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A Review of Methods Used for Purification of Biogas

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Abstract

Biogas produced from the fermentation process of methane contains 59-71% methane with some other components such as water, small quantities of oxygen and nitrogen, hydrogen sulphide, carbon dioxide, and water vapors. Due to these ingredients, the heating value and efficiency of biogas reduces and can also cause corrosion of instruments if used as fuel. Also, gases like CO_2 and H_2S have a very pungent smell and are toxic in nature thus making the food toxic and low quality. These are the reasons why we need to purify biogas. Biogas purification involves removal of CO_2 and H_2S, the toxic gases. The methods we use for purification of biogas involve physical methods, chemical methods, and biological methods. Purified biogas i.e. biomethane is a promising renewable energy which will help in increasing food quality as well as reduce pollution. Purified biogas is a very cost-effective as well as a very good alternative to natural gas.

Keywords: biogas, biogas purification, chemical absorption, membrane technology, carbon capture.

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1. Introduction

Anaerobic digestion of organic matter including animal waste, food scraps, plant material, wastes from agricultural products, household wastes, industrial waste, and energy crops produces a renewable fuel which is called biogas [1]. At both domestic and commercial levels, biogas is used as a fuel for cooking food, producing power, and producing heat. Biogas contains 59-71% methane with some other components such as water, small quantities of oxygen and nitrogen, hydrogen sulphide, carbon dioxide, and water vapors. Due to these ingredients, the heating value and efficiency of biogas reduces and can also cause corrosion of instruments if used as fuel. Presence of these gases especially CO_2 and H_2S have a very pungent smell and are very toxic in nature thus making the food toxic and reducing food quality. Because of the presence of these component gases in the biogas, biogas need purification before we can use biogas as a source of energy. Treatment of biogas removes it toxicity, increases heating value of biogas, removes any pungent and unwanted smell, and reduces pollution caused by used of biogas. CO_2 can cause suffocation and death while H_2S when combined with water forms sulphuric acid which is a highly toxic and corrosive material. These are reasons why we need easy, safe & cost-effective methods for purification of biogas.

Biogas is quite different from natural gas and the difference between the two gases lies between its CO_2 content. The composition of biogas can also differ on the basis of their origin [2]. These component gases are the key to toxicity and pollution caused by biogas in comparison to natural gas. Both of these gases have different compositions of the component gases and the variation in the composition of biogas originates because the source of its production. This difference in the composition of both gases can be seen in the table 1. Biogas mainly consists of CH_4 and CO_2 [3]. Biogas is delivered to a wide number of markets and applications, including power, heat, and transportation fuels [4]. The most popular, simple, and easiest way of obtaining energy from biogas is combustion of biogas. Although biogas is a source of the renewable energy at domestic as well as industrial level, it is not easy and safe to use biogas as raw gas. Gases like CO_2 and H_2S present in biogas cause food toxicity, reducing food quality, and health issues at domestic level and corrosion of instruments, pollution, and H_2SO_4 production at industrial level. These are the reasons why biogas needs purification before using.

2. Methods of purification

The methods used for biogas purification involve physical, chemical, and biological methods (Figure 1).

2.1. Physical methods

Physical methods are very efficient in removing CO_2 and H_2S but in order to do so these require a large amount of water. Also, it requires high temperature, pressured water, water recirculation, and treatment of wastewater. It also requires electrical energy for water sprinkling and purification. Majority of the studies in the field of physical treatments focus on laboratory-scale water purification systems. In a sophisticated industrial process, a specialized apparatus is required due to the large quantity of water being used and a treatment plant for the purification of wastewater producing in this process.

2.1.1. Removal of CO_2

Water is a universal solvent and it has the ability to dissolve CO_2 [5]. CO_2 dissolves in water and forms carbonic acid [6].

$$CO_2 + H_2O \leftrightarrow H_2CO_3 \leftrightarrow H_2CO_3^- + H^+$$

Methane and water generate carbon monoxide which can cause reduction in heating value and efficiency and can increase toxicity.

$$CH_4 + H_2O \rightarrow CO + 3H_2$$

In the pressurized water purification method, biogas is fed from the bottom in to a packed bed column, and pressurized water is sprayed from the top of the column, the biogas purification is a counter-current process. In this way, the CO_2 is dissolved in the water and exits through the bottom of the tower, allowing an efficiency of process to increase with 95% CO_2 removed (Figure 2) [7]. According to the studies, purification of biogas depends on the water flow rate, pressure in the scrubbing column, and the biogas flow rate in the tower. The table 2 shows a comparison between different methods of physical absorption of CO_2.

2.1.2. Removal of H_2S

At 20^0C, H_2S dissolves in water at a rate of 0.41g per 100ml and yields sulphur and hydronium ions [6]. The solubility of H_2S is influenced by temperature in a direct relation.

$$H_2S + H_2O \rightleftarrows H_2S_{(aq)}$$
$$H_2S_{(aq)} \rightleftarrows H^+ + HS^-$$
$$HS^- \rightleftarrows H^+ + S^{2-}$$

Removal of H_2S was measured in a series of reaction with different levels of water and result of the experiments is given in the table 3.

2.2. Chemical methods

In order to maximize its utilization and increase process efficiency, it is crucial to research different conditions. The use of organic materials in form of activated carbon has been a novel material for purification of biogas.

2.2.1. Removal of CO_2

In chemical methods, strong bases including calcium hydroxide ($Ca(OH)_2$), sodium hydroxide (NaOH), and potassium hydroxide (KOH) have been used to remove CO_2 and H_2S from biogas [8].

$$CO_2 + H_2O \rightarrow CO_{2(aq)}$$
$$NaOH + CO_2 \rightarrow NaHCO_3$$
$$Ca(OH)_2 + NaHCO_3 \rightarrow CaCO_3 + H_2O + NaOH$$

The removal of CO_2 in different reagents was checked and the results are discussed in the table 4. CO_2 can be removed chemically using a variety of chemical reagents but the drawback to this method is that these chemicals can be expensive, hard to find, and not easy to implement on the large scale industries [6]. It is important to know that the bases used in this process must be strong diluted bases.

2.2.2. Removal of H_2S

For the removal of H_2S from biogas, iron oxide (Fe_2O_3) is used. Iron oxide is easy to make and can efficiently separate H_2S. The preparation of Fe_2O_3 is given as below:

$$4Fe + 3O_2 \rightarrow 2Fe_2O_3$$

The process of removal of H2S from biogas by using Fe_2O_3 takes place by the following process:

$$Fe_2O_3 + H_2S \rightarrow Fe_2S_3 + H_2O$$

Fe_2O_3 can be easily regenerated from Fe_2S_3 by reacting it with oxygen.

$$2Fe_2S_3 + 3O_2 \rightarrow Fe_2O_3 + 6S$$

The removal of H_2S in different reagents was checked and the results are discussed in the table 5 [6].

