

Upgrading Biogas to Biomethane by Physical Absorption Process

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Abstract

Biogas is generated from anaerobic digestion of organic wastes by micro organisms. It is a relatively simple and economical method to produce a fuel from waste. It has a composition of methane as the main component with 50-70 percent, carbon dioxide around 30-40 percent and, hydrogen sulphide and moisture in trace quantities. The composition of biogas varies depending upon the substrates used. The raw biogas from the biogas digesters is properly scrubbed and purified to remove the unwanted gases like CO₂, H₂S and moisture upto a certain required level. In this work, a biogas upgrading system of 2 m³/h capacity has been developed for the removal of carbon dioxide and hydrogen sulphide from biogas, is indispensable to get biomethane. Developed biogas upgrading system, upgrades the biogas up to 96- 97% biomethane at 10 bar column-operating pressure with 1.5- 2.5 m³ /h of biogas in-flow rate and 1.75 m³/h of water inflow rate.

Highlights

- Highest CO₂ absorption 93.13% was observed at 10 bar gas pressure.
- Increasing water flow rate behind 1.75 m³/h there is no effect on CO₂ absorption percentage.
- Bureau of Indian Standards for biogas utilization in vehicles meet out by physical absorption process.

Keywords: Methane, CO₂ and H₂S absorption, Water scrubbing, Biogas upgrading

Biogas which is produced through the anaerobic degradation of organic materials is proposed as one of the most promising renewable fuels. Biogas can be produced in different environments, with sewage digesters producing a mixture containing a volumetric fraction of 55-65% methane (CH₄), 35-45% carbon dioxide (CO₂) and 1% nitrogen (N₂); organic waste/cattle dung digesters 60-70% CH₄, 30-40% CO₂ and 1% N₂; landfills 45-55% CH₄, 30-40% CO₂ and 5 - 15% N₂ (Rasi *et al.*, 2007).

The total potential of biogas availability in India from all sources has been estimated to be 48,382 million m³/year (Vijay, 2014). Currently, biogas is mainly applied in small to medium scale heat and power production. However, the high CO₂ content reduces the fuel calorific value and in turn limits the engine peak power. In addition, at low engine loads (i.e. at low combustion temperatures), engines fuelled with biogas suffer from combustion instability and high CH₄ concentrations in the exhaust



(Nathan *et al.*, 2010). Therefore, instead of combusting the biogas directly, removing CO_2 content and using the biomethane (from biogas) for combustion has been seen as an attractive fuel for IC engines, for power generation, and also in transportation.

Although biogas production in India is increasing, establishment of biogas upgrading system is very limited. Biogas upgrading scenario reveals that represent only laboratory-scale and pilot-level demonstration plants are available at present. Bhattacharya *et al.*, (1988) developed a water scrubbing system that produces 100% pure CH_4 , but is dependent on factors like dimensions of scrubbing tower, gas pressure, and composition of raw biogas, water flow rates and purity of water used. Similarly, Vijay, (1989) developed a packed bed type scrubbing system using locally available packing materials and reported that CO_2 removal was 30–40% more by volume compared with the scrubbing systems without a packed bed. The quality of biomethane is also affected by the water flow rate, scrubber dimensions and the number of scrubbers. In a continuous counter-current type scrubber with gas flow rate of $1.8 \text{ N m}^3 \text{ hour}^{-1}$ at 0.48 bar pressure and water in flow rate of $0.465 \text{ m}^3 \text{ hour}^{-1}$, CO_2 concentration was reduced from 30% at inlet to 2% at outlet by volume (Khapre, 1989).

