only be the best alternative of traditional biogas upgrading process when electricity is surplus and H<sub>2</sub> is cheaper. Therefore, these limitations have to be addressed to introduce its commercial application.

## 2.8. Economic assessment of biogas upgrading technologies

The technical availability, operating and maintenance costs associated with various upgrading technologies for 1000 m<sup>3</sup>/h of raw biogas are summarised in Table 6. The availability of membrane separation, water scrubbing, and organic physical scrubbing is highest due to their low maintenance costs. Maintenance cost is one of the key factors for the selection of any upgrading technology as it increases the operational cost of the process. It is worthwhile that the operating and maintenance costs for different upgrading technologies are different and can increase the capital cost [38,121,122].

The amount of energy required to upgrade raw biogas is another important factor when selecting a technology. Table 7 summarises the energetic performance of various upgrading technologies and concludes that water scrubbing and organic physical scrubbing are cheaper and consistent with a range of 0.20–0.43 kWh/Nm<sup>3</sup> and 0.4–0.5 kWh/Nm<sup>3</sup>, respectively. PSA is less consistent with a range of 0.24–0.60 kWh/Nm<sup>3</sup>. The possible reason is that different adsorbents need numerous regeneration energies that could make the process expensive and variable. Similarly, amine scrubbing shows a wide variation in the energy requirement due to high thermal energy to regenerate the amine absorbent. Different types of membrane materials and operating pressures results in a high variation in energy needed to upgrade biogas.

The capital and operating costs of biogas upgrading technologies largely depend on the selected process, quality of raw biogas, desired product quality, and more importantly, the capacity of the plant [16]. The specific investment cost of various upgrading technologies significantly depends on the plant capacities; the smaller the capacity

**Table 7**  
Energy requirements of various biogas upgrading technologies.

Technology	Energy requirement (kWh/m <sup>3</sup> of upgraded biogas)					
	Collet et al. [128]	Patterson et al. [127]	Götz et al. [129]	Ncibi et al. [130]	Olsson et al. [131]	Meier et al. [126]
PSA	0.5–0.6	0.24	0.335	0.285	–	–
HPWS	0.3	0.2	0.43	0.391	–	–
OPS	0.4	–	0.49	0.511	–	–
CSP	0.15	0.12	0.646	0.126	–	–
MS	–	0.19	0.769	–	0.27	0.378
CS	–	–	–	–	0.42	–

**Table 8**  
Capital investment cost of different upgrading technologies.

Technology	Cost
PSA	0.40 €/Nm <sup>3</sup> of biogas [46] Capital costs for 250, 500, 600, and 1000 m <sup>3</sup> /h are 5.5, 3.2, 2.4, and 2.2 kUSD/(m <sup>3</sup> /h), respectively [16,39,128,129]
HPWS	0.13 €/Nm <sup>3</sup> of biogas [46] Capital costs for 250, 500, 660, and 1000 m <sup>3</sup> /h are 1.22, 2.7, 2.78, and 2 kUSD/(m <sup>3</sup> /h), respectively [16,39,128,129]
OPS	Capital costs for 250, 500, and 1000 m <sup>3</sup> /h are 4.8, 3.8, and 2.4 kUSD/(m <sup>3</sup> /h), respectively [16,39,128,129]
CSP	0.17 €/Nm <sup>3</sup> of biogas [46] Capital costs for 100, 250, 500, and 1000 m <sup>3</sup> /h are 10.5, 5.5, 3.6, and 2.6 kUSD/(m <sup>3</sup> /h), respectively [16,39,128,129].
MS	0.12 €/Nm <sup>3</sup> of biogas [46] Capital costs for 100, 600, and 700–1400 m <sup>3</sup> /h are 6.6, 2.5, and 2.2 kUSD/(m <sup>3</sup> /h), respectively [16,39,128,129]

(0–100 Nm<sup>3</sup>/h), the higher the specific investment cost and vice versa [30]. For a plant with low capacity, more or less the same number of equipment, sensors, control valves, and pipes are required as for a plant with much larger capacity. The dimensions of the pipes and valves will be small, but the investment cost will be high. The capital investment costs of various upgrading technologies are quite similar and decreasing only with plant capacity as presented in Table 8.

2.9. Technology development and commercialisation

The biogas upgrading technologies which dominate today are membrane separation, pressure swing adsorption, water scrubbing, and amine scrubbing [30,132,133]. A few years ago, the membrane separation was considered as an expensive and unestablished technology but now it is rapidly gaining interest and market share due to the availability of highly selective and low cost polymeric materials [27]. There are some major challenges for membrane technology such as process stability for a long time and CH<sub>4</sub> concentration and its degradation by raw biogas components such as NH<sub>3</sub> and H<sub>2</sub>S, thus requiring a precleaning step to protect it which increases upgrading cost. Multistage membrane process can remove acid gases and water vapours from raw biogas and produces CH<sub>4</sub> with high concentration (> 98%) with low operational cost [30]. Moreover, only membrane units are available at low throughput capacities for small scale biogas upgrading. The increasing trend of membrane based biogas upgrading plants can