2.3. Biological methods

For the purification of biogas, biological techniques are both cost-effective and fascinating. Chemotrophic or photosynthetic bacteria are the primary focuses of research in this area. Biological methods of biogas purification are ecologically friendly, cost-effective, require no chemical products, use little energy, have high rates of degradation, and are supported by biofilms. However, biological processes are inefficient and easily contaminated by isolated microbes [9].

2.3.1. Removal of CO_2

The experimental process for the removal of CO_2 biologically was carried out using the photoautotrophic microalgae (*Chlorella vulgaris*), the results show that yield of 91% CH_4 is obtained (Figure 3). This process was carried out using different species and the results discovered are shown in the table 6 [6].

2.3.2. Removal of H_2S

For removal of H_2S biological methods have been studied but on the laboratory scale. Microorganisms can remove H_2S from biogas by the following pathway:

$$HS^- \leftrightarrow H^+ + S^{2-}$$
$$0.5O_2 + HS^- \rightarrow OH^- + S^0$$
$$1.5O_2 + S^0 + H_2O \rightarrow 2H^+ + SO_4^{2-}$$
$$2O_2 + HS^- \rightarrow H^+ + SO_4^{2-}$$

There is a variety of microbes which can remove H_2S from biogas which are given in the table 7 [6].

3. Recent Advances in Biogas Purification Technologies

The techniques studied in order to accelerate biogas disintegration have been widely studied which include heat-induced steam explosion of waste material for intracellular biomass disintegration [10], sonification [11], and water plasma expansion using phytomass disintegration [1-12]. The recent work done in this field focuses on removal of CO_2 and H_2O more efficiently (Figure 4).

3.1. Pressure Swing Adsorption (PSA)

Adsorption system contains a solute which transfers with the stream of gas working as an adsorbent [3]. In PSA different gases like CO_2, N_2, and O_2 are separated at different pressures by using either activated carbon or zeolites [13]. Adsorption of the gases in PSA is a reversible process. H_2S can also be adsorbed in this process, but the process is irreversible, making it unfavorable for H_2S [6]. In order to protect the adsorbent material, H_2S is separated individually [14]. The yield of biomethane obtained using this technique ranges from 90% to 99% by different authors. This technique is very useful because it requires low energy consumption, no chemical is required, and it is compact, thus it is very common in low-scale industries. The drawbacks of this technique are that it is less efficient than other techniques and it requires treatment to remove H_2S and water before the process starts (Figure 5).

3.2. Absorption

The purification of biogas by the process of absorption requires liquid scrubbing so that CO_2 is absorbed using either chemical or physical processes [15].

3.2.1. High pressure water scrubbing (HPWS)

For this process it is required to know the solubility of all the ingredients of biogas because the principle of HPWS lies in the fact that CO_2 and H_2S have high solubility in H_2O, which is used as an absorbent, as compared to other components [16]. HPWS is carried out using packed columns with optimized conditions in order to achieve the highest emission of CO_2 [17]. Biomethane produced from this technique ranges between 95%-97%. This technique is easy to use, cheap, and able to remove NH_3 and H_2S from biogas. It requires low electricity consumption and is flexible in capacity, thus it is the most applied technique in industries. The drawbacks to this technique are that CH_4 obtained is of medium-high composition, losses of CH_4 are about 2%, is not very flexible in terms of composition of biogas, and a drying stage is required after scrubbing (Figure 6).

3.2.2. Organic physical scrubbing (OPS)

OPS is quite identical to that of water scrubbing except that the solvent used in it as an absorbent. In this technique an organic solvent like polyethylene glycol is used instead of water [18-19]. Solubility of CO_2 is higher in polyethylene glycol than in water, thus we require less adsorbent and a smaller equipment in OPS than HPWS [6]. Both techniques give almost the same yield [20]. Polyethylene glycol used in this process can be regenerated along with H_2O, H_2S, and O_2 [6]. On a commercial level, other adsorbents like selexol and genosorb are used [15-21]. CO_2 and NH_3 are more soluble in selexol than in H_2O, also selexol has low freezing point and is noncorrosive in nature [6]. Selexol has the ability to dissolve H_2S in higher quantities than CO_2 and for regeneration of H_2O and H_2S from selexol high temperature is required [6]. Low losses of CH_4 are faced in this technique than HPWS. It is also able to eliminate H_2S, ammonia, and other impurities, but a pre-treatment is required. Yield obtained from OPS is similar to that of HPWS i.e. CH_4 obtained is of medium-high composition, requires high investment for operation, and is difficult to operate because this technique requires high temperature for a complete regeneration of gases (Figure 7).

3.2.3. Chemical scrubbing

Chemical scrubbing involves chemical reaction between the absorbent and the absorbed gases [6]. The absorbent used in this technique is mostly an amine for the removal of CO_2 or H_2S either in the form of Monoethanolamine (MEA), Diethanolamine (DEA), or Methyldiethanolamine (MDEA) [6]. Chemical reactions occurring with amine in chemical scrubbing are highly selective in order to keep the losses below 0.1% and purity of biomethane above 99.9% [15-21]. Optimized conditions required for chemical scrubbing are 120^0C and 1-2 bar pressure [22-23]. The amine solution is also lost in this technique due to evaporation [14]. The regeneration of gases occurs at high temperature and requires high energy consumption [15]. This technique gives the highest purity of biomethane (>99%) and low losses are obtained are very minimum (<0.1%). There are no pressurization of biogas and High CO_2 elimination takes place. The drawbacks to this technique are that it requires a high investment cost, high temperature for regeneration of gases, causes corrosion and salt precipitation. In this technique, foam is produced and removal of H_2S requires pre-treatment (Figure 8).

3.3. Membrane separation

In membrane separation technique, membranes which are permeable to CO_2, H_2O, and NH_3 are used for the purification of biogas [6]. H_2S and O_2 also pass through the membrane. CH_4 passes through the membrane in a very low proportion and this makes the separation of gases possible [15]. The purification of biogas by membrane separation can take place by either wet process or dry process [6]. This technique can be simply constructed, the operation requires a little maintenance, and has modular configuration. This technique requires no heating and no chemical products. It is highly reliable and smaller gas flows can be treated without any greater cost increase. The drawbacks of this technique are that membrane has low, several stages are required to achieve high purity of biomethane, medium to high losses of CH_4. This technique is not suitable for biogas with undetermined pollutants, such as landfill gas or wastewater biogas. This technique has a high cost because of membranes and is not widespread in industries (Figure 9).

3.4. Cryogenic separation

The cryogenic separation technique is based on the difference in boiling point of different components for the purpose of purification of biogas. In this technique biogas is compressed at a low temperature so that CO_2 condenses and can be separated as liquid [6]. By this technique, high purity biomethane is produced, causes low methane losses (<1%), pure CO_2 is obtained as a byproduct, and no chemicals are required. The drawbacks of this technique are that it requires a high cost for operation, requires pre-treatment to eliminate contaminants, efficiency is low, and technology is not very demonstrated. This technique is very recently implemented on large-scale industries (Figure 10).

