Biogas scrubbing, compression and storage: perspective and prospectus in Indian context

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Received 8 May 2003; accepted 23 September 2004

Abstract

Biogas is a clean environment friendly fuel. Raw biogas contains about 55-65% methane (CH\textsubscript{4}), 30-45% carbon dioxide (CO\textsubscript{2}), traces of hydrogen sulfide (H\textsubscript{2}S) and fractions of water vapours. Presently, it can be used only at the place where it is produced. There is a great need to make biogas transportable. This can be done by compressing the gas in cylinders which is possible only after removing its CO\textsubscript{2}, H\textsubscript{2}S and water vapour components. Pilot level trials to compress the biogas have been carried out by a number of earlier investigators working on the subject. This paper reviews the efforts made to improve the quality of biogas by scrubbing CO\textsubscript{2} and the results obtained. There is a lot of potential if biogas could be made viable as a transport vehicle fuel like CNG by compressing it and filling into cylinders after scrubbing and drying. Thus the need emerges for a unified approach for scrubbing, compressing and subsequent storage of biogas for wider applications.

Keywords: Biogas; Scrubbing; Compression; Storage

1. Introduction

Biogas is produced by anaerobic digestion of biological wastes such as cattle dung, vegetable wastes, sheep and poultry droppings, municipal solid waste, industrial waste water, land fill, etc. It is an environment friendly, clean, cheap and versatile fuel. Historically, the biogas technology is over a hundred years old in India. The National Project on Biogas Development (NPBD) was launched by Government of India in 1981.
A total of about 3.4 million family size biogas plants had been installed all over India by Dec. 2002. This is only 28.3% of the total potential (12 million) of the family size biogas plants that can be put up in India [1].

Also, more than 3380 Community Biogas Plants (CBP), Institutional Biogas Plants (IBP) and Night-soil based Biogas Plants (NBP) have been installed all over the country. Biogas plant having capacity 1-10 cum is categorized as family size biogas plant whereas above 10 cum is known as large size biogas plant (CBP/IBP/NBP). Installed biogas plants are estimated to be saving 3.96 million tonnes of fuel-wood besides producing about 920 thousand tonnes of enriched organic manure every year. Apart from these economic benefits, biogas plants have provided many indirect social benefits; such as reduction in the drudgery of rural women and children involved in the collection of fuel materials from long distances, reduction in the incidences of lung and eye diseases from cooking in smoky kitchens and an overall improvement in the standard of living. During year 2000-2001 alone, 164 thousand biogas plants were constructed, generating employment to the tune of 5 million man-days. By linking biogas plants to the toilets, about 160 thousand families in the rural areas and 350 thousand people living in the slums in and around cities have been provided with good sanitation facilities [2].

Production of biogas could be a continuous process. However, its use is limited near to the site of the biogas plant. The presence of uncombustible gases like CO2, H2S and water vapour reduce its calorific value and make it uneconomical to compress and transport to longer distances. It is therefore necessary to remove these gases before compression.

2. CO2 scrubbing from biogas

A variety of processes are being used for removing CO2 from natural gas in petrochemical industries. Several basic mechanisms are involved to achieve selective separation of gas constituents. These may include physical or chemical absorption, adsorption on a solid surface, membrane separation, cryogenic separation and chemical conversion.

2.1. Physical absorption

For biogas scrubbing physical/chemical absorption method is generally applied as they are effective even at low flow rates that the biogas plants are normally operating at. Also the method is less complicated, requires fewer infrastructures and is cost effective.

One of the easiest and cheapest method involves the use of pressurized water as an absorbent. The raw biogas is compressed and fed into a packed bed column from bottom; pressurized water is sprayed from the top. The absorption process is, thus a counter-current one. This dissolves CO2 as well as H2S in water, which are collected at the bottom of the tower. The water could be recycled to the first scrubbing tower [3]. This perhaps is the simplest method for scrubbing biogas.
Bhattacharya et al. [4] developed one such water scrubbing system. The process provides 100% pure methane but is dependent on factors like dimensions of scrubbing tower, gas pressure, composition of raw biogas, water flow rates and purity of water used.

Vijay [5] developed a packed bed type scrubbing system using the locally available packing materials removing 30^-0% more CO2 by volume compared with the scrubbing systems without a packed bed.

Khapre [6] designed a continuous counter-current type scrubber with gas flow rate of 1.8 m$^3$/h at 0.48 bar pressure and water inflow rate of 0.465 m$^3$/h. It continuously reduced CO2 from 30% at inlet to 2% at outlet by volume.