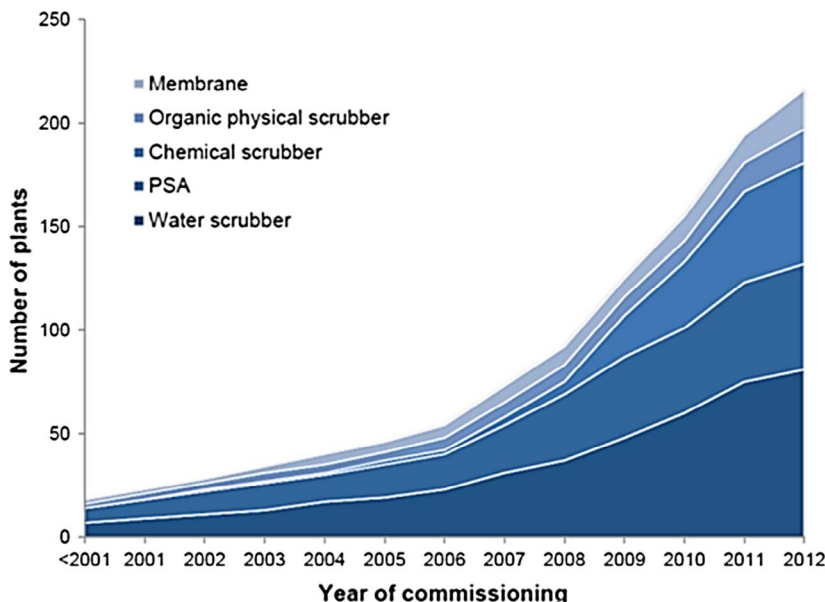


Fig. 9. The progress of biogas upgrading plants with increasing years [38].

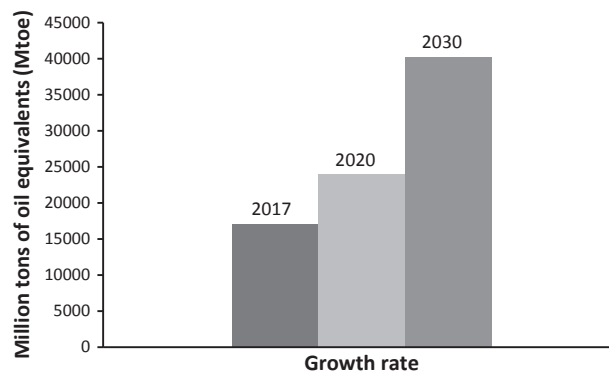


Fig. 10. Growth of biogas production in European countries for 2017, 2020, and 2030 [38].

also be seen in Fig. 9. The production of biogas in Europe has increased rapidly over the last decade. It is predicted that biogas production will be increased from the current level of 16.9 Mtoe to 40.2 Mtoe in 2030 (Fig. 10). The market of biogas has grown from \$124 billion in 2010 to \$217 billion in 2016 [133]. Thus, the global biogas plant market is expected to reach \$8.98 billion in 2017, contributed mainly by the European countries. Among them, Germany has the higher biogas production; UK, Sweden, Austria, Switzerland, and Italy are the other major biogas markets in the continent (Table 9).

The recent market for matured biogas upgrading technologies highly depends on its optimisation design with low operational cost [10]. Although water scrubbing, amine scrubbing, and PSA have developed technologies since many years, a lot of research has been conducted to improve their efficiency. For instance, water scrubber process has been optimised to reduce the fresh water demand [132]. PSA has been developed to reduce the number of expensive pressure valves and also the use of the high-performance low-cost absorbent materials, and amine process has been developed with cost-effective regeneration process for amine solution [133]. Cryogenic upgrading technology is the best for liquefied biogas (LBG) production but it is still not available for full-scale commercial operation. The use of several devices and equipment in cryogenic separation makes it an expensive option for the upgrading of biogas. Therefore, it makes a small market share in biogas upgrading sector [134]. This technology may have a breakthrough within a short period if the operational and economic problems are properly fixed.

**Table 9**  
Biogas plants, biogas upgrading plants, and their upgrading capacities in selected European countries [135,136].

Country	Biogas plants	Biogas upgrading plants	Upgrading capacity (Nm <sup>3</sup> /h)
Germany	94	120	204,082
Austria	9066	10	5160
Italy	1264	1	540
Sweden	187	53	38,858
Netherlands	211	16	16,720
UK	–	–	18,957
Switzerland	–	–	6310

**Table 10**  
Guidelines for impurities removal for specific biogas applications [23,39].

Applications	H <sub>2</sub> S	CO <sub>2</sub> (% vol)	H <sub>2</sub> O (% vol)
Gas Heating (Boiler)	< 250 ppm	No removal required (25–30)	No removal required (6)
Kitchen Stove	< 10 ppm	No removal required (25–30)	No removal required (6)
Stationary Engine (CHP)	< 1000 ppm	No removal required (25–30)	Avoid condensation (< 3)
Vehicle Fuel	Removal required (5 mg/m <sup>3</sup> )	Recommended (< 4)	Removal required (< 3)
Natural Gas Grid	Removal required (2–15 mg/m <sup>3</sup> )	Removal required (≤ 3)	Removal required (1 – 8)