4. Future Perspectives

In all the methods we have studied so far, the purification process mainly focuses on removal of CO_2 and H2S. It is important that when a new study is carried out for purification of biogas, the substrate considered for use must be of low cost, environment-friendly, and safe to use. The process of biogas upgradation is intended to generate a sustainable and profitable process with zero waste. These considerations in the process will allow the user to produce biogas simply with the available material at a low cost. Use of biomethane helps in reducing waste, pollution, toxicity, and greenhouse gases, thus it is important to continue studies in this field. Since the yield of biomethane obtained from these processes is quite satisfactory, it is important that the future work done in this field should focus on improving the economics of this process and finding the better substrates for this process.

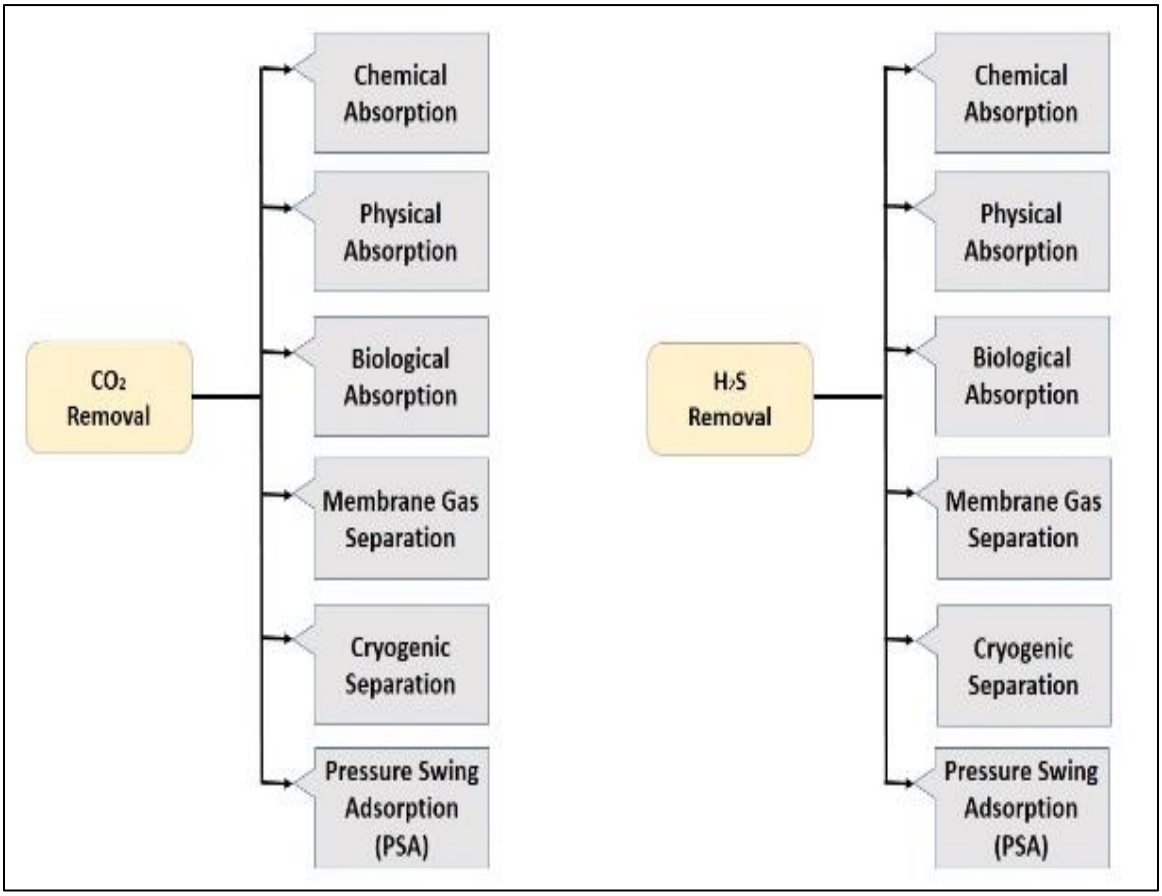

Figure 1: Different methods used for purification of biogas [2]

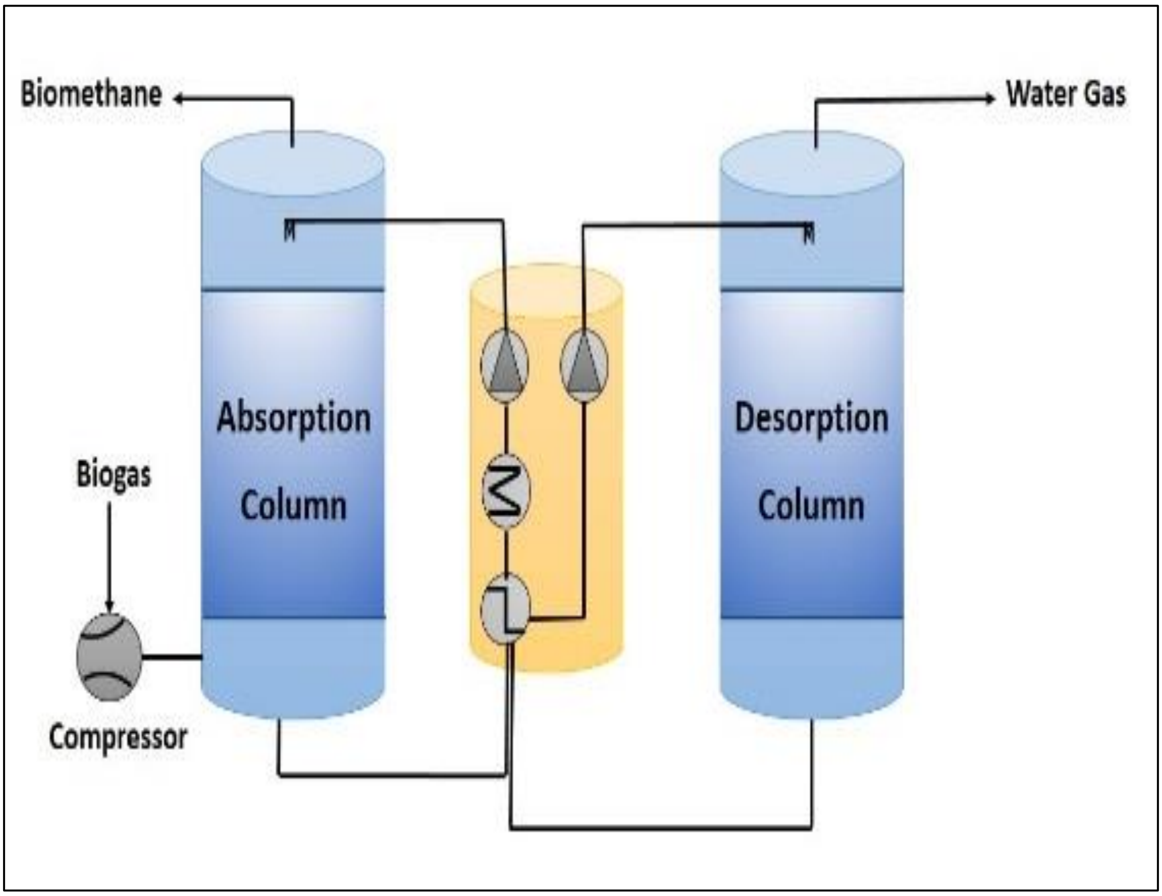

Figure 2: General scheme of biogas purification by water [7]