Ramanathan, (1996) developed a single packed column using water as absorbent. Complete removal of CO_2 was obtained at a water flow rate of 0.1 L/s and a gas flow rate of 0.3 L/s at gas pressures upto 5 bar. Water recirculation was possible upto 20 minutes in packed column for 100 percent CO_2 removal. Dubey, (2000) reported that the CO_2 absorption is influenced by the flow rates of gas and water, but not by the diameter of scrubbers. The G.B. Pant University of Agriculture and Technology, Pantnagar, India 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 $2 \text{ Nm}^3 \text{ hour}^{-1}$, while water was being circulated through the tower. A maximum of 87.6% of the CO_2 present could be removed from the raw biogas. Vijay, (2006) designed a water scrubbing system using a gas inlet pressure and flow rate of 1 bar and $1.5 \text{ m}^3/\text{h}$ respectively, while the corresponding water flow rate was $1.8 \text{ m}^3/\text{h}$.

In order to convert biogas into biomethane, two major steps are required: (1) a cleaning process to remove the trace components and (2) an upgrading process to adjust the calorific value. In these processes, reducing CO_2 and H_2S content will significantly improve the quality of biogas. Various technologies have been developed for upgrading biogas to biomethane. These include absorption by physical absorption, chemical solvents, cryogenic separation, membrane separation and CO_2 fixation by biological or chemical methods (Abatzoglou and Boivin, 2009). The most common method used is the absorption of carbon dioxide in water at elevated pressure. The method is called water scrubbing and can be out-lined with or without regeneration of the water. The second most common method is an adsorption process called, Pressure Swing Adsorption (PSA) process. Carbon dioxide is adsorbed on activated carbon at elevated pressure and released when the pressure is reduced down to vacuum. It is a costly process.

All upgrading methods involve some loss of methane in the process. Since methane is a strong green house gas (GHG) with about 20 times stronger than carbon dioxide, it become necessary important to reduce the methane losses. For water scrubber and PSA have 2% maximum methane losses in upgrading plants (Persson, 2007). One of the simple and cheapest, less methane loss method involves use of water as an absorbent liquid for removal of CO_2 , due to simplicity, it suits for rural applications also (Vijay, 2007).

In physical absorption for gas separation, the main requirements are that the gas be brought into intimate contact with the liquid, and the effectiveness of equipment will largely be determined by the creation of degree of success contact between two phases. The biogas is allowed from the bottom and the liquid is sprayed from the top in a counter current manner in a shell filled with packing materials. The purified gas is obtained at the top, while the liquid along with the absorbed gas (CO_2 and H_2S) is drained out at the bottom. The packing helps to increase the mass transfer area. Based on physical absorption process, a biogas water scrubbing system has been developed for the removal of carbon dioxide and hydrogen sulphide from biogas to get biomethane.



Materials and Methods

Two very important and basic parameters for the design of packed bed absorption column for the removal of gases are : (i) the amount of gas to be purified and (ii) the degree of purification. These two parameters determine the size of the absorption column. The liquid flows down the column over the packing surface and gas flows counter currently up the column. The performance of packed column is dependent on the maintenance of good liquid and gas distribution throughout the packed bed.

Based on above principle, a pilot plant of 2 m³/ h with packed column and water as absorbent is developed and installed in the Department of Bioenergy, TNAU, Coimbatore. This consists mainly of a PVC shell, provisions for the gas and liquid inlet and outlet, packing with necessary supporting and redistribution flash tower system. The diameter and height of the packed scrubbing column and packing material height are 0.30, 5.0 and 3.0 m respectively. A schematic diagram, showing the main features of the biogas upgrading system is given in Figure 1.