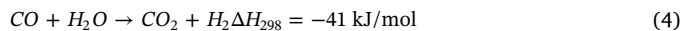
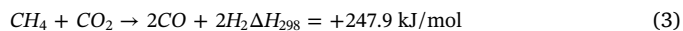
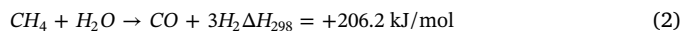
**3. Biogas utilisation and conversion to bio-compressed natural gas (Bio-CNG)**

As mentioned previously, biogas has other impurities except for CO<sub>2</sub> that should be removed. The concentrations of those impurities depend on the biogas source [7]. Hence, the biogas is preferred to be upgraded before any utilisation. In fact, the levels of biogas impurities should be controlled based on the requirements of each application as shown in Table 10. Biogas can be used for different applications either as raw or upgraded. The selection of upgrading technologies relies on various

factors, such as availability of resources, utilisation of biogas, and investment cost. The cheapest technology does not always mean the best solution if it is not able to meet the requirements for utilisation [137]. Production and utilisation of biogas has numerous environmental advantages: (1) It is used as a renewable energy source. (2) It reduces the CH<sub>4</sub> emission to the environment. (3) It can be used as a substitute for fossil fuels. (4) It reduces the emission of CO<sub>2</sub> from combustion. (5) It can be used in all natural gas appliances after upgrading. Biogas is mainly utilised in five different applications: (1) H<sub>2</sub> production from biogas. (2) Electricity and power generation with combined heat and power production (CHP). (3) Injection into the natural gas grids after upgrading. (4) Production of heat and steam. (5) As a vehicular fuel in upgraded and compressed form. Fig. 11 shows the different potential applications of biogas after upgrading to the required level.

**3.1. H<sub>2</sub> production from biogas**

Reforming of biogas on supported catalysts is an interesting method to produce hydrogen. The use of biogas instead of CH<sub>4</sub> for the production of H<sub>2</sub> is also a promising way to reduce CO<sub>2</sub> emission [138,139]. The presence of H<sub>2</sub>S is poisonous for nickel (Ni) catalyst so it has to be removed prior to the reforming process. Currently, the biogas derived H<sub>2</sub> is used in the field of the hydrogen-base fuel cell which efficiently converts the H<sub>2</sub> to electricity at a commercial level. The main drawback in the reforming process is the formation of carbon monoxide (CO) as a gaseous by-product [140]. It acts as a poison to the fuel cell in the 50 ppm range [141]. Therefore, the controlled steam reforming (Eqs. (2) and (3)) followed by CO shift reactions (high-temperature, HT, and low-temperature, LT) (Eq. (4)) are proved to be the feasible and economical solution for CO reduction.



Also, there are various advantages of using biogas for the production of H<sub>2</sub>: (1) It is a domestic and local energy resource. (2) It is a cheap feed

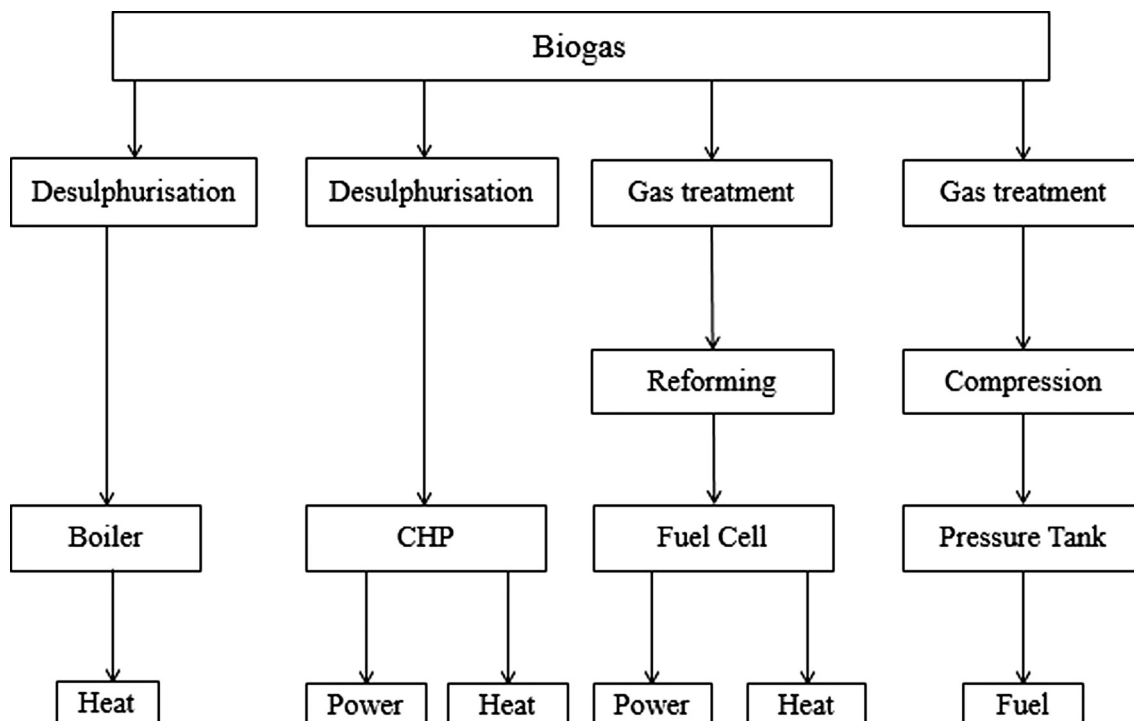


Fig. 11. Potential applications of biogas.