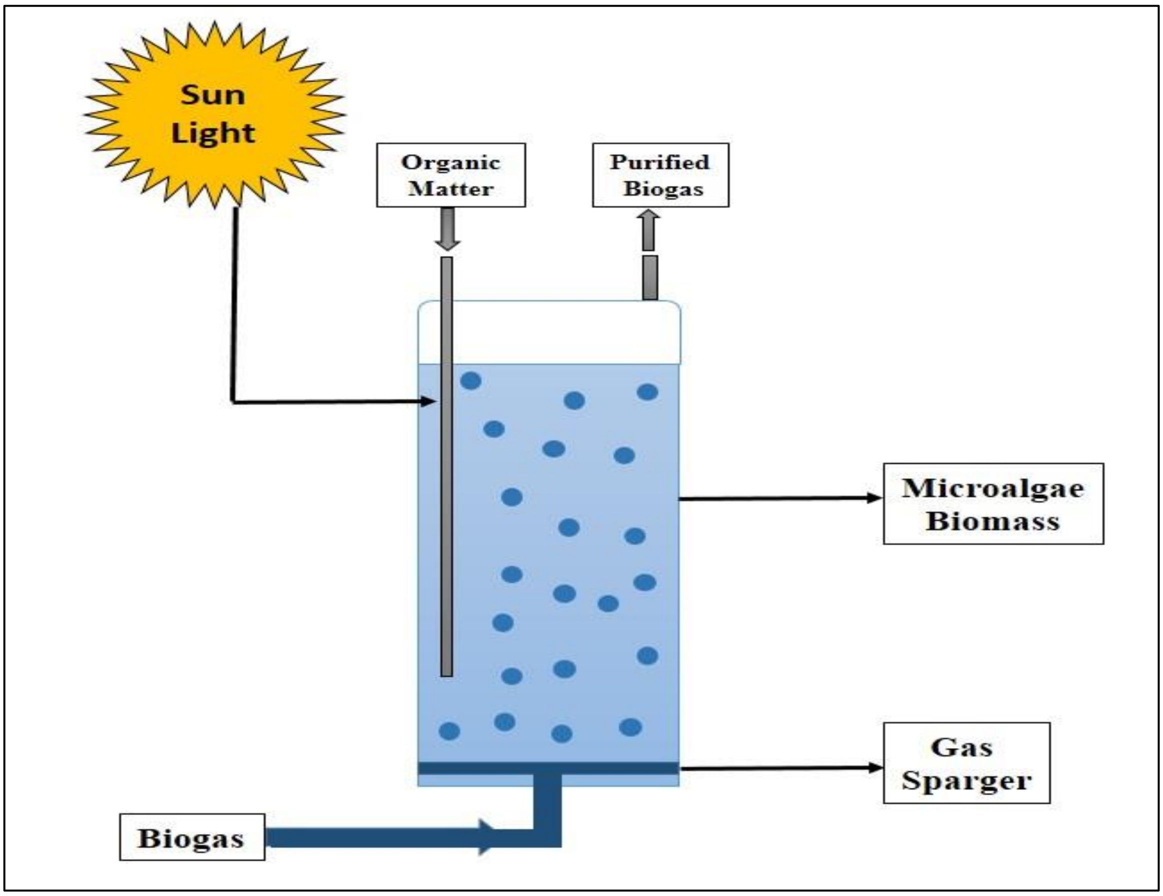

Figure 3: Involvements for microalgae growth [24]

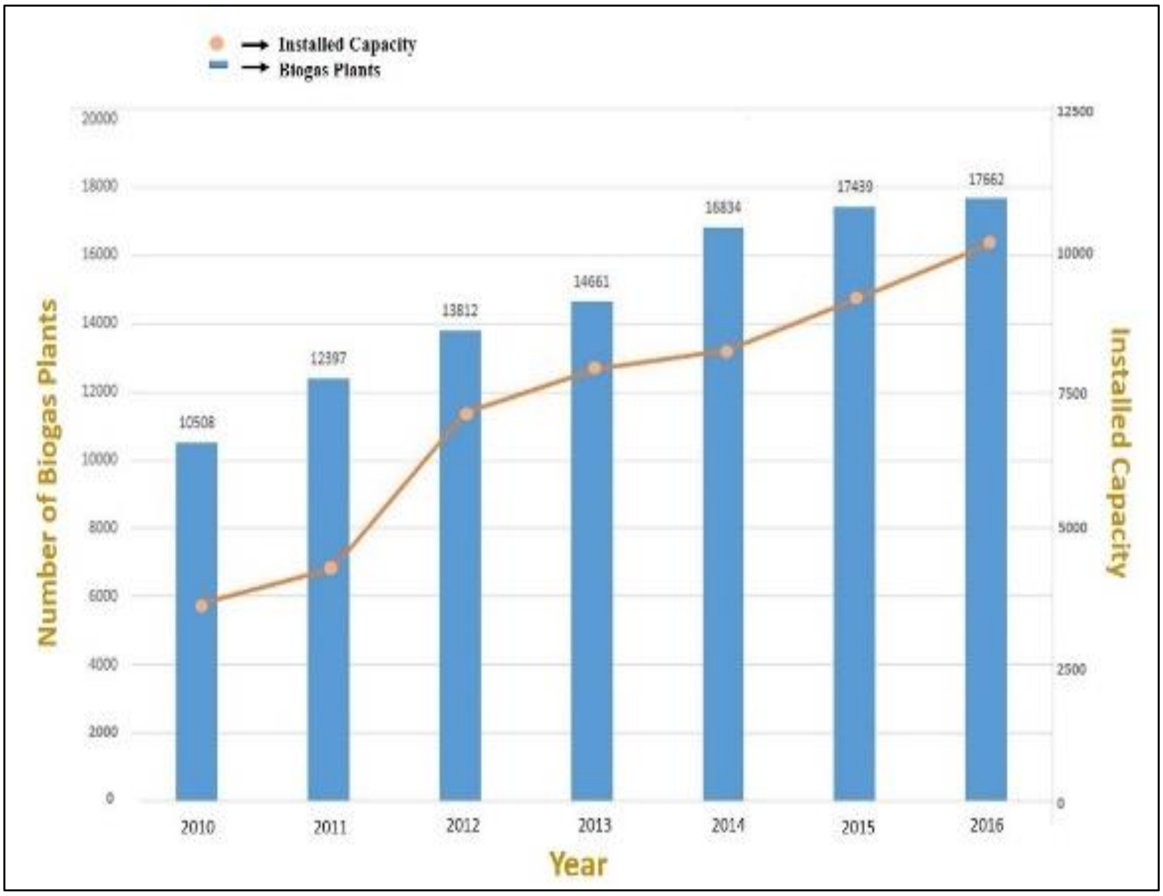

Figure 4: Plot between number of biogas plants VS installed capacity in Europe 2010–2016 [25]