Dubey [7] tried three water scrubbers having diameters 150 mm (height: 1.5 m), 100 mm (height: 10 m) and 75 mm (height: 10 m) to absorb CO2 present (37-41%) in the biogas. He found that the CO2 absorption is influenced by the flow rates of gas and water than different diameters of scrubbers.

The G.B. Pant University of Agriculture and Technology, Pantnagar, India [8] developed a 6 m high scrubbing tower, packed up to 2.5 m height with spherical plastic balls of 25 mm diameter. The raw biogas compressed at 5.88 bar pressure was passed at a flow rate of 2 m$^3$/h while water was circulating through the tower. A maximum of 87.6% of the CO2 present could be removed from the raw biogas.

Water scrubbing method is popular for CO2 removal in sewage sludge based biogas plants in Sweden, France and USA. The results show that 5-10% CO2 remains in biogas after scrubbing [9].

2.2. Chemical absorption

Chemical absorption involves formation of reversible chemical bonds between the solute and the solvent. Regeneration of the solvent, therefore, involves breaking of these bonds and correspondingly, a relatively high energy input. Chemical solvents generally employ either aqueous solutions of amines, i.e. mono-, di- or tri-ethanolamine or aqueous solution of alkaline salts, i.e. sodium, potassium and calcium hydroxides.

Biswas et al. [10] reported that by bubbling biogas through 10% aqueous solution of mono-ethanolamine (MEA), the CO2 content of the biogas was reduced from 40 to 0.5-1.0% by volume. MEA solution can be completely regenerated by boiling for 5 min and thus can be used again.

Savery et al. suggested that the three agents NaOH, KOH and Ca(OH)$_2$ can be used in chemical scrubbing of biogas. The absorption of CO2 in alkaline solution is assisted by agitation. The turbulence in the liquid aids to diffusion of the molecule in the body of liquid and extends the contact time between the liquid and gas. Another factor governing the rate of absorption is concentration of the solution. The rate of absorption is most rapid with NaOH at normality’s of 2.5-3.0.

2.3. Adsorption on a solid surface

Adsorption process involves the transfer of solute in the gas stream to the surface of a solid material, where they concentrate mainly as a result of physical or Vander wall forces. Commercial adsorbents are generally granular solids with a large surface area per unit
volume. By a proper choice of adsorbent, the process can remove CO₂, H₂S, moisture and other impurities either selectively or simultaneously from biogas.

Gas purification can also be carried out using some form of silica, alumina, activated carbon or silicates, which are also known as molecular sieves [12].

Adsorption is generally accomplished at high temperature and pressure. It has good moisture removal capacities, simple in design and easy to operate. But it is a costly process with high pressure drops and high heat requirements.

Schomaker et al. [13] reported that CO₂ could be removed from biogas by pressure swing adsorption which consists of at least three active carbon beds. One of the beds is fed with biogas under pressure (6 bar) CO₂ is adsorbed. When there is saturation of CO₂ in the adsorption bed, the process is shifted to the second bed. The saturated bed is depressurized to ambient pressure. The efficiency of this process is up to 98%.

Continuous monitoring of a small-scale installation (26 m³/h) in Sweden using pressure swing adsorption technique through carbon molecular sieves have given excellent results in terms of clean gas, energy efficiency and cost [9].

Pandey and Fabian [14] used naturally occurring zeolite-Neopoliton Yellow Tuff (NYT) for adsorption. They found that the active component for CO₂ adsorption is chabazite, which has adsorption capacity of 0.4 kg CO₂ per kg of chabazite at 1.50 bar and 22 °C. During the adsorption process the H₂S content is also reduced.

2.4. Membrane separation

The principle is that some components of the raw gas could be transported through a thin membrane (∼1 mm) while others are retained. The transportation of each component is driven by the difference in partial pressure over the membrane and is highly dependent on the permeability of the component in the membrane material. For high methane purity, permeability must be high. Solid membrane constructed from acetate-cellulose polymer has permeability for CO₂ and H₂S up to 20 and 60 times, respectively, higher than CH₄. However, a pressure of 25^1-0 bar is required for the process [15].

Wellinger and Lindberg [9] described two basic systems of gas scrubbing with membranes: a high pressure gas separation with gas phases on the both sides of the membrane and a low pressure gas liquid absorption on separation where a liquid absorbs the molecule diffusing through the membrane. The high pressure gas separation membranes can last up to 3 years which is comparable to the life time of membranes used for natural gas purification—which last typically 2-5 years.