**Table 11**  
Biogas requirements for injection into natural gas grids [148,149].

Component	Sweden	France	Switzerland	Germany	Netherlands	Austria
CH <sub>4</sub> (% vol)	≥ 97	≥ 86	≥ 96	≥ 96	≥ 85	≥ 96
CO <sub>2</sub> (% vol)	≤ 3	≤ 2.5	≤ 6	≤ 6	≤ 6	≤ 3
O <sub>2</sub> (% vol)	≤ 1	≤ 0.01	≤ 0.5	≤ 0.5	≤ 0.5	≤ 0.5
H <sub>2</sub> (% vol)	≤ 0.5	≤ 6	≤ 4	≤ 5	≤ 0.5	≤ 4
CO (% vol)	–	≤ 2	–	–	≤ 1	–
H <sub>2</sub> S (mg/Nm <sup>3</sup> )	≤ 10	≤ 5	≤ 5	≤ 5	≤ 5	≤ 5
Total sulphur (mg/Nm <sup>3</sup> )	≤ 23	≤ 30	≤ 30	≤ 30	≤ 16.5	≤ 10
NH <sub>3</sub> (mg/Nm <sup>3</sup> )	≤ 20	≤ 3	≤ 20	–	≤ 3	0
H <sub>2</sub> O (mg/Nm <sup>3</sup> )	≤ 3	–	–	–	–	–
Water dew point (°C)	≤ –5	≤ –5, P <sub>max</sub>	–	Soil temp	≤ –8, 70 bar	≤ –8, 40 bar
Heavy metals (mg/Nm <sup>3</sup> )	–	≤ 1	≤ 5	≤ 5	–	–
Siloxanes (mg/Nm <sup>3</sup> )	–	–	–	–	≤ 5	≤ 10
Halogens (mg/Nm <sup>3</sup> )	–	≤ 1 (Cl) ≤ 10 (F)	≤ 1	0	≤ 50/25 (Cl/F)	0
Mercaptans (mg/Nm <sup>3</sup> )	–	≤ 6	≤ 5	≤ 15	≤ 6	≤ 6

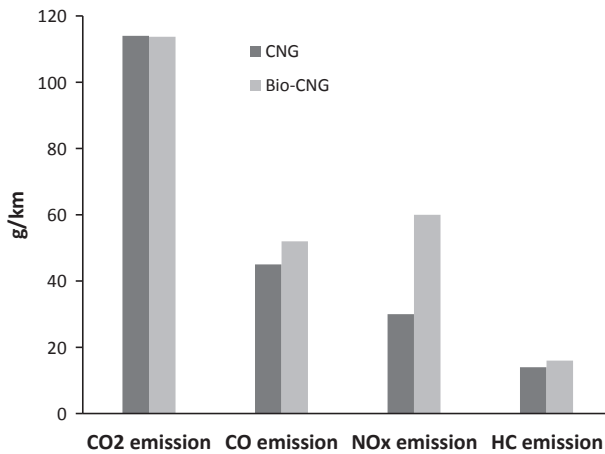


Fig. 12. Comparison of emissions for CNG and bio-CNG [154].

**Table 12**  
Compositions of raw biogas, CNG, and bio-CNG [157].

Component	CNG (volume%)	Bio-CNG (volume%)	Raw biogas (volume%)
CH <sub>4</sub>	89.14	93	65
CO <sub>2</sub>	4.38	4	33
H <sub>2</sub>	0.01	0.06	0.02
N <sub>2</sub>	0.11	2.94	1.98
C <sub>2</sub> H <sub>6</sub>	4.05	–	–
C <sub>3</sub> H <sub>8</sub>	0.83	–	–
Iso-C <sub>4</sub> H <sub>10</sub>	0.28	–	–
Neo-C <sub>4</sub> H <sub>10</sub>	0.66	–	–
Iso-C <sub>5</sub> H <sub>12</sub>	0.09	–	–
Neo-C <sub>5</sub> H <sub>12</sub>	0.28	–	–
C <sub>6</sub> H <sub>14</sub>	0.17	–	–
H <sub>2</sub> S	–	20 ppm	500 ppm

**Table 13**  
The gaseous emissions from heavy vehicles using different types of fuel [40]

Fuel	CO (g/km)	HC (g/km)	NO <sub>x</sub> (g/km)	CO <sub>2</sub> (g/km)	Particulates (g/km)	Calorific value (kJ/kg)
Diesel	0.20	0.40	9.73	1053	0.100	44,800
Natural Gas	0.40	0.60	1.10	524	0.22	50,000
Biogas	0.08	0.35	5.44	223	0.5	35,000
Bio-CNG	0.02	0.12	0.48	100	0.1	52,000

**Table 14**  
Biomethane production plants, gas, and biomethane filling stations in selected European countries [135,136].