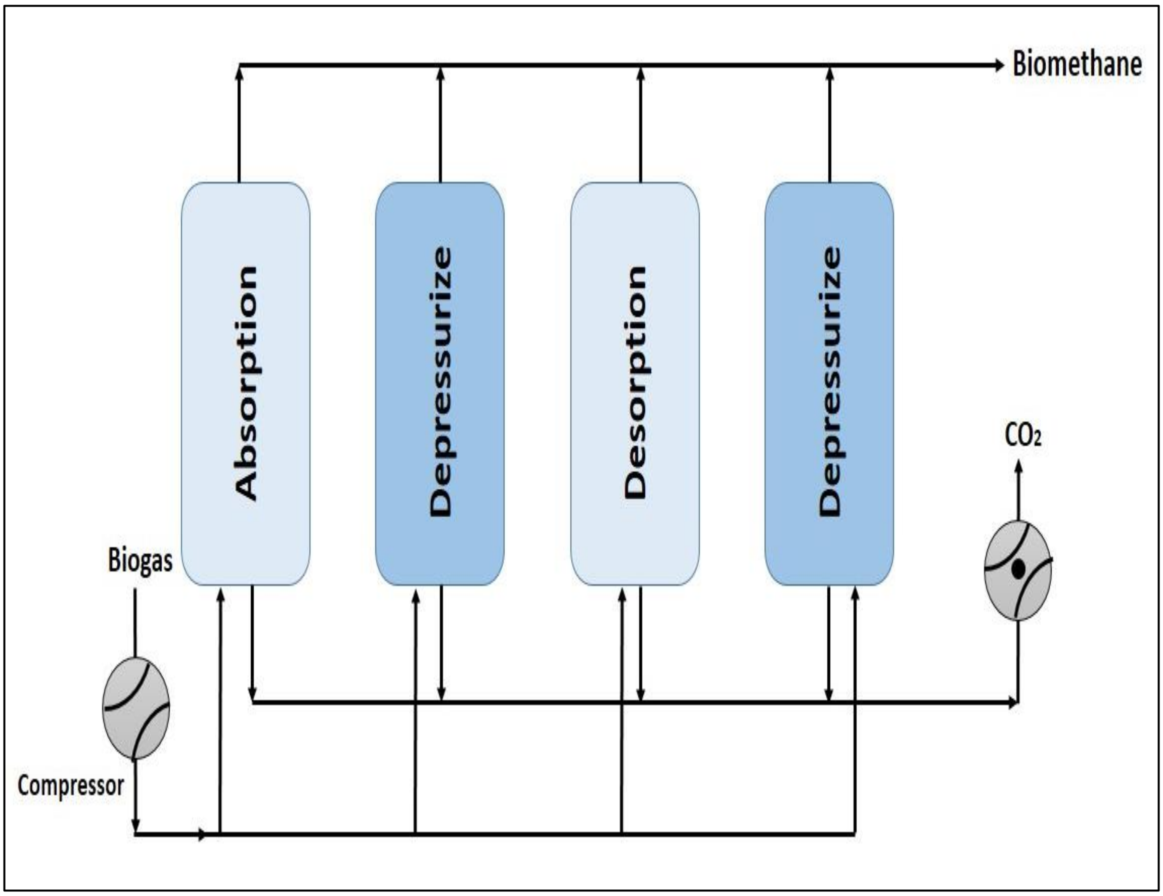

Figure 5: PSA System [14]

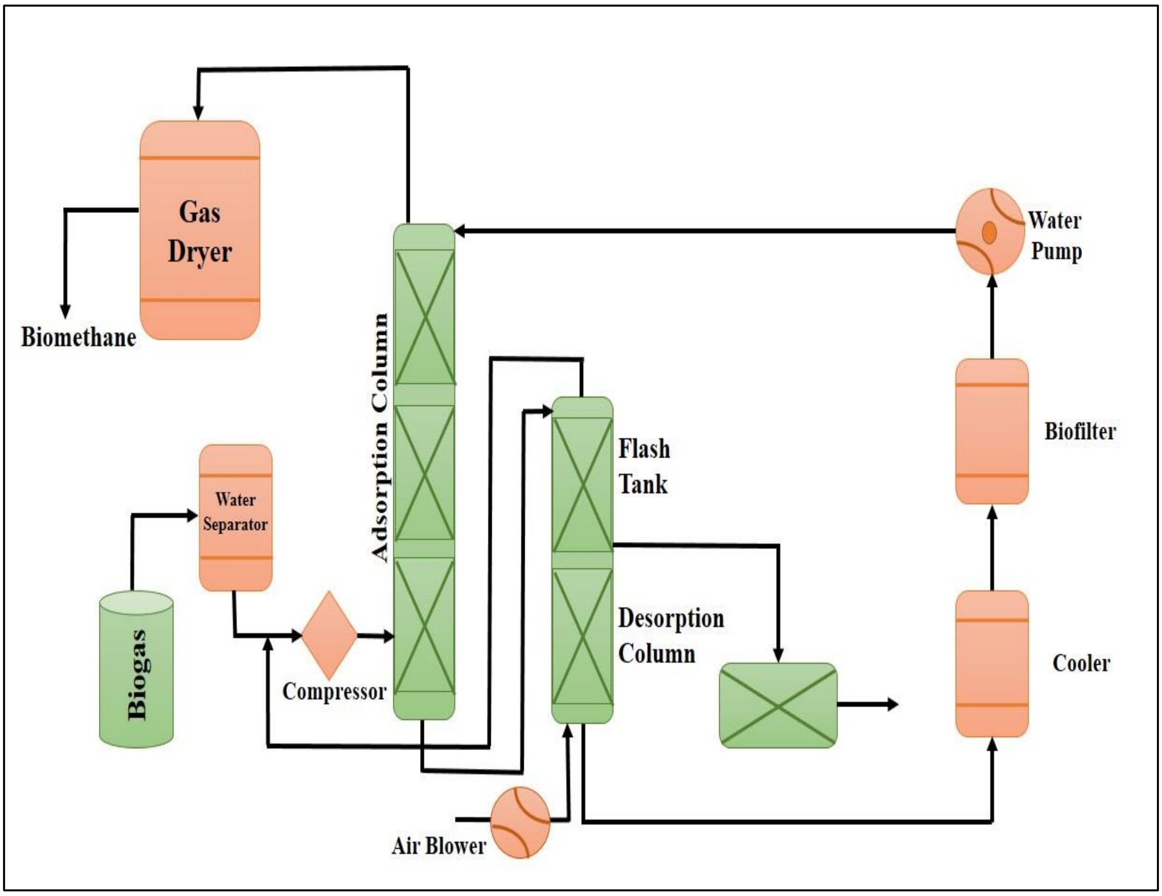

Figure 6: HPWS [16]

