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Review Article

Mechanism of Methane Production in Biogas Plant and its Upgrading Methods

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Abstract

To meet the energy demands of the growing population, shift towards renewable sources of energy is the need of the hour. The wastes generated in the households and farms can be efficiently treated by biomethanisation technology. In the biogas digester, the organic wastes undergo anaerobic degradation in the presence of different groups of micro-organisms to produce a mixture of gas with higher amounts of methane. To efficiently use the produced biogas for various purposes, it is necessary to upgrade the biogas to biomethane. This review includes, the biochemical processes involved in the biogas production along with the parameters affecting the operations and also the various methods involved in upgrading it.

Keywords: Anaerobic Digestion, Biogas, Biomethane, Renewable, Upgrading

Introduction

Energy is the most fundamental requirement of every nation as it progress through the ladder of development. To meet the food security of the ever-growing human population, there is also an increase in the livestock and poultry farms in most nations. As the animal population increases in the farm, the amount of animal waste produced also increases. In order to maintain healthy and hygienic conditions in the farm, it is necessary to treat the wastes scientifically. One of the easy and highly accepted technologies is the production of biogas, utilising wastes from different sources (Achinas *et al.*, 2017).

Biogas technology is the anaerobic bacterial degradation of the organic matter which produces a gas rich in methane which can be used for cooking, electricity generation and as vehicle fuel. The plant also produces an additional byproduct- digestate, which can be directly applied on the field to increase the nutritive quality of the soil and enhance the soil bacteria (Goswami *et al.*, 2016). The biogas production technology involves two stages: 1. Acid fermentation and 2. Methane fermentation (Hagos *et al*., 2016). The micro-organisms involved in the process are archae bacteria belonging to two different biological kingdoms (Adekunle and Okolie, 2015).

History

The history of flammable gas production from decaying organic matter goes back to 17th century which was recorded by Van Helmont. John Dalton and Humphrey Davy in 1804-1808, found that, the combustible gas produced during organic decay was methane. The methane production involved microbiological process was given by Bechamp in 1868. In 1890s, the microbes responsible for hydrogen, acetic and butyric acid formation were isolated by Omelianski (Abbasi *et al*., 2011).

Anaerobic Digestion Process

Anaerobic digestion is an oxygen independent process, which occurs naturally during the decomposition of the organic matter. In this process, the complex and insoluble organic matter are broken down to simpler components by a group of archae and symbiotic bacteria (Christy *et al*., 2014). The four biochemical steps involved in the biogas production are: hydrolysis, acidogenesis, acetogenesis and methanogenesis (Zieminski and Frąc, 2012).

Hydrolysis

Hydrolysis, the first step in biogas production involves depolymerisation of complex organic substances like sugars, proteins and lipids into the smaller compounds like simple sugars, alcohols and fatty acids, brought about by a large number of hydrolytic bacteria like *Bacteroides, Butryvibrio, Clostridia, Fusobacterium, Selenomonas, Micrococcus, Peptococcus* and *Streptococcus*. The micro-organisms involved in the reaction can be either facultative or obligatory anaerobes. The action of the bacteria is mainly by the secretion of the hydrolytic enzymes like amylase, cellulase, hemicellulase, proteinase, pectinase, lipase, etc. which carry out the break down process (Adekunle and Okolie, 2015). The hydrolysis reaction involves two phases. The first phase involves covering the solid surface of organic matter by the hydrolytic bacteria where, the micro-organisms release the hydrolytic enzymes on the surface bringing about the depolymerisation action. Thus, the monomers produced by the action of hydrolytic enzymes will be used either by the same micro-organisms or by another group of micro-organisms. During the second phase, the degradation of the particle size occurs at a constant rate (Christy *et al*., 2014).

$$(C_6H_{10}O_5)_n + nH_2O = n(C_6H_{10}O_5)$$ (Deepanraj *et al*., 2014)