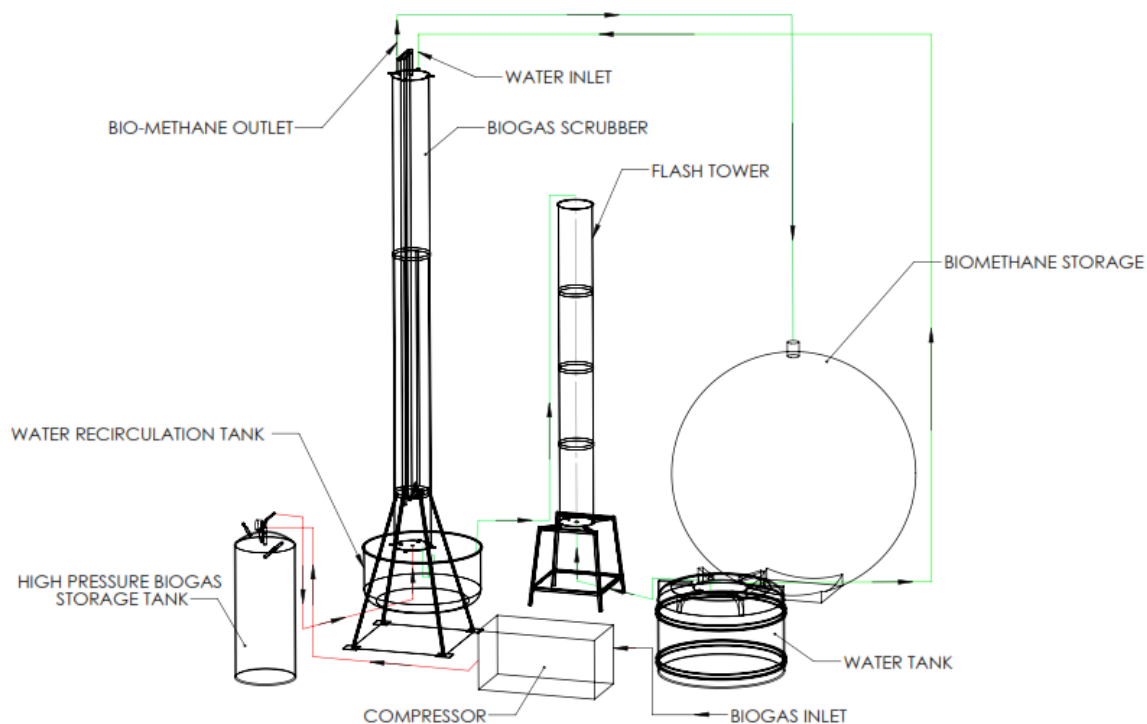


Fig. 1. Biogas Upgrading System

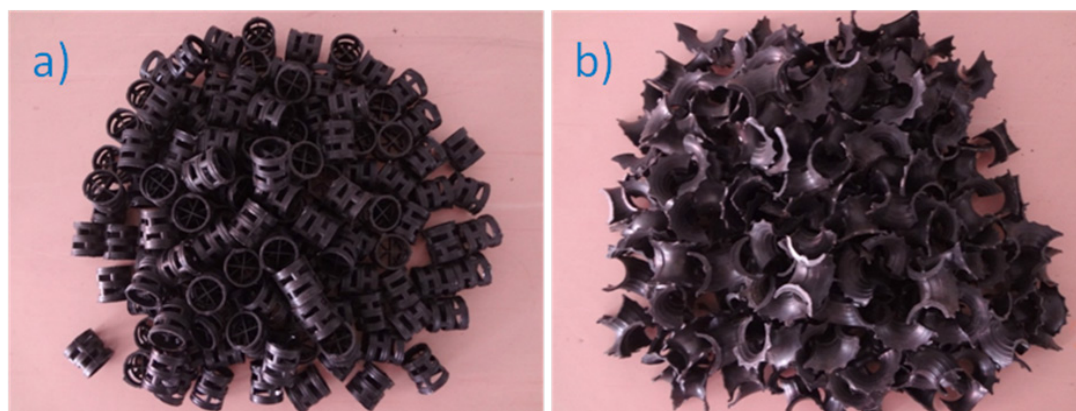


Fig. 2. Packing Materials (a) Pall ring (b) Intalox saddle

Packing material and size

Technically the function of the packing material is to provide an extensive area of liquid surface in contact with the gas phase under the conditions favouring mass transfer. In this study, random packing plastic (polypropylene) materials such as pall ring (1.5 m) and super intalox saddle (1.5 m) are used shown in Figure. 2.