Rautenbach et al. [16] designed a pilot plant for the removal of CO₂ from biogas using membrane separation technique. He reported that monsanto and acetate cellulose membranes are more permeable to CO₂, O₂ and H₂S than CH₄. The best separation occurred at 25 °C temperature and 5.50 bar pressure.

The gas flux across the membrane increases proportionally with the partial pressure difference. Thus, higher the pressure difference, the smaller is the membrane area required. However, maximum pressure which membrane can withstand must be taken into consideration [17].
2.5. Cryogenic separation

The cryogenic method of purification involves the separation of the gas mixtures by fractional condensations and distillations at low temperatures. The process has the advantage that it allows recovery of pure component in the form of a liquid, which can be transported conveniently. However, attempts to apply the cryogenic process for the removal of CO2 from digester gas by Los Angelus County sanitation have not proven successful. Rather complicated flow streams are involved and thermal efficiency is low. Capital cost and utility requirements are also high [12].

In a cryogenic method, crude biogas is compressed to approximately 80 bar. The compression is made in multiple stages with inter-cooling. The compressed gas is dried to avoid freezing during cooling process. The biogas is cooled with chillers and heat exchangers top K45 8C, condensed CO2 is removed in a separator. The CO2 is processed further to recover dissolved methane, which is recycled to the gas inlet. By this process more than 97% pure methane is obtained. No data is available on investment and operational cost [15].

2.6. Chemical conversion method

To attain extremely high purity in the product gas, chemical conversion method can be used. It reduces the undesirable gas concentrations to trace levels. Usually the chemical conversion process is used after bulk removal has been accomplished by other methods. One such chemical conversion process is methanation, in which CO2 and H2 are catalytically converted to methane and water. Chemical conversion process is extremely expensive and is not warranted in most biogas applications [17].

Due to highly exothermic nature of the methanation reactions, the removal of the heat from the methanator is a major concern in the process design. The requirement of the large amount of pure hydrogen also makes the process generally unsuitable.

3. Scrubbing of H2S

H2S is always present in biogas, although concentrations vary with the feedstock. It has to be removed in order to avoid corrosion in compressors, gas storage tanks and engines [9].

H2S is poisonous and corrosive as well as environmentally hazardous since it is converted to sulfur dioxide by combustion. It also contaminates upgrading process. H2S can be removed either in the digester, from the crude biogas or in upgrading process [15].

The most commonly used H2S removal process can be classified into two general categories namely: (1) dry oxidation process and (2) liquid phase oxidation process [12].

3.1. Dry oxidation process

It can be used for removal of H2S from gas streams by converting it either into sulfur or oxides of sulfur. This process is used where the sulfur content of gas is relatively low and high purities are required. Some of these methods are described below.
3.1.1. Introduction of air/oxygen into the biogas system

A small amount of oxygen (2-6%) is introduced in the biogas system by using air pump. As a result, sulfide in the biogas is oxidized into sulfur and \( \text{H}_2\text{S} \) concentration is lowered.

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

This is a simple and low cost process. No special chemicals or equipments are required. Depending on the temperature, the reaction time and place where the air is added, the \( \text{H}_2\text{S} \) concentration can be reduced by 95% to less than 50 ppm. However, care should be taken to avoid overdosing of air, as biogas in air is explosive in the range of 6-12%, depending on the methane content [9].

3.1.2. Adsorption using iron oxide

\( \text{H}_2\text{S} \) reacts with iron hydro-oxides or oxides to form iron sulfide. The biogas is passed through iron oxide pellets, to remove \( \text{H}_2\text{S} \). When the pellets are completely covered with sulfur, these are removed from the tube for regeneration of sulfur. It is a simple method but during regeneration a lot of heat is released. Also the dust packing contains a toxic component and the method is sensitive to high water content of biogas.

Wood chips covered with iron oxide have a somewhat larger surface to volume ratio than plain steel. Roughly 20 g of \( \text{H}_2\text{S} \) can be bound per 100 g of iron oxide chips. The application of wood chips is very popular particularly in USA. It is a low cost product, however, particular care has to be taken that the temperature does not rise too high while regenerating the iron filter [9].

\( \text{H}_2\text{S} \) can be adsorbed on activated carbon. The sulfur containing carbon can then either be replaced with fresh activated carbon or regenerated. It is a catalytic reaction and carbon acts as a catalyst [15].

3.2. Liquid phase oxidation process

This process is primarily used for the treatment of gases containing relatively low concentration of \( \text{H}_2\text{S} \). It may be either: (a) physical absorption process or (b) chemical absorption process.