Country	Biomethane plants	Gas filling stations	Biomethane filling stations
Germany	178	203	308
Austria	14	904	3
Italy	5	903	–
Sweden	59	190	218
Netherlands	21	150	60
UK	37	80	–
Switzerland	24	137	130

which helps to decrease the end-use cost. (3) It is considered as an environmentally friendly process.

3.2. Electricity production with combined heat and power production (CHP)

A significant percentage of the global electricity demand is expected to be generated using biogas in the near future [142]. Biogas is an economical fuel for the generation of electricity and heat [143]. Internal combustion engines, gas turbines, micro-turbines, Stirling engines, and fuel cells have been successfully generating electricity using biogas [144,145]. Gas engines do not require high quality gas but it is preferred to condense the water vapours in raw gas to avoid condensation in gas lines. The required level of H<sub>2</sub>S is below 250 ppm to avoid corrosion of boiler and gas engine in combined heat and power plants (CHPs) [17]. A low calorific value of biogas is one of the main barriers for its utilisation in combined heat and power generation [23].

3.3. Biogas injection into natural gas grids

Biogas utilisation as a substitute of natural gas has gained significant importance in recent years due to depletion and low quality of natural gas resources. Purified biogas can be injected into the natural gas grids [146]. Upgrading biogas as equal quality as natural gas by an efficient appropriate method is very important for its injection into the existing natural gas grids. Sweden, Germany, Switzerland, and France have set their own standards for biogas injection into the natural gas grids to avoid corrosion of equipment (Table 11). These standards could be maintained using the existing biogas upgrading processes [27,147].

3.4. Biogas for the production of heat and steam

In most developing countries, biogas is used for cooking and lighting [150]. Biogas is also used as fuel in the boiler for steam production in different industrial applications. Burning biogas in a boiler is one of the reliable technologies because low quality biogas can also be

**Table 15**  
Bio-CNG fuel standards in various countries [38,154].

Specification	France	Sweden	Netherlands	Germany	Austria	Switzerland
CH <sub>4</sub> (% vol)	96	> 97	–	–	96	> 96
CO <sub>2</sub> (% vol)	< 2.5	< 4	< 6	< 6	< 3	< 6
H <sub>2</sub> S (mgS/Nm <sup>3</sup> )	< 5	< 15	< 5	< 5	< 5	< 5
H <sub>2</sub> (%vol)	< 6	–	< 12	< 5	< 4	< 4
Mercaptans (mgS/Nm <sup>3</sup> )	< 6	–	< 10	< 16	< 6	< 5
Total Sulphur (mgS/Nm <sup>3</sup> )	< 30	< 23	< 45	< 30	< 10	< 30
O <sub>2</sub> (%vol)	< 1	< 1	< 0.5	< 0.5	< 0.5	< 0.5
Water (H <sub>2</sub> O) dew point	< – 5 °C	< – 9 °C, 200 bar	< – 10 °C, 8 bar	At ground temperature	< – 8 °C, 40 bar	< – 8 °C
Wobbe index (MJ/Nm <sup>3</sup> )	48.24–56.52	44.7–47.3	43.46–44.41	46.1–56.5	47.7–56.5	47.9–56.5
Calorific value (MJ/Nm <sup>3</sup> )	38.52–46.08	–	31.6–38.7	30.2–47.2	38.5–46	38.5–47.2

**Table 16**  
Commonly used storage devices at various pressures [153].

Pressure	Storage device	Material
Low (0.138–0.414 bar)	Water sealed gas holder	Steel
Low	Gas bag	Rubber, plastic, vinyl
Medium (1.05–1.97 bar)	Propane or butane tanks	Steel
High (200 bar)	Commercial gas cylinders	Alloy

used with pressure around 8–25 mbar [16]. Normally, biogas can be used in a boiler without upgrading, however, H<sub>2</sub>S level below 1000 ppm should be considered to avoid corrosion of boiler [23,151].

### 3.5. Biogas conversion to bio-CNG

After the removal of unwanted components from biogas to ≥ 97% CH<sub>4</sub>, it can be converted into bio-compressed natural gas (bio-CNG) at 20–25 MPa (2900–3600 psi) [102,152]. Compression also reduces its storage volume with high energy and pressure [153]. Although compression at high pressure is very expensive, high heating value and highly valuable product of bio-CNG justify this cost [48]. Compression is a physical approach for the conversion of biogas to bio-CNG. Also, the properties of bio-CNG are the same as that of CNG. However, the logistical investigation is also important with biogas upgrading and compression techniques.