Acidogenesis

Acidogenesis being the second step in anaerobic digestion occurs at a fastest rate. The fermentative micro-organisms like *Streptococcus, Bacillus, Lactobacillus, E. coli, Desulfovibrio, Selenomonas,* etc., act on the products of hydrolysis to produce the organic acids like acetic, propionic and butyric acids, along with some small chain fatty acids, alcohols, hydrogen and carbon-dioxide (Zhou *et al.*, 2018). The production of organic acids results in reduced pH of 4.5-5.5. The intermediate products of glucose metabolism will produce pyruvic acid by following the glycolytic Embden- Meyerhoff Parnas pathway. The pyruvic acid, depending on the micro-organisms present undergoes puruvic acid fermentation to produce Volatile Fatty Acids (VFA). The facultative bacteria sometimes,

depend on the amino acids as a source of carbon and energy (Christy *et al*., 2014).

$$C_6H_{12}O_6 + 2H_2O \longrightarrow 2CH_3COOH + 4H_2 + CO_2$$

$$C_6H_{12}O_6 + 2H_2 \longrightarrow 2CH_3CH_2COOH + 2H_2O$$

$$C_6H_{12}O_6 \longrightarrow CH_3CH_2CH_2COOH + 2H_2 + 2CO_2$$

$$C_6H_{12}O_6 \longrightarrow 2CH_3CH_2OH + 2CO_2$$

$$C_6H_{12}O_6 \longrightarrow 2CH_3CHOHCOOH$$

Acetogenesis

Acetogenesis being the third step is mainly for those acidogenic products which cannot be directly used as methanogenic substrate by methanogens in methane production. The VFAs with carbon chains more than two and alcohols containing carbon chains more than one undergo acetogenesis in the presence of obligatory acetogenic bacteria to produce acetate and hydrogen (Drake, 1994). The acetogens are the strict anaerobic bacteria that synthesise acetate by using carbon-dioxide by any of the three mechanisms- Acetyl-CoA pathway, Reductive citric acid cycle and Glycine-synthase dependent pathway, but most of the organisms follow the Acetyl-CoA pathway. Some of the acetogenic bacteria include '*Syntrophomonas wolfeii', 'Syntropho-bacter wolinii'*. There exists a syntropic relationship between the acetogens and hydrogenotropic methanogens (Goswami *et al*., 2016).

$$CH_3CH_2OH + H_2O \longrightarrow CH_3COOH + 2H_2$$

$$2CH_3CH_2OH + 2CO_2 \longrightarrow CH_4 + 2CH_3COOH$$

$$CH_3CH_2COOH + 2H_2O \longrightarrow CH_3COOH + 3H_2 + CO_2$$

$$CH_3CH_2CH_2COOH + 2H_2O \longrightarrow 2CH_3COOH + 2H_2$$

$$CH_3CHOHCOOH + H_2O \longrightarrow CH_3COOH + CO_2 + 2H_2$$

Methanogenesis

The final stage of anaerobic digestion wherein, the two different groups of methanogens use the important substrates like hydrogen, carbon-dioxide, acetic acid, formic acid, methanol, methylamine and dimethyl sulphate to produce methane. The two different groups of methanogens include- aceticlastic and hydrogenotropic methanogens (Sekiguchi *et al*., 2001). The acetotropes use acetate as a substrate and they split the acetate to produce carbon-dioxide from the carboxyl group and methane from the methyl group. The hydrogenotropes use hydrogen and carbon-dioxide as substrates to produce methane. As of now, only two groups of acetotropes are known which include- Methanosaeta and Methanosarcina which differ in growth rates and are sensitive to the changes in the acetate concentration. The Methanosarcina group activates acetate to acetyl CoA by acetate kinase phosphotransacetylase system. The Methanosaeta uses Adenosine Mono Phosphate (AMP) for the conversion. The different groups of hydrogenotropes include Methanobacterium, Methanococcus, Methanobrevibacter and Methanogenium. The hydrogenotropes produce methane by stepwise reduction of carbon-dioxide in the presence of the special coenzymes like methanofuran, tetrahydromethanoptein and coenzyme M with, methyl-coenzyme M reductase being the key enzyme which helps in the reduction of methyl-coenzyme M to methane (Zieminski and Frąc 2012).