In order to increase the surface area of contact between the liquid and gas, the pall rings are provided with grate like structure, which will further help the easy flow of fluids through the openings. The characteristics of packing material are shown in Table 1.

Table 1. Characteristics of packing material

S.No.	Parameters	Pall ring	Intalox Saddle
	Material	Polypropylene	Polypropylene
	Thickness	1.106 mm	-
	Size	25 mm	-
	Packing factor	52.0	50.0
	Dry Bed P.F	55.0	50.0
	Bulk density	88.0 kg/m ³	80.0 kg/m ³
	Surface area	207.0 m ² /m ³	240.0 m ² /m ³
	Voidage	90.0%	90.0%
	Quantity, Nos	50800 per m ³	40000 per m ³

Performance evaluation of biogas upgrading system was carried out at various pressures (4, 6, 8, 10 bar), gas flow rates (1.50, 1.75, 2.00, 2.25, 2.50 m³/h) and water flow rates (1, 1.5, 2 m³/h) to achieve methane percent of biogas as spell out by the biogas standard BIS 16087:2013, developed by the Bureau of Indian Standards (BIS) for biogas utilization in vehicles (Table 2).

Table 2. Requirements for Biogas into Biomethane (IS 16087 : 2013)

SI. No.	Characteristic	Requirements
1.	CH ₄ , percent, <i>Minimum</i>	90
2.	Moisture, mg/m ³ , <i>Maximum</i>	16
3.	H ₂ S, mg/m ³ , <i>Maximum</i>	30.3
4.	CO ₂ +N ₂ +O ₂ , percent, <i>Maximum</i> (v/v) basis	10
5.	CO ₂ , percent, <i>Maximum</i> (v/v) basis	4
6.	O ₂ , percent, <i>Maximum</i> (v/v) basis	0.5

Results and Discussion

In water the acidic components of the biogas such as CO₂ and H₂S are more easily dissolved than the hydrophobic, non polar components such as methane. Performance of biogas upgrading unit with respect to CO₂ absorption in under the conditions of varying pressures, gas flow rates and water flow rates are shown in Table 3.

The inlet gas flow rates were varied from 1.5 to 2.5 m³/h at 4 bar. The percent absorption of CO₂ increases as the water flow rates increases from 1.5 to 2 m³/h. However, CO₂ absorption percent decreases, as biogas flow rate increases. Similar trend has been observed for 6 to 10 bar operating pressures. It is observed that based on the maximum percent of CO₂ absorption, the biogas flow rate of 1.5 m³/h is optimised under varying column pressures.

Table 3. Performance of biogas upgrading unit in terms of CO₂ absorption percent.

Column pressure, bar	Biogas in-flow rate, m ³ / h	CO ₂ absorption,%		
		Water in-flow rate, m ³ /h		
		1.5	1.75	2.00
4	1.50	74.67	74.89	76.22
	1.75	73.11	73.33	75.11
	2.00	71.33	72.67	73.56
	2.25	70.67	70.89	73.33
	2.50	69.33	70.22	72.67
6	1.50	78.22	78.00	81.11
	1.75	77.56	77.33	80.00
	2.00	76.00	76.67	78.89
	2.25	75.56	75.78	78.67
	2.50	75.11	75.56	78.44
8	1.50	85.56	89.78	90.44
	1.75	84.89	89.33	89.56
	2.00	84.22	88.44	88.89
	2.25	83.56	86.89	88.00
	2.50	82.89	87.33	87.78
10	1.50	87.11	93.11	93.13
	1.75	86.89	92.67	92.67
	2.00	86.44	91.78	92.00
	2.25	86.00	91.56	91.78
	2.50	85.33	90.67	90.67