### 3.6. Bio-CNG as vehicular fuel

Global warming, energy crisis, and the increasing cost of fossil fuels are the driving forces for the development of alternative vehicular fuels. The compressed biomethane, also called as bio-CNG, has the same properties as CNG in terms of engine performance, gas consumption, and efficiency, when used as vehicular fuel [18,146]. The CO<sub>2</sub> is one of the GHGs emission and important parameters for any fuel in view of global warming and climate changes. There is no significant difference

in CO<sub>2</sub> emission level for both fuels, however, the emission levels of NO<sub>x</sub>, HC, and CO for bio-CNG are slightly higher than CNG when used as a vehicular fuel (Fig. 12) [127,154]. The temperature is low during the warm up period of the vehicle engine. The decrease in temperature reduces the oxidation rate of CO into CO<sub>2</sub> resulting in high CO emission [155]. Ethane and propane in CNG (Table 12) have lesser activation energies than methane resulting in better combustion compared to bio-CNG. Subsequently, bio-CNG gives slightly higher CO emission when used as a vehicular fuel compared to CNG. The reason of high hydrocarbon carbon (HC) emission is also due to the incomplete combustion caused by poor oxidation at low temperature during the warm up period. Bio-CNG contains a higher content of N<sub>2</sub> compared to CNG which favours the NO<sub>x</sub> formation during combustion. Furthermore, there is no significant difference in fuel economy between biogas (24.11 km/kg) and CNG (24.38 km/kg) [154]. Bio-CNG has a great potential for becoming one of the most sustainable, economical, and environmentally friendly vehicular fuels in the near future [156]. The low density and high thermal efficiency of bio-CNG make it an economic vehicular fuel and [40,158]. For heavy vehicles, the use of bio-CNG is very advantageous because it has higher calorific value and more environmentally friendly compared to any other fuels (Table 13). There is a reduction of 63% in the emission of GHGs when bio-CNG is used instead of CNG for heavy vehicles [102,134,159].

Also, it produces a less amount of dangerous and hazardous chemicals such as CO<sub>2</sub>, sulphur, lead, and other heavy hydrocarbons into the atmosphere [160,161]. Bio-CNG is widely used in Sweden, Italy, Germany, Switzerland, France, Austria, Netherlands, and England as vehicular fuel (Table 14). Sweden was a pioneer by using 73% of biomethane in CNG vehicles in 2015 [162]. Sweden aims to achieve fossil fuel independent road transport system by 2030 and completely carbon neutral transport system in all modes of transportation by 2050 [136]. Germany also has the vision to use 100% bio-CNG as a vehicular fuel produced mainly using municipal waste by 2021 [154]. The fuel specifications of bio-CNG for various countries are summarised in Table 15. These countries have their own quality standards for biogas as

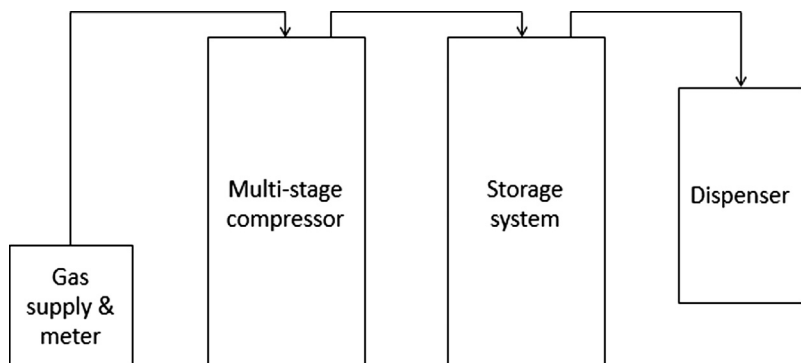


Fig. 13. Process flow diagram of a typical bio-CNG filling station.

Fig. 14. Bio-CNG production, storage, and distribution.

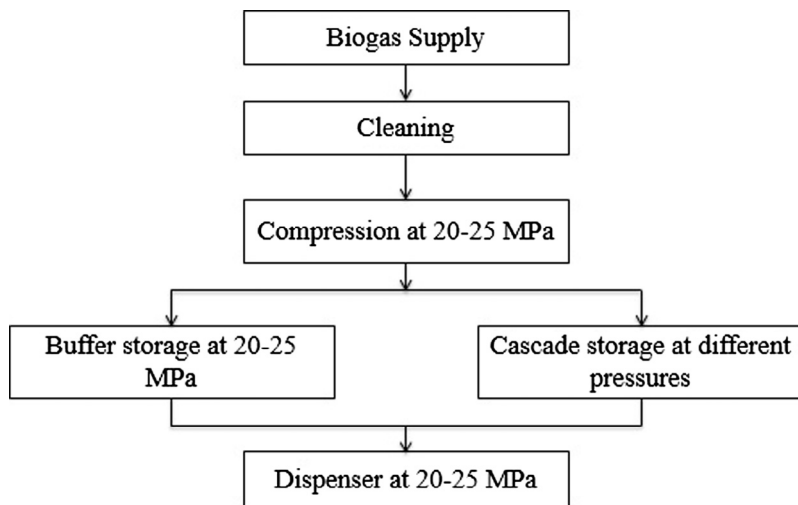


Fig. 15. Buffer storage system for bio-CNG [43].