$$CH_3COOH \longrightarrow CH_4 + CO_2$$

$$CO_2 + 4\,H_2 \longrightarrow CH_4 + 2H_2O$$

Rate-Limiting Step

The rate limiting step in the anaerobic digestion is the one which causes the process failure under the kinetic stress conditions. Kinetic stress is mainly due to the reduced solid retention time in the digesters which causes washing out of the micro-organisms mainly methanogens from the digester (Adekunle and Okolie, 2015). Most commonly

methanogenesis is considered as the rate limiting step.

In some cases, with optimum retention time and substrate concentration, the rate of hydrolysis can be the rate limiting step. If the rate of hydrolysis is slow then, the substrate availability for the acidogenic and methanogenic bacteria will reduce, thereby causing caessation of methane production (Liu *et al*., 2016).

Important Parameters Affecting the Anaerobic Digestion

The important factors which affect the anaerobic digestion can depend on the type of the substrate and also on the operating parameter conditions. The factors relating to the substrate include the toxic elements, pH, inhibitory substances or rate of hydrolysis which causes an increased amount of VFAs. At the same time, the operating parameters like temperature, pH, organic loading, moisture level etc can also affect the biogas production (Khalid *et al.*, 2011).

Operating Parameters

Temperature

In the anaerobic digestion, temperature plays an important role. The methanogens can perform their activities in three different temperature groups: Psychrophilic (<20℃), Mesophilic (20-40℃) and Thermophilic (45-60℃). But for biogas production, only mesophilic and thermophilic temperature are preferred (Goswami *et al.*, 2016). As the temperature increases, the rate of gas production also increases till the temperature reaches 60-70℃ (Scherer *et al*., 2000). When the temperature exceeds 60℃, the methanogen's activity is reduced by higher percentage than the acid forming bacteria, which causes the accumulation of fatty acids.

Mesophilic Digestion

In this, the optimum temperature ranges between 35-37℃. But when the temperature falls below this optimum range, the organisms which are less sensitive to the temperature alone replicate and carry out the process.

Thermophilic Digestion

Most of the thermophilic digesters work best at the range of 50-55℃. At higher temperature the organisms work faster, with the increased availability of organic compounds (from the increased solubility) which reduces the viscosity of the material thereby, facilitating mixing (Schnurer and Jarvis, 2010). The high temperature also kills the pathogenic organisms maintaining the sanitation in the digester (Sahlstrom, 2003). When compared between mesophilic and thermophilic, the mesophilic temperature is more preferred as the microbial diversity is varied and rich and also the fluctuating parameters won't affect the gas production to greater degree (Khalid *et al.*, 2011).

pH

The pH range varies for different biochemical reactions. The optimum range of pH for the methanogenesis is 6.8-8.5 whereas; it is 5.5 and 6.5 for hydrolysis and acidogenesis respectively (Lee *et al.*, 2009). The ideal pH for the optimal anaerobic reaction is 6.8-7.2 (Ward *et al.*, 2008). When the digesters are fed with the easily fermentable carbohydrates, the production of VFAs is greater than the rate of methanogenesis, which results in the lowering of pH in the digester. For optimal working of the biogas plants, it is necessary to maintain the volatile acid to total alkalinity ratio of 0.5 (Nijaguna, 2006).

Hydraulic Retention Time (HRT)

HRT is the duration of time (in days) during which the substrate should remain in the digester for complete digestion and for maximum gas production. The retention time varies depending on the composition of the substrate and the temperature (Amani *et al.*, 2010). When the substrate added contains higher amount of easily fermentable carbohydrates like starch and sugar, the retention time is lower. If the substrate contains higher amount of fibre and cellulose then, the retention time is longer. Nijaguna (2006) stated that for mesophilic bacteria, the digestion period ranges between 20-30 days whereas, for the thermophilic bacteria, the period of digestion is between 3-10 days.