The effect of pressure on CO₂ absorption with different gas flow rate

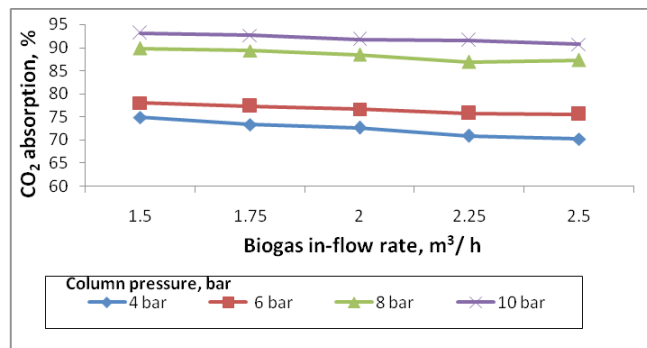


Fig. 3. The effect of pressure on CO₂ absorption with different gas flow rate.

The effect of gas pressure on percent absorption of CO₂ was determined for different gas flow rates corresponding water flow rate 1.75 m³/h and the relationships shown in figure. 3. It is obvious from figure that, higher the pressure, higher will be the CO₂ absorption percent. The percentage of CO₂ absorption ranged from 69.33 to 76.22% at 4 bar, 71.11 to 81.11% at 6 bar, 82.89 to 90.44 at 8 bar and 85.33 to 93.13% at 10 bar at the water flow rates of 1.5, 1.75 and 2 m³/h respectively. The highest percentage of CO₂ absorption is observed for the gas flow rate of 1.5 m³/h for different pressure variation between 4 to 10 bar. The highest CO₂ absorption percent 93.13 is observed at 10 bar gas pressure. This is in agreement with report given by Wellinger and Lindberg (1999) and Vijay (2007) that at 10 bar pressure for inlet gas can give more than 90% of CH₄ in upgraded biogas.

The effect of water flow rate on CO₂ absorption percent under varying column pressures.

Figure 4 shows the effect of water flow rate on CO₂ absorption for the gas flow rate of 1.5 m³/h. At 8 and 10 bar pressures, CO₂ absorption percent is found to the same for different water flow rates 1.75 and 2 m³/h. Hence, it can be concluded that the water flow rate of 1.75 m³/h can be optimised for the optimised gas flow rate 1.5 m³/h.

Table 4. Optimized values of biogas upgrading system at 10 bar operating pressure

Column pressure, Bar	Biogas in-flow rate, m ³ /h	Water in-flow rate, m ³ /h	CO ₂ in raw biogas	CH ₄ content in upgraded biogas (bio-methane) %	CO ₂ remained in upgraded biogas, %	CO ₂ absorption, %
10	1.50	1.75	45	97.00	3.00	93.11
	1.75	1.75	45	96.70	3.30	92.67
	2.00	1.75	45	96.40	3.60	91.78

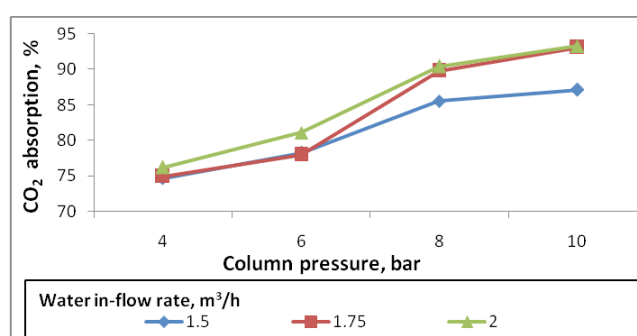


Fig. 4. The effect of water flow rate on CO₂ absorption with 1.5m³/h gas flow rate.

The optimized values for 10 bar operating pressure are shown in Table 4. The highest carbon dioxide percent absorption of 93.11 was observed at 10 bar column-operating pressure with a methane purity of 97% in gas outlet. This can be compared with the results of Chandra *et al.*, 2012 that carbon dioxide absorption is 90.6 and 97 methane percent in upgraded biogas.

It can be concluded from the above table that the biogas flow rate of 2.0 m³/h at water flow rate of 1.75 m³/h for the 10 bar pressure can be recommended considering the economy of scrubbing process.