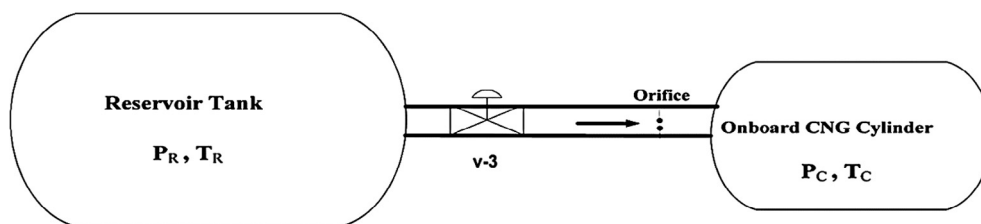
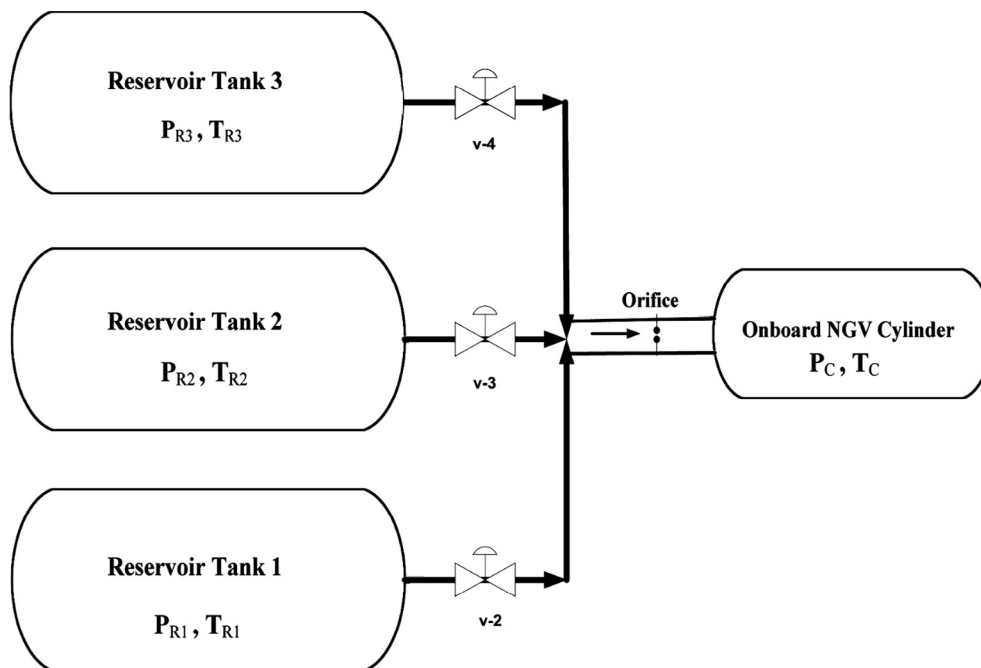


Fig. 16. Cascade storage system for bio-CNG [43].



a vehicular fuel. Although, the most important biogas quality indicators for vehicular fuel are the concentrations of CH<sub>4</sub> and CO<sub>2</sub>, which should be ≥ 96% and < 2.5%, respectively [18,147,156]. These requirements for bio-CNG could also be taken as a quality standard for other developing countries such as Pakistan, India Malaysia, and Indonesia.

#### 4. Storage of bio-CNG

Storage of bio-CNG or regular CNG is an important stage because it affects cylinder filling efficiency, filling time, charged mass, safety, and maintenance cost of the compressor [43]. Like CNG, bio-CNG can be

stored in steel airtight tanks for future usage [32,163]. Most commonly used storage devices are given in Table 16.

Fig. 13 shows a typical bio-CNG filling station. Bio-CNG from the distribution pipeline, usually with a low or medium pressures of less than 0.4 or 1.6 MPa, respectively, is compressed using large multistage compressors. Generally, a storage system consists of several high pressure large cylinders, so that the bio-CNG flows to the vehicle under differential pressures. Typically, the storage system operates at a pressure of 20.5–25 MPa, while the vehicle’s maximum on-board cylinder pressure is 20 MPa [16,164]. At the bio-CNG filling station, the gas is stored in the storage system for more efficient and economical

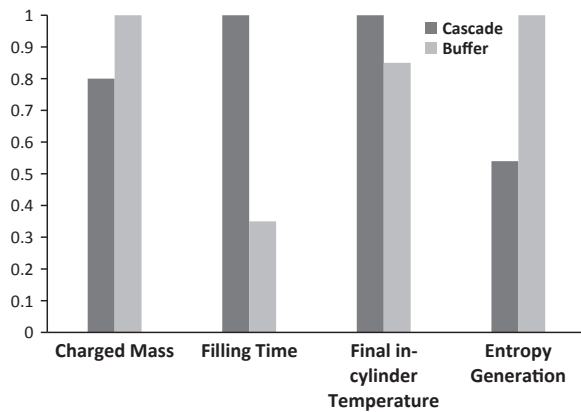


Fig. 17. Dimensionless comparisons between important parameters in buffer and cascade storage systems [43].

distribution. There are two common systems for storing bio-CNG at filling stations, namely buffer and cascade storage systems. Fig. 14 shows the approaches to produce bio-CNG, its storage, and distribution.