Carbon-Nitrogen (C/N) Ratio

For the normal growth and activity of the microbes in the digester, it is necessary to maintain the optimum C/N ratio of around 25 to 30:1. The carbon provides energy while, the nitrogen helps in building the cell structure. The C/N ratio varies for different substrates and it mainly depends on the biodegradability index. The ideal biodegradable carbon to available nitrogen ratio should be 25:1 for the biogas production. The higher ratio will speed up the usage of nitrogen by the methanogens while, the lower ratio results in ammonia accumulation (Ohorhoro *et al*., 2016).

Water Content

For most of the activities in the digester, water is essential. The slurry added to the digester should be neither too thick nor too thin. The optimum concentration of total solids in the digester should be in the range of 7-25 % (Deepanraj *et al*., 2014). The high moisture level in the slurry reduces digester temperature and the low moisture content will cause accumulation of acids affecting the fermentation process.

Organic Loading Rate (OLR)

OLR is the rate at which the biomass is added to the digester and is expressed in grams of volatile solids per unit volume of digester per day. Angelidaki *et al*., 2006 stated that during the start of new process, the loading rate should be low and the rate should be gradually increased for the optimal growth of micro-organisms. If the plant is initially loaded with higher amount of organic matter then, there are chances of complete imbalance in the entire digestion process.

Table 1: Composition of Biogas (Munoz *et al*., 2015)

Composition	Percentage
Methane	50-70%
Carbon-dioxide	30-50%
Hydrogen Sulphide	0-10000 ppm
Water vapour	5-10%
Nitrogen	0-3%
Oxygen	0-1%
Hydrocarbons	0-200 mg/m^3
Siloxanes	0-41 mg/m^3

The nature of the substrate and pH of the digester causes variation in the content of CH_4 and CO_2. The calorific value of raw biogas is low due to high concentrations of CO_2 and N_2 (Ryckebosch *et al*., 2011). The high levels of O_2 may results in explosion and the H_2S upon combustion will produce SO_2 which causes corrosion (Petersson and Wellinger., 2009). To avoid the problems from different gases and also to increase the calorific value of biogas, the gas has to be purified.The conversion of biogas to biomethane involves two processes- 1. Cleaning process, to remove the trace elements 2. Upgrading process, to increase the calorific value of methane, mainly by removing carbon-dioxide (Kougias *et al*., 2017).

Biogas Cleaning

Apart from methane, the rest all gases in the raw biogas are referred as pollutants. In the biogas cleaning, the pollutants like water vapour, hydrogen sulphide, oxygen, nitrogen, ammonia, hydrocarbons, siloxanes and particulates are removed.

Water Removal

Biogas leaving the digester, gets saturated with water vapour and this can be removed by absorption, adsorption, cooling and compression. With the increase in the pressure or decrease in the temperature of the biogas, there will be condensation of the water thereby, removing it. Adsorption of the water can be carried out by using activated carbon, molecular sieves or silicon dioxide. The use of glycol solution or some hygroscopic salts helps in removal of water by absorption mechanism (Petersson and Wellinger, 2009). Activated Alumina balls are used in commercial

plants to remove water.

Hydrogen Sulphide Removal

The hydrogen sulphide which is a poisonous gas is removed either during the digestion or after the digestion. The concentration of hydrogen sulphide can be reduced by precipitation, adsorption, absorption or by biological method.

Before Digestion

It can be carried out by using Fe^{3+} or Fe^{2+} ions in the form of $FeCl_2$, $FeCl_3$, or $FeSO_4$ in the digester. The H_2S reacts with Fe ions to form insoluble iron sulphide which can be removed along with digestate. Other method is biological treatment- by giving air dose to the digester, which is based on the aerobic oxidation reaction carried out by the sulphate oxidizing bacteria like Thiobacillus and Sulpholobus. (Ryckebosch *et al*., 2011).