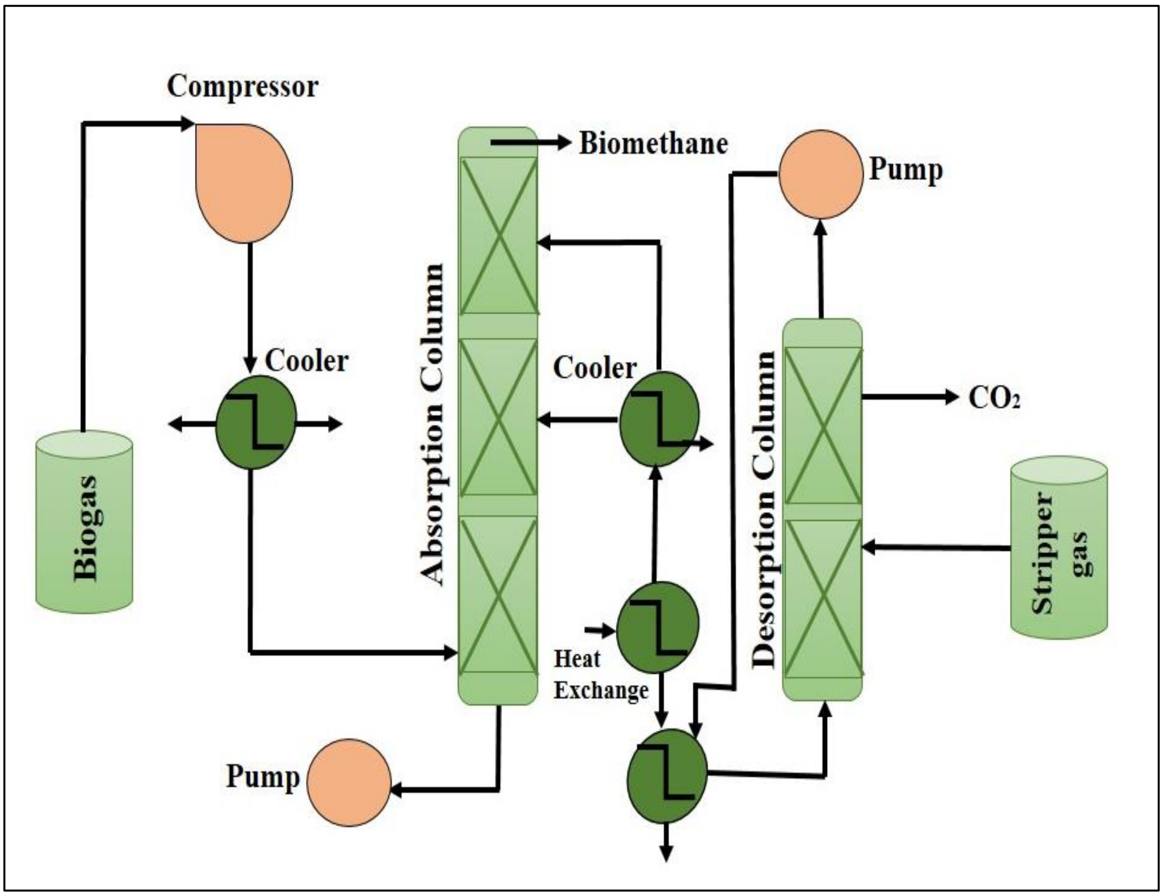

Figure 7: OPS process [26]

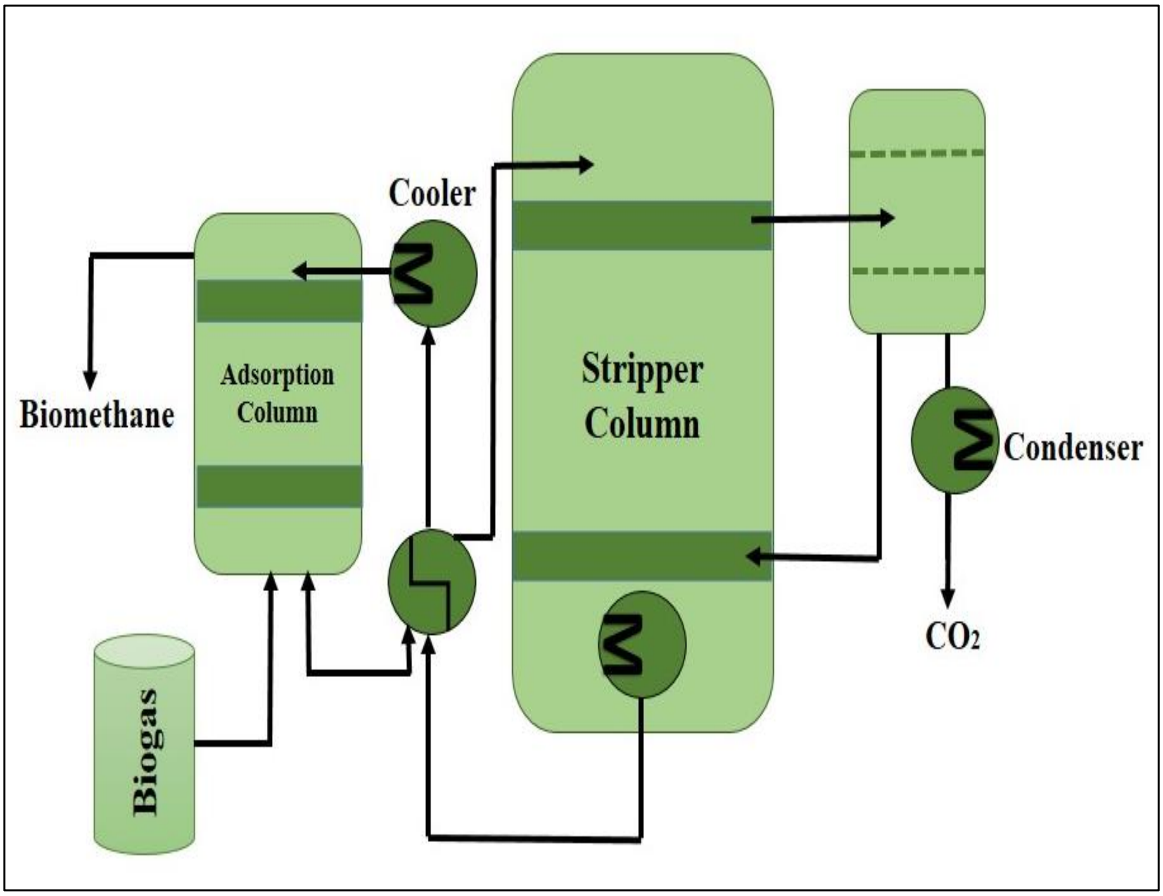

Figure 8: Chemical Scrubbing [27]

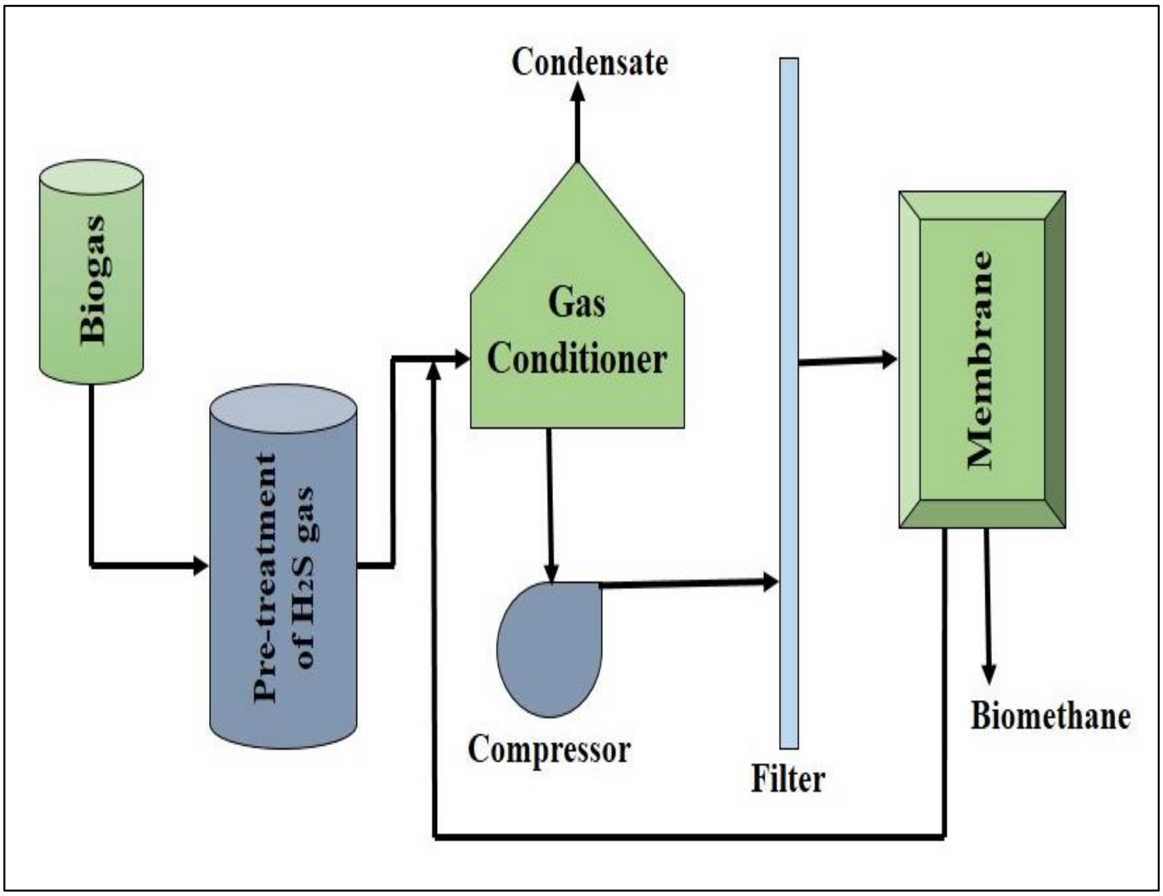

Figure 9: Membrane Separation [15]

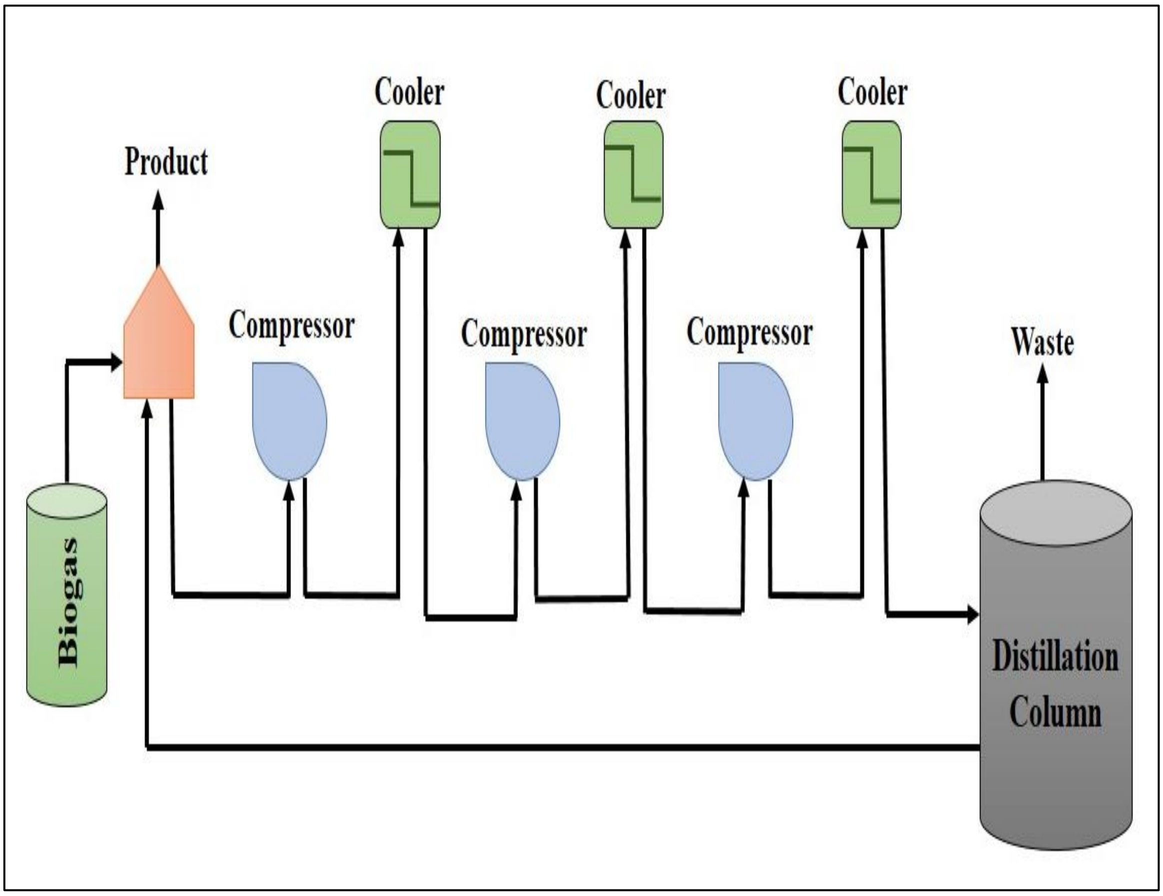

Figure 10: Cryogenic Separation [26]