#### 4.1. Buffer storage system

In buffer storage system, the pressure of bio-CNG is maintained at 3000–3600 psi (20.7–24.8 MPa, 207–248 bar) and provides a maximum gas pressure of 2900 psi (20 MPa, 200 bar) to the vehicle's cylinders at 300 K. In this storage system, all filling station reservoir cylinders are connected and maintained at the same pressure during all time [152]. Fig. 15 shows typical buffer storage process of bio-CNG.

#### 4.2. Cascade storage system

In cascade storage, three reservoirs at different pressures (low, medium, and high) are used to fill the cylinders with bio-CNG. Each of these reservoirs contains several large cylinders which are used into an order of ascending pressure. Fig. 16 shows the typical cascade storage system using three reservoirs. During the gas filling, initially, the low pressure reservoir is connected to the cylinder. When the pre-set level is attained, it automatically switches to the reservoir of medium pressure and finally, high pressure reservoir is connected to ensure the maximum amount of gas.

In cascade storage system, bio-CNG filling from three reservoirs at different pressures takes more time compared to buffer storage system. Therefore, the cascade storage system is chosen for filling big vehicles that normally takes a longer filling time, while the buffer storage system is preferred for small family cars with fast cylinder filling [16,43,152,164].

Farzaneh-Gord et al. [43] studied the effect of storage type on the performance of gas filling stations and gave a theoretical analysis of various important parameters. Fig. 17 shows a dimensionless comparison between important parameters for these storage systems. The dimensionless parameters have been calculated by dividing the parameters to a higher value in each case. They identified that the filling time required to fill the vehicle cylinder to its final pressure in the buffer storage system was about 66% less than the cascade storage system. Also, the charged mass for buffer storage system was about 20% higher than the cascade storage system which gives an advantage to buffer system over cascade one. The biggest advantage of the cascade storage system over the buffer system is 50% less entropy generation for this configuration, leading to a lower compressor input work compared to buffer system. Temperature has a great effect on charged mass, where high temperature decreases the amount of charged mass in the cylinder. The cascade storage system has a higher final temperature that results in less charged mass compared to the buffer storage system.

## 5. Conclusions and future direction

Cost minimisation is not the only criterion for selecting biogas upgrading technology. It is also essential that the specific technology can satisfy the specific requirement. Therefore, it is attempted to provide critical considerations on available biogas upgrading technologies, various utilisation of biogas, conversion to bio-CNG, and its storage for further use. Although biogas production is a commercially developed technology, its worldwide utilisation is still limited due to stringent purification requirements before its usage. This review found that a lot of researches are still needed to reduce the CH<sub>4</sub> loss, environmental impacts, maintenance cost, and energy consumption. The developed biogas upgrading technologies are water scrubbing, pressure swing adsorption, and amine scrubbing, but membrane technology has altered the situation due to high market demand. Membrane technology can replace all traditional separation processes in the future due to its economic and environmental aspects. However, the future membrane material developments should focus on the enhanced compatibility with the high variety of biogas components rather than on the achievement of very high selectivity. Also, the use of membrane technology in other steps of biogas upgrading like desulphurisation, drying, or VOC removal is still rather rare. Some novel technologies such as cryogenic separation and ex situ biological upgrading methods represent the recent developments in biogas upgrading technologies. However, they are still under development and not available on a commercial scale. Therefore, more efforts are needed to channel the knowledge gap between such new methods and large scale operations. Biogas can be used as a substitute for natural gas in all applications but it is difficult to find the gas requirements for some specific applications.

Furthermore, biogas can be converted into bio-CNG as a vehicular fuel as an alternative to regular CNG. Bio-CNG would play a vital role to minimise environmental pollution contributed by other carbon-based fossil fuels. It is found that biogas production is increasing in Europe and around the globe, and subsequently increases its utilisation as a vehicular fuel in compressed form. Furthermore, CO<sub>2</sub> emission level for both fuels is almost the same. CO<sub>2</sub> emissions for the bio-CNG and regular CNG are 113.72 and 113.98 g/km, respectively. Safety precaution for bio-CNG storage is also an important aspect that requires attention. Generally, two common methods are used for storing bio-CNG, namely buffer and cascade storage systems. The performance of storage systems could be improved by balancing various parameters such as charged mass, filling time, and compressor input work. It is identified that the performance of bio-CNG filling station is improved by reducing the filling time and compressor input work and increasing the charge mass into the vehicle's cylinders. In addition, more researches and developments are required to construct such a storage system that combine the advantages of buffer and cascade storage systems. This provides an opportunity for future developments and improvements of the storage systems.

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