In physical absorption process the \( \text{H}_2\text{S} \) can be absorbed by the solvents. One of the solvent is water. But the consumption of water is very high for absorption of small amount of \( \text{H}_2\text{S} \). If some chemicals like NaOH are added to water, the absorption process is enhanced. It forms sodium sulfide or sodium hydrosulfide, which is not regenerated and poses problems of disposal.

Chemical absorption of \( \text{H}_2\text{S} \) can take place with iron salt solutions like iron chloride. This method is extremely effective in reducing high \( \text{H}_2\text{S} \) levels. The process is based on the formation of insoluble precipitates. FeCl3 can be added directly to the digester slurry. In small anaerobic digester system, this process is most suitable. All other methods of \( \text{H}_2\text{S} \) removal are suitable and economically viable for large-scale digesters. By this method the final removal of \( \text{H}_2\text{S} \) is about 10 ppm.
4. Biogas compression and storage

Biogas, containing mainly methane, could not be stored easily, as it does not liquefy under pressure at ambient temperature (critical temperature and pressure required are -82.5°C and 47.5 bar, respectively).

Compressing the biogas reduces the storage requirements, concentrates energy content and increases pressure to the level required overcoming resistance to gas flow. Compression is better in the scrubbed biogas. Most commonly used biogas storage systems are given in Table 1 [18].

Integrated units with facilities for scrubbing, compressing and storing have been developed in certain developed countries. For instance a water scrubber coupled with a gas compressor is being promoted for uniform use in New Zealand. Similarly, the biogas produced from poultry manure is being dried, scrubbed, compressed and stored at a pressure of 4 bar in 0.2 m$^3$ steel tanks in Belgium [19].

Khapre [6] conducted a study on scrubbing and compression of biogas and subsequently used it for domestic cooking. He found reduced requirement of scrubbed and compressed biogas (0.353 m$^3$) than raw biogas (0.591 m$^3$) for cooking a day’s meal of a six member family. He stored the scrubbed and compressed biogas at a pressure of 7 bar in cylinder of 0.1 m$^3$ capacity.

By purifying the biogas produced from the distillery wastes, scientists of Jadavpur University, Kolkatta, India [20] claimed to have generated huge quantities of compressed methane, a gas with an immense potential and an alternative source of vehicle fuel. Experimenting with bulk distillery wastes, from alcohol manufacturing breweries, researchers produced the gas by bio-methanation of the effluents.

Similar results have also been reported from Netherlands, UK, Australia, New Zealand and USA. All these results indicate that biogas is one of the potential substitutes for present day fuels including CNG, petrol, diesel and LPG [9].

Nema and Bhuchner [21] stressed on value addition to biogas by scrubbing and compressing, making it as good as the compressed natural gas (CNG). They reported the economic feasibility of producing energy from solid wastes of Delhi city. From 5000 tonnes wastes generated per day in Delhi, 100,000 Nm3/day biogas can be produced which is equivalent to 309.5 m$^3$ CNG worth US $ 70,000 per day. Beside this, by adopting this technology 117 tonnes/day CO2 gas can be prevented from entering into the atmosphere.

<table>
<thead>
<tr>
<th>Pressure</th>
<th>Storage device</th>
<th>Material</th>
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<tbody>
<tr>
<td>Low (0.138-0.414 bar)</td>
<td>Water sealed gas holder</td>
<td>Steel</td>
</tr>
<tr>
<td>Low</td>
<td>Gas bag</td>
<td>Rubber, plastic, vinyl</td>
</tr>
<tr>
<td>Medium (1.05-1.97 bar)</td>
<td>Propane or butane tanks</td>
<td>Steel</td>
</tr>
<tr>
<td>High (200 bar)</td>
<td>Commercial gas cylinders</td>
<td>Alloy</td>
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</table>
5. Conclusions

The aim of this study is to explore the potential of biogas production in India from animal waste and its prospects in wider perspective. Presently, biogas is mainly used for cooking purpose in India. To tap full potential of biogas, need emerges for its commercialization by making it transportable. Therefore biogas scrubbing and compression at high pressure for storage in cylinders are essential.

Different methods of scrubbing are reviewed and found that water scrubbing is simple, continuous and less expensive method for CO$_2$ removal from biogas for Indian conditions. It simultaneously also removes H$_2$S. After removal of CO$_2$, biogas is enriched in methane and becomes equivalent to natural gas. It can be used for all such applications for which natural gas is being used viz. as a fuel for vehicles, CHP, electricity generation, etc.

References