Table 1: Difference of composition between natural gas and biogas

Components	Natural gas	Biogas	References
Methane	80-90 (% vol)	59-71 (% vol)	[14-15]
Hydrocarbons	3-10 (% vol)	-	[15-28]
Carbon dioxide	0.65-1.1 (% vol)	31-41 (% vol)	[15-28]
Nitrogen	0.0-0.20 (% vol)	0.27-14.5 (% vol)	[14]
Hydrogen Sulphide	0.1-4000.5 (ppm)	-	[14-15]
Ammonia	0.1-100.5 (ppm)	-	[14-28]

Table 2: Comparison of different reagents used for physical absorption of CO_2

Reagent	Flow of biogas	Flow of reagent	Temperature in °C	Initial CO_2 conc. (%)	Final CO_2 conc. (%)	Initial CH_4 conc. (%)	Recovered conc. Of CH_4 (%)	Reference
H_2O	1.5 m^3h^{-1}	1.80 m^3h^{-1}	AT*	40	1	60	85.0	[29]
H_2O	10.0 bar	15.1 Ls^{-1}	10.0	21.1	6.91	45.0	85.0	[30]
H_2O	25.0 $Lmin^{-1}$	20 $Lmin^{-1}$	26.0	–	–	69.0	93.0	[31]
H_2O	–	–	AT*	–	8	55-65	75-95	[32]
H_2O	–	50 $Lmin^{-1}$	AT*	15	11.62	66.6	–	[33]
H_2O	60 Ls^{-1}	0.149 Ls^{-1}	AT*	20.2	0.111	51.11	70.5	[34]
H_2O	1.29-1.81 $Lmin^{-1}$	0.20 $Lmin^{-1}$	AT*	42.72	4.981 L	54.631	90.0	[35]

Table 3: Comparison of different reagents used for physical absorption of H_2S

Reagents	Biogas flow	Reagent flow	Temperature (°C)	Initial H_2S	Final H_2S	CH_4 initial (%)	CH_4 recovered (%)	Reference
H_2O/ activated carbon/sawdust	0.651 m^3h^{-1}	20.01 $Lmin^{-1}$	27.00	8800.1 ppm (0.881%)	10.00 ppm (0.010%)	68.571	93.151	[6]
H_2O	140 $Lmin^{-1}$	–	AT*	6000 ppm (6%)	2933.3 ppm (0.29%)	–	–	[36]
H_2O	7.41 Nm^3h^{-1}	5.5-11 $Lmin^{-1}$	20-25	100-166.0 ppm (0.010-0.01660%)	13.21 ppm (0.00131%)	50.81-57.91	90.20	[37]
H_2O	–	50.01 $Lmin^{-1}$	–	18.40 mgL^{-1}	–	66.61	–	[33]

Table 4: Comparison of different reagents used for chemical absorption of CO_2

Reagent	Biogas flow	Reagent flow	Temperature (°C)	Initial CO_2 (%)	Final CO_2 (%)	Initial CH_4 (%)	Biomethane (%)	Reference
Soda lime	1 L min^{-1}	5 bar	AT*	–	1.340	41.50	97.7	[8]
$Ca(OH)_2$	5 L min^{-1}	10 L min^{-1}	AT*	39.60	–	51.0	89.30	[38]
NaOH	–	–	AT*	46.0	16.0	52.0	80	[39]
MEA	–	–	AT*	46.0	15.0	52.0	85	[40]

AT* = Ambient Temperature

Table 5: Comparison of different reagents used for chemical absorption of H_2S

Reagents	Biogas inflow	Initial H_2S (ppm)	Final H_2S (ppm)	Removal H_2S (%)
Charcoal	$7.94\ m^3$ (28 $Lmin^{-1}$)	68.09	1.98	97.08
Bokashi	$1.70\ m^3$ (6 $Lmin^{-1}$)	92.61	3.71	96.01
Iron filings	–	–	250	94.08
Iron shaving	111.6 Lh^{-1}	180	9.37	94.79
Iron shaving	111.6 Lh^{-1}	180	0.75	99.58
Activated carbon	–	1000	610	39
FeO / activated carbon	–	1000	515	48.5
Commercial activated carbon	–	1000	687.5	31.25
Fe_2O_3/H_2O	–	1500	8	99.46
CaO	7 kPa	370	225	40

Table 6: Comparison of different species used for biological absorption of CO_2

Species	Biogas flow ($Lmin^{-1}$)	Initial CO_2 conc. (%)	Final CO_2 conc. (%)	Initial CH_4 (%)	Final CH_4 (%)
Chlorella vulgaris	0.9	28	8.56	71	91
Chlorella vulgaris	1.8	28	15	71	85
Arthrospira platensis	–	19.5	–	76	–
Chlorella vulgaris (10%w L^{-1})	–	39.6	32.2	60.4	67.6
Chlorella vulgaris (30%w L^{-1})	–	39.6	25.5	60.4	74.5
Chlorella vulgaris (60%w L^{-1})	–	39.6	39	60.4	60.5
Chlorella vulgaris /*G. lucidum*	–	33.7	23.5	64.59	–

Table 7: Comparison of different species used for biological absorption of H_2S

Bacteria	Genus
Aerobic	*Acidianus, Acidithiobacillus, Aquaspirilum, Aquifex, Bacillus, Beggiota, Methylobacterium, Paracoccus Pseudomonas, Starkeya, Sulfolobus, Thermitiobacillus, Thiobacillus y Xanthobacter.*
Anaerobic	*Allochromatium, Chlorobium, Rhodopseudomonas, Rhodovulum Thiocapsa*

5. Conclusions

Biogas is generated from the fermentation of organic matter, which consists of gases like CO_2 and H_2S, making biogas a toxic and pungent gas with corrosive properties. Due to these component gases, biogas cannot be used as raw biogas as a source of fuel, and it needs purification. For the purification of biogas, different methods are used which mainly focus on removal of H_2S and CO_2. Water is employed as a natural absorbent in physical processes, however the volume of water required makes this procedure not entirely feasible on a large scale. The most effective methods for eliminating CO_2 and H_2S (over 90%) have been chemical processes; however, use of diluted chemicals may produce additional residues that need to be treated or neutralized. When it comes to purifying biogas, biological techniques are new and have proven to be more than 90% successful. However, research employing biological techniques has only been carried out in lab settings. When significant CH_4 losses are prohibited, cryogenic technologies have shown to be successful. In terms of various costs, membrane separation plants have the lowest CC, while water scrubbing has the lowest OMC. The structure of the biogas plant must be taken into consideration when determining method of conditioning the raw biogas because certain specifics may be needed. These methods are being used for the purification of biogas and studies in this field are also trying to improve the process day by day in terms of using safe and environment-friendly substrates and making the process with less cost. The use of purified biogas helps in reducing pollution, toxicity, and emission of greenhouse gases caused by use of raw biogas.

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