After Digestion

Adsorption using activated carbon, where the H_2S will get oxidised to S in the presence of water and oxygen thereby facilitating adsorption. To speed up the reaction, the activated carbon can be impregnated with zinc oxide or potassium permanganate or iodide. The activated carbon can be regenerated by supplying air. Absorption of H_2S can be carried either physically or chemically. Most commonly water is used as a solvent and in physical method, H_2S gets dissolved in water. In the chemical absorption, the commonly used chemicals can be NaOH, $FeCl_2$ etc. The reaction of NaOH with H_2S yields Na_2S or NaHS which get precipitated and Na salts cannot be regenerated. With the $FeCl_2$, there is a formation of insoluble FeS which has to be separated.

Membrane Separation

The H_2S can also be removed by semi-permeable membranes along with O_2, CO_2, H_2O and NH_3, but CH_4 cannot pass through the membrane (Schomaker *et al*., 2000). The membrane separates the liquid and gas phase. When the gas flows in one direction it diffuses through the membrane and the gas molecules will be absorbed by the liquid on the other side by counter current mechanism. Most commonly NaOH is used as absorbing liquid (Wellinger and Lindberg, 1999).

Biogas Upgrading

To increase the calorific value of the biogas, it is necessary to separate carbon-dioxide from methane. There are different physical and chemical methods employed for upgrading biogas and these methods involve different processes like absorption, adsorption and membrane separation.

Pressure Swing Adsorption (PSA)

In this, different gases are separated based on their molecular characteristics and their adsorption affinity for adsorbents. When the pressure of the gas is increased, large amount of gas will get adsorbed and upon decreasing the pressure, the gases are released. The process involves four steps- adsorption, blow-down, purge and pressurization (Augelletti *et al*., 2017).

Cryogenic Separation Process

In this, the upgrading is carried out by drying and compressing the gas to 80 bars and then, decreasing the temperature gradually till it reaches -110℃ (Ryckebosch *et al*., 2011). The pollutant gases along with CO_2 will be gradually removed leaving behind the pure biomethane.

Biological Technologies

In this, the chemoautotropic or photosynthetic mechanisms are used to upgrade the biogas and also, the CO_2 in the biogas will be converted to other high energy containing product. The chemoautotropic upgrading method is based on the principle of hydrogenotropic methanogenesis which use H_2 and CO_2 to produce CH_4. To make this upgrading method renewable, trials are going on to generate H_2 from the renewable electricity (from wind mills and solar

energy) which is used to hydrolyse the water to H_2 and O_2. This has resulted in the development of concept called Power to Gas (P2G). Thus, in this method the CO_2 is converted to CH_4 which results in a high energy output, which is referred as wind gas (if the renewable electricity is from wind mills) or solar gas (if the electricity is from solar panels) (Kougias *et al*., 2017).

In the photosynthetic upgrading method, CO_2 is converted to CH_4 using photoautotropic micro-organisms like prokaryotic cyanobacteria and eukaryotic microalgae. The raw biogas will be injected directly into the photobioreactors or in the absorption column which is circulated with the microalgal broth stream. The micro-organisms utilize CO_2 in the presence of solar irradiation, water and nutrients to produce the biomass, oxygen and heat. Therefore, the yield of the methane will be increased and the CO_2 level can be minimized to 2-6% in the final pure gas (Munoz *et al*., 2015).

Conclusion

To meet the energy demand of the growing population, it is necessary to shift towards the sustainable and eco-friendly source of energy, one of which could be biogas. To enhance the productivity of the biogas, it is necessary to understand each and every step-in detail along with the eco-physiology of different microbes and the operational parameters. Apart from enhancing the gas productivity, it is of importance to upgrade the produced gas. Not only the physico-chemical upgrading technologies but also, the biological upgrading technologies can be developed to integrate different renewable sources of energy and bring about the development of green economy. The upgraded methane rich gas can be compressed in the cylinders to be supplied as compressed biomethane. Also, by applying high pressure and low temperature, the gas can be liquefied to produce liquefied biogas.

Conflict of Interests

There is no conflict of interest.

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