Removal of hydrogen sulphide

The traces of H₂S available in the biogas was removed completely in water scrubbing packed column. The removal was confirmed by exposing the scrubbed gas



to the lead acetate impregnated strip. The colour of the lead acetate paper was not changed, which indicates the absence H_2S .

Conclusion

In order to meet out the requirements of biogas standard BIS 16087:2013, a biogas upgrading system designed and developed for the removal of carbon dioxide and hydrogen sulphide in one stage. This upgrading biogas to biomethane system, upgrades the biogas up to 96- 97% biomethane at 10 bar column-operating pressure for the biogas in-flow rate of 2 m³/h and water inflow rate of 1.75 m³/h. The upgradation of biogas confirming to the above said biogas standard helpful: (a) to provide the safe operation of the engine whether stationary or automotive (b) to protect the fuel system from the detrimental effects of corrosion, poisoning and liquid or solid deposition; and (c) do not emit any pollutant or the greenhouse gases after combustion, beyond prescribed limit.

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References

- Abatzoglou, N. and Boivin, S. 2009. A review of biogas purification processes. *Biofuels, Bioproducts and Biorefining* 3: 42–71.
- Bhattacharya, T. K., Mishra, T. N. and Singh B. 1988. Techniques for removal of CO_2 and H_2S from Biogas. Paper presented at XXIV annual convention of ISAE, held at PKV, Akola.
- Chandra, R., Vijay, V. K. and Subbarao. P. M. V. 2012. Vehicular Quality Biomethane Production from Biogas by Using an Automated Water Scrubbing System. *ISRN Renewable Energy* 1-6.
- Dubey, A.K. 2000. Wet scrubbing of carbon dioxide. Annual report of CIAE, Bhopal, India.
- IS 16087 (2013): Biogas (Biomethane) – Specification- PCD 3: Petroleum, Lubricants and their Related Products, Bureau of Indian Standards, New Delhi.
- Khapre, U. L. 1989. Studies on biogas utilization for domestic cooking; Paper presented at XXV annual convention of ISAE, held at CTAE, Udaipur.
- Nathan, S. S., Mallikarjuna, J.M. and Ramesh, A. 2010. An experimental study of the biogas-diesel HCCI mode of engine operation. *Energy conservation and Management* 51(7):1347-53.
- Persson, M., Jönsson, O. and Wellinger, A. 2006. Biogas Upgrading to Vehicle Fuel standards and Grid Injection. IEA Bioenergy, Task 37 – Energy from Biogas and Landfill Gas.
- Ramanathan, M. 1996. Studies on carbondioxide scrubbing of biofuel gases and production of hydrogen. Thesis Ph.D. (Bioenergy). Department of Bioenergy, Tamil Nadu Agricultural University, Coimbatore, India.
- Rasi, S., Veijanen, A. and Rintala, J. 2007. Trace compounds of biogas from different biogas production plants. *Energy* 32(8): 1375-80.
- Vijay, V. K. 1989. Studies on utilization of biogas for improved performance of dual fuel engine. Thesis M E (Ag). CTAE, Udaipur, India.
- Vijay, V. K. 2007. Biogas refining for production of Bio-Methane and its bottling for automotive applications and holistic development. Proceeding of International symposium on Eco Topia Science. Nagoya University, Nagoya, Japan. November 23-25: 623-628.
- Vijay, V. K. 2014. Upgraded bottled biogas, A green and low-cost fuel for automobiles in India. *Akahay Urja* 7(5): 20-23.
- Vijay, V. K., Chandra, R., Subbarao, P. M. V., and Kapid, S. 2006. Biogas purification and Bottling into CNG Cylinders: Producing Bio-CNG from Biomass for Rural Automotive Applications. A paper presentation at The 2nd Joint International Conference on Sustainable Energy and Environment (SEE) on 21-23 November 2006 Bangkok, Thailand.
- Wellinger, A. and Lindberg, A. 1999. Biogas upgrading and utilisation. IEA Bioenergy Task 24: Energy from Biological conversion of organic waste 